

Neutron inelastic scattering, recent experiments and their interpretation

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Abstract

Measurements of inelastic scattering and (n,xn) -cross sections with the $(n,xn\gamma)$ -technique are performed at the GELINA neutron time-of-flight facility with two arrays consisting of high purity germanium detectors, GAINS and GRAPHEME. These measurements provide important nuclear data for criticality, reactivity and power distribution estimates in current and advanced power reactors, for the development of active material interrogation techniques for security and safeguards, and for background studies supporting the search for neutrinoless double-beta decay in experiments like GERDA, and MAJORANA and for weakly interacting massive particles. Despite significant advances in modeling, such cross sections still pose a major challenge to nuclear theory at the level of the required accuracy. GAINS is an array consisting of 12 large volume detectors used to study inelastic scattering from C to Bi with high incident neutron energy resolution. GRAPHEME using four planar detectors, is tailored for the actinides. Recent and ongoing experimental work for ^{23}Na , ^{76}Ge , W and ^{232}Th is presented. The experimental work is supported and complemented by state-of-the-art nuclear modeling with the well-known TALYS code using both a phenomenological and a microscopic approach, and with resonance analysis for selected nuclides. Advances and open issues will be shown. For carbon interesting complementary results were obtained using single-crystal diamond detectors.

1 Introduction

Remarkably, there is still a strong current interest in neutron inelastic scattering and $(n,xn\gamma)$ -reactions that derives from innovation in nuclear energy [1, 2], the development of active material interrogation techniques for security and safeguards, and from background studies supporting the search for neutrinoless double-beta decay in experiments like GERDA [3], and candidate dark matter particles [4]. Remarkable since the history of neutron inelastic scattering is a long one dating back from the period shortly after the discovery of the neutron. A brief recap.

Conclusive experimental evidence [5] for $(n,2n)$ reactions (on ^{63}Cu and ^{65}Zn) was first established at the N.V. Philips Gloeilampenfabrieken, Eindhoven, Holland, by an activation method confirming the half-life, employing radiochemistry to eliminate the end products were neighboring elements and

deflecting the emitted particles to establish β^+ -decay [6]. Inelastic scattering was established a year later. Following several indications of "excitation without capture" of materials by fast neutrons [5], as summarised by Livingston and Bethe in Part C of an extensive review of nuclear physics, experimental proof of neutron inelastic scattering was first established by Seaborg, Gibson and Grahame using a radium-beryllium neutron source, various configurations involving a large lead-block and a Geiger-Mueller counter [7]. The experiments demonstrated negligible loss of neutrons traversing the lead block with or without various other materials around the source, the ability of neutrons to excite the lead even after deep penetration, the minor role of slow neutrons in producing these gamma-rays and the reduced production of gamma-rays when other materials shield the source. Thus, it was established that 1) neutrons are not significantly captured as they produce gamma-rays and 2) their ability to excite lead is reduced when they lose energy. Implications for the nuclear reaction theory of Weisskopf and Ewing were sought by Dunlap and Little using D-D neutrons and a cloud chamber [8]. They were unsuccessful as the 2.5 MeV neutrons mostly scattered from discrete levels. The suggested discrete energies of the outgoing neutrons following inelastic scattering were exhibited using photographic emulsions and the Li(p,n) source reaction by Stelson and Preston for the first level of ^{56}Fe [9]. Quantitative studies detecting gamma-rays took off with the advent of NaI scintillator counters [10, 11] and for detection of neutrons through time-of-flight measurements at quasi mono-energetic pulsed neutron sources with fast hydrogenous scintillators as detectors. The latter technique was pioneered by Cranberg and Levin at Los Alamos for iron [12]. The highest resolution measurements of this type were developed much later by Haouat and co-workers and applied to ^{238}U [13]. The first neutron-gamma coincidence measurements were performed early fifties as well with the aim of curbing the ever important background in neutron experiments [14].

