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Phys. Rev. C **95**, 054605 — Published 4 May 2017

DOI: 10.1103/PhysRevC.95.054605

Total and isomeric-state cross sections for the ^{76}Ge(n,2n)^{75}Ge reaction from threshold to 14.8 MeV

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(Dated: April 4, 2017)

The cross section for the reaction ^{76}Ge(n,2n)^{75}Ge to both the isomeric and ground state of ^{75}Ge has been measured with the activation method between 10 and 15 MeV in small energy steps to help resolve inconsistencies in the existing database. The ^{197}Au(n,2n)^{196}Au reaction with its known cross section was used for normalization of the data, which are compared to experimental and evaluated data of the EXFOR, EAF, JENDL, ENDF, and TENDL libraries. Model calculations using the TALYS-1.8 code are presented which also allow for the extrapolation to higher neutron energies. The data are important to estimate potential neutron-induced backgrounds in currently running large-scale experiments aimed at the discovery of neutrinoless double-beta decay of ^{76}Ge.

PACS numbers: 25.40.Fq,29.30.Kv,24.60.Dr

I. INTRODUCTION

The **GER**manium **D**etector **A**rray (GERDA) and MAJORANA collaborations use Germanium-diode detectors in their searches for neutrinoless double-beta decay ($0\nu\beta\beta$) of ^{76}Ge [1, 2]. These highly enriched detectors (86% ^{76}Ge, 14% ^{74}Ge) serve as both source and detector. Common to all $0\nu\beta\beta$ decay searches is the requirement that background events in the energy region of interest, a narrow energy band centered at the Q-value for $0\nu\beta\beta$ decay, must be extremely small. To help achieving this goal, the experiments are being performed deep underground, resulting in a substantial reduction of cosmogenic background. However, some of the remaining muons may interact with nuclei in the vicinity of the detector or within the detector itself, producing so-called spallation neutrons. During the slowing-down process of these neutrons, (n,xn) reactions with $n \geq 2$ play an important role. These reactions tend to multiply the incident spallation neutron yield by typically one order of magnitude for a 100 MeV spallation neutron.

The ^{76}Ge(n,2n)^{75}Ge reaction is of special importance because its (n,2n) cross section is of hundreds of mb. A partial level scheme of ^{75}Ge and its β-decay daughter ^{75}As is shown in Fig. 1. The ^{76}Ge(n,2n)^{75}Ge reaction populates the isomeric state $7/2^+$ state of ^{75}Ge at 139.7 keV, which either decays via an isomeric transition IT to the $1/2^-$ ground state with $T_{1/2}$=47.7 seconds and a branching ratio of 99.97%, or via β decay to ^{75}As with a branching ratio of 0.03%. The ground state of ^{75}Ge in turn β decays with $T_{1/2}$ = 82.78 minutes to ^{75}As.

In this work, we report on the activation cross sections for the reactions ^{76}Ge(n,2n)^{75m}Ge (isomeric state), ^{76}Ge(n,2n)^{75}Ge (total) and ^{76}Ge(n,2n)^{75g}Ge (ground state) at eleven neutron energies from threshold to 14.8 MeV. The associated Q-values are -9.55 MeV and -9.69 MeV, respectively. The results are compared to estimates of recent evaluated data libraries and data from the literature. In addition, we have compared the measured cross-section data of these reactions with theoretical model calculations performed with the TALYS code (version 1.8) [4] in the neutron energy range of 10 to 16 MeV. The calculations were made using different level-density options to match the cross-section data measured in the present work.

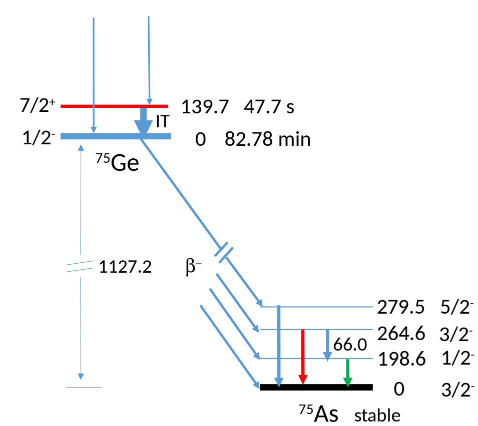

FIG. 1: (Color online). Partial level scheme relevant to the ^{76}Ge(n,2n)^{75m}Ge and ^{76}Ge(n,2n)^{75}Ge reactions. All energies are given in keV. Data taken from [3].

