Nucleon-Induced Fission Cross Sections of Heavy Nuclei in the Intermediate Energy Region

BY

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ABSTRACT


Fission is the most important nuclear reaction for society at large today due to its use in energy production. However, this has raised the problem of how to treat the long-lived radioactive waste from nuclear reactors. A radical solution would be to change the composition of the waste into stable or short-lived nuclides, which could be done through nuclear transmutation. Such a concept requires accelerator-driven systems to be designed, where those for transmutation are reactor hybrids. This thesis is a contribution to the knowledge base for developing transmutation systems, specifically with respect to the computational modeling of the underlying nuclear reactions, induced by the incident and secondary particles. Intermediate energy fission cross sections are one important type of such data. Moreover, they are essential for understanding the fission process itself and related nuclear interactions.

The experimental part of this work was performed at the neutron beam facility of the The Svedberg Laboratory in Uppsala. Fission cross sections of $^{238}$U, $^{209}$Bi, $^{208}$Pb, $^{197}$Au, $^{nat}$W, and $^{181}$Ta were measured for neutrons in the range $E_n = 30-160$ MeV using thin-film breakdown counters for the fission fragment detection. A model was developed for the determination of the efficiency of such detectors.

A compilation of existing data on proton-induced fission cross sections for nuclei from $^{165}$Ho to $^{239}$Pu was performed. The results, which constitute the main body of information in this field, were added to the worldwide EXFOR database. The dependences of the cross sections on incident energy and target nucleus were studied, which resulted in systematics that make it possible to give estimates for unmeasured nuclides.

Nucleon-induced fission cross sections were calculated using an extended version of the cascade exciton model. A comparison with the systematics and the experimental data obtained in the present work revealed significant discrepancies. A modification of the model was made that significantly improved the predictions.

Key words: Fission, cross section, (n,f) reaction, (p,f) reaction, intermediate energy, database, nuclear reaction, neutron.

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"To work well means to work a lot"

Isaac Newton

To my Teachers
The present thesis is based on the following papers, which will be referred to in the
text by the Roman numerals I-V.

I. V.P. Eismont, A.V. Prokofiev, A.N. Smirnov, K. Elmgren, J. Blomgren, H.
Condé, J. Nilsson, N. Olsson, T. Rönnqvist, and E. Tranéus, Relative and
Absolute Neutron Induced Fission Cross Sections of $^{208}\text{Pb}$, $^{209}\text{Bi}$ and $^{238}\text{U}$ in

II. A.V. Prokofiev, Compilation and Systematics of Proton-Induced Fission Cross
Section Data. Nuclear Instruments and Methods in Physics Research A (in

III. A.V. Prokofiev, S.G. Mashnik, and A.J. Sierk, Cascade-Exciton Model
Analysis of Nucleon-Induced Fission Cross Sections of Lead and Bismuth at
Energies from 45 to 500 MeV. Nuclear Science and Engineering 131 (1999)
78.

IV. A.V. Prokofiev and N. Olsson, Fission Fragment Detection Efficiency of
Thin-Film Breakdown Counters in Sandwich Geometry. Uppsala University

V. A.V. Prokofiev, P.-U. Renberg, and N. Olsson, Measurement of Neutron-
Induced Fission Cross Sections for $^{nat}\text{Pb}$, $^{208}\text{Pb}$, $^{197}\text{Au}$, $^{nat}\text{W}$, and $^{181}\text{Ta}$ in the
Intermediate Energy Region. Uppsala University Neutron Physics Report UU-
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Other related publications not included in this thesis.


In addition, there are 12 contributions to international conferences published in conference proceedings.
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1 Introduction

Fission is the most important nuclear reaction for society at large today due to its use in nuclear power plants. In these, $^{235}\text{U}$ is used as a source of energy, which is released through neutron chain reactions. The uranium reactors are driven by fission neutrons of low energy ($E_n < 2 \text{ MeV}$), and the corresponding research field of neutron reactions is very well studied.

Fission reactions can also be induced by intermediate energy neutrons and protons. These are of interest in their own right for the purpose of providing nuclear physics knowledge, which is at the same time the knowledge base for applications in transmutation of nuclear waste using accelerator driven systems (ADS), as well as in beam monitoring or dosimetry for radiation protection, cancer therapy, studies of particle-induced effects in electronics (single event effects, SEE), etc.

This thesis deals with experimental studies, phenomenological systematics, and theoretical model calculations for fission reactions induced by intermediate energy neutron and proton beams. They have involved the determination of fission cross sections and their dependence on projectile energy, and on charge and mass of the target nucleus. The overall motivation and guide for these studies have been application considerations, especially in the field of ADS. The studies have been made possible through development of measurement techniques, especially with respect to the use of thin-film breakdown counters (TFBC) for fission fragment detection. Along this line, detector calibration problems have been considered, and data analysis and interpretation models have been developed.

My contribution to the project is as follows. Paper II has been published by me as a single author. In Papers III and IV, I did the major part of work, in both cases with proper supervision and/or advice from the co-authors. Finally, in the experimental studies at TSL, discussed in Papers I and V, my role was writing the on-line and off-line software, responsibility for data acquisition, participation in setting up the in-beam arrangements and the electronics, and participation in beamtime, off-line analysis, and in preparation of publications.
2 Properties of the fission reaction

Fission is a spectacular nuclear reaction of great practical importance. In 1938, Hahn and Strassmann [1] irradiated uranium with neutrons and found that it results in isotopes of elements that are not neighbouring to the target nucleus, but belong to the middle of the periodic table (e.g., barium). Meitner and Frisch [2] proposed an explanation of the new phenomenon similar to the splitting of a vibrating liquid drop into two smaller ones. They proposed the term "fission" in analogy to the division of biological cells. The first complete theoretical description of the fission process was published by Bohr and Wheeler [3] shortly after the discovery.

In 1940, Petrzhak and Flerov [4] found that fission of some heavy nuclei may occur spontaneously, without any external stimulation. A well-known example is $^{252}$Cf. As a general trend, nuclei become less and less stable with respect to spontaneous fission with increasing fissility parameter, which is proportional to $Z^2/A$, where $Z$ and $A$ are the charge and mass of the nucleus, respectively. Thus, it is in fact spontaneous fission that limits the periodic table of the elements.

It is easy to understand why fission is energetically possible. The binding energy per nucleon of medium-weight nuclei is about 8.5 MeV, while in the uranium region it is only 7.6 MeV. If a heavy nucleus is split into two fragments, the total binding energy will be increased by about 200 MeV, which is a very large value in comparison with other nuclear reactions. The major part of this energy is released in the form of kinetic energy of the resulting fragments. The latter have usually an excess of neutrons, and therefore fission is accompanied by neutron emission, which can take place either promptly or after a beta-decay of the fission products (so called delayed neutrons). The latter phenomenon makes it possible to control the chain fission reaction at nuclear power plants reliably.

The fission cross section, which expresses the probability of fission induced by, e.g., neutrons, depends strongly on the specific nucleus and incident energy. Neutrons with energies up to a few MeV, which are dominating in spectra from fission reactions, are able to induce fission only in heavy nuclei (e.g., actinides). Fission of some heavy nuclei (e.g., $^{233}$U, $^{235}$U, $^{239}$Pu) can be induced by neutrons of very low energy, and in particular by thermal neutrons ($E_n = 0.025$ eV). The corresponding fission cross section is very high (e.g., ~600 b for $^{235}$U). As a general trend, the cross section decreases with increasing energy above the thermal value, but in addition, there is a region where it fluctuates strongly with energy. This so-called resonance region extends, e.g., from ~0.5 eV to ~1 keV for $^{235}$U. With further increase of the incident energy up to a few MeV, fission becomes energetically possible even after emission of a light particle (usually a neutron). This "second-chance fission" is manifested by a step in the dependence of the fission cross section on the incident energy. With further increase of the incident energy, higher multiple-chance fission channels open up, and thus a stepwise structure is observed in the excitation function.

Nuclei in the sub-actinide region possess larger fission barriers (e.g., ~20-25 MeV for bismuth and lead isotopes [5]), and therefore fission can be induced only by particles of a higher energy. The TSL neutron beam with energies up to 180 MeV has made it possible to study fission of nuclei in a wide region of atomic number, from the heaviest ones and down to $^{181}$Ta. Moreover, there is no doubt that fission of even lighter nuclei can be observed using a neutron beam with sufficiently high energy and intensity.

Fission can be induced not only by neutrons, but also by protons, light and heavy ions, photons, electrons, $\pi$-mesons etc. Fission of such a light element as nickel ($Z=28$) by 1-GeV protons has for instance been studied [6]. The fission mechanism remains in operation up to energies as high as 300 GeV. At high energies, it can result in three and even four comparable fragments with a measurable probability (see, e.g., [7]).
The particle that induces fission may leave a part of its linear momentum to the fissioning nucleus, or transfer the momentum completely, which corresponds to fission of the true compound nucleus. Due to this effect of linear momentum transfer (LMT), the folding angle between two complementary fission fragments is no longer equal to $180^\circ$ in the laboratory frame, and fragments are preferentially emitted in the forward hemisphere. Measurements of the distribution of fission fragments with respect to the folding angle (see, e.g., [8]) provide LMT data, which are of interest not only in fission physics, but also for fundamental theories of nuclear reactions.

