Finding focus positions from an electron scattering experiment and using their deviation from the Rowland circle to predict focus positions for a stimulated RIXS experiment

Abstract

It has been found that a number of focus positions from an electron scattering experiment deviate from their theoretical position on the circumference of the Rowland circle. The deviation of the y-position has a linear relationship with the y-position; the deviations are in agreement with a theoretical analysis of the optics of the Rowland circle. A self-updating spread sheet has been made which can predict more focus positions by considering the theoretical position and the experimentally found deviation.

Introduction

Radiation from a scattering experiment can provide us with information about the atomic and sub-atomic structure of the scattering target. The greater the detail of this radiation which can be resolved, the more information we can acquire, and therefore high resolution is desired. When radiation is diffracted by a grating, the resolving power is proportional to the order of diffraction. As the order of diffraction increases, the resolution increases but the intensity of radiation decreases. The stimulated RIXS group at Uppsala will perform a scattering experiment using radiation from a free electron laser (FEL) at the Linac Coherent Light Source (LCLS) in Stanford, USA, to probe the atomic structure of solid state samples. Radiation from an FEL is so intense that there will still be plenty of intensity at higher orders, making it possible to utilise the high resolution at these high orders.

As I will explain in more detail, constructively interfering radiation which has been diffracted by a curved diffraction grating focuses on the circumference of the Rowland circle, which has half the radius of the curved grating. There are however theoretical and experimental corrections, meaning that in reality the focus positions deviate from the Rowland circle. In this project I performed an electron scattering experiment using solid state samples, directing the subsequent radiation onto a curved diffraction grating, therefore observing the focus positions of constructive interference close to the Rowland circle. The same grating will be used at LCLS and so the purpose of my project was to use data from my experiment to provide accurate predictions of the focus positions for any sample, at any diffraction order. If my project is successful, the stimulated RIXS group from Uppsala can save time when looking for focus positions during their beam time at LCLS.

Theory

This section describes the relevant theory behind the experiment. I will focus on the theory of the Rowland circle, because this is required for finding focus positions, and for offering explanations for their deviation from its circumference. Firstly, however, I will discuss the stimulated resonant Inelastic X-ray scattering (stimulated RIXS) process, which will provide the source of radiation incident on the diffraction grating at LCLS.

1 Såthe, C., 2011, Applications of Soft X-Ray Spectroscopy, Uppsala, ACTA, page 43
**Stimulated RIXS**

Inelastic x-ray scattering occurs when an x-ray photon incident on an atom transfers some or all of its energy to the excitation of electrons to higher energy levels in the atom. The x-ray photon will therefore emerge with less energy than before the scattering process. Resonant Inelastic x-ray scattering (RIXS) occurs when the incident x-ray photon is tuned to a specific resonant absorption frequency such that it is completely absorbed into the atom, transferring all of its energy to the excitation of a core electron to the conduction band. The core hole is subsequently filled by the de-excitation of a valence electron, via the emission of an x-ray photon. Thus, this is a photon-in, photon-out process.\(^3\)

The de-excitation of the valence electron in the RIXS process happens spontaneously with a certain probability as the electrons tend towards lower energy states. The de-excitation has a corresponding transition state with a dipole field of a specific frequency. If an incoming photon of the same frequency is present, the electric field which it provides can stimulate the de-excitation process, increasing the probability for it to occur. This process is known as 'stimulated RIXS' and requires two incoming photons of specific frequencies; the resonant absorption frequency and the frequency of the dipole field of the transition state are different.

At LCLS photons of a range of energies from a free electron laser will include the appropriate energies for absorption and stimulated emission. These photons will be incident on the surface of solid material samples. At Uppsala the excitations were driven by an incoming beam of electrons of a range of energies from an electron gun. Therefore the process at Uppsala was not a RIXS process but an electron scattering process. The de-excitation energies are the same for both processes, which is the important part of the process for my project. The x-rays emitted in the de-excitation process were passed through a slit and then incident on a curved reflection diffraction grating. In our experiment we looked for photons of known x-ray emission energies.

**Plane diffraction grating**

A plane reflection diffraction grating involves a row of reflecting strips, separated by spacing \(d\). A photon incident on a reflecting surface imparts energy to the charged particles in surface atoms, which subsequently oscillate. From electrodynamics, accelerating charged particles emit radiation, which is emitted in all directions. The result is a superposition of outgoing going waves from the reflecting strips. The diagram shows light incident at an angle \(\Theta_i\) and light outgoing at an arbitrary angle \(\Theta_r\).

---

We can approximate both sets of beams to be parallel because we are interested in effects which are relatively very far away where the ‘parallel’ beams coincide. For constructive interference we require the path difference between the two outgoing parallel beams to equal an integer number of wavelengths. We can use trigonometry to show the required angles at which constructive interference occurs:

\[ d \sin \theta_i + d \sin \theta_r = n \lambda \]  
\[ \theta_r = \sin^{-1} \left( \frac{n \lambda}{d} - \sin \theta_i \right) \]

Thus for a given incident energy, we can expect diffraction maxima at a range of angles, \( n \) gives the order of diffraction. Equation 1 is known as the grating equation.4

Curved diffraction grating

In this experiment, a curved reflection diffraction grating was used. I will now present the more detailed optical geometry of this curved grating and show that the above grating equation will only hold if certain approximations are made (namely the radius of curvature is infinite). This more detailed analysis will shed light on some more interesting phenomena; the constructively interfering rays of light from the curved grating will focus (with a certain error) on the perimeter of a circle, known as the Rowland circle, which has half the radius of the grating.5

Consider the curved grating in the diagram. The centre of the curved surface lies at the origin \( O \) of an \( X,Y,Z \) co-ordinate system. The \( X \)-axis is normal to the plane of the grating, the \( Y \) and \( Z \)-axes are as shown on the diagram. We will consider three points in this co-ordinate system. \( A \) is the source of light which is incident on the grating, with co-ordinates \((a,b,c)\). \( P \) is the point of the grating surface which is hit by the light, with co-ordinates \((x,y,z)\). \( B \) is a point on the light’s subsequent path after diffraction from the grating, with co-ordinates \((a',b',c')\).