II. EXPERIMENT AND PROCEDURE

The cross-section measurements were performed using the neutron-activation technique. Irradiations were carried out at the 10 MV FN Tandem Van de Graaff accelerator at the Triangle Universities Nuclear Laboratory (TUNL) using two different neutron source reactions. First, the ^{2}H(d,n)^{3}He reaction (Q = 3.269 MeV) was

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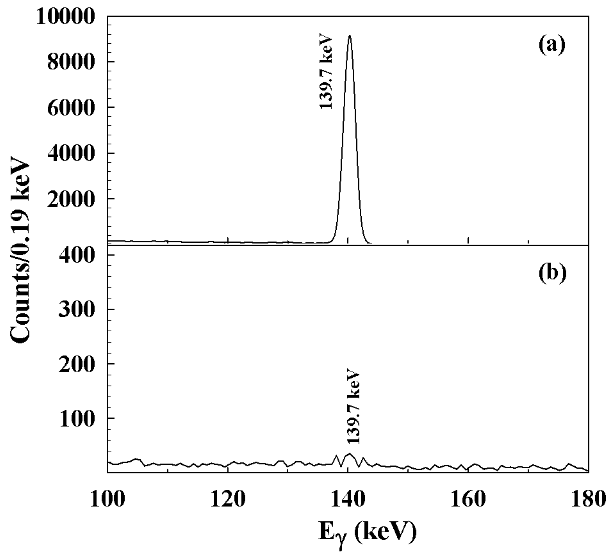

FIG. 2: (Color online). (a) ^{75}Ge γ-ray line at 139.7 keV measured for 30 seconds with a HPGe detector starting 40 seconds after the 3 minutes irradiation time of ^{76}Ge with 12.87 MeV neutrons, (b) same as above, but with a starting time of 3.5 minutes after irradiation.

used to produce quasimonoenergetic neutrons between 9.9 and 14.5 MeV employing a deuterium gas target cell. The gas pressure was adjusted to 4 atm to provide the desired neutron energy spread in the energy range investigated. Typically the neutron energy spread was ± 150 keV at 0°. Second, at 14.8 MeV, the ^{3}H(d,n)^{4}He reaction (Q = 17.59 MeV) was employed by replacing the deuterium gas cell by a tritiated target assembly. It consists of a 2 mg/cm^2 thick titanium layer loaded with 2.5 Ci of tritium and evaporated onto a 0.4 mm thick copper disk. A metallic germanium slab of 10 mm × 10 mm area and thickness of 2 mm (resulting in a mass of about 1.5 g) with the same isotopic composition as that of the enriched HPGe detectors used by the GERDA and MAJORANA collaborations was supported by a thin plastic foil and positioned 1.9 cm from the end of the deuterium gas cell at 0° relative to the direction of the incident deuteron beam (see Fig. 1 of Ref. [5]). A total of three slabs were used one at a time during the course of the measurements to optimize the efficiency of our irradiation and counting procedure. The deuteron beam current was typically 2 μA.

In order to normalize the neutron flux at the Ge slab position, high-purity Au foils of the same area and thickness of 0.025 mm were placed on the front and backside of the Ge slabs, enabling us to use the ^{197}Au(n,2n)^{196}Au reaction with $T_{1/2}$ = 6.17 days, E_γ = 355.73 keV and I_γ = 87% as monitor reaction. The neutron activation cross-section data for this reaction were obtained from Ref. [6]. The average neutron flux produced at the Ge slab position ranged from 1.4×10^7 to 4.4×10^7 n/(cm^{2}s). A 1.5 inch × 1.5 inch BC-501A based neutron detector was placed at 0° relative to the incident deuteron beam. During irradiation, the detector operated in the multichannel-scaling acquisition mode to record the time profile of the neutron flux, allowing to make off-line correction for any beam current variation.