Another closely related effect is the anisotropy of the fission fragment angular distribution in the frame of the fissioning nucleus. The fact that more fragments are emitted at $0$ and $180^\circ$ than at $90^\circ$ with respect to the incident beam direction, is a consequence of the angular momentum transfer. The theory of anisotropy, developed by Halpern and Strutinsky [9] and more recently by Ignatyuk and co-workers (see, e.g., [5]), links the experimental observables to fundamental nuclear quantities, e.g., the nuclear moment of inertia.

During the last decade, great interest has been paid to dynamic effects in the fission process, i.e., effects which are not taken into account in the statistical description of the process. An appropriate way to account for fission dynamics is to consider fission as a diffusion process over the fission barrier. In the framework of such an approach, suggested long ago by Kramers [10], the fission width, which serves as a measure of the fission probability, depends on a dissipation coefficient, which characterizes the viscosity of nuclear matter. In addition, formation of such a large-amplitude collective motion as the fission process requires a finite time, and during that time fission will be suppressed, while competing decay channels are active. Calculations of Grange and Weidenmüller [11] show that these effects grow rapidly with increasing excitation energy of the fissioning nucleus. A number of dynamic effect studies were performed with high-energy heavy ion beams (see, e.g., [12]). Taking such effects into account has been found to be necessary also for the description of nucleon-induced fission at intermediate energies (see, e.g., a recent evaluation of Ignatyuk et al. [13]).

In the past, the most extensive studies were performed for neutron-induced fission at low energies, especially for nuclides that are relevant for nuclear power engineering. Consequently, evaluated neutron data libraries have been produced, which traditionally have covered the energy region up to 20 MeV. During the last decades, such studies have largely shifted to higher energies. Besides the theoretical interest, this is motivated by requests due to new applications of the fission reaction (see Sect. 3). Yet, neither experimental nor theoretical studies of fission performed at intermediate energies have been sufficient to meet these requests. Even the fission cross section, which represents a very basic characteristic of the reaction, is often unmeasured and cannot be predicted with the precision that satisfies the practical needs, especially for incident neutrons. As a response to this, the present work includes experimental studies of (n,f) cross sections for a number of target nuclei. The experimental information on proton fission cross sections is more extensive, but it has never been thoroughly overviewed. This has been done in another part of this thesis (see Sect. 4.2), resulting in systematics of the (p,f) cross sections for a wide range of energy and nuclei.
3 New applications of nucleon-induced fission reactions

The traditional application of fission for energy production has raised the problem of how to treat the long-lived radioactive waste from nuclear reactors. A radical solution would be to change the composition of the waste into stable or short-lived nuclides, which could be done through nuclear transmutation. A large majority of the transmutation concepts (see, e.g., [14]) suggests the construction of accelerator-driven systems (ADS), which would combine the features of high-energy accelerators and nuclear reactors.

The basic idea of ADS is to feed a subcritical reactor core with external neutrons, produced via spallation of heavy nuclei by a high-intensity ion beam. The prospective spallation target materials are thorium, lead, and lead-bismuth eutectics; in addition, mercury, tungsten, and tantalum are often considered. The optimal incident particles are protons or deuterons. A beam of high energy (~1-2 GeV) and intensity (20-100 mA) is required to produce the neutrons, which could have a flux a few orders of magnitude larger than in conventional power reactors. The resulting neutron spectrum has a peak at an energy of a few MeV, and extends up to about the primary beam energy. The neutrons are subsequently used in the surrounding reactor core. The core can be loaded with, e.g., long-lived isotopes of actinide elements, or with fission products from spent fuel. Neutron capture or fission reactions can then result in short-lived or stable nuclides. The excessive released energy could, in turn, be delivered to the electric power grid. In a longer perspective, similar methods might be used for accelerator-driven energy production.

The advantage of accelerator-driven transmutation over other related concepts is that it allows incineration of long-lived minor actinides that are difficult to use as fuel in facilities based on self-supporting chain reactions. For instance, some nuclides are troublesome to transmute in conventional reactors because of the very low fraction of delayed neutrons, which makes the reactivity control difficult. Another advantage of ADS is that the associated reactor can be operated in a subcritical mode, which improves the inherent safety of the system.

Fission is one of the important processes that occur in the spallation target and the reactor core. The fission channel contributes to the radioactivity produced in the target, as well as to the chemical and radiological toxicity of the reaction products. For example, fission products in a lead target irradiated by 1.6-GeV protons will contribute 10-15% of the overall residual activity after one year of cooling [15].

The techniques used in the present work to measure intermediate-energy nucleon-induced fission cross sections are useful in several other applications, mainly as a tool for beam monitoring or for radiation dosimetry. This is especially true for neutrons, which have no charge and therefore do not ionize, thus making them difficult to detect in a reliable way.

The low sensitivity of thin-film breakdown counters (TFBCs) to gamma radiation makes them, in conjunction with fissile samples, a good choice for neutron and proton dosimetry in situations where gamma rays are present. This is the case in, e.g., fast neutron therapy, which is employed at more than a dozen hospitals or institutes around the world [16]. At those facilities, a 50 - 70 MeV proton or deuteron beam is incident upon a thick target of lithium or beryllium. The resulting continuous neutron spectrum, extending up to the incident beam energy, is accompanied by an intense flux of gamma rays, which also contributes to the total dose. To be able to single out the neutron contribution is of course of great value.

Recently, the dose received by aircrew due to cosmic radiation has been discussed. Cosmic-ray protons, continuously bombarding the atmosphere, create secondary neutrons by spallation reactions in the atomic nuclei of the air constituents. These neutrons show essentially a 1/E spectrum, extending up to several GeV. The neutrons can penetrate deep into the atmosphere, and are especially troublesome at altitudes of 10 - 30 km. For aircrew,
spending a large fraction of their time at altitudes of 10 km or more, the neutrons give a substantial fraction of - or even dominate - the total annual dose. The TFBC technique, using fission reactions in $^{209}$Bi, has recently been employed to calibrate dosimeters for such applications [17].

The neutrons created in the atmosphere have sufficient energy and intensity to be a potential threat not only for the aircrew, but also for electronic components in the aircraft. These neutrons can induce soft- or hardware errors, called single event upsets (SEU), in sensitive devices, e.g., memory chips. The susceptibility of aircraft electronics to neutron-induced SEU is a problem that has received widespread attention (see, e.g., [18-20]).

For several of the mentioned applications, a better understanding of the fundamental interactions of neutrons with various nuclides is needed. Such data can only be acquired in experiments employing a neutron beam. The techniques based on fission reactions and TFBCs have shown to be useful for neutron beam monitoring in those measurements.

Recently, intermediate-energy fission of some subactinide nuclei (Bi, Tl, Hg) has been employed in the manufacturing of high-temperature superconductive compounds, which enhances their performance to such an extent that new temperature records have been obtained (see, e.g., [21]). A widespread application of this technology may require appropriate engineering calculations, which in turn require a reliable database for the underlying nuclear reactions.
4 Studies of fast nucleon-induced fission cross sections

4.1 Experimental techniques for fission fragment detection

A variety of experimental techniques have been implemented for fission fragment detection since the discovery of fission. Since a specific experiment may focus on some specific characteristics of the fission process (e.g., cross section, LMT, distribution of fragments versus angle, energy, mass and charge, etc.), and experimental conditions may vary depending on the specific reaction studied, a single technique cannot be optimal in all cases. For the further discussion, emphasis is put on the application of different techniques in fission cross section measurements.

The radiochemical method was among the first ones to be applied in fission detection, and it was used in a fair number of studies, in combination with β- and/or γ-spectrometry of fission products. However, uncertainties associated with the isotope separation procedures are intrinsically large, and therefore the quality of such cross section determinations is always low. A more recent version of this technique has come into use with the advent of high-resolution γ-spectrometers, which have made chemical separation of the products unnecessary, and thus the related uncertainties could be eliminated. The method is applicable for measurements of cross sections for fission product formation in both (p,f) and (n,f) reactions (see, e.g., [22, 23]), although it may involve interpretation of very complicated γ-spectra. A possible solution of the problem is to separate products of fission and other reactions using a multi-layer catcher system [24]. This technique, however, must rely on model calculations of the detection efficiency of the setup. Moreover, the method requires a relatively large fluence of incident particles, which is not easy to achieve for neutrons.

When applied to integral fission cross section measurements, the discussed techniques have, as a common problem, the uncertainties associated with the integration of the mass yield curve. In addition, they are intrinsically not intended for correlation measurements, and therefore a problem may arise in isolating contributions from different reaction mechanisms to the mass curve, especially at high energies.

Nuclear photoemulsions also belong to the first generation of detection techniques. They played an important role as a means of direct, straightforward visualization of the fission process. However, problems due to detection efficiency, non-fission and intrinsic backgrounds, together with tedious track counting, lead to limited application in cross section measurements. One of the modifications of the technique included loading of the emulsion with a chemical compound of the studied fissile material, so that the emulsion itself combined the sample and the detector (see, e.g., [25]).

Solid state nuclear track detectors (SSNTDs) of different types have been extensively employed in fission studies. When used in "remote geometry", they allow high homogeneity of the tracks and, thus, high reliability in the track identification [26]. Another variant of the technique allows detection of correlated fragments using a pair of SSNTDs in sandwich geometry with a thin fissile sample, which is transparent for the fragments. By re-matching the detectors after irradiation and track etching, it is possible to observe the correlated tracks originating from the point where the fission event occurred. Such a setup is, however, sensitive to event selection criteria, which may introduce a large uncertainty in the cross section determination (see studies of Hudis and Katcoff [27, 28]). Similar to the nuclear emulsions, SSNTDs require tedious track counting, although there have been attempts to make this procedure automatic (see, e.g. [29]).