The grating is covered in reflecting strips, parallel to the \( Z \)-axis, with spacing \( d \) in the \( Y \)-direction. Even though these strips have some width which leads to a periodic reflectivity with periodicity \( d \), we can approximate these strips as thin lines, neglecting any spread in reflectivity. So in our co-ordinate

---

system, the Y co-ordinate of point P has only discrete allowed values; \( y/d \) is an integer. For constructive interference, we require the path difference between two beams which reflect off different strips separated by \( y \) to be an integer number of wavelengths. For adjacent strips, this leads simply to:

\[
\text{path difference} = m\lambda, \text{ (adjacent strips)}
\]

(3)

where \( m \) is the order number. In general for strips separated by \( y/d \) this leads to:

\[
\text{path difference} = \left( \frac{y}{d} \right) m\lambda
\]

(4)

This allows constructive interference of light from each strip in-between the strips in question. For example, let us consider strip #1 and strip #4, so \( y/d = 3 \). For first order \((m=1)\) constructive interference we require a path difference of \( 3\lambda \). This allows a path difference of \( \lambda \) and \( 2\lambda \) between strips #1 and #2, and strips #1 and #3, respectively. Thus all strips will contribute to constructive interference.

I can now write a light path function, \( F \), which is required for constructive interference. Let us consider light following the path \( A \rightarrow P \rightarrow B \), for arbitrary \( P \) on the grating surface. Light following any different path from \( A \rightarrow B \) must satisfy the following function if it is to focus on point \( B \):

\[
F = AP + PB + \left( \frac{y}{d} \right) m\lambda
\]

(5)

Fermat’s principle of least time requires light to take the shortest possible path between points\(^6\), i.e. the light path function should be a minimum. For a fixed point \( A \) and an arbitrary point \( P \), point \( B \) will be positioned to satisfy Fermat’s principle. Light reflecting from all other points on the grating surface should also satisfy Fermat’s principle, so each light path function should be a minimum with respect to the co-ordinates of the arbitrary point \( P \). A minimum is located where the derivative is zero, so mathematically Fermat’s principle leads to the following conditions:

\[
\frac{\partial F}{\partial x} = 0
\]

(6)

\[
\frac{\partial F}{\partial y} = 0
\]

(7)

\[
\frac{\partial F}{\partial z} = 0
\]

(8)

The task now is to express \( F \) in terms of \( x, y \) and \( z \), and then differentiate the result to find out the conditions for constructive interference and focusing. The distances \( AP \) and \( PB \) as follows:

\[
(AP)^2 = (a - x)^2 + (b - y)^2 + (c - z)^2
\]

(9)

\[
(PB)^2 = (a' - x)^2 + (b' - y)^2 + (c' - z)^2
\]

(10)

The minus sign in the final term represents the fact that the Z co-ordinate of the incoming and outgoing light will necessarily have opposite signs. The orientation of the reflecting strips, however, allows the signs of Y co-ordinates to be the same. Regarding the X co-ordinate; the whole light path is in the positive X-axis.

A cylindrical co-ordinate system can also be used, with the same origin, and the following substitutions, where the angles ($\theta_i, \theta_r$) and lengths ($r, r'$) are in the XY plane:

\[
a = r \cos \theta_i \\
b = r \sin \theta_i \\
a' = r' \cos \theta_r \\
b' = r' \sin \theta_r
\]

These substitutions allow $(AP)^2$ and $(PB)^2$ to be written in terms of $r$ and $r'$:

\[
(AP)^2 = r^2 + c^2 + x^2 + y^2 + z^2 - 2ax - 2r \sin \theta_i y - 2cz \\
(PB)^2 = r'^2 + c'^2 + x^2 + y^2 + z^2 - 2a'x - 2r' \sin \theta_r y + 2c'z
\]

We can also write and re-arrange an equation for the sphere, radius $R$ which describes the curvature of the grating (notice that the origin of this sphere lies at $(R,0,0)$ in the co-ordinate system):

\[
(R - x)^2 + y^2 + z^2 = R^2 \\
x^2 - 2Rx + y^2 + z^2 = 0
\]

By rearranging equation 18 for $2Rx$ and substituting the expression into equations 15 and 16 we obtain:

\[
(AP)^2 = r^2 + c^2 + 2Rx - 2ax - 2r \sin \theta_i y - 2cz \\
(PB)^2 = r'^2 + c'^2 + 2Rx - 2a'x - 2r' \sin \theta_r y + 2c'z
\]

In order to expand these expressions further we can solve equation 18 for $x$ using the general formula for quadratic equations:

\[
x = R \pm \sqrt{R^2 - (y^2 + z^2)}
\]

