Measurement of the differential neutron-deuteron scattering cross section in the energy range from 100 keV to 600 keV using a proportional counter

R. Nolte$^1$, J. Beyer$^1$, A. Plompen$^2$, S. Röttger$^1$

$^1$Physikalisch-Technische Bundesanstalt (PTB), Bundesallee 100, 38116 Braunschweig, Germany

$^2$Institute for Reference Materials and Measurements (IRMM), Retieseweg 111, 2440 Geel, Belgium

Abstract

The angular distribution of neutron-deuteron scattering was investigated using the proportional counter P2 simultaneously as scattering target and detector for the recoil deuterons. The measurements were carried out using monoenergetic neutrons in the energy range from 150 keV to 500 keV. Various techniques were employed to reduce distortions of the experimental pulse-height distribution by photon-induced events. The experimental data were compared with realistic simulations which were carried out using different evaluated data sets. This comparison allows to conclude on inconsistencies in the evaluations.

1 Introduction

Next to the neutron-proton (n-p) scattering ($N = 2$), neutron-deuteron scattering ($N = 3$) is the most fundamental interaction process in a few-body quantum system consisting of $N$ nucleons. It can be described by the re-formulated Fadeev three-body equations [1] using the well-developed nucleon-nucleon potentials for the interaction between the three nucleons involved. These calculations covered the energy range from 3 MeV to 19 MeV which were later extended to a lower limit of 50 keV [2].

In addition to its relevance for understanding quantum mechanical few-body systems, the differential neutron-deuteron (n-d) scattering cross section is also relevant for nuclear technology, in particular for the design and safe operation of heavy-water moderated reactors, e.g. of the CANDU design. Several critical and subcritical benchmark experiments for heavy-water moderated configurations demonstrated the sensitivity of the effective neutron multiplication factor $k_{\text{eff}}$ and the coolant void reactivity (CVR) to the angular distribution of the neutron-deuteron scattering cross sections [3]. In particular, significant changes in the calculated $k_{\text{eff}}$ and CVR values were observed when the data of the ENDF/B-VI.3 [4] library were replaced by data from later releases.

It is very striking to see that the experimental data base supporting the evaluations are rather scarce and partially inconsistent, with some of the measurements dating back to the 1950’s and 1960’s. As an example, Figure 1 shows the experimental data available from EXFOR [5] for the energy range from 100 keV to 2 MeV. The data are grouped into four narrow energy intervals and compared with angular distributions from ENDF/B-VII.0 [6] calculated for the mean energy of each energy interval. The inconsistency of the available experimental data and, in particular, the difficulty to reproduce the results of benchmark experiments have prompted the inclusion of the differential neutron-deuteron scattering cross section in the OECD high-priority request list (HPRL) [7] for urgent nuclear data measurements.
As for neutron-proton scattering, the differential neutron-deuteron scattering cross sections can be measured either by detecting the scattered neutron or the recoiling deuteron. While the first approach is most suited for higher neutron energies, it poses difficulties at lower energies because efficient and well-characterized neutron detectors are difficult to find in the energy range below a few hundred keV. The present contribution reports results of new measurements using a proportional counter simultaneously as target and detector for the recoiling deuterons. This technique was already used for the measurements of some of the earlier data available from EXFOR. Therefore, the present work also aims at improving the potential of the method by proper modelling of deteriorating experimental influences.

Fig. 1: Experimental angular distributions (symbols) for neutron-deuteron scattering for the energy range from 100 keV to 2000 keV compared with evaluated angular distributions from ENDF/B-VII.0 (solid lines) calculated for the mean neutron energy of the data sets.

2 Experimental Technique

For elastic neutron scattering in non-relativistic approximation a simple relation exists between the energy \( E_R \) of the recoil nucleus in the laboratory (LAB) system and the scattering angle \( \Theta'_n \) in the centre-of-mass (CM) system,

\[
E_R = E_n \left( \frac{4A}{A+1} \right)^2 \frac{1 + \cos(\Theta'_n)}{2}
\]

where \( A = m_T/m_n \) denotes the ratio of the mass \( m_T \) of the target nucleus to the mass \( m_n \) of the neutron. Because of this relation the energy distribution of the recoil nuclei in the LAB system is directly related to the differential scattering cross section \( (d\sigma/d\Omega_n) \) in the CM system,

\[
\left( \frac{d\sigma}{dE_R} \right) = \left( \frac{d\sigma}{d\Omega_n} \right) \left( \frac{E_R}{E_R^{\text{max}}} \right)
\]

Here, \( E_R^{\text{max}} = 4A/(A+1)^2 \) \( E_n \) is the maximum energy of the recoil nucleus in the LAB system. For neutron-proton and neutron-deuteron scattering the kinematical factor \( 4A/(A+1)^2 \) is approximately 1 and 8/9, respectively. Hence the angular distribution in the CM system is directly proportional to
the distribution of energies deposited by the recoil nuclei. In an ideal detector for recoil nuclei, this distribution is identical to the pulse-height (PH) distribution.