There are very few applications of ionization chambers for (p,f) reaction [30], because of the difficulties due to the background produced by the incident beam. On the other hand, ionization chambers (IC) dominate for (n,f) studies [32-44], which do not suffer from the
mentioned problem. There is still a complication in the interpretation of pulse height spectra from ICs, however, because the contributions from fission and other reactions interfere. If fission is much less probable than competing reactions, as is the case, e.g., for interactions of neutrons with sub-actinide nuclei, this interference is especially severe. An important modification of the technique is the Frisch-gridded ionization chamber, which allows to measure also the emission angle of the fragment relative to the chamber axis, and thus to reject the non-fission background. The technique has been recently applied at the TSL neutron beam by Tutin et al. [45].

The parallel plate avalanche counter (PPAC) is a rather new and prospective type of fission detectors with excellent timing characteristics. Its use in cross section measurements has been, however, very limited [7].

Semiconductor detectors have the great advantage that they allow measurements of fragment energy simultaneously with obtaining timing information. The possibility to make on-line coincidence measurements ensures a reliable selection of fission events. However, the accompanying problems (poor radiation stability and susceptibility to different kinds of background) make the technique more suitable for fundamental multi-parameter studies at a single projectile energy than for routine cross section measurements in a wide energy range. Thus, the number of cross section measurements with this technique are few (see, e.g., [7, 46]), but the results are normally very reliable.

Thin-film breakdown counters (TFBCs), devised by Tommasino et al. [47], are metal-oxide-silicon capacitors intended for detection of heavy ions and, in particular, fission fragments. The operation principle of the TFBC is based on the phenomenon of electric breakdown in a metal-oxide-silicon structure caused by an ion passing through a thin silicone dioxide layer. The breakdowns are non-shorting, since they lead to vaporization of a small part of the electrode area and leave no conducting path between the electrodes. The features of the TFBCs are: the threshold properties, i.e., the insensitivity to light charged particles, neutrons and γ-radiation, real-time operation and good timing properties, ease of operation (no high voltage required, no gases, large output signals, which makes preamplifiers unnecessary), low cost, compact design, and long-term stability under heavy radiation conditions. TFBC properties have been studied by Smirnov and co-workers [48-55], Klein [56], and Streubel and co-workers [57, 58]. At present, TFBCs have been developed to mature nuclear instruments, useful in a number of applications, such as nuclear cross section measurements, particle beam monitoring, neutron dosimetry, and studies of the performance of nuclear reactors (see ref. [54] and references therein).

In a typical application, a primary particle beam induces nuclear reactions in a sample (referred to as radiator), which acts as a source of strongly ionizing secondary particles (e.g., fission fragments), detected by one or several TFBCs. There are two possibilities for the mutual positioning of the TFBC and the radiator, namely in remote geometry or sandwich geometry. Remote geometry implies that the TFBC and the radiator are separated by a distance, which is comparable to or larger than the dimensions of the detector sensitive area. In many cases, it is possible to choose a radiator-detector distance and a bias voltage that allow detection of all fragments entering the sensitive area of the TFBC. In this case, the detection efficiency as a function of the bias voltage reaches a plateau, and the efficiency is defined solely by the available solid angle. The simplicity of the detection efficiency determination is an attractive feature of the remote geometry. On the other hand, this design is not suitable in applications involving low event rates or limitations in the available space.

In sandwich geometry, the sensitive surface of the TFBC and the surface of the radiator are situated parallel to each other at a distance that is small in comparison with the dimensions of the detector sensitive area. The fragments can travel this distance in air without significant energy loss, and thus, evacuation of the setup is not necessary, which makes the
sandwich design more practical than the remote one. A more significant advantage of the sandwich geometry is that it gives maximum possible sensitivity of the setup to the primary beam. This is the reason for using the sandwich design in applications involving low-intensity beams, e.g., intermediate-energy neutrons. One example is the TFBC-based neutron fission monitor [55] recently installed at the neutron beam facility at TSL [59, 60]. More examples can be found in a review paper [54].

All experimental studies included in this thesis and published as Papers I and V have been performed using TFBCs in sandwich geometry (see Sect. 5.1). The approaches employed for the determination of the detection efficiency in the present work are discussed in Sect. 5.2.

4.2 Status of experimental data

Several experimental studies on intermediate-energy nucleon-induced fission cross sections have been performed recently or are in progress. The cost of experimental work at accelerators is high, and beam time is scarce. It is therefore of increasing importance that existing experimental data are properly documented. The optimal tool for this activity is the format for international nuclear data exchange, EXFOR [61]. However, high-energy fission data are scarcely represented in the EXFOR databases. As a step to fill this gap, a systematic literature search and an EXFOR compilation of proton-induced fission cross section data have been performed in Paper II. The compilation concerns (p,f) cross sections for $^{165}$Ho, $^{187}$Ta, $^{183,184}$W, $^{185}$Re, $^{194,195,196}$Pt, $^{197}$Au, $^{205}$Tl, $^{204,206,207,208}$Pb, $^{209}$Bi, $^{232}$Th, $^{233,235,238}$U, $^{237}$Np, and $^{239}$Pu at energies from 0 up to 30 GeV. The choice of nuclei was motivated by the fact that tantalum, tungsten, lead and heavier elements are considered as prospective materials for ADS spallation targets, while $^{165}$Ho and $^{187}$Yb were included in order to reveal trends of the data in the region around Z=70 where only sparse results exist. The experimental data processed are plotted in Fig. 1 for the subactinide nuclei and in Fig. 2 for the actinides.

In the course of the compilation, data uncertainties were analyzed and a comparative critical analysis of the experimental techniques and the data was performed. A phenomenological expression for the cross section as a function of proton energy, which contains four independent parameters, was fitted for each of the target nuclei with sufficient energy coverage. These fits are shown as solid lines in Figs. 1 and 2. In order to be able to predict cross sections for unmeasured nuclides and elements (e.g., Hg), the parameters were in turn further expressed as a function of $Z^2/A$ for the compound system by fitting polynomials of power two to the corresponding individual values. The fitting was done separately for
Fig. 1.
The (p,f) cross sections of $^{165}$Ho, $^{181}$Ta, $^{197}$Au, $^{205}$Tl, $^{208,207,206,204}$natPb, and $^{209}$Bi nuclei versus incident proton energy. The solid and open symbols represent the rejected and accepted data, respectively. The solid lines are the individual best fits and the dashed lines are the predictions of the overall systematics obtained in the present work. For the $^{nat}$Hg(p,f) cross section, no data exist, and only the systematics prediction is shown.
subactinides and actinides, and the result is shown as dashed lines in Figs. 1 and 2, respectively. As can be seen, the overall systematics give a good representation of the data in most cases.

The compilation and the systematics may be helpful for comparisons with theoretical calculations as well as for practical cross section estimates, although a few “problem spots” have been revealed. Further measurements in the region of tantalum-tungsten should be given high priority due to its practical importance and the unresolved data discrepancies. Actinide data above 1 GeV are scarce and often discrepant, especially for $^{232}$Th, and should be supplemented by further experimental studies. In addition, data for separated isotopes (e.g., of W, Hg, Pb) may be important for further development of the theoretical description of the fission process in the intermediate energy region.

![Fig. 2.](image)

The (p,f) cross sections of $^{232}$Th, $^{238,235,233}$U, $^{237}$Np, and $^{239}$Pu nuclei versus incident proton energy. The solid and open symbols represent the rejected and accepted data, respectively. The solid lines are the individual best fits and the dashed lines are the predictions of the overall systematics obtained in the present work.

Fission induced by intermediate-energy neutrons has been studied much less extensively than with protons, and the available (n,f) cross section data sets are often discrepant. It would therefore be premature to search for systematics for a wide range of target nuclei. Moreover, standards for (n,f) cross section measurements are still under discussion and may need improvement.

Early experimental data of Kelly and Wiegand [62], Goldanskiy et al. [63], Reut et al. [64], Dzhelilov et al. [65], and Pankratov et al. [66] are of a rather qualitative character, because of various problems with the characterization of the neutron beam, the fission
samples, and the detection efficiency. More recent measurements for a few sub-actinide nuclei in the energy region 18-23 MeV were performed by Vorotnikov [67] and Vorotnikov and Larionov [69], respectively, using a d-T neutron source and solid state nuclear track detectors. In many cases, only upper limits could be established, because the cross sections are extremely small in this near-barrier region.

The most extensive work in the field was previously performed by Lisowski and co-workers at the LANL "white" spectrum neutron facility, using parallel-plate ionization chambers. The (n,f) cross section for a few actinide nuclei was measured relative to that of $^{235}$U. The latter cross section was made absolute using the benchmark datum at 14 MeV and the shape of the spectral neutron fluence versus incident energy. The results have been published in conference proceedings (see [39] and references therein). However, none of the listed references contain a detailed description of the experimental procedure. Moreover, earlier results from the same group [32, 35] differ significantly (by tens of percent) from those of ref. [39], but no explanation to these changes has been given.

Further measurements with a similar experimental arrangement were performed by Vonach et al. [43] and Staples et al. [40-42] in the early 90ies. From these studies, only the (n,f) cross section data for Pu isotopes [42] have the status of being final, while the measurements for sub-actinides [40, 41, 43] have not yet resulted in a final publication.

Similar techniques have been employed by Donets et al. [69] at the neutron facility in Gatchina. At present, only preliminary data are available.

The $^{238}$U(n,f) cross section was measured by Newhauser and co-workers [44]. This group continues experimental activities in the field, and new data are expected, while the older ones are under revision [70].

