

## Isomeric cross-section ratios for the formation of $^{75}\text{Ge}^{m,g}$ in $(n,p)$ , $(n,\alpha)$ , and $(n,2n)$ reactions from 6 to 15 MeV

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The isomeric cross-section ratio  $\sigma_m/(\sigma_m + \sigma_g)$  for the formation of  $^{75}\text{Ge}^{m,g}$  was measured in  $^{75}\text{As}(n,p)$ ,  $^{78}\text{Se}(n,\alpha)$ , and  $^{76}\text{Ge}(n,2n)$  processes in the neutron energy range of 6.3 to 14.7 MeV. Use was made of the activation technique in combination with high-resolution  $\gamma$ -ray spectrometry. The ratio is relatively high ( $>0.70$ ) even at low incident particle energies and appears to increase only slightly with increasing energy. Statistical model calculations, taking into account precompound effects, were performed for the total reaction channel as well as for the isomer ratio. In either case, good agreement was obtained. For the investigated isomeric pair, no significant effect of reaction channels on the isomeric cross-section ratio was observed.

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### I. INTRODUCTION

Isomeric cross-section ratios have attracted considerable attention in recent years. The status of experimental and theoretical information available was recently reviewed (cf. Ref. [1]). The yield of the high-spin isomer increases with increasing projectile energy (cf. Refs. [2–6]), and the isomer ratio appears to be primarily governed by the spins of the levels involved, rather than their separation and excitation energies (cf. Ref. [6]). As far as nuclear model calculations are concerned, cross sections for the formation of isomeric states are more difficult to predict than those for the total reaction channels. Calculations on the isomeric states depend critically on the input level scheme of the product nucleus (cf. Refs. [2,3]) as well as on the spin distribution of the level density, characterized in terms of the effective moment of inertia (cf. Refs. [2,3,5]). Despite some of these recent interesting results, our knowledge regarding the formation of isomeric states is still rather scanty. Two areas of investigations deserving more attention (cf. Ref. [1]) include (a) the effect of reaction channels and (b) the effect of a systematic increase in spin. The present work attempts to study the effect of reaction channels on the isomeric cross-section ratio.

We chose to study the isomeric pair  $^{75}\text{Ge}^{m,g}$ , a simplified level and decay scheme of which is given in Fig. 1. It can be formed via three different routes, viz.,  $^{75}\text{As}(n,p)$ ,  $^{78}\text{Se}(n,\alpha)$ , and  $^{76}\text{Ge}(n,2n)$ . Investigations were performed on these reactions in the neutron energy range of 6–15 MeV.

### II. EXPERIMENTAL DETAILS

Total cross sections for the formation of  $^{75}\text{Ge}^{m,g}$  in the three processes  $(n,p)$ ,  $(n,\alpha)$ , and  $(n,2n)$  were determined by activation and identification of the ground state after the complete decay of the metastable state. The details have already been given [7].

In the work on the metastable state described here, only

relative measurements were carried out; i.e., the neutron flux density was not determined. As in previous studies [7], measurements in the 6–12 MeV energy range were carried out using quasimonoenergetic neutrons produced via the  $dd$  reaction on a  $\text{D}_2$ -gas target at the variable energy compact cyclotron CV28 of KFA Jülich. Investigations with 14.7 MeV neutrons were performed using the  $dt$ -neutron generator at TU Dresden.

Samples of natural isotopic composition were used. For irradiations with  $dd$  neutrons at Jülich,  $\text{As}_2\text{O}_3$  (2.5 g, 99.5%, supplied by Riedel de Haen and Merck) and Se (2.5 g, 99.5%, Merck) were pressed to 1.3 cm diam  $\times$  0.5 cm thick disks, and  $\text{GeO}_2$  (5 g, 99.999%, Heraeus) was filled into a 1.3 cm diam  $\times$  3.1 cm long polyethylene tube of wall