Measurements at incident-neutron time-of-flight facilities with a white neutron spectrum were established much later. At the Karlsruhe cyclotron a Ge(Li) detector was used for several elements [15], while at the Oak Ridge electron linear accelerator ORELA initially a NaI detector was employed [16] which was replaced with a germanium detector in later work [17]. A new impulse to this line of experimentation was due to the installation of the GEANIE high purity germanium array at the WNR spallation time-of-flight facility of Los Alamos [18]. This new facility gave easy access to gamma-rays from (n,xn) reactions tackling important targets such as ^{239}Pu [19] and ^{238}U [20]. The installation of GEANIE was inspired by the work of Vonach et al. who first demonstrated the potential of ($n, xn\gamma$)-measurements at WNR [21].

Early inelastic scattering studies drew inspiration from the Wolfenstein-Hauser-Feshbach model allowing a qualitative rather than a quantitative agreement with experimental results [22]. Detailed angular distribution measurements could be described by an extension of the WHF model allowing the derivation of transition multipolarities and the inference of level spins and sometimes parities [23]. Despite significant advances in modeling, predicting cross sections still poses a major challenge to nuclear theory at the level of the required accuracy. In particular, accurate criticality and reactivity estimates of advanced fast reactors and the power distribution in PWRs warrant low uncertainties (2 – 8%) for inelastic scattering cross sections of the most important isotopes (^{23}Na , ^{56}Fe and ^{238}U). Depending on the concepts considered the list may be extended to include Mg, Si, Cr, Ni, Zr, Mo and Th.

To meet these challenges accurate experiments must be complemented by state-of-the-art nuclear model calculations to take optimum benefit of the data and provide the required quantities. What may be achieved was recently demonstrated for the $^{241}\text{Am}(n,2n)^{240}\text{Am}$ reaction where consistent phenomenological model calculations from different origin were beautifully confirmed by experiment [24, 25]. In addition, we may now expect a performance from WHF calculations using level densities [26], strength functions [27] and optical model potentials [28] from (semi-)microscopic calculations at the level of the phenomenological approach [29]. The phenomenological approach itself has recently seen considerable development [30] through new dispersive (coupled-channels) optical-models [31–34], imposing Lane-consistency on optical models [35], investigating the minimum number of coupled-channels to attain

convergence [36], WHF calculations by Monte Carlo to understand coincidence data [37], and the determination of the spin distribution of residuals populated by the pre-equilibrium process [38]. For the light nuclei impressive results are obtained using an algebraic coupled-channel approach that takes account of the Pauli principle and describes bound and scattering states [39–42]. Model calculations are facilitated through a comprehensive numerical compilation of nuclear model parameters [43].

In the present collaboration neutron-inelastic scattering is studied experimentally at the IRMM GELINA neutron time-of-flight facility by observation of the emitted gamma-rays using two arrays based on high purity germanium detectors. GAINS, an array consisting of 12 large volume detectors, is used to study inelastic scattering from C to Bi with high incident neutron energy resolution [29, 44–48]. GRAPHEME, developed by IPHC and using four planar detectors, is tailored for the actinides and was also used for lead [49–51]. Recent and ongoing experimental work that concerns ^{23}Na , ^{76}Ge , W and ^{232}Th is presented. Interesting complementary results for carbon are shown as well.

2 ^{23}Na

Inelastic scattering data for sodium are important for the estimation of the void coefficient in advanced fast reactors, in particular when multiple recycling of high level radioactive waste is emphasized [1, 52]. For a sodium cooled fast reactor configured as a transuranic burner, the target uncertainty between 0.5 and 1.35 MeV is 4% on the energy average and 9% between 1.35 and 2.2 MeV. For other concepts such as the European Fast Reactor or the Advanced Breeder Test Reactor the requirement is less stringent (8–10%), but the currently achieved uncertainties are much worse (15–25%).

