

$M1$ γ Strength for Zirconium Nuclei in the Photoneutron Channel

H. Utsunomiya,¹ S. Goriely,² T. Kondo,¹ T. Kaihori,¹ A. Makinaga,¹ S. Goko,³ H. Akimune,¹ T. Yamagata,¹ H. Toyokawa,⁴ T. Matsumoto,⁴ H. Harano,⁴ S. Hohara,⁵ Y.-W. Lui,⁶ S. Hilaire,⁷ S. Péru,⁷ and A. J. Koning⁸

¹*Department of Physics, Konan University, Okamoto 8-9-1, Higashinada, Kobe 658-8501, Japan*

²*Institut d'Astronomie et d'Astrophysique, Université Libre de Bruxelles, Campus de la Plaine, CP-226, 1050 Brussels, Belgium*

³*Japan Atomic Energy Agency, Tokai-mura, Naka, Ibaraki 319-1195, Japan*

⁴*National Institute of Advanced Industrial Science and Technology, Tsukuba 305-8568, Japan*

⁵*Atomic Energy Research Institute, Kinki University, Kowakae 3-4-1, Osaka 577-8502, Japan*

⁶*Cyclotron Institute, Texas A&M University, College Station, Texas 77843, USA*

⁷*Département de Physique Théorique et Appliquée, Service de Physique Nucléaire, B.P. 12 - F-91680 Bruyères-le-Châtel, France*

⁸*Nuclear Research and Consultancy Group, P.O. Box 25, NL-1755 ZG Petten, The Netherlands*

(Received 30 November 2007; published 24 April 2008)

Photoneutron cross sections were measured for ^{91}Zr , ^{92}Zr , and ^{94}Zr near the neutron separation energy with quasimonochromatic γ rays. The data exhibit some extra components around the neutron threshold. A coherent analysis of the photoneutron data for ^{92}Zr together with the neutron capture on ^{91}Zr based on the microscopic Hartree-Fock-Bogoliubov plus quasiparticle random-phase approximation model for the $E1$ strength has revealed the presence of an $M1$ resonance at 9 MeV. The microscopic approach systematically shows the same $M1$ strength in the photoneutron cross section for ^{91}Zr and ^{94}Zr . The total $M1$ strength is about 75% larger than the strength predicted by the systematics, being qualitatively consistent with the giant $M1$ resonance observed in the inelastic proton scattering.

DOI: [10.1103/PhysRevLett.100.162502](https://doi.org/10.1103/PhysRevLett.100.162502)

PACS numbers: 25.20.-x, 21.10.Pc, 21.60.Jz, 27.60.+j

There exist 35 neutron-deficient nuclides that, unlike the majority of the elements heavier than iron, neither the slow nor the rapid neutron capture processes can produce. The nucleosynthesis of these nuclides is referred to as the p process (for a review, see [1]) in which the photodisintegration of preexisting s - and r -process nuclei in a hot stellar plasma at typically 2 to 3×10^9 degrees plays a primary role. Since nuclei are thermally equilibrated in the photon bath, the γ strength function near, both above and below, the neutron separation energy is a key nuclear ingredient for the p process. Measurements of photoneutron cross sections immediately above the neutron separation energy [2] supplemented with the quasiparticle random-phase approximation (QRPA) calculation [3,4] have enabled us to investigate the $E1$ γ -ray strength function of direct relevance to the p process. However, investigations have so far been limited to the $E1$ γ strength function. Despite the experimental efforts in the (γ, n) and (γ, γ') channels, little is known on the magnetic-dipole strength function near neutron threshold [5].

Magnetic-dipole strength was observed for ^{90}Zr in the (e, e') [6], the (p, p') [7–10], and the (γ, γ') [11] experiments at excitation energies $E_x = 8$ –10 MeV. While the strength is weak and fragmented in (e, e') , it is strong like a giant $M1$ resonance in (p, p') reactions. The $M1$ excitation in ^{90}Zr found in (p, p') reactions is rather consistent with the excitation of the Gamow-Teller resonance in the analogue (p, n) channel [7,9,12]. The strong $M1$ strength is also observed for ^{92}Zr , ^{94}Zr , and ^{96}Zr in the very same energy region in the inelastic proton scattering [9]. It is interesting to note that, although it lies below the neutron

separation energy ($S_n = 11.97$ MeV) in ^{90}Zr , the $M1$ strength lies above $S_n = 8.635$ MeV for ^{92}Zr , 8.220 MeV for ^{94}Zr , and 7.854 MeV for ^{96}Zr .