Because of the high thresholds of the ^{76}Ge(n,2n)^{75}Ge and ^{76}Ge(n,2n)^{75m}Ge reactions, and the gap of about 5.5 MeV between the energy of the monoenergetic neutrons of the ^{2}H(d,n)^{3}He reaction and the maximum energy of the breakup continuum, the (n,2n) reactions are not sensitive to breakup neutrons in the energy range studied in the present work.

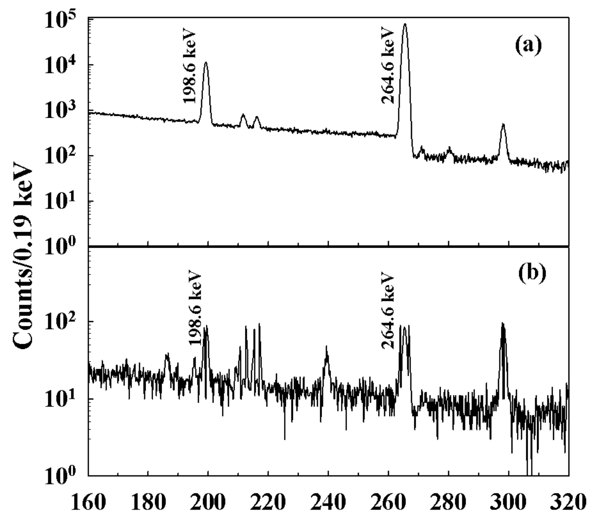

FIG. 3: (Color online). (a) ^{75}As γ-ray lines at 198.6 keV and 264.6 keV measured for 2 hours with a HPGe detector starting 0.3 hours after the 1 hour irradiation time of ^{76}Ge with 12.87 MeV neutrons, (b) same as above, but with a starting time of 10.5 hours after irradiation.

High-resolution γ-ray detection systems located at TUNL's low background counting facility were used to record γ-ray spectroscopy data off-line for the irradiated samples and monitors foils. Two 60% HPGe detectors combined with a Canberra Multiport II multichannel analyzer and a 16 K ADC, supported by the Genie 2000 data-acquistion system, were employed. These detectors were properly shielded with lead blocks in order to reduce the contribution of natural radioactivity from the environment. The sample activities were determined using the counts in the full-energy peak of the γ-ray transition. For this it was important to know the absolute photopeak efficiency and the energy calibration. For energy and efficiency calibration, a mixed source consisting of the isotopes ^{241}Am (E_γ = 59.5 keV), ^{109}Cd (E_γ = 88 keV), ^{57}Co (E_γ = 122.1 keV), ^{139}Cs (E_γ = 165.9 keV), ^{203}Hg (E_γ = 279.2 keV), ^{113}Sn (E_γ = 391.7 keV), ^{134}Cs (E_γ = 604.7 keV), ^{137}Cs (E_γ = 661.7 keV), ^{54}Mn (E_γ = 834.8 keV), ^{65}Zn (E_γ = 1115.5 keV) and, ^{88}Y (E_γ = 1836.1 keV) was used. The energy resolution was found to be ∼ 1.8 keV for 1836.2 keV γ rays emitted from ^{88}Y. The residual activity of samples and monitor foils was counted at 5 cm distance from the center of the detector window. The choice of this distance was the result of a compromise between assuring acceptable count rate and reducing coincidence summing effects. Prior to irradiation, background measurements were performed with non-irradiated Ge/Au foils to check for any interference in the pulse-height region of interest. The induced activities in the germanium samples were determined by measuring the γ rays associated with the decay of ^{75m}Ge and ^{75}Ge at 139.7 keV (39.51%) and 264.6 keV (11.4%), respectively.

Typical spectra are shown in Figs. 2(a) and 2(b) for the 139.7 keV transition recorded 40 seconds and 3.5 minutes after the end of irradiation, respectively. Figures 3(a) and 3(b) present similar spectra for the 198.6 keV and 264.6 keV transitions of interest. The peak-area analysis was done with the program TV [7]. For the activity determination, half-lives, emission probability, γ-ray attenuation, spatial difference of the γ-ray efficiency, and coincidence-summing corrections were taken into account. The decay data for both ^{75}Ge and ^{196}Au used in the analysis were taken from Ref. [3].

