The Absorption of Laser Light by Rough Metal Surfaces

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Doctoral Thesis

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Luleå, February 2008
Dedicated to the Curious Mind.
Cover figure: A laser profilometer image of the topography of a hot rolled stainless steel surface (middle), with a photograph of an experimental setup for absorptance measurements using an integrating sphere (left) and a computer-generated Gaussian random rough surface illustrating the principles of ray-tracing (right).
Acknowledgements

The work presented in this Doctoral thesis is devoted to the theoretical and experimental study of the light absorption mechanisms in laser processing of rough metals and metallic alloys. The Doctoral thesis consists of a short introduction and a series of six papers, all dealing with different aspects of this topic.

This work has been carried out at the Division of Manufacturing Systems Engineering at Luleå University of Technology (LTU) in Luleå, Sweden, and at the Department of Engineering, Physics and Mathematics at Mid Sweden University (MIUN) in Östersund, Sweden, from 2003 to 2008.

I would like to express my gratitude to my supervisors Prof. Alexander Kaplan (LTU) and Prof. Torbjörn Carlberg (MIUN) for their advice and support during these past years. My deepest thanks go to Prof. John Powell (Laser Expertise Ltd., Nottingham, UK) for always keeping me focused on the essentials. I’d like to thank him for all the time he has devoted in the discussions of different thoughts and ideas and for proof-reading all my written work. It’s been a pleasure working with you and Alexander and I wish and hope for further collaboration in the future.

Thanks are also due to M.Sc. Rickard Olsson and his colleagues at Lasernova AB in Östersund, for once upon a time introducing me to this exciting subject and for their support and friendship. I’m also indebted to Patrik Frihlén of Azpect Photonics AB and M.Sc. Ingemar Eriksson for helping me setting up the equipment for the laser lab in Östersund. Further thanks go to Doc. Mats Tinnsten and Dr. Leon Dahlén for providing the financial circumstances in making this work possible. I would also like to thank my colleagues at my university department in Östersund, especially my fellow PhD student Marianne Olsson for sharing countless cups of coffee over conversations about the trials and tribulations of postgraduate studies as well as of topics beyond.

Finally I would like to thank my family for their support and encouragement and my girlfriend Susanne for her love, patience and understanding.

Östersund, February 2008

David Bergström
Abstract

In Laser Material Processing of metals, an understanding of the fundamental absorption mechanisms plays a vital role in determining the optimum processing parameters and conditions. The absorptance, which is the fraction of the incident laser light which is absorbed, depends on a number of different parameters. These include laser parameters such as intensity, wavelength, polarisation and angle of incidence and material properties such as composition, temperature, surface roughness, oxide layers and contamination. The vast theoretical and experimental knowledge of the absorptance of pure elements with smooth, contamination-free surfaces contrasts with the relatively sparse information on the engineering materials found in real processing applications. In this thesis a thorough investigation of the absorption mechanisms in engineering grade materials has been conducted, both experimentally and theoretically. Integrating sphere reflectometry has been employed to study the impact of surface conditions on Nd:YAG and Nd:YLF laser absorptance of some of the most common ferrous and non-ferrous metallic alloys found in Laser Material Processing. Mathematical modelling and simulations using ray-tracing methods from scattering theory have been used to analyze the influence of surface topography on light absorption.

The Doctoral thesis consists of six papers:

Paper 1 is a short review of some of the most important mathematical models used in describing the interaction between laser light and a metal surface.

Paper 2 is a review of experimental methods available for measuring the absorptance of an opaque solid such as a metal.

Papers 3 and 4 are experimental investigations of the absorptance of some of the most frequently found metallic alloys used in Laser Material Processing today.

Paper 5 presents results from 2D ray-tracing simulations of random rough metal surfaces in an attempt to investigate the influence of surface roughness on laser scattering and absorption.

Paper 6 is a full 3D ray-tracing investigation of the interaction of laser light with a rough metallic surface, where some comparisons also are made to the previous 2D model.
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Paper 2: Laser Absorption Measurements in Opaque Solids


Paper 5: A Ray-Tracing Analysis of the Absorption of Light by Smooth and Rough Metal Surfaces

Paper 6: The Absorption of Light by Rough Metal Surfaces - A Three-Dimensional Ray-Tracing Analysis
1. Motivation of the thesis

Metal processing with lasers has reached a high level of maturity and acceptance in industry. It is used for cutting, drilling, welding, forming, engraving, marking, hardening and various forms of surface treatment of metals in a broad spectrum of modern industries, including the automotive and aerospace industries, the shipbuilding industry, the microelectronics industry and the medical instrument industry to name a few.

Common to all these applications, is that the laser light is transformed into heat. This is the fundamental process of light absorption and can often be a determining factor in whether a process will be successful for a particular material under certain circumstances or not.

In the field of material optics the studies of light interaction with metals have usually been concentrated on pure metallic elements with surfaces as smooth and clean as possible. This is understandable from the point of view of finding correlation with existing theories from solid state and condensed matter physics. If any handbook of chemistry or physics is picked up from a library, this is usually the experimental data that will be found. However, the engineering grade metals and metallic alloys found in real life processing with lasers have various forms of texture and roughness on their surfaces and also have layers of oxides on top of them. The sparse amount of experimental data and mathematical models for these kinds of non-perfect but more realistic surfaces is a limitation to the modellers of laser metal processing who need accurate heat input data for their heat conduction problems. The main motivation for this research was to provide more realistic information about the absorption of laser light by engineering metal surfaces.

2. Methodological approach

The methodological approach of this Doctoral thesis can be understood by reference to the structure and order of the papers included. The work began by making an overview of the existing mathematical models of laser absorption in
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metals (see Paper 1 for details) and this lead to a realisation of the lack of theory and experimental data for the engineering grade metals and metallic alloys found in real life processing applications.

A study and review of the available experimental methods for measuring laser absorption in metals was then initiated and a potential candidate was found in the method of measuring reflectance using an integrating sphere. For opaque solids like metals, which do not transmit light unless they are very thin, a direct correspondence between reflectance and absorptance can be assumed (see Paper 2 for details).

The discovered lack of experimental data for rough and oxidised metals lead to an experimental investigation of the reflectance and absorptance of some of the most frequent steels and non-ferrous alloys found in processing with lasers today, including mild and stainless steel, copper, aluminium and brass, etc. The results of these investigations can be found in Papers 3 and 4.

These experimental studies revealed that surface conditions play a very important role for the total absorptance, including the influence of oxide layers and surface roughness which may vary substantially between different as-received surfaces depending on the manufacturing and surface finishing processes used. Most of the previous modelling of how surface topography affects absorptance has considered either of two extreme roughness regimes; when $\delta \ll \lambda$ or when $\delta \gg \lambda$, where $\delta$ is the roughness parameter (usually the $R_a$ or the $R_q$ value) and $\lambda$ is the wavelength. For Nd:YAG or Nd:YLF laser light, the wavelengths of the fundamental and the frequency-doubled modes can very well be in the order of the roughness parameter, in which case more exact approaches from EM wave scattering theory have to be used. These are computationally very intensive methods, usually involving the necessity of super computers for accurate solutions and approximate methods are therefore highly desirable. Ray-tracing, or the geometric optics approximation as it is also known as in scattering theory, provides a promising approximate method. In Papers 5 & 6, ray-tracing methods have been developed to study the influence of surface roughness on absorption in two and three dimensions, respectively.
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3. Laser absorption in metals – a short introduction

Shortly after the advent of the first ruby laser in 1960, experimental investigations of laser effects on materials began to appear. The first lasers developed were too weak and too unstable for any industrial use, but since the early 60’s the field of lasers and photonics has evolved and expanded at such a rate that modern lasers are capable of cleaning, marking, cutting, welding, drilling and surface treating diverse forms of materials with remarkable precision. As higher powers, better beam qualities and an expanded number of wavelengths have become available, more and more applications are being invented, investigated and brought into practical use. The future for these technologies seems bright as we enter the 21st century, which by optimists is predicted to become “the century of the photon”.

One of the two true workhorses of laser metal processing is the Nd:YAG laser (the other one being the CO\textsubscript{2}), operating in the near infrared just outside the visible wavelength region (see Figure 1). The Nd:YAG laser (or the similar in wavelength (colour) Nd:YLF laser) is used routinely to cut and weld metals, metallic alloys and ceramics. Most of the experimental work in this Doctoral thesis is devoted to the study of the laser absorption mechanisms involved when using a Nd:YAG or a Nd:YLF laser to process metals, but many of the results and conclusions are applicable to other laser sources as well.

![Infrared, visible and ultraviolet spectral wavelength regions](image)

Figure 1: The infrared, visible and ultraviolet spectral wavelength regions, with the positions of the CO\textsubscript{2}, the Nd:YAG and the Nd:YLF laser wavelengths. SHG denotes the second harmonic generated (double-frequency, half wavelength) versions of the latter two.

For a laser metal processing application to be possible, the electromagnetic energy of the laser light needs to be transformed into thermal energy inside the metal. The amount of transformed energy is determined by the light absorption mechanisms in the metal. It is this “secondary” type of energy, the absorbed
energy, rather than the laser beam itself, that is available for heating the metal and producing the effect that we want, whether it be cutting, welding, drilling or so on. Laser absorption in a metal depends on a number of different parameters, involving both the laser and the metal.

The laser parameters of importance are the wavelength (or colour) of the light, the angle with which the beam impinges on the metal surface (see Figure 2) and the polarization of the beam, which is related to how the electric field in the light wave is oriented. It can also, in some circumstances, be dependent on the intensity, which is a combination of the power and focal spot size of the laser beam.

![Figure 2: Depicting the angle $\theta$ with which a laser beam strikes a surface.](image)

The primary material parameter determining the amount of absorbed light is the composition of the material, whether we are dealing with a pure element (such as copper, iron, aluminium, etc.) or an alloy (such as brass or steel). Regardless of the composition, light always interacts with the electrons inside the metal or the alloy, since light is an electromagnetic wave and electric and magnetic fields only interact with charged matter (atomic nuclei are usually so heavy that they cannot be moved around easily and their influence is often neglected). The electrons will be accelerated by the electric field and through various collisions with the other constituents of the metal, energy will be transferred to the lattice (the 3D atomic structure of the solid).
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As energy is transferred to the metal, it will heat up and as the temperature is elevated the amount of absorbed light may change, since both the electrons and the lattice atoms in the metal gain kinetic energy which will influence the collision frequency.

Absorption is also heavily dependent upon the surface properties of the metal or alloy. Most real life surfaces are not perfectly flat (not even near-perfect mirrors) and have certain degrees of texture and roughness to them, which will influence their optical behaviour. Pits and valleys (see Figure 3) may, for instance, “trap” some of the light and thereby enhance absorption.

![Figure 3: Left hand side: Profilometry scan of a rough metal surface. Right hand side: The texture of a surface may “trap” some of the light and enhance absorption.](image)

Metals also naturally have a layer (or several layers) of oxides on the surface and the chemical and optical properties of the oxides can often be very different from the properties of the metal or alloy underneath. We may then, for instance, get the situation depicted in Figure 4 where the light is “caught” by the oxide layer which may further increase the absorption.
Finally, contamination such as dirt, oil or dust also changes the absorptive potential of a metal surface. This can involve substances left by earlier processing steps (such as polishing or finishing), from handling or even from the fabrication of the metal or the alloy itself.

4. Conclusions of the thesis

The Doctoral thesis consists of six papers:

The first paper is a short review of some of the most important mathematical models on laser absorption in metals and explains the laser and metal parameters of interest in this respect. These are the laser parameters of wavelength, angle of incidence, polarization and intensity and the material parameters of composition, temperature, roughness, oxide layer chemistry and thickness and various forms of contaminations in the bulk and on the surface of the material.

The second paper is a review of a number of experimental methods available for the scientist and engineer to measure and quantify how much laser light is absorbed in a metal. The absorbance can be measured “directly” using laser calorimetry. It can be determined indirectly by measuring the reflectance, using reflectometers like gonioreflectometers, integrating spheres and integrating mirrors, or by measuring emittance, which is done by emittance radiometry.
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Papers three and four are experimental investigations of the laser absorptance of some of the most commonly used engineering grade metals and alloys used in modern material processing with lasers, including mild and stainless steel, aluminium, copper and brass. In this work, it was found that the measured absorptances of as-received engineering grade metals and alloys can differ substantially from the tabulated values found in literature, since the latter are usually for pure, clean and smooth surfaces.

The final two papers are theoretical studies of the influence of surface roughness or topography on the laser absorptance, using Monte Carlo simulations based upon ray-tracing methods from EM wave scattering theory on random rough surfaces with Gaussian surface statistics. Paper 5 analyses the two-dimensional scattering problem for one-dimensional rough surface profiles, whereas Paper 6 treats the full three-dimensional scattering problem for two-dimensional rough surfaces. In both papers it is concluded that the absorptance is a function of the RMS slope which is proportional to $\sigma/\tau$, where $\sigma$ is the RMS height and $\tau$ is the correlation length. At normal incidence, it is found that the absorptance has a threshold at a certain RMS slope, after which it increases quite sharply due to the onset of multiple scattering. The effect of diminishing returns makes surfaces that are more reflective in the smooth state more sensitive to roughness than less reflective ones. At oblique incidence, the multiple scattering threshold is lowered and shadowing inhibits further increase of scattering, causing the absorptance to be more stagnant or even to decrease in certain slope ranges. At grazing incidence, single scattering is a dominant feature and the behaviour depends very strongly on the specific Fresnel absorptivity curve of the metal in question. While the phenomenology is very similar between the 3D and 2D models, it is concluded in Paper 6 that the 3D model shows an increased level of scattering (and therefore absorption) as compared to the 2D model, mainly owing to the different inclination angle distributions that exist for two- and one-dimensional surfaces, respectively.

5. Future outlook

Laser absorptance in metals is a complex but vital subject for improving the understanding and the efficiency of many laser material processing applications. The main focus of this PhD thesis has been on the influence of surface conditions, both experimentally through a series of reflectometry measurements of
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engineering grade metal surfaces and theoretically through the development and analysis of two ray-tracing models.

The ray-tracing models have been used on isotropic Gaussian random rough surfaces, exclusively, with only one roughness scale (one specific correlation length). A direct and quite straight-forward idea to continue this research would be to investigate other types of surfaces, for instance non-isotropic surfaces, surfaces with two or more roughness scales or surfaces with non-Gaussian statistics (e.g. exponential, fractal, etc.).

The ray-tracing models could also be extended to include interactions through a semi-transparent and absorbing layer, e.g. an oxide layer. Tang & Buckius developed an augmented ray-tracing model where equations from the theory of thin film optics were thought to incorporate the interference effects of such a layer [2]. In that model, it is assumed that the familiar Snell’s law and Fresnel’s equations are valid and that absorption through the oxide layer can be modelled as a path loss by using a simple exponential law (e.g. Beer’s law). This may be justified if the imaginary component of the complex refractive index is very small, but for many oxides in the visible and infrared wavelength regions this is not the case. In that scenario a more rigorous theory, involving complex wave vectors and inhomogenous plane waves (i.e. waves for which the normals to the planes of constant phase and constant amplitude do not coincide) has to be applied. Important work in this field has been done by Dupertuis [3] and Chang et.al. [4], who have discussed the implementation and the consequences of such an approach.

Experimental verification of the ray-tracing models is highly desired. Most of the previous work in this direction has been concentrated on specially fabricated surfaces with specially designed surface statistics, e.g. gold-coated diffuse reflectance standards as in the work of Tang & Buckius [5] or photoresist materials imprinted with light speckle patterns and subsequently coated with gold or platinum (see method as described by for instance Gray [6] or O’Donnell & Mendez [7]). In both of those cases the resulting surfaces are highly isotropic and Gaussian. Some experimental verification has also been done for more anisotropic and non-Gaussian surfaces such as silicon wafers (see for instance Lee [8]). More studies are needed for the kind of surfaces that are most frequent in laser material processing, e.g. engineering grade metal surfaces manufactured with different
surface finishes (such as cold rolled, hot rolled, polished, brushed or bright annealed surfaces).

The existing experimental data on the temperature dependence of light absorption is also sparse, much owing to the fact that the relevant experiments are cumbersome to set up. This data is, of course, of tremendous value to modellers in the field of laser metal processing since heating is the name of the game. Further research in this field is therefore required, both experimentally and theoretically.

Finally, to reach a more complete comprehension of the implications of light absorptance in laser material processing it is also highly desirable to try to abridge our knowledge of the properties of the separate absorption mechanisms into a more complete theoretical foundation. A theoretical basis that is simple enough for easy implementation in different processing models and simulations but complex enough to be realistic and accurate with our observations. It is of paramount interest to gain an understanding of the intricate details of how the absorptance changes dynamically during a processing application and how this will affect the manufacturing process and the quality of the final product. For this matter, more suitable and accurate experimental methods for in situ measurements also have to be developed.

6. Appendix

In this section some additional results are presented, which either didn’t fit into any of the papers listed or which weren’t seen to be mature enough for publication.

6.1 Further comparison between 2D and 3D ray-tracing models

In the final two papers, results of 2D and 3D ray-tracing models are presented from investigations of the influence of surface roughness on the laser absorptance. The 2D model, introduced in Paper 5, was found as a good starting point for these investigations for a number of reasons; it is computationally less intensive, it is easier to envision and comprehend phenomenologically and in light scattering theory it is the only model for which rigorous validation has been made to the more exact wave-theoretical calculations. It can also be a useful model, in its own right, in the study of quasi one-dimensional surfaces, such as the rippled, liquid
surfaces of cut or weld fronts or surfaces with a strong and well-defined direction of lay (e.g. surfaces brushed in a specific direction). However, a 2D ray-tracing model has its limitations since light rays are only confined to single planes and a more realistic 3D model, capable of treating broader types of surfaces, was therefore developed. The results of this model are presented in Paper 6. In comparing the results of the two models, both applied to Gaussian random rough surfaces, the phenomenology was found to be very similar in the two papers. However, some of the results were different and worth some further analysis (for the following the reader is strongly advised to read Papers 5 & 6 first).

Fig. 5 shows the average number of scattering events per incident ray for the four different angles analysed in the two ray-tracing models. For normal and 30° incidence, the overall behaviour is very similar but the level of scattering (and therefore absorption) is much higher in the 3D modelling for all RMS slopes above the multiple scattering threshold.

![Figure 5: Level of scattering in 2D and 3D ray-tracing models vs. RMS slope $\sigma/\tau$ (from simulations presented in Papers 5 & 6).](image)
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The main reason for the increased level of scattering in 3D ray-tracing (of 2D surfaces) as compared to the 2D ray-tracing case (of 1D surface profiles), is due to the different inclination angle distributions for Gaussian random rough surfaces in two and one dimensions, respectively. The inclination angle $\alpha$ of a surface point is the angle between the tangent plane at the surface point and the mean plane of the rough surface (e.g. the x-y plane), or equivalently the angle between the local surface normal and the z-axis (see Fig. 6). Due to Snell’s reflection law, larger inclination angles will force incident rays to be scattered more downwards (e.g. closer to the mean plane of the surface) which will promote multiple scattering.

Fig. 6 attempts to explain qualitatively why the inclination angle distributions will be different and illustrates the relationship between the surface normal vectors and the inclination angles of 2D and corresponding 1D surface models. Since a 1D surface profile results from an intersection of the 2D surface with a perpendicular plane, this will result in a projection of the corresponding 2D surface normal vectors into this intersecting plane, as it is seen in Fig. 6. This projection will result in a redistribution of the surface normal vectors towards smaller inclination angles, so that $\alpha_{1D} < \alpha_{2D}$.

![Figure 6: Relationship between normal vectors for 2D and 1D rough surfaces (the 1D surface profile is in this case located in the yz-plane) and the corresponding inclination angles $\alpha_{2D}$ and $\alpha_{1D}$. Since $\mathbf{N}_{1D}$ results from a projection of $\mathbf{N}_{2D}$ onto the yz-plane, $\alpha_{1D} < \alpha_{2D}$.](image-url)
More quantitatively, theorems from statistical theory can be used to explain this redistribution of normal vectors and inclination angles. The inclination angle $\alpha$ at a specific surface point is related to the slope at that particular point. More specifically $\tan(\alpha) = \sqrt{\xi_x^2 + \xi_y^2}$, where $\xi_x = \partial \xi / \partial x$ and $\xi_y = \partial \xi / \partial y$ are the slopes in x and y directions, respectively ($z = \xi(x,y)$ defines the surface). Due to isotropy, both $\xi_x$ and $\xi_y$ are Gaussian distributed with zero mean and the same standard deviation (or variance), e.g. $\xi_{x,y} \sim \text{N}(0, w^2)$, where $w = \sqrt{2} \sigma/\tau$ is the RMS slope. It is well known from the theory of statistics and probability, see for instance Meyer [1], that if the elements of a two-element vector are independent, Gaussian distributed with zero mean and the same variance, then the magnitude of the two-element vector will be Rayleigh distributed.

Thus, for a 2D Gaussian random rough surface, the tangent of the inclination angle, $\tan(\alpha) = \sqrt{\xi_x^2 + \xi_y^2}$, will be Rayleigh distributed. This is to be compared to the situation where the surface is one-dimensional (and $\xi_y = 0$) and in which case the distribution of $\tan(\alpha) = \xi_x$ will be reduced to a simple Gaussian. As Fig. 7 shows, these distributions (here normalized so the integral sum is equal to one, e.g. equal number of surface points) are substantially different for a specific RMS slope $\sigma/\tau$ (here the solid lines represent 2D Gaussian random rough surfaces and the dashed lines represent 1D Gaussian random rough surfaces of the same RMS slope). In comparison to the Gaussian distribution, the Rayleigh distribution is skewed towards larger inclination angles and will therefore promote multiple scattering more. An example is shown in the figure for normal incidence, where the approximate threshold for multiple scattering at normal incidence is indicated by the dash-dotted vertical (e.g. at $\alpha = 45^\circ$) and where a significant difference in the number of surface points with inclination angles above this threshold can be noticed for $\sigma/\tau \geq 0.3$.

For $60^\circ$ and $80^\circ$ incidence, the higher level scattering is also seen for the larger RMS slopes (see Figures 5c and 5d) and for basically the same reasons as explained above. Since two-dimensional surfaces are sloped in two directions instead of one, the inclination angle distributions will be skewed right towards larger angles. Phenomenologically, at these angles of incidence, this will result in lateral (side) scattering which does not exist for corresponding one-dimensional
surfaces. For smaller RMS slopes, the scattering of rays with large and grazing angles of incidence is approximately two-dimensional (since very little lateral scattering is involved) and the two models therefore largely coincides up until roughly $\sigma/\tau \sim 0.2$ for $60^\circ$ incidence and $\sigma/\tau \sim 0.3$ for $80^\circ$ incidence. 

Figure 7: Normalized distributions of the tangent of the inclination angles, $\tan(\alpha)$, of 2D (solid) and 1D (dashed) Gaussian random rough surfaces of various RMS slopes $\sigma/\tau$. The dash-dotted vertical line indicates the approximate limit of multiple (double) scattering at normal incidence, e.g. $\alpha = 45^\circ$.

6.2 AFM characterization and ray-tracing of real engineering grade metal surfaces

In the ray-tracing models presented in Papers 5 & 6, simulations and analysis are made for isotropic random rough surfaces, computer generated with Gaussian surface statistics (one- and two-dimensional, respectively) and with only one roughness scale (e.g. one unique correlation length). These surfaces are only approximate models of the real engineering grade metal surfaces found in laser material processing applications, where surfaces can both exhibit non-isotropic and non-Gaussian behaviour and where several roughness scales can be
superimposed on each other. A batch of four real engineering grade metal surfaces (taken from the larger set which was investigated experimentally in Papers 3 & 4) was selected for surface characterization and ray-tracing analysis and which are listed in Table 1.

<table>
<thead>
<tr>
<th>Label</th>
<th>Metal</th>
<th>Surface finish</th>
</tr>
</thead>
<tbody>
<tr>
<td>Surf1</td>
<td>Stainless steel 430</td>
<td>Bright annealed</td>
</tr>
<tr>
<td>Surf2</td>
<td>Stainless steel 304</td>
<td>Hot rolled</td>
</tr>
<tr>
<td>Surf3</td>
<td>Stainless steel 3Cr12</td>
<td>Hot rolled</td>
</tr>
<tr>
<td>Surf4</td>
<td>Zinc-coated Mild steel</td>
<td>Zintec</td>
</tr>
</tbody>
</table>

The surfaces were characterized using Atomic Force Microscopy (AFM) which provided surface data as well as suitable images for ray-tracing analysis. The AFM instrument, which was located at SCA R&D centre in Sundsvall, is a Veeco diDimension™ 3100 model, with sub-nm height resolution and a lateral resolution in the order of 20 nm (being limited mainly by the size of the probe tip and being slightly dependent upon selected scan size). The instrument was operated in tapping mode using a cantilever frequency of ~ 300 kHz (which means that topography is mapped by lightly tapping the surface using an oscillating probe tip).

Three images were taken for each separate surface (see Figures 8 & 9) with a scan size of 10 μm, which was found to be the largest scan size that can be used while retaining the resolution limit of the instrument. The surface data was then analysed in MATLAB. Height distribution functions (HDF) of each surface were calculated by adding the surface data of the three corresponding images (see Figure 10). As Figure 10 shows, Surf1 (the hot rolled stainless steel 430 surface) has near-Gaussian surface statistics, while the other three show more peaked HDFs. RMS heights, correlation lengths and RMS slopes were also calculated for each separate image, the results of which are shown in Table 2.
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Figure 8: AFM images of two surfaces; a bright annealed stainless steel (430) surface on the left and a hot rolled stainless steel (304) surface on the right.

Figure 9: AFM images of two surfaces; a hot rolled stainless steel (3Cr12) surface on the left and a zinc-coated (Zintec) mild steel surface on the right.
The AFM images were inserted into a 3D ray-tracing simulation (3D-RT) where laser light were assumed to be incident normally on the mean surface plane and the average number of scattering events per incident ray was calculated. Figure 11 shows the results of the surface data analysis and the ray-tracing simulations.

Based upon these results only, in the ray-tracing approximation Surf2 (the hot rolled stainless steel 304 surface) will be the only surface that will show any significant roughness-induced absorption since it is the only surface where the AFM images display topography that will promote multiple scattering. Surf1 (the
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bright annealed stainless steel 430 surface) is the surface with the least amount of multiple scattering in the batch and will, in the ray-tracing approximation, show an extremely small deviation from a smooth surface absorptance value. However, it is difficult to draw any conclusions based upon such few images and a thorough investigation should involve more images taken at several different scan sizes as larger roughness scales may well be hidden at the particular scan size studied here.

<table>
<thead>
<tr>
<th>Surfaces</th>
<th>Label</th>
<th>Image</th>
<th>RMS height</th>
<th>Corr. lengths</th>
<th>RMS slopes</th>
<th>Scat. events</th>
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<td></td>
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<td>0.91</td>
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<td>0.0367</td>
<td>0.675</td>
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<td>0.054</td>
<td>0.036</td>
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<tr>
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<td></td>
<td>0.0229</td>
<td>0.773</td>
<td>2.06</td>
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<td>0.317</td>
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<td></td>
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<td>0.020</td>
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</table>

References

Introduction


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Mathematical Modelling of Laser Absorption Mechanisms in Metals: A Review

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Abstract

In Laser Material Processing, an understanding of the fundamental absorption mechanisms plays a vital role in determining the optimum processing parameters and conditions. To this end, a combination of experimental as well as of theoretical work is required. In this paper, results of some of the most important mathematical models of laser-metal interactions are reviewed, including models for absorptance dependence on wavelength, polarization, angle of incidence, workpiece temperature, surface roughness, defects, impurities and oxides.

1 Introduction

The development of laser material processing requires extensive empirical research and the development of accurate, robust mathematical models. Many of these models have concentrated on various aspects of cutting [1, 2, 3], welding [4, 5, 6, 7] and drilling [8, 9]. All models of different laser machining processes involve the understanding and modelling of the fundamental physical and chemical laser absorption mechanisms, i.e. the mechanisms that rule the photon absorption phenomena. Photon-matter interactions are complex, but it is evident [10] that the mechanisms determining energy coupling into the workpiece, represent a key issue for the understanding of laser material processing and for improving the efficiency and reliability of this technology. "The Investigation of the dynamics of absorptivity variation during the process of laser irradiation is one topic of paramount scientific and practical interest“ to cite Prokhorov et. al. [11].
2 Laser absorption in metals

Light impinging on a material surface can be reflected, transmitted or absorbed. In reality, all three occur to some degree. In order for laser machining to be practical, the laser light must be absorbed by the material.

To yield an efficient process, it is necessary to couple as much of the incident intensity to the workpiece as possible. This coupling efficiency is described by the sample absorptivity $A$ (in some parts of literature this is also referred to as absorbance, absorption coefficient or just absorption). The absorptivity is defined as the ratio between the absorbed energy and the incident energy. Absorptivity changes during the heating process and is a function of the sample’s optical properties as well as the properties of the electromagnetic wave.

The beam and material properties of importance in this respect are [12]:

**Laser Beam:**

- Intensity
- Wavelength ($\lambda$),
- Angle of Incidence ($\alpha$)
- Polarization, $p$ or $s$ (parallel or perpendicular to the plane of incidence)

**Material:**

- Composition (eg. pure metals, alloys, polymers, ceramics, composites, etc.)
- Temperature ($T$)
- Surface roughness
- Surface and bulk defects and impurities (eg. dust particles, abrasives, cracks, pores, oxides, etc.)
The aim of this paper is to review some of the different methods that have been employed describing the influence of the above properties on absorption. Let us begin this task with a brief overview of the basics of light-matter interaction and the terms associated with it. There are two ways of describing this interaction. One is by treating light as a wave and the other considers light particles (photons). In most of the following treatment the first approach is used.

