Determination of neutron induced nuclear reaction cross sections by time-of-flight measurements

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Abstract

Neutron Time-of-Flight (TOF) technique is mainly used for neutron cross section measurements. Neutron induced cross sections give probability for neutron interaction with nuclei, in the seminar their energy dependence will be described. Throughout the seminar different facilities for neutron time of flight measurements are presented. Types of experimental uncertainties and their impact on measurements will be explained.
1 Introduction

Major facilities for Neutron Time-of-Flight (TOF) measurements [1] are GELINA [2, 3], n_TOF in CERN [4, 5] and ORELA [6]. The main purpose of this method is neutron induced cross section measurement [7, 8]. Knowledge of the neutron cross sections is needed in many different research fields such as nuclear reactor calculations [9], medical physics [10], radiotherapy, industry or even in search for oil. Theoretical models can describe the energy dependence of the cross section only to a certain degree. Therefore cross sections are mainly determined experimentally and a lot of effort is devoted to minimize their uncertainties. When analysing, measuring and using neutron cross sections there must be taken into account whether isotope is free or bounded inside a molecule (this effect is very important in free hydrogen or hydrogen bounded inside water molecule). Neutrons can be produced by an e$^-$ or p$^+$ accelerator or inside a nuclear reactor. Inside the nuclear reactor neutrons are distributed through entire reactor core, which increases uncertainty in position. With accelerators neutrons can be produced in concentrated ns duration bursts, which decreases uncertainty in time. The great advantage of neutron time-of-flight technique is that it allows measurements of neutron cross sections in entire energy region and uncertainties even smaller than 1% can be achieved.

Neutron time-of-flight technique is the most accurate and therefore the most commonly used method for analysing resonance region of neutron cross section, but also other neutron cross section measuring methods can be used. In neutron activation analysis (NAA) [11] sample is bombarded with neutrons, which are captured inside material nuclides leading to radioactive isotopes, they emit $\gamma$-rays with energies corresponding to the excitation energy. Lead Slowing-Down Spectroscopy (LSDS) [12] is a technique in which neutrons are led on a massive lead block with inside channels for detectors and samples. Mean energy of neutrons causing the fission in sample can be calculated from slowing-down time inside lead. From measurements with
quasi-monoenergetic neutrons neutron cross section value only for this quasi-monoenergy can be obtained (example: spallation process in p+ accelerators) [13]. In integral type of measurements energy dependence of neutron cross section can not be obtained, it can only be confirmed (example: Neutron Activation Analysis). Another type of neutron cross section measurements includes also ultra cold neutrons. Ultra cold neutrons are neutrons with so low energies that their speed is comparable with human running. For such low energy neutrons their gravity is no longer negligible.

2 Resonances in neutron cross section

Neutrons do not have any electric charge and can therefore travel through several cm of matter without any interaction. When neutron interacts with nucleus many reactions can take place, among the most important are: scattering (n,n), neutron capture (n,γ) or fission (n,f). Neutron cross sections are proportional probabilities for these reactions and are commonly retrieved from nuclear data libraries. At some energies reaction cross section suddenly rises for a few orders of magnitude, those peaks are called resonances. Resonance structure of reaction cross section is typical for each nuclei. In Figure 2.1 elastic scattering (Fig. 2.1b) and radiative capture (Fig. 2.1a) cross section for 238U are shown. Each of these resonances, corresponding to a nuclear state μ, is characterized by resonance energy $E_\mu$ and total natural width $\Gamma$, which is related to the lifetime of the resonance state via the Heisenberg uncertainty principle [1]. In Figure 2.1 different shapes of resonances are shown: resonances in capture cross section have symmetric shape, while in scattering cross section resonances have asymmetric shape with minimum, where probability for interaction drops for few orders of magnitude, which results in transparency of material for neutrons of that energy (neutrons go through the material without any interaction). The asymmetric resonance shape is a consequence of quantum interference between the potential and resonance components of the elastic cross section. This effect is very important when choosing proper material for protection against radiation.

Figure 2.1: 238U cross sections: capture cross section (left) and scattering cross section (right).