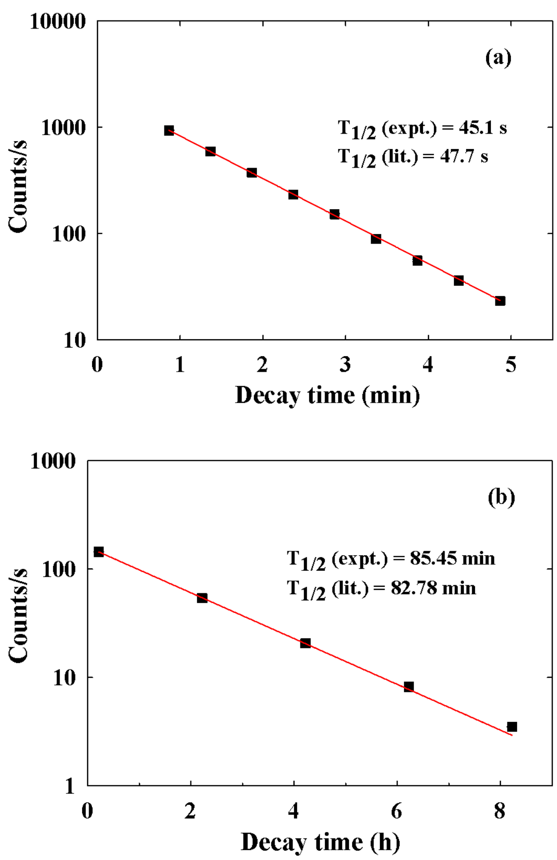

FIG. 4: (Color online). Decay curves for (a) the 139.7 keV γ-ray line of ^{75m}Ge and (b) the 264.6 keV γ- ray line of ^{75}As obtained after irradiation with E_n = 12.87 MeV neutrons.

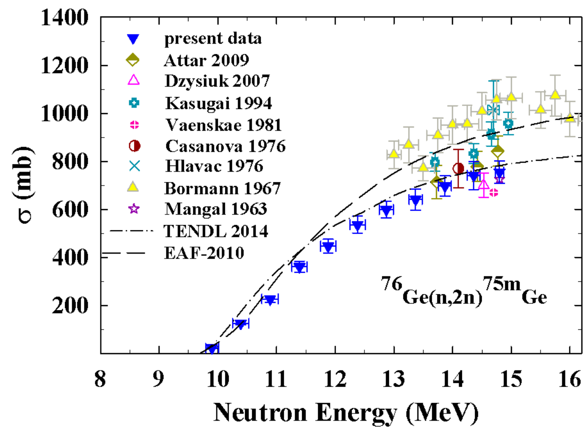

FIG. 5: (Color online) Experimental results for ^{76}Ge(n,2n)^{75m}Ge reaction compared with results from earlier measurements [14–21] and the model calculation TENDL-2014 [10] and the EAF-2010 [11] evaluation.

For the study of the ^{76}Ge(n,2n)^{75m}Ge reaction, a HPGe detector was mounted just outside of the target room to limit the time between irradiation and counting to typically 20 seconds. The sample was irradiated for three half lives and the induced γ-ray activity of the 139.7 keV transition from the ^{75m}Ge decay was measured for a period of ten half lives. Figs 4(a) and 4(b) show the intensities of both the 139.7 keV and 264.6 keV γ rays as a function of cooling time after irradiation.

The neutron fluence and cross-section values were derived using the well-known activation formula, closely following the procedure explained in Ref. [8]. The yields were corrected for dead time, γ-ray emission probability, γ-ray self-absorption including the size and shape of the samples and monitor foils, efficiency of the detector, time-dependence of the neutron flux, and source-size geometry.