In a recent publication we describe measurements performed with the GAINS array at the neutron time-of-flight facility GELINA [44] that meet the target uncertainty for the inelastic scattering cross-sections averaged over the above-mentioned energy ranges. An uncertainty of less than 2.5% was claimed. The experiment was performed at the 200 m flight station where an energy dependent resolution is obtained (being about 1 keV at 1 MeV) that is largely determined by a fixed time-of-flight uncertainty of about 10 ns. Eight 8 cm diameter 8 cm long high purity germanium detectors were used which are placed 4 by 4 at angles of about 110° and 150° degrees for optimal integration over non-trivial gamma-ray angular distributions (Fig. 1). For the case of sodium the ratio of the 150° yield over the 110° yield was one within the uncertainties for the transitions (Fig. 1) for which cross sections were determined. Thus no significant deviation from isotropy was found.

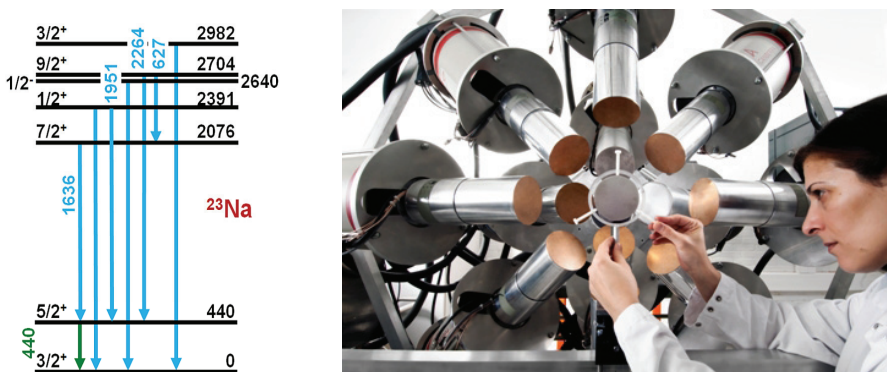


Fig. 1: Left: Partial level scheme of ^{23}Na showing the transitions measured in this work. Right: The current configuration of GAINS has twelve detectors at 110° , 125° and 150° degrees.

The gamma-ray efficiency determinations are done by Monte Carlo simulations with detector models optimized by calibrations with well characterised sources [53]. The normalization to neutron flux is

obtained by a measurement with a ^{235}U fission ionization chamber that is placed less than 2 m upstream from the sample [54, 55]. The Na sample was a high purity metallic sample encased in an Al container. The sample diameter was about 80 mm diameter and 4.2 mm thick with an areal density of $0.389(1)$ g/cm 2 . Further details may be found in reference [44].

In Rouki et al. also a complete overview of the results is given. These results show differences with the ENDF/B-VI.1 and JEFF-3.1 evaluations for energies above 1 MeV. Improvements are presently being sought. Since detailed nuclear modeling of $n+^{23}\text{Na}$ reactions is out of scope of WHF calculations due to the resonance structure and since the algebraic model mentioned above is currently only applied to still lighter nuclei, the best that may be done is a description of the cross section using an R-matrix parametrization. Such a parametrization is being undertaken and will still require a number of modifications to come from the present status, which corresponds with JEFF-3.1(.1), to an agreement with the newly measured data (Fig. 2) at the higher energies.

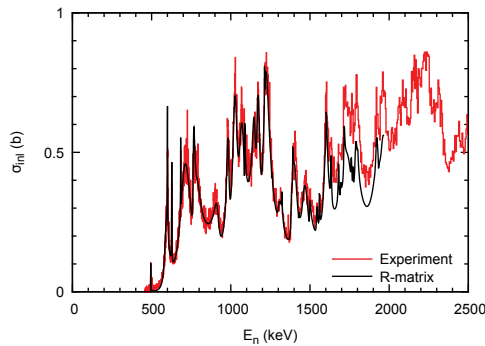


Fig. 2: Results obtained for sodium compared with a new R-matrix fit.