We are interested in the solution with the negative sign: $x = R - \sqrt{R^2 - (y^2 + z^2)}$ because the solution with the positive sign represents points on the opposite side of the sphere. This solution can be expanded $^7$ in powers of $(y^2 + z^2)$:

\[
x = \frac{y^2 + z^2}{2R} + \frac{(y^2 + z^2)^2}{8R^3} + \frac{(y^2 + z^2)^3}{16R^5} + \frac{5(y^2 + z^2)^4}{128R^7} + \cdots
\]

By replacing all \(x\) terms in equations 19 and 20 with this series, and taking the square root, we obtain a series expansion of \(AP\) and \(PB\). The continued derivation of this series is presented in the appendix, here I display the most significant terms. Terms with higher inverse powers of \(R, r\) and \(r'\) are less significant because these terms are much greater than \(x, y\) and \(z\).

\[
AP = AP_1 + AP_2 + AP_3 + AP_4 + AP_5 + \cdots
\]

\[
PB = PB_1 + PB_2 + PB_3 + PB_4 + PB_5 + \cdots
\]

\[
AP_1 = r - y\sin\theta_i
\]

\[
AP_2 = \left[\frac{1}{2}y^2 \left(\frac{\cos^2\theta_i}{r} - \frac{\cos\theta_i}{R}\right) + \frac{1}{2}y^3 \frac{\sin\theta_i}{r} \left(\frac{\cos^2\theta_i}{r} - \frac{\cos\theta_i}{R}\right) + \frac{1}{2}y^4 \frac{\sin^2\theta_i}{r^2} \left(\frac{\cos^2\theta_i}{r} - \frac{\cos\theta_i}{R}\right) + \cdots\right]
\]

\[
AP_3 = \frac{1}{2}z^2 \left(\frac{\sin\theta_i}{r} - \frac{\cos\theta_i}{R}\right) - \frac{zc}{r} + \frac{c^2}{2r}
\]

\[
AP_4 = \frac{1}{2}z^2 \frac{y\sin\theta_i}{r} \left(\frac{1}{r} - \frac{\cos\theta_i}{R}\right) + \frac{y\sin\theta_i}{2r^2} \left(c^2 - 2zc\right)
\]

\[
AP_5 = \frac{(y^2 + z^2)^2}{8R^2} \left(\frac{1}{r} - \frac{\cos\theta_i}{R}\right)
\]

\[
PB_1 = r' - y\sin\theta_r
\]

\[
PB_2 = \frac{1}{2}y^2 \left(\frac{\cos^2\theta_r}{r'} - \frac{\cos\theta_r}{R}\right) + \frac{1}{2}y^3 \frac{\sin\theta_r}{r'} \left(\frac{\cos^2\theta_r}{r'} - \frac{\cos\theta_r}{R}\right) + \frac{1}{2}y^4 \frac{\sin^2\theta_r}{r'^2} \left(\frac{\cos^2\theta_r}{r'} - \frac{\cos\theta_r}{R}\right)
\]

\[
PB_3 = \frac{1}{2}z^2 \left(\frac{1}{r'} - \frac{\cos\theta_r}{R}\right) + \frac{zx}{r'} + \frac{c^2}{2r'}
\]

\[
PB_4 = \frac{1}{2}z^2 \frac{y\sin\theta_r}{r'} \left(\frac{1}{r'} - \frac{\cos\theta_r}{R}\right) + \frac{y\sin\theta_r}{2r'^2} \left(c^2 + 2zc'\right)
\]

\[
PB_5 = \frac{(y^2 + z^2)^2}{8R^2} \left(\frac{1}{r'} - \frac{\cos\theta_r}{R}\right)
\]

So finally we have an expanded expression for the light path function:

\[
F = AP_1 + AP_2 + AP_3 + AP_4 + AP_5 + \cdots + PB_1 + PB_2 + PB_3 + PB_4 + PB_5 + \cdots + \left(\frac{\gamma}{d}\right)m\lambda
\]

Because of the differing powers of \(R, r\) and \(r'\) in the successive terms, each term will have a very different magnitude, therefore we can subject each term separately to conditions 6, 7 and 8 (corresponding terms in the \(AP\) and \(PB\) series should be grouped together because they have similar magnitude). The terms of highest magnitude are \(AP_4, PB_1\) and \(\left(\frac{\gamma}{d}\right)m\lambda\). We can collectively call these terms \(F_1\).

\[
F_1 = r - y\sin\theta_i + r' - y\sin\theta_r + \left(\frac{\gamma}{d}\right)m\lambda = r + r' - y(\sin\theta_i + \sin\theta_r) + \left(\frac{\gamma}{d}\right)m\lambda
\]

\[
\frac{\partial F_1}{\partial y} = -(\sin\theta_i + \sin\theta_r) + \frac{m\lambda}{d} = 0
\]

\[
\frac{m\lambda}{d} = \sin\theta_i + \sin\theta_r
\]
The result is the grating equation for a plane grating, which I derived earlier using the geometry of the plane grating. Indeed, if we consider a plane grating, all higher order terms in the expansion of $F$ are zero, because $R$, $r$ and $r'$ are all infinity for the plane grating. This equation also applies to the curved grating, for a given wavelength incident at a given angle, the angular positions of regions of constructive interference can be calculated. Because of the absence of $r$ and $r'$ this equation gives no focusing conditions. In the case of the plane grating, rays are assumed to converge at infinity so one can merely observe the pattern far from the grating. For a curved grating, however, we can examine the terms which have the next highest magnitude, $AP_2$ and $PB_2$, which I will call $F_2$.