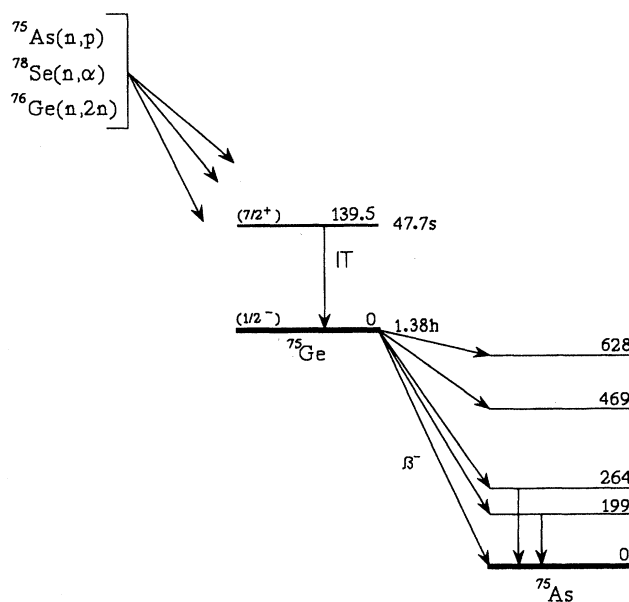


FIG. 1. Simplified representation of formation and decay of the isomeric pair  $^{75}\text{Ge}^{m,g}$ . The nuclear level energies are in keV.

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thickness 0.05 cm. In work with  $dt$  neutrons at Dresden, materials of the same purity and from the same suppliers were used, but the dimensions of the samples were somewhat different.  $\text{As}_2\text{O}_3$  (1.5 g) was pressed to a 0.9 cm diam  $\times$  0.45 cm thick disk and Se and Ge (each 2.5 g) to 1.5 cm diam  $\times$  0.5 cm thick pellets.

All irradiations were done in the  $0^\circ$  direction relative to the deuteron beam and the samples were placed at a distance of 0.8 cm behind the beam stop (in the case of the gas target) and 1.1 cm from the neutron source (in the case of the neutron generator). The duration of each irradiation varied between 5 and 10 min. The flux during the irradiations with  $dd$  neutrons was constant, as checked by monitoring the deuterium gas pressure in the target and the deuteron beam current. For the  $dt$  source, small flux variations during the irradiations were corrected using the count rates of the associated  $\alpha$  particles.

The irradiation time of 5–10 min chosen in this work was a compromise to study both the 47.7 s isomeric state and the 1.38 h ground state of  $^{75}\text{Ge}$ . Shorter irradiations would have been beneficial to the study of the isomeric state. However, the ground state would have been hardly observed. Since the major error in the experimentally determined isomeric cross-section ratio originated anyway from the poor counting statistics involved, shorter irradiations would have resulted in larger errors. The irradiation time of 5–10 min led to results of reasonable accuracy.

The neutron energy effective at each sample was calculated using the program NEUT in the 6–12 MeV range (cf. Ref. [8]) and PROFIL in the 14 MeV region (cf. Ref. [7]). In the case of  $\text{GeO}_2$  samples irradiated at Jülich, the neutron attenuation was taken into account by using the results of neutron transport calculation (the MNCP code [9]). The mean neutron energies had therefore slightly larger spreads. In an earlier work [7] we showed that the deuteron energies recorded at the compact cyclotron CV28 via adjustment of cyclotron parameters were somewhat high and a normalization factor had to be used. Recent accurate energy measurements [10] confirmed that observation.

After the end of irradiation, the sample was transferred within  $\sim 40$  s to a HPGe detector coupled to an ORTEC MCA plug-in card which was connected to an IBM-compatible PC-AT. Several  $\gamma$ -ray spectra were recorded at 30–60 s intervals. They served to characterize and measure the activity of  $^{75}\text{Ge}^m$  ( $T_{1/2}=47.7$  s) via its 139.5 keV  $\gamma$  ray ( $I_\gamma=38.8\%$ ). After about 15 min, i.e., after the complete decay of  $^{75}\text{Ge}^m$  to  $^{75}\text{Ge}^g$  ( $T_{1/2}=1.38$  h), the sample was counted in the same geometry for longer periods of 15–20 min and the activity of  $^{75}\text{Ge}^g$  determined via the characteristic 264.7 keV  $\gamma$  ray ( $I_\gamma=11.3\%$ ). The count rates of both  $^{75}\text{Ge}^m$  and  $^{75}\text{Ge}^g$  at the end of bombardment (EOB) were obtained by applying corrections for decay. For  $^{75}\text{Ge}^m$ , it was straightforward. In the case of  $^{75}\text{Ge}^g$  the parent-daughter decay relationship was taken into account. The count rates were converted to activities (decay rates) at the EOB by applying corrections for  $\gamma$ -ray intensity, detector efficiency, and self-absorption.