### 2.1 Basic electromagnetic theory and optical constants

Electromagnetic waves propagating through material can be described by Maxwell’s equations [13]. A solution to the wave equation for the electric field strength $E(z,t)$ in the case of a plane wave propagating along the z-axis can be written as

$$E(z,t) = E_0 e^{-\omega/c} e^{i(\omega/c)nz} e^{-i\omega t},$$

where $E_0$ is the amplitude of the field strength, $n$ the index of refraction, $k$ the extinction coefficient, $\omega$ the angular frequency of the wave and $c$ is the light velocity in the medium. The first exponential on the right hand side describes an attenuation (damping) of the wave, whereas the last two represent the characteristics of "free" propagation. As the intensity of an electromagnetic wave is proportional to the square of the amplitude, the intensity will decrease over distance when the wave is passing through an absorbing medium (see fig. 1). According to Beer’s law, for homogeneous media $^1$:

$$I(z) = I_0 e^{-\alpha z},$$

where $\alpha = \frac{4\pi nk}{\lambda_0}$ is the absorption coefficient and $\lambda_0$ is the vacuum wavelength. The reciprocal of $\alpha$ is called the absorption length, $l_\alpha$, and is the distance after which the intensity is reduced by a factor of $1/e$.

The optical constants $n$ and $k$ can be calculated from the complex dielectric permittivity

$^1$In heterogeneous media $n$ and $k$ depend on position and may in certain crystals even depend on direction of propagation. In that case $\alpha$ must be replaced by $\alpha(z)$ in Eq. (2) and integrated along the propagation path.
Figure 1: Absorption of electromagnetic radiation where the intensity of the wave is exponentially absorbed upon penetration. Figure taken from the Handbook of the EuroLaser Academy [10].

\[ \epsilon = \epsilon_1 - i\epsilon_2, \]  

using the following equations

\[ n^2 = (\epsilon_1 + \sqrt{\epsilon_1^2 + \epsilon_2^2})/2, \]

\[ k^2 = (-\epsilon_1 + \sqrt{\epsilon_1^2 + \epsilon_2^2})/2. \]  

The behavior of the optical constants, \( n \) and \( k \), with respect to light and material parameters such as wavelength and temperature is clearly of great interest and has been for a long time. Various methods have therefore been developed to experimentally determine these properties and extensive databases exist [14, 15], although these are mostly for pure materials at room temperature with as clean and smooth surface conditions as possible.
2.2 Fresnel absorption

From Fresnel’s formulas [13] we can derive the following two fundamental relations, which show the absorptivity dependence on polarization and angle of incidence:

\[
A_p = \frac{4n \cos \theta}{(n^2 + k^2) \cos^2 \theta + 2n \cos \theta + 1},
\]
\[
A_s = \frac{4n \cos \theta}{n^2 + k^2 + 2n \cos \theta + \cos^2 \theta}.
\]

These equations are valid when \(n^2 + k^2 \gg 1\), which is the case for metals at a wavelength \(\lambda \geq 0.5 \mu m\) (see fig. 2 for an example of Iron at 0.5\(\mu m\).) For other cases the full, exact expressions must be used, see for instance [13]. In Eqs. (5) the suffixes \(p\) and \(s\) denote linearly polarized radiation parallel and perpendicular to the plane of incidence (defined as the plane containing the direction of beam propagation and a line perpendicular to the surface), respectively. \(\theta\) is the inclination angle measured against the workpiece surface’s normal. In practice circular polarization is often used. The absorption is in this case the arithmetic mean value of the \(p\) and \(s\) components, ie. \(A_c = \frac{1}{2}(A_p + A_s)\). For normal incidence (\(\theta = 0\)), Eqs. (5) simplify to

\[
A_{p,s}(\theta = 0) = \frac{4n}{n^2 + k^2},
\]

where the absorptivity is now polarization independent.

2.3 Absorption in real metals

In discussing absorption in metals and its dependency on the properties listed in the beginning of this chapter, it is quite useful and instructive to separate the different contributions to the absorptivity \(A\) [11]. Following the treatment of Prokhorov et al. we can thus write

\[
A = A_{int} + A_{ext} = A_D + A_A + A_{IB} + A_r + A_{ox} + A_{id},
\]

where \(A_{int} = A_D + A_A + A_{IB}\) is the absorptivity determined by the intrinsic (bulk) properties of the metal and consists of terms from the normal (\(D\) as in
Figure 2: Absorption as a function of polarization and angle of incidence for Iron (Fe) at 0.5μm. Figure taken from the Handbook of the EuroLaser Academy [10]. Note the maximum occurring in $A_p$ for the angle $\theta_B$, the Brewster angle.

Drude) and anomalous skin effect ($A$) as well as from interband transitions ($IB$). $A_{ext} = A_r + A_{ox} + A_{id}$ is the function determined by the external (surface) conditions and contains terms from surface roughness ($r$), oxides ($ox$) and impurities and defects ($id$). All the separate terms will be explained and dealt with in the following sections.

### 2.4 Drude’s model - Intraband absorption

The absorptivity of metals shows a general tendency to increase when the frequency of the incident radiation is increased from the infrared to the ultraviolet spectral range (see fig. 3). An early attempt to describe the frequency/wavelength dependence on absorptivity was carried out in 1903 by Hagen and Rubens [16], who obtained
where $\sigma_0$ is the electrical dc conductivity. This simple model was only applicable at normal incidence ($\theta = 0$) and turned out to be an acceptable approximation only for longer wavelengths (see fig. 4). At short wavelengths deviations from experimental results were severe, although it could be seen that its range of validity was shifted towards shorter wavelengths for decreasing conductivity.

Modelling of optical constants from the elementary electron theory of metals is based on the work of Paul Drude [17] in the early 1900’s. Drude’s model builds on the assumption that the electron dynamics can be described as free electrons being accelerated by the electric field and being damped by collisions with phonons, other electrons and with lattice imperfections. In connection with the collisions a relaxation time $\tau$ and a relaxation wavelength $\lambda$ can be defined. This is called

\[ A_{HR} \simeq \sqrt{\frac{2\omega}{\pi \sigma_0}}, \quad (8) \]
intraband absorption as only electrons from the conduction band are involved.

In the study of the optical properties of free carriers in metals, we can roughly distinguish three different frequency regions [11, 18]

- \( \omega \tau \ll 1 \ll \omega_p \tau \) (non-relaxation region, far IR):

\[
n(\omega) \approx k(\omega) \approx \sqrt{\frac{2\pi \sigma_0}{\omega}} \Rightarrow A_D(\theta = 0) \approx \sqrt{\frac{2\omega}{\pi \sigma_0}},
\]

which is the same result as that reached by Hagen and Rubens.
In the low frequency limit, i.e., in the non-relaxation region, the Hagen-Rubens equation is seen to be the asymptote of the Drude theory and the correspondence between theory and experiments is rather good. The optical constants of various metals (Ag, Al, Au, Cu, Pb, W) in the infrared agree well to experimental values, see [19], but this is not true in the visible and UV regions.

In the relaxation region, one would expect $A_D \to 0$ as the temperature $T \to 0$ since the relaxation time $\tau$ increases when temperature is decreased [11]. However experiments performed in the 1930’s indicated that not even the transition to superconductivity was accompanied by a substantial decrease in $A_D$. This behavior was called the anomalous skin effect and occurs whenever the mean free path of the electrons becomes comparable with the radiation wavelength and the penetration depth. It is not incorporated in the Drude model which is referred to as the normal skin effect and must therefore be added if one wants an expression for the total absorptivity. In the case of a crystalline lattice with a spherical Fermi surface, the contribution is given by [11]

$$A_A = \frac{3}{4} \frac{<v_F>}{c} \frac{1}{1 + \frac{1}{\omega^2 \tau}} (1 - f) + \frac{\omega_p^2}{2 \omega^2} \frac{<v_F>}{c^3} f,$$

where $<v_F>$ is the mean electron velocity and $0 \leq f \leq 1$ is a parameter which depends on the surface roughness. For an average roughness 100-200 Å one can safely assume $f \simeq 0$ and for a roughness not exceeding about 30 Å, $f \simeq 1$ [11].
The above model assumes only one kind of conduction electrons. In 1900 Drude first actually proposed a formula based on the postulated existence of two kinds of free charge carriers. Later he abandoned this idea, since it seemed inconsistent with the electron theory being developed at that time. In 1955 Roberts [20] went back to Drude’s original idea, which he interpreted in view of modern solid state physics as conduction electrons originating from the s and d bands, respectively. For the complex dielectric permittivity he wrote

$$\epsilon = (n - ik)^2 = \epsilon_\infty - i\frac{\sigma_\infty \lambda}{2\pi \epsilon \epsilon_0} - \frac{\lambda^2}{2\pi \epsilon \epsilon_0} \left( \frac{\sigma_1}{\lambda_1 - i\lambda} + \frac{\sigma_2}{\lambda_2 - i\lambda} \right).$$

(13)

Here $\lambda_i$ and $\sigma_i$ are relaxation wavelengths and electrical conductivities for the two sets of carriers, respectively. $\epsilon_\infty$ accounts for the contribution of bound electrons at long wavelengths compared to their resonance wavelengths, $\sigma_\infty$ is a conductivity introduced as a correction factor for surface effects and $\lambda$ is the wavelength of the light. In this manner, good coincidence with room temperature data was obtained at wavelengths above the so called X-point, which is a unique wavelength that is special for each metal and is explained in section 2.10 (see fig. 5).

Another extension of Drude’s theory worth mentioning at this point was performed by Wieting and Schriempf, who used it to calculate the absorption of alloys [21]. Good agreement was found for wavelengths above 10 $\mu$m, but large deviations were observed for shorter wavelengths.

### 2.5 Interband absorption

So far we have only treated intraband absorption. For photon energies high enough, *interband* absorption also takes place, i.e. transitions between valence and conduction bands. For noble metals this is the case in the visible and UV part of the spectrum and for transition metals it begins at lower energies [22].

To remedy the shortcomings of the Drude theory at shorter wavelengths, Roberts therefore wrote a second paper in which he generalized his equation by taking interband absorption into account [23].
\[ \epsilon = 1 - \frac{\lambda^2}{2\pi c \epsilon_0} \sum_n \frac{\sigma_n}{\lambda - \lambda_n} + \sum_m \frac{K_{0m}}{\lambda^2 - \lambda_{sm}^2 + i\delta_m \lambda_{sm} \lambda}, \]  

where all parameters except \( \lambda, c \) and \( \epsilon_0 \) are considered arbitrary and which are adjusted independently to characterize any given metal. Since terms containing \( \sigma_n \) and \( \lambda_{rn} \) all contribute to the dc conductivity, they may be attributed to conduction electrons or free electrons. In the same way, those terms containing \( K_{0m} \) do not contribute to the dc conductivity and are therefore associated with bound electrons.

The above equation was used by Dausinger [24] who fitted it to room temperature data of iron by Weaver et al. [22]. Fairly good agreement could be achieved by taking 4 interband terms into account (see fig. 6).
Figure 6: Absorption of iron at perpendicular incidence for a few temperatures. Experimental data are taken from literature. Modelling by Dausinger including both intraband and interband transitions. Figure taken from the Handbook of the EuroLaser Academy [10].

2.6 External absorptivity

The appreciable scatter in absorption data in the literature and the often large disagreement between data and the theory, mainly stems from the fact that the quality of the surfaces of the irradiated samples is far from ideal. Consequently the total absorptivity cannot be described only by the bulk properties of the metals and the formula $A = A_D + A_A + A_{IB}$, but must also, and often to a large extent, include contributions from the surface conditions. The main contributions of interest here are the surface roughness, defects and impurities and oxide layers.

The optical behavior becomes almost completely dominated by surface effects when the dimension of the metal is shrunk to values of the order of the absorption length, such as in extremely thin evaporated films or in aggregate structures consisting of small insulated metallic particles [25].
2.7 Surface roughness

The influence of surface roughness on absorptivity can manifest itself in different ways [11]. First there is the increase of absorption from surface areas where the radiation angle of incidence $\theta \neq 0$. There is also a contribution from grooves and cracks which favor waveguided propagation and were there may be multiple reflections down the surface irregularities.

Fig. 7 shows the change in reflectivity with roughness for copper and aluminium, after the metal surface was roughened by sandpaper with various grits (experimental results taken from Xie and Kar [26]).

![Figure 7: Effect of surface roughness on CO$_2$ laser reflectivity for copper and aluminium. Experiment and figure taken from Xie and Kar [26], where stationary and moving samples of Cu and Al were roughened with sandpapers of various grits.](image)

In studies of the roughness dependence of absorption two essential regimes have been considered. The first one is where the roughness, characterized by the roughness parameter $\delta$, is very small compared to the laser wavelength, i.e. $\delta/\lambda \ll 1$. In this regime first-order perturbation with respect to the intrinsic absorptivity $A_{\text{int}}$ can
be used. A simple estimation of absorptivity for a rough surface, $A_r$, as compared to the absorptivity of a very smooth surface of the same material, $A \approx A_{int}$, can be obtained with the expression [27]

$$A_r \approx 1 - (1 - A_{int})e^{-(4\pi\delta/\lambda)^2}.$$  \hspace{1cm} (15)

This equation was established on the assumption of a gaussian distribution of the roughness height on the surface. The theory of Elson and Sung [28, 11] can be used for other, more general, roughness distribution functions.

For large scale roughness $\delta/\lambda \gg 1$ other models have been used, see for instance that of Ang, Lau, Gilgenbach and Spindler [29], who modelled roughness as rectangular wells with height and width described by gaussian-related distribution functions. Calculations showed that an absorptivity increase of an order of magnitude may be possible for a UV laser bound on an aluminium target (as compared to a flat Al surface).

Not all roughness geometries lead to enhanced absorption though. This was observed in an experimental and theoretical analysis performed by Matsuyama et al. [30]. They showed that the peak absorptivity decreased using an asymmetric triangulated surface model.

### 2.8 Oxide layers

The surfaces of conventional metals, when kept in air, are most often covered by an oxide layer that sometimes shows a multilayer structure. Oxide layers can cause an increase in sample absorptivity by as much as an order of magnitude. [31].

The thickness $x$ and the structure of the oxide layer are the main properties that determine this absorptivity contribution, $A_{ox}$. These properties may well change during the laser processing because of their temperature dependence (see for instance [11] for a few different models of oxidation kinetics due to Cabrerra et al.). The effect is also highly influenced by the wavelength used, which has been confirmed by experiments.
Figure 8: Absorption can be significantly enhanced due to oxide layers on metals. This effect is mainly due to the interference phenomena occurring inside the layer, where the beam is partly absorbed, partly reflected at the metal and partly reflected back again at the oxide-atmosphere boundary.

To obtain a correct dependence $A_{ox}(x)$ it is necessary to consider the interference phenomena occurring within the metal-oxide layer (see fig. 8). In the simple case of a layer of a single type of oxide that grows with uniform thickness on the surface of the metal sample, the following formula has been presented [32]

$$A_{ox}(x) = \frac{r_{12}e^{-2i\varphi(x)} + r_{23}}{e^{-2i\varphi(x)} + r_{12}r_{23}},$$

(16)

where

$$\varphi(x) = \frac{\omega}{c_0} x \sqrt{\epsilon_{ox}}, \quad r_{12} = \frac{1 - \sqrt{\epsilon_{ox}}}{1 + \sqrt{\epsilon_{ox}}},$$

$$r_{23} = \frac{r_{12} - r_{13}}{r_{12}r_{13} - 1}, \quad 1 - |r_{13}|^2 = A_M.$$  

(17)

Here $r_{12}$ and $r_{13}$ are the amplitude coefficients of radiation reflection on oxide and metal, $\epsilon_{ox} = n_{ox} + ik_{ox}$ is the complex dielectric permittivity of the oxide and $A_M$ is the metal absorptivity. In fig. 9 the absorptivity as a function of both oxide layer thickness and angle of incidence can be seen for a circular polarized beam in a typical metal. In the work of Arzuov et al. the following result was derived using
Figure 9: Absorptivity as function of oxide layer thickness (here normalized with respect to the wavelength) and angle of incidence for a typical metal. Figure taken from Franke [33]. Observe the original Fresnel curve for no oxide layer.

the approximations that (1): the oxide absorption is much larger than the metal absorption, ie. \(1 - |r_{12}| \gg A_M\), (2): the oxide layer \(x \ll 1\) m and (3): the refraction index of the oxide \(n_{ox} > 1\)

\[
A_{ox}(x) = \frac{n_{ox}^2 A_M + 2 k_{ox} (\beta x - \sin \beta x)}{n_{ox}^2 + (1 - n_{ox}^2) \sin^2(\beta x/2)},
\]

(18)

where \(\beta = 2 \omega n_{ox}/c_0\).

Simpler formulas have also been used in different papers, eg. [32]

\[
A(x) = A_M + 2 \alpha x,
\]

(19)

which was derived from Eq. (18) under the conditions \(x \gg \lambda/(4 \pi n_{ox})\), \(n_{ox} - 1 \ll 1\) and \(2 \alpha x \ll 1\). Another one is

\[
A(x) = A_M + bx^2,
\]

(20)

where \(b = 4 \pi^2 (n_{ox}^2 - 1) A_M / \lambda^2\) and was seen suitable for thin oxide films [34].
2.9 Other external absorptivity contributions

Impurities scattered on and in the metal surface can increase the absorption of radiation. One example is dust particles of different size and shape, which may lead to centers of sharply enhanced local absorptivity. Another is abrasive particles left behind by polishing, whose contribution to the absorption is largely determined by the species of embedded particles and their absorptivity properties at the laser wavelength in use. Therefore, as a general rule to reduce/eliminate the influence of absorption by the inclusion of particles, the polishing materials should be transparent at the appropriate wavelength [11].

Apart from impurities we may also have additional absorption from defects occurring in the bulk of the material, such as pores, cracks and grooves. Their role in increasing locally absorptivity has been shown in the following references [35, 36, 37].

Also, flaws in the metal sample itself may cause locally enhanced absorptivity, such as for instance flakes. Flakes are examples of parts of the metal that are thermally insulated and therefore are overheated compared to the surrounding material during the laser processing. A formula for how to calculate the overheating is suggested in [11].

These effects make up the final absorptivity contribution, $A_{id}$ of Eq. (7). Other external effects than those mentioned here might also contribute, such as effects from the surrounding plasma cloud, diffuse electron scattering (particularly in thin films) and plasmon excitations $^4$, etc.

As we have covered the different terms in (7) we now turn our attention to the variation of absorptivity with temperature, which is an important factor in understanding the dynamic behavior during a laser process.

$^4$Plasmons are collective oscillations of conduction electrons in metals and semiconductors. Plasmon excitation occurs by p-polarized light at an incident angle slightly above critical angle for total internal reflection.
2.10 Variation of absorptivity with temperature

A knowledge of the variation of absorptivity with temperature is of great practical importance in calculations. The temperature dependence of the optical properties of metals has been studied theoretically in many papers, e.g. [37, 38, 39].

In the Drude model regime, i.e. when the interband contributions are negligible, it can be shown that absorptivity increases with temperature [11]. The reason is essentially that both the dc conductivity and the relaxation time decreases with temperature and therefore \( \frac{dA_D}{dT} > 0 \) according to Eqs. (9) and (10).

Let us now examine the dependence \( A(T) \) in two different temperature ranges: from room temperature up to the melting point and for temperatures in the liquid phase.

2.10.1 Solid-phase metals

Most of the available data on temperature dependence on absorptivity, obtained experimentally or theoretically (see fig. 10 for examples of experimental curves of a heated aluminium sample), may generally be described by a simple linear equation

\[
A(T) = A_0 + A_1 T.
\]  

(21)

This relation was inferred by fitting data available in the infrared range with a general polynomial. For high-purity metals in spectral ranges where the interband influence was small, the coefficients of second and higher order terms were negligible and formula (21) worked well. However for real metal surfaces the fit might be less obvious because of surface roughness and defects which affect the absorptivity.

Seban [40] applied Roberts’ two-electron intraband approach, Eq. (13), to an elevated temperature (850 °C) by making the damping frequency of one electron type inversely proportional to the electrical conductivity and keeping that of the other type constant. Reasonable agreement with experimental results was obtained, but only above the X-point (see fig. 5).

When interband transitions have to be considered the temperature dependence changes. We have already mentioned the X-point as a unique wavelength for each metal un-
Temperature dependence of CO\textsubscript{2} laser absorptivity for an aluminium sample. Figure taken from Prokhorov [11]. Curves 1, 2 and 3 corresponds to the same sample heated subsequently, where $A(T_0)$ is seen to decrease with every step due to the surface cleaning effect.

By making the following assumptions to Eq. (13) for higher temperatures

$$\sigma_1(T) = \sigma_0(T) - \sigma_2,$$

$$\delta_m(T) = \frac{\sigma_1(T \simeq 300K)}{\sigma_1(T)} \delta_m(T \simeq 300K),$$

and assuming $\sigma_2$, $\lambda_2$ and the quotient $\sigma_1/\lambda_1$ all are temperature independent, the X-point behavior was satisfactorily described by Dausinger [24] (see fig. 6).

Interband modelling with the temperature dependence assumptions of Eq. (22) has also been used for low alloys steels [42] with very good experimental agreement.
This is also true for Aluminium, where one Drude term and two interband terms were used [10].

### 2.10.2 Liquid-phase metal

At the melting temperature, when the metal goes through a solid-liquid phase change, the number of conduction electrons increases simultaneously with metal density and DC resistivity. This results in a stepwise increase of absorptivity, the amplification of which may well be in the order of 150-200%. The absorptivity then continues to increase and the same linear or polynomial approximations as in the solid phase can be used [11] (see fig. 11 for some computed curves).

![Figure 11: Some computed curves showing the increase of the intrinsic absorptivity (at the CO$_2$ wavelength) with temperature of some pure metals before and after melting. Figure taken from Prokhorov [11].](image)

It should also be mentioned that in several publications [43, 44] there has been evidence of an anomalously high as well as low absorptivity at the melting point. This cannot be accounted for by the previously reviewed theory. To interpret these effects other processes involved in intense laser irradiation of metallic surfaces have to be considered, such as dynamic surface thermal deformations, resonant surface
periodical structures and heat transfer from plasma to the sample [11].

3 Summary

A thorough knowledge and understanding of the fundamental absorption mechanisms underlying all material processing with lasers is clearly of great significance for the future development of this technology. In this paper the results of some of the most important mathematical models explaining the dependence of a metal’s absorptivity on different laser and material properties such as wavelength, polarization, angle of incidence, temperature, surface roughness, oxides, defects and impurities have been reviewed.

For the absorptivity dependence on polarization and angle of incidence Fresnel’s formulas [13] can successfully be employed.

To explain the dependence of light frequency on absorptivity both intraband and interband transitions must be taken into account. For the intraband contribution, where absorption takes place due to free electrons in the metal, the theory is well described by the simple Drude model [17] which regards metals as a classical gas of electrons executing a diffusive motion. The interband contribution, which is due to transitions between valence and conduction bands in the metal, is often modelled using Roberts’ theory [20, 23] and explains the resonant and oscillatory behaviour of absorptivity in the high frequency range.

Absorptivity as a function of temperature is an important consideration in practice and is usually modelled successfully as a linear function except around the melting point of the metal where there is a significant jump due to the sudden change of conduction electron density.

Many external properties of the material being processed influence the absorptivity, among which oxide layers and surface roughness are perhaps the most important and for which good theoretical models exist. The absorptivity depends on the roughness geometry and is in most cases enhanced because of multiple reflections down the surface irregularities. Models exist both for small and large roughness

parameter regimes [27, 28, 45, 29, 30]. The absorptivity is also enhanced in oxide layers due to interference effects and depends strongly on oxide layer thickness, structure and laser wavelength [32, 34]. Other surface properties such as defects and impurities and bulk properties such as pores, cracks, grooves and flakes that may lead to local variation of absorption have also been mentioned.

Although a lot of experimental results seem to exist for pure metals at room temperature, more data is needed for model evaluation at elevated temperatures and for non-smooth, non-clean surface conditions.

As a last remark I would like to point out that this survey has not considered the relatively new ultra-short lasers and their physical effects in metal processing. Ultra-short laser pulses with their ultra-high intensity levels involves non-equilibrium physics and therefore the standard values of the metals’ optical properties cannot be used, since they have been experimentally determined and theoretically calculated using steady state assumptions [46].

4 Further reading

Many of the models and theories that have been brought forward in this paper have been presented quite briefly, mostly in terms of the main results. For those of you who are interested in more detail I refer to the references given at the end of this paper. I can also recommend a few books that treat this topic more generally.

In the literary material that has been surveyed and presented I found the book ”Laser Heating of Metals” by Prokhorov et al. [11] the most comprehensive and complete description of the subject, treating both intrinsic and external material properties.

Another good book of reference for this subject is the Handbook of the EuroLaser Academy and specifically its first chapter written by Hügel and Dausinger [10]. Although not as comprehensive as it treats mostly intrinsic absorption contributions, it gives a good historical overview of how the subject has been developed, has useful references to important advances that have been made and has a nice collection of

5Femtosecond lasers has pulse durations comparable to or shorter than electronic relaxation times.

illustrative and informative figures.

I would also like to mention "Laser-Beam Interactions with Materials - Physical Principles and Applications" by von Allmen and Blatter [47] as a very good introductory book.

Another good handbook for experimental material data seems to be that of Palik, "Handbook of Optical Constants" [14]. An alternative option to this may be the "CRC Handbook of Chemistry and Physics" by Weast [15].

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Paper 2: Laser Absorption Measurements in Opaque Solids

Laser Absorption Measurements in Opaque Solids

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Paper 2: Laser Absorption Measurements in Opaque Solids
Abstract

In Laser Material Processing, an understanding of the fundamental absorption mechanisms plays a vital role in determining the optimum processing parameters and conditions. The absorptance, which characterizes how much a material absorbs, can alter during a processing application, as a result of melting, boiling, structural changes of the surface, oxidation, plasma formation, etc. To gain knowledge of these complex processes, a combination of experimental as well as of theoretical work is required. In this paper, an overview of the most common experimental methods and techniques to measure laser absorption in opaque solids, such as metals, will be presented.

Keywords: absorptance, reflectance, emittance, calorimetry, integrating sphere, integrating mirror

1 Introduction

The development of laser material processing requires extensive empirical research as well as the design of accurate, robust mathematical models. Many of these models have concentrated on various processing applications, such as cutting [1, 2, 3], welding [4, 5, 6, 7], drilling [8, 9] and so on. All models of different laser machining processes involve an understanding and modelling of the fundamental physical and chemical laser absorption mechanisms, ie. the mechanisms that rule the photon absorption phenomena. Photon-matter interactions are complex, but it is evident that the mechanisms determining energy coupling into the workpiece, represent a key issue for the understanding of laser material processing and for improving the efficiency and reliability of this technology. "The Investigation of the dynamics of absorptivity variation during the process of laser irradiation is one topic of paramount scientific and practical interest” to cite Prokhorov et. al. [10].
In a previous paper a number of different mathematical models of the various laser absorption mechanisms present in metals were reviewed [11]. These models dealt with the dependence of absorption on laser parameters such as wavelength, polarisation and angle of incidence as well as on material parameters such as composition, temperature and various surface properties, eg. roughness and oxide layers. Mathematical modelling is a powerful tool in the hands of the scientist or the engineer, making it possible to predict the behaviour of a complex physical system. But to reach this stage the models have to go from Mathematics into Physics, and this is only achieved through experimental verification.

2 Basic theory of radiative properties of solids

In this paper, a brief overview of the various experimental methods available for the measurement of laser absorption in opaque solids will be presented. Although special focus will be given to metals, many of the methods will apply to other types of materials, eg. semiconductors, isolators, polymers, ceramics, composites, wood, etc., as long as they are non-transmitting. The review is not to be seen as a complete survey of the whole field and we will specifically exclude measurements concentrated on the determination of optical constants (complex refractive indices, complex dielectric functions or complex conductivities), such as the methods of ellipsometry, polarimetry and Kramer-Kronig integrating which require pure, flat and oxide-free surfaces, conditions that are rarely achieved in engineering applications.

Before reviewing the different experimental techniques, we first need some basic knowledge of the theory of the radiative properties of solids. The reason for this is two-fold. First of all, it will let us get acquainted with the nomenclature commonly used in the field and secondly, it will also give us an insight into the different ways of getting the information that we seek.

2.1 Thermal radiation and emittance

All materials with a temperature above absolute zero have one property in common; they all continuously emit radiation by lowering atomic and molecular energy levels. This is known as thermal radiation. The emitted spectrum at a specific temperature varies from material to material and the emissive ability of a specific material is usually compared to that of a so called blackbody. The blackbody is a
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Theoretical construct having the properties of absorbing all radiation that strikes it (only a few real materials approach this limit, examples being carbon black, platinum black, gold black and some specially formulated black paints on absorbing substrates). By a variation of Kirchhoff’s law from basic thermodynamics, it can be seen that this same property of being a perfect absorber also implies that it’s a perfect emitter, no other material emitting as much radiation at a specific temperature (see Figure 1).

![Figure 1: Consider a blackbody inside a blackbody enclosure, both held at the same temperature. By definition, all radiation that strikes the blackbody from the surface element of the enclosure must be absorbed. To maintain thermal equilibrium and isotropic radiation throughout the enclosure, the radiation emitted back into the incident direction must equal that received.](image)

The spectral distribution of the blackbody radiation is given by the famous Plank’s law and the wavelength of maximum intensity is well described by Wien’s displacement law. These laws tell us that the hotter the material is the shorter the emitted wavelengths from it. Let us now define a nondimensional property called the emittance, $\epsilon$, which is given by

$$\epsilon = \frac{\text{Power emitted from a surface}}{\text{Power emitted by a black body surface at the same temperature}}.$$  

(1)
Thus, emittance is a radiative property that can vary between 0 and 1 and for a black body we have $\epsilon = 1$. The emittance of a body can be a function of temperature, wavelength, direction and various surface properties.