In this Letter, we present (γ, n) cross sections for ^{91}Zr , ^{92}Zr , and ^{94}Zr near the neutron separation energy (S_n) that are found to be strongly enhanced with respect to theoretical predictions and threshold behavior. It is shown that the enhancement can be systematically explained by an extra $M1$ strength.

We measured the photoneutron cross sections for ^{91}Zr , ^{92}Zr , and ^{94}Zr near neutron threshold with quasimonochromatic γ -ray beams produced from laser inverse-Compton scattering (LCS) at the National Institute of Advanced Industrial Science and Technology. Enriched samples of ^{91}Zr (90.4%), ^{92}Zr (91.4%), and ^{94}Zr (92.6%) in the chemical form ZrO_2 were irradiated. A major isotopic impurity present in the target samples is ^{90}Zr (3.7%–5.8%) with the high neutron threshold energy. A Nd:YVO₄ Q-switch laser was operated at 20 kHz in the second harmonics ($\lambda = 532$ nm). The γ -ray beams had the same macroscopic time structure of 80 ms beam-on and 20 ms beam-off as that of the laser. A 4π -type neutron detector consisting of 20 ^3He counters embedded in a polyethylene moderator was used. Background neutrons were detected during the 20 ms beam-off. Photoneutron cross sections were determined at the average γ -ray energies with the Taylor expansion method [13]. A measurement at 8.16 MeV (the maximum energy of an LCS γ beam) showed that the cross section for ^{94}Zr is vanishing below the neutron threshold. The uncertainty of the present photoneutron cross section associated with the isotopic target impurities was estimated

to be 3.6%–7.4% for ^{91}Zr , 1.7%–6.2% for ^{92}Zr , and 1.8%–3.0% for ^{94}Zr . The systematic uncertainty for the cross section is 4.8%–9.1%; its breakdown is, besides the target impurities, 3.2% for the neutron detection efficiency, 3% for the number of incident γ rays, and a few percent for the beam size effect. Further experimental details are found in [13].

Results of the present photoneutron cross section measurement for ^{91}Zr , ^{92}Zr , and ^{94}Zr are shown in Fig. 1. For comparison, the experimental cross sections of [14] are also shown. The present measurement fills previously unexplored energy regions near neutron threshold from 7.33 to 10.71 MeV for ^{91}Zr and from 8.66 to 9.97 MeV for ^{92}Zr . Note that the present cross sections for ^{92}Zr are significantly larger than those of [14] below 11 MeV, while for ^{94}Zr , the data are in good agreement with each other except the two data points of [14], which show nonvanishing cross sections below S_n .