The $(n, xn\gamma)$ -technique does not allow to extract angular distributions of the scattered neutrons. As a prestudy for new work and to facilitate new evaluations of earlier work the Mårten et al. [56] data and their R-matrix analysis by Kopecky et al. [57] were re-analysed [58]. The R-matrix results are available for future evaluations. These concern the total cross section measured at ORNL and the inelastic cross section obtained by Mårten et al. The elastic scattering data from that work are also of interest since they offer valuable experience with obtaining angle-dependent data. Figure 3 shows the result of a numerical integration of the differential cross section data for elastic scattering. Added to the inelastic scattering cross section these should yield the total cross section. It is shown that two methods of integration of the experimental data have negligible differences but the differences with the total cross section are substantial and energy dependent (Fig. 3).

Since the R-matrix fit provides a fairly good description of the total and the inelastic data, it is no surprise that the R-matrix estimates for elastic scattering and for the mean-cosine of the scattering angle differ substantially from the experimental data (Fig. 3). The original data of the experiment are no longer available and important aspects such as multiple scattering corrections cannot be undone and redone. It is therefore of utmost interest to reinvestigate these angular distributions by new measurements. Theoretical guidance for this still rather light nucleus with significant resonance structure in the range of interest would also be of high value.

3 ^{76}Ge

With a Q-value of 2039.0 keV the nucleus ^{76}Ge is one of a small set of nuclides that may exhibit (neutrinoless) double-beta decay. In the case of regular double-beta decay two neutrinos will be emitted and

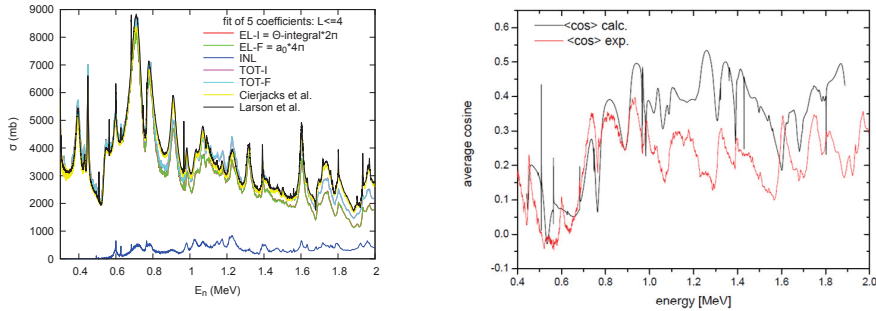


Fig. 3: Na data of reference [58]. Left: Elastic differential cross section data obtained were numerically integrated in the center of mass system (EL-I) or fitted with a 4th order Legendre polynomial to obtain the integral (EL-F). Adding the experimental data for inelastic scattering (INL) results in two estimates for the total cross section (TOT-I and TOT-F). These are compared with data for the total cross section of Cierjacks et al., and Larson et al. Since the EL-I and EL-F, resp., the TOT-I and TOT-F curves are nearly identical, the “-I” are hidden behind the “-F” curves. Right: Mean cosine of the scattering angle in the center of mass system.

the sum of the energies of the two electrons will be a characteristic continuous distribution limited above by the Q-value. Neutrinoless double-beta decay goes beyond the standard model being possible only if the neutrino is its own antiparticle. The important characteristic is that the sum of the energies of the electrons is exactly the Q-value. The current lower limit on the process half life is $1.6 \cdot 10^{25}$ y [59]. The GERDA experiment [3] attempts to establish this mode of decay by employing a number of high purity germanium detectors 86% enriched in ^{76}Ge , following up on an early claim of observation of this decay mode [60, 61]. The detectors are suspended in an Ar cryostat for cooling and more importantly for shielding against background. The cryostat has 2 m radius and is further shielded by 3 m of water. The primary concern for the shielding are gamma-rays from the rock and concrete, next come the neutrons (same source) and finally the cosmic rays. The latter are vetoed using the water shield as Cerenkov counter. The experiment aims at a background at 2039 keV of less than 10^{-3} counts per year, per keV and per kg of germanium.