Resonance region is defined as region, where resonances can be separated one from the other, and the average distance between the resonances $D$ is larger than resonance width $\Gamma$ ($D > \Gamma$). With increasing energy resonances are more closely together (average resonance width decreases) and the resonance width increases, therefore they start to overlap. Although at intermediate energies, the resonance structure still exists ($D > \Gamma$), the structure can no longer
be resolved due to limited instrumental resolution $\Delta E$ [1]. Therefore, in the resonance region, one distinguishes between the resolved resonance region (RRR), for which $D > \Delta E$, and the unresolved resonance region (URR), for which $D < \Delta E$ (see Figure 2.2) [1]. Continuum region is the region at higher energies, where resonance structure disappears, because the distance between the resonances becomes smaller than their width.

In general, the level density increases with the increase in atomic mass, such that resonances are more closely spaced for heavier nuclei and the unresolved resonance region starts at lower energies [1]. Theoretical model that could predict resonance structure does not exist yet, therefore resonances are determined experimentally. In general, measurements of cross section are fitted with resonance parameters, which are later used to reconstruct resonance structure.

Figure 2.2: Comparison of the cross section for neutron induced capture and elastic scattering in $^{238}$U. The resolved resonance region (RRR), unresolved resonance region (URR), and continuum region are indicated [1].

3 Physical background of neutron TOF

The TOF technique is based on the measurement of the time that a neutron needs to travel a given distance $L$ [1]. This time $t$ can be used to determine the neutron speed $v$ by [1]:

$$v = \frac{L}{t}. \quad (1)$$

In nonrelativistic case, when neutron has low speed, neutron kinetic energy can be calculated as:

$$E = \frac{1}{2}mv^2. \quad (2)$$

When neutron has high speed (compared to speed of light), its kinetic energy must be calculated using relativistic expression:

$$E = mc^2(\gamma - 1), \quad (3)$$

where $\gamma$ stands for relativistic Lorentz factor:

$$\gamma = \frac{1}{\sqrt{1 - (v/c)^2}}. \quad (4)$$
where \( c \) represents the speed of light. Experimentally, the TOF of a neutron is determined from the difference of the arrival time and a start signal [1]. The TOF \( t \) corresponding to the distance \( L \) is different from the observed time difference \( t' \), which is obtained from the difference between the stop signal delivered by the detector and the start signal obtained from the pulsed beam [1]. The TOF \( t \) is in fact a bit smaller than observed time, the difference is due to a correction factor \( dt \), which represents neutron transport inside target, moderator and detector:

\[
t = t' - dt.
\]  

(5)

The neutron transport inside the target and moderator is usually calculated and measured time is corrected, the difference between measured and real time of flight depends mostly on neutron energy and material and thickness of target and moderator. From measuring neutron time-of-flight, its energy can be determined. Two different measurement types can be performed: transmission and reaction analysis. The major difference is in type of particles that are being detected.

### 3.1 Transmission measurement

In neutron reaction transmission analysis (NRTA), the measured quantity is the fraction of the neutron beam that traverses the sample without any interaction [1]. \( \phi_{in} \) is the incident neutron beam and \( \phi_{out} \) is the neutron beam leaving the target. Experimentally, the transmission \( T_{exp} \) is obtained from the ratio of the counts of a sample-in measurement \( C_{in} \) and a sample-out measurement \( C_{out} \), after proper subtraction of the background contributions \( B_{in} \) and \( B_{out} \), respectively [1]:

\[
T_{exp}(E) = \frac{\phi_{in}}{\phi_{out}} = \frac{C_{in}(E) - B_{in}(E)}{C_{out}(E) - B_{out}(E)} = e^{\Sigma_{tot}(E)d},
\]

(6)

where \( d \) represents sample thickness and \( \Sigma_{tot}(E) \) total macroscopic cross section of sample material, which represents probability for neutron interaction in unit of traveled distance. Taking into account relation between microscopic and macroscopic cross section, microscopic cross section \( (\sigma) \) can be subtracted:

\[
\Sigma(E) = \sigma(E)nd,
\]

(7)

\[
\sigma_{tot} = \frac{\ln \frac{\phi_{in}}{\phi_{out}}}{nd},
\]

(8)

where \( n \) stands for atomic density of target. This method is very simple and therefore it is most commonly used. Typically more samples of different thickness are placed inside neutron beam for optimal resolution of different resonances. From transmission (eq. (6)) can be seen that \( \Sigma_{tot}d \) must be \( \sim 1 \), because of that thin samples are used for observation of large resonances, while thick samples are used for minor resonances.