III. RESULTS

The cross-section values measured in the present work along with their uncertainties are presented in Table I. The first column shows the neutron energy and its energy spread. The second column gives the ^{197}Au(n,2n)^{196}Au reaction cross-section values used to calculate the neutron flux. Columns 3 and 4 represent the cross-section results σ_m and σ_t for the reactions ^{76}Ge(n,2n)^{75m}Ge and ^{76}Ge(n,2n)^{75}Ge, respectively. To determine the (n,2n) cross section σ_g to the ground state, the relation $\sigma_g = \sigma_t$ - σ_m was used. This cross section is shown in column 5. Finally, column 6 gives the isomeric to ground state cross-section ratio σ_m/σ_g. As a by-product, the ^{74}Ge(n,α)^{71m}Zn cross section was obtained at 14.8 MeV and found to be (3.24±0.17) mb, in good agreement with the previous datum of [9]. Because of unfavorable threshold, decay-time and γ ray energy values, other neutron-induced reaction cross-section determinations on ^{74}Ge

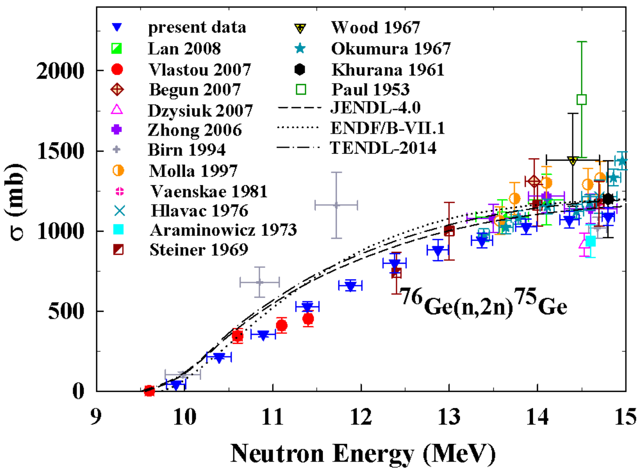

FIG. 6: (Color online) Cross-section results for the ^{76}Ge(n,2n)^{75}Ge reaction compared with results from earlier measurements [15, 17, 19, 22–33], the model calculation TENDL-2014, and the JENDL-4.0 [12] and ENDF/B-VII.1 [13] evaluations.

and ^{76}Ge were not attempted in the present work.

The sources of errors considered in the activation measurements are shown in Table II: nuclear constants (half-life, γ-ray intensities), instrumental factors (time of irradiation, cooling and measurements), and uncertainties related to the determination of the correction factors. The uncertainties of the measured cross-section data vary from 5.2% to 8.3%. By considering the uncertainities involved in the measurement of each parameter, the total uncertainty was obtained by taking the square root of the sum of the squares of the individual uncertainties.

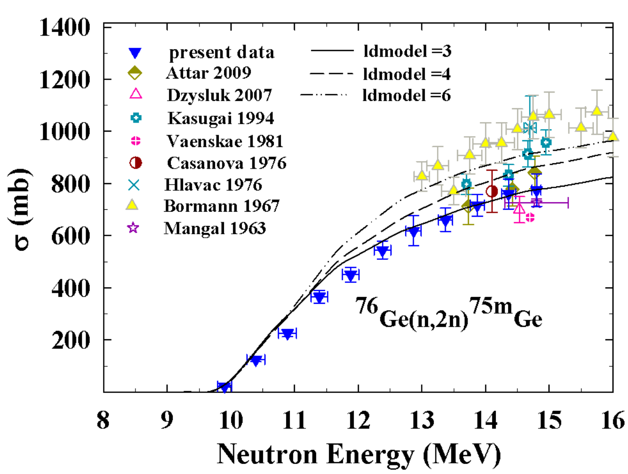

FIG. 7: (Color online) Comparison of cross-section data [14–21] and TALYS calculations for the ^{76}Ge(n,2n)^{75m}Ge reaction using different level-density choices (see text). Our data are best described by the generalized superfluid model.