A possible background is through the excitation of a level at 3951.89 keV by neutron inelastic scattering. This level emits a 2040.7 keV gamma-ray with a probability of 3.6(9)% per decay. The energy of this gamma-ray is sufficiently close to the Q-value to be of concern and thus it was decided to study the cross section for the production of this level by neutron inelastic scattering with GAINS at GELINA. In the experiment the 2040.7 keV gamma-ray was not observed. Also the transitions with energy (emission probability) 3951.7 (46(4)%) and 3388.8 (31(2)%) keV were not observed. The inferred upper limit for the cross section of producing a 2040.7 keV gamma-ray by neutron inelastic scattering is 3 mb. Using the neutron-fluxes ($3 \cdot 10^{-7}$ n/cm²/s [62, 63]) determined at LNGS where GERDA is located for unshielded detectors this implies an upper limit of 6-8 10^{-2} kg⁻¹y⁻¹ emissions of 2040.7 keV gamma-rays. The GERDA shielding easily reduces this to rates that are insignificant compared to the present goal for the background. TALYS model calculations show that the cross section could actually be much smaller ($<0.5\mu\text{b}$) allowing an unshielded detector at LNGS. For the present generation of experiments this does not require further investigation, however future experiments may have considerably more stringent requirements.

Using the samples shown in figure 4 cross sections could be measured for four gamma-rays (of energy 562.9, 545.5, 431.0 and 1348.1 keV). Two of these are shown in comparison with TALYS model calculations in figure 5. The typical uncertainty of the measurement is about 10% and is primarily due to the irregular sample shape.

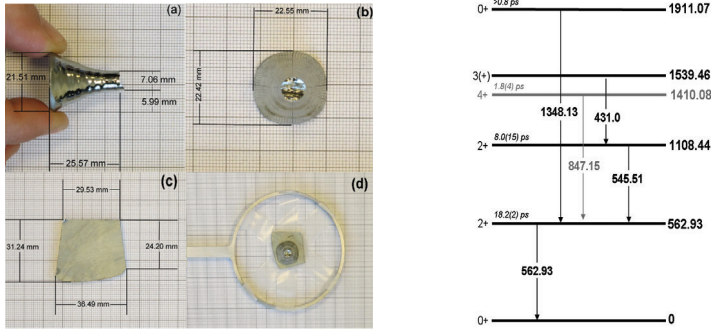


Fig. 4: Left: Samples used for the experiment on ^{76}Ge . Right: Portion of the level scheme of ^{76}Ge showing the gamma-rays observed in this work in black. The decay of the 4^+ level at 1410.08 keV was not observed.

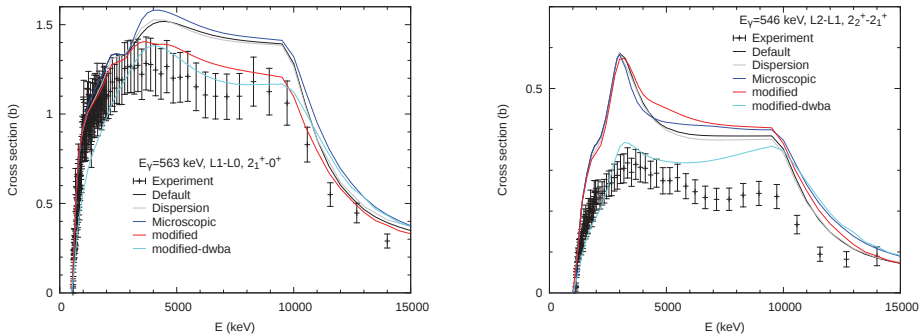


Fig. 5: Two gamma-ray production cross sections of ^{76}Ge .