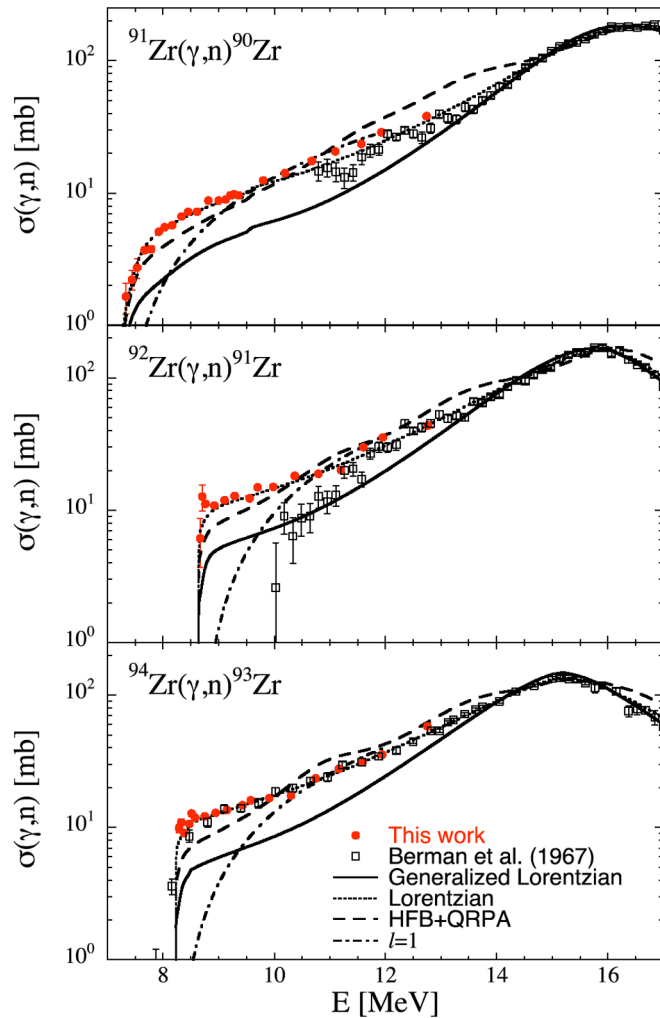


FIG. 1 (color online). Comparison of experimental and theoretical photoneutron cross sections. The theoretical estimates are based on different $E1$ γ -ray strengths, as described in the text, but they all consider the same standard $M1$ strength from systematics. The dash-dotted line shows the expected threshold behavior $\sigma(E_\gamma) \propto (E_\gamma - S_n)^{\ell+1/2}$ with $\ell = 1$.

The threshold behavior of reaction cross sections is well elucidated [15]. The dependence of photoneutron cross sections on neutron energy follows $\sigma(E_\gamma) \propto (E_\gamma - S_n)^{\ell+1/2}$ near the threshold, where ℓ is the orbital angular momentum of neutrons. In terms of the detailed balance of (γ, n) and (n, γ) , this energy dependence arises from the properties (de Broglie wavelength and the matrix element) of neutron channel, not those of photon channel. In the $E1$ excitation of $^{91,92,94}\text{Zr}$, s -wave neutron emission is inhibited, the lowest ℓ allowed being 1. The present photoneutron cross sections exhibit a very strong enhancement from the $\ell = 1$ law (see Fig. 1), which was unobserved in nuclei studied in the past (^{181}Ta [2], ^{139}La , ^{141}Pr [13], ^{188}Os , ^{187}Re , ^{185}W [16]), being indicative of the presence of extra strength that are attributable to $E1$, $M1$, or other multipoles.

Theoretically, the present experimental data have been analyzed on the basis of the TALYS reaction code [17] and different global predictions for the γ -ray strength function. These include three different models, namely, the Lorentzian model of [18,19], the generalized E -dependent Lorentzian model of [20], and the Skyrme Hartree-Fock-Bogoliubov (HFB) plus QRPA model of [4]. Note that in the case of the Lorentzian-type models, the $E1$ resonance energy, width and strength at maximum are all deduced from the photoabsorption data [14]. In contrast, these quantities are predicted by the global HFB-QRPA model, and in this case, some deviations in the resonance properties could arise since no renormalization is performed. On top of such an $E1$ description, the contribution of the $M1$ strength is included on the basis of the usual global systematics defined in [19,21], i.e., a Lorentzian function of centroid energy $E_{M1} = 41A^{-1/3}$ MeV, width $\Gamma = 4$ MeV, and strength normalized to $f_{M1} = 1.58 \cdot 10^{-9} A^{0.47}$ MeV $^{-3}$ at the reference energy of 7 MeV. On the basis of such γ -ray strength models, it can be seen in Fig. 1 that only the parametrized [19] Lorentzian model can decently reproduce the present experimental photoneutron cross section close to the neutron separation energy, and that both the more elaborated models fail.

A priori, the underestimate of the cross section around the neutron threshold seen in Fig. 1 could be due to a lack of some either $E1$ or $M1$ strength. In particular, a simple increase of the $E1$ strength, as properly simulated by the Lorentzian model, could solve the discrepancy. There is, however, some additional data that can help us constrain the low-energy tail of the γ -ray strength. It concerns the inverse radiative neutron capture available for ^{91}Zr . The keV-neutron capture cross section is sensitive to the γ -ray strength below the neutron separation energy. It is clearly seen in Figs. 1 and 2 that although the Lorentzian model reproduces well the $^{92}\text{Zr}(\gamma, n)^{91}\text{Zr}$ cross section (Fig. 1, middle panel), it overestimates significantly the $^{91}\text{Zr}(n, \gamma)^{92}\text{Zr}$ cross section. This means that the tail of the dipole strength cannot be artificially increased just to reproduce photoabsorption data. Note that the neutron