### 2.2 Reflectance, absorptance and transmittance

We now move on by contemplating the situation depicted in Figure 2, where a laser beam is impinging on a piece of material. We ask ourselves what happens when light strikes the surface of the material. Well, initially it can only go into two directions, it can either be reflected away from the front surface or it can penetrate and start propagating through the interior of the material. As the light propagates it may be scattered and absorbed and depending on whether the material is a weak or strong absorber internally and on the material thickness, some of the light may also make it through to the opposite side and be transmitted (some of the light may also be reflected from the back surface, but this is not considered in the following discussion). These are the processes of reflection, absorption and transmission.

![Diagram showing light reflection, absorption, and transmission](image)

**Figure 2:** When a laser beam hits a material sample of finite thickness, some of the light may be reflected, some may be absorbed and the rest may be transmitted.
Please note here, that it is customary to distinguish between internal (or bulk) absorption and total absorption, including surface effects. A shiny material such as a metal is a very strong internal absorber but it also reflects light very well, so that the total amount absorbed is still quite small. For a material to be a strong total absorber it thus needs to have low surface reflection and high internal absorption (examples of that are certain metal powders, like the already mentioned platinum or gold black).

The quantities that are most often used describing the three processes in Figure 2 are called reflectance, absorptance and transmittance. The definitions of these quantities are given as simple ratios, as follows. The reflectance, \( R \), is given by

\[
R = \frac{\text{Reflected laser power from the sample surface}}{\text{Total laser power incident on the surface}},
\]

while absorptance, \( A \), is

\[
A = \frac{\text{Absorbed laser power in the sample}}{\text{Total laser power incident on the surface}},
\]

and transmittance, \( T \), is defined by

\[
T = \frac{\text{Transmitted laser power through the sample}}{\text{Total laser power incident on the surface}}.
\]

These quotients give dimensionless numbers that all have values between 0 and 1 and are often given as percentages. Since these are the only processes available, the following relationship, which is simply a conservation of energy statement, must always be fulfilled

\[
R + A + T = 1.
\]

2.3 Opaque solids

At this point, we pause to make an important distinction, that between materials that do not transmit any radiation (\( T = 0 \)) and materials that do (\( T \neq 0 \)). These are termed opaque and semi-transparent, respectively. In this paper we will limit

\[1\] Also known in literature with an -ivity ending. NIST has recommended the use of the ending -ivity for optical properties describing pure and smooth surfaces and the ending -ance for rough and contaminated surfaces (ie. real surfaces).
ourselves to opaque materials, and hardly speak of transmittance again. As already mentioned, whether a material transmits radiation or not, depends on both the material type and the material thickness at the wavelength used. Metals are opaque at visible and IR wavelengths, unless we are dealing with extremely thin films. This is because metals are characterized by their large concentrations of free electrons, which is the property that gives them their highly reflective nature and shiny appearance. Metals have typical skin depths (defined as the depth from the surface where the intensity has exponentially decreased to \(1/e^2 \approx 0.13\) of its original value) of only a few nanometers at visible wavelengths (ie. a few atomic diameters). This is also the case for most non-metals where absorption usually takes place within a few microns [14], although obvious exceptions exist like quartz and glass.

### 2.4 Spectral vs. total properties

The definitions of the optical properties of emittance, reflectance and absorptance in equations (1), (2) and (3) were seen to be simple enough. However, because of the spectral and directional dependencies of these quantities, it is customary to refine the definitions somewhat. The first distinction is that between spectral and total values, where spectral refers to the value at a specific wavelength and where total is the averaged, integrated value over all wavelengths. In notation we usually indicate a spectral property by adding a \(\lambda\) subscript to the quantity, as in \(E_\lambda\), and a total property by adding a \(\text{tot}\) subscript, as in \(A_{\text{tot}}\). The spectral and total values are then simply related according to

\[
A_{\text{tot}} = \int_0^\infty A_\lambda(\lambda)d\lambda,
\]

where absorptance was used as an example.

In this paper we are interested in experiments using a laser as the light source. The laser is a light producing tool with the special properties of high spectral purity and directionality. For this reason the following discussion will only involve spectral values of emittance, absorptance and reflectance.

### 2.5 Directional vs. hemispherical properties

The directional dependence of the values in equations (1), (2) and (3) forces us to separate them into directional and hemispherical values (see Figures 3 and 4), where
directional means the value to and/or from a specific direction and hemispherical means the integrated value over all directions (incoming or outgoing). We denote directional values by adding a prime superscript, as in $A_{\lambda}'$, and a hemispherical by adding a half circle, as in $A_{\lambda}^{\text{tot}}$. The hemispherical and directional absorptances are then related through an integration

$$A_{\lambda}^{\text{tot}} = \int A_{\lambda}'(\theta_i, \phi_i) \cos(\theta_i) d\Omega_i,$$

where $d\Omega_i$ is an elemental solid angle around the angle of incidence $\Delta(\theta_i, \phi_i)$.

For reflectance this is slightly more complicated since reflectance depends on two directions, both the incoming and outgoing ones. Most information is contained in the so called spectral, bidirectional reflectance function, denoted $R''_{\lambda}$ (also known in literature as the bidirectional reflectance distribution function, BRDF). This is a function of wavelength as well as of angles of incidence and reflection (see Figure 5). By averaging over wavelengths and/or directions one can then define total values as well as directional-hemispherical, hemispherical-directional and bihemispherical values (see Table 1). The spectral directional-hemispherical reflectance is, for example, denoted $R_{\lambda}^{\text{tot}}$ and is defined in terms of $R''_{\lambda}$ as
Figure 4: A pictorial description of a hemispherical radiative property, in this case the hemispherical emittance which is emitted into all directions of the hemisphere.

\[ R'_\lambda(\theta, \phi) = \int R''_\lambda(\theta, \phi, \theta_r, \phi_r) \cos(\theta_r) d\Omega_r, \]  

(8)

where \( d\Omega_r \) is an elemental solid angle around the angle of reflected radiation \( \Delta(\theta_r, \phi_r) \).

A similar procedure is of course possible for \( R'_\lambda \), but with an integration over angles of incidence instead.

In fact, according to Helmholtz Reciprocity Principle,

\[ R''_\lambda(\theta_i, \phi_i, \theta_r, \phi_r) = R''_\lambda(\theta_r, \phi_r, \theta_i, \phi_i), \]  

(9)

from which we can see that

\[ R'\lambda = R'_\lambda, \]  

(10)

which means that \( R'\lambda \), \( R'_\lambda \) and \( R'_\lambda \) are not totally independent quantities.
2.6 Specular and diffuse reflection

The reason why we needed to define these different directional definitions is because, although we might have a very well defined incoming direction from a laser source, the outgoing, reflected direction doesn’t need to be. If the surface is perfectly smooth the incoming parallel rays of the laser beam are all reflected in the same direction, as in Figure 6A. This is known as **specular** reflection. However, if the surface is non-smooth and irregular, the incoming parallel rays will be reflected in a number of different directions, which is seen in Figure 6B. This is termed **diffuse** reflection. Observe that Snell’s law of reflection (angle of incidence = angle of reflection) still holds for each ray in both cases. But since a laser beam hits a spot of perhaps small but finite area, the reflected radiation will be spread over a range of different directions.

Whether a surface is to be considered smooth or rough in a given situation is not only a question of surface topology but is also dependent on the wavelength of the incoming light as well as on the angle of incidence. The Rayleigh criterion, which is one of the simpler criterions for surface roughness, states that a surface

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Figure 5: A pictorial description of the bidirectional reflectance, depending on both incoming and outgoing (reflecting) directions.

Figure 6: A) Specular reflection. B) Diffuse reflection.
can be treated as smooth (specular) only under two conditions [19], either when \( h/\lambda \to 0 \) or when \( \theta \to \pi/2 \), where \( h \) is the height of the irregularities, \( \lambda \) is the wavelength and \( \theta \) is the angle of incidence. Absolutely perfectly smooth surfaces do not exist, not even in the case of high precision optical mirrors, so all attempts at measuring the total reflectance will necessarily also include a diffuse part, to lesser or greater extent.

### 2.7 Measurement options

The quantity that is the main focus of this paper is the spectral, directional absorptance, denoted \( A'_{\lambda} \). This is the quantity that measures a solid's ability to absorb an incoming laser beam of a specific wavelength and from a specific direction. But as we will see, \( A'_{\lambda} \) can also be measured by a couple of indirect methods.

The first indirect method is evident by considering Kirchhoff’s law. Kirchhoff’s law is a law relating the emitting and absorbing abilities of a body. Take another look at Figure 1. To maintain an energy balance (thermal equilibrium) between the element of the blackbody in the centre and the surface element of the black enclosure, the absorbed and emitted energies of both elements must be equal. Hence, Kirchhoff’s law states that

\[
\epsilon'_{\lambda} = A'_{\lambda},
\]

from which we can see that as an alternative to measuring the spectral directional absorptance we can measure the spectral directional emittance.

Equ.(11), only holds strictly true for each component of polarization of the light, separately [12]. For equ.(11) to be valid for all incident energy the incident radiation must have equal components of polarization. Furthermore, Kirchhoff’s law was proven for thermodynamic equilibrium in an isothermal enclosure and is therefore strictly true only when there is no net heat transfer to or from the surface. In actual applications this may not be the case and equ.(11) must therefore be seen as an approximation. There is much experimental evidence of the validity of this approximation, though, showing that in most applications materials are able to maintain themselves in local thermodynamic equilibrium in which the populations of energy states that take part in the absorption and emission processes are given, to a very close approximation, by their equilibrium distributions [12]. Thus, materials usually do not depend on the surrounding radiation field.
The other way to collect information about $A'_\lambda$ is seen by considering equ.(5), which states that the sum of reflectance, absorptance and transmittance must be unity. This means that we can derive the absorptance ($A$) if we have measured values for $R$ and $T$. For opaque materials, for which $T = 0$, it suffices to measure the reflectance only. To determine the directional absorptance $A'_{\lambda}$, either the full bidirectional, the directional-hemispherical or the hemispherical-directional reflectance must be known.

After this brief tour of some theory of the radiative properties of matter we move on to the next section, which is intended to give a short description of the various experimental means we have at our disposal for measuring these properties.

3 Experimental methods for measuring laser absorptance

The experimental methods for measuring laser absorptance can be divided into two main groups, calorimetric methods that measure the absorptance ($A$) directly and radiometric methods that measure absorptance indirectly via other radiative properties, eg. reflectance ($R$) and emittance ($\epsilon$).

3.1 Laser calorimetry

Laser calorimetry represents the only direct way of measuring laser absorptance, without going via other radiative properties such as emittance or reflectance. It’s not a direct measurement in the sense that we actually measure the absorbed photons in the material from the laser (an impossibility of course). But all the absorbed radiation eventually heats the material and laser calorimetry is a method that registers the temperature signal that a material produces after it has been illuminated by a laser beam. The strong advantage of laser calorimetry lies in its simplicity, which is one of the reasons why it has been selected as the standard method for measurement of laser absorption in optical laser components (ISO 11551) [15]. It requires relatively cheap equipment and calibration is easy.

Look at Figure 7 for a typical experimental setup. A laser beam is produced and focused down on a sample, which usually sits in a stabilization chamber for thermal protection and which, initially, is in thermal equilibrium with its surroundings. After a sufficient amount of time for the temperature of the sample to be raised above ambient the laser is switched off and the sample is left to cool down to
ambient temperature again. The temperature of the sample is registered via thermocouples (or possibly a pyrometer), while the energy of the laser pulse is measured by reflecting off a small known part via a beam-splitter to a laser power meter.

3.1.1 Theoretical model

For most applications the temperature model underlying the evaluation of the measured data is based on three important assumptions [16]:

1. A homogeneous temperature of the sample, ie. the sample shows infinite thermal conductivity (which implies that metals probably are more suitable for this model than most other materials.)

2. A linear approximation of thermal losses, which can be justified if the temperature rise of the sample is small compared to ambient temperature, ie. if $T(t) - T_0 \ll T_0$, where $T(t)$ is the sample temperature and $T_0$ is the ambient temperature.

3. Sample is at thermal equilibrium with its surroundings prior to measurement, ie. $T(t < t_0) = T_0$ where $t_0$ marks the beginning of the measurement.
From the first of the assumptions (homogeneous temperature) and simple thermodynamics we can derive the following equation, describing the sample temperature change rate:

\[
\frac{dT}{dt} = \frac{1}{C} \left( \frac{dQ_a}{dt} - \frac{dQ_e}{dt} \right),
\]

where \( C \) is the sample heat capacity, \( dQ_a/dt \) is the power absorbed and \( dQ_e/dt \) is the power lost by emission.

The power absorbed is the power incident from the laser, \( P(t) \), multiplied by the spectral, directional absorptance, ie.

\[
\frac{dQ_a}{dt} = A_\lambda' P(t).
\]

Generally for the second term on the right hand side of equ.(12), we can write

\[
\frac{dQ_e}{dt} = SH(T - T_0),
\]

where \( S \) is the total surface area of the sample and \( H(T - T_0) \) is the total heat conductance between the sample and its surroundings, originating from conduction, convection and radiation. If the temperature difference between the sample and its surroundings is small, as we assumed above, we can use a linear approximation and write

\[
\frac{dQ_e}{dt} = Sh(T - T_0),
\]

where \( h \) is the surface heat loss coefficient ([\( W \) \( K \) cm\(^2\)]). Expressions (13) and (15) inserted into equ.(12) then lead to the first order linear differential equation

\[
\frac{dT}{dt} = \frac{A_\lambda' P(t)}{C} - \gamma(T - T_0),
\]

where \( \gamma = \frac{Sh}{C} \) and is known as the temperature-loss coefficient ([\( 1/s \)]).

If we assume the laser power to be constant during irradiation (where \( t_1 \) marks the beginning and \( t_2 \) the end of laser illumination), ie. \( P(t_1 < t < t_2) = P_0 \), then equ.(16) is an autonomous equation and the solution is easily seen to be
By knowing the mass and the specific heat of the sample, one can then deduce the absorptance by correct interpretation of the temperature data.

3.1.2 Evaluation methods

A number of different ways of evaluating the data have been identified (often a combination of more than one are useful for increasing accuracy). Willamowski et al. describes five of them; the gradient method, the pulse method, the exponential method, the integral method and the response method [17], of which we will concentrate on the first three.

The **gradient method** allows one to calculate the absorptance by graphically determine two gradients of the heating and cooling curve, one during irradiation and one after, but both at the same temperature (see Figure 8). According to equ.(16) we have

\[
\frac{A'_{\lambda}P_0}{C} = \frac{\dot{T}_h}{t_h} + \gamma (T(t_h) - T_0) = \frac{\dot{T}_c}{t_c} + \gamma (T(t_c) - T_0) = \frac{\dot{T}_h - \dot{T}_c}{t_h},
\]

where \( t_1 < t_h < t_2 \) and \( t_c > t_2 \) and from which we can see that the absorptance can be determined by a simple summation of the two gradients.

In the **pulse method** the cooling curve is extrapolated (see Figure 9) to a time \( t = t_1 + t_p/2 \) in the middle of the irradiation interval \( (t_p = t_2 - t_1 \) is the duration of the laser irradiation) and the extrapolated temperature \( T_{extr} \) is interpreted as the temperature rise due to an instantaneous energy input \( E_{pulse} = P_0t_p \):

\[
T_{extr} - T_0 = \frac{A'_{\lambda}P_0}{\gamma mc_p}(1 - e^{-\gamma t_p})e^{\gamma t_p/2} \approx \frac{A'_{\lambda}E_{pulse}}{mc_p}(1 + \frac{(\gamma t_p)^2}{24}).
\]

We see from equ.(19) that if \( (\gamma t_p)^2 \ll 1 \), i.e. for small thermal losses or for short irradiation times (hence the name ”pulse” method), the absorptance can be calculated directly from the extrapolated temperature.
Figure 8: In the gradient evaluation method two gradients are determined (see upper graph), one during irradiation/heating (at time $t_h$) and one later when the sample is cooling down (at time $t_c$), but both at the same temperature. In the lower graph the laser pulse is depicted, where $t_1$ and $t_2$ mark the beginning and end of laser irradiation.

The third and most general evaluation method is the **exponential method** which is performed by fitting two exponential functions of the types in equations (17), one for the heating curve ($A_h - B_h e^{-c_h t}$) and one for the cooling curve ($A_c - B_c e^{c_c t}$) [17]. The fitting parameters can then be used in (17) to calculate both the absorptance $A'_\lambda$ and the thermal loss $\gamma$.

All these evaluation methods are based on the temperature model of equations (16) which was derived using the approximations listed in the beginning of 3.1.1. Deviations from these approximations can introduce severe errors in some cases and therefore a number of different sources of these deviations and what can be done about them will now be described.

### 3.1.3 Errors in laser calorimetric measurements

Willamowski *et al.* mentions three sources for signal distortions in laser calorimetric measurements: temperature drift, influence of finite thermal conductivity and effects of scatter irradiation.
Figure 9: In the pulse evaluation method, the cooling curve is extrapolated to a time in the middle of the laser irradiation interval and it is seen, through equ. (19), that if the thermal loss is small or the laser irradiation interval short then the absorptance $A_{\lambda}$ of the sample can be derived from the extrapolated temperature $T_{\text{extr}}$.

Fluctuations of the ambient temperature can be reduced by thermally shielding the sample from the environment, i.e. to enclose and isolate it in a chamber. If temperature drift is still present, especially if the drift is non-linear, it is a good idea to delay the measurement since thermal equilibrium might not have been reached yet. If this doesn’t help the situation a constant or linear drift can usually be compensated for in the equations (16) via a term $T_0(t) = T_0(t_0) + \zeta(t - t_0)$ (see [16] for more information and the solution).

Real samples usually show a finite thermal conductivity, contrary to the assumption used in the temperature model discussed in this paper. Bernal [18] considers the case of thin cylindrical samples which renders this solution,

$$\nu(\rho, t) = \frac{A_{\lambda}}{C} \begin{cases} 0 & t \leq t_1 \\ \sum_i g_i(\rho) \left(1 - e^{-\gamma_i(t-t_1)}\right) & t_1 < t < t_2 \\ \sum_i g_i(\rho) \left(1 - e^{-\gamma_i t_p}\right) e^{-\gamma_i(t-t_2)} & t > t_2 \end{cases}$$

where $\nu(\rho, t) = T(\rho, t) - T_0$. For the homogeneous temperature model $g_0 = 1$ and $g_{i>0} = 0$, but in this case the higher order terms are non-zero and can be calculated from the laser beam profile and the physical properties of the sample. $\gamma_0$
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corresponds to the thermal loss coefficient $\gamma$ used in Eq. (16) while the values $\gamma_{i>0}$ are typically orders of magnitude higher and usually lead to a delay of the temperature rise as compared to the simple homogeneous model. For more complete derivations, see [18] or [16]. Willamowski et al. [17] also speaks of the possibility of replacing $C$ with an effective heat capacity $C_{eff}$ to take the higher order terms into consideration. $C_{eff}$ can be determined by calibration and can, in practice, be described by a term such as $C_{eff} = C_0 + fC$, where $f$ is a number close to 1 and $C_0$ represents the sample holder’s contribution to the heat capacity.

A finite thermal conductivity usually results in an inhomogeneous temperature profile of the sample surface, making it very important to choose the right position on the sample for reading the temperature signal. For sensitive measurements Willamowski et al. suggests a preferable position at an adapted location on the sample where the influence of thermal conductivity is small and nearly negligible. [16] mentions a position 7mm from the sample and laser beam center of a 25mm diameter thin cylindrical sample.

Scatter irradiation can also be a nuisance in laser calorimetric measurements. The two main sources are scattering from the incoming laser beam and from the sample surface. Scattering from the laser beam can be minimized by careful alignment of the sample with respect to the beam and by using apertures. Scattering from the sample surface is more difficult to find a remedy for but it is possible to detect errors from it by analyzing the temperature data ([17] gives good graphical examples of different signal distortions and their causes).

Finally, the temperature models described above require knowledge of the specific heat capacity of the sample, $c$. If absorptance is to be measured for elevated temperatures it also requires information of $c(T)$. This information may not always be available. It can also be noted that if the temperature dependence of $A'_{\lambda}$ is required, both the sample and ambient temperature must be elevated (see the second assumption in 3.1.1), thus requiring the use of a furnace.

### 3.2 Reflectometry

As mentioned earlier, for opaque materials reflectance provides us with indirect means to calculate the absorptance through equ.(5). Radiometric methods are employed to measure either the bidirectional ($R'_{\lambda}$), the directional-hemispherical ($R'_{\lambda}^{\text{hemi}}$) or the hemispherical-directional reflectance ($R'_{\lambda}^{\text{hemi}}$). Most information is of course gained through knowledge of the bidirectional values, but this is also the most diffi-
cult reflectance function to measure.

Reflectometry can be absolute or relative to a reference standard. Absolute measurements are obtained with an optical system to directly measure the intensities of the beams incident on and reflected from the sample. The reflectance is then obtained from the ratio of the two measurements. In relative measurements the beam reflected by the sample is compared with a known standard, a procedure which requires simpler optics, but which may lead to a reduction of accuracy if the standard is of bad quality and is not handled with great care (standards may undergo degradation due to age and handling).

3.2.1 Bidirectional reflectometers

The bidirectional reflectance can be measured by irradiating the sample with a collimated beam and then collecting the reflected radiation over a various number of small solid angles. Of course, for this kind of measurement to be practical for absorptance (or emittance) determination, we need to know the full distribution of the reflectance over the entire hemisphere above the surface for both incidence and reflection angles.

Modern instruments used for BRDF evaluation are known as gonioreflectometers and have a long history of development. Early reflectometer design, eg. Birkebak and Eckert [20] and Torrance and Sparrow [21], used a multi-frequency light source, such as a globar, and a monochromator and various apertures to achieve a collimated monochromatic light beam. These devices often shared the main problem of producing too low a level of reflected radiation for detection, even with the advent of FTIR spectrometers and highly sensitive detectors. This problem was much helped with the introduction of lasers as sources, eg. as in the early work of Hsia and Richmond [22].

Take a look at Figure 10 for a typical experimental setup, where a beam-splitter is used to send a small fraction of the laser beam into a power meter. The sample is placed in a holder on a turntable, which allows rotation of both the sample around three mutually perpendicular axis as well as rotation of the detector which is placed on an arm extending from the sample and holder positions [22]. Modern designs use rotating arms for both source and detector as in the work of White et al. [23], which could work in laser applications if a fiber-coupled laser source is used.

To achieve the angular resolution necessary for integration in absorptance measurements a number of requirements are put on the experimental equipment.
First, the alignment of laser, sample and detector must be precise. A knowledge of the angles of incidence and reflection must be known to better than at least $1^\circ$ (White et al. accomplished an angular resolution of $0.3^\circ$ [23]). The solid angle of the collector (detector) must be small, at least on the order of one degree square. Baffles and apertures in front of the detector can be used for this as well as for blocking scattered radiation from reaching the detector.

A high angular resolution means a lot of data. BRDF evaluation for points $1^\circ$ apart over the entire hemisphere involves 400 million data points, so it is highly desirable to have the measurement process automated by the use of stepping motors [22] controlled by a personal computer.

Gonioreflectometers are available commercially but are still under development.
3.2.2 Directional-hemispherical and hemispherical-directional reflectometers

Reflectometers used for measuring the directional-hemispherical and hemispherical-directional reflectances are usually divided into three categories:

- Heated cavity reflectometers
- Integrating sphere reflectometers
- Integrating mirror reflectometers

**Heated cavity reflectometers** are used to measure hemispherical-directional reflectances. They are also known as Gier-Dunkle reflectometers to honor their inventors in 1954 [24]. Heated cavity reflectometers generally consist of a uniformly heated hohlraum (enclosure) with two openings. In one opening the water-cooled sample is placed and in the other one a viewport is fitted, as in Figure 11.

The radiation striking the sample from the isothermal hohlraum is assumed to be essentially that of a blackbody of the cavity-wall temperature, $T_w$. The radiation from the hohlraum is therefore isotropic and the sample is uniformly illuminated over its entire surface. Images of the sample and a spot on the cavity wall are alternately focused onto the entrance slit of a monochromator. The signal from the sample, $I_s$, consists both of emission from the sample (at the sample temperature $T_s$) and reflection of the cavity-wall’s blackbody intensity, $I_{b\lambda}(T_w)$. Since the signal from the cavity wall, $I_w$, is proportional to $I_{b\lambda}(T_w)$, the ratio of the two signals is [14]

$$\frac{I_s}{I_w} = \frac{R^{\Sigma'} I_{b\lambda}(T_w) + \epsilon'_\lambda I_{b\lambda}(T_s)}{I_{b\lambda}(T_w)}.$$  \hspace{1cm} (21)
If the sample is relatively cold, i.e. \( T_s \ll T_w \), emission can be neglected and the reflectometer simply measures the hemispherical-directional reflectance \( R^{\circ}_{\lambda} \).

For higher sample temperatures, we can use \( R^{\circ}_{\lambda} = R^{\circ} = 1 - A'_{\lambda} \), and write

\[
\frac{I_s}{I_w} = 1 - A'_{\lambda} \left[ 1 - \frac{I_{b\lambda}(T_s)}{I_{b\lambda}(T_w)} \right],
\]

but errors increase severely with the sample temperature and the method is generally only used for low sample temperatures.

The method is extremely sensitive to any deviations from a true black body cavity. The greatest sources of error come from non-uniform temperature and a non-homogeneous field of view. Although attempts have been made to improve the design (e.g. Hembach et al. [25]), it is a method which is hardly used today in reflectance measurements and not many papers have been written on the matter since the 60’s.

**Integrating sphere reflectometers** are the most commonly employed devices for reflectance measurements and are commercially available in a number of different forms. Integrating spheres can be thought of as light collecting devices and may also be used for lamp flux measurements, LED measurements, laser power measurements and various other radiance and irradiance applications. In reflectance applications, they either measure the directional-hemispherical or the hemispherical-directional reflectance, depending on whether they work in the direct or indirect (reciprocal) mode (see Figure 12).

The basis for the function of the integrating sphere is the highly reflective lambertian properties of the coating of the inside of the sphere. A lambertian surface is an ideal surface which reflects radiation equally well from and in all directions. Examples of coating materials used for integrating spheres are smoked magnesium oxide and barium sulfate for visible and near-infrared wavelength regions and roughened gold for the far-infrared wavelength region.

The integrating sphere is perforated with openings or ports on the perimeter of the sphere, where detectors, samples and light sources can be fitted.

In the direct mode, light enters through an entrance port, strikes and reflects off the sample and will then, due to the strong, diffuse reflectance character of the coating and the sphere geometry, undergo multiple reflections and be scattered uniformly around the interior of the sphere. This uniform diffuse light can then be detected at a port configured with a detector device. Let us denote this signal as
Similar readings are also taken on a comparison standard reference of known reflectance $R_r$, under the same conditions (see Figure 12A). Let us denote the signal from the reference $S_r$. The sample may either be replaced by the standard in the same port, which is called the substitution method or a single beam sphere, or there may be separate sample and standard holders, which are alternately irradiated by the light source, which is called the comparison method or a double beam sphere (as in Figure 12A)).

Ideally, the ratio of the two measured signals will then be

$$\frac{S_s}{S_r} = \frac{R_s}{R_r},$$

(23)

where $R_s$ is the sample reflectance.

In the substitution method however, the average reflectance of the sphere changes when the sample is substituted for the reference material. This changes the overall throughput of the system, which is defined as the flux reaching the detector. The true measurement in a single beam sphere is

$$\frac{S_s}{S_r} = \frac{R_s}{R_r} \frac{1 - \overline{R}_r}{1 - \overline{R}_s},$$

(24)

where $\overline{R}_r$ and $\overline{R}_s$ is the average wall reflectance with reference and sample fitted in the sample holder, respectively.
The average reflectance with the sample material in place, $\overline{R}_s$, cannot easily be determined since it also depends on $R_s$.

In a double beam sphere, the sample and reference beams are present in the sphere at essentially the same time. In this case, sample and reference beams each “see” the same sphere and there is no substitution error.

In the indirect mode, light from the source does not illuminate the material directly, but only after multiple diffuse scatterings inside the sphere, as in Figure 12B. The radiance scattered into a specific direction by the sample is then viewed directly by a detector of small angular aperture [26].

Integrating spheres with center mounted and rotatable sample holders exist, allowing for determination of the reflectance as a function of incidence angle and also the possibility of simultaneous measurement of transmittance (sometimes called "transflectance").

Until quite recently, reflectance measurements using integrating spheres have been limited to moderate temperature levels, due to the temperature sensitivity of the coating materials used. However, Zhang and Modest was able to rapidly heat up only the sample by using a two laser configuration, where one laser is used as a heating source and the other one as the probe [27]. Sample temperatures up to 3000°C were reported for reflectance measurements of various ceramic materials.

Errors in integrating sphere measurements are caused mainly by coating imperfections, port losses, problems associated with baffles, detector field of view (FOV) and the so called substitution error involved in single beam spheres.

Imperfections of the sphere coating, causing deviations from the ideal lambertian surface, creates imperfect diffuse reflections inside the sphere. These errors have also been found, through Monte-Carlo calculations, to be a strong function of wall reflectance [28]. It is minimized for reflectances approaching 1.0 and increases as the reflectance value decreases, but are relatively independent of the detector’s field of view. Selection of a good coating material is therefore essential.