In real detectors, however, several effects deteriorate this simple relation because the energy deposition is only the first step of the detection process and is followed by production and transport of scintillation light in case of scintillation detector or the electron-ion pairs in case of gas detectors. In a proportional counter, incomplete energy deposition by particles leaking out of the sensitive volume (wall effects), sensitivity of the counter to parasitic photons and a possible energy dependence of the mean energy $W$ required to produce an electron-ion pair distort the PH spectra and have to be accounted for in the analysis of the measurements. This can be achieved by an iterative comparison of a realistic Monte Carlo simulation with the experimental PH spectra and an adjustment of the angular distributions.

The present measurements were carried out using the PTB recoil proton proportional counter (RPPC) P2. A sketch of the RPPC is shown in Fig. 2. P2 is routinely used to measure the total fluence for neutron energies below 1.2 MeV. The use of this type of detector was described in detail by Skyrme et al. [8]. The RPPC P2 was constructed in compliance with this reference, with only slight modifications of the mechanical details. It consists of a cylindrical stainless steel housing, 0.5 mm thick, 76 mm in diameter and 360 mm in length. The thickness of the stainless steel entrance window is 0.5 mm. The size of the sensitive volume within this housing, 55.5 mm in diameter and 193.3 mm in length is restricted and defined by a cylindrically shaped cathode made of aluminium, 0.3 mm thick, and by guard tubes arranged at both ends of the anode wire.

Fig. 2: Recoil proton proportional counter P2 used at the PTB as the primary reference instrument for fluence measurement of neutrons with energies up to 1.2 MeV. C: cathode cylinder, A: anode wire (gold-plated tungsten wire 100 µm in diameter), F: field tube, G: guard tube, S: sensitive volume (shaded in grey). The neutrons are usually incident along the axis of the counter.

The guard tubes are held at ground potential, while the potential of anode and cathode are selected such that the cylindrical equipotential surfaces extent undisturbed into the volumes before and behind the sensitive volume between the guard tubes. The gas of the RPPC must meet the requirements of a well-known hydrogen content and a high gas amplification. The optimal gas filling depends on the neutron energy. For neutron energies below 300 keV, P2 is operated with a mixture of $\text{H}_2$ (96.5 vol%) and $\text{CH}_4$ (3.5 vol%) while propane ($\text{C}_3\text{H}_8$) is used at higher energies to reduce the range of recoil protons and limit the influence of incomplete energy deposition by recoil particles escaping the sensitive volume through the annular or rear surface or entering it through the front surface. For deuterated gases the use of a $\text{D}_2/\text{CD}_4$ mixture can be extended to about 500 keV because of the smaller ranges of the deuterons at a given kinetic energy.

For the present experiments, P2 was operated with the $\text{D}_2/\text{CD}_4$ mixture at a pressure of 1000 hPa (965 hPa $\text{D}_2$ and 35 hPa $\text{CD}_4$). In addition, measurements were also carried out using a $\text{H}_2/\text{CH}_4$ mixture at the same pressure and $\text{C}_3\text{H}_8$ at 600 hPa to identify a possible distortion of the
PH spectra. The isotopic purity of the deuterium in the D₂ and CD₄ was 99.8% and 99.9%, respectively. The chemical purity of the D₂, H₂ and CH₄ gases were better than 99.999% while the purity of the CD₄ was only 99.9%. Therefore, oxisorb cartridges were used to remove traces of oxygen and water from the gas during the filling process.

The neutrons fields were produced in open geometry using the $^7\text{Li}(p,n)^7\text{Be}$ reaction. The measurements were carried out in the low scatter hall of the PTB ion accelerator facility PIAF using proton beams from the 3.7 MV Van-de-Graaff accelerator. Data were taken for mean neutron energies $E_n$ of about 145 keV, 200 keV, 250 keV, 300 keV and 500 keV with and without a polyethylene shadow cone, 300 mm in length, for subtraction of room-return neutrons. The neutron fields had a 2% - 4% contribution of non-monoenergetic neutrons resulting from neutron scattering in the target. The spectral distribution of these neutrons was calculated using the TARGET code [9].