In the early 90ies, (n,f) cross section measurements started at TSL in Uppsala within a collaboration between V.G. Khlopin Radium Institute and Uppsala University. Measurements have been performed using two different techniques for fission fragment detection, namely, a Frisch-gridded ionization chamber [71, 72] and thin-film breakdown counters (TFBC) [73-76]. In a few cases, the (n,f) cross sections were measured relative to that for n-p scattering. Some of the results obtained with TFBCs can be found in Papers I and V of this thesis, others are available elsewhere [73, 75, 76].

In 1997, IAEA/NDS adopted the $^{235}$U, $^{238}$U, and $^{209}$Bi(n,f) cross sections as standards in the intermediate-energy range [77]. The recommended data for the former two cross sections were represented by parameterizations based on the LANL experimental data [39], and the adopted $^{209}$Bi(n,f) cross section standard was based on the data of Vorotnikov and Larionov [68] and Eismont et al. from Paper I and ref. [71, 73] available at that time.

Coming publications with new experimental results, together with the mentioned lack of clarity concerning the LANL data, may necessitate changes in the standards in the near future. In particular, a new analysis of the TSL data for the $^{209}$Bi(n,f) reaction is presented in ref. [75].

### 4.3 Theoretical modeling

Proton- and neutron-induced fission cross sections can be calculated in the framework of the cascade-exciton model (CEM), developed at the Joint Institute for Nuclear Research in Dubna, Russia [78]. The model assumes that a nuclear reaction occurs in three stages: the cascade, pre-equilibrium and compound nucleus stage. In the past, a large variety of experimental data for reactions induced by nucleons, pions, and photons have been analyzed in the framework of the CEM, and the general validity of this approach has been confirmed. The CEM was found to have one of the best predictive powers for double differential cross sections of nucleon emission in comparison with other available models [79]. Recently, the
CEM has been extended by taking into account competition between particle emission and fission at the compound nucleus stage [80] and an improved description of nuclear level densities [81].

In Paper III, the CEM, as realized in the CEM95 code, was applied to the calculations of cross sections and momentum transfer in proton- and neutron-induced fission reactions with $^{209}\text{Bi}$ and $^{208}\text{Pb}$ nuclei. Fig. 3 shows the calculated average LMT for the studied reactions. For comparison, experimental data [46, 82-84], which are scarcely available, especially for the (n,f) reactions, are shown. Moreover, a large spread exists between the data from different studies and even within the same data set [83]. From the comparison, one can conclude that the description of the LMT does not contradict the available experimental data.

The calculations of fission cross sections (Fig. 4) performed with the original version of the CEM95 code reproduce the general trends of the experimental data (represented by parameterizations; shown as solid lines in Fig. 4). However, the model predicts too steep an increase of the excitation functions above 100 MeV. As shown in Paper III, this systematic discrepancy cannot be eliminated by any reasonable variation of the input parameters of the model. On the other hand, such a variation has no significant influence on the calculated LMT.

In order to improve the description of the fission cross section data, a modification of the model was undertaken as follows. The CEM95 code makes use of the expression suggested by Ignatyuk et al. [85] for the level density parameter:

\[
a(E^*) = \tilde{a} \left(1 + \delta W_{gs} \frac{1 - \exp(-\gamma E^*)}{E^*}\right),
\]

where $\tilde{a} = \alpha A + \beta A^{2/3} B_s$ is the asymptotic value of the level density parameter at high excitation energies, with $A$ being the atomic mass of the nucleus and $B_s$ the ratio of the surface area of the nucleus to the surface area of a sphere of the same volume. For nuclei with equilibrium deformation, $B_s = 1$, while at the fission saddle point $B_s > 1$. Further, $\delta W_{gs}$ is the shell correction and $E^* = E - \Delta$, where $E^*$ is the excitation energy and $\Delta$ is the pairing correction. Finally, $\alpha$, $\beta$, and $\gamma$ are phenomenological constants. Eq. (1) reflects the correlation between the level density and the shell correction for low excitation energies, and the fade-out of the shell effects at high excitations [85].

In the original version of the CEM95 code, Eq. (1) is applied to all decay channels of the excited nucleus, except for the fission channel. For the latter, the level density parameter at the saddle point, $a_s$, is calculated using the corresponding parameter for the neutron emission channel, $a_n$, and a constant ratio for $a_f/a_n$, which serves as a fitting parameter of the model. Thus, the influence of the shell effects on the level density in the neutron emission channel is automatically conveyed to the level density at the saddle point. Contrary to this, the shell correction at the saddle point for nuclei in the studied mass range is not expected to have any relation to that for the ground state, but should be much smaller. This is due to the large saddle-point deformation and, therefore, the reduced symmetry of the saddle-point shape compared to the nearly spherical ground-state configuration close to the doubly magic nucleus $^{208}\text{Pb}$.

To estimate the importance of this effect, we have performed cross section calculations with the parameter $a_f$ being energy-independent, which is equivalent to the removal of the shell-effect influence on the level density at the saddle point. The parameter $B_s$ was adjusted to the experimental data.

The results of the calculations with the modified model are shown in Fig. 4 as dashed lines. Much better agreement with experiment has been achieved in comparison with the
original version. The modifications indicate the crucial importance of incorporating an appropriate level density description in the model, and motivate the search for a consistent model of barriers, ground-state masses, and level densities, which may further improve the predictive power of the model.

![Graph 1](image1.png)

**Fig. 3.**
The average fraction of the longitudinal LMT as a function of the projectile energy for the following reactions: $^{209}$Bi(p,f) (a), $^{209}$Bi(n,f) (b), $^{208}$Pb(p,f) (c), and $^{208}$Pb(n,f) (d). The solid symbols show experimental data from Refs. [46, 82-84]. The open squares connected with the dashed lines show the calculations of this work.

![Graph 2](image2.png)

**Fig. 4.**
The cross sections of the following reactions: $^{209}$Bi(p,f) (a), $^{209}$Bi(n,f) (b), $^{208}$Pb(p,f) (c), and $^{208}$Pb(n,f) (d). The solid lines show the parameterizations of the experimental data. The dotted and dashed lines represent the calculations with the original and modified versions of the CEM95 code, respectively.
5 Measurements of neutron-induced fission cross sections at TSL

The experimental part of this work, described in Papers I and V, was performed at the neutron beam facility of TSL in Uppsala. Paper I presents a study of neutron fission cross sections of $^{238}$U, $^{209}$Bi, and $^{208}$Pb in the energy range 73-160 MeV. Paper V is devoted to (n,f) cross section measurements for $^{nat}$Pb, $^{208}$Pb, $^{197}$Au, $^{nat}$W, and $^{181}$Ta in the range $E_n = 30$-140 MeV. Both studies employed thin-film breakdown counters for the fission fragment detection. In Paper I, the (n,f) cross sections were measured relative to that for $n-p$ scattering, and thus information was obtained that allowed to establish secondary cross section standards. In Paper V, the measurements were performed relative to the $^{209}$Bi(n,f) cross section, which was adopted as a standard, mainly based on the experimental data from Paper I.

5.1 Experimental apparatus and procedure

5.1.1 Neutron facility and irradiation positions

The $^7$Li(p,n) reaction is used for the neutron production at TSL [59, 60]. The proton beam from the Gustaf Werner cyclotron is directed onto a thin disc of lithium, highly enriched in $^7$Li (see Fig. 5). Downstream the target, the proton beam is deflected by two magnets into an 8 m long tunnel, where it is focussed onto a beam dump. The neutrons produced within a 60-$\mu$sr cone around 0° pass a collimating system before reaching the experimental hall at a distance of about 8 m from the production target.

The incident neutron spectrum is not monoenergetic, but includes a high-energy peak accompanied by a low-energy tail, which also contributes to the fission reaction rate. To determine the (n,f) cross section at the energy of the peak, one needs to know the fraction of fission events due to that peak. Two different approaches to determine this fraction have been employed in Papers I and V, respectively.

The first approach is to use the time-of-flight (TOF) spectrum of the fission events relative to the cyclotron RF. In order to separate the peak from the tail in the TOF spectrum, a sufficiently long neutron flight path is required (Irradiation position 2 in Fig. 5). This, in turn, limits the available neutron flux density, which is inherently low for secondary beams. Consequently, the count rate of fission events is sufficient only for measurements of large cross sections (tens of millibarns or more). This is the case for neutron-induced fission of $^{238}$U and $^{209}$Bi, studied in Paper I. Fission of $^{208}$Pb was under study in that early experiment as well, but insufficient statistics and significant background contributions, which increase with decreasing peak neutron energy, necessitated a search for an alternative approach.

Since the neutron flux density is inversely proportional to the distance from the production target squared, this distance has to be as short as possible. At the same time, the neutron field in the irradiation position has to be free from contamination by primary or scattered protons. In addition, locations close to the 0° direction are preferable, because the production of high-energy neutrons is strongly forward-peaked. The given conditions could be satisfied by placing the experimental setup between the proton bending magnet and the first neutron collimator, at a distance of about 2 m from the production target (see Irradiation position 1 in Fig. 5). To facilitate simultaneous experiments in the neutron beam line, the setup was placed outside the vacuum tube at an angle of about 4° to the beam axis. Because of the short flight path, TOF separation of the peak fissions from the tail ones was not possible any more. The fraction of peak fission events was therefore obtained in an iterative unfolding procedure, taking into account relative fission reaction rates at as many incident neutron
Fig. 5.
An overview of the neutron beam facility. The positions of the experimental setup in the present experiments are indicated as Irradiation position 1 and 2.
energies as possible, together with corresponding information on the shape of the neutron spectra. This technique was implemented in Paper V.

