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Isomeric Cross Section Study of Neutron–Induced Reactions on Ge Isotopes

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Abstract In the present work, natural Ge targets have been irradiated by neutron beams of 17.7 and 19.3 MeV, at the 5 MV Tandem Accelerator of NCSR “Demokritos”. The cross section of the $^{72,74}\text{Ge}(n,\alpha)^{69,71}\text{Zn}$ and $^{76}\text{Ge}(n,2n)^{75}\text{Ge}$ reactions, leading to the population of isomeric states, has been deduced with respect to the $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ and $^{93}\text{Nb}(n,2n)^{92m}\text{Nb}$ reference reactions. After the neutron irradiations, the induced γ -ray activity of the Ge target and reference foils, was measured with high-resolution HPGe detectors.

Keywords cross section, Monte Carlo simulations, neutron activation technique, Ge

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INTRODUCTION

Studies of neutron–induced reactions on medium mass nuclei are of considerable significance, both for their importance to fundamental research in nuclear physics and astrophysics and for practical applications in nuclear technology, medicine and industry [1–3]. Natural Ge consists of five isotopes and from all the neutron induced reactions, six of them produce long lived residual nuclei and can thus be studied by using the activation technique, while three of them produce the residual nucleus in an isomeric state. Isomeric cross sections are of special interest for the study of the formation and deexcitation of the compound nucleus, via its decay to isomeric and normal states, which is governed by the spins of the discrete levels involved as well as the spin distribution of the continuum phase space [3–5].

A survey on the available experimental neutron cross sections on Ge isotopes, revealed a plethora of datasets at energies around 14 MeV and very few ones at lower and higher energies, with many discrepancies among them.

In view of these remarks, the neutron facility at the 5MV tandem accelerator of NCSR “Demokritos”, has been used for the measurement of neutron induced reaction cross sections on natural Ge at 17.7 and 19.3 MeV neutron energies, implementing the activation technique.

EXPERIMENTAL METHOD

Quasi–monoenergetic neutron beams were produced via the $^3\text{H}(d,n)^4\text{He}$ reaction using a Ti tritiated target, of 373GBq activity, on a Cu backing, at a flux of the order of $\sim 10^5$ n/(cm² sec). The absolute flux of the beam was deduced with respect to the $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ and $^{93}\text{Nb}(n,2n)^{92m}\text{Nb}$ reference reactions, while its variation was monitored by a BF₃ detector placed at a distance of 3 m from the neutron source. The spectra of the BF₃ monitor were stored at regular time intervals of ~ 200 sec in a separate ADC, during the irradiation process.

Samples of high purity natural Ge pellets of 1.3 cm diameter and ~1.3 g weight, doped with 6% high purity cellulose, were placed between Al and Nb reference foils and exposed to the neutron beam for about 30 h continuous irradiation. A Au foil was also used to cross check the experimental neutron flux, as well as the simulated one, via the $^{197}\text{Au}(n,\gamma)$ reaction. The induced activity of product radionuclides in both target and reference foils was measured with HPGe detectors of 80% and 16% efficiency, properly shielded with lead blocks to reduce the contribution of the natural radioactivity. The efficiency of the detectors at the position of the activity measurements (10 cm) was determined via calibrated ^{152}Eu , ^{133}Ba , ^{57}Co , ^{60}Co , ^{109}Cd , ^{54}Mn , ^{137}Cs and ^{214}Bi point sources. The characteristic γ -rays were corrected for self-absorption in the target, summing effects of cascading transitions and counting geometry. A characteristic spectrum with all the important gamma-rays of the various reactions from the irradiated Ge target are shown in Fig. 1.