The proportional counter is also sensitive to photons. Hence, photon-induced events can deteriorate the determination of the angular distribution of neutron-deuteron scattering from the PH distributions. Therefore, several measures were undertaken to minimize photon interference as much as possible. The contamination of the neutron field with photons was reduced as much as possible by employing a metallic lithium target. The lithium mass per unit area was 70 µg/cm² and the target backing consisted of tantalum, 0.5 mm in thickness. The pressure in the counter was adjusted such that the energy deposition by Compton electrons directed parallel to the counting wire did not exceed 40 keV in case of the D₂/CD₄ mixture at 1000 hPa and 125 keV for the C₂D₄ gas at 600 hPa, while still keeping the wall effects for recoil particles below an acceptable level (see below).

For some neutron energies a cylindrical lead absorber, 21 mm in thickness and 82 mm in diameter, was placed between the target and the counter to further suppress the photon contamination. At the energies used in the present work, neutrons interact with lead only by elastic scattering which does not affect the spectral distribution of the field significantly. A PH distribution produced by photons only was measured at a proton energy of 1880 keV, i.e. below the $^7\text{Li}(p,n)^7\text{Be}$ threshold at 1881 keV. Assuming that the spectral distribution of the contamination photons has a weak energy dependence, this PH distribution was used to correct for the photon contribution for the neutron beams with energies between 145 keV and 250 keV, i.e. for proton energies between 1945 keV and 2021 keV. It should be noted here that a considerable fraction of the photon contamination is subtracted anyway by the shadow cone measurement. The residual photon interference is only caused because some photons are absorbed in the shadow cone which makes the subtraction of the photon component incomplete.

The sensitivity of the counter to the residual photons was suppressed by an analogue rise time discrimination scheme [10]. In a proportional counter the tracks caused by Compton electrons are much longer than those from recoil proton or deuterons. Hence, as shown in Fig. 3, the drift times of secondary electrons from a long electron track show a considerably larger spread than those from the short track of a recoil particle, unless the tracks are almost collinear with the counting wire. This spread in drift times is reflected in the rise time (RT) of the anode signal. In the present experiment, the rise time of the anode signal after shaping by a charge-sensitive preamplifier and a fast-filter amplifier was determined from the time difference between the outputs of a leading edge discriminator (LE) set just above the noise level and a constant-fraction discriminator (CFD) triggering at about 40% of the maximum signal amplitude ($f = 0.4$).

The events were sorted in a RT versus PH matrix. In this matrix the recoil events cluster on a ridge while the electron events have a wider distribution at low PH. Fig. 4 shows a RT vs. PH matrix for a neutron energy of 300 keV. The events contained in the shaded region are those effected by recoil deuterons. Of course, this RT discrimination cannot be perfect because at higher
energies the track length of recoil particles becomes similar to that of electrons in the sensitive volume.

**Fig. 3:** Discrimination of recoil proton or deuteron events (short tracked labelled p) from those produced by Compton electrons (long track labelled e) using the different spread of the drift times of secondary electrons which is reflected in the rise time of the anode signal. The rise time is determined from the time difference of the outputs of two discriminators, one (LE) operating just above the noise level and one (CFD) at about 40% of the maximum signal amplitude.

**Fig. 4:** RT (vertical axis) versus PH (horizontal axis) matrix for a neutron energy of 300 keV. The data points inside the shaded polygon are those produced by recoil deuterons.

**Fig. 5** shows a comparison of the suppression of photon-induced events by the RT discrimination technique to that achieved using either the lead absorber (right panel) or the subtraction of a sub-threshold PH distribution (left panel). Obviously all techniques result in almost equivalent net PH distributions, except for very low pulse-height, where the lead-absorber seems to give slightly better results.
Fig. 5: PH distributions after subtraction of the shadow cone measurement. The black histograms are without further photon suppression. The red histograms show the effect of the RT discrimination. The blue histogram in the left panel was obtained by subtraction of a sub-threshold PH distribution from the black histogram. The blue histogram on the right panel shows the effect of using a lead absorber instead of applying the RT discrimination technique.