Ideally an integrating sphere would have no openings or ports. As this is a physical impossibility (we need at least an entrance port, a sample port and a detector port), errors are introduced by diffuse and specular light escaping out of the sphere through these ports. The best way to reduce these errors is to minimize both the number and the size of the ports. A common recommendation is to let the ports constitute no more than 5% of the total sphere surface area.
In using integrating spheres, it is important that the light reaching the detector does not include direct irradiation from the light source or direct reflections from the sample surface, which would give erroneous results. In spheres working in the comparison mode it is also necessary to shield the sample and reference from each other, so that no direct interchanging radiation is possible. This is usually a question of port geometry as well as the introduction of baffles to the inside of the sphere. Baffles (see Figure 13) limit the detector FOV but at the same time introduce deviations from the ideal sphere, thus both reducing and adding new errors to the measurements.

Detector FOV is a tricky problem, because besides the necessity of shielding it from the light source and sample surface, the FOV needs to be as large as possible to see as much of the sphere as possible which usually leads to a compromise when using baffles. There is usually also the problem with many detectors that as their FOV is increased, their detectivity is decreased. This makes them less sensitive and thus lowers the sphere throughput, which is the flux reaching the detector. There is also the fact that no detectors show a perfect isotropic response to radiation incoming over a large off-normal incidence angle range.

A promising solution to problems associated with detector FOV, which still seem relatively new and not so widely used, are non-imaging concentrators. For integrating spheres, compound elliptic concentrators (CEC) and compound parabolic concentrators (CPC) (see Figures 14 and 15) can be used for a number of advantageous reasons [29]: 1) detector FOV can be varied with no loss in throughput, 2) detector FOV can be sharply defined and 3) losses between detector and sphere can be minimized by proper coupling between concentrator and sphere.

The substitution error in single beam spheres is a systematic, predictable and non-random error which can be corrected to a great extent by using a proper center mount sample holder design [30] and by using reflectance standard references with reflectance values close to the sample reflectance. Limiting the sample size and/or...
increasing the sphere size is also a good way of reducing the error [31].

Figure 14: The working principle of a CPC/detector combination. The ray traces show that CPCs limit the incidence angle on the detector to less than $30^\circ$ for various angles ($20^\circ$ – $80^\circ$) incident on the CPC opening, thus increasing the angular response of the detector.

Figure 15: Integrating sphere geometry which eliminates detector FOV related errors. The CEC views exactly one-half of the sphere, for samples having a BRDF with mirror symmetry around the plane of incidence, equal amounts of radiation are reflected in and out of detector/CEC FOV.

**Integrating mirror reflectometers** share some similar design concepts to integrating spheres, also functioning as light collecting devices. But as opposed to integrating spheres they are designed to gather all radiation from a scattering sample with a minimum number of reflections on a high reflectance surface (usually copper or aluminum) and directing it onto the detector. The main advantage of mirror reflectometers are therefore that the throughputs can be orders of magnitude higher. On the other hand, the influence of misalignment errors is much worse, requiring large detector areas. Three different types of integrating mirror designs have been developed: hemispherical [32], paraboloidal [33] and ellipsoidal [34, 35, 36, 37](see Figures 16 and 17).
The operating principles of the three mirror designs are the same. Each design has two conjugate focal points, which means that if a point source of light is placed in one focal point, all radiation will, after reflection off the mirror, be focused and concentrated to the second focal point. Thus in the integrating mirror reflectometer technique, an external laser beam enters through an entrance port in the mirror and is focused onto the sample, which is placed at one of the focal points. The reflected radiation, regardless of direction, will then be reflected by the mirror and be collected at the second focal point, where the detector is located. This yields, after comparison with a reference signal, the directional-hemispherical reflectance $R'_\lambda$. Alternatively, the integrating mirror can work in a reciprocal mode, where a blackbody source is placed at one of the focal points and where the emitted radiation leaving the blackbody, which will have equal intensity in all directions, will reflect off the mirror and be focused on the sample at the other focal point. Radiation reflected off the sample can then be probed through a small aperture in the mirror and spectrally resolved, by a monochromator or an FTIR spectrometer, yielding the hemispherical-directional reflectance $R'_\lambda$ instead.

Because of mirror limitations measurements have generally been limited to relatively large wavelengths, $\lambda > 2.5\,\mu m$.

Reflectance measurements for samples heated up to temperatures of 1000°C have been reported by Battuello and co-workers [37] and Freeman et al. [38] using a two laser configuration, one heating laser and one probing laser (see Figure 16). Even higher temperatures, over 2000°C, were attainable using oxy/acetylene torches as heating sources, which was reported by Markham and co-workers [39].

For integrating mirror reflectometers the main sources of error come from absorption by the mirror, radiation losses through entrance and viewing slots, nonuni-
form angular response of detectors and mirror aberrations leading to detector over-
filling (ie. radiation missing the detector).

To minimize aberrations, ellipsoids are generally preferred over hemispheres and large detector areas are wanted. As for integrating spheres, non-imaging concentrators (CPC and CEC) have been successfully utilized for improving the angular response and FOV of the detectors [40].

Because of wavelength limitations and sensitivity to flux losses and misalignment errors, integrating mirrors are somewhat less popular than integrating spheres for reflectance measurements [14].

3.3 Emittance radiometry

Spectral directional emittances are most commonly determined by comparing the emission from a sample with the emission from a blackbody at the same wavelength with both bodies held at the same temperature [14]. The same detector should be used in the two measurements as well as an identical or equivalent optical path. Under these conditions, the two signals will both be proportional to the emitted
intensity (with the same proportionality constant) and the spectral directional emittance can be found as their ratio

$$\epsilon'_\lambda(T, \lambda, \theta, \psi) = \frac{S_{s\lambda}(T, \lambda, \theta, \psi)}{S_{b\lambda}(T, \lambda)},$$

(25)

where $S_{s\lambda}$ and $S_{b\lambda}$ are the signals from sample and blackbody, respectively.

The blackbody may either be a separate blackbody held at the same temperature as the sample or it may be an integral part of the sample or the sample chamber. The main problems involved in these kinds of measurements are to ensure equality of sample emitting surface and blackbody temperatures and the uniformity of the temperature over all the active area of the sample surface [13]. Great care must be taken in the design of the heating system of the sample, the thermocouples implementation, calibration and of the temperature control system. The integral version is therefore generally preferred at high temperatures, where temperature control is difficult or at short wavelengths where small temperature deviations can cause large errors.

In the separate reference blackbody method the blackbody is kept separate from the sample chamber, while both are heated to the same temperature. To measure the emission from the blackbody and the sample via an identical path, either two identical paths have to be constructed or sample and blackbody must be alternately placed into a single optical path (see Figure 18 for a possible experimental setup).

In the integrated reference blackbody method, the blackbody is incorporated into the design of the sample furnace. If the sample rests at the bottom of a deep cylindrical and isothermal cavity, the radiation leaving the sample (by emission and reflection) corresponds to that of a black body surface. If the hot side wall is removed or replaced by a cold one, radiation leaving the sample is due to emission only. The ratio of these two signals will then determine the spectral, directional emittance through equation (25).

Two different methods of removing the reflection component of the signal have been identified. Several researchers [41, 42, 43] have used a tubular furnace with the sample mounted on a movable rod. When the sample is deep inside the cavity the signal corresponds to a blackbody. The sample is then rapidly moved to the exit of the furnace and the signal is then due to emission only. Some disadvantages of this approach can be the difficulties in maintaining isothermal conditions of the whole tube, maintaining sample temperature during and after displacement and...
Figure 18: An emissometer set-up for direct measurement of spectral directional emittance, taken from Sacadura et al. The sample and blackbody are kept separate but emissions from them are combined into the same beam by means of a semi-transparent germanium plate. The two signals are separated by chopping them at two slightly differing frequencies.

avoiding stress on the high-temperature sample during the rapid movement [14].

The second approach, which was used by Postlewaith et al. (see Figure 19) is to replace the hot cavity with a cold one. This can be done by dropping a cold tube into the blackbody cavity. Measurements must be taken quite rapidly (in a few seconds time), before the cold tube starts heating up and the sample starts cooling down.

Sources of errors in emittance measurements are sample and blackbody temperature errors, interreflections between the sample and its enclosure and stray radiation. As with laser calorimetry, emittance spectroscopy requires a homogeneous sample temperature. The method is therefore best suited for materials that have a high thermal conductivity. There is also the problem of not obtaining a large enough flux, usually limiting emittance measurements to temperatures $T > 500 \degree C$ and wavelengths $\lambda > 2\mu m$ [13]. Emittance measurements also lack the possibility of acquiring intensity and polarisation dependent information, the latter constraint because all emission due to thermal radiation is unpolarized.
Figure 19: *Schematic of a drop-tube emissometer.*
4 Summary

As we have seen, a number of different methods exist for the measurement of laser absorptance. Laser calorimetry represents the only "direct" way of determining $A'_\lambda$. The indirect means were seen to be reflectometry and emittance spectroscopy. Reflectometry included gonioreflectometers for BRDF measurement ($R'_{\lambda}$), heated cavity reflectometers for obtaining $R_{\lambda}^c$ and integrating spheres and mirrors for determination of both $R_{\lambda}^c$ and $R_{\lambda}^\infty$.

Which method to select for a particular application, is determined by laser intensity, wavelength, angle of incidence, polarisation, sample composition, temperature and available funds.

Laser calorimetry is relatively cheap and calibration is simple. It offers several "degrees of freedom" concerning experimental parameters that can be changed, but may prove a little more difficult for elevated temperatures where a furnace is needed. It also requires some pre knowledge of the sample at hand, especially of its specific heat (as a function of temperature).

Gonioreflectometers are a fairly new tool but are now commercially available in a few different forms. Problems involve getting adequate enough information about the full BRDF function for spatial integration to be accurate. Gonioreflectometers are probably the most complete solution, offering almost full versatility concerning parameters like wavelength, angle of incidence, polarisation, temperature, etc., but need further development [13].

Heated cavity reflectometers are relatively complicated to build and are not commercially available. They can only be used for low temperature measurements. The viewport is usually fixed so unless a turnable holder is utilized only fixed angle measurements can be performed. No intensity or polarisation effects can be studied.

Integrating sphere reflectometers have been around for quite some time and are commercially available from the ultraviolet (250nm) to far infrared wavelengths (20μm). They are versatile but can be sensitive to high temperatures and require a library of suitable reflectance standards.

Integrating mirrors tend to be less popular than integrating spheres because of their sensitivity to misalignment errors, flux losses and mirror aberrations. They also share the same limitations concerning high temperatures.

Emittance spectroscopy is limited to the IR wavelength region ($\lambda > 2\mu m$) and for high temperatures ($> 500 ^\circ C$). No intensity or polarisation effects can be studied.
Paper 2: Laser Absorption Measurements in Opaque Solids

This review is by no means meant to be exhaustive or complete. The aim was to give an overview of some different options available for the measurement engineer wanting to determine how much of his laser energy is absorbed by a specific, opaque sample. Methods based on optical constants determination, such as ellipsometry, polarimetry or Kramer-Krönig integration have been left out, since they either are very sensitive to surface conditions and/or require a vast amount of spectral data that is not readily available to the laser engineer (unless a tunable laser with a very wide spectral band is at hand).

5 Further reading

A good introductory treatment of the optical properties of solids dealing with absorption, reflection, transmission, etc. can be found in Mark Fox’ ”Optical Properties of Solids” (2001) [45].

A good standard text and reference to the field of thermal radiation can be found in the classic book by Robert Siegel and John Howell called ”Thermal Radiation Heat Transfer” (2002) [12], and also in Michael F. Modest’s ”Radiative Heat Transfer” (2003) [14]. In the latter book, an overview of different measurement methods is presented as well as a useful introduction to the most common optical components used in radiative property measurements.

Many of the references used in this paper were collected from Jean-Francois Sacadura’s excellent report ”Measurement techniques for thermal radiation properties” (1990) [13].

Vast databases of optical constants of pure and smooth solids can be found in eg. Palik’s ”Handbook of Optical constants of Solids” [46], as well as explanations of the experimental means of their determination.
References


Paper 2: Laser Absorption Measurements in Opaque Solids


[31] Quantitation of single beam substitution correction in reflectance spectroscopy accessories, Labsphere Application Note No. 1.


Table 1: *Summary of definitions for radiative surface properties of solids*

<table>
<thead>
<tr>
<th>Property</th>
<th>Symbol</th>
<th>Comments</th>
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<tr>
<td><strong>Emittance</strong></td>
<td></td>
<td></td>
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<td>Spectral</td>
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<td></td>
</tr>
<tr>
<td>directional</td>
<td>$\epsilon'_\lambda$</td>
<td>depends on wavelength and outgoing direction</td>
</tr>
<tr>
<td>hemispherical</td>
<td>$\epsilon_{\lambda}$</td>
<td>average over outgoing directions of $\epsilon'_\lambda$</td>
</tr>
<tr>
<td>Total</td>
<td></td>
<td></td>
</tr>
<tr>
<td>directional</td>
<td>$\epsilon_{\text{tot}}$</td>
<td>spectral average over $\epsilon_{\lambda}$</td>
</tr>
<tr>
<td>hemispherical</td>
<td>$\epsilon_{\text{ tot}}$</td>
<td>directional and spectral average of $\epsilon'_\lambda$</td>
</tr>
<tr>
<td><strong>Absorptance</strong></td>
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<td></td>
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<td>directional</td>
<td>$A'_\lambda$</td>
<td>depends on wavelength and incoming direction</td>
</tr>
<tr>
<td>hemispherical</td>
<td>$A_{\lambda}$</td>
<td>average over incoming directions of $A'_\lambda$</td>
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<td>Total</td>
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<tr>
<td>directional</td>
<td>$A_{\text{tot}}$</td>
<td>spectral average over $A_{\lambda}$</td>
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<tr>
<td>hemispherical</td>
<td>$A_{\text{tot}}$</td>
<td>directional and spectral average of $A'_\lambda$</td>
</tr>
<tr>
<td><strong>Reflectance</strong></td>
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<td></td>
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<tr>
<td>bidirectional</td>
<td>$R'_\lambda$</td>
<td>depends on wavelength and both incoming and outgoing directions</td>
</tr>
<tr>
<td>directional-hemispherical</td>
<td>$R'_{\lambda}$</td>
<td>average over outgoing directions</td>
</tr>
<tr>
<td>hemispherical-directional</td>
<td>$R_{\lambda}$</td>
<td>average over incoming directions</td>
</tr>
<tr>
<td>bihemispherical</td>
<td>$R_{\lambda}$</td>
<td>average over incoming and outgoing directions</td>
</tr>
<tr>
<td>Total</td>
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<td>bidirectional</td>
<td>$R_{\text{tot}}$</td>
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<td>bihemispherical</td>
<td>$R_{\text{tot}}$</td>
<td>average over both directions of $R'_{\text{tot}}$</td>
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</table>
Paper 2: Laser Absorption Measurements in Opaque Solids
The Absorptance of Steels to Nd:YLF and Nd:YAG Laser Light at Room Temperature

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Paper 3: The Absorptance of Steels to Nd:YLF and Nd:YAG Laser Light at Room Temperature
The Absorptance of Steels to Nd:YLF and Nd:YAG Laser Light at Room Temperature

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Abstract

The measurement of absorptance is important for the analysis and modelling of laser-material interactions. Unfortunately, most of the absorptance data presently available considers only polished pure metals rather than the commercially available (unpolished, oxidised) alloys which are actually being processed in manufacturing. This paper presents the results of absorptance measurements carried out at room temperature on as-received engineering grade steels including hot and cold rolled mild steel and stainless steels of various types. The measurements were made using an integrating sphere with an Nd:YLF laser at two wavelengths (1053nm and 527nm – which means that the results are also valid for Nd:YAG radiation at 1064nm and 532nm). The absorptance results obtained differ considerably from existing data for polished, pure metals and should help improve the accuracy of laser-material interaction models. Some clear trends were identified; For all materials studied, the absorptance was considerably higher than the previously published values for the relevant pure metals with polished surfaces. For all 15 samples the absorptance was higher for the green than for the infrared wavelength. No clear trend correlating the absorptance with the roughness was found for mild steel in the roughness range \(0.4 \text{ -- } 5.6\) microns. A correlation between absorptance and roughness was noted for stainless steel for \(S_a\) values above 1.5 microns.
1. Introduction

In Laser Material Processing of metals, an understanding of the fundamental absorption mechanisms plays a vital role in determining the optimum processing parameters and conditions. The absorptance\(^1\), which is the fraction of the incident laser light which is absorbed, depends on a number of different parameters, the most important of which are listed in Table 1 [1-4].

<table>
<thead>
<tr>
<th>Laser properties</th>
<th>Metal properties</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wavelength</td>
<td>Chemical composition</td>
</tr>
<tr>
<td>Polarization</td>
<td>Temperature</td>
</tr>
<tr>
<td>Angle of Incidence (Intensity,</td>
<td>Roughness / topography</td>
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<tr>
<td>non-linear effects)</td>
<td>Oxide layers</td>
</tr>
<tr>
<td></td>
<td>Contamination (dust, dirt, surface and bulk defects, etc.)</td>
</tr>
</tbody>
</table>

When modelling laser processing applications, such as cutting, welding, drilling, cladding, hardening, etc., it is important to model the absorption correctly. In very sophisticated models it may be necessary to have an analytical function for the different absorption mechanisms. In other, simpler, cases it may be sufficient to have an accurate average absorptance value to plug in. In any case, published data for the absorptance and reflectance of metals are usually presented only for perfectly pure, clean and flat surfaces which are free from oxide layers (measurements are commonly done in vacuum). This is quite different from the normal situation found in a real life material processing applications. Most metal surfaces are rough to some extent and oxide layers are more of a rule than an exception. Also, the concentration on pure metals has meant that there is very little information available for the absorptance of alloys (stainless steel, brass, etc.), polished or otherwise. In real processing environments alloys are far more commonly used than pure metals. It is therefore generally inappropriate to use the published values of absorptance for pure polished surfaces in mathematical models of laser processing.

\(^1\) Also known as absorptivity. NIST recommends using the –ivity ending for perfectly flat and pure surfaces and –ance for rough and contaminated surfaces (ie. all real surfaces).
Figure 1 demonstrates some of the complexities of photon absorption by an engineering grade metal surface. Such a surface has a characteristic roughness and is always covered by an oxide layer. The surface roughness can result in multiple reflections which involve multiple absorption events (Fig. 1b). Surface roughness can also involve the incoming radiation in absorption events at high angles of incidence, close to the Brewster angle. This can result in very high local absorptance levels. The oxide layer may be absorbing in its own right or may be responsible for multiple reflections or wave guiding (Fig. 1c). In addition to these effects the surface may be contaminated with material which has a higher absorptance than the underlying metal.

Fig. 1: Some of the mechanisms which increase the absorptivity of real engineering surfaces: (a) typical cross section of an engineering surface, (b) high (Brewster) angle absorptance and multiple reflections due to surface roughness, (c) multiple reflections within an oxide layer.

All of the above phenomena increase the absorptance of the surface in question and the use of alloying elements will also tend to complicate the situation. For this reason the best way to establish the absorptance of an engineering metal surface is by direct measurement.
It is, of course, extremely difficult to measure the absorptance of the molten, turbulent surfaces typical of laser welding or cutting. However, it is possible to accurately measure the absorptance of the solid material at room temperature and the values obtained can provide an insight into how the materials behave at elevated temperatures. This can be argued from the fact that, for wavelengths around 1\( \mu \)m, the temperature dependence of absorptance is small. It is well established that metals usually have a so called X-point, a wavelength point or band where the temperature coefficient of absorptance changes sign [5]. The reason for this kind of behaviour lies in the different temperature coefficients for intraband (free-electron) and interband (bound-electron) absorption, the two intrinsic absorption mechanisms prevalent in metals. Intraband absorption increases with temperature, simply because the free electrons gain kinetic energy while the phonon population grows, which increases the electron-phonon collision frequency (ie. the energy conversion efficiency between the light, the free electrons and the lattice). Interband absorption, on the other hand, is much more complicated and depends on the particular band structure of the metal. The general principle is that as the temperature is elevated, the electron Fermi energy distribution function is smoothed out and the absorption bands are broadened. The competition between these two effects leads to the X-point, which for most metals lies near 1\( \mu \)m [6].

In any case, the absorptance of the solid material as opposed to the molten state is in itself very important for two reasons:

1. During the initiation of any weld or cut the laser is interacting with the solid surface.
2. During welding or cutting the leading edge of the laser beam in many cases interacts with the solid surface of the material.

This paper presents the results of an experimental survey of the room-temperature absorptance of a wide range of commercially available steels in the as-received state.

A comprehensive literature survey with 46 references on experimental absorption measurement methods is given in [7] by the present authors. In addition the present authors have provided a comprehensive literature survey on theory and modelling of the absorption mechanism in metals in [1], with reference
to 47 publications. General treatments of laser absorption mechanisms can be found in [1-4].

There are a number of different experimental methods available for measuring laser absorptance in opaque solids. Some of the more commonly used are; laser calorimetry, gonioreflectometry, integrating sphere or integrating mirror reflectometry and emittance spectroscopy [7-9].

In this paper, reflectometry using an integrating sphere was selected, mainly because integrating spheres are commercially available, relatively inexpensive and easy to use while at the same time being very accurate and versatile. By measuring the reflectance $R$ of a sample, we get immediate information regarding the absorptance $A$, since $A = 1 - R$ for opaque solids.

2. Experimental setup

2.1 Instrumentation

The reflectance measurements were carried out in the laser laboratory at the Mid Sweden University, in Östersund. The laser used as the radiation source was a 1W cw Nd:YLF laser with a fundamental wavelength 1053nm, equipped with a SHG (Second Harmonic Generation) crystal to also allow production of green light at 527nm.

The reflectance of the various metals was measured by using an integrating sphere in a double beam setup, as shown in Fig. 2. The 150mm diameter 6-port integrating sphere has a barium sulphate based coating called Spectraflect, which has high reflective Lambertian properties for wavelengths between 300 and 2400 nm.

The Nd:YLF laser beam was directed onto a rotatable mirror (M2 in Fig. 2) that was flipped between two positions to produce sample and reference beams. The flipping frequency was set to approximately 1 Hz, to ensure mirror stability. Mirrors 3 and 4 (M3 and M4 in Fig. 2) then directed the beams onto the sample and reference, respectively, at an angle of incidence of 8°. As reference a port plug of Spectraflect was used. After spatial integration by the sphere, the reflected
light was then detected by a reverse biased 5.7 mm² Si photodiode, with a spectral response between 190nm and 1100nm. The voltage signal of the photodiode was finally digitized in an A/D-converter and sampled by a PC.

2.2 Error correction methods

Several correction methods were applied to the reflectance measurements to improve accuracy and minimize errors. Clarke and Compton [10] have identified the following main sources of errors in integrating sphere reflectometers:
(a) Light losses through ports in the sphere wall,
(b) Unequal illumination of the sample and standard and changes in throughput when a single beam method is used,
(c) Directional dependence of light scattering from the sample and standard surfaces,
(d) Errors due to diffraction effects in apertures,
(e) Errors due to imperfect diffusion of reflected light from the sphere walls (non-lambertian characteristics) and
(f) Baffling.
To minimize these errors the following procedures and correction methods were applied to the measurements:

(a) Light losses:
With the aim of reducing light losses, all ports not in use were closed with port plugs.

(b) Unequal illumination of sample and standard:
The double beam configuration used in these experiments avoids the substitution error which is often a feature of single beam measurements (and can be as large as 4-5 % according to the manufacturers [11]). The double beam configuration also has the advantage of reducing the influence of possible laser power fluctuations.

(c) Directional dependence of light scattering:
Light behaves slightly different inside the sphere when reflected from a rough surface as compared to a relatively flat one. Light incident on a flat surface is subjected to mirror-like, specular reflection in contrast to rough surfaces where the reflected light is scattered diffusively into many different directions. This produces differences in the number of reflections needed inside the sphere before the light is homogenously scattered, which will lead to detector throughput changes. This error can be corrected for by using a combination of diffuse (rough) and specular (flat) reflectance standards.

By knowing the sample diffusivity, $D_s$, which is the fraction of reflected light which is scattered diffusively (specular part excluded), the reflectance of the sample, $R_s$, can then be obtained by

$$R_s = S_s \left[ (1 - D_s) \frac{R_{s,r}}{S_{s,r}} + D_s \frac{R_{d,r}}{S_{d,r}} \right],$$

where $S_s$ is the signal measured with the sample, $R_{s,r}$ is the reflectance of the specular standard (mirror), $S_{s,r}$ is the signal measured with the specular standard, $R_{d,r}$ is the reflectance of the diffuse standard and $S_{d,r}$ is the signal measured with the diffuse standard[12].
The diffusivity can be measured by opening the specular exclusion port (or attaching a light trap) and comparing the signal from the sample to a signal from the diffuse standard, see Fig. 3. The diffusivity is then given by

\[
D_s = 1 - \frac{S_{s,\text{spec}} / S_s}{S_{d,\text{spec}} / S_{d,r}},
\]

where \( S_{s,\text{spec}} \) is the signal measured with sample and with specular exclusion port opened and \( S_{d,\text{spec}} \) is the signal measured with the diffuse standard and specular exclusion port opened. Using this method, a diffusivity of 100 % was assigned to the diffuse standards and 0 % to the specular mirror standard. This error was empirically seen to be almost negligible for \( \lambda = 527 \) nm, but could amount to more than 1 % in absolute reflectance for \( \lambda = 1053 \) nm.

(d) Diffraction effects in apertures:

To get a suitable laser spot size on the sample, a lens was put in front of the sample beam entrance port. Due to diffraction, this produces a characteristic halo around the sample and sample port, which causes a slight error to the measurements. Empirically this was shown to be around 0.2 % in absolute reflectance. This error was easily corrected for by measuring the signal with the sample port empty and subtracting this from the signal with the sample in place.
Errors (e) and (f) are due to the specific sphere designs and are not easily corrected for.

Another error was identified in the case of structured samples with contoured surfaces (termed ‘lay’ in the literature on surface properties). According to Roos [13] this error can be substantial if care is not taken in positioning the samples correctly in the sample ports. A contoured surface produces a reflected band of light (termed specular-diffuse radiation by Roos [13]), which will produce errors if the band crosses the detector (giving too high a reflectance value) or passes through any open beam ports (giving too low a reflectance value). No other corrections than correct positioning was applied in this case.

The measurements were performed in a dark room to minimize the influence of external light sources, such as lamps. Corrections were also made to the current flowing through the photodiode in absence of light. These dark current measurements were performed before each measurement, which were then subtracted from the measured signals.

Using the correction methods outlined above, the reflectance values of all the diffuse and specular standards were seen to be reproducible to at least within 0.3% in absolute reflectance. The same reliability can approximately be attributed to the following results for the samples investigated.

2.3 Samples and measurements

Table 2 lists the different samples of steels (ferrous samples) examined in the survey in terms of their standards denominations and their chemical composition. Note that a similar study on aluminium alloys, copper alloys and zinc-coatings was also carried out and is under preparation as a separate publication.
Table 2: List of the steels examined in this survey and their chemical composition.

<table>
<thead>
<tr>
<th>No.</th>
<th>Mild steels</th>
<th>Fe*</th>
<th>C*</th>
<th>Cr*</th>
<th>Ni*</th>
<th>Mn*</th>
</tr>
</thead>
<tbody>
<tr>
<td>MS1 – MS2</td>
<td>CR4</td>
<td>99.0%</td>
<td>0.1%</td>
<td>0.0%</td>
<td>0.0%</td>
<td>0.6%</td>
</tr>
<tr>
<td>MS3 – MS7</td>
<td>43A, DIN 17100, St44-2</td>
<td>98.0%</td>
<td>0.2%</td>
<td>0.0%</td>
<td>0.0%</td>
<td>1.5%</td>
</tr>
<tr>
<td>Stainless steels</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SS1, SS3, SS6</td>
<td>AISI 304SS, DIN 1.4301</td>
<td>70.4%</td>
<td>0.0%</td>
<td>18.0%</td>
<td>9.5%</td>
<td>2.0%</td>
</tr>
<tr>
<td>SS2, SS4, SS8</td>
<td>AISI316SS, DIN 1.4401</td>
<td>68.4%</td>
<td>0.0%</td>
<td>17.0%</td>
<td>12.5%</td>
<td>2.0%</td>
</tr>
<tr>
<td>SS5</td>
<td>AISI3Cr12, DIN 1.4003</td>
<td>88.0%</td>
<td>0.0%</td>
<td>11.5%</td>
<td>0.5%</td>
<td>0.0%</td>
</tr>
<tr>
<td>SS7</td>
<td>AISI430SS, DIN 1.4016</td>
<td>81.4%</td>
<td>0.0%</td>
<td>17.0%</td>
<td>0.5%</td>
<td>1.0%</td>
</tr>
</tbody>
</table>

*wt.%

The corresponding technical surfaces for the 15 samples studied are described in Table 3. The roughness values given in this table are supported by SEM and optical microscopy together with profilometry imaging in the results section of this work.