The experimental data obtained in the present work (downward looking triangles) are shown in Figs. 5 and 6 along with values from the literature and results from the available comprehensive evaluations TENDL-2014 [10], EAF-2010 [11], JENDL-4.0 [12] and ENDF/B-VII.1 [13] databases. The results shown in Fig 5 reveal that our results for the ^{76}Ge(n,2n)^{75m}Ge reaction above 13 MeV favor the lower cluster of the previous experimental data [14, 15, 17, 18, 21], while the data of Bormann *et al.* [20], Kasugai *et al.* [16], and the datum of Hlavac *et al.* [19] provide larger cross-section values. Our data below 13 MeV are the first data in this energy range. They give an accurate determination of the cross section in the important energy region above the (n,2n) threshold. The TENDL-2014 predictions are in much better agreement with the present data than the EAF-2010 evaluation, which favors the upper cluster of the previously available experimental data.

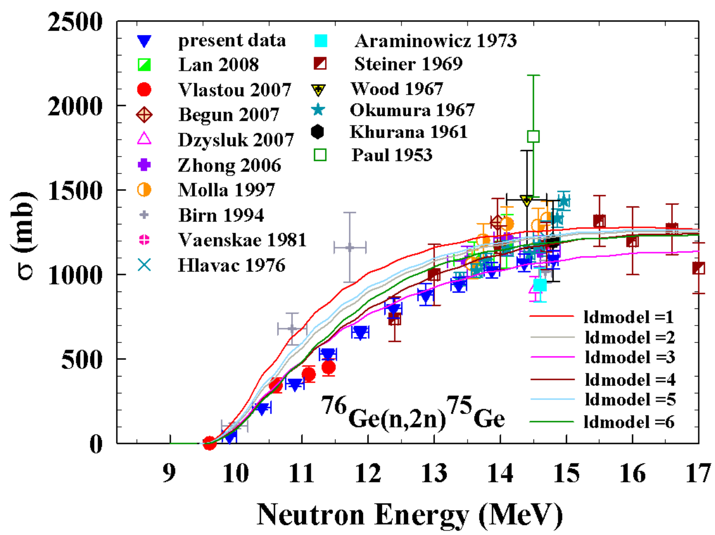

FIG. 8: (Color online) Comparison of cross-section data [15, 17, 19, 22–33] and TALYS calculations for the ^{76}Ge(n,2n)^{75}Ge reaction using different level-density choices (see text). Our data are best described by the generalized superfluid model.

Inspecting Fig. 6, we note that our data for the ^{76}Ge(n,2n)^{75}Ge reaction in the 14 MeV region are in good agreement with the lower set of the previous cross-section data. At lower energies the present data confirm the energy dependence established by the two data points of [29] near 12.5 and 13 MeV and the five data points of [22, 23] below 11.5 MeV. Because the model calculation TENDL-2014, and the evaluations JENDL-4.0, and ENDF/B-VII.1 are trying to reproduce the average of the data in the 14 MeV energy region, they clearly overestimate the ^{76}Ge(n,2n)^{75}Ge cross-section data in the 11 to 12.5 MeV energy range. Figs. 7 and 8 show the measured cross-section data in comparison with nuclear-model code TALYS calculations (for explantion of curves see Sec. IV). Our results for the deduced cross section σ_g

TABLE I: Measured cross sections and deduced isomeric yield ratio obtained in the present work at neutron energies from E_n = 9.9 to 14.8 MeV.