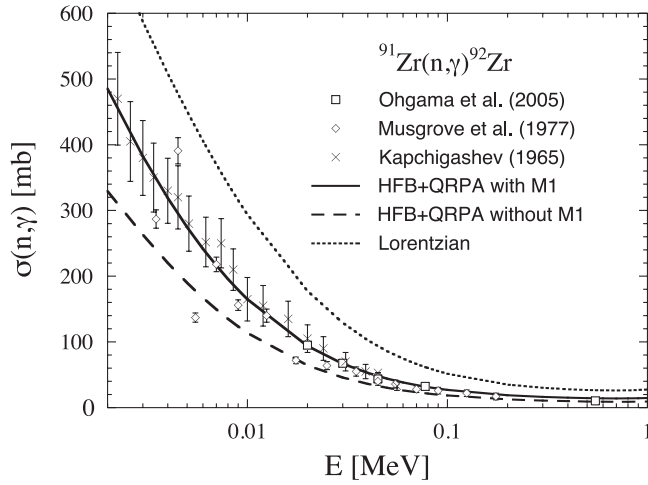


FIG. 2. Comparison of experimental [26–28] and theoretical radiative neutron capture cross section $^{91}\text{Zr}(n, \gamma)^{92}\text{Zr}$. The dotted line corresponds to the calculation based on the $E1$ and $M1$ Lorentzian model recommended in Ref. [19] and shown in Fig. 1. The solid (dashed) line corresponds to the calculation using the HFB + QRPA $E1$ strength with (without) an additional $M1$ contribution as explained in the text.

capture cross section is also relatively sensitive to the nuclear level density. The model adopted here is the HFB plus combinatorial model [22]. Experimental data exist to constrain the level density in $^{91,92}\text{Zr}$ around the neutron binding energy, namely, the s -wave resonance spacing [19], so that the uncertainties affecting the level density predictions were not found to change the above-mentioned conclusion.

Thus, no solution to both channels can be found by increasing globally the low-energy tail of the $E1$ strength. In contrast, a local increase of the dipole strength could provide an interesting solution or, more exactly, a confirmation of the presence of the strong $M1$ resonance already measured. Indeed, a broad giant resonance has been observed by inelastic scattering of 200 MeV protons from the different Zr isotopes [9]. This resonance located systematically around 9 MeV and of a FWHM approximately 1.5 MeV has been identified as being most probably an $M1$ giant resonance. For this reason, we have renormalized the above-mentioned $M1$ resonance considered in the TALYS calculation, taking the total strength as a free parameter, but with the centroid energy and width constrained by the scattering measurements, i.e., more precisely a peak energy at 9 MeV and a width of $\Gamma = 2.5$ MeV. Any additional $M1$ strength in the Lorentzian model [18] obviously would not cure the overestimate of the radiative neutron capture cross section. Adopting the generalized E -dependent Lorentzian [20], it is found that the $M1$ component could fill the gap seen in Fig. 1 (middle panel) around the threshold but not up to 13 MeV excitation energy (a wider $M1$ resonance would be needed), and even in that case, the (n, γ) cross section is also overestimated. With the $E1$ contribution of [20], the only solution found

for a coherent description of both channels would be to locate the $M1$ resonance at higher energy, i.e., around 10–11 MeV with a strength about 4 times the one expected from the systematics, i.e., a peak cross section of $\sigma_0^{M1} = 10$ mb. Such a high resonance centroid energy is, however, in conflict with the inelastic scattering data [9].

The best way we have found to coherently reproduce both the neutron capture and inverse photoneutron cross sections is to adopt the $E1$ strength from the HFB + QRPA model [4] and to add an $M1$ contribution of strength $\sigma_0^{M1} = 7$ mb with $\Gamma = 2.5$ MeV at 9 MeV in Lorentz shape, i.e., a peak cross section about 2.8 times larger and a width about twice smaller than those recommended by the systematics [19]. The integrated strength is consequently about 75% larger than the recommended one. The corresponding cross sections are compared in Figs. 2 and 3 (middle panel) with experimental data. As a comparison,