From the activities of the two isomers at the EOB, viz.,  $A(^{75}\text{Ge}^m)$  and  $A(^{75}\text{Ge}^g)$ , the isomeric cross-section ratio  $\sigma_m/(\sigma_m + \sigma_g)$  was obtained using the expression

$$\frac{\sigma_m}{\sigma_m + \sigma_g} = \left\{ \left[ \frac{A(^{75}\text{Ge}^g)}{A(^{75}\text{Ge}^m)} - \frac{\lambda_g}{\lambda_g - \lambda_m} \frac{e^{-\lambda_g t} - e^{-\lambda_m t}}{1 - e^{-\lambda_m t}} \right] \frac{1 - e^{-\lambda_m t}}{1 - e^{-\lambda_g t}} \right\}^{-1},$$

where  $\lambda_m$  and  $\lambda_g$  denote the decay constants of the two isomers and  $t$  is the time of irradiation.

At each neutron energy several irradiations were done, yielding several isomeric cross-section values. The averaged value at each energy was then obtained by taking the arithmetic mean of the measured values.

Irradiations with empty gas cells were not done; i.e., the corrections for activations from background neutrons were ignored, since they were assumed to be the same for the two isomers. This may not be strictly correct since the thresholds for the formation of the two isomers differ by about 0.14 MeV. However, the error involved is expected to be small. In the irradiations done at Dresden, a small correction for the nonconstancy in the neutron flux was applied.

The errors in isomeric cross-section ratios should generally be smaller than those in absolute cross sections, since uncertainties in monitor reaction cross sections and background neutron contributions have not to be taken into account. However, due to relatively low count rates the statistical errors were rather large (up to 22%). The uncertainty in the efficiency of the detector ( $\sim 5\%$ , including self-absorption correction) and the uncertainties in the decay constants were considered as correlated errors. Combining the two errors in quadrature, a total error ranging between 6% and 24% was obtained.

Since  $\text{GeO}_2$  of natural isotopic abundance was used ( $^{70}\text{Ge}$ , 20.5%;  $^{72}\text{Ge}$ , 27.4%;  $^{73}\text{Ge}$ , 7.8%;  $^{74}\text{Ge}$ , 36.5%,  $^{76}\text{Ge}$ , 7.8%), the  $^{75}\text{Ge}^{m,g}$  states can be produced via the reactions  $^{76}\text{Ge}(n,2n)$  and  $^{74}\text{Ge}(n,\gamma)$ . While investigating the  $^{76}\text{Ge}(n,2n)^{75}\text{Ge}^{m,g}$  process, a correction for the contribution from the  $^{74}\text{Ge}(n,\gamma)^{75}\text{Ge}^{m,g}$  reaction was therefore necessary. This was done as follows: The isomeric cross-section ratio  $\sigma_m/(\sigma_m + \sigma_g)$  in the  $^{74}\text{Ge}(n,\gamma)$  process with thermal neutrons is known to be 0.33. This value was assumed also at neutron energies in the MeV region (where unfortunately no data exist). For the different incident neutron energies, the ratios of the activities induced via  $(n,\gamma)$  and  $(n,2n)$  processes were obtained by neutron transport calculations [7]. The correction factors were then calculated using the isotopic abundances of  $^{74}\text{Ge}$  and  $^{76}\text{Ge}$  (0.365 and 0.078, respectively) and assuming the isomeric cross-section ratio of  $^{75}\text{Ge}^{m,g}$  via the  $^{76}\text{Ge}(n,2n)$  process being constant and equal to 0.8. At 10 MeV the correction amounted to about 20% and at 14.7 MeV to about 2%, with a relative uncertainty of almost 100%. This uncertainty in the correction was included in the estimation of the total error.