### 3.2 Reaction measurement

When neutron interacts with target nuclei many different reactions can take place: scattering, fission, capture or charged particle emission. Among different neutron reactions, neutron capture is the most common, therefore hereinafter more about reaction of neutron capture will be described.

Neutron resonance capture analysis (NRCA) is based on the detection of the prompt \( \gamma \)-radiation
emitted after a neutron has been captured in the sample [1]. The quantity determined in a capture experiment is the fraction of the incident neutrons undergoing a reaction in the sample and creating signal in the detection system [1]. This fraction depends on capture yield and can be expressed as [1]:

\[ Y(E) = F(E)n\sigma(\gamma) + Y_M(E), \]  

(9)

where \( \sigma(\gamma) \) is microscopic capture cross section and \( Y_M \) accounts for the contribution of neutrons scattered at least once before their capture in the sample [1]. In most cases when sample is thin, the first term in Equation 9 adds the majority of signal and multiple scattering contribution of \( Y_M \) can be neglected. This simplification is justified when target thickness is much smaller than the average mean free path of the neutron inside the target. The self-shielding factor \( F(E) \), caused by the attenuation of the neutron beam in the sample, is defined as [1]:

\[ F(E) = \frac{1 - e^{-\frac{n\sigma_{tot}}{n}}} {n\sigma_{tot}}, \]  

(10)

where \( \sigma_{tot} \) is total microscopic cross section.

For detecting \( \gamma \) rays produced at capture experiment, detectors need to be placed all around the target, example of detector position is presented in Figure 3.3.

![Figure 3.3: View of CERN n-TOF \( \gamma \) detectors [15].](image)

4 Facilities for TOF measurements

In the TOF facility, neutrons used for the neutron cross section measurements are produced by the impact of a short pulse of high-energy particles (electrons or protons) on a neutron-producing target [2]. Three main facilities for neutron TOF measurements are presented below, their characteristics are later compared and analysed.

4.1 GELINA (IRMM)

The Geel Electron LINear Accelerator Facility (GELINA) at the Institute for Reference Materials and measurements (IRMM) of the European Commission’s Directorate-General Joint Research Centre (JRC) is especially designed for TOF measurements. GELINA can perform simultaneous experiments within 12 different flight paths. Neutrons are produced in bunches of less than 1 ns duration, the total neutron production in the target is \( 3.4 \times 10^{13} \) neutrons/s.
Energy resolution (specially in the resonance region) is the most important design criterion, and GELINA has among the white spectrum neutron sources the best energy resolution (below 1%) in the world.

Main components of GELINA is a linear electron accelerator, which generates a 10 ns long electron pulse. The energy of the electrons in the pulse leaving the accelerator varies linearly from 140 MeV at the start of the pulse to 70 MeV at the end of the pulse, due to the beam loading [2]. Before hitting the target, the electrons make a “looping” in a specially designed 360° compression magnet [2]. Inside the magnetic field, electrons with higher energy will have longer trajectory (because of bigger bending radius). The magnet is designed so that all electrons of a 10 ns pulse, entering the magnet, will leave the compression magnet within a time bin of 1 ns [2].

After leaving the magnet, electrons are led upon a U-Mo neutron producing target, with 10 wt.% of Mo. The target is rotating in the beam [3]. Each electron produces about 6 neutrons. Mercury is chosen as a coolant, mainly to avoid neutron moderation [3]. The electrons inside target decelerate and produce high-energy photons via the Bremsstrahlung process. These photons may interact with target nuclei via photon induced nuclear ($\gamma$,n) reaction and to much lesser extent by fission ($\gamma$,f) reaction [2]. In order to have significant number of neutrons in the energy region below 100 keV, two light water moderators are placed above and below the target [3]. Experimental configuration enables two types of flux: Direct Flux Configuration (DFC) and Moderated Flux Configuration (MFC).

Around neutron producing target 12 flight paths for TOF measurements are installed in star shape (see Figure 4.4). The flight tubes are under vacuum, have a diameter of 50 cm and a length ranging up to 400 m [3]. Several measurement stations are installed at different distances along the flight path [2]. These experimental stations are equipped with a wide variety of detectors and data acquisition systems, especially designed for neutron-induced total and partial cross-section measurements with exceptional precision and energy resolving power [3].