3 Results

The data analysis for the present experiment at PTB is based on a dedicated Monte Carlo code which simulates the scattering of neutrons on hydrogen ($^1$H), deuterium ($^2$H) and carbon ($^{12}$C). The differential cross sections are sampled from Legendre expansions. For $^1$H the coefficients from ENDF/B-VII.0 were used directly. For $^2$H the coefficients were determined from fits to the tabulated angular distributions of the ENDF/B-VII.0, JENDL 4.0 [11] and ENDF/B-V1.3 libraries. For $^{12}$C data from ENDF/B-VII.0 were used. A logarithmic-linear interpolation scheme was used to obtain coefficients for all neutron energies. The recoil particles are tracked using range data calculated with the SRIM2013 code [12]. With the employed option of the code energy and angular straggling are not simulated. For $^2$H/CH$_4$ and $^3$H/CD$_4$ an energy-independent $W$ value was used for protons and deuterons based on the data of Breitung [13]. A linear dependence of the $W$ value for carbon ions on $\log(E/\text{keV})$ was assumed with a slightly modified slope compared with the data of Posny et al. [14] for propane. The neutron transport in the other counter materials was not modelled because calculations using MCNPX showed that neutron scattering on structural materials had a negligible effect on the shape of the PH distributions.

Figures 6 - 8 show a comparison of simulated and experimental PH distributions for the five neutron energies between 145 keV and 500 keV and for the two hydrogen isotopes. The simulated distributions were folded with a Gaussian response function of constant relative width to model the PH resolution of the instrument. The maximum order of Legendre polynomials used for fitting the angular distributions for $^2$H was $l_{\text{max}} = 1$ below 500 keV and $l_{\text{max}} = 2$ at 500 keV.
**Fig. 6:** PH spectra measured for 145 keV neutrons (left panels) and 201 keV neutrons (right panels) with D₂/CD₄ (upper panels) and H₂/CH₄ (lower panels) at a pressure of 1000 hPa (histograms). The solid lines show the calculated spectra obtained with the differential n-d and n-p cross sections from ENDF/B-VII.0 (red line), JENDL 4.0 (blue line) and ENDF/B-VI.3 (green line). The calculations were fitted to the experimental data in the recoil energies range above 35 keV (left panels) and 50 keV (right panels), corresponding to a neutron-deuteron scattering angle $\Theta_n$ of 117.4° and 116.1°, respectively. The experimental data were obtained using rise time discrimination of photon-induced events. In addition a lead absorber was employed to reduce the photon contamination of the neutron field.

The good agreement of the measured pulse-height spectra with the calculations, except for the very low pulse heights, confirms the angular distributions from ENDF/B-VII.0 or JENDL 4.0 which are quite similar at these low neutron energies.

**Fig. 7:** Same as for Fig. 6 but for a neutron energy of 248 keV (left panels) and 297 keV (right panels). The calculations were fitted to the experimental data in the recoil energy range above 60 keV (left panels) and 100 keV (right panels), corresponding to a neutron-deuteron scattering angle $\Theta_n$ of 117.1° and 104.1°, respectively.
Fig. 8: The upper panel shows measured calculated PH spectra for 498 keV neutrons. The lower panel shows the angular distributions in the CM system from ENDF/B-VII.0 (red line), JENDL 4.0 (blue line) and ENDF/B-VII.3 (green line) for a neutron energy of 500 keV. The calculations were fitted to the experimental data in the recoil energies range above 120 keV, corresponding to a neutron-deuteron scattering angle $\Theta_n$ of 117.3°.

At 300 keV, the fit of experimental and calculated spectra was restricted to recoil energies higher than 100 keV because there is a mismatch for smaller recoil energies for the data taken with both gases. Since the ranges of photons and deuterons differ by about a factor of about $\sqrt{2}$ for protons and deuterons of the same energy, it is not very likely that this mismatch is due to problems with the description of wall effects, but it could be due to a residual contribution of photon-induced events to the experimental spectra.

At 500 keV there is a clear difference in the angular distributions from ENDF/B-VII.0 and JENDL 4.0 as well as ENDF/B-VI.3. Only the ENDF/B-VII.0 distribution fits the measured spectra almost over the entire range of recoil deuteron energies. Unfortunately, it was not possible to obtain data for recoil protons stopped in $\text{H}_2/\text{CH}_4$ gas at this energy because the ranges already become too high so that wall effects would dominate the shape of the PH distribution.

4 Conclusions

From the present experiments it can be concluded that the angular distributions for $^2\text{H}(n,n)^2\text{H}$ at 500 keV are less backward peaked than predicted in JENDL 4.0 and are better represented by the ENDF/B-VII.0 distributions. The angular distribution from ENDF/B-VI.3 is considerably off for almost all neutron energies investigated. However, there are still open questions left about the influence of either a possible non-linearity in the PH response of the proportional counter or the incomplete discrimination of photon-induced events. Hence, further investigations are required.

Acknowledgment

The authors would like to thank the staff of the PTB ion accelerator facility for providing the beams and S. Löh and M. Thiemig for their support during the measurements. This work was partly supported by the Commission of the European Community through the ERINDA project (grant agreement no. 269499).
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