\[
F_2 = AP_2 + PB_2 = \frac{1}{2} y^2 \left(\frac{\cos^2 \theta_i}{r} - \frac{\cos \theta_i}{R}\right) + \frac{1}{2} y^3 \frac{\sin \theta_i}{r} \left(\frac{\cos^2 \theta_i}{r} - \frac{\cos \theta_i}{R}\right) + \cdots + 
\frac{1}{2} y^2 \left(\frac{\cos^2 \theta_r}{r'} - \frac{\cos \theta_r}{R}\right) + \frac{1}{2} y^3 \frac{\sin \theta_r}{r'} \left(\frac{\cos^2 \theta_r}{r'} - \frac{\cos \theta_r}{R}\right) + \cdots 
\]

(39)

\[
\frac{\partial F_2}{\partial y} = 0 = y \left(\frac{\cos^2 \theta_i}{r} - \frac{\cos \theta_i}{R} + \frac{\cos^2 \theta_r}{r'} - \frac{\cos \theta_r}{R}\right) + \frac{3}{2} y^2 \left(\frac{\sin \theta_i}{r} \left(\frac{\cos^2 \theta_i}{r} - \frac{\cos \theta_i}{R}\right) + \frac{\sin \theta_r}{r'} \left(\frac{\cos^2 \theta_r}{r'} - \frac{\cos \theta_r}{R}\right)\right) 
\]

(40)

The first term of this expression is zero if:

\[
\cos \theta_i \left(\frac{\cos \theta_i}{r} - \frac{1}{R}\right) + \cos \theta_r \left(\frac{\cos \theta_r}{r'} - \frac{1}{R}\right) = 0
\]

(41)

This relation can be satisfied if:

\[
r = R \cos \theta_i
\]

(42)

\[
r' = R \cos \theta_r
\]

(43)

It can be shown that the second term of equation 40 is also zero given equations 42 and 43. Equations 42 and 43 describe a circle, of radius $R/2$, with points $A$, $B$ and $P$ falling on the circumference of the circle. The diagram below shows this.
This result is significant; it means that if we place a curved grating of radius $R$ tangent to the circumference of a circle of radius $R/2$ and we place a source of radiation at another point on this circumference, then the interference maxima from the diffraction from the grating will focus to another point on the circumference. The circle, of radius $R/2$ is called the ‘Rowland Circle’ after the man who discovered its significance. So far I have just examined the two most significant terms in the expansion of $AP$ and $PB$. If these were the only terms then we could expect the focus positions from a diffraction experiment to fall exactly on the circumference of the Rowland Circle. However, there are higher order terms in the expansion of the light path function, so we should expect some deviation.

The terms of next highest significance are given by $AP_3$ and $PB_3$:

$$F_3 = AP_3 + PB_3 = \frac{1}{2} z^2 \left( \frac{1}{r} - \frac{\cos \theta_i}{R} + \frac{1}{r'} - \frac{\cos \theta_r}{R} \right) - \frac{zc}{r} + \frac{c^2}{2r} + \frac{zcr}{r'} + \frac{cr^2}{2r'} \quad (44)$$

These terms will slightly increase the light path function and will cause the focus positions to deviate slightly from the circumference of the Rowland circle. The values of $\theta_i, r', r'$ and $c'$ depend on the wavelength and order number, and $z$ can take any value on the surface of the grating. Therefore the expression for the exact expected deviation is complicated; it also depends on higher order terms in the expansion ($F_4, F_5, \ldots$).

It is the purpose of this experiment to investigate the deviation of the focus positions from the circumference of the Rowland circle. Possible reasons for the deviations will be discussed, which may be subsequently related to the expression for $F_3$ and higher order terms.

It should be noted at this stage, that higher order terms such as $F_4$ and $F_5$ not only add to the light path function but also define the coma and aberration of the image when they are subject to the conditions 6, 7 and 8. The coma and aberration of the image place constraints on the design parameters of the grating. I will not discuss these terms further here.

**Experiment**

**Apparatus**

The diagram below shows the X-ray spectrometer used in this experiment.

---


Photons incident on the slit on the right came from the solid samples which were held at a vacuum inside a vessel where they were targeted with incoming electrons/photons. The samples included oxygen and zinc. The diagram shows three gratings, as there are locations for three; however we just used one in our experiment, with the following properties:

Strips/mm: 1200  
Radius of curvature: 5m  
Angle of incidence: 88.1°

The detector is a micro channel plate detector (MCP). Each photon hits one of the channels on the plate, where it loses its energy to an electron, this in turn loses energy to another electron, and the process continues so that each single photon leads to a cascade of secondary electrons. These electrons illuminate a fluorescent screen which is detected with a camera. The camera is connected to a computer program which displays the number of photon counts on each channel of the MCP.

Method

The first task was to find focus positions for a range of spectra. The spectra appeared as a number of photon counts on each channel of the MCP. A perfect focus position would be where all the photons struck one channel of the MCP; however, both for theoretical reasons which I have discussed and experimental reasons, we could expect a spread of the photons across a number of channels. I define the focus position as being the spatial position of the detector which gave rise to the minimum spread of photon counts across the channels (i.e. the minimum standard deviation on a graph of counts against channel number). For each sample and order number, this was the method for finding the focus position:

1. Use the grating equation (equation 2) to calculate the angle of diffraction. Wavelength $\lambda$ was found using known x-ray emission energies and the following equation relating the wavelength and energy of a photon:

$$ E = \frac{hc}{\lambda} $$

2. Use the geometry of the Rowland Circle to find the theoretical focus position on the circumference of the Rowland Circle.