![Figure 4.4: GELINA flight paths star shape configuration [2].](image)

### 4.2 n_TOF (CERN)

At the n_TOF facility at CERN (see Fig. 4.5), a highly intense, pulsed neutron beam is produced by spallation reactions of a pulsed 20 GeV proton beam from the CERN Proton Synchrotron on a massive lead target [4]. Each proton produce about 300 neutrons, which is a major advantage of the n_TOF CERN facility. Produced neutrons have relatively large energies and are slowed down, first within the target and then by 5 cm water shield surrounding the target, which serves as moderator and as coolant. The resulting neutron flux approximates an energy dependence proportional to $1/E_n$ and ranges from thermal (25 meV) up to few GeV. Neutrons produced in the target, enter 200 m time-of-flight tunnel. Along the flight path, two collimators placed at 135 and 180 m from the spallation target, are used to shape the neutron beam [5]. A sweeping magnet placed at 40 m upstream the experimental area is used
to deflect outside the beam charged particles travelling along the vacuum pipe [5]. For an efficient background suppression, several concrete and iron walls are placed along the time-of-flight tunnel [5]. The measuring station is located inside the tunnel, centred at 187.5 m from the spallation target, and delimited by two concrete walls 7.5 m apart [5]. An escape line, 12 m long and ending in a polyethylene block, ensures a negligible background from the backscattered neutrons and capture γ-rays [5].

4.3 ORELA (ORNL)

The Oak Ridge Electron Linear Accelerator (ORELA) was recently shut down permanently. ORELA consists of a 180 MeV electron linear accelerator; a neutron-producing, water-cooled Ta target; underground and evacuated flight tubes; sophisticated detectors and data acquisition systems [6]. Pulse widths from 4 - 30 ns are available at a repetition rates from 12 - 1000 pulses per second. Simultaneous measurements are possible at 18 detector stations on 10 separate flight paths at distances between 9 and 200 m from the neutron source [6].

4.4 Comparison of different facilities

Above three main neutron TOF facilities are described their basic information are gathered in Table 4.1.

Table 4.1: Characteristics of TOF facilities used for neutron cross-section measurements [7].

<table>
<thead>
<tr>
<th>Facility</th>
<th>Type</th>
<th>Particle Energy [MeV]</th>
<th>Target</th>
<th>Pulse width</th>
<th>Frequency [Hz]</th>
<th>Flight path length [m]</th>
</tr>
</thead>
<tbody>
<tr>
<td>GELINA</td>
<td>e</td>
<td>80 - 140</td>
<td>U</td>
<td>1</td>
<td>40 - 800</td>
<td>10 - 400</td>
</tr>
<tr>
<td>n_TOF</td>
<td>p</td>
<td>20000</td>
<td>Pb</td>
<td>6</td>
<td>0.4</td>
<td>185</td>
</tr>
<tr>
<td>ORELA</td>
<td>e</td>
<td>140</td>
<td>Ta</td>
<td>2 - 30</td>
<td>1 - 1000</td>
<td>10 - 200</td>
</tr>
</tbody>
</table>

The Figure 4.6 demonstrates the difference between neutron TOF experiment based on spallation source (n_TOF in CERN) and white neutron source from photnuclear reactions (GELINA, ORELA).
The resonance width of n_TOF facility in CERN resonance is larger than from GELINA or ORELA even though neutron flight path at CERN is larger. Neutrons at n_TOF facility are produced from spallation process and are primary particles, while in GELINA and ORELA first electrons produce primary $\gamma$-rays, which than produce secondary neutrons with continuous energy. Energy of neutrons produced by spallation process can be changed by changing the accelerator voltage or by flight paths at different angles. The difference between GELINA and ORELA is mainly due to different flight paths, and because GELINA had longer flight path, its resonance width is smaller (because of smaller resolution broadening) compared to ORELA. Due to different characteristics, these facilities complement each other. For measurements with high energy neutrons n_TOF in CERN is the best option, while on the other hand for measurements with better resolution GELINA or ORELA can be used.

5 Uncertainties

The main purpose of neutron TOF measurements is to determine neutron cross section as precisely as possible, with as low experimental uncertainties as possible. In order to achieve this goal, the below-stated causes of uncertainty have to be minimized.