Table 3: Surface conditions of the 15 samples studied

<table>
<thead>
<tr>
<th>No.</th>
<th>Mild steels</th>
<th>Surface</th>
<th>Sa [μm]</th>
<th>Sq [μm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>MS1</td>
<td>CR4</td>
<td>Cold Rolled</td>
<td>0.92</td>
<td>1.16</td>
</tr>
<tr>
<td>MS2</td>
<td>CR4</td>
<td>Cold Rolled</td>
<td>1.45</td>
<td>1.68</td>
</tr>
<tr>
<td>MS3</td>
<td>43A</td>
<td>Hot Rolled</td>
<td>1.73</td>
<td>2.17</td>
</tr>
<tr>
<td>MS4</td>
<td>43A</td>
<td>Hot Rolled</td>
<td>0.46</td>
<td>0.58</td>
</tr>
<tr>
<td>MS5</td>
<td>43A</td>
<td>Hot Rolled</td>
<td>1.24</td>
<td>1.51</td>
</tr>
<tr>
<td>MS6</td>
<td>43A</td>
<td>HR, pickled, oiled</td>
<td>2.68</td>
<td>3.23</td>
</tr>
<tr>
<td>MS7</td>
<td>43A</td>
<td>HR, corroded</td>
<td>5.58</td>
<td>6.47</td>
</tr>
<tr>
<td>Stainless steels</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SS1</td>
<td>304SS</td>
<td>Cold Rolled</td>
<td>0.29</td>
<td>0.34</td>
</tr>
<tr>
<td>SS2</td>
<td>316SS</td>
<td>Cold Rolled</td>
<td>0.15</td>
<td>0.19</td>
</tr>
<tr>
<td>SS3</td>
<td>304SS</td>
<td>Hot Rolled</td>
<td>3.26</td>
<td>4.17</td>
</tr>
<tr>
<td>SS4</td>
<td>316SS</td>
<td>Hot Rolled</td>
<td>2.25</td>
<td>3.57</td>
</tr>
<tr>
<td>SS5</td>
<td>3Cr12</td>
<td>Hot Rolled</td>
<td>2.47</td>
<td>3.48</td>
</tr>
<tr>
<td>SS6</td>
<td>304SS</td>
<td>Bright annealed</td>
<td>0.09</td>
<td>0.12</td>
</tr>
<tr>
<td>SS7</td>
<td>430SS</td>
<td>Bright annealed</td>
<td>0.05</td>
<td>0.08</td>
</tr>
<tr>
<td>SS8</td>
<td>316SS</td>
<td>Dull polished (brushed)</td>
<td>1.38</td>
<td>1.83</td>
</tr>
</tbody>
</table>
The standard engineering phrases which describe the surface condition of the steels (hot rolled, pickled and oiled etc.) need some explanation here;

Mild Steel – Cold Rolled; In this case the as-received material has a surface which has a microscopic surface texture as a result of the rolling process. Cold rolled mild steel has a clean surface which is covered in a very thin layer of oxide which is transparent to the naked eye. (The material is also usually coated in a thin layer of oil - which was removed prior to absorptance testing). The material is approximately 99% pure iron.

Mild Steel – Hot rolled; Hot rolled mild steel is covered in a material called ‘Mill Scale’ which is an oxide layer several tens of microns deep. The laser therefore interacts primarily with this surface mixture of iron oxides rather than the underlying 98% iron alloy.

Mild Steel – HR (Hot Rolled), Pickled and Oiled; Mild steel in this condition has been hot rolled to the required thickness and subsequently ‘Pickled’ in acid to remove the ‘mill scale’ oxide surface. After pickling, the sheets of steel are coated in oil as a protection against corrosion (This oil was, of course, removed prior to our reflectance measurements). The surface texture of this material is rougher than that of cold rolled mild steel. The surface can also have residual pockets of oxide in some of the deeper surface pits (see Fig. 1).

Mild Steel – HR (Hot Rolled), Corroded; Cold rolled and hot rolled pickled and oiled mild steel is coated in oil and therefore resistant to corrosion during storage. Hot rolled material is corrosion resistant as a result of its surface layer of oxides. However, if hot rolled mild steel is stored outside for any length of time, it can accumulate a patchy layer of brown corrosion products because water can penetrate the oxide layer. As part of this survey of as-received engineering grade steels we included samples of partially corroded hot rolled steel.

Stainless steel – Cold rolled; This material has a clean, microscopically textured surface.

Stainless steel – Hot rolled; Unlike its mild steel counterpart, hot rolled stainless steel is not coated with oxides as a result of the rolling process. Hot rolled stainless does, however, have a rougher surface texture than the cold rolled material.
Stainless steel – Bright Annealed; As the name implies, this grade of stainless steel has been rolled to produce a bright, almost mirror quality surface.

Stainless Steel – Dull Polished (Brushed). This grade of stainless steel has been surface ground in one direction to give the ‘brushed’ finish commonly used in display work and hotel bars etc.

The samples were laser or mechanically cut from sheets into 30mm squares with thicknesses varying between 2 and 6 mm. For each sample, measurements were taken at five separate locations which were then averaged and a standard deviation was calculated. Each measurement for every location is in turn an average of over 300 data points.

All integrating sphere measurements are done relative to a standard of known reflectance. In these experiments a combination of diffuse and specular reflectance standards were utilized. The diffuse standards had reflectances between 2 and 99 %. The specular standard consisted of an Al-coated mirror, which was calibrated at the Ångström Laboratory in Uppsala, Sweden. It had a reflectance of 97.7 % at 1053nm and 98.9 % at 527nm.

The reliability of the absorptance measurement method was regarded as satisfactory. The variance $\sigma$ of the measured $A$ values was low. In 17 of 30 cases (15 material samples, 2 wavelengths) the variance of the five measurements was less than 1.5%, only in one case (SS3, IR) it was higher than 10%.

2.4 Equipment for surface characterization

For characterizing the surfaces of the different samples investigated in this study, a Nikon Eclipse L200 optical microscope, a CamScan MV2300T Scanning Electron Microscope (SEM) and a Wyko NT1100 optical profiler were used. The optical profiler, used for measuring the roughness of the sample surfaces, had a horizontal resolution of 1.6 $\mu$m and a vertical resolution in the sub-nanometer range.
3. Results

Figures 4-7 for mild steel and Figs. 9-11 for stainless steel present the optical microscopy (OM), Scanning Electron Microscope (SEM), profilometry and absorptance results obtained in this survey. In each figure, the magnification used is 100 times for the optical microscopy pictures, 2000 times for the SEM photos and 5 times for the 3-D optical profilometry scans. In the table below the photos the average absorptance A and standard deviation σ are given for both wavelengths as well as the results of the optical profilometry measurements. Figures 8 and 12 show the measured absorptance plotted as a function of the surface roughness for the mild and Stainless steels respectively.

3.1 Mild Steels

![Image of cold rolled mild steel](image-url)

<table>
<thead>
<tr>
<th>λ=1053nm</th>
<th>λ=532nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>σ</td>
</tr>
<tr>
<td>62.0%</td>
<td>2.2%</td>
</tr>
</tbody>
</table>

Sa[µm] 0.92  Sq[µm] 1.16

Fig. 4: Cold rolled mild steel: (a) MS1: CR4

\[ S_a = \iint_a Z(x,y) \, dx \, dy \quad \text{and} \quad S_q = \sqrt{\iint_a (Z(x,y))^2 \, dx \, dy}. \]
Paper 3: The Absorptance of Steels to Nd:YLF and Nd:YAG Laser Light at Room Temperature

<table>
<thead>
<tr>
<th>λ=1053nm</th>
<th>λ=532nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>σ</td>
</tr>
<tr>
<td>48.1%</td>
<td>1.4%</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Sa[μm]</th>
<th>Sa[μm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.45</td>
<td>1.68</td>
</tr>
</tbody>
</table>

Fig. 4: Cold rolled mild steel: (b) MS2: CR4

<table>
<thead>
<tr>
<th>λ=1053nm</th>
<th>λ=532nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>σ</td>
</tr>
<tr>
<td>84.3%</td>
<td>0.6%</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Sa[μm]</th>
<th>Sa[μm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.73</td>
<td>2.17</td>
</tr>
</tbody>
</table>

Fig. 5: Hot rolled mild steel: (a) MS3: 43A
The Absorptance of Steels to Nd:YLF and Nd:YAG Laser Light at Room Temperature

<table>
<thead>
<tr>
<th>$\lambda=1053\text{nm}$</th>
<th>$\lambda=532\text{nm}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>$\sigma$</td>
</tr>
<tr>
<td>81.4%</td>
<td>0.4%</td>
</tr>
</tbody>
</table>

$S_a[\mu m]$ 0.46 $S_q[\mu m]$ 0.58

Fig. 5: Hot rolled mild steel: (b) MS4: 43A

<table>
<thead>
<tr>
<th>$\lambda=1053\text{nm}$</th>
<th>$\lambda=532\text{nm}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>$\sigma$</td>
</tr>
<tr>
<td>86.8%</td>
<td>0.2%</td>
</tr>
</tbody>
</table>

$S_a[\mu m]$ 1.24 $S_q[\mu m]$ 1.51

Fig. 5: Hot rolled mild steel: (c) MS5: 43A
Paper 3: The Absorptance of Steels to Nd:YLF and Nd:YAG Laser Light at Room Temperature

<table>
<thead>
<tr>
<th>λ=1053nm</th>
<th>λ=532nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>σ</td>
</tr>
<tr>
<td>67.8%</td>
<td>0.7%</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Sa[μm]</th>
<th>Sq[μm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.68</td>
<td>3.23</td>
</tr>
</tbody>
</table>

Fig. 6: MS6: mild steel 43A, hot rolled, pickled and oiled

<table>
<thead>
<tr>
<th>λ=1053nm</th>
<th>λ=532nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>σ</td>
</tr>
<tr>
<td>87.6%</td>
<td>1.3%</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Sa[μm]</th>
<th>Sq[μm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.58</td>
<td>6.47</td>
</tr>
</tbody>
</table>

Fig. 7: MS7: mild steel 43A, hot rolled and corroded

106
The measured absorptance as a function of the roughness is plotted in Fig. 8 for the two wavelengths applied. No clear dependence on the roughness can be identified.

![Graph showing absorptance as a function of roughness](image)

**Fig. 8:** Absorptance as a function of the surface roughness for the seven mild steel cases studied (ref. values in Figs. 4-7)

As Table 2 demonstrates, mild steel is >98% iron. However, the absorptance of cold rolled mild steel (52% at 1053nm and 67% at 527nm) is considerably higher than the published figure for polished iron (36% at 1053nm and 43% at 527nm [14]). This increase in absorptance is to be expected as our mild steel samples are not polished and are, to some extent, oxidised.

As would be expected, the absorptance of mild steel increases with increasing levels of surface oxide. The relatively clean metal surface of cold rolled mild steel is more reflective than a pickled and oiled hot rolled surface. Hot rolled mild steel does not have a metallic surface as it is coated in a combination of iron oxides. The reflectances of FeO, Fe₂O₃ and Fe₃O₄ are shown in Table 4.

It is clear from the results that the presence of iron oxides, either as mill scale or corrosion products will increase the absorptance of the steel surface. It is also clear that the presence, or otherwise, of a thick oxide surface affects the absorptance at these two wavelengths by different amounts. For the two cold rolled samples the green light absorptance value was higher than the infra-red value by 16% in one case and 34% in the other. The differences for the hot rolled, oxide coated material were considerably smaller than this, with an average value
of 3%. This indicates a disproportionate rise in the absorptance at the infra-red wavelength in the presence of thick layers of oxide.

### Table 4. Reflectances of the iron oxides

<table>
<thead>
<tr>
<th>Oxide</th>
<th>Reflectance</th>
<th>(\lambda=1053\text{nm})</th>
<th>(\lambda=527\text{nm})</th>
</tr>
</thead>
<tbody>
<tr>
<td>FeO ([15])</td>
<td>19%</td>
<td>20%</td>
<td></td>
</tr>
<tr>
<td>Fe(_2)O(_3) (hematite) ([16])</td>
<td>31%</td>
<td>3%</td>
<td></td>
</tr>
<tr>
<td>Fe(_3)O(_4) (magnetite) ([17])</td>
<td>20%</td>
<td>17%</td>
<td></td>
</tr>
</tbody>
</table>

### 3.2 Stainless steels

<table>
<thead>
<tr>
<th>(\lambda=1053\text{nm})</th>
<th>(\lambda=532\text{nm})</th>
<th>(A)</th>
<th>(\sigma)</th>
<th>(A)</th>
<th>(\sigma)</th>
</tr>
</thead>
<tbody>
<tr>
<td>37.4%</td>
<td>0.3%</td>
<td>44.6%</td>
<td>0.2%</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Sa[(\mu\text{m})]</th>
<th>0.29</th>
<th>Sq[(\mu\text{m})]</th>
<th>0.34</th>
</tr>
</thead>
</table>

**Fig. 9**: Cold rolled stainless steel: (a) SS1: 304SS
Paper 3: The Absorptance of Steels to Nd:YLF and Nd:YAG Laser Light at Room Temperature

<table>
<thead>
<tr>
<th>λ=1053nm</th>
<th>λ=527nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>σ</td>
</tr>
<tr>
<td>37.2%</td>
<td>0.3%</td>
</tr>
</tbody>
</table>

Sa[μm] 0.15  Sq[μm] 0.19

Fig. 9: Cold rolled stainless steel: (b) SS2: 316SS

<table>
<thead>
<tr>
<th>λ=1053nm</th>
<th>λ=527nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>σ</td>
</tr>
<tr>
<td>57.5%</td>
<td>3.2%</td>
</tr>
</tbody>
</table>

Sa[μm] 3.26  Sq[μm] 4.17

Fig. 10: Hot rolled stainless steel: (a) SS3: 304SS
### Table 1: Absorptance of Steels to Nd:YLF and Nd:YAG Laser Light at Room Temperature

<table>
<thead>
<tr>
<th>λ (nm)</th>
<th>A (%)</th>
<th>σ (%)</th>
<th>λ (nm)</th>
<th>A (%)</th>
<th>σ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1053</td>
<td>54.6%</td>
<td>9.4%</td>
<td>532</td>
<td>64.1%</td>
<td>6.5%</td>
</tr>
<tr>
<td>Sa[μm]</td>
<td>2.25</td>
<td>Sq[μm]</td>
<td>3.57</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Fig. 10: *Hot rolled stainless steel: (b) SS4: 316SS*

### Table 2: Absorptance of Steels to Nd:YLF and Nd:YAG Laser Light at Room Temperature

<table>
<thead>
<tr>
<th>λ (nm)</th>
<th>A (%)</th>
<th>σ (%)</th>
<th>λ (nm)</th>
<th>A (%)</th>
<th>σ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1053</td>
<td>44.2%</td>
<td>1.8%</td>
<td>527</td>
<td>54.8%</td>
<td>3.8%</td>
</tr>
<tr>
<td>Sa[μm]</td>
<td>2.47</td>
<td>Sq[μm]</td>
<td>3.48</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Fig. 10: *Hot rolled stainless steel: (c) SS5: 3Cr12*
Paper 3: The Absorptance of Steels to Nd:YLF and Nd:YAG Laser Light at Room Temperature

<table>
<thead>
<tr>
<th>$\lambda=1053\text{nm}$</th>
<th>$\lambda=527\text{nm}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A$</td>
<td>$\sigma$</td>
</tr>
<tr>
<td>32.4%</td>
<td>0.2%</td>
</tr>
</tbody>
</table>

$Sa[\mu\text{m}]$ 0.09 $Sq[\mu\text{m}]$ 0.12

Fig. 11: Specially prepared stainless steel surfaces: (a) SS6: 304SS, bright annealed

<table>
<thead>
<tr>
<th>$\lambda=1053\text{nm}$</th>
<th>$\lambda=527\text{nm}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A$</td>
<td>$\sigma$</td>
</tr>
<tr>
<td>37.0%</td>
<td>0.3%</td>
</tr>
</tbody>
</table>

$Sa[\mu\text{m}]$ 0.05 $Sq[\mu\text{m}]$ 0.08

Fig. 11: Specially prepared stainless steel surfaces: (b) SS7: 430SS, bright annealed
Fig. 11: Specially prepared stainless steel surfaces: (c) SS8: 318SS, dull polished (brushed)

The measured absorptance as a function of the surface roughness is plotted in Fig. 12. In this case there are indications of a roughness-absorptance correlation at roughness values above $Sa = 1.5$ microns.

![Image of specially prepared stainless steel surfaces]

<table>
<thead>
<tr>
<th>$\lambda=1053\text{nm}$</th>
<th>$\lambda=527\text{nm}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A$</td>
<td>$\sigma$</td>
</tr>
<tr>
<td>37.3%</td>
<td>2.3%</td>
</tr>
</tbody>
</table>

$Sa[\mu\text{m}]$ 1.38  $Sg[\mu\text{m}]$ 1.83

Fig. 12: Absorptance as a function of the surface roughness for the eight stainless steel cases studied (ref. values in Figs. 9-11)
Comparison of the surface profilometry maps and data in Figs. 9 and 10 reveals that the hot rolled material has Sa values which are an order of magnitude greater than those for the cold rolled material. This has resulted in higher absorptance values for the hot rolled material. The correlation between surface roughness and absorptance seems, however, to disappear at low levels of surface roughness. For example; the standard cold rolled 316SS sample has a roughness measurement which is an order of magnitude larger than the dull polished (brushed) sample but, in this case, both Sa values are below 1.5 microns and the absorptances of the two surfaces are very similar. The whole subject of roughness-absorptance correlation will be the subject of future work by the present authors.

Table 5 lists a summary of the absorptance and reflectance measurements taken in these experiments.

Table 5. A summary of the average absorptance and reflectance measurements obtained from this survey

<table>
<thead>
<tr>
<th>Material</th>
<th>$\lambda=1053\text{nm}$</th>
<th>$\lambda=527\text{nm}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>A</td>
<td>R</td>
</tr>
<tr>
<td>MS1,2: Cold rolled mild steel (averaged)</td>
<td>52%</td>
<td>48%</td>
</tr>
<tr>
<td>MS3-5: Hot rolled mild steel (averaged)</td>
<td>86%</td>
<td>14%</td>
</tr>
<tr>
<td>MS6: Hot rolled, pickled + oiled mild steel</td>
<td>68%</td>
<td>32%</td>
</tr>
<tr>
<td>MS7: Hot rolled, corroded mild steel</td>
<td>88%</td>
<td>12%</td>
</tr>
<tr>
<td>SS1,2: Cold rolled stainless steel (aver.)</td>
<td>37%</td>
<td>63%</td>
</tr>
<tr>
<td>SS3,4: Hot rolled stainless steel (aver.)</td>
<td>56%</td>
<td>44%</td>
</tr>
<tr>
<td>SS5: Hot rolled 3Cr12</td>
<td>44%</td>
<td>56%</td>
</tr>
<tr>
<td>SS6,7: Bright annealed stainless steel (av.)</td>
<td>35%</td>
<td>65%</td>
</tr>
<tr>
<td>SS8: Brushed stainless steel</td>
<td>37%</td>
<td>63%</td>
</tr>
</tbody>
</table>

4. Conclusions

Reliable absorptance results have been measured for a range of mild and stainless steels for Nd:YLF and Nd:YAG lasers at their infrared and green wavelengths. These results have, as expected, been found to differ considerably from existing published data for pure, polished metals.
Several trends could be identified for the 15 ferrous samples:

- All measured absorptance values were considerably higher than the previously published ones for pure, polished metals. The increases in absorptance can be attributed to; surface oxides, surface contamination, surface roughness and the presence of alloying elements. Further studies will be needed to isolate the contributions and importance of each factor.

- The absorptance of the ferrous samples was always higher for the green wavelength compared to the infrared.

- For stainless steel a trend of increasing absorptance for increasing roughness could be seen for Sa roughness values above 1.5 microns. Below this value there was no roughness-absorptance correlation. No clear roughness-absorptance correlation was observed for the mild steel samples for Sa values below 6 microns.

### Bibliography


Paper 3: The Absorptance of Steels to Nd:YLF and Nd:YAG Laser Light at Room Temperature
The Absorptance of Non-Ferrous Alloys to Nd:YLF and Nd:YAG Laser Light at Room Temperature

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²Luleå University of Technology, Luleå, Sweden
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Paper 4: The Absorptance of Non-Ferrous Alloys to Nd:YLF and Nd:YAG Laser Light at Room Temperature
The Absorptance of Non-Ferrous Alloys to Nd:YLF and Nd:YAG Laser Light at Room Temperature

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Abstract

The measurement of absorptance is important for the analysis and modelling of laser-material interactions. Unfortunately, most of the absorptance data presently available considers only polished pure metals rather than the commercially available (unpolished, oxidised) alloys which are actually being processed in manufacturing. This paper presents the results of absorptance measurements carried out at room temperature on as-received engineering grade non-ferrous metals (Al, Cu and Zn alloys). The measurements were made using an integrating sphere with an Nd:YLF laser at two wavelengths (1053nm and 527nm – which means that the results are also valid for Nd:YAG radiation at 1064nm and 532nm). The absorptance results obtained differ considerably from existing data for polished, pure metals and should help improve the accuracy of laser-material interaction models. Some clear trends were identified. For all 22 cases studied the absorptance was higher than for ideal pure, polished metals. For all Al- and Cu-samples the absorptance was higher for the green than it was for the infrared wavelength, while for all Zn-coatings this trend was reversed. No clear correlation between absorptance and surface roughness was found at low roughness values (Sa 0.15 – 0.60) but one, rougher set of samples (Sa 2.34), indicated a roughness-absorptance correlation at higher roughness levels.
1. Introduction

In laser materials processing of metals, an understanding of the laser absorption mechanisms plays a vital role in determining the optimum processing parameters. The absorptance, which is the fraction of the incident laser light which is absorbed, depends on a number of different parameters, the most important of which are listed in Table 1. (‘Absorptivity’ is a term restricted to flat, pure surfaces, ‘Absorptance’ is a more general term).

When modelling laser processing applications, such as cutting, welding, drilling, cladding, hardening, etc., it is important to model the absorption correctly. In very sophisticated models it may be necessary to have an analytical function for the different absorption mechanisms. In other, simpler, cases it may be sufficient to have an accurate average absorptance value to plug in. In any case, published data for the absorptance and reflectance of metals are usually presented only for perfectly pure, clean and flat surfaces which are free from oxide layers (measurements are commonly done in vacuum). This is quite different from the normal situation found in a real life material processing applications. Most metal surfaces are rough to some extent and oxide layers are more of a rule than an exception. Also, the concentration on pure metals has meant that there is very little information available for the absorptance of alloys (stainless steel, brass, etc.), polished or otherwise. In real processing environments alloys are far more commonly used than pure metals. It is therefore generally inappropriate to use the published values of absorptance for pure polished surfaces in mathematical models of laser processing.

<table>
<thead>
<tr>
<th>Laser properties</th>
<th>Metal properties</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wavelength</td>
<td>Chemical composition</td>
</tr>
<tr>
<td>Polarization</td>
<td>Temperature</td>
</tr>
<tr>
<td>Angle of Incidence</td>
<td>Roughness / topography</td>
</tr>
<tr>
<td>Intensity</td>
<td>Oxide layers</td>
</tr>
<tr>
<td></td>
<td>Contamination (dust, dirt, surface defects)</td>
</tr>
</tbody>
</table>

Figure 1 demonstrates some of the complexities of photon absorption by an engineering grade metal surface. Such a surface has a characteristic roughness and
is always covered by an oxide layer. The surface roughness can result in multiple reflections which involve multiple absorption events (Fig. 1b). Surface roughness can also involve the incoming radiation in absorption events at high angles of incidence, close to the Brewster angle. This can result in very high local absorptance levels. The oxide layer may be absorbing in its own right or may be responsible for multiple reflections or wave guiding (Fig. 1c). In addition to these effects the surface may be contaminated with material which has a higher absorptance than the underlying metal.

Fig. 1: Some of the mechanisms which increase the absorptivity of real engineering surfaces: (a) typical cross section of an engineering surface, (b) high (Brewster) angle absorptance and multiple reflections due to surface roughness, (c) multiple reflections within an oxide layer.

All of the above phenomena increase the absorptance of the surface in question and the use of alloying elements will also tend to complicate the situation. For this reason the best way to establish the absorptance of an engineering metal surface is by direct measurement.
It would, of course, be extremely difficult to measure the absorptance of the molten, turbulent surfaces typical of laser welding or cutting. However, it is possible to accurately measure the absorptance of the solid material at room temperature and the values obtained can provide an insight into how the materials behave at elevated temperatures. This can be argued from the fact that for metals, at wavelengths around 1µm, the temperature dependence of absorptance is small. It is well established that metals usually have a so called X-point, a wavelength point or band where the temperature coefficient of absorptance changes sign. The reason for this kind of behaviour lies in the different temperature coefficients for intraband (free-electron) and interband (bound-electron) absorption, the two intrinsic absorption mechanisms prevalent in metals. Intraband absorption increases with temperature, simply because the free electrons gain kinetic energy while the phonon population grows, which increases the electron-phonon collision frequency (ie. the energy conversion efficiency between the light, the free electrons and the lattice). Interband absorption, on the other hand, is much more complicated and depends on the particular band structure of the metal. The general principle is that as the temperature is elevated, the electron Fermi energy distribution function is smoothed out and the absorption bands are broadened. The competition between these two effects leads to the X-point, which for most metals lies at a wavelength near 1µm.

In any case, the absorptance of the solid material as opposed to the molten state is in itself very important for two reasons:

1. During the initiation of any weld or cut the laser is interacting with the solid surface.
2. During welding or cutting the leading edge of the laser beam in many cases interacts with the solid surface of the material.

A comprehensive literature survey with 46 references on experimental absorption measurement methods has been prepared by the present authors. In addition the authors have provided a comprehensive literature survey on the theory and modelling of the absorption mechanisms in metals, with reference to 47 publications.

This paper presents the results of an experimental survey of the room-temperature absorptance of a wide range of as-received commercially available alloys. There are a number of different experimental methods available for
measuring laser absorptance in opaque solids. Some of the more commonly used are; laser calorimetry, gonioreflectometry, integrating sphere or integrating mirror reflectometry and emittance spectroscopy.

In this paper, reflectometry using an integrating sphere was selected, mainly because integrating spheres are commercially available, relatively inexpensive and easy to use while at the same time being very accurate and versatile. By measuring the reflectance $R$ of a sample, we get immediate information regarding the absorptance $A$, since $A = 1 - R$ for opaque solids.

Although all the following measurements were made using an Nd:YLF laser, the results are also valid for Nd:YAG lasers because the wavelengths of the two types of laser (1053nm or 527nm and 1064nm or 532nm respectively) differ by less than 1%.

2. Experimental setup

2.1 Instrumentation

The reflectance measurements were carried out in the laser laboratory at the Mid Sweden University, in Östersund. The laser used as the radiation source was a 1W cw Nd:YLF laser with fundamental wavelength 1053nm, equipped with a SHG (Second Harmonic Generation) crystal to also allow production of green light at 527nm.

The reflectance of the various metals was measured by using an integrating sphere in a double beam setup, as shown in Fig. 2. The 150mm diameter 6-port integrating sphere, has a barium sulphate based coating called Spectraflect, which has high reflective Lambertian properties for wavelengths between 300 and 2400 nm.

The Nd:YLF laser beam was directed onto a rotatable mirror (M2 in Fig. 2) that was flipped between two positions to produce sample and reference beams. The flipping frequency was set to approximately 1 Hz, to ensure mirror stability. Mirrors 3 and 4 (M3 and M4 in Fig. 2) then directed the beams onto the sample and reference, respectively, at an angle of incidence of 8°. As reference a port
plug of Spectraflect was used. After spatial integration by the sphere, the reflected light was then detected by a reverse biased 5.7 mm$^2$ Si photodiode, with a spectral response between 190nm and 1100nm. The voltage signal of the photodiode was finally digitized in an A/D-converter and sampled by a PC.

![Fig. 2: The experimental setup for measuring reflectance.](image)

The laser beam was partially focussed from its original diameter of 10mm to approximately 3mm on the sample surface by using a lens with a diameter of 25mm and a focal length of 150mm (see Fig. 2). The port diameters (see Fig. 2) were 25mm. Samples were cleaned with isopropyl alcohol and dried before measurements were taken. In all cases the samples were checked for differences in reflectance as a function of any directional surface structure. This was done by rotating the sample between measurements. For these samples no correlation between surface structure orientation and reflectance was observed.
2.2 Error correction methods

Several correction methods were applied to the reflectance measurements to improve accuracy and minimize errors. Clarke and Compton⁵ have identified the following main sources of error in integrating sphere reflectometers:

(a) light losses through ports in the sphere wall,
(b) unequal illumination of the sample and standard and changes in throughput when a single beam method is used,
(c) directional dependence of light scattering from the sample and standard surfaces,
(d) errors due to diffraction effects in apertures.
(e) errors due to imperfect diffusion of reflected light from the sphere walls (non-lambertian characteristics) and
(f) baffling.

To minimize these errors the following procedures and correction methods were applied to the measurements:

(a) Light losses:
With the aim of reducing light losses, all ports not in use were closed with port plugs.

(b) Unequal illumination of sample and standard:
The double beam configuration used in these experiments avoids the substitution error, which is often a feature of single beam measurements (and can be as large as 4-5 % according to the manufacturers⁶). The double beam configuration also has the advantage of reducing the influence of possible laser power fluctuations.

(c) Directional dependence of light scattering:
Light behaves slightly different inside the sphere when reflected from a rough surface as compared to a relatively flat one. Light incident on a flat surface is subjected to mirror-like, specular reflection in contrast to rough surfaces where the reflected light is scattered diffusively in many different directions. This produces differences in the number of reflections needed inside the sphere before the light is homogenously scattered, which will lead to detector throughput
changes. This error can be corrected for by using a combination of diffuse (rough) and specular (flat) reflectance standards.

By knowing the sample’s optical diffusivity, $D_s$, which is the fraction of reflected light which is scattered diffusively, the reflectance of the sample, $R_s$, can be obtained by

$$R_s = S_s \left[ (1 - D_s) \frac{R_{s,r}}{S_{s,r}} + D_s \frac{R_{d,r}}{S_{d,r}} \right],$$

(1)

where $S_s$ is the signal measured with the sample, $R_{s,r}$ is the reflectance of the specular standard (mirror), $S_{s,r}$ is the signal measured with the specular standard, $R_{d,r}$ is the reflectance of the diffuse standard and $S_{d,r}$ is the signal measured with the diffuse standard.