Note: the X-Y co-ordinate system which our detector position was measured in was offset from the co-ordinate system with the grating at the origin, so the appropriate adjustment had to be made. The diagram below shows how they are orientated relative to one another. Our x-axis and our grating were angled 15° and 1.1° respectively to a fixed slab.
This information alone was not enough to translate positions from the grating frame of reference to the detector frame of reference. It was also necessary to take one experimental focus position and assume zero error (i.e. assume the experimental position was also the theoretical position). The focus position I chose to assume zero error was 2\textsuperscript{nd} order Oxygen because the we took the most data points when calculating this focus position so it is likely that this was the most accurate.

3. Place detector in theoretical x-position and adjust y-position so the photon-count peak falls roughly in the centre of the detector (middle channel number).
4. Take a photon count for 5 minutes
5. Move the detector 5mm in the x-direction, adjust the y-position accordingly, take another count for 5 minutes
6. Repeat step 5 on either side of the theoretical x-position appropriately such that data exists on either side of the focus position
7. In excel, plot standard deviation vs. x-position and standard deviation vs. y-position.
8. In each graph, plot two straight lines of best fit, for points on either side of the focus position.
9. Use the equations of these lines to see where they intersect. Points of intersection give the x- and y-positions of experimental focus

An example of this method will be displayed in the results section. The next task was to see how the experimental focus positions deviated from their theoretical positions on the circumference of the Rowland circle. For each experimental focus position found, its x- and y-co-ordinates were compared to the theoretical x- and y-co-ordinates of focus. The difference in these values was plotted as a function of the position to look for trends in the deviation.

**Results**

**Focus positions**

3\textsuperscript{rd} order Oxygen

Here I will present the results, step-by-step, for the focus position of 3\textsuperscript{rd} order Oxygen:
Steps 1 and 2

<table>
<thead>
<tr>
<th>element</th>
<th>Oxygen</th>
</tr>
</thead>
<tbody>
<tr>
<td>energy (eV)</td>
<td>524.9</td>
</tr>
<tr>
<td>energy (J)</td>
<td>8.40982E-17</td>
</tr>
<tr>
<td>order</td>
<td>3</td>
</tr>
<tr>
<td>angle of diffraction (rad)</td>
<td>-1.436134751</td>
</tr>
<tr>
<td>x theoretical (grating frame)</td>
<td>-0.665197624</td>
</tr>
<tr>
<td>y theoretical (grating frame)</td>
<td>0.09012197</td>
</tr>
<tr>
<td>x theoretical (instrument frame)</td>
<td>-0.007171319</td>
</tr>
<tr>
<td>y theoretical (instrument frame)</td>
<td>-0.014514598</td>
</tr>
</tbody>
</table>

The difference between the theoretical positions in the grating frame and in the instrument frame are due to these two frames of reference being offset, as explained previously.

Steps 7 and 8

The difference between the theoretical positions in the grating frame and in the instrument frame are due to these two frames of reference being offset, as explained previously.

The difference between the theoretical positions in the grating frame and in the instrument frame are due to these two frames of reference being offset, as explained previously.

The difference between the theoretical positions in the grating frame and in the instrument frame are due to these two frames of reference being offset, as explained previously.

The difference between the theoretical positions in the grating frame and in the instrument frame are due to these two frames of reference being offset, as explained previously.

The difference between the theoretical positions in the grating frame and in the instrument frame are due to these two frames of reference being offset, as explained previously.
Step 9

Finding the co-ordinates where the lines in the above graphs meet give us the focus position:

<table>
<thead>
<tr>
<th>x focus, instrument frame (cm)</th>
<th>-1,20078</th>
</tr>
</thead>
<tbody>
<tr>
<td>y focus, instrument frame (mm)</td>
<td>-5,95132</td>
</tr>
</tbody>
</table>

In Uppsala I helped find four focus positions; Oxygen 2nd order, 3rd order, 4th order and Zinc 4th order. The positions are presented below, the corresponding graphs of standard deviation vs. x- and y-position can be found in Appendix 2.

<table>
<thead>
<tr>
<th>element</th>
<th>Zn</th>
<th>O</th>
<th>O</th>
<th>O</th>
</tr>
</thead>
<tbody>
<tr>
<td>order</td>
<td>4</td>
<td>2</td>
<td>3</td>
<td>4</td>
</tr>
<tr>
<td>x experimental (instrument frame)</td>
<td>0,120499763</td>
<td>0,108151227</td>
<td>-0,01200783</td>
<td>-0,104287619</td>
</tr>
<tr>
<td>y experimental (instrument frame)</td>
<td>-0,010167548</td>
<td>-0,010504908</td>
<td>-0,005951321</td>
<td>-0,007718147</td>
</tr>
</tbody>
</table>

Deviation of experimental positions from theoretical positions

In order to examine how these experimental positions deviated from the theoretical positions, I plotted the error (theoretical position – experimental position) against the x- or y-value:

![x error vs x](image-url)
It is difficult to see a trend in the x-error, however there does appear to be a linear relationship between the y-error and the y position, which I have fitted a line to. It is possible that there is a similar relationship between the x-error and x-position which we cannot see because we only have four data points.