5.1.2 Neutron spectrum

Since an unfolding procedure was needed to obtain fission cross sections from the reaction rates measured in Paper V, precise information on the neutron spectrum at the various incident neutron energies was required. A complete set of characterization measurements is not available for the TSL neutron beam at present. We therefore rely on the following assumptions about the neutron spectrum:

- The spectrum is a sum of two components. One of them originates directly from the $^7\text{Li}$ target. The other is a background arising from interactions of primary protons with the beam transport system, the beam dump, the walls, and other material in the surroundings, with subsequent propagation and slowing-down of secondary neutrons.
- In Irradiation Position 2, the background component does not have any significant influence on the (n,f) cross section measurements for the studied nuclides. On the other hand, the background in Irradiation Position 1 dominates in the low-energy end of the spectrum and vanishes at neutron energies of about 10 MeV [86]. This component could hamper cross section measurements for reactions with a threshold at lower energy, e.g., $^{238}\text{U}(n,f)$. It is not important, however, for fission reactions in sub-actinide nuclei, which possess thresholds of about 20 MeV or more (see, e.g., [5]).
- The energy and angular distribution of neutrons in the first component is defined only by the double-differential cross section (DDX) of the $^7\text{Li}(p,n)$ reaction.

Support for the last assumption is given by the fact that the neutron spectra measured at TSL agree with data from other sources. Spectra reconstructed from earlier n-p scattering studies at TSL [59, 87], as well as from other facilities [88-90] are shown in Fig. 6. The peak neutron energy was 94 MeV (a), 133 MeV (b), and 160 MeV (c). All spectra are normalized so that the area under the high-energy peak is unity.

As seen in the figure, the neutron spectrum consists of a high-energy peak and a low-energy tail. The high-energy peak corresponds to the $^7\text{Li}(p,n)$ reactions that leave the $^7\text{Be}$ nucleus in the ground state or in the first excited state at 0.43 MeV. The low-energy tail is related to the excitation of higher states in $^7\text{Be}$ and to break-up reactions.

An evaluated nuclear data library for the $^7\text{Li}(p,n)$ reaction has been developed recently by Mashnik et al. [91]. It provides an accurate representation of the angle-integrated high-energy peak cross section. However, the angular distribution of the peak neutrons at forward angles is not reproduced well, especially at higher energies where the evaluated and experimental data disagree by up to a factor of two. Therefore, we have adopted another technique to calculate the differential cross section of the $^7\text{Li}(p,n)^7\text{Be}$ (g.s.+0.43 MeV) reaction at 0° in the laboratory frame:

$$
\frac{d\sigma_{\text{peak}}}{d\Omega}(0^\circ) = \begin{cases} 
\sigma_{\text{peak}} R(E_p), & \text{if } E_p < 60 \text{ MeV}, \\
35.5 \text{ mb/sr}, & \text{if } E_p > 60 \text{ MeV}, 
\end{cases}
$$
Neutron spectra from the $^7$Li(p,n) reaction at 0º. The peak energy is 94 MeV (a), 133 MeV (b), and 160 MeV (c). The filled circles represent measurements at the TSL neutron facility [59, 87]. The open symbols are data from other facilities [88-90]. The dashed line in panel (a) represents the neutron spectrum calculation based on the evaluation [91] (see text). The thick solid lines in panels (a) and (b) show the calculated spectra folded with the time resolution of the measurements at TSL. The thin solid line in panel (c) is drawn through the experimental data to guide the eye. All spectra are normalized so that the area under the high energy peak is unity.
where $\sigma_{\text{peak}}$ is the angle-integrated $^7\text{Li}(p, n)^7\text{Be} (\text{g.s.} + 0.43 \text{ MeV})$ cross section from the evaluation [91], and $R(E_p)$ is the "index of forwardness" introduced by Uwamino et al. and fitted to experimental data as a polynomial function of the incident proton energy $E_p$ [92]. The saturation value of the cross section at higher energies comes from the analysis of experimental data in the proton energy range 60-400 MeV performed by Watson et al. [93].

Existing experimental data on the continuum neutron production in the $^7\text{Li}(p, n)$ reaction are too scarce to be used directly for the unfolding procedure in the study of Paper V. Therefore, the evaluation of Mashnik et al. [91] was employed to deduce double-differential cross sections (DDX) of continuum neutron production in the laboratory frame. Figs. 6a and b show a comparison of the calculated DDX at 0° (solid lines) with the neutron spectra measured at TSL [59, 87] and at other facilities [88-90]. The evaluation satisfactorily reproduces the high-energy part of the experimental neutron spectra, which is of primary importance for the study of Paper V. On the other hand, the evaluation overestimates the production of low-energy neutrons (below about 20-30 MeV), although few experimental data sets are available in this energy region. However, the uncertainty in the neutron spectrum in the low energy region has little influence in the present work, again due to the high thresholds of the studied fission reactions.

As has been mentioned, the experimental setup was placed at an angle of 4° with respect to the primary proton beam direction. Because of the strong forward-peaking of the high-energy neutron production, the difference between the neutron spectra at 0° and 4° had to be taken into account in the unfolding calculations.

5.1.3 Fission fragment detectors and experimental chambers

The fission fragments were detected by thin-film breakdown counters (TFBC, see Sect. 4.1). Excellent stability of TFBCs under heavy radiation conditions was of primary importance for the experiment of Paper V, because of the severe $\gamma$-radiation background in irradiation position 1.

Fig. 7.
The design of an experimental chamber.

The low beam intensity necessitated the use of sandwich geometry, i.e., where the detector is situated as close as possible to the fission sample. Since the short distance between
the sample and the detector sensitive area could be passed by fission fragments in air without any significant energy loss, evacuation of the chamber is not necessary. Relative fission cross sections can be measured using detectors sandwiched with samples of different nuclides and being irradiated by the same neutron beam.

The design of an experimental chamber of the type employed in Paper V is shown in Fig. 7. It consists of a mosaic arrangement of detectors, a similar arrangement of samples, and a thin mechanical housing. The bias voltage and the signal output were common for all detectors in the mosaic. The whole experimental setup consisted of a few such chambers, stacked along the neutron beam direction, so that each set of sample-detector sandwiches was exposed to virtually the same neutron fluence. Each chamber was equipped either with samples of one of the studied nuclides (natPb, $^{208}$Pb, $^{197}$Au, natW, and $^{181}$Ta) or with the monitor samples ($^{209}$Bi).

In Paper I, detector-sample arrangements of two different types were employed. First, mosaic arrangements with natU, $^{209}$Bi, and $^{208}$Pb samples were used, which were similar to the ones shown in Fig. 7, except of a larger number of sandwiches per mosaic. Second, single sandwiches with $^{238}$U and $^{209}$Bi samples of a larger area were used. TOF techniques were feasible only with the arrangements of the first type, while those of the second type could only give the count rate of fission events induced by the whole neutron spectrum, because of the much larger spread in signal propagation time.

5.1.4 Fission samples

The samples were prepared by vacuum evaporation ($^{238}$UF$_4$, $^{209}$Bi, natPb, $^{208}$Pb, $^{197}$Au, and natWO$_3$), magnetron evaporation ($^{181}$Ta), and multi-layer smearing (natU$_3$O$_8$) onto stainless steel or aluminum backings. In all cases, the area of the sample exceeded the sensitive area of the respective TFBC, and therefore the latter defined the effective area of the sandwich.

The actinide contamination of the sub-actinide samples was checked by $\alpha$-activity measurements using semiconductor or nuclear track detectors. In addition, irradiations with 14-MeV or 21-MeV neutron beams were performed for different samples sandwiched with TFBCs. Because of the very small fission cross sections of the sub-actinide nuclei at this energy, virtually all detected fission events could be attributed to actinide contaminants.

5.1.5 Electronics and data acquisition system

The signals from the mosaic TFBC arrangements were fed into a fast multichannel leading-edge discriminator, logically summed and fed into the start input of a time-to-digital converter (TDC). A pulse, phase-locked to the cyclotron RF, served as the stop signal for the TDC. In addition, the discriminated signal from each fission chamber was recorded in a scaler, and this information was used in the analysis to separate the TOF spectra of fission events from the different chambers. The TOF spectra and the counting-rate data were stored in a computer on an event-by-event basis and could be inspected on-line.

As discussed above, it was not possible in the experiment of Paper V to fully separate the high-energy peak fissions from those due to the low-energy tail using TOF techniques. Nevertheless, TOF techniques were useful for rejection of intrinsic detector background events, as well as those from spontaneous fission of contaminating nuclides. In addition, inspection of the low-energy part in the TOF spectra allowed us to check that no significant actinide contamination was present in the fission samples.

The signals from the large-area TFBCs employed in Paper I passed a shaping amplifier and a discriminator, and were recorded by a scaler. No TOF information was provided in this case.
5.1.6 Neutron flux measurement

Part of the irradiations described in Paper I, namely, those with peak neutron energy of 135 and 162 MeV, were performed simultaneously with experiments on $n$-$p$ scattering [87], and thus it was possible to obtain the neutron flux relative to the $n$-$p$ scattering cross section by the use of a proton-recoil spectrometer [59, 87]. The $n$-$p$ scattering data were normalized to the experimental total cross section, with a correction for undetected angular regions using predictions of phase-shift analyses solutions (see, e.g., [94]). In this way, absolute neutron-induced fission cross sections could be obtained for $^{238}$U, $^{209}$Bi, and $^{208}$Pb.