$E_n \pm \triangle E_n$ (MeV)	σ_{mon} (mb)	^{76}Ge(n,2n)^{75m}Ge (mb)	^{76}Ge(n,2n)^{75}Ge (mb)	^{76}Ge(n,2n)^{75g}Ge (mb)	σ_m/σ_g
9.90 ± 0.11	964.21 ± 29.50	22.14 ± 1.84	44.91 ± 2.59	22.77 ± 2.30	0.97 ± 0.13
10.39 ± 0.14	1411.41 ± 39.52	124.85 ± 7.12	214.30 ± 12.22	89.45 ± 7.21	1.40 ± 0.14
10.89 ± 0.14	1701.31 ± 44.06	226.25 ± 13.21	355.81 ± 18.46	129.56 ± 10.12	1.75 ± 0.17
11.39 ± 0.14	1573.60 ± 42.30	360.94 ± 22.88	528.75 ± 30.23	167.81 ± 14.33	2.15 ± 0.23
11.88 ± 0.13	1706.34 ± 44.19	446.51 ± 28.58	660.35 ± 33.82	213.84 ± 17.53	2.10 ± 0.22
12.38 ± 0.15	1828.14 ± 44.79	535.83 ± 35.36	800.07 ± 59.32	264.24 ± 26.23	2.03 ± 0.24
12.87 ± 0.15	1938.13 ± 44.00	598.72 ± 39.64	881.83 ± 65.38	283.11 ± 28.14	2.11 ± 0.25
13.37 ± 0.15	2038.87 ± 37.92	640.87 ± 43.32	941.42 ± 45.02	300.55 ± 24.89	2.13 ± 0.23
13.87 ± 0.07	2116.77± 26.46	696.85 ± 42.09	1026.67 ± 46.78	329.82 ± 24.95	2.11 ± 0.21
14.36 ± 0.11	2153.29 ± 24.12	740.11 ± 43.67	1070.21 ± 53.18	330.10 ± 25.46	2.24 ± 0.22
14.80 ± 0.07	2164.20 ± 22.83	754.56 ± 46.86	1090.26 ± 54.71	335.70 ± 26.80	2.25 ± 0.23

TABLE II: Uncertainty budget for the ^{76}Ge(n,2n)^{75}Ge, ^{76}Ge(n,2n)^{75m}Ge and monitor reaction cross-section values.

Parameter	Ge (%)	Monitor (%)
Photo-peak area	0.1-2.48	0.36-6.32
Reference cross sections		1.05-3.06
Detector efficiency	2.30-5.92	0.62-5.30
Source geometry and self-absorption of γ-rays	<0.2	<0.2
Half-life	< 1.1	0.01
γ-ray intensity	-	-
Irradiation time	< 1	< 1
Decay time	< 1	< 1
Counting time	< 1	< 1
Neutron flux correction	-	<2
Neutron flux fluctuation	<1	< 1

and the isomeric-to-ground state ratio σ_m/σ_g are shown in Figs. 9 and 10. (see Sec. IV).

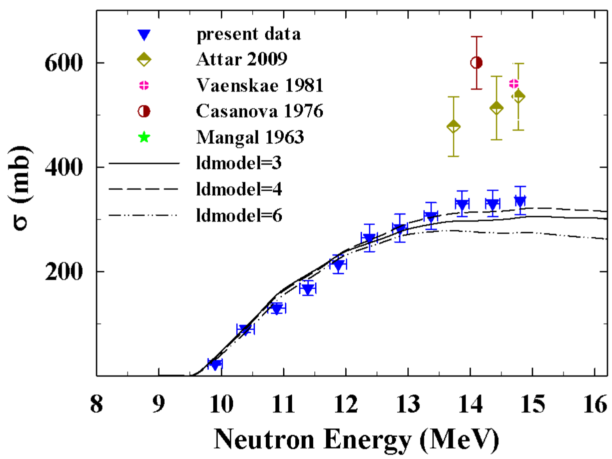

FIG. 9: (Color online) Comparison of cross-section data [14, 17, 18] and TALYS calculations for the ^{76}Ge(n,2n)^{75g}Ge reaction using different level-density choices (see text). Our data are best described by the generalized superfluid model.

IV. NUCLEAR-MODEL CALCULATIONS

The cross sections for the ^{76}Ge(n,2n)^{75m}Ge, ^{76}Ge(n,2n)^{75}Ge and ^{76}Ge(n,2n)^{75g}Ge reactions were calculated in the neutron energy range from 9 MeV to 16 MeV using the recent version (version 1.8) of the nuclear-model code TALYS [4]. In the present work the calculations have been performed mainly with input parameters given by default settings in TALYS. However, one exception deals with the nuclear level density. Here, particular models were used to investigate their sensitivity.