**Predicted Positions**

The group who will perform the stimulated RIXS experiment at LCLS require a method of quickly and accurately predicting focus positions in order that they do not waste time finding this positions. Having found 4 focus positions in Uppsala using electron scattering, my task was to produce a spreadsheet in which one could enter the element, known energy gap between valence and core and the order number being looked for. My spreadsheet should then predict the focus position. Given the linear nature of the y-error graph I will assume a linear relationship for x-error against x, hence I have added a line of best fit to this graph. I can now predict positions in the following way:

\[
\begin{align*}
    x_{predicted} &= x_{theoretical} + \Delta x \\
    &= x_{theoretical} + mx_{theoretical} + c \\
    &= x_{theoretical}(1 + m) + c
\end{align*}
\]

where \( m \) and \( c \) are the gradient and intercept respectively of the relevant error versus position graph.

I designed my spreadsheet such that when a new focus position had been found at LCLS, a new data point could be added to the error versus position graph, updating its gradient and intercept, thus
creating a more accurate prediction of the next focus position. This is an extract from my spreadsheet, showing the data from my experiment and space for new data points to be added:

<table>
<thead>
<tr>
<th>angle of incidence (°)</th>
<th>88.1</th>
</tr>
</thead>
<tbody>
<tr>
<td>angle of incidence (rad)</td>
<td>1.53763507</td>
</tr>
<tr>
<td>d (grating)</td>
<td>8.33333E-07</td>
</tr>
<tr>
<td>h</td>
<td>6.63E-34</td>
</tr>
<tr>
<td>c</td>
<td>299792458</td>
</tr>
<tr>
<td>radius of grating (m)</td>
<td>5</td>
</tr>
<tr>
<td>radius of Rowland circle (m)</td>
<td>2.5</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>element</th>
<th>Zn</th>
<th>O</th>
<th>O</th>
<th>O</th>
</tr>
</thead>
<tbody>
<tr>
<td>energy (eV)</td>
<td>1034.7</td>
<td>524.9</td>
<td>524.9</td>
<td>524.9</td>
</tr>
<tr>
<td>energy (J)</td>
<td>1.65777E-16</td>
<td>8.40982E-17</td>
<td>8.40982E-17</td>
<td>8.40982E-17</td>
</tr>
<tr>
<td>order</td>
<td>4</td>
<td>2</td>
<td>3</td>
<td>4</td>
</tr>
<tr>
<td>angle of diffraction (rad)</td>
<td>-1.45847471</td>
<td>-1.45921524</td>
<td>-1.43613475</td>
<td>-1.41645081</td>
</tr>
<tr>
<td>x theoretical (grating frame)</td>
<td>-0.55689639</td>
<td>-0.55328618</td>
<td>-0.66519762</td>
<td>-0.75952949</td>
</tr>
<tr>
<td>y theoretical (grating frame)</td>
<td>0.06281588</td>
<td>0.06199376</td>
<td>0.09012197</td>
<td>0.11816983</td>
</tr>
<tr>
<td>x theoretical (instrument frame)</td>
<td>0.10445461</td>
<td>0.10815122</td>
<td>-0.00717131</td>
<td>-0.10558149</td>
</tr>
<tr>
<td>y theoretical (instrument frame)</td>
<td>-0.01071619</td>
<td>-0.01050490</td>
<td>-0.01451459</td>
<td>-0.01372640</td>
</tr>
<tr>
<td>x experimental (instrument frame)</td>
<td>0.12049976</td>
<td>0.10815122</td>
<td>-0.01200783</td>
<td>-0.10428761</td>
</tr>
<tr>
<td>y experimental (instrument frame)</td>
<td>-0.01016754</td>
<td>-0.01050490</td>
<td>-0.00595132</td>
<td>-0.00771814</td>
</tr>
<tr>
<td>x error</td>
<td>0.01604514</td>
<td>0</td>
<td>-0.00483651</td>
<td>0.00129387</td>
</tr>
<tr>
<td>y error</td>
<td>0.00054864</td>
<td>3.29597E-17</td>
<td>0.00856327</td>
<td>0.00600825</td>
</tr>
<tr>
<td>x predicted (instrument frame)</td>
<td>0.11076413</td>
<td>0.11460972</td>
<td>-0.00536032</td>
<td>-0.10773643</td>
</tr>
<tr>
<td>y predicted (instrument frame)</td>
<td>-0.01027089</td>
<td>-0.01048834</td>
<td>-0.00636157</td>
<td>-0.00717278</td>
</tr>
<tr>
<td>gradient of x error graph</td>
<td>0.0403</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>constant of x error graph</td>
<td>0.0021</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>gradient of y error graph</td>
<td>-2.0292</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>constant of y error graph</td>
<td>-0.0213</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The instructions for using my spreadsheet are given as follows:

1. Enter element and energy of x-ray emission in eV
2. Enter the order of diffraction which you plan to observe
3. Use 'x predicted' and 'y predicted' cells to find the expected focus position, based on theoretical position and linear error function
4. Once an experimental position is found, enter this into the appropriate cell (IN METRES)
5. Set the data on the x-error graph (y-error graph) to include the new point of x error (y error) against x theoretical instrument frame (y theoretical instrument frame)
6. Using the new lines on the graphs, enter the gradients and constants of these lines, so that more accurate predicted positions are located.
The spreadsheet above was given to the group who travelled to LCLS, Stanford. They were able to locate three positions, of oxygen (2nd and 3rd order) and nickel (4th order) and use the data from valence to core de-excitations to explore the atomic structure of the materials used.