The optical diffusivity can be measured by opening the specular exclusion port (or attaching a light trap) and comparing the signal from the sample to a signal from the diffuse standard, see Fig. 3. The optical diffusivity is then given by

$$D_s = 1 - \frac{S_{s,\text{spec}} / S_s}{S_{d,\text{spec}} / S_{d,r}},$$

(2)

where $S_{s,\text{spec}}$ is the signal measured with sample and with specular exclusion port opened and $S_{d,\text{spec}}$ is the signal measured with the diffuse standard and specular exclusion port opened. Using this method, a diffusivity of 100 % was assigned to the diffuse standards and 0 % to the specular mirror standard. This error was empirically seen to be almost negligible for $\lambda=527$ nm, but could amount to more than 1 % in absolute reflectance for $\lambda=1053$ nm.
Fig. 3: Optical diffusivity can be measured by excluding the specular fraction of the reflected light, through an empty port or by fitting a light trap.

(d) Diffraction effects in apertures:

To get a suitable laser spot size on the sample, a lens was put in front of the sample beam entrance port. Due to diffraction, this produces a characteristic halo around the sample and sample port, which causes a slight error to the measurements. Empirically this was shown to be around 0.2 % in absolute reflectance. This error was easily corrected for by measuring the signal with the sample port empty and subtracting this from the signal with the sample in place.

Errors (e) and (f) are due to the specific sphere designs and are not easily corrected for.

Another error was identified in the case of structured samples with contoured surfaces (termed lay in literature on surface properties). According to Roos\(^8\) this error can be substantial if care is not taken in positioning the samples correctly in the sample ports. A contoured surface produces a reflected band of light (termed specular-diffuse radiation by Roos\(^8\)) , which will produce errors if the band crosses the detector (giving too high a reflectance value) or passes through any open beam ports (giving too low a reflectance value). No other corrections than correct positioning was applied in this case.

In experiments such as this it is always important to eliminate the possibility of error as a result of polarisation effects. In this work polarisation of the laser beam was identified as having a trivial effect upon the results because the angle of incidence of the beam was very close to normal (between eight and nine degrees). At these low angles of incidence polarisation effects are minimal.
The measurements were performed in a dark room to minimize the influence of external light sources, such as lamps. Corrections were also made to the current flowing through the photodiode in absence of light. These dark current measurements were performed before each measurement, which were then subtracted from the measured signals.

Using the correction methods outlined above, the reflectance values of all the diffuse and specular standards were seen to be reproducible to at least within 0.3 % in absolute reflectance. The same reliability can be attributed to the following results for the samples investigated.

2.3 Samples and Measurements

Table 2 lists the different non-ferrous samples examined in the survey in terms of their standards identification numbers and their chemical composition. Three types of material groups were studied; aluminium alloys, copper alloys and zinc coatings on mild steel (the substrate having no importance because the light interacts only with the zinc surface). Note that a similar study on mild and stainless steel was performed also, and is under preparation as a separate publication.

Table 2: List of the alloys examined in this survey and their chemical composition

<table>
<thead>
<tr>
<th>No.</th>
<th>Aluminium alloys</th>
<th>Al</th>
<th>Fe</th>
<th>Cu</th>
<th>Zn</th>
<th>Mn</th>
<th>Si</th>
<th>Mg</th>
<th>P</th>
</tr>
</thead>
<tbody>
<tr>
<td>A11</td>
<td>AA1050, Al, cp aluminium</td>
<td>99%</td>
<td>0,4%</td>
<td>*</td>
<td>*</td>
<td>*</td>
<td>0,25%</td>
<td>*</td>
<td>-</td>
</tr>
<tr>
<td>A12, A13</td>
<td>AA 5251, AlMg, NS4</td>
<td>95,5%</td>
<td>0,50%</td>
<td>0,15%</td>
<td>0,15%</td>
<td>0,8%</td>
<td>0,4%</td>
<td>2,0%</td>
<td>-</td>
</tr>
<tr>
<td>A14</td>
<td>AA6082, AlMgSi, T6</td>
<td>96%</td>
<td>0,5%</td>
<td>*</td>
<td>0,2%</td>
<td>0,7%</td>
<td>1,0%</td>
<td>0,9%</td>
<td>-</td>
</tr>
</tbody>
</table>

Copper alloys

| Cu1 | Pure copper, C106 - Cu7n37 | - | - | 99,8% | - | - | - | - | * |
| Cu2, Cu3 | Brass, CZ108 or Cu DHP | - | - | 63% | 37% | - | - | - | - |

Zinc coatings

| Zn1 | Zintec, electroplated | >99% |
| Zn2 | Galvatite, electroplated | >99% |
| Zn3, Zn4 | Hot dipped – liquid coated | >99% |

* trace (<0,1%)

† surface composition
The corresponding technical surfaces for the 11 samples studied are described in Table 3. The roughness values given in this table are supported by SEM and optical microscopy together with profilometry imaging in the results section of this work. In the case of the Cu and Al alloys investigated here, the final cold rolling process gives a directional grain to the surface roughness of these materials. The two electroplated zinc coatings have a fine isotropic grain structure and the hot-dipped zinc coating has a distinctive ‘spangled’ appearance. All the metal surfaces were measured in the “as received” condition.

The samples were laser or mechanically cut from sheets into 30mm squares with thicknesses varying between 2 and 6 mm. For each sample, absorptance measurements were taken at five separate locations which were then averaged and a standard deviation was calculated. Each measurement for every location is in turn an average of over 300 data points.

<table>
<thead>
<tr>
<th>No.</th>
<th>Aluminium alloys</th>
<th>Surface</th>
<th>Al %</th>
<th>Iso-</th>
<th>Sa [μm]</th>
<th>Sq [μm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>A11</td>
<td>AA1050</td>
<td>Cold rolled</td>
<td>99%</td>
<td>N</td>
<td>0,25</td>
<td>0,32</td>
</tr>
<tr>
<td>A12</td>
<td>AA5251</td>
<td>Cold rolled, 3 mm thick</td>
<td>95,5%</td>
<td>N</td>
<td>0,28</td>
<td>0,35</td>
</tr>
<tr>
<td>A13</td>
<td>AA5251</td>
<td>Cold rolled, 6 mm thick</td>
<td>95,5%</td>
<td>N</td>
<td>0,23</td>
<td>0,29</td>
</tr>
<tr>
<td>A14</td>
<td>AA6082</td>
<td>Cold rolled</td>
<td>96%</td>
<td>N</td>
<td>0,30</td>
<td>0,36</td>
</tr>
</tbody>
</table>

| Copper alloys | Cu % | | | |
| Cu1 | Pure Cu | Cold rolled | 99,8% | N | 0,16 | 0,20 |
| Cu2 | Brass | Cold rolled, non-polished | 63% | N | 0,50 | 0,62 |
| Cu3 | Brass | Cold rolled, polished | 63% | N | 0,54 | 0,66 |

| Zinc coatings | Zn % | | | |
| Zn1 | Zintec | Galvanized - electroplated | >99% | Y | 2,34 | 2,77 |
| Zn2 | Galvatite | Galvanized - electroplated | >99% | Y | 0,52 | 0,68 |
| Zn3 | Hot-dip Galv. | Hot-dipped - liquid coated | >99% | Y | 0,33 | 0,43 |
| Zn4 | Hot-dip Galv. | Hot-dipped - liquid coated | >99% | Y | 0,58 | 0,78 |

All integrating sphere measurements were carried out relative to a standard of known reflectance. In these experiments a combination of diffuse and specular reflectance standards were utilized. The diffuse standards had reflectances
between 2 and 99 %. The specular standard consisted of an Al-coated mirror, which was calibrated at the Angström Laboratory in Uppsala, Sweden. It had a reflectance of 97.7 % at 1053nm and 98.9 % at 527nm.

The reliability of the measurement method was regarded as satisfactory. The variance $\sigma$ of the measured $A$ values was low. In 15 of 22 cases (11 material samples, 2 wavelengths) the variance of the five measurements taken for each sample was less than 2.0%, only in one case (Zn3, green) was the variance higher than 4% (namely 6.6 %).

### 3. Results and discussion

#### 3.1 General

Figures 4-6 for the Al-alloys, Figs. 8-9 for the Cu-alloys and Figs. 11-13 for the Zn-coatings, present the optical microscopy (OM), Scanning Electron Microscope (SEM), profilometry and absorptance results obtained in this survey. In each figure, the magnification used is 100 times for the optical microscopy pictures, 2000 times for the SEM photos and 5 times for the 3-D optical profilometry scans. The surface was measured with a Wykon NT1100 optical profiler, achieving a horizontal resolution of 6.8 $\mu$m and a vertical resolution in the nm-range. In the table below the photos the average absorptance $A$ and standard deviation $\sigma$ are given for both wavelengths as well as the results of the optical profilometry measurements. $S_a$ and $S_q$ are the average roughness and root mean square (rms) roughness evaluated over the complete 3D surface respectively. If the surface elevation is given by $Z(x,y)$, $S_a$ and $S_q$ are evaluated mathematically as follows (with surface area $A$):

$$S_a = \left[ \int_A |Z(x,y)| \, dx \, dy \right]$$

$$S_q = \left( \int_A (Z(x,y))^2 \, dx \, dy \right)^{1/2}$$

(3)

Figures 7, 10, 14 show the measured absorptance plotted as a function of the surface roughness for Al-, Cu- and Zn-alloys, respectively.
3.2 Aluminium alloys

| \( \lambda = 1053 \text{nm} \) | \( \lambda = 532 \text{nm} \) |
| A | \( \sigma \) | A | \( \sigma \) |
| 15.7% | 1.3% | 32.4% | 2.8% |

\[ \text{Sa[\mu m]} = 0.25 \quad \text{Sq[\mu m]} = 0.32 \]

Fig. 4: Al1: AA1050, commercially pure aluminium

| \( \lambda = 1053 \text{nm} \) | \( \lambda = 532 \text{nm} \) |
| A | \( \sigma \) | A | \( \sigma \) |
| 20.7% | 0.7% | 42.6% | 1.2% |

\[ \text{Sa[\mu m]} = 0.28 \quad \text{Sq[\mu m]} = 0.35 \]

Fig. 5: AA5251, AlMg: (a) Al2: AlMg, 3mm thick
Paper 4: The Absorptance of Non-Ferrous Alloys to Nd:YLF and Nd:YAG Laser Light at Room Temperature

Fig. 5: AA5251, AlMg: (b) Al3: AlMg, 6mm thick

<table>
<thead>
<tr>
<th>$\lambda$=1053nm</th>
<th>$\lambda$=532nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A$</td>
<td>$\sigma$</td>
</tr>
<tr>
<td>26,9%</td>
<td>1,3%</td>
</tr>
</tbody>
</table>

$S_a[\mu m]$ | 0,23 |
$S_q[\mu m]$ | 0,29 |

Fig. 6: Al4: AA6082, AlMgSi

<table>
<thead>
<tr>
<th>$\lambda$=1053nm</th>
<th>$\lambda$=532nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A$</td>
<td>$\sigma$</td>
</tr>
<tr>
<td>23,9%</td>
<td>1,3%</td>
</tr>
</tbody>
</table>

$S_a[\mu m]$ | 0,30 |
$S_q[\mu m]$ | 0,36 |
The measured absorptance as a function of the roughness is plotted in Fig. 7 for the four aluminium samples and for the two wavelengths used. It is clear from this figure that there is no clear correlation between surface roughness and absorptance.

Previously published figures put the absorptances of aluminium as 4.8% at 1053nm and 7.6% at 527nm. The figure we measured for our commercially pure (99% Al, sample Al1) aluminium sheet for 1053nm was ~ 16% and this threefold increase is clearly due to the oxidised and unpolished nature of our sample. The average figure of ~ 32% we obtained for 527nm is surprisingly higher than the previously published value but the accuracy of this new value is supported by the fact that all the results for 527nm were considerably higher than for 1053nm over the whole range of aluminium samples studied.

The increased absorption for the AA5XXX (samples Al2, Al3) and AA6XXX (sample Al4) grades compared with the ‘pure’ aluminium sample Al1 is probably related to the increase in alloying elements in these grades.

It is interesting to note that there is an absorptance peak for aluminium at a wavelength of 800nm due to the atomic band structure of the material. This may, in the future, have positive consequences for aluminium processing with high power diode lasers in this wavelength region.
3.3 Copper alloys

<table>
<thead>
<tr>
<th>$\lambda=1053\text{nm}$</th>
<th>$\lambda=532\text{nm}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A$</td>
<td>$\sigma$</td>
</tr>
<tr>
<td>5,6%</td>
<td>1,0%</td>
</tr>
</tbody>
</table>

$\text{Sa[}\mu\text{m}]$ 0,16 $\text{Sq[}\mu\text{m}]$ 0,20

Fig. 8: Cu1: commercially pure copper

<table>
<thead>
<tr>
<th>$\lambda=1053\text{nm}$</th>
<th>$\lambda=532\text{nm}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A$</td>
<td>$\sigma$</td>
</tr>
<tr>
<td>10,2%</td>
<td>1,0%</td>
</tr>
</tbody>
</table>

$\text{Sa[}\mu\text{m}]$ 0,50 $\text{Sq[}\mu\text{m}]$ 0,62

Fig. 9: Brass: (a) Cu2: brass, non-polished
The measured absorptance as a function of the roughness is plotted in Fig. 10 for the three copper-based samples and for the two wavelengths applied. Once more there is no clear correlation between roughness and absorptance.

Once again the ‘pure’ copper has a higher absorptance (6% at 1053nm and 58% at 527nm) than the previously published values (e.g. 2.4% at 1053nm and 38.2% at 527nm). The explanation for the quite large difference in absorptance...
between the two wavelengths lies in the details of the band structure of copper, showing a typical interband absorption edge at $\lambda \sim 550\text{nm}$. The addition of 37% of zinc to the copper (to produce brass) increases the absorptance at 1053nm but decreases it at 527nm for reasons which will be discussed below for the zinc coatings.

3.4 Zinc coatings

![Zinc coatings](image)

<table>
<thead>
<tr>
<th>$\lambda$=1053nm</th>
<th>$\lambda$=532nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>$\sigma$</td>
</tr>
<tr>
<td>77,6%</td>
<td>0,2%</td>
</tr>
<tr>
<td>A</td>
<td>$\sigma$</td>
</tr>
<tr>
<td>60,5%</td>
<td>0,3%</td>
</tr>
</tbody>
</table>

$S_a[\mu m]$ 2,34 $S_q[\mu m]$ 2,77

Fig. 11: ZnI: Zintec
Paper 4: The Absorptance of Non-Ferrous Alloys to Nd:YLF and Nd:YAG Laser Light at Room Temperature

\[
\begin{array}{c|c|c|c|c}
\lambda &=& 1053\text{nm} & \lambda &=& 532\text{nm} \\
A & \sigma & A & \sigma \\
\hline
53.8\% & 1.9\% & 33.4\% & 1.4\% \\
\end{array}
\]

\[
\begin{array}{c|c}
\text{Sa[\mu m]} & 0.52 \\
\text{Sq[\mu m]} & 0.68
\end{array}
\]

Fig. 12: Zn2: Galvatite

\[
\begin{array}{c|c|c|c|c}
\lambda &=& 1053\text{nm} & \lambda &=& 532\text{nm} \\
A & \sigma & A & \sigma \\
\hline
63.1\% & 3.7\% & 48.7\% & 3.9\%
\end{array}
\]

\[
\begin{array}{c|c}
\text{Sa[\mu m]} & 0.33 \\
\text{Sq[\mu m]} & 0.43
\end{array}
\]

Fig. 13: Hot-dip galvanized (spangled): (a) Zn3: sample I
The measured absorptance as a function of the roughness is plotted in Fig. 14 for the four zinc coated samples and for the two wavelengths applied. Once again there is no clear correlation between roughness and absorptance at the lower roughness values. There is, however, a noticeable increase in absorptance for the samples with roughness values above two microns.

![Graph showing absorptance as a function of roughness for zinc-coated samples at two wavelengths.](image)

**Fig. 14: Absorptance as a function of the surface roughness for the four zinc coating samples studied (ref. values in Figs. 11-13)**

Zinc coatings come in a wide range of finishes and three of the most commonly available have been tested here. Of course, we are no longer dealing
with a mild steel surface in this case and the laser is interacting only with the zinc. It can be observed from Fig. 14 that in every case except where zinc is involved, the absorptance of the metals at 527nm is higher than it is at 1053nm. In the case of brass (37% Zn) and the zinc coated mild steel this trend is reversed. The reason for this lies in the band structure and absorptance spectra of zinc, the latter of which has a sharp peak at $\lambda \sim 1080\text{nm}^{10}$.

The absorptance of the Zintec sample is considerably higher than that of the Galvatite or galvanized (spangled) samples. The SEM photographs and surface roughness measurements indicate that the reason behind this higher absorptance is the considerably rougher surface texture of the Zintec (The Zintec sample gave roughness values which were more than three times greater than any other sample in this whole series of tests).

### 3.5 Summary of the absorptance measurements

Table 4 provides a summary of the absorptance and reflectance values obtained for all the various alloys investigated. All these measurements demonstrated that engineering grade metal surfaces have absorptances which are considerably higher than those published for pure, polished surfaces. In the case of copper and aluminium alloys the absorptance is higher for a wavelength of 527nm than for 1053nm. For Zinc this trend was reversed.

All the absorptance values are shown in Fig. 15 as a function of the surface roughness with the exception of the Zintec sample.

### Table 4. A summary of the absorptance and reflectance measurements

<table>
<thead>
<tr>
<th>Material</th>
<th>$\lambda=1053\text{nm}$</th>
<th>$\lambda=527\text{nm}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Absorptance</td>
<td>Reflectance</td>
</tr>
<tr>
<td>Al1: Commercially pure Al</td>
<td>16%</td>
<td>84%</td>
</tr>
<tr>
<td>Al2,3: AA5251, AlMg (average)</td>
<td>24%</td>
<td>76%</td>
</tr>
<tr>
<td>Al4: AA6082, AlMgSi</td>
<td>24%</td>
<td>76%</td>
</tr>
<tr>
<td>Cu1: Commercially pure Cu</td>
<td>6%</td>
<td>94%</td>
</tr>
<tr>
<td>Cu2,3: Brass, CuZn37 (average)</td>
<td>12%</td>
<td>88%</td>
</tr>
<tr>
<td>Zn1-4: Zinc coatings (average)</td>
<td>64%</td>
<td>36%</td>
</tr>
</tbody>
</table>
It is clear from Fig. 15 that if the surface roughness of the sample is lower than $S_a = 0.6$ then there is no clear correlation between roughness and absorptance. On the other hand it appears that absorptance does increase if the surface roughness measurement $S_a$ has a value above 2.3 ($\text{Zintec; } S_a: 2.34, A: 77.6 \text{ for } 1053\text{nm}, 60.5 \text{ for } 532\text{nm}$) this phenomenon is the subject of an on-going investigation by the present authors.

### 4. Conclusions

Reliable absorptance results have been measured for a range of non-ferrous alloys for Nd:YAG and Nd:YLF lasers at their infrared and green wavelengths. These results have, as expected, been found to differ substantially from existing published data for pure, polished metals.

The increases in absorptance for engineering grade surfaces can be broadly attributed to surface oxides, roughness and alloying ingredients but further study is necessary to isolate the contribution of each factor.

Over the range of surface roughness from approximately $S_a 0.15 – 0.60$ there was no clear correlation between surface roughness and absorptance. However,
one much rougher sample ($S_a: 2.34$) indicated that outside this range there could be a roughness-absorptance correlation.

For the aluminium and copper alloys studied here the absorptance was higher for the green wavelength than for the infra-red. This trend was reversed for the zinc surfaces as a result of the band structure and absorption spectra of zinc.

**Bibliography**


A Ray-Tracing Analysis of the Absorption of Light by Smooth and Rough Metal Surfaces

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Paper 5: A Ray-Tracing Analysis of the Absorption of Light by Smooth and Rough Metal Surfaces
A Ray-Tracing Analysis of the Absorption of Light by Smooth and Rough Metal Surfaces

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Abstract

Ray tracing has been employed to investigate the absorption of light by smooth and random rough metal surfaces. For normally incident light the absorptance of the surface increases with surface roughness. However, for light incident at a tangent to the surface the absorptance – surface roughness relationship is more complex. For example, in certain cases the absorptance can rise, fall and rise again as the surface roughness increases. In this paper this complex absorptance – roughness relationship is defined and explained. The wavelengths of the light chosen for this study correspond to the primary and secondary output wavelengths of Nd:YAG lasers.

Keywords; Absorptance, Light, Surface roughness, Angle of Incidence, Laser.

1. Introduction

An understanding of the various laser absorption mechanisms is of vital importance to the study of laser processing of metals. Laser absorption depends on a number of different parameters, involving both laser and metal properties [1]. Due to the very short penetration depths in metals for infrared and visible light (in the order of tens of nanometers), absorption is very much a surface phenomenon and depends very strongly upon the surface properties of the metal such as the roughness and texture and the existence and structure of oxide layers.

In this paper, Monte Carlo simulations are used to numerically calculate the absorptance of one-dimensional Gaussian random rough metal surfaces with various RMS slopes (roughness) using the geometric optics (GO) approximation. The GO approximation is a commonly used approximation in rough surface
scattering theory [2-6], due to its relatively simple numerical implementation and
the reduced computational requirements compared to the numerical integration
techniques needed for rigorous electromagnetic wave analysis [7-11]. The
approximation is a ray-tracing approach, where energy bundles are traced
throughout their interactions with the surface until they leave it. The
approximation is regarded as valid when the normalized correlation length, \( r / \lambda \), as
well as the normalized rms roughness, \( \sigma / \lambda \), are larger than unity (\( \lambda \) being the
wavelength of the light involved). Tang et al. [2] have shown that the GO
approximation corresponds well to the exact wave-theoretical methods for
\( \sigma \cos(\theta_0) / \lambda > 0.17 \) (\( \theta_0 \) being the angle of incidence, \( \theta_0 = 0^\circ \) meaning normal
incidence) and for surfaces with \( \sigma / r < 2.0 \). Fig. 1 and Table 1 show the regions of
validity (the RMS heights and the correlation lengths) for the scattering and
absorptance results presented in this paper. Two wavelengths of importance to
laser processing with metals were used; \( \lambda = 1064nm \) corresponding to the
fundamental wavelength of the Nd:YAG laser and \( \lambda = 532nm \) corresponding to
second harmonic generated light for the same laser source. Four discrete angles of
incidence were investigated; \( 0^\circ, 30^\circ, 60^\circ \) and \( 80^\circ \).

**Figure 1.** Plots of the regions of validity for the geometric optics approximation
for the wavelengths (a) \( \lambda = 1064nm \) and (b) \( \lambda = 532nm \) and the angles of
incidence (\( 0^\circ, 30^\circ, 60^\circ \) and \( 80^\circ \)) used in the simulations [2].
Table 1. Region of validity of the simulation results [2].

<table>
<thead>
<tr>
<th>θ₀, [°]</th>
<th>σ [µm]</th>
<th>τ [µm]</th>
<th>θ₀, [°]</th>
<th>σ [µm]</th>
<th>τ [µm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>0°</td>
<td>≥ 0.20</td>
<td>≥ α/2</td>
<td>0°</td>
<td>≥ 0.40</td>
<td>≥ α/2</td>
</tr>
<tr>
<td>30°</td>
<td>≥ 0.23</td>
<td>≥ α/2</td>
<td>30°</td>
<td>≥ 0.47</td>
<td>≥ α/2</td>
</tr>
<tr>
<td>60°</td>
<td>≥ 0.40</td>
<td>≥ α/2</td>
<td>60°</td>
<td>≥ 0.81</td>
<td>≥ α/2</td>
</tr>
<tr>
<td>80°</td>
<td>≥ 1.16</td>
<td>≥ α/2</td>
<td>80°</td>
<td>≥ 2.33</td>
<td>≥ α/2</td>
</tr>
</tbody>
</table>

2. Theory and Analysis

2.1 Radiative properties

When light is incident on a material surface the energy will either be reflected, transmitted or absorbed. To describe and quantify the angular distribution of the reflected or scattered light, the bidirectional reflection distribution function (BRDF) is defined as [12]

\[
\rho_\lambda^s(\Omega_i, \Omega_s) = \frac{\pi}{\cos(\theta_\lambda)} \left( \frac{d\Phi_s}{d\Omega_s} \right) \left( \frac{d\Phi_i}{d\Omega_i} \right),
\]

where \(\theta_\lambda\) is the angle of scattered light, \(\Phi_i\) and \(\Phi_s\) are the incident and scattered radiant powers and \(\Omega_i\) and \(\Omega_s\) are the incident and scattered solid angles, respectively. A similar definition can, of course, also be made for the bidirectional transmittance. The bidirectional reflectance is a fundamental quantity in radiative theory, from which all reflective properties can be derived and is the quantity most often derived in scattering models.

Integration of the bidirectional reflectance over the entire hemisphere yields the directional-hemispherical reflectance [12]

\[
\rho_\lambda(\Omega_i) = \frac{1}{\pi} \int_{-\pi}^{\pi} \rho_\lambda^s(\Omega_i, \Omega_s) \cos(\theta_\lambda) d\Omega_s.
\]

For opaque materials such as metals (except when dealing with extremely thin films), all non-reflected light can be regarded as absorbed and by conservation of energy the spectral directional absorptance can be found from
Since there exists a direct correspondence between emittance and absorptance from Kirchhoff’s law in thermodynamics, BRDF models in scattering theory can also be used to quantify the emissive properties of a material body.

2.2 Gaussian random rough surfaces

In light scattering theory there are two main classes of models used to describe rough surfaces; surfaces of precisely given profiles (sinusoidal, saw-tooth, rectangular, etc.) and surfaces with random irregularities. These classes are named deterministic and random rough surfaces, respectively, and usually differ in their general treatment and in their applications. The treatment is a lot simpler for the first class, but most naturally occurring and man-made surfaces fall in the latter category.

A random rough surface, given by the function \( z = \zeta(x, y) \), is described in statistical terms using two distribution functions, the height probability distribution, \( p(\zeta(x, y)) \), and the autocovariance function, \( C(\tau) \). The height probability distribution describes the surface height deviation from a certain mean reference level (usually \( <\zeta(x, y)> = 0 \)) and the autocovariance function describes the variance of these heights laterally along the surface (i.e. the crowdedness of the hills and valleys – see Fig. 2). A commonly used model is to approximate the height probability distribution as a Gaussian. Thomas [13] has demonstrated that this Gaussian assumption is valid as long as the height at any point on the surface is not produced by a single one-off event (which is true of most surfaces). The Gaussian height probability assumption gives us

\[
A'_\lambda(\Omega_v) = 1 - \rho'_\lambda(\Omega_v). \tag{3}
\]

where \( \sigma \) is the root mean square (RMS) height (which equals the standard deviation). The autocovariance function can be defined in various ways. It is most common to use either a Gaussian or an exponential function. In this paper we assume it is described by a Gaussian so that

\[
p(\zeta) = \frac{1}{\sigma \sqrt{2\pi}} e^{-\frac{\zeta^2}{2\sigma^2}}, \tag{4}
\]
\[ C(\tau) = \langle \zeta(x_1)\zeta(x_2) \rangle = \sigma^2 \exp \left( -\frac{|x_1 - x_2|^2}{\tau^2} \right) \], \quad (5) 

where \( x_1 \) and \( x_2 \) are two different points along the surface and \( \tau \) is the correlation length (see Fig. 2 for two surfaces with the same rms height (\( \sigma = 1 \mu m \)) but with different correlation lengths). For random rough surfaces where both the rms height distribution function and the autocovariance function are given by Gaussians (as above), it can be shown [14] that the RMS slope will be given by \( \sqrt{2} \sigma / \tau \). For simplicity the factor of \( \sqrt{2} \) will be omitted in the following treatment and when we speak of the RMS slope or slope we will only use the ratio \( \sigma / \tau \).

![Figure 2](image_url)

**Figure 2.** Two Gaussian random rough surfaces with the same RMS height (\( \sigma = 1 \mu m \)) but with different correlation lengths. The profile in (a) has correlation length \( \tau = 10 \mu m \) while the profile shown in (b) has \( \tau = 1 \mu m \).

For simulation purposes, one-dimensional Gaussian random rough surfaces of this kind can easily be generated using the Spectrum method by Thorsos [15]. The method starts with the Fourier transform of the autocovariance function in Eq. (5), which yields the power spectral density function:

\[ W(k_x) = \mathfrak{F}[C(x)](k_x) = \frac{\sigma^2 \tau^2}{4\pi} \exp \left( -\frac{k_x^2 \tau^2}{4} \right) \], \quad (6) 

where \( \mathfrak{F} \) denotes the Fourier transform and \( k_x \) is the variable in the spatial frequency domain.
This power spectral density function is then related to the discrete Fourier transform of the height function through the following relationship

\[
F(k_{x_n}) = 2\pi L \sqrt{W(k_{x_n})} \begin{cases} 
N(0,1) + iN(0,1) & m \neq 0, \frac{N}{2} \\
\sqrt{2} & m = 0, \frac{N}{2}
\end{cases},
\]

where \( k_{x_n} = 2\pi m/L \), \( L \) is the length of the rough surface and \( N(0,1) \) is a zero-mean, unit-variance normal distribution. The height function, \( z = \zeta(x) \), is then obtained by taking the discrete inverse Fourier transform of \( F(k_{x_n}) \):

\[
z = \zeta(x) = \frac{1}{L^2} \sum_{m=-N/2}^{N/2-1} F(k_{x_n}) \exp(ik_{x_n}x),
\]

where for negative values of \( m \) it is necessary to use the complex conjugation of \( F(k_{x_n}) \) to ensure that \( f(x) \) is real. Fig. 3 shows one realization of a random rough surface using the method described above, together with the height distribution function and the normalized autocovariance function (also known as the correlation function) calculated from the generated data.
Figure 3. (a) shows a realization of a Gaussian random rough surface using the Spectrum Method with $\sigma = \tau = 1\mu m$. (b) displays the normalized autocovariance function from the surface data compared to an exact Gaussian, while (c) plots the surface height distribution in comparison with an exact Gaussian.