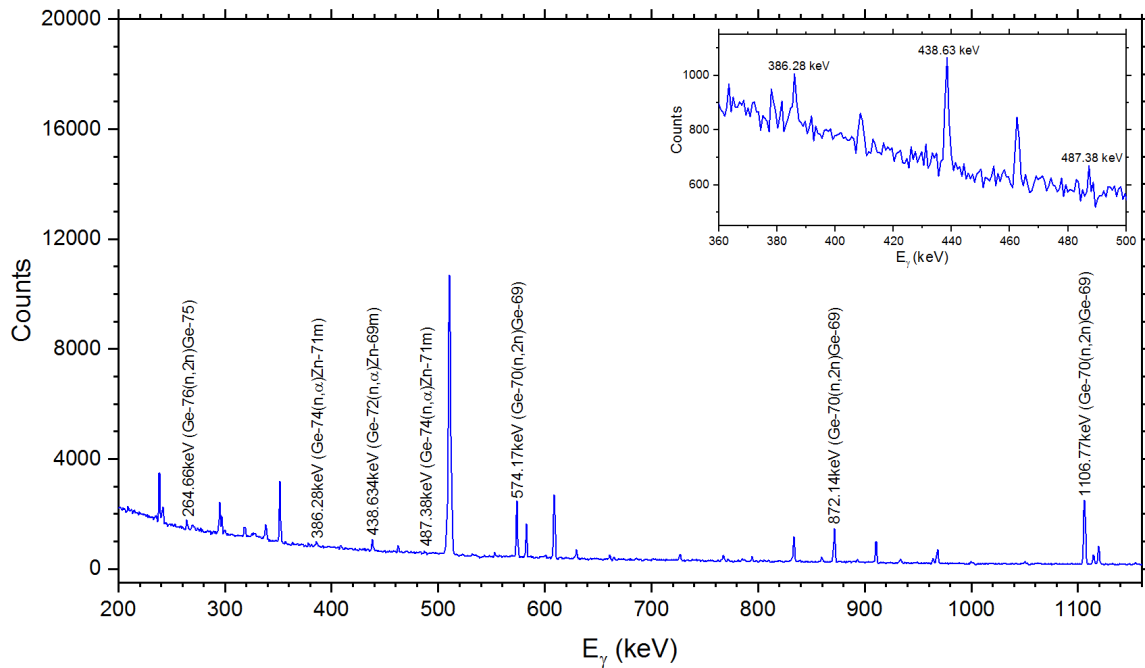


Figure 1. Off-line γ -ray energy spectrum observed after the neutron irradiation of natural Ge target at 19.3 MeV.

Of all isotopes contained in natural germanium ($^{70,72,73,74,76}\text{Ge}$), only the $^{72,73}\text{Ge}(n,p)^{72,73}\text{Ga}$, $^{72,74}\text{Ge}(n,\alpha)^{69,71}\text{Zn}$ and $^{70,76}\text{Ge}(n,2n)^{69,75}\text{Ge}$ reactions, can in principle be studied using the activation technique. From these six reactions the $^{72,74}\text{Ge}(n,\alpha)^{69,71}\text{Zn}$ and $^{76}\text{Ge}(n,2n)^{75}\text{Ge}$ ones are of special interest as the residual nuclei are produced in a high spin isomeric state.

In more detail, ^{69}Zn is produced both in its $1/2^-$ ground state with a 56.4 min half life and its metastable $9/2^+$ state with a half life of 13.76 h. The ground state of ^{69}Zn decays directly to the ground state of ^{69}Ga , while the metastable state decays to the ground state of ^{69}Zn , emitting the characteristic 438.6 keV gamma ray, with 94.85 % intensity, which can be used for the determination of the σ_m cross section. ^{71}Zn is produced both in its $1/2^-$ ground state with a half life of 2.45 min and its metastable state $9/2^+$ with a half life of 3.96 h. Due to the short lifetime of the ground state of ^{71}Zn , only the activity of the metastable state could be measured via the activation technique. Thus, the σ_m could be determined by analyzing the 386.3 keV (with intensity 91.4%) transition of ^{71}Ga which is fed by the de-excitation of the metastable state of ^{71}Zn .

As for the $^{76}\text{Ge}(n,2n)^{75}\text{Ge}$ reaction, it leads to the formation of the unstable nucleus ^{75}Ge both in its $1/2^-$ ground state with half life 82.78 min and its metastable $7/2^+$ state with half life 47.7 s. The de-excitation of the metastable to the ground state in fraction 99.97%, along with its short lifetime leads

to the measurement of the total cross section σ_{m+g} , corresponding to the production of both ground and metastable states of the $(n,2n)$ reaction, via the 264.6 keV (with intensity 11.4%) characteristic transition of ^{75}As . All these γ -rays are indicated in the spectrum presented in Fig. 1.

RESULTS AND DISCUSSION

The results for the cross section for all the three above mentioned reactions, deduced from their characteristic gamma-rays, are presented in Figs. 2, 3 and 4, along with data from literature. A more detailed description of the experimental procedure for the data analysis and deduction of cross section is given in Ref. [6].