Interpretation of results, conclusions

Reasons for error function

There are a couple of possible explanations for the deviation of the focus positions from the circumference of the Rowland circle. The first explanation is theoretical. When I showed that the focus position would like on the circumference of the Rowland circle, only the first two terms of the light path function were considered. As is shown in equation 44 the next most significant term of the light path function has terms which depend on \( r', \theta, \) and \( c' \) which are all different for each focus positions. This term of the light path function depends on these terms in a complicated way which is difficult to predict, but it is possible that our (roughly) linear error function is due to the combined dependence of the third term of the light path function on these different parameters.

Equation 35 also shows that there are higher order terms in the light path function. These terms are less significant but may also play a role in our experimental error function.

There are also some experimental explanations for the error which I have observed. The most significant is the possibility that the slit in our experimental set up was not actually the source for the optics. In a perfect scenario, the slit would be a point source; however our source has a small width, meaning that the source of the optics may be slightly behind the slit. In order for the focus position to be on the Rowland circle the source must also be, so if the source deviated from the circumference of the Rowland circle, the focus position would also deviate.

There are other experimental reasons which could lead to error in the focus positions. For example when we examined the number of photon counts on the MCP, there was a spread of results. We placed the detector such that the peak of this spread was in the central channel. However, the spread was not a perfect Gaussian curve so it was difficult to precisely locate the central point in the spread of results.

Improvements to the experimental procedure

There are some improvements that could be made to the experimental procedure in order to make more accurate predictions for the focus positions. We could reduce the width of the slit to be surer that this was acting as the source of the optics. This would lead to less photons incident on the grating and thus on the MCP, therefore we would have to take statistics for longer in order to find the focus positions. This leads me to another experimental improvement; taking data for a longer period of time. We were somewhat time-constricted in this experiment because of the need for its completion before the group travelled to LCLS. Given more time, we could take statistics for longer, the spread of photon counts would be a smoother curve, thus enabling a more accurate placement of the centre of the peak in the middle of the detector.

As I have explained, when a new focus position is found, its error can be added to the graph, the line of which helps us more accurately predict the next focus position. Therefore, by finding focus positions for more samples, we could more accurately predict further positions.
Conclusion

By describing the theory of the curved diffraction grating, I showed that if we neglect higher order terms of the light path function, the constructively interfering radiation will focus onto the circumference of the Rowland circle (which has half the radius of the curved grating). However, we can expect this not to be perfectly true if we consider higher order terms. Experimentally I have shown that this is indeed not true. There is a deviation in both the x- and y-position focus positions from their theoretical position on the Rowland circle. The y-error has a linear relationship to the y-position. By assuming a similar linear relationship for the x-error I have produced a self-updating spreadsheet which enables more focus positions to be predicted, taking into account their expected deviation from the Rowland circle.

Bibliography


Såthe, C., 2011, Applications of Soft X-Ray Spectroscopy, Uppsala, ACTA

Appendices

Appendix 1: Expansion of \((AP)^2\) and \((PB)^2\) (main reference: Beutler, H., The theory of the concave grating)

From equations 19 and 20 we have:

\[(AP)^2 = r^2 + c^2 - 2r \sin \theta_i y - 2cz + (2R - 2a)x\]  \hspace{1cm} (47)

\[(PB)^2 = r'^2 + c'^2 - 2r' \sin \theta_i y + 2c'z + (2R - 2a')x\]  \hspace{1cm} (48)

I will work through \((AP)^2\) first and then \((PB)^2\), which follows the same process. By substituting in the expansion of \(x\) from equation 22 into equation 47 we obtain:

\[(AP)^2 = r^2 + c^2 - 2r \sin \theta_i y - 2cz + (2R - 2a) \left( \frac{y^2 + z^2}{2R} + \frac{(y^2 + z^2)^2}{8R^3} + \frac{(y^2 + z^2)^3}{16R^5} + \cdots \right)\]  \hspace{1cm} (49)

\[(AP)^2 = r^2 + c^2 - 2r \sin \theta_i y - 2cz + \left(1 - \frac{r \cos \theta_i}{R}\right) \left( y^2 + z^2 + \frac{(y^2 + z^2)^2}{4R^2} + \frac{(y^2 + z^2)^3}{8R^4} + \cdots \right)\]  \hspace{1cm} (50)
We can approximate the square root by series developments\textsuperscript{10}, giving:

\[ (AP)^2 = r^2 - 2r \sin \theta_i y + y^2 (\sin^2 \theta_i + \cos^2 \theta_i) - y^2 \frac{r \cos \theta_i}{R} + z^2 \left( 1 - \frac{r \cos \theta_i}{R} \right) + c^2 - 2cz + \left( 1 - \frac{r \cos \theta_i}{R} \right) \left( y^2 + z^2 \right)^2 \left( \frac{1}{4R^2} + \frac{(y^2+z^2)^3}{8R^4} + \cdots \right) \]  
\[ (AP)^2 = \left( r - y \sin \theta_i \right)^2 + y^2 \left( \cos^2 \theta_i - \frac{r \cos \theta_i}{R} \right) + z^2 \left( 1 - \frac{r \cos \theta_i}{R} \right) + c^2 - 2cz + \left( \frac{(y^2+z^2)^2}{4R^2} \left( 1 - \frac{r \cos \theta_i}{R} \right) + \frac{(y^2+z^2)^3}{8R^4} \left( 1 - \frac{r \cos \theta_i}{R} \right) + \cdots \right) \]  