2.3 Geometric Optics Approximation

The geometric optics approximation is an approximation to the exact numerical integration methods available from electromagnetic wave theory. As an approximation it is, of course, limited to certain roughness parameters (as depicted in Fig. 1), but contrary to many other approximate methods, like the Kirchhoff approximation [15-18], the small perturbation theory [17-19], the phase perturbation theory [20-21] and the small slope approximation [22-23], it is a method that naturally incorporates both shadowing and multiple scattering (see
Fig. 4. It is also easily implemented for computational purposes, using intuitive geometrical arguments.

![Figure 4. (a) depicts shadowing where a part of the surface is not “seen” from the incident direction, while (b) is an illustration of multiple scattering (second-order in this case).](image)

The GO approximation is a ray-tracing approach where the incident energy bundle is traced through its interactions with the surface until it leaves the surface. The surface is approximated as locally smooth so that each scattering event on the surface is treated as a specular reflection (this is known as the Fresnel approximation).

The successive steps of the ray-tracing algorithm will now be explained.

STEP 1. The first step consists of generating the rough surface, using for instance the Spectrum method outlined in section 2.2. The slopes and normals of all points are calculated and the angle of incidence of the light \( \theta_0 \) is selected.

STEP 2. A first reflection point is then chosen. The number of first reflection points can be as large as the number of surface points and can be distributed randomly or equidistant along the surface.

STEP 3. For the first reflection point chosen, a strike ability test is performed to check if the reflection point can be struck by the incident ray. The tangent angle of the reflection point is compared to the angle of the incident ray (see Fig. 5 for the scattering geometry). If the ray is
incident at an angle larger than the tangent angle of the reflection point, another first reflection point is chosen.

**STEP 4.** It is then tested whether the first reflection point is shadowed due to other irregularities along the surface. This test is of most importance for larger angles of incidence (for normal incidence it is unnecessary). The angle of the incident ray is compared to the angles of the ratio of differential changes between the first reflection point and any other point on the surface (all surface points to the left if the ray is incident from the left and all points to the right if the incident ray approaches from the right). If the ray is incident at an angle larger than the angle of any ratio of differential changes, the reflection point is shadowed and another first reflection point is selected.

![Rough surface scattering geometry](image)

**Figure 5.** Rough surface scattering geometry where $\vec{i}$ and $\vec{o}$ are the incident and scattered rays, respectively, and $\vec{n}$ is the surface normal. $\theta_i$ and $\theta_s$ are the angles of incidence and scattering, respectively (notice their definition of positive direction).

**STEP 5.** If the first reflection point is strikeable and non-shadowed, the scattered ray is then calculated using the surface normal and Snell’s reflection law:
where \( \bar{i} \) and \( \bar{o} \) are the incident and scattered rays, respectively, and \( \bar{n} \) is the surface normal (see Fig. 5).

The energy of the incident ray, \( E_i \), is calculated using

\[
E_i = \cos(\theta_{loc}) \frac{L}{N \cos(\alpha)},
\]

where \( \theta_{loc} = \theta_0 + \alpha \) is the local angle of incidence (the angle between the incident ray and the surface normal), \( \alpha \) is the tangent angle, \( L \) is the surface length and \( N \) is the number of surface points. In this equation, the second factor represents the locally smooth segment area, while the first factor gives the area projected normal to the incident ray. In this case we are assuming that the energy incident is equally distributed along the surface. It is also possible to use other types of distributions (e.g. a Gaussian).

The energy of the scattered ray, \( E_s \), is then found through a multiplication of \( E_i \) with the Fresnel coefficient

\[
E_s = F(n, \kappa, \theta_{loc}) E_i,
\]

where the Fresnel coefficient \([11]\) is a function of the local angle of incidence as well as the optical constants of the material; the refractive index \( n \) and the extinction coefficient \( \kappa \). The amount of energy absorbed in the scattering event is then simply the difference in energy between incident and scattered rays.

STEP 6. The existence and position of any possible second reflection point is then determined by comparing the tangent of the scattered ray with the local topology (similar to the shadowing test in step 4).
STEP 7. If a new reflection point is found, the scattered ray is transformed into an incident ray and the scattering process with successive reflections is continued until the energy leaves the surface. The amount of absorbed energy is then found by subtracting the incident energy (Eq. 10) with the energy scattered off the surface.

A new first reflection point is then selected and the ray-tracing process continues until all first reflection points have been accounted for.

STEP 8. When all first reflection points have been treated, the scattered energy at each scattering angle is divided by the total amount of energy incident on the surface, which is the differential reflection coefficient. The bi-directional reflection distribution function is then found by dividing the differential reflection coefficient by the cosine of the scattering angle and the size of the scattering region \( d\Omega \), in radians (dependent on the angular resolution required), and multiplying by \( \pi \) (as in Eq. (1)). In principle the absorptance then can be calculated using Eq. (2) and (3), i.e. through an integration of the BRDF and using energy conservation, but it is more conveniently found by dividing the total amount of absorbed light (from all scattered rays) by the total amount of incident energy.

STEP 9. To get statistically accurate results, the process outlined above is then repeated for several realizations of the rough surface (with the same values for \( \sigma \) and \( \tau \)).

STEP 10. Finally the results are averaged.

3. Results and Discussion

3.1 Scattering results

Fig. 6 displays some scattering plots produced by the ray tracing algorithm for normal incidence on pure copper surfaces with a few different values for the slope ratio \( \sigma/\tau \) at the fundamental Nd:YAG wavelength \( \lambda = 1064\text{nm} \) (the complex index of refraction \( n + ik \) was found from the SOPRA database [24]). These plots were
numerically calculated using the average simulation results for 30 surface realizations (each surface was defined by 75000 points using 10000 first reflection points). In the plot the differential reflection coefficient, defined as $\rho_s^* \cos(\theta_s)$, is displayed vs. the angle of scattering $\theta_s$, instead of the more common BRDF function, $\rho_s^*$, as defined in Eq. (1). The reason for this is that the differential reflection coefficient is more closely related to the amount of scattered light energy for the different directions.

For small RMS slopes the scattering is typically specular in nature, where for $\sigma/\tau = 0.01$ the scattered light distribution resembles a delta function around $\theta_s = 0^\circ$. For $\sigma/\tau = 0.1$ light is still specularly scattered but with broadening due to small changes of the local angles of incidence. As the slope increases further, eg. at $\sigma/\tau = 0.3$, a more diffuse scattering behaviour is observed where light is spread across all scattering angles. For the very rough surfaces, $\sigma/\tau = 1$ and 2, backscattering (also known as retroreflection) can be seen where a significant

**Figure 6.** Surface scattering plots for a few different RMS slopes (roughness levels). The differential reflection coefficient, $\rho_s^* \cos(\theta_s)$, is shown vs. the scattering angle $\theta_s$ for light at normal incidence.
amount of light is scattered back in approximately the incident direction (see Fig. 4b).

Backscattering effects are even more evident when considering the scattering of light beams with non-normal incidence, as in Figs. 7 and 8, which depict the results of the geometric optics approximation for light incident on rough surfaces with angles $\theta_0 = 30^\circ$ and $60^\circ$, respectively. In both figures two peaks are visible for the surfaces with higher slopes, one for the regular specular direction and one in the backscattered direction. An off-specular peak can also be seen, especially for $\theta_0 = 60^\circ$, where the maximum amount of scattered light is found at an angle smaller than the expected specular direction. Both the phenomena of off-specular peaks and backscattering have been verified experimentally by independent investigators (eg. [25-30]).

Figure 7. Scattering plots for copper surfaces with various RMS slopes (roughness levels) for light incident with an angle $\theta_0 = 30^\circ$. 

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3.2 Absorptance results

3.2.1 Normal incidence

Figs. 9a and 9b show absorptance results for light normally incident on rough copper surfaces with slopes varying between 0.01 and 2. The plots were generated for two sets of values for the complex refractive index representing copper at the Nd:YAG wavelengths of 1064 nm and 532 nm. All points in the plots are numerical results for 30 surface realizations for each slope, using 10000 first reflection points on surfaces defined by at least 50000 points. The standard deviation of the absorptance results presented in Fig. 9 and subsequent figures was less than 2% in all cases.
Figure 9. Absorptance is plotted as a function of the RMS slope $\sigma/\tau$ (roughness) for light normally incident on rough copper surfaces at (a) $\lambda = 1064\text{ nm}$ and (b) $\lambda = 532\text{ nm}$. Optical constants taken from the SOPRA database [24].

The increase in ‘$A$’ at a RMS slope of $\sigma/\tau \approx 0.2$ correlates with the threshold for multiple scattering as shown in Fig. 10, which shows the average number of scattering events as a function of the RMS slope (this function is material independent as it is only dependent upon the surface topography).

Figure 10. The average number of scattering points per incident ray as a function of the RMS slope (roughness) $\sigma/\tau$ for normal incidence.
This threshold is also demonstrated in Figs. 11a and 11b, which are plots of the scattered energy separated into first, second and higher order scattering for the two sets of optical constants used to generate Figs. 9a and 9b. (Rays reflected only once by the surface before leaving constitute first order scattering, rays reflected twice are second order and so on.) It can be seen, in both Figs. 11a and 11b, that second order scattering starts gaining influence at about \( \sigma / \tau \approx 0.2 \) and third order scattering begins at \( \sigma / \tau \approx 0.5 \).

![Figure 11. Scattered energy distributed in different orders and normalized to total reflectance, for copper at (a) \( \lambda = 1064 \text{ nm} \) and at (b) \( \lambda = 532 \text{ nm} \).](image)

In Fig. 12 values of absorptance as a function of roughness are given after being normalized to the value for a flat, smooth surface (the Fresnel absorptance at normal incidence) for several metals at \( \lambda = 532 \text{ nm} \) and \( \lambda = 1064 \text{ nm} \). Fig. 12 demonstrates that the increase in absorptance as a function of roughness is most pronounced for high reflectivity wavelength-material combinations such as Aluminium or Copper at 1064 nm.

Although an increase in roughness (above the threshold value of 0.2) always results in an increase in absorptance, the effect is reduced for higher absorptivity materials. This is simply a consequence of the ‘diminishing returns’ which are to be expected when comparing multiple reflections from high and low absorptivity interactions. For example; If the absorptivity of a surface is 50% then the first scattering event will result in an absorption of 50%, and the second will result in absorption of a further 25% (of the original energy), taking the total absorption...
level up to 75%. If, on the other hand, the absorptivity of the material is only 10%, then the absorption figures from the primary and secondary scattering events will be 10% and 9% - making a total of 19%. This increase from 10% to 19% is considerably bigger than the increase from 50% to 75% and this is the principle demonstrated in Fig. 12.

The practical consequence of the above is that roughening the surface of a high reflectivity metal may increase its absorptance to Nd:YAG laser light by several hundred percent. The effect of roughening the surface of low reflectivity metals will not be so dramatic.

**Figure 12.** Ratios of the absorptances of rough and smooth surfaces for normally incident light shown as a function of slope (roughness) for the metals listed in the legend (the number after the atomic symbol indicates the wavelength of the light involved). The smooth surface absorptance at normal incidence is given in parentheses.
3.2.2 Non-normal incidence, angle of incidence dependency

In some laser processing applications, such as surface treatment or laser cleaning, it is assumed that process efficiency can be increased by utilizing the Brewster maximum of the Fresnel absorptance, an angle usually situated in the range of 70°-90° (see Figs. 13 and 14 for two examples).

The Fresnel absorptance is only an approximation for perfectly smooth surfaces ($\sigma/\tau = 0$) and this dependency on angle of incidence (AOI) cannot be expected for rougher surfaces. Figs. 13, 14 and 15 show numerical results for the absorptance as a function of AOI for the valid slopes regime of the GO approximation. Fig. 13 and 14 shows results for copper and aluminium at $\lambda = 1064$ nm, respectively, while Fig. 15 shows for copper at $\lambda = 532$ nm. It can clearly be seen that the AOI dependence resembles the Fresnel curve only when $\sigma/\tau \ll 0.1$. As the slope increases up to $\sigma/\tau \approx 0.3$, the absorptance curve is flattened and both the AOI dependency and the Brewster maximum disappear (as is seen in Fig. 13a and Fig. 14a). As the slope is further increased a maximum close to the normal angle of incidence is established (see Figs. 13b, 14b and 15b). For copper at $\lambda = 532$ nm there is no pronounced Brewster maximum, as is seen in Fig. 15, but the situation is otherwise similar.
Figure 13. The absorptance vs. angle of incidence for copper at $\lambda = 1064$ nm, for a range of different slopes (roughness). In (a) $A(\theta)$ is shown for the range of small and medium slopes where $0.01 \leq \sigma/\tau \leq 0.5$, while in (b) it is given for the range of larger slopes where $0.5 \leq \sigma/\tau \leq 2.0$. The Fresnel absorptance curve has been included for reference.
Figure 14. The absorptance vs. angle of incidence for aluminium at $\lambda = 1064$ nm, for a range of different slopes (roughness). In (a) $A(\theta)$ is shown for the range of small and medium slopes where $0.01 \leq \sigma/\tau \leq 0.5$, while in (b) it is given for the range of larger slopes where $0.5 \leq \sigma/\tau \leq 2.0$. The Fresnel absorptance curve has been included for reference.
Figure 15. The absorptance vs. angle of incidence for copper at $\lambda = 532$ nm, for a range of different slopes (roughness). In (a) $A(\theta)$ is shown for the range of small and medium slopes where $0.01 \leq \sigma/\tau \leq 0.5$, while in (b) it is given for the range of larger slopes where $0.5 \leq \sigma/\tau \leq 2.0$. The Fresnel absorptance curve has been included for reference.
Fig. 12 demonstrates that, at normal incidence, an increase in roughness has the effect of increasing the absorptance of a surface. Figs. 13, 14 and 15 show that this relationship does not hold true at high angles of incidence. This point is supported by a comparison of Figs. 16 and 17 which show the same data as Fig. 12 but for angles of incidence of 30° and 60°, respectively. Fig. 16 follows the same trends as Fig. 12 but the data presented in Fig. 17 reveals that, at high angles of incidence (60° in this case), the relationship between surface roughness and absorptance is complex. Many of the metals plotted in Fig. 17 (e.g.; Al, Cu) demonstrate a decrease in absorptance with increasing roughness between the slope values of 0.2 and 0.7. Above a value of 0.7 there is a general increase in absorptance with roughness, but the lines for the different metals have differing inclinations and therefore intersect each other.

![Figure 16](image)

**Figure 16.** Ratios of the absorptances of rough and smooth surfaces for \( \theta_0 = 30^\circ \) shown as a function of slope (roughness) for the metals listed in the legend (the number after the atomic symbol represents the wavelength of the light involved). The smooth surface absorptances at incidence angle \( \theta_0 = 30^\circ \) is given in parentheses.
Figure 17. Ratios of the absorptances of rough and smooth surfaces for $\theta_0 = 60^\circ$ shown as a function of slope (roughness) for the metals listed in the legend (the number after the atomic symbol represents the wavelength of the light involved). The smooth surface absorptances at incidence angle $\theta_0 = 60^\circ$ is given in parentheses.

If we consider a typical set of results from Fig. 17 (e.g. Rh at 1064nm) we can divide the surface roughness/absorptance relationship into three segments as follows:

1. Roughness range $0 < \sigma/\tau < 0.2$

As we saw in Figs. 9-12, the absorptance of metals does not change for normally incident light in this low roughness range because only first order scattering takes place. However, the situation is different at high angles of incidence.
If the low roughness surface shown in Fig. 18a was exposed to normal incidence light there would only be a trivial amount of second order scattering. At high angles of incidence however, second order reflections can easily take place even on low roughness surfaces – as demonstrated in Fig. 18a. The increase of this second order scattering events with increasing roughness is the reason why the surface absorptance increases in the range $0 < \sigma/\tau < 0.2$.

2. Roughness range $0.2 < \sigma/\tau < 0.6$

As the roughness of the surface is increased into this range a considerable proportion of the surface becomes shadowed as a consequence of its topology and the high angle of incidence of the light. This phenomenon is demonstrated by Fig. 18b. To understand the effect of shadowing on the absorptance we need to refer back to Fig. 18a; here we can see that second order reflection at a high angle of incidence must involve the reflection of the ray off both sides of the same ‘valley’. If the roughness is low there will be no shadowing and the whole of both sides of any valley are available to take part in multiple absorption events (although not all primary reflections will give rise to secondary ones). As the roughness is increased shadowing becomes a feature and the sides of the valleys closest to the light source become increasingly unavailable as sites for primary reflections. The number of multiple absorption events will therefore decrease and this will lead to a reduction in absorptance.

3. Roughness range $0.6 < \sigma/\tau < 2$

The increase in absorptance as the roughness increases through the range $0.6 < \sigma/\tau < 2$ is due to an increase in the level of backscattered multiple absorption events demonstrated by Fig. 18c. In this case the multiple reflections once more involve both sides of the same valley but the order of reflection is reversed from the forward scattering shown in Fig. 18c. This effect increases with surface roughness (within the limits shown here) and a gradual increase in absorptance is the result.
Figure 18. Three different regions of scattering behaviour for light incident with a relatively large angle, here illustrated for $\theta_0 = 60^\circ$. (a) Low roughness – multiple scattering in the forward direction is possible, (b) Intermediate roughness – shadowing results in a decrease of multiple scattering in the forward direction, (c) High levels of roughness – multiple scattering in the backward direction (back scattering).

If the light is incident on the surface at a glancing angle the backscattering phenomenon illustrated in Fig. 18c becomes suppressed. Fig. 19 demonstrates this point, showing the scattering experienced by a beam with $\theta_0 = 80^\circ$. 
Figure 19. Three different regions of scattering behaviour for light incident at a glancing angle, here illustrated for $\theta_0 = 80^\circ$. (a) Low roughness – minimal multiple scattering, (b) Intermediate roughness – shadowing - minimal multiple scattering, (c) High levels of roughness – only the top part of the far valley is non-shadowed – minimal multiple scattering or backscattering.

Here we can see that the geometry of the situation generally favours only single scattering interactions even if the surface roughness is increased (which is confirmed by simulations, see Fig. 20). At this high angle of incidence one side of each valley is in shadow in nearly all cases. Also, the light can generally only interact with the upper part of the far slope of the valley in question. The geometry of this arrangement ensures that light is not scattered back onto the shadowed slope. Fig. 21 presents the ratio of the absorptance for increasingly rough surfaces compared to the flat surface absorptance at an angle of incidence.
of $80^\circ$. Here we can see that the absorptance tends to level out as $\sigma/\tau$ rises above a value of 0.6. This is because the number of multiple scattering events does not rise with roughness (as seen in Fig. 20).

**Figure 20.** The average number of scattering points per incident ray as a function of the RMS slope (roughness) $\sigma/\tau$ for $80^\circ$ incidence.

**Figure 21.** Absorptance ratios shown as a function of slope (roughness) for optical constants representing the metals listed in the legend (the number after the atomic symbol represents the wavelength involved). The smooth surface absorptance at incidence angle $\theta_0 = 80^\circ$ is given in parentheses.
It may seem surprising that some of the metal/wavelength combinations in Fig. 21 show a decrease in absorptance with increasing roughness and then remain at a level which is lower than 1.0 (the flat surface value). This phenomenon can be explained with the help of Fig. 22 which considers the local angle of incidence experienced by individual rays rather than the macroscopic overall angle of 80 degrees.

![Figure 22. Mean average local angle of incidence as a function of surface roughness (slope).](image)

Fig. 22 shows that as the roughness ($\sigma/\ell$ or slope) increases, the average local angle of incidence changes from 80° to approximately 30° (the standard deviation curve demonstrates that, for a flat surface, the incident angle is exactly 80°, but variance around the mean increases as the roughness increases). This change in mean local angle of incidence is clearly demonstrated in Fig. 19 which shows that, as the surface roughness increases, the light interacts with an increasingly specific portion of the surface i.e. the upper part of the far side of each valley. The geometry of this area means that the local angle of incidence is generally considerably smaller than 80°. To understand why this downwards drift in local angle of incidence gives us ‘A rough/A smooth’ ratios of less than 1.0 (see Fig. 21) we need to refer to the absorptance vs. angle of incidence curves for the materials in question.
Fig. 23a clearly shows that, for Al at a wavelength of 532nm, the absorptance is much greater at an angle of incidence of 80° than it is for angles of approximately 30°. This being the case it is easy to understand that, if single scattering is predominant and if an increase in surface roughness gives a decrease in local angles of incidence, the overall absorptance of the surface will decrease (which is confirmed in Fig. 21). Conversely, if we look at the absorptance vs. angle of incidence curve for Au at a wavelength of 532nm (Fig. 23b), we can see that a reduction in angle of incidence from 80° to 30° will result in an increase in absorptance. This result is also confirmed in Fig. 21 which shows that, for gold, rough surfaces (with lower local angles of incidence) are more absorptive than a smooth surface.

![Figure 23. Absorptance (A) vs. angle of incidence (θ₀) for perfectly smooth (a) Aluminium and (b) Gold, both at λ=532nm. A(80°) is indicated in the figure.](image)

**Conclusions**

The conclusions below have, of course, only been demonstrated within the confines of the roughness and wavelength limits of this paper. However the authors feel that the principles will remain valid over a much wider range of roughness and wavelengths. Some of the points raised in the following conclusions are well established but have been included here in the interests of providing a full set of observations.
1. When light is normally incident on a flat smooth metal surface it is reflected in a specular manner back in the direction it came from. If the surface is slightly roughened the reflected light diverges from the specular reflection angle. If the surface is roughened beyond a certain limit then multiple reflection events take place which tend to concentrate the beam back in the direction it came from.

2. When light is incident at an angle on a flat, smooth metal surface it is reflected off the surface at the same angle in a specular manner. If the surface is slightly roughened the beam diverges away from this specular reflection angle. If the surface is roughened above a threshold value multiple scattering can take place which, eventually, can direct a substantial proportion of the beam back in the approximate direction it came from.

3. For normally incident light the absorptance of the surface increases with roughness after a certain roughness threshold has been exceeded. This phenomenon is the result of the onset of multiple scattering events. (Double scattering has its own roughness threshold and triple scattering has a higher one etc.).

4. The increase of absorptance with roughness noted above is most pronounced for metals which have a low absorptivity in the flat, smooth state.

5. For small angles of incidence (up to approximately 30° * in the case of this study) the change in absorptance with surface roughness follows the same trends as for normal incidence (noted above). * 0° = Normal incidence.

6. For large angles of incidence (approximately 60° in the case of these results) absorptance first rises, then falls, then rises again as the surface roughness is increased. The initial rise is due to increasing levels of multiple scattering (the multiple scattering roughness threshold is minimised at large angles of incidence). The fall is caused by part of the surface falling into shadow, a phenomenon which inhibits multiple scattering. The eventual rise in absorptance is due to advent and rise of backscattered multiple scattering events.

7. For very large angles of incidence (approximately 80° in the case of this work) the absorptance eventually remains fairly uniform with increasing surface roughness after rising or falling to a level above or below the reflectivity level for a smooth flat surface. The rise or fall in this case is due to the fact that the average local angle of incidence must be considered
when ray tracing on rough surfaces. The average local angle of incidence falls with increasing roughness and the absorptance vs. angle of incidence relationship (the Fresnel curves) for the material will determine whether the absorptance rises or falls with increasing roughness.

8. At very large angles of incidence the light – material interaction is predominantly governed by single scattering events whatever the roughness involved. This results in only small changes in absorptance with increasing roughness once the ‘local angle of incidence’ (7 above) settles down to a fairly constant value.

References


Paper 5: A Ray-Tracing Analysis of the Absorption of Light by Smooth and Rough Metal Surfaces
The Absorption of Light by Rough Metal Surfaces
- A Three-Dimensional Ray-Tracing Analysis

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Paper 6: The Absorption of Light by Rough Metal Surfaces – A Three-Dimensional Ray-Tracing Analysis
The Absorption of Light by Rough Metal Surfaces
— A Three-Dimensional Ray-Tracing Analysis

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Abstract

The laser absorptance of rough surfaces has been investigated using Monte Carlo simulations based upon three-dimensional ray-tracing. The influence of multiple scattering, shadowing and Fresnel-equation based angle dependence is discussed. The 3D results are compared to previously published results from a 2D ray-tracing analysis and the different applications of the two models are explained.

Keywords; Absorptance, Light, Surface roughness, Angle of Incidence, Laser.

I. Introduction

In Laser Material Processing, an understanding of the various mechanisms for light absorption is important for the modelling of particular applications (such as welding, cutting, surface treatment, etc.) as well as for optimizing the use of the applied laser energy. The energy of the laser beam is converted into heat in the workpiece through the process of light absorption. In metals, light is absorbed mainly via free electrons (intraband absorption) and/or bound electrons (interband absorption). On a macroscopic level we can define the absorptance¹ as the ratio of the absorbed laser power to the incident laser power. This ratio depends on several parameters including laser wavelength, angle of incidence, polarization, metal oxide layer thickness and structure, surface contamination and surface roughness or topography [1].

¹ The convention used here follows that of NIST; for radiative properties the –ivity ending is reserved for pure and smooth surfaces and the –ance ending for all others (i.e. all real surfaces).
In a previous study, two-dimensional ray-tracing involving one-dimensional surface profiles was used to investigate the influence of surface topography on light absorption [2]. In the present paper, this method has been extended to a full three-dimensional analysis on two-dimensional rough surfaces. Ray-tracing, also known as the geometric optics (GO) approximation in scattering theory [3-4], is a common tool in various scientific and engineering applications where the scattering properties of light are of interest, e.g. in computer graphics, optical instrument design, communications, remote sensing, heat transfer analysis, etc. [5-7]. In the GO approximation, energy bundles are traced throughout their interactions with the surface until they leave it. At each interaction point, the directions and amplitudes of the individual energy bundles or rays are determined from Snell’s law of reflection and Fresnel’s equations, respectively.

For one-dimensional surfaces, which are associated with two-dimensional scattering, ray-tracing has been verified by Tang & Buckius [3] to be a good approximation to the more exact electromagnetic wave theoretical models based upon Maxwell’s equations. The authors demonstrated that the approximation is valid as long as \( \sigma \cos(\theta_0)/\lambda > 0.17 \) (\( \sigma \) being the RMS roughness, \( \lambda \) representing the wavelength of light and \( \theta_0 \) the angle of incidence where \( \theta_0 = 0^\circ \) means normal incidence) and for surfaces with \( \sigma/\tau < 2.0 \) (where \( \tau \) is the surface correlation length). Although no rigorous studies of the regions of validity for 3D ray-tracing (on two-dimensional surfaces) have been conducted, comparisons made to the few existing results from wave theory and to various experimental results seem to indicate that the approximation is valid for the same range as in the 2D case [4].

The following section presents some basic and useful quantities from radiative theory and describes the generation and properties of Gaussian random rough surfaces as well as providing a detailed description of the ray-tracing model.

**II. Theory and Analysis**

**A. Radiative properties**

When light is incident on a material surface the energy will either be reflected, transmitted or absorbed. To describe and quantify the angular distribution of the
reflected or scattered light, the bidirectional reflection distribution function (BRDF) is defined as [8]

\[ \rho_i^s(\Omega_s, \Omega_i) = \frac{\pi}{\cos(\theta_s)} \frac{d\Phi_s}{d\Omega_s} \frac{d\Phi_i}{d\Omega_i} \]  

(1)

where \( \theta_s \) is the polar (zenith) angle of the scattered light, \( \Phi_i \) and \( \Phi_s \) are the incident and scattered radiant powers and \( \Omega_i(\theta_i, \varphi_i) \) and \( \Omega_s(\theta_s, \varphi_s) \) are the incident and scattered solid angles, respectively (see Fig. 1). The BRDF is a fundamental quantity in radiative theory, from which all reflective properties can be derived and is the quantity most often derived in scattering models.

Fig. 1: Definition of the BRDF in a global reference frame. The x-y plane here defines the mean plane of the rough surface and the z-axis is normal to this plane. The directions of the incident (subscript i) and scattered (subscript s) rays of light, subtended by the solid angles \( d\Omega \), are specified by the zenith and the azimuth angles, \( \theta \) and \( \varphi \) respectively.

Integration of the bidirectional reflectance over the entire hemisphere yields the directional-hemispherical reflectance [8]

\[ \rho_i^s(\Omega_i) = \frac{1}{\pi} \int_{2\pi} \rho_i^s(\Omega_s, \Omega_i) \cos(\theta_s) \ d\Omega_s. \]  

(2)
For opaque materials such as metals (except when dealing with extremely thin metal films), all non-reflected light can be regarded as absorbed and, by conservation of energy, the spectral directional absorptance can be found from

\[
A'_{\lambda}(\Omega_i) = 1 - \rho'_{\lambda}(\Omega_i). \tag{3}
\]

Since there exists a direct correspondance between emittance and absorptance from Kirchhoff’s law in thermodynamics, BRDF models in scattering theory can also be used to quantify the emissive properties of a material body.