5.2 Data analysis

5.2.1 Fission cross section

In Paper I, the absolute fission cross section was determined as follows:

$$\sigma_f = \frac{N_f k_{\text{low}} k_{\text{anis}} k_{\text{LMT}}}{\varepsilon S_{\text{TFBC}} j_n},$$

where $N_f$ is the number of detected fission fragments, $k_{\text{low}}$ is the fraction of detected fissions due to the high-energy peak, $k_{\text{anis}}$ and $k_{\text{LMT}}$ are corrections for anisotropy and LMT, respectively. Furthermore, $\varepsilon$ and $S_{\text{TFBC}}$ are the efficiency and the sensitive area of the TFBC, respectively, $\rho$ is the number of nuclei in the sample per unit area, and $j_n$ is the neutron flux density, determined with the $n$-$p$ spectrometer and scaled to the position of the fissile sample. The product $\varepsilon S_{\text{TFBC}}$ was determined in a calibration measurement with a $^{252}$Cf sample:

$$\varepsilon S_{\text{TFBC}} = n_{sf} / a_{sf},$$

where $n_{sf}$ is the fragment count rate in the calibration measurement and $a_{sf}$ is the spontaneous fission activity of the calibration sample per unit area.

In Paper V, the following expression was used for determination of the fission cross section ratio (at peak energy) with a pair of sandwich arrangements $X$ and $Y$, containing samples of different materials:

$$\frac{\sigma_{f0(X)}}{\sigma_{f0(Y)}} = \frac{N_{f0(X)} k_{\text{low}(X)} R_{(X)}^2 \rho_{(X)} k_{e0(Y)} n_{sf(Y)}}{N_{f0(Y)} k_{\text{low}(Y)} R_{(Y)}^2 \rho_{(Y)} k_{e0(X)} n_{sf(X)}},$$

where $R$ is the distance between the production target and the experimental chamber, and $k_{e0}$ is the relative detection efficiency for peak fissions (including the effects of LMT and anisotropy). The quantities $R$, $\rho$, $n_{sf}$, and $N_f$ were obtained in direct measurements for the respective chambers. The latter quantity was corrected for intrinsic detector background on the basis of the obtained TOF spectra of fission events. The determination of the remaining parameters in Eq. (4), $k_{e0}$ and $k_{\text{low}}$, is discussed in Sect. 5.2.2 and 5.2.3, respectively.

5.2.2 Detection efficiency determination

Different techniques were employed for the detection efficiency determination in the measurements of Papers I and V. In the first case, the method of imitators was employed. This means that the absolute detection efficiency for a specific fissile sample is determined in a
measurement of the count rate of spontaneous fission fragments provided by a substitute sample (imitator) containing $^{252}$Cf. The imitator was designed in order to simulate the linear energy loss (LET) in the detector sensitive layer for fission fragments from the studied sample. However, it was difficult to reproduce the LET distribution sufficiently well, and thus the method was only approximate.

An alternative approach has been implemented in the experiment of Paper V. The energy-dependent relative detection efficiency was calculated for the actual fissile samples using a model developed in Paper IV. The model is based on a simple and general formulation of the assumed physics of TFBC particle detection, and the used sample-detector geometry. The derivation yielded the following expression for the detection efficiency:

$$
\varepsilon = \frac{M \bar{k}_a}{2} \tilde{\varepsilon}(T),
$$

where $M$ is the mean multiplicity of the fragments ($M = 2$ for binary fission) and $\varepsilon$ is the reduced detection efficiency, which depends on the linear energy transfer (LET) of the fragments in the material of the detector sensitive layer (SiO$_2$) and on the properties of a specific TFBC at a given bias voltage. The latter are characterized by two free parameters, $L_{th}$ and $L_e$. The threshold LET, $L_{th}$, is the minimum LET that a particle with perpendicular incidence must have in order to cause a breakdown. The excess LET, $L_e$, represents the additional LET a particle must have to be detected at any incident angle. The efficiency for detection of such particles in a sandwich geometry would approach 100%, which is never the case in reality. The parameters $L_{th}$ and $L_e$ were obtained from experimental data for the actual type of TFBC and bias voltage.

The correction factor $k_a$ accounts for the anisotropy of the fragment angular distribution in the laboratory frame, and is thus unity for $^{252}$Cf spontaneous fission. The determination of $k_a$ requires information on the linear momentum transfer (LMT) from the projectile to the fissioning nucleus, as well as the anisotropy of the fragment angular distribution in the frame of the fissioning nucleus. Corresponding experimental data from the literature were therefore compiled in order to reveal the systematic behavior of the relevant quantities.

5.2.3 Correction for low-energy neutrons

The TOF techniques employed in Paper I allowed determination of the correction for the low-energy neutrons, $k_{low}$, by decomposition of the time spectrum of fission events. In order to separate the peak from the tail, the contribution of the latter in the peak region was approximated by a linear function. This simple algorithm was verified at a single energy of 162 MeV by folding the neutron spectrum measured by Stamer et al. [89] with the $^{238}$U(n,f) cross section obtained by Lisowski et al. [39], and by subsequent folding with the energy resolution in the experiment of Paper I. A reasonable agreement was obtained between the calculated and experimentally observed TOF spectra, as well as between the $k_{low}$ values obtained with the two procedures.

In the cross section ratio measurements of Paper V, an iterative unfolding procedure was used for the determination of $k_{low}$. A similar procedure was implemented in an analysis of neutron-induced single-event upsets performed by Johansson et al. [95]. However, the present study makes use of a more sophisticated description of the incident neutron spectrum (see Sect. 5.1.2). The correction for the $^{209}$Bi(n,f) reaction was calculated using the standard
cross section [77] as a function of energy. To get a first estimate of \( k_{\text{low}} \) for the other studied reactions, the measured count rate was employed as a trial input. From the trial cross section, the neutron spectrum, and the relative detection efficiency, \( k_{\text{low}} \) was calculated for each beam energy employed in the study. The respective fission cross sections were then calculated using Eq. (4) and the standard \(^{209}\text{Bi}(n,f)\) cross section. The folding was applied again, and the next approximation of \( k_{\text{low}} \) was calculated using a smooth curve fitted to the obtained cross section values. The procedure was repeated until convergence was reached. Typically, a few iterations were needed. In all cases, the result was found to be independent of the initially assumed cross section.

5.3 Experimental results

The relative cross sections are presented in Table 1 and are shown in Fig. 8 together with previously reported data. In a few cases, only upper limits for cross section ratios were determined, using the prescriptions of Schmidt et al. [96] for analysis of data with poor counting statistics. The uncertainties of the relative data stem from the sample thickness determinations, the counting statistics in the Cf calibration and beam measurements, the calculation of corrections to the detection efficiency, and the determination of the fraction of peak fission events.

The absolute cross sections are given in Table 2 and are shown in Fig. 9 together with data from previously reported studies. Three different methods have been employed to obtain the absolute data:

1. The experiments at 135 and 162 MeV presented in Paper I yielded absolute fission cross sections based on the neutron flux determined using the \( n-p \) spectrometer. The additional components of the uncertainty in the absolute data are due to the determination of the spontaneous fission activity of the calibration sample per unit area and the determination of the neutron flux density with the \( n-p \) spectrometer. The latter component included the uncertainty in the standard \( n-p \) cross section.

2. Cross sections at the other energies in Paper I, namely at 73 and 96 MeV, were made absolute using the \(^{238}\text{U}(n,f)\) cross sections of Lisowski et al. [39]. Later, Carlson et al. [77] provided a parameterization of the data of Lisowski et al., which was adopted as a standard. The parameterization differs from the values used in Paper I by less than 1.5%. Since this error is much smaller than the overall uncertainty, no re-normalization has been made. In Paper I, the uncertainties of the absolute cross sections obtained in this way did not include those of the standard, since the latter had not been documented at that time. In the later work of Carlson et al. [77], the uncertainty in the \(^{238}\text{U}(n,f)\) standard is estimated to be 3-4% in this energy region. This would not significantly contribute to the overall uncertainty (10-13%) of the present data, and was therefore not included in the given uncertainties.

3. In the study of Paper V, the cross sections were made absolute using the standard \(^{209}\text{Bi}(n,f)\) cross section [77]. According to ref. [77], the uncertainty of the standard amounts to 50% at 35 MeV and varies from 13% at 46 MeV to 11% at 133 MeV. This uncertainty is not included in the total uncertainties of the data given in Table 2 and shown in Fig. 9.
Table 1.
Relative neutron-induced fission cross sections.