### III. NUCLEAR MODEL CALCULATIONS

In order to describe the measured isomeric cross-section ratios, nuclear model calculations were performed using the computer code STAPRE [11]. The method was similar to that described earlier [4], utilizing the exciton model formalism for preequilibrium particle emission, the width-fluctuation-corrected Hauser-Feshbach formula for first chance emission

TABLE I. Properties of discrete levels of  $^{75}\text{Ge}$  considered in the statistical model calculations.

$E$ (MeV)	$J^\pi$	$E$ (MeV)	$J^\pi$
0.0	$1/2^-$	1.1369	$3/2^-$
0.1397	$7/2^+$	1.1900	$9/2^+$
0.1922	$5/2^+$	1.2407	$5/2^-$
0.1999	$9/2^+$	1.2570	$9/2^+$
0.2531	$1/2^-$	1.3944	$5/2^+$
0.3168	$5/2^-$	1.4080	$9/2^+$
0.4571	$5/2^-$	1.4162	$3/2^-$
0.5747	$3/2^-$	1.4274	$7/2^+$
0.5844	$5/2^+$	1.5015	$1/2^-$
0.6510	$5/2^-$	1.5144	$1/2^+$
0.6737	$1/2^+$	1.5377	$5/2^+$
0.7622	$3/2^+$	1.6030	$7/2^-$
0.8855	$1/2^-$	1.6820	$9/2^+$
0.9870	$7/2^+$	1.6990	$5/2^-$
1.0622	$9/2^+$	1.7184	$5/2^+$
1.1278	$7/2^+$		

from the equilibrated system, and the evaporation formula for higher chance emission. Direct interactions were not considered, but their contributions should be  $< 10\%$ .

The particle transmission coefficients were generated via the spherical optical model using the program ABACUS [12]; individually, the following global parameter sets were used: Wilmore and Hodgson [13] for neutrons, Mani *et al.* [14] for protons, and Huizenga and Igo [15] for  $\alpha$  particles. The transmission coefficients for the latter were reduced by 6% as the absorption cross sections achieved by this optical potential are known to be quite high. The  $\gamma$ -ray strength functions for  $E1$  transitions were derived according to the Brink-Axel model [16], but with slightly modified values for the position, width, and peak cross section of the giant-dipole resonance, and for all the other transitions according to the Weisskopf model [17]. The  $E1$  strength-function value as well as the  $M1/E1$  ratio at the neutron binding energy was prescribed according to McCullagh *et al.* [18], whereas the normalization of the strengths of the other radiation types relative to  $E1$  was done according to Weisskopf's estimate.

Regarding the exciton model incorporated in STAPRE, we

chose the energy and mass dependence of the effective matrix element for internal transitions as  $|M|^2 = (FM)A^{-3}E^{-1}$ , with  $FM = 237 \text{ MeV}^3$ . This value was deduced from some experimental data (cf. Ref. [19]), on the condition that at an excitation energy of 21 MeV the transition rate follows the relation  $\lambda_+(3) = 5 \times 10^{21} \text{ s}^{-1}$ . The preformation factor for  $\alpha$  particles in the formulation of the emission rates was adopted as 0.28 (cf. Ref. [20]).

Particle emission was treated in the usual way: The excited states of the product nuclei were described by the available information on discrete levels [21]; at higher excitation energies, however, the levels were treated as a continuum described by the back-shifted Fermi gas model. In particular, for  $^{75}\text{Ge}$  we included 31 levels whose properties are listed in Table I. The choice of level density parameters was guided by the compilation of Dilg *et al.* [22]. The parameters were verified by checking the reproduction of cumulative level densities and resonance spacings. For this purpose, the experimental information on discrete levels was used much higher up in excitation energy than in the actual statistical model calculations, as no attention was paid to completeness of angular momentum and parity assignment. The spin distribution of the level density was characterized by the ratio of the effective moment of inertia,  $\theta_{\text{eff}}$ , to rigid body moment of inertia,  $\theta_{\text{rig}}$  ( $\eta = \theta_{\text{eff}}/\theta_{\text{rig}}$ ). The calculations were performed for  $\eta = 1.0$  and 0.5 to investigate its effect on the isomeric cross-section ratio.

## IV. RESULTS AND DISCUSSION

### A. Experimental data

The experimentally determined isomeric cross-section ratios  $[\sigma_m/(\sigma_m + \sigma_g)]$  for the isomeric pair  $^{75}\text{Ge}^{m,g}$  produced in three different nuclear reactions are given in Table II. In the literature some data above 13 MeV have been given. Our data in the neutron energy range of 6–12 MeV describe the first measurements.