5.1 Final temperature (Doppler resonance broadening)

Thermal motion of the target nuclei causes Doppler broadening of the resonance peaks: as the target temperature increases, the peaks become broader while their areas remain practically constant (see Figure 5.7) [8]. Doppler broadening can be avoided with cooling the sample to temperature $\sim 0$ K, which is experimentally very challenging. In practise neutron cross sections are needed at different temperatures and their values for different temperatures are reported in nuclear data libraries or are measured experimentally.
5.2 Time and distance resolution

The energy resolution obtained at TOF spectrometer depends on the speed resolution $\Delta v$ and is determined by [1]:

$$\frac{\Delta E}{E} = (\gamma + 1)\gamma \frac{\Delta v}{v}. \quad (11)$$

At low speeds, $\gamma \approx 1$ and the relative energy resolution is twice the relative speed resolution [1]. The uncertainty in the neutron speed results from the resolution-broadening $\Delta t$ and uncertainty in distance $\Delta L$, respectively [1]:

$$\frac{\Delta v}{v} = \sqrt{\left(\frac{\Delta t}{t}\right)^2 + \left(\frac{\Delta L}{L}\right)^2}. \quad (12)$$

The distance $L$ is determined by metric measurements is the one between the outer surface of moderator and the front surface of detector (sample) in the case of transmission (capture) measurements [1]. The uncertainty in $L$ is less than $1$ mm ($\Delta L < 1$ mm). The speed resolution $\Delta v$ is almost completely determined by the broadening due to determination of the TOF resulting from [1]:

- finite duration of accelerator burst,
- neutron transport in the target and moderator,
- neutron transport in the detector in the case of transmission measurements,
- time resolution of the detector and electronics [1].

In Figure 5.8 the impact of the flight path length on the energy resolution is presented. At low energies the main impact on resolution is due to Doppler broadening and there is no observed difference due to neutron flight path length (left figure). At higher energies, the resolution starts to dominate and the observed width will be smaller with increasing flight path distance as can be deduced from Equation (12) (right figure). Since the neutron flux is in first approximation inversely proportional to the square of the distance, a compromise between the resolution and intensity has to be made when choosing the distance [1].
Figure 5.8: The results of capture measurements on a 0.12-mm-thick Au metal disk at 12.5 and 30 m flight path at GELINA are compared in two energy regions [1].

5.3 Background (γ,n)

The background in TOF transmission measurement can be considered as a sum of a time independent and time dependent components:

\[ B(t) = B_0 + B_\gamma(t) + B_{no}(t) + \ldots \]  

Time independent component \( B_0 \) can be subtracted by background measurements before the experiment. \( B_\gamma \), time dependent contribution is mainly due to neutron capture in hydrogen (slowing down process in moderator), which results in 2.2 MeV \( \gamma \) rays. \( B_{no} \) time dependent component represents overlap neutrons\(^1\) - this contribution is mainly present at longest flight paths. Time dependent components can mainly be subtracted by insertion of different filters inside neutron beam [17].

6 Conclusion

Precise knowledge of the neutron induced cross sections is very important in neutron transport calculations and other applications such as activation analysis of samples. Neutron induced cross sections have very strong energy dependence, its important part are resonances, which are typical for each nuclide. Some theoretical models of resonances exist but can only describe the shapes (energy dependence) but not resonance energies and widths, therefore effort is placed into experimental determination of the resonance structure. Neutron time-of-flight technique is a very precise way to analyze the whole energy region of cross section and it enables high energy resolution needed for resonance structure measurements. The uncertainty in measurements depend mostly on time resolution of experimental configuration (duration of neutron burst, \ldots). Among different types of neutron TOF technique, from the uncertainty point of view, the best is white neutron source, produced from Bremsstrahlung of electrons from accelerator inside target. With optimal experimental configuration, uncertainties smaller than 1 % can be achieved. The uncertainties in neutron cross section remain main source of uncertainties in nuclear reactor calculations. This facilities provide satisfactory precision in neutron cross section measurements, but in the future when new techniques will be developed, the uncertainties in neutron cross sections can be even reduced.

\(^1\)Detected neutrons, which were produced in previous cycle.
7 References