<table>
<thead>
<tr>
<th>$E_n$ (MeV)</th>
<th>$^{209}$Bi/$^{238}$U</th>
<th>$^{208}$Pb/$^{238}$U</th>
<th>natPb/$^{209}$Bi</th>
<th>$^{208}$Pb/$^{209}$Bi</th>
<th>$^{197}$Au/$^{209}$Bi</th>
<th>natW/$^{209}$Bi</th>
<th>$^{181}$Ta/$^{209}$Bi</th>
</tr>
</thead>
<tbody>
<tr>
<td>34.6</td>
<td>-</td>
<td>-</td>
<td>0.17±0.09</td>
<td>&lt;0.079</td>
<td>&lt;0.021</td>
<td>&lt;0.039</td>
<td>&lt;0.041</td>
</tr>
<tr>
<td>46.1</td>
<td>-</td>
<td>-</td>
<td>0.189±0.028</td>
<td>0.089±0.019</td>
<td>0.053±0.015</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>66.4</td>
<td>-</td>
<td>-</td>
<td>0.282±0.018</td>
<td>0.185±0.015</td>
<td>0.105±0.010</td>
<td>&lt;2.3·10⁻³</td>
<td>-</td>
</tr>
<tr>
<td>73</td>
<td>0.0097±0.0009</td>
<td>0.0026±0.0004</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>73.8</td>
<td>-</td>
<td>-</td>
<td>0.319±0.016</td>
<td>0.213±0.013</td>
<td>0.118±0.009</td>
<td>(4.2±0.8)·10⁻³</td>
<td>(1.4±0.4)·10⁻³</td>
</tr>
<tr>
<td>94.0</td>
<td>-</td>
<td>-</td>
<td>0.402±0.020</td>
<td>0.303±0.018</td>
<td>0.151±0.011</td>
<td>(8.0±1.0)·10⁻³</td>
<td>(3.8±0.5)·10⁻³</td>
</tr>
<tr>
<td>96</td>
<td>0.018±0.002</td>
<td>0.0053±0.0006</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>133.0</td>
<td>-</td>
<td>-</td>
<td>0.431±0.021</td>
<td>0.300±0.019</td>
<td>0.202±0.015</td>
<td>(16.3±2.2)·10⁻³</td>
<td>(11.5±2.0)·10⁻³</td>
</tr>
<tr>
<td>135</td>
<td>0.028±0.003</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>162</td>
<td>0.040±0.004</td>
<td>0.016±0.002</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

Table 2.
Absolute neutron-induced fission cross sections.

<table>
<thead>
<tr>
<th>$E_n$ (MeV)</th>
<th>$^{238}$U</th>
<th>$^{209}$Bi</th>
<th>natPb</th>
<th>$^{208}$Pb</th>
<th>$^{197}$Au</th>
<th>natW</th>
<th>$^{181}$Ta</th>
</tr>
</thead>
<tbody>
<tr>
<td>34.6</td>
<td>-</td>
<td>0.027±0.014</td>
<td>&lt;0.013</td>
<td>&lt;3.3·10⁻³</td>
<td>&lt;6.3·10⁻³</td>
<td>&lt;6.5·10⁻³</td>
<td></td>
</tr>
<tr>
<td>46.1</td>
<td>-</td>
<td>0.37±0.06</td>
<td>0.17±0.04</td>
<td>0.102±0.029</td>
<td>-</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>66.4</td>
<td>-</td>
<td>3.45±0.22</td>
<td>2.26±0.18</td>
<td>1.28±0.12</td>
<td>&lt;0.032</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>73</td>
<td>-</td>
<td>15±1.5</td>
<td>3.9±0.5</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>73.8</td>
<td>-</td>
<td>5.08±0.25</td>
<td>3.39±0.20</td>
<td>1.88±0.14</td>
<td>0.067±0.013</td>
<td>0.022±0.007</td>
<td></td>
</tr>
<tr>
<td>94.0</td>
<td>-</td>
<td>10.3±0.5</td>
<td>7.8±0.5</td>
<td>3.9±0.3</td>
<td>0.205±0.025</td>
<td>0.098±0.012</td>
<td></td>
</tr>
<tr>
<td>96</td>
<td>-</td>
<td>25±2</td>
<td>-</td>
<td>7.5±0.9</td>
<td>-</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>133.0</td>
<td>-</td>
<td>17.7±0.9</td>
<td>12.3±0.8</td>
<td>8.3±0.6</td>
<td>0.67±0.09</td>
<td>0.47±0.08</td>
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</tr>
<tr>
<td>135</td>
<td>1440±160</td>
<td>40±5</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>162</td>
<td>1310±120</td>
<td>53±5</td>
<td>-</td>
<td>21±3</td>
<td>-</td>
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<td></td>
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</tbody>
</table>
Neutron-induced fission cross section ratios $^{209}$Bi/$^{238}$U (a), $^{208}$Pb/$^{238}$U (b), natPb/$^{209}$Bi (c), $^{208}$Pb/$^{209}$Bi (d), $^{197}$Au/$^{209}$Bi (e), natW/$^{209}$Bi (f), and $^{181}$Ta/$^{209}$Bi (g) versus incident energy. The results of Papers I and V are shown as filled squares. Data from other studies are shown as open squares [73], triangles [72, 98], and circles [41].

Fig. 8.
Fig. 9.
Absolute neutron-induced fission cross sections of $^{238}$U (a), $^{209}$Bi (b), $^{nat}$Pb (c), $^{208}$Pb (d), $^{197}$Au (e), $^{nat}$W (f), and $^{181}$Ta (g). The results of the present study are shown as filled squares. Our earlier data for $^{209}$Bi and $^{208}$Pb from Paper I and ref. [73] are shown as crosses. Open squares, circles, up triangles, down triangles, and stars represent data of Lisowski et al. [x], Newhauser et al. [x], Staples et al. [41], Vorotnikov [67, 68], and Eismont et al. [x], respectively. Results of Reut et al. [64] and Dzhelepov et al. [65] are shown as diamonds, with horizontal bars that represent the energy spread of the neutron beam.
5.4 Discussion

Experimental cross sections for the $^{238}\text{U}$, $^{209}\text{Bi}$, and $^{208}\text{Pb}(n,f)$ reactions are presented in Paper I. The $^{238}\text{U}(n,f)$ data (see Fig. 9a) agree well with the results of Lisowski et al. [39]. The $^{209}\text{Bi}(n,f)$ cross sections (see Fig. 9b) are compatible with results of our group published elsewhere [73], as well as with data of Vorotnikov and Larionov [68]. In addition, they agree with preliminary results of Staples et al. [41] and with earlier data of Eismont et al. [71] obtained with a Frisch-gridded ionization chamber (FGIC). The situation, however, has become less clear after the new FGIC measurements of Eismont et al. [72, 97]. While the FGIC data at $E_n < 90$ MeV still agree with the TFBC ones, the two data sets are discrepant by 20-25% at higher energies. Further efforts are needed to disentangle this problem.

The $^{208}\text{Pb}(n,f)$ cross section (see Fig. 9d) has been studied both in Papers I and V, employing different techniques, and the data agree in the overlapping energy region. The difference between the result in Paper V at $E_n \approx 46$ MeV and the earlier one, published by our group elsewhere [73], amounts to about two standard deviations of the earlier datum. The results in Paper V are obtained with better counting statistics, less background, and more sophisticated data processing techniques.

Paper V has yielded cross sections for the $^{nat}\text{Pb}$, $^{197}\text{Au}$, $^{nat}\text{W}$, and $^{181}\text{Ta}(n,f)$ reactions as well, for which virtually no published results are available in the literature. Earlier measurements for $^{197}\text{Au}$ and $^{nat}\text{Pb}$ in the energy region 18-23 MeV were performed by Vorotnikov [67] and Vorotnikov and Larionov [68], respectively, using a d-T neutron source and solid state nuclear track detectors. The upper limits for the $^{nat}\text{Pb}(n,f)$ cross section established in ref. [68] are compatible with the present data (see Fig. 9c). This is not the case, however, for the $^{197}\text{Au}(n,f)$ cross section. The upper limit established in the present experiment is smaller by more than a factor of 5 than the value expected from a smooth interpolation between the data from ref. [67] and those at $E_n > 45$ MeV (see Fig. 9e).

A measurement on $^{nat}\text{Pb}$ and $^{197}\text{Au}$ was performed by Staples et al. [41] using a parallel-plate ionization chamber irradiated by neutrons from the LANSCE facility with a "white" spectrum. Only preliminary data are available (see Fig. 8c,e and Fig. 9c,e), and they are in reasonable agreement with the present ones, with the exception of the $^{nat}\text{Pb}(n,f)$ cross section at energies below 50 MeV. In the latter case, the data of Staples et al. are distinctly larger. The disagreement increases with decreasing incident energy and amounts to about one order of magnitude at $E_n=55$ MeV (see Fig. 9c). Furthermore, the $^{nat}\text{Pb}/^{209}\text{Bi}$ ratios deduced from the data of Staples et al. (see Fig. 8c) show an unexpected energy dependence: as the neutron energy decreases to below 50 MeV, the smooth decrease of the ratio turns into a sharp rise, which is difficult to understand, having in mind that the fission barrier for lead isotopes is higher than that for bismuth [5]. This leads to the suggestion that some background contribution may not have been fully taken into account in the LANSCE measurements.

Early measurements of Reut et al. [64] for $^{nat}\text{Pb}$ and $^{197}\text{Au}$ (see Fig. 9c,e) and Dzhelepov et al. [65] for $^{nat}\text{W}$ (see Fig. 9f) were made using neutrons from the Cu(d,n) reaction with a broad spectrum, as indicated by the horizontal bars in Fig. 9. These results agree qualitatively with the more recent and precise data.

The data presented in Fig. 9 allow one to conclude on some common features of sub-actinide neutron fission cross sections. The cross section increases with neutron energy and with the atomic number of the target nucleus. (Because of these two trends, the cross sections for the lightest studied nuclei, $^{nat}\text{W}$ and $^{181}\text{Ta}$, are so small at energies below about 70 MeV, that only upper limits could be determined in the present experiment). The slope of the cross section versus energy is steepest in the near-barrier region (20-25 MeV), and becomes flatter with increasing energy. The slope at a specific incident energy is steeper for lighter nuclei. The properties summarized above are similar to those of the (p,f) data (see, e.g., [98]).
6 Outline of the papers

Paper I

RELATIVE AND ABSOLUTE NEUTRON INDUCED FISSION CROSS SECTIONS OF $^{208}\text{Pb}$, $^{209}\text{Bi}$ AND $^{238}\text{U}$ IN THE INTERMEDIATE ENERGY REGION

Measurements of neutron-induced fission cross sections for $^{208}\text{Pb}$, $^{209}\text{Bi}$ and $^{238}\text{U}$ have been performed in the 70-160 MeV energy region at the neutron beam facility at the The Svedberg Laboratory in Uppsala using the $^7\text{Li}(p,n)$ reaction as neutron source. The fission fragments were detected by thin-film breakdown counters. The neutron flux was measured relative to the n-p scattering cross section with a proton recoil spectrometer.