The TALYS model calculations use various options available in the code. The so-called default calculation is a fully phenomenological calculation with parameters obtained earlier [64]. This is also the basis of the calculations labeled "Dispersion", "modified" and "modified-dwba". The "Dispersion" calculation uses the optical model potential of [64] adding the dispersive correction to the real potential. No significant differences are found. The modified calculation adjusts the optical model potential for better agreement with the data above 3 MeV incident neutron energy for the 563 keV gamma. The modified-DWBA calculation uses in addition a DWBA rather than a coupled-channels calculation to account for the vibrational character of the first excited states. This results in better agreement with the data for the 546 keV gamma in particular. The microscopic calculation uses the optical model of Bauge et al. [35], the level densities of Hilaire et al. [26] and the gamma-ray strength functions of Goriely et al. [27]. The result using ingredients from microscopic calculations is comparable in quality to that of the phenomenological calculation. It is however clear that model improvements are of interest in order to come to an overall satisfactory description of the experimental data.

4 W and ^{232}Th

Measurements with the GRAPHME array of IPHC Strasbourg and installed at the GELINA time-of-flight facility in Geel at a 30 m flight path currently address the actinides. Recent work with this array for ^{235}U and ^{238}U is summarised in a separate contribution to this conference [49]. There too details are presented of this setup, which presently consists of four planar germanium detectors placed 2 by 2

at 110 and 150 degrees. A particular focus of work at this experimental setup is the Th/U fuel cycle. Data for $(n, xn\gamma)$ -cross section were obtained for ^{232}Th and measurements for ^{233}U are being planned. In view of the difficulties of detecting low-energy gamma-rays which for actinides is compounded by natural radioactivity and gamma-rays due to the fission process, measurements were also made of natural and enriched tungsten samples. Since data for tungsten are simpler to obtain such data also allow to better study the experimental and analysis methods. Furthermore, the physics of tungsten nuclei is similar to that of the actinides in the sense that these are well-deformed rotational nuclides emitting low-energy intra-band gamma-rays and higher energy inter-band gamma-rays. Thus, comparisons with model calculations cover a wider mass range allowing a broader impact of the data. Some preliminary results are shown in figure 6 in comparison with model calculations with the TALYS code. The data analysis is still ongoing.

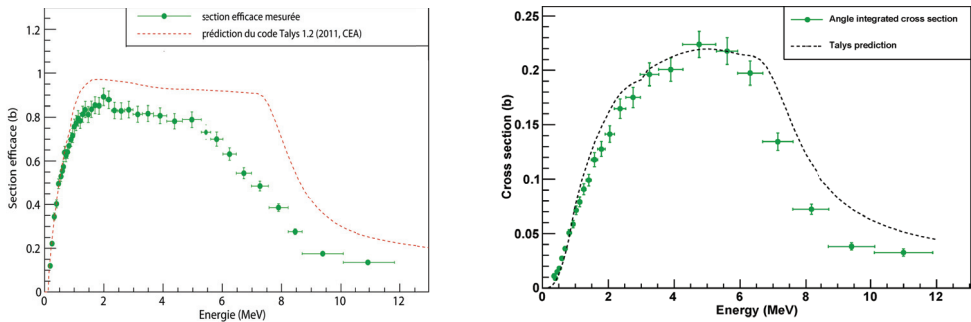


Fig. 6: Experimental inelastic scattering cross sections for the emission of the 122.64 keV gamma-ray of the $2_1^+ \rightarrow \text{gs}$ -transition in ^{186}W (Left) and the 112.75 keV gamma-ray of the $4_1^+ \rightarrow 2_1^+$ -transition in ^{232}Th (Right).