We can approximate the square root by series developments\textsuperscript{10}, giving:

\[ AP = r - y \sin \theta_i + \frac{1}{2} y^2 \left( \cos^2 \theta_i - \frac{r \cos \theta_i}{R} \right) + \frac{1}{2} z^2 \left( 1 - \frac{r \cos \theta_i}{R} \right) - \frac{1}{2} \left( \cos^2 \theta_i - \frac{r \cos \theta_i}{R} \right)^2 + \cdots \]
\[ AP = r - y \sin \theta_i + \frac{1}{2} y^2 \left( \cos^2 \theta_i - \frac{r \cos \theta_i}{R} \right) + \frac{1}{2} z^2 \left( 1 - \frac{r \cos \theta_i}{R} \right) - \frac{1}{2} \left( \cos^2 \theta_i - \frac{r \cos \theta_i}{R} \right)^2 + \cdots \]
\[ \frac{(y^2+z^2)^2}{8R^2} \left( 1 - \frac{r \cos \theta_i}{R} \right) - \cdots + \frac{1}{2} y^2 \frac{\sin \theta_i}{r} \left( \cos^2 \theta_i - \frac{r \cos \theta_i}{R} \right) + \frac{1}{2} z^2 \frac{y \sin \theta_i}{r} \left( 1 - \frac{r \cos \theta_i}{R} \right) - \cdots + \frac{1}{2} y^4 \frac{\sin^2 \theta_i}{r^2} \left( \cos^2 \theta_i - \frac{r \cos \theta_i}{R} \right)^2 + \cdots \]

The terms are presented separately in the main text. Here is the same process for \((PB)^2\):

\[ (PB)^2 = r'^2 + c'^2 - 2r' \sin \theta_r y + 2c'z + (2R - 2a') \left( \frac{y'^2+z'^2}{2R} + \frac{(y'^2+z'^2)^2}{8R^3} + \cdots \right) \]  
\[ (PB)^2 = r'^2 + c'^2 - 2r' \sin \theta_r y + 2c'z + (1 - \frac{r' \cos \theta_r}{R}) \left( y'^2 + z'^2 + \frac{(y'^2+z'^2)^2}{4R^2} + \cdots \right) \]  
\[ (PB)^2 = r'^2 - 2r' \sin \theta_r y + y^2 (\sin^2 \theta_r + \cos^2 \theta_r) - y^2 \frac{r' \cos \theta_r}{R} + z^2 \left( 1 - \frac{r' \cos \theta_r}{R} \right) + c'^2 + 2c'z + \left( 1 - \frac{r' \cos \theta_r}{R} \right) \left( y'^2 + z'^2 \right)^2 \left( \frac{1}{4R^2} + \frac{(y'^2+z'^2)^3}{8R^4} + \cdots \right) \]  

(PB)^2 = (r' - ysin\theta_r)^2 + y^2\left(\cos^2\theta_r - \frac{r'\cos\theta_r}{R}\right) + z^2\left(1 - \frac{r'\cos\theta_r}{R}\right) + c'^2 + 2c'z + \frac{(y^2+z^2)^2}{4R^2}\left(1 - \frac{r'\cos\theta_r}{R}\right) + \frac{(y^2+z^2)^3}{8R^4}\left(1 - \frac{r'\cos\theta_r}{R}\right) + \cdots \tag{58}

PB = r' - ysin\theta_r + \frac{1}{2}y^2\left(\cos^2\theta_r - \frac{r'\cos\theta_r}{R}\right) + \frac{1}{2}y^2\left(1 - \frac{r'\cos\theta_r}{R}\right) + \frac{1}{8}(c'^2 + 2cz) + \frac{(y^2+z^2)^2}{8R^2}\left(1 - \frac{r'\cos\theta_r}{R}\right) + \frac{(y^2+z^2)^3}{8R^4}\left(1 - \frac{r'\cos\theta_r}{R}\right) + \cdots \tag{59}

PB = r' - ysin\theta_r + \frac{1}{2}y^2\left(\cos^2\theta_r - \frac{r'\cos\theta_r}{R}\right) + \frac{1}{2}y^2\left(1 - \frac{r'\cos\theta_r}{R}\right) + \frac{1}{2}y^2\left(\frac{1}{r'} - \frac{r'}{R}\right) + \frac{1}{2}y^2\left(\sin\theta_r\frac{1}{r'} - \frac{1}{R}\right) + \frac{1}{2}y^2\left(\frac{1}{r'} - \frac{r'}{R}\right) + \cdots + \frac{1}{2}y^2\left(\sin^2\theta_r\frac{1}{r'} - \frac{r'}{R}\right) + \frac{1}{2}y^2\left(\frac{1}{r'} - \frac{r'}{R}\right) + \cdots \tag{60}

Appendix 2: Standard deviation vs. x- and y- position for Oxygen 2\textsuperscript{nd} order, Oxygen 4\textsuperscript{th} order and Zinc 4\textsuperscript{th} order
O 2nd s.dev vs y

\[ y = 2.6966x + 78.207 \]

\[ y = -2.4799x + 22.494 \]

O 4th s.dev vs x

\[ y = -3.3744x + 16.029 \]

\[ y = 0.9007x + 60.613 \]
O 4th s.dev vs y

\[ y = -0.7616x + 45.298 \]

\[ y = 2.5607x + 70.94 \]

Zinc 4th s.dev vs x

\[ y = -7.9276x + 131.74 \]

\[ y = 4.2862x - 15.436 \]
Zinc 4th s.dev vs. $y$

$y = 4.5315x + 82.146$

$y = -2.4814x + 10.842$

Standard deviation (number of channels)

$y$ (mm)