Paper II

COMPILATION AND SYSTEMATICS OF PROTON-INDUCED FISSION CROSS SECTION DATA

A compilation of proton-induced fission cross section data in EXFOR has been performed. Cross section parameterizations for target nuclei from $^{165}\text{Ho}$ to $^{239}\text{Pu}$ have been made on the basis of comparative critical analysis and selection of the data. The overall systematics that was created allows predictions to be made for nuclei for which experimental data do not exist.

Paper III

CASCADE-EXCITON MODEL ANALYSIS OF NUCLEON-INDUCED FISSION CROSS SECTIONS OF LEAD AND BISMUTH AT ENERGIES FROM 45 TO 500 MeV

An extended version of the cascade-exciton model (CEM) of nuclear reactions is applied to analyze nucleon-induced fission cross sections for $^{209}\text{Bi}$ and $^{208}\text{Pb}$ nuclei in the 45-500 MeV energy range. The available data on linear momentum transfer are analyzed as well. The results are compared with parameterizations resulting from a comparative critical analysis of all available experimental data. Systematic discrepancies between calculations and experimental data are revealed. A modification of the CEM is proposed, which significantly improves the model predictions for projectile energies above 100 MeV.

Paper IV

FISSION FRAGMENT DETECTION EFFICIENCY OF THIN-FILM BREAKDOWN COUNTERS IN SANDWICH GEOMETRY

A model for calculating the efficiency for fission fragment detection in thin-film breakdown counters in sandwich geometry is developed. The model, which is presented in an analytic form, is applied in efficiency calculations for fragments from intermediate-energy neutron-induced fission. Information on the angular distribution and linear momentum transfer in fission reactions is needed as input to the model. Corresponding experimental data from the literature have therefore been compiled in order to reveal the systematic behavior of these quantities. Predictions of the systematics, as well as of the efficiency model, are found to be consistent with experimental data.
MEASUREMENT OF NEUTRON-INDUCED FISSION CROSS SECTIONS FOR \(^{\text{nat}}\)Pb, \(^{208}\)Pb, \(^{197}\)Au, \(^{\text{nat}}\)W, AND \(^{181}\)Ta IN THE INTERMEDIATE ENERGY REGION

Neutron-induced fission cross sections for \(^{\text{nat}}\)Pb, \(^{208}\)Pb, \(^{197}\)Au, \(^{\text{nat}}\)W, and \(^{181}\)Ta have been measured relative to \(^{209}\)Bi in the 30 - 140 MeV energy range using the neutron beam facility at The Svedberg Laboratory in Uppsala. The \(^7\)Li(p,n) reaction was employed as a neutron source. The fission fragments were detected by thin-film breakdown counters. Cross sections at specific energies were determined using unfolding techniques with respect to the excitation function and the neutron spectra, the latter obtained from an evaluated nuclear data library. The \(^{181}\)Ta(n,f) cross section have been measured for the first time. The results for other nuclei were compared with earlier data, and discrepancies were found for \(^{\text{nat}}\)Pb and \(^{197}\)Au(n,f) cross sections in the lower part of the studied energy region.
7 Conclusions and outlook

This thesis is devoted to fission studies at intermediate energies. The neutron-induced fission cross section measurements at TSL have been the central issue of the work, covered in Papers I and V. Before the work began, very little information had been available, especially for sub-actinide nuclei. Nowadays, the results presented here comprise the major part of the world database, although the possibilities for improvement are certainly far from being exhausted. This concerns, e.g., the unfolding techniques and the detection efficiency. The treatment of the latter in Paper IV is not the final word, but rather a first step in the development of an approach to the problem. On the other hand, despite its simplicity (which is believed to be an advantage), the model works well at least for the relative efficiency calculations in the forward hemisphere, and thus a reasonable solution has been found for the long-standing problem of efficiency corrections in the cross section experiments.

Phenomenological systematics of proton-induced fission cross sections, developed in Paper II, is more of a final character. The available experimental data are thoroughly covered, and a few "problem spots" are located, which may help in planning further measurements. The systematics can be employed in applications and provide an input in theoretical modeling. The latter is the subject of Paper III, where the (n,f) and (p,f) data met, and their description was attempted in the framework of the cascade-exciton model. Since only a limited success was achieved with the original version of the model, a modification, related to the level density in the fission channel, has been undertaken, which improved the predictions spectacularly. This finding is valuable for further model development.

An important aspect of the experimental work at TSL has been the adaptation of the TFBC technique at high-energy neutron beams in general. In parallel with the cross section measurements, methodical studies were undertaken, resulting in the development of beam monitors for different applications. The approaches to measurements and data processing, developed for the cross section studies, are largely applicable also for the beam monitoring. Moreover, these two directions of work are linked by the common field of cross section standards. The measured standard cross section data, after proper evaluation, can be used for monitoring applications at any neutron beam. These applications have been growing rapidly during the last few years, and so does the number of neutron facility users who need the beam monitoring. Examples are studies of different neutron-induced reactions (e.g., production of radionuclides and neutron elastic scattering), calibration of neutron dosemeters for aircrew, single-event effect studies, etc.

The two last applications are closely related to the problems of human safety and health, and they alone would be sufficient to justify the present study for a broader scientific/technological community and for society at large. Still, an even more ambitious field is accelerator-driven systems (ADS). Such systems might provide a radical solution of the global problem of long-lived radioactive waste. Successful development of ADS requires knowledge of data for underlying nuclear reactions, in particular for fission.

The discovery of fission in 1939 rapidly led to applications, first the development of nuclear weapons, and later civil nuclear power. This motivated huge efforts in both basic and applied research. During the last twenty years, the applications above have reached maturity, and therefore the demand for new nuclear data from the applied community has decreased markedly. This in turn has affected fundamental nuclear physics which has been steadily declining during the last decade.

At present, we are experiencing a rapidly increasing interest in novel nuclear physics applications. Accordingly, nuclear data for applications is again an expanding field. Looking back in the historical mirror, it can be seen that nuclear physics had its strongest moments when fundamental and applied work were united. The present thesis represents a good
example of that this is possible also today. Fission studies are motivated by potential large-scale applications, but at the same time, fission itself provides insight into the basic mechanisms governing the phenomena taking place in nuclei. It is among the most complex processes in nature we attempt to understand from knowledge of the fundamental interaction of its constituents. Thus, research in this area can unify fundamental and applied aspects, combine complexity with a strive for simplicity, and serve as a raw model for future subatomic research.
Acknowledgements

This Ph.D. study has been a long and rather dramatic development across the borders of different countries and the walls of different institutions. Consequently, recalling people whom I wish to thank, I often need to mentally cover large distances in space and time.

First thanks come to my past and present supervisors, in Russia and Sweden respectively. Following an old tradition, I mention them in an alphabetical order: Prof. Vilen P. Eismont, Prof. Jan Källne, Dr. Nils Olsson, and Dr. Andrey N. Smirnov.

Prof. Eismont have been heading the group at KRI for many years, in parallel with being a Head of Nuclear Physics Department. Many valuable ideas in experiment, analysis and modeling were suggested by him. His efforts in the development of new projects and steadily running the current ones during the uneasy past years deserve much respect and gratitude.

It has been a bright experience to study under supervision of Prof. Jan Källne. I was surprised most of all by his ability to see things in an unexpected and non-trivial aspect. Particularly, his exams will be remembered for long. In addition, his help and advice are greatly appreciated in settling living arrangements in Uppsala.

Both Jan Källne and Nils Olsson have been carrying a hard task of fighting my "perfectionism", and I believe that this thesis is quite a step in this way. Nils has been providing me with supervision and advice during the last year. His patience in uncountable iteration of drafts and in teaching me writing scientific papers have been enormous. His help and advice in practical arrangements is much appreciated, too. Special thanks come to him for adjusting his schedule to my one, often far beyond normal working hours, and for giving me rides to Flogsta in hectic days before submission of this thesis to print!

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Next acknowledgements come to those whose efforts allowed the beginning of the joint Swedish-Russian fission research project: Prof. Em. Henri Condé and Dr. Alexander A. Rimski-Korsakov. It was their involvement at that hard time, which has opened our group the window to Sweden and Europe. At that early years, we did not know much about TSL, Uppsala, Sweden at large, and Prof. Condé spent a lot of his time, introducing me and my colleagues with new people and possibilities. This is much appreciated and certainly not forgotten!

Positive attitude of the TSL leaders, Dr. Leif Nilsson and later Dr. Curt Ekström, contributed to the success of the work, as well as excellent skills of the laboratory staff. It is not possible to mention everybody involved here, but first I wish to thank Drs. Per-Ulf Renberg and Olle Jonsson, whose efforts resulted in excellent beams.

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To conclude with the Scandinavian side of Baltic, I wish to thank all people from TSL and INF who know me, who helped here and there with everyday issues, or just made good company without borders.

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