The data are shown in Figs. 2–4 as a function of incident neutron energy. In addition to our own measurements (cf. Table II), results available in the literature are also given for the energy range above 13 MeV (cf. Refs. [23–26]). The solid and dashed lines represent results of nuclear model calculations which are described in Sec. IV C.

TABLE II. Measured isomeric cross-section ratios.

$^{75}\text{As}(n,p)^{75}\text{Ge}^{m,g}$		$^{78}\text{Se}(n,\alpha)^{75}\text{Ge}^{m,g}$		$^{76}\text{Ge}(n,2n)^{75}\text{Ge}^{m,g}$	
$E_n^a$	$\frac{\sigma_m}{\sigma_m + \sigma_g}$	$E_n^a$	$\frac{\sigma_m}{\sigma_m + \sigma_g}$	$E_n^a$	$\frac{\sigma_m}{\sigma_m + \sigma_g}$
(keV)		(keV)		(keV)	
6340 ± 140	0.69 ± 0.08				
7320 ± 160	0.69 ± 0.09				
8260 ± 180	0.67 ± 0.09	8120 ± 145	0.71 ± 0.12		
9180 ± 200	0.71 ± 0.09	9030 ± 160	0.74 ± 0.17		
10 090 ± 220	0.75 ± 0.09	9910 ± 180	0.73 ± 0.17	9980 ± 200	0.79 ± 0.12
10 980 ± 240	0.71 ± 0.11	10 790 ± 190	0.73 ± 0.10	10 850 ± 220	0.79 ± 0.06
11 870 ± 255	0.75 ± 0.09	11 650 ± 190	0.70 ± 0.12	11 720 ± 240	0.73 ± 0.05
14 680 ± 100	0.80 ± 0.06	14 680 ± 100	0.69 ± 0.09	14 680 ± 100	0.80 ± 0.04

<sup>a</sup>The deviation gives the uncertainty of the mean neutron energy. The full width at half maximum (FWHM) of the neutron energy distribution can be found in Ref. [7].

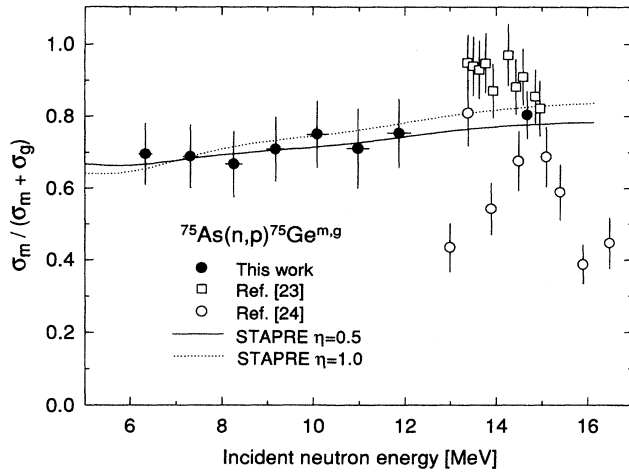


FIG. 2. Isomeric cross-section ratio for the isomeric pair  $^{75}\text{Ge}^{m,g}$ , formed via the  $(n,p)$  reaction on  $^{75}\text{As}$ , plotted as a function of incident neutron energy.

For the  $^{75}\text{As}(n,p)^{75}\text{Ge}^{m,g}$  process (cf. Fig. 2) two reports exist in the literature [23,24]. The data of Grochulski *et al.* [24] show large fluctuations, mainly due to large errors in total  $(n,p)$  cross sections. The values of Okumura [23], on the other hand, are more consistent. Our 14 MeV value is in agreement with the latter data within the experimental errors. Considering all the data in the investigated energy range of 6–14 MeV, the isomeric cross-section ratio appears to increase from 0.69 to 0.80, i.e., slowly with increasing neutron energy. The high-spin metastable state ( $I=7/2^+$ ) is thus preferred even at 6 MeV and this preference increases slowly with increasing neutron energy.