Due to the fact that the germanium target was of natural isotopic composition, some of the activation products may be formed via several interfering reactions. The $^{76}\text{Ge}(n,2n)^{75}\text{Ge}$ threshold reaction cross section is contaminated by the $^{74}\text{Ge}(n,\gamma)^{75}\text{Ge}$ one, which is activated by low energy parasitic neutrons. The parasitic neutrons may come from deuteron break-up reactions, from the TiT target itself, from reactions with materials of the beam-line and from scattering in the materials of the experimental room. In order to estimate this neutron background contribution to the activation of ^{75}Ge and make the appropriate corrections, detailed simulations of the irradiation setup by means of the NeuSDesc [7] and MCNP5 codes [8,9] have been performed. These simulations were executed for a number of 10^9 simulated particles for sufficient statistics, the geometry of the experimental area was described in great detail and a very low neutron energy cut-off (10^{-11} MeV) was implemented. The resulted neutron energy distribution $\Phi(E)$ was normalized in order to comply with the experiment and to agree with the data obtained by the analysis of the $^{197}\text{Au}(n,2n)^{196}\text{Au}$ reaction ($E_{\text{threshold}} \sim 8$ MeV) for the main neutron energy peak. It was then validated by means of the $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$ reaction, which is a non-threshold reaction ($E_{\text{threshold}} = 0$ MeV) and therefore can be readily activated by the low energy tail of parasitic neutrons. The simulated and experimental results were in good agreement, thus verifying the reliability of the simulations. The reaction rate $R.R$ for the $^{74}\text{Ge}(n,\gamma)^{75}\text{Ge}$ reaction was then calculated from the following expression :

$$R.R = \sum_{\Delta E} \sigma(E) \cdot \Phi(E)$$

using the simulated neutron fluence $\Phi(E)$ and the excitation function $\sigma(E)$ of the $^{74}\text{Ge}(n,\gamma)^{75}\text{Ge}$ reaction, taken from the ENDF/B-VII.1 library. The expected number of counts were then calculated and subtracted from the experimental number of counts for the 264.6 keV characteristic γ -ray of the ^{75}Ge decay. A detailed description of this methodology can be found in Ref. [10]. The correction of the $^{76}\text{Ge}(n,2n)^{75}\text{Ge}$ reaction cross section due to the interference of the $^{74}\text{Ge}(n,\gamma)^{75}\text{Ge}$ one, was estimated to be 0.8% at 17.7 and 19% at 19.3 MeV. The resulting cross section values are shown in Fig. 2 as solid circles before the corrections and as solid squares after the corrections, while the solid triangles are older measurements by our group. They are seen to be in fair agreement with the only data existing in literature from Ref. [11].

Furthermore, the $^{72}\text{Ge}(n,\alpha)^{69m}\text{Zn}$ reaction was contaminated by the $^{73}\text{Ge}(n,\alpha)^{69m}\text{Zn}$ one. The appropriate corrections were performed by using the cross section of the contaminant reaction from EAF2010 [12] library and were found to be 27% and 35% at 17.7 and 19.3 MeV, respectively. The final cross section values are shown in Fig. 3 as solid circles before the corrections and as solid squares after the corrections, while the solid triangles are older measurements by our group. Finally, the data from the $^{74}\text{Ge}(n,\alpha)^{71m}\text{Zn}$ reaction are shown in Fig. 4 as solid squares.

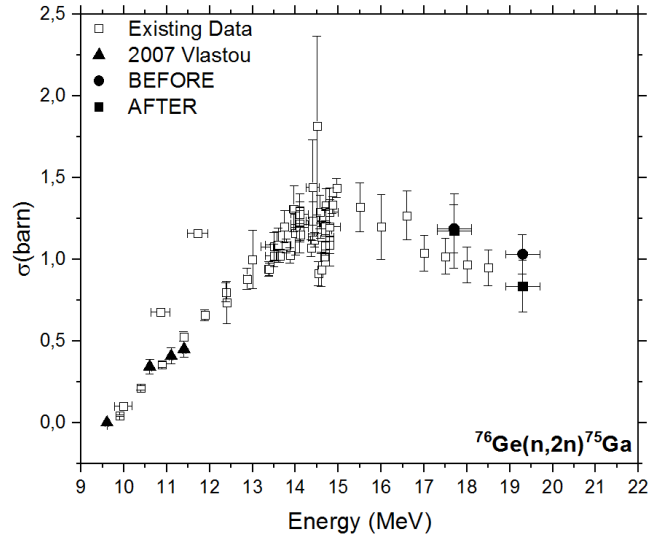


Figure 2. Experimental values of the $^{76}\text{Ge}(n,2n)^{75}\text{Ga}$ reaction at 17.7 and 19.3 MeV, along with EXFOR data from literature. The solid circles correspond to the cross section values before the corrections due to the interfering $^{74}\text{Ge}(n,\gamma)^{75}\text{Ge}$ reaction and the solid squares after the corrections.