5 ^{12}C

Inelastic neutron scattering on ^{12}C can be studied in a way quite different from the $(n, xn\gamma)$ -technique and the neutron time-of-flight methods mentioned above. In recent work [65, 66] single crystal diamond detectors were exposed to quasi mono-energetic neutron fields at the IRMM van de Graaff laboratory. These detectors register the energy deposited by the charged particles left in the crystal following excitation of the carbon atoms by neutron inelastic scattering. The resulting pulse height spectrum in these very pure carbon detectors has better than 50 keV energy resolution and is determined by the Q-value of the reaction plus the incident neutron energy minus the sum of the emitted neutron and gamma energies. Gamma-emission is the dominant decay mode for the first level (2_1^+ , $E_x = 4438.91$ keV) but is negligible for the higher lying levels. These decay into $\alpha + ^8\text{Be}$ or 3α s. In view of the range of energies assumed by the outgoing neutron a range of energies in the pulse height spectrum is contributed by each of the excited levels in ^{12}C . In addition one observes in the detector full-energy peaks that are associated with the dissociation of the compound nucleus ^{13}C into charged particles only. In particular one observes the following binary exit channels: $\alpha + ^9\text{Be}$, $p + ^{12}\text{B}$, or $d + ^{11}\text{B}$. For these channels cross sections are readily obtained. A first attempt at modeling was undertaken by inspecting the data available in the ENDF/B-VII neutron library using MCNP. Using this Monte Carlo simulation code with a specially developed tally-ing subroutine it is possible to check the energy deposited by looking at the difference in energy of the incident neutron and the outgoing neutron(+gamma) [67]. The comparison of data and simulation is shown in figure 7. Here the data are taken for 16.6 MeV neutrons with a standard spread of 0.2 MeV.

At the highest deposited energy the $^{12}\text{C}(n, \alpha)^9\text{Be}$ contribution is evident. From 4.5 to 9.5 MeV deposited energy the response is dominated by 3α breakup continuum. For deposited energies less than 4.5 MeV the response is dominated by elastic scattering for which this is the maximum deposited

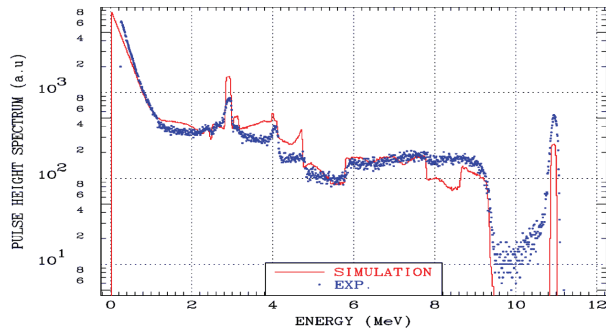


Fig. 7: Comparison of experimental data and Monte Carlo simulations based on ENDF/B-VII cross sections for $n+^{12}\text{C}$. The data correspond to the pulse height spectrum observed with a $4.7 \times 4.7 \times 0.5 \text{ mm}^3$ single crystal diamond detector obtained from Diamond Detectors Ltd.

energy [68]. The discrete peaks on top of the (in-)elastic scattering distribution correspond with the $^{12}\text{C}(n,p)^{12}\text{B}$ and $^{12}\text{C}(n,d)^{11}\text{B}$ reactions.

The figure clearly shows that some of the features in the spectrum are adequately described while others are not. In particular it appears that the description of inelastic scattering at large energy deposition and elastic scattering near the maximum recoil energy could be improved. Hence these data appear to offer an interesting test ground for the algebraic approach to coupled channel calculations for carbon described in references [41,42]. As is evident from our publication pulse height spectra and cross sections were obtained in the energy range from 7.3 to 20.5 MeV and the numerical data are available on request.

A good description of these data is of interest to applications aiming at neutron fluence and neutron spectrum measurements in various radiation fields in fission and fusion energy and in accelerator based neutron fields.

6 Summary

An overview is presented of recent measurements with the $(n, xn\gamma)$ -technique with the GAINS and GRAPHENE setups. Cross sections were shown for ^{23}Na , ^{76}Ge , ^{186}W , ^{232}Th and in an accompanying contribution to this conference: $^{235,238}\text{U}$. The data are compared with calculations in the interest of improving nuclear models and making the most of the data in the interest of applications. For applications in nuclear energy such data are in high demand and there remains considerable room for improved measurements and improved model calculations. Also shown are neutron inelastic scattering and reaction data obtained with a single-crystal diamond detector. These should be of interest to $n+^{12}\text{C}$ model calculations that were recently performed. Describing these data better is of interest for the use of these detectors in complex neutron fields and involves the excitation spectrum of ^{12}C and ^{13}C and the angular distribution of emitted neutrons.

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