In the case of the  $^{78}\text{Se}(n,\alpha)^{75}\text{Ge}^{m,g}$  reaction (cf. Fig. 3) only one isomeric cross-section ratio measurement was reported at 14.8 MeV [25]. Our result at 14.7 MeV is very different from that value. Our investigations suggest that the

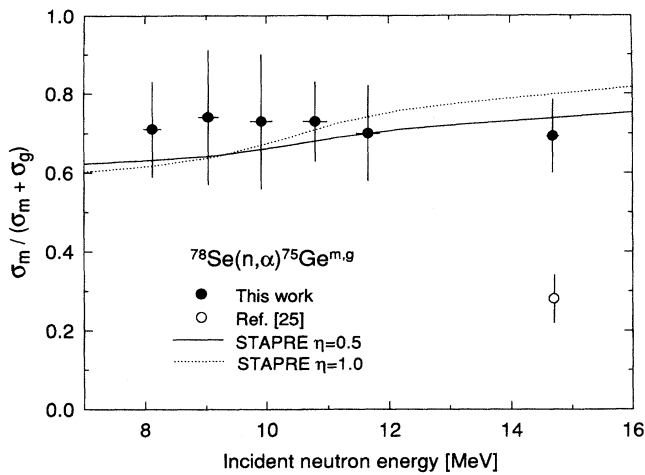


FIG. 3. Isomeric cross-section ratio for the isomeric pair  $^{75}\text{Ge}^{m,g}$ , formed via the  $(n,\alpha)$  reaction on  $^{78}\text{Se}$ , plotted as a function of incident neutron energy.

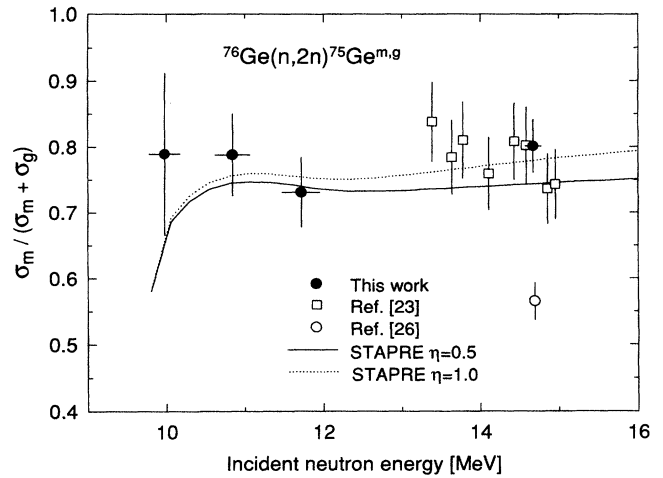


FIG. 4. Isomeric cross-section ratio for the isomeric pair  $^{75}\text{Ge}^{m,g}$ , formed via the  $(n,2n)$  reaction on  $^{76}\text{Ge}$ , plotted as a function of incident neutron energy.

ratio has a high value of 0.71 even at 6 MeV; however, the value remains almost constant over the whole investigated energy range.

For the  $^{76}\text{Ge}(n,2n)^{75}\text{Ge}^{m,g}$  reaction (cf. Fig. 4) two experimental studies have been described around 14 MeV [23,26]. Our 14.7 MeV result agrees with the data of Okumura [23]. Again the ratio has a relatively high value ( $> 0.70$ ) even near the threshold of the reaction. Similar to  $(n,p)$  and  $(n,\alpha)$  reactions, the increase in the preferential population of the high-spin isomer with increasing neutron energy, if any, is not well defined.