The level density is an essential ingredient for calculation of reaction cross sections. It is a key parameter in any statistical model calculation at excitation energies where discrete level information is not available or incomplete. The most important step for determining a reliable theoretical prediction of cross sections, energy spectra, angular distributions, and other nuclear reaction observables is to use the correct level density together with the appropriate optical-model potential parameters. In TALYS 1.8, the level density can be calculated via six different choices, corresponding to the input parameter ldmodel equal 1 to 6. The three phenomenological and the three microscopic options for level densities are ldmodel=1: constant temperature plus fermi gas model, ldmodel=2: back-shifted Fermi gas model, ldmodel=3: generalised super-fluid model, ldmodel=4: microscopic level densities (Skyme force) from Goriely's tables, ldmodel=5: microscopic level densities (Skyme force) from Hilaire's combinatorial tables, ldmodel=6: microscopic level densitites (tempearature dependent HFB, Gogny force) from Hilaire's combinatorial tables.

Moreover, in these TALYS calculations, for each level-density choice, the default proton and neutron optical-model potentials using the local and global parameterization of Koning and Delaroche can be applied [35]. These potentials provide the necessary reaction cross sections and transmission coefficients for the statistical model calculations. The TALYS nuclear structure database has been generated from the Reference Input Parameter Library (RIPL) [36]. The Hauser-Feshbach model is used for the calculation of the compound-nucleus contribution [37]. In addition, the pre-equilibrium reactions were included via the two component exciton model of Kalbach [38].

A comparison of experimental data for the reactions of interest with predictions of the TALYS-1.8 code utilizing different level-density model options is presented in Figs. 7-10. It can be clearly seen that for our experimental results the best agreement is achieved using the calculations with ldmodel = 3. For completeness, Fig. 8 shows calculations with all the level-density options provided with the TALYS-1.8 code, in contrast to Figs. 7, 9-10, where only a subset of level-density choices are considered.

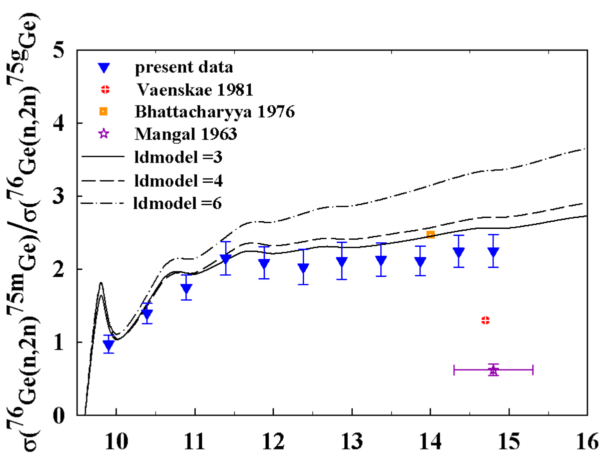

FIG. 10: (Color online) Comparison of the energy dependence of the measured isomeric to ground state cross-section ratio for the ^{76}Ge(n,2n)^{75}Ge reaction with the predictions by the TALYS code, and the existing previous measurements [17, 21, 34].

V. CONCLUSION

The total and isomeric cross sections of the reaction ^{76}Ge(n,2n)^{75}Ge were measured from threshold to 14.8 MeV to guide evaluations and model calculations to provide reliable cross-section data throughout the energy range of interest for tracking neutrons in future large-scale HPGe detectors currently envisioned for searches of $0\nu\beta\beta$ of ^{76}Ge. Neutron-induced background reactions are a major concern because they have the potential to mimic the signal of interest. Our measured cross-section data follow the trend of the few previous data below 13 MeV, but are lower in magnitude than most of the data in the heavily researched 14 MeV energy range, resulting in evaluations and the model calculation TENDL-2014 to miss our data for energies above 11 MeV. Previous data for the isomeric cross section do not exist below 13 MeV. However, above this energy our data favor the lower band of the available data. Our TALYS calculations performed with the level density of the generalized super-fluid model give an overall satisfactory description of the measured total and isomeric and the deduced ground state cross section of the ^{76}Ge(n,2n)^{75}Ge reaction.

VI. ACKNOWLEDGEMENT

This work was supported in part by the US Department of Energy, Office of Nuclear Physics under Grant No. DE-FG02-ER41033.

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