**B. Gaussian random rough surfaces**

A two-dimensional random rough surface, \( z = \zeta(x,y) \), is commonly characterized by its height distribution and its autocovariance function, the former describing the surface height deviation from a mean surface level and the latter describing how the peaks and valleys are laterally distributed along the surface. In this paper, both the height distribution function (HDF), \( p_h(\zeta) \), and the autocorrelation function (ACF), \( C(x,y) \), are assumed to have a Gaussian distribution, thus obeying the following properties

\[
p_h(\zeta) = \frac{1}{\sqrt{2\pi\sigma^2}} \exp\left(-\frac{\zeta^2}{2\sigma^2}\right), \tag{4}
\]

\[
C(x,y) = \exp\left(-\frac{x^2}{\tau_x^2} - \frac{y^2}{\tau_y^2}\right), \tag{5}
\]

where \( \sigma \) is the RMS height and \( \tau_x \) and \( \tau_y \) are the correlation lengths in \( x \) and \( y \), respectively. Thomas has demonstrated that this Gaussian assumption is valid as long as the height at any point on the surface is not produced by a single one-off event (which is true of most surfaces) \[9\]. It is also assumed in this work that the surfaces are isotropic, i.e. showing no preferred roughness direction so that \( \tau_x = \tau_y = \tau \).
As a result of the fact that both the height distribution and the autocorrelation function are Gaussians, the slope distribution function (SDF), \( p_s(\xi) \), will also be a Gaussian, i.e.

\[
p_s(\xi_x, \xi_y) = \frac{1}{2\pi w_x w_y} \exp\left( -\frac{\xi_x^2}{2w_x^2} - \frac{\xi_y^2}{2w_y^2} \right),
\]

where \( \xi_x = \partial \xi / \partial x \) and \( \xi_y = \partial \xi / \partial y \) are the slopes in x and y directions, respectively. The RMS slopes are \( w_x \) and \( w_y \) and isotropy results in their equality so that \( w_x = w_y = w \). It can be shown that the RMS slope for a Gaussian random rough surface will be given by \( w = \sqrt{2} \sigma / \tau \) [10]. For simplicity the factor of \( \sqrt{2} \) will be omitted in the following treatment and when we speak of the RMS slope or slope we will only use the ratio \( \sigma / \tau \).

For simulation purposes, two-dimensional random rough surfaces with the above properties can be generated using the method outlined by Garcia and Stoll. Working on a discrete mesh of points in the x-y plane, an uncorrelated Gaussian random rough surface distribution, \( \xi_u(x,y) \), is generated using a Gaussian random number generator. To achieve correlation of surface points, this distribution is then convolved with a Gaussian filter

\[
F(x, y) = \frac{2}{\tau \sqrt{\pi}} \exp\left( -2(x^2 + y^2) / \tau^2 \right),
\]

which means that we set

\[
\xi(x, y) = \int \int F(x-x', y-y') \xi_u(x', y') dx' dy'.
\]

In practice this can be implemented using a Fast Fourier Transform (FFT) algorithm. Fig. 2 shows two realizations of 2D Gaussian random rough surfaces generated using this method, for two different RMS slopes. Fig. 3 shows statistical data for a surface generated with RMS slope \( \sigma / \tau = 1.0 \).
Fig. 2: Two realizations of two-dimensional Gaussian random rough surfaces, with (a) $\sigma/\tau = 0.1$ and (b) $\sigma/\tau = 1.0$ (the correlation length $\tau$ was $1\mu m$ in both cases).
Fig. 3: Surface statistics for a Gaussian random rough surface generated with $\sigma = 1\mu m$ and $\tau = 1\mu m$. Fig. 3(a) shows a normalized histogram of the height distribution function (HDF) while (b) is a graph of the autocorrelation function (ACF), here shown in the x direction as an example. In both figures the exact Gaussians are included for reference as dashed lines. Fig. 3(c) shows the corresponding normalized 2D histogram of the slope distribution function (SDF), which also follows a Gaussian distribution approximatively.
C. The Geometric Optics Approximation

The Geometric Optics (GO) approximation is an approximation to the more exact numerical integration methods from electromagnetic wave theory. As an approximation it is, of course, limited to certain roughness parameters, which have been discussed by Tang & Buckius [3-4]. Although no rigorous study has been made of the region of validity of the 3D scattering approximation, the authors projected that the parameters derived for the 2D case would very likely also hold for the 3D case [4].

The GO approximation is a ray-tracing approach, where energy bundles or rays are traced throughout their interactions with the surface until they leave it, see Fig. 4. The surface is assumed to be locally flat so that light is specularly reflected at each interaction point (this is known as the Fresnel approximation). Unlike many other approximations, the GO approximation treats the phenomena of multiple scattering and shadowing automatically. Multiple scattering describes the situation when light is scattered more than once from the surface (an example is given in Fig. 4 which shows double scattering), which becomes more important the rougher the surface. Shadowing is when some parts of the surface are blocked from the incident light due to obstruction from other sections (see the greyed areas in Fig. 4 for an illustration). Shadowing is important for oblique angles of incidence and becomes more important the higher the angle.

Fig. 4: The geometry of three-dimensional ray-tracing, illustrating multiple (double) scattering and shadowing phenomena (shadowed regions are darker grey).
The first step of the ray-tracing technique consists of generating the Gaussian random rough surface for the pre-specified correlation length, RMS height and surface length. The slopes and normals for all surface points are then estimated (see for instance Thümer and Wüthrich [11]). The angles of incidence; the polar (zenith) angle $\theta_0$ and the azimuth angle $\varphi_0$, are then chosen and an incident ray vector is formed as $\mathbf{s} = -[\sin(\theta_0)\cos(\varphi_0), \sin(\theta_0)\sin(\varphi_0), \cos(\theta_0)]$.

Next, a first reflection point is chosen. The first reflection points can be chosen from a random or an equally spaced distribution of points over the surface. The first reflection point chosen is checked for shadowing by analysing all surface points in the direction of the incident ray (more specifically the surface points lying in the plane formed by $\mathbf{s}$ and the projection of $\mathbf{s}$ onto the x-y plane). If any surface point in this plane lies above the incident ray vector, the first reflection point is considered as shadowed and the next one is considered. If the reflection point is not shadowed, the direction of the reflected ray is calculated from Snell’s law: $\mathbf{s}_r = \mathbf{s} - 2\mathbf{n}(\mathbf{n} \cdot \mathbf{s})$, where $\mathbf{n}$ is the local surface normal.

The energy of all incident rays, $G_i$, are equally distributed but are compensated for different area projections at the first reflection points according to Lambert’s cosine law. The energy of the reflected ray, $G_r$, can essentially be found by multiplying the incident energy by the Fresnel coefficient $R_{s,p}(\theta_0, \varphi_0, n, k)$, which is found from Fresnel’s equations and is dependent upon the incident polarization (s or p), the angles of incidence $(\theta_0, \varphi_0)$ and the optical constants of the medium (refractive index $n$ and extinction coefficient $k$). But, because of the random orientations of the local surface normal vectors, the incident polarization will change upon reflection, i.e. depolarization will occur. To account for this, the incident energy is decomposed into s- and p-polarized components, $G_{i,s}$ and $G_{i,p}$, respectively. These components are defined in a global reference frame defined by the z-axis unit vector $\mathbf{z}$ and the direction of the incident ray $\mathbf{s}$, the two vectors that define the global plane of incidence. In a similar manner, $\mathbf{z}$ and the reflected ray $\mathbf{s}_r$ will define the plane of reflection, where the polarized components of the reflected energy, $G_{r,s}$ and $G_{r,p}$, are formed.
The incident and reflected energies are then related according to

\[
\begin{bmatrix}
G_{r,s} \\
G_{r,p}
\end{bmatrix} =
\begin{bmatrix}
\rho_{ss} & \rho_{ps} \\
\rho_{sp} & \rho_{pp}
\end{bmatrix}
\begin{bmatrix}
G_{i,s} \\
G_{i,p}
\end{bmatrix},
\]

(9)

where \(\rho_{ss}\) and \(\rho_{pp}\) are the co-polarized reflectivities and \(\rho_{sp}\) and \(\rho_{ps}\) are the cross-polarized reflectivities (the first and second index stand for the incidence and reflection, respectively).

The calculation of these polarized reflectivities involves two conversions of polarization components. First, the s- and p-polarized components in global coordinates are transformed into their counterparts in the local frame of the specific interaction point (the frame defined by local surface normal and incident ray vector). The local polarization components are multiplied by the Fresnel amplitude reflection coefficients and are then converted back into the global frame. Accordingly, the polarized reflectivities can be calculated from [13,14]

\[
\begin{align*}
\rho_{ss} &= \left( (\mathbf{v}_r \cdot \mathbf{s}) (\mathbf{v}_i \cdot \mathbf{s}) r_s + (\mathbf{h}_r \cdot \mathbf{s}) (\mathbf{h}_i \cdot \mathbf{s}) r_p \right)^2 \left| \mathbf{s} \times \mathbf{s} \right|^4, \\
\rho_{sp} &= \left( (\mathbf{h}_r \cdot \mathbf{s}) (\mathbf{v}_i \cdot \mathbf{s}) r_s - (\mathbf{v}_r \cdot \mathbf{s}) (\mathbf{h}_i \cdot \mathbf{s}) r_p \right)^2 \left| \mathbf{s} \times \mathbf{s} \right|^4, \\
\rho_{ps} &= \left( (\mathbf{v}_r \cdot \mathbf{s}) (\mathbf{h}_i \cdot \mathbf{s}) r_s - (\mathbf{h}_r \cdot \mathbf{s}) (\mathbf{v}_i \cdot \mathbf{s}) r_p \right)^2 \left| \mathbf{s} \times \mathbf{s} \right|^4, \\
\rho_{pp} &= \left( (\mathbf{h}_r \cdot \mathbf{s}) (\mathbf{h}_i \cdot \mathbf{s}) r_s + (\mathbf{v}_r \cdot \mathbf{s}) (\mathbf{v}_i \cdot \mathbf{s}) r_p \right)^2 \left| \mathbf{s} \times \mathbf{s} \right|^4.
\end{align*}
\]

In Eq. (10), \(r_s\) and \(r_p\) are the Fresnel amplitude reflectivities (see for instance Modest [8]). \(\mathbf{h}\) represents the direction of the s- or horizontal polarized components and \(\mathbf{v}\) the direction of the p- or vertical polarized components where incident vectors have subscripts \(i\) and reflected vectors have subscripts \(r\). Hence, the unit vectors \(\mathbf{h}_i\) and \(\mathbf{v}_i\) will be perpendicular and parallel to the plane of incidence, respectively, and similarly \(\mathbf{h}_r\) and \(\mathbf{v}_r\) will be perpendicular and parallel to the plane of reflection, respectively.
These vectors can be calculated from

\[ \vec{h}_i = \frac{\vec{z} \times \vec{s}_i}{|\vec{z} \times \vec{s}_i|}, \quad \vec{v}_i = \vec{h}_i \times \vec{s}_i, \]

\[ \vec{h}_r = \frac{\vec{z} \times \vec{s}_r}{|\vec{z} \times \vec{s}_r|}, \quad \vec{v}_r = \vec{h}_r \times \vec{s}_r. \]  

To determine whether the scattered ray, \( \vec{s}_r \), strikes the surface again, a check similar to the one for shadowing is carried out. All surface points in the direction of \( \vec{s}_r \) are checked and if any surface point in the scattering plane (defined by \( \vec{s}_r \) and the projection of \( \vec{s}_r \) onto the x-y plane) lies above \( \vec{s}_r \), then a new interaction point has been found and a new scattered ray is calculated. Once the scattering process has been completed, the remaining energy of the incident ray which finally leaves the surface is added to the BRDF.

When all the first reflection points have been considered and all the rays have been traced and scattered, the BRDF can be found from Eq. (1). The directional-hemispherical reflectance can then, in principle, be found from an integration over all scattered angles as in Eq. (2), but can more easily and straightforwardly be found by dividing the sum of the energies of all the rays which leave the surface by the total incident energy. The absorptance is subsequently found from Eq. (3), i.e. by taking one minus the reflectance. To get statistically accurate results, the above procedure is then repeated for several realizations of surfaces having the same set of values for the correlation length and the RMS height and an overall average can thus be calculated.

### III. Results and Discussion

The results presented below are overall averages for each RMS slope \( \sigma/\tau \) taken from Monte Carlo simulations of 15-20 surface realizations (which was considered sufficient due to the small relative standard deviations involved, typically 1% or less) of sample sizes 100 x 100 \( \mu \text{m} \).
The following analysis will initially be separated into normal and oblique incidences (sections A and B, respectively). Normal incidence is a somewhat special case, because of the absence of shadowing, but an important one nonetheless since this is the angle used in the initiation phase of most laser material processing applications.

In section C the predictions of Fresnel’s equations and the Brewster angle are discussed in relation to rough surfaces. In section D a comparison is made to some earlier published results from 2D ray-tracing simulations. Section E concludes the results and discussion section with some general notes on roughness in the context of laser-materials processing.

A. Normal incidence, $\theta_0=0^\circ$

Fig. 5 displays the absorptance results at normal incidence, i.e. when light is incident perpendicular to the mean surface plane. In the figure, the absorptance is plotted as a function of RMS slope, after being normalized to the absorptance of a flat, smooth surface for the same metal/wavelength combination (i.e. the Fresnel absorptance at normal incidence). Two distinctly different regions can be identified:

Roughness range $0 < \sigma/\tau < 0.15$: Single scattering regime

The results show a minimal variation in absorptance over this roughness range. A comparison with Fig. 6 (see the blue line for normal incidence), which shows the average number of scattering points per ray as a function of slope, reveals this as a single scattering regime where rays are scattered only once before leaving the surface and thus absorptance only will depend upon the distribution of local surface normals and local angles of incidence. These angles are very small in this range of slopes and hence the absorptance shows no noticeable changes from the smooth surface value.

Roughness range $0.15 < \sigma/\tau < 2$: Multiple scattering regime

At around $\sigma/\tau \sim 0.15$ the absorptance starts to increase more sharply and a comparison with Fig. 6 explains that this is the threshold for double scattering. Beyond this threshold, the average number of scattering points increases almost
linearly with slope. As each scattering point contributes energy to the surface, the absorptance will also increase in this slope range for all metal/wavelength combinations. However, as we can see in Fig. 5, less absorptive (i.e. more reflective) materials are found to be more sensitive to roughness than more absorptive ones. This is simply a consequence of ‘diminishing returns’ where each new scattering point along a ray path contributes less and less energy for absorption due to the limited amount of energy available in the bundle. For example; If the absorptivity of a surface is 50% then the first scattering event will result in an absorption of 50%, and the second will result in absorption of a further 25% (of the original energy), taking the total absorption level up to 75%. If, on the other hand, the absorptivity of the material is only 10%, then the absorption figures from the primary and secondary scattering events will be 10% and 9% - making a total of 19%. This increase from 10% to 19% is considerably larger than the increase from 50% to 75% and this is the principle demonstrated in Fig. 5.

The practical consequence of this principle is that the deliberate roughening of a high reflectance material, such as Aluminium at 1064nm, increases the absorption relatively more than it would for a material that already absorbs well in the smooth state (compare the relative increases in absorptance for Aluminium and Titanium in Fig. 5).

Fig. 5: Ratios of the absorptances of rough and smooth surfaces for normally incident light shown as a function of RMS slope (roughness) for the metals listed in the legend (the number after the atomic symbol indicates the wavelength of the light involved). The smooth surface absorptances at normal incidence are given in parentheses.
Fig. 6: The average number of scattering events per incident ray as a function of the RMS slope (roughness) $\frac{\sigma}{\tau}$ for the different angles of incidence (AOI) in the study.

B. Oblique incidence

At oblique incidence the analysis becomes more complex, mainly due to the influence of shadowing. In the simulations, three different oblique angles of incidence were analysed; 30°, 60° and 80°. At 30° incidence the results are very similar to those for normal incidence (compare Fig. 7 to Fig. 5), except for a small decrease of the double scattering threshold and slightly less overall scattering (and therefore absorption) for the larger slopes (as seen in Fig. 6). These effects are even more apparent for 60° (see Fig. 8), for which they will be discussed in greater detail.
Fig. 7: Ratios of the absorptances of rough and smooth surfaces for $\theta_0 = 30^\circ$ shown as a function of RMS slope (roughness) for the metals listed in the legend (the number after the atomic symbol represents the wavelength of the light involved). The smooth surface absorptances at incidence angle $\theta_0 = 30^\circ$ are given in parentheses.

Fig. 8: Ratios of the absorptances of rough and smooth surfaces for $\theta_0 = 60^\circ$ shown as a function of RMS slope (roughness) for the metals listed in the legend (the number after the atomic symbol represents the wavelength of the light involved). The smooth surface absorptances at incidence angle $\theta_0 = 60^\circ$ are given in parentheses.
At 60° incidence, the analysis is most effectively treated by dividing the absorbance/roughness relationship into the following four segments;

**Roughness range** \(0 < \sigma/\tau < 0.1\): Single scattering regime

As for normal incidence, this range of very small slopes involves only single scattering events and roughness therefore does not have a significant effect on the absorbance.

**Roughness range** \(0.1 < \sigma/\tau < 0.2\): Multiple forward scattering regime

A comparison of Figures 8 and 7 reveals that the roughness threshold for double scattering reduces with increasing angle of incidence. This point is also confirmed by Fig. 6. This threshold reduction can be understood from Fig. 9. A simple condition for the introduction of double scattering can be put forward as this; if the heights of the bumps on the sample surface are increased continuously from a flat state to the geometry shown in Fig. 9, double scattering will, in a first approximation, initially occur for surface patches where the first scattering point produces a horizontally reflected ray (i.e. a ray parallel to the mean reference plane). This corresponds to a ray with zenith angle of 90° (the zenith angle is the angle between the ray and the positive z-axis). At normal incidence this occurs for surface patches where the normal vectors have inclination angles \(\varphi_N = 45°\) (the inclination angle is the angle between the surface normal and the z-axis). As the angle of incidence increases, this inclination angle limit for double scattering decreases (as seen in the figure) according to \(\varphi_N = \pi/4 - \theta_0\), so that for instance \(\varphi_N(\theta_0=30°) = 30°\), \(\varphi_N(\theta_0=60°) = 15°\) and \(\varphi_N(\theta_0=80°) = 5°\).

Double scattering in this range involves the type of forward scattering events illustrated in Fig. 10b, where the rays are scattered mainly in the forward direction with respect to the incident ray. These kinds of events will become more probable the higher the slope and thus scattering and absorption will increase in this range.
Fig. 9: The condition for double scattering is changed as the angle of incidence $\theta_0$ is increased, since the limit of the surface inclination angle $\varphi_N$ required for generating a horizontally scattered ray is reduced.

Fig. 10: Four different regions of scattering behaviour for light incident with a relatively large angle, here illustrated for $\theta_0 = 60^\circ$. (a) Very low roughness – single scattering only, (b) Low roughness – multiple scattering in the forward direction is possible, (c) Intermediate roughness – shadowing inhibits multiple scattering in the forward direction, (d) High levels of roughness – multiple scattering in the backward and sideways directions.
Roughness range $0.2 < \sigma/\tau < 0.5$: Shadow-inhibited regime

In this region we find that the absorptance either levels out or decreases with increasing surface roughness. This contra-intuitive result is explained as follows:

As the roughness of the surface is increased into this range a considerable proportion of the surface becomes shadowed as a consequence of its topography and the high angle of incidence of the light. Fig. 11 shows the average number of reflection points being shadowed and the phenomenon is demonstrated by Fig. 10c. To understand the effect of shadowing on the absorptance we need to refer back to Fig. 10b; Here we can see that second order reflection at a high angle of incidence must involve the reflection of the ray off both sides of the same ‘valley’. If the roughness is low there will be no shadowing and the whole of both sides of any valley are available to take part in multiple absorption events (although not all primary reflections will give rise to secondary ones). As the roughness is increased shadowing becomes a feature and the sides of the valleys closest to the light source become increasingly unavailable as sites for primary reflections. This has the effect of inhibiting further increase of multiple forward scattering in this range (as seen in Fig. 6) and is the explanation for the levelling out or reduction in absorptance which is visible in Fig. 8.

Fig. 11: Shadowing, i.e. the average fraction of first reflection points being shadowed, as a function of RMS slope.

Roughness range $0.5 < \sigma/\tau < 2.0$: Multiple back- and sidedscattering regime

As the roughness is increased into this range, the absorptance is found to rise again. The effect of shadowing is in this case overcome by the type of higher
order scattering events depicted in Fig. 10d, where rays are scattered laterally (sideways) as well as in the backward direction (backscattering). These events make the shadowed regions once again available for scattering and absorption, which leads to an increase in absorptance.

For the grazing angle of incidence 80° (see the results in Fig. 12), we divide the roughness range into the following two regions:

Roughness range $0 < \sigma/\tau < 0.5$: Shadow-inhibited single scattering regime

The strong influence of shadowing (see the 80° line in Fig. 11) in this region inhibits multiple scattering to the extent that the absorptance is largely determined by single scattering events, as seen in Figures 13a-c (and Fig. 6 which shows the low level of scattering) involved. As single scattering is the dominant interaction, the absorptance will therefore mainly be a function of the distribution of local angles of incidence (as was the case for the single scattering, low roughness regimes discussed earlier).

Fig. 12: Ratios of the absorptances of rough and smooth surfaces for $\theta_0 = 80°$ shown as a function of RMS slope (roughness) for the metals listed in the legend (the number after the atomic symbol represents the wavelength of the light involved). The smooth surface absorptances at incidence angle $\theta_0 = 80°$ are given in parentheses.
Fig. 13: Four different regions of scattering behaviour for light incident with a relatively large angle, here illustrated for $\theta_0 = 80^\circ$. (a) Very low roughness – single scattering only, (b) Low roughness – single scattering remains the dominant interaction due to shadowing, (c) Intermediate roughness – shadowing continues to inhibit multiple forward scattering events, (d) High levels of roughness – multiple scattering is strongly inhibited in the forward and backward directions, but may occur laterally as seen by the non-shadowed regions along the sides of the hills.

The results in Fig. 12 show a clear division between metal/wavelength combinations for which the absorptance increases with RMS slope and combinations for which it doesn’t. The determining factors here are; a. Where the Brewster angle of the metal/wavelength is situated relative to the global angle of incidence and b. The shape of the Fresnel (angle of incidence/absorption) curve for the particular metal/wavelength combination. Fig. 14 shows how the average local angle of incidence changes with slope (roughness) while Fig. 15 shows the Fresnel absorptances of the different metal/wavelegths in the study. Since the
average local angle of incidence experienced by the incident rays decreases with increasing roughness from 80° to approximately 50°, we can see from Fig. 15 that the absorptance of Aluminium at 1064nm and 532nm as well as of Rhodium at 1064nm will decrease with roughness while Titanium, Gold and Copper at 532nm will increase, which is what the results of the simulations in Fig. 12 suggest.

Fig. 14: The average local angle of incidence as a function of RMS slope, for rays incident with a global angle of incidence $\theta_0 = 80^\circ$.

Roughness range $\sigma/\tau > 0.5$ : Multiple sidescattering regime

For the larger slopes, e.g. for $\sigma/\tau > 0.5$, backscattering is strongly suppressed because of the geometry and it is mainly sidescattering which causes a moderate increase of the absorptance in this range.
Fig. 15: Fresnel absorptances of the metal/wavelengths in the study, normalized to the value at normal incidence ($\theta_0 = 0^\circ$). The Fresnel absorptance at 80° is indicated in the figure.

C. Fresnel absorptance and the Brewster angle for rough surfaces

In some processing applications, such as laser cleaning and laser hardening (as well as other surface treatment processes), it is believed that a laser applied at or close to the Brewster angle of incidence can be beneficial for maximizing absorption. The Brewster angle is an angle usually situated in the range 60° - 85° (see Fig. 15 for a few examples) that can be found from Fresnel’s equations. However, Fresnel’s equations are defined for a perfectly flat and smooth surface and cannot be expected to hold for rougher surfaces. Fig. 16 and Fig. 17 show the simulation results for the angular dependence of the absorptance for Aluminium at 1064nm and for Copper at 532nm, respectively. As the roughness is increased from an almost smooth state, $\sigma/\tau = 0.01$, to a medium rough state, $\sigma/\tau = 0.3$, the Brewster maximum is reduced and eventually disappears for Aluminium (see Fig. 16a) and for both Aluminium and Copper the Fresnel predictions are replaced by a more flat angular response (see Fig. 16a and 17a, respectively). As roughness is increased further to rough and very rough surfaces, a new maximum is formed at or close to normal incidence, as Fig. 16b and Fig. 17b show.
Fig. 16: The absorptance vs. angle of incidence for aluminium at $\lambda = 1064$ nm, for a range of different RMS slopes (roughness). In (a) $A(\theta)$ is shown for the range of small and medium slopes where $0.01 \leq \sigma/\tau \leq 0.5$, while in (b) it is given for the range of larger slopes where $0.5 \leq \sigma/\tau \leq 2.0$. The Fresnel absorptance curve has been included for reference.
Fig. 17: The absorptance vs. angle of incidence for copper at $\lambda = 532$ nm, for a range of different RMS slopes (roughness). In (a) $A(\theta)$ is shown for the range of small and medium slopes where $0.01 \leq \sigma/\tau \leq 0.5$, while in (b) it is given for the range of larger slopes where $0.5 \leq \sigma/\tau \leq 2.0$. The Fresnel absorptance curve has been included for reference.
D. Comparison between 2D and 3D ray-tracing models

In a previous publication by the present authors, a 2D ray-tracing model was developed to study the effects of surface roughness on light absorption [2]. This 2D model had a limited application to real surfaces and this is the main reason for the extension of the work to the isotropic 3D model surface shown in Fig. 2. However, the rippled liquid surface inside a laser cutting zone is one type of topography which may be better understood by reference to the 2D work. Fig. 9 is typical of the figures produced in the 2D model because it involves no Y component to the light-surface interaction. Results from this type of 2D analysis can be directly used for real surfaces of the type shown in Fig. 18 (i.e. a real surface covered in parallel ripples). This surface is closer to the topography of the liquid in a laser cutting melt than the isotropic surface described in Fig. 2 and thus, in this case, the 2D model may be more useful as a starting point. One other area of application of the 2D model may be in the absorption of light by a surface which has been ground or machined with parallel grooves.

Fig. 18: The type of surface (covered in parallel ripples or grooves) which may be better analysed using the 2D rather than the 3D model.

Although the 2D and 3D models give the same phenomenological results, they exhibit quantitative differences as shown in Fig. 19. This figure shows the average number of scattering points for the four different angles analysed in the two ray-tracing models. In all cases the overall behaviour is very similar but the level of scattering is progressively higher in the 3D modelling case for surfaces above a certain roughness (slope) threshold.
The main reason for the increased level of scattering in 3D modelling as compared to the 2D modelling case can easily be appreciated by comparison of Figures 18 and 2. Fig. 2 describes the type of surface modelled in the 3D model and Fig. 18 is an equivalent description of the 2D model surface. It is clear that there are ‘hills and valleys’ in all directions in the case of Fig. 2 whereas the surface only undulates in one direction in Fig. 18. This means that, in the 3D model case (Fig. 2), there are more slopes available to scatter the incident light rays downwards. This improved efficiency of the isotropic surface to scatter light downwards results in an increase in the level of multiple scattering in the 3D model results, as shown in Fig. 19. This increase in multiple scattering will, of course, be accompanied by an increase in absorptance.
E. Surface roughness in the context of laser-materials processing

The results of this work have revealed a number of relationships between roughness and absorptance which should be of use to workers in the field of laser-materials processing. However, it should be appreciated that simply measuring the roughness in terms of RMS height (Ra or Rq) is inadequate for estimations of absorptance. This commonly used measure of roughness gives no information about the crowdedness of the hills and valleys which go to make up the material surface roughness. Thus, there is no information about the RMS slope associated with the surface roughness, which has been demonstrated in this paper to be a major influence on absorptance.

IV. Conclusions

In this paper, 3D ray-tracing of two-dimensional Gaussian random rough surfaces has been used to study the effects of roughness on laser absorption. Although the investigation has been focused on specific metals (aluminium, copper, gold, rhodium and titanium) at the particular wavelengths of the Nd:YAG laser (1064nm and 532nm), the authors feel that the phenomenological results and conclusions should be valid over a much wider range as long as the simulation parameters (wavelengths, angles of incidence, RMS heights and correlation lengths) are within the confines of the validity region of the Geometric Optics Approximation as shown by Tang & Buckius [3].

For light incident normally (0°) or with a relatively small angle (30°), the laser absorptance was found to increase with roughness (RMS slope) after the threshold for multiple (double) scattering had been reached. The increase of absorptance with roughness is most pronounced for metals which are very reflective in the flat, smooth state.

For light incident at relatively large angles of incidence (60° in the case of these results), the threshold for multiple scattering is lowered. After the threshold, the absorptance first increases with roughness (RMS slope) but then levels out or even decreases in a medium roughness range where shadowing inhibits multiple scattering. As the surface roughness is further increased the absorptance rises
again due to higher order scattering events which make shadowed regions once again available for absorption.

For grazing incidence ($\theta_0$ in the case of these results), single scattering dominates the behaviour for surfaces with small and medium roughness (RMS slope). The absorptance increases or decreases with increasing roughness depending on the location of the Brewster angle of the particular metal/wavelength combination involved. For very rough surfaces, the absorptance increases with roughness due to the introduction of higher order scattering events.

In a study of the full angular dependence of absorption it was found that the Fresnel angle of dependency only was satisfied for very smooth surfaces (with very small RMS slopes). For medium rough surfaces, the Brewster maximum is suppressed and there is a flatter angular response. For very rough surfaces, there is a maximum at or close to normal incidence.

It was also found that a previous 2D model of ray tracing described similar phenomena to the present 3D model but predicted generally lower levels of multiple scattering and therefore absorption. The 2D model could be useful however, in situations where the laser is impinging on a rippled or non-isotropic machined surface.

Finally, this paper has highlighted the point that any roughness index which relies only on height measurements is of little use when analysing the absorptance of light by a surface. For the purpose of this type of analysis it is important to know the RMS slope which is typical of the surface in question.

References