## B. Validation of calculational method

Prior to a calculation of isomeric cross-section ratios, it appeared important to validate the calculational method. The program STAPRE [11] generally reproduces the total  $(n,2n)$ ,  $(n,p)$ , and  $(n,\alpha)$  reaction cross sections up to 20 MeV with reasonable accuracies, provided the optical model and level density parameters are properly chosen. We consider here the excitation functions of the  $^{76}\text{Ge}(n,2n)^{75}\text{Ge}^{m,g}$ ,  $^{75}\text{As}(n,p)^{75}\text{Ge}^{m,g}$ , and  $^{78}\text{Se}(n,\alpha)^{75}\text{Ge}^{m,g}$  reactions which were measured earlier [7]. Now nuclear model calculations using the same global parameters were performed for all the reactions. In each case good agreement was found. As an example we present in Fig. 5 the experimental [7,25,27–30] and calculated results (this work) for the  $(n,\alpha)$  reaction. This process is somewhat more difficult to reproduce than the other two reactions, since it involves the emission of a light complex particle. In Fig. 5 most of the experimental and calculated data agree well over the whole investigated energy range; a change in the parameter  $\eta$  (see above) has almost no effect on the results. We also achieved satisfactory reproduction of the cross sections in the competing channels  $^{75}\text{As}(n,\alpha)^{72}\text{Ga}$ ,  $^{75}\text{As}(n,2n)^{74}\text{As}$ , and  $^{78}\text{Se}(n,p)^{78}\text{As}$  measured in the same experiment [7]. This agreement added confidence to our calculational method.

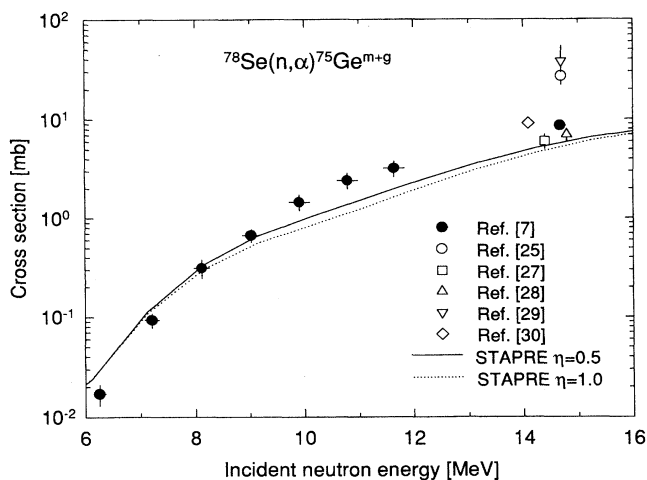


FIG. 5. Excitation function of the  $^{78}\text{Se}(n,\alpha)^{75}\text{Ge}^{m+g}$  process.

### C. Comparison of experimental and calculated data on isomeric cross-section ratios

The results of nuclear model calculations on the isomeric cross-section ratios are shown in Figs. 2–4 together with the experimental data. In all the three cases, viz.,  $(n,p)$ ,  $(n,\alpha)$ , and  $(n,2n)$  reactions, good agreement is observed. A change in  $\eta$  (0.5 or 1.0) does not have any strong effect on the isomer ratio for incident neutron energies up to 10 MeV. At  $E_n \geq 10$  MeV, however, the calculated isomeric cross-section ratios are consistently higher when  $\eta=1.0$  is used. On the other hand, considering the error limits of the experimental data, it is obvious that either of the two  $\eta$  values (0.5 or 1.0) leads to acceptable results. This is not surprising, since the spins of the ground and isomeric states are about equally distant from the spin values, at which—under usage of  $\eta=0.5$  or  $\eta=1.0$ —the level densities and hence the populations show a maximum.

A prescription assuming a  $(2J+1)$  population of each state would yield a value of 0.8 for  $\sigma_m/(\sigma_m + \sigma_g)$ , which appears to be a reasonable description of the isomeric ratios in all three reactions investigated here. However, such a prescription does not consider the individual details of the level scheme, which govern the branching of the  $\gamma$ -ray cascades; nor does it allow for a dependence of the isomeric cross-section ratio on incident energy.

As a variation of model options, we also tried an  $M1$  strength function derived from a giant-dipole resonance rather than constant with energy. It turned out that the isomeric cross-section ratio increases by less than 1%. The use of an exciton-number-dependent matrix element [31] for computing the internal-transition rates in the exciton-model part of the calculations causes no difference to the ratio over the incident-energy range considered.

## V. CONCLUSION

From the experimental and theoretical studies performed on  $^{75}\text{Ge}^{m,g}$ , it is concluded that the higher-spin metastable state  $^{75}\text{Ge}^m(I=7/2^+)$  is preferentially populated in all three reactions investigated. The increase in the isomeric cross-section ratio over the incident neutron energy range of 6–15 MeV is, however, small. No significant effect of reaction channels on the isomeric cross-section ratio was observed.

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