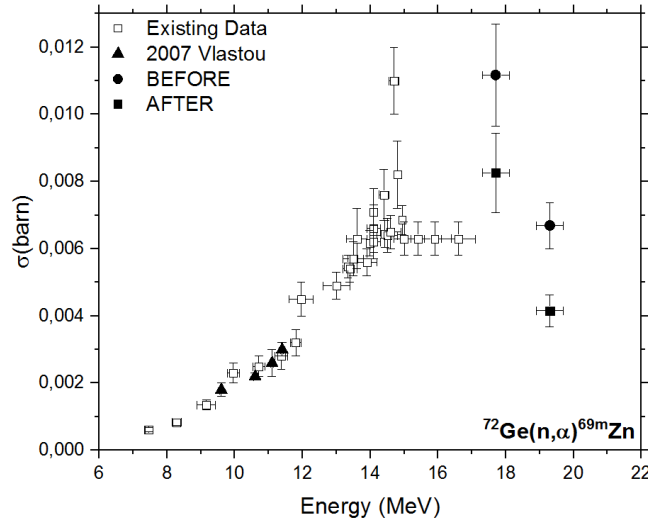


Figure 3. Experimental values of the $^{72}\text{Ge}(n,\alpha)^{69m}\text{Zn}$ reaction at 17.7 and 19.3 MeV, along with EXFOR data from literature. The solid circles correspond to the cross section values before the corrections due to the interfering $^{73}\text{Ge}(n,\alpha)^{69m}\text{Zn}$ reaction and the solid squares after the corrections.

Due to the low cross section of the (n,α) reactions, the statistics of the corresponding gamma-rays was poor (see the inset of Fig. 1), leading to large uncertainties in the cross section values shown in Figs. 3 and 4. Measurements with much higher neutron beam flux or with mono-isotopic targets are required in order to improve the experimental accuracy of the (n,α) cross section values.

FUTURE PERSPECTIVES

Additional measurements are planned in the near future mainly at energies between 15 and 17 MeV to cover this important plateau region where the cross sections reach their maximum values and to provide more experimental information which will help to resolve discrepancies among the existing experimental data. In addition, theoretical calculations based on the compound nucleus theory of Hauser–Feshbach, will be performed in the energy range $\sim 5\text{--}30$ MeV, using the codes “EMPIRE” (3.2.2 version) [13] and TALYS [14], for all reactions under study. The effect of different optical

model potentials and level density models will be investigated. The level densities described either by the Enhanced Generalized Superfluid Model (EGSM) [15] or by the RIPL–3 microscopic Hartree–Fock–Bogoliubov method [16] will be tried and the investigation will be further focused on the sensitivity of the calculations to variations of the level density parameter $\tilde{\alpha}$, which is very sensitive to the production of high spin isomeric states. Furthermore, the pre–equilibrium emission mechanism will be studied, implementing the classical exciton model [17] via the PCROSS module [13].

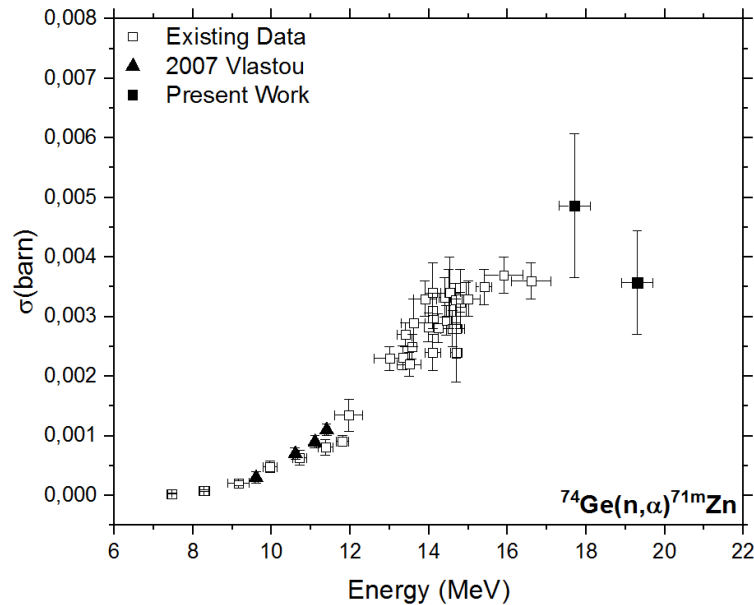


Figure 4. Experimental values of the $^{74}\text{Ge}(n,\alpha)^{71m}\text{Zn}$ reaction at 17.7 and 19.3 MeV, along with EXFOR data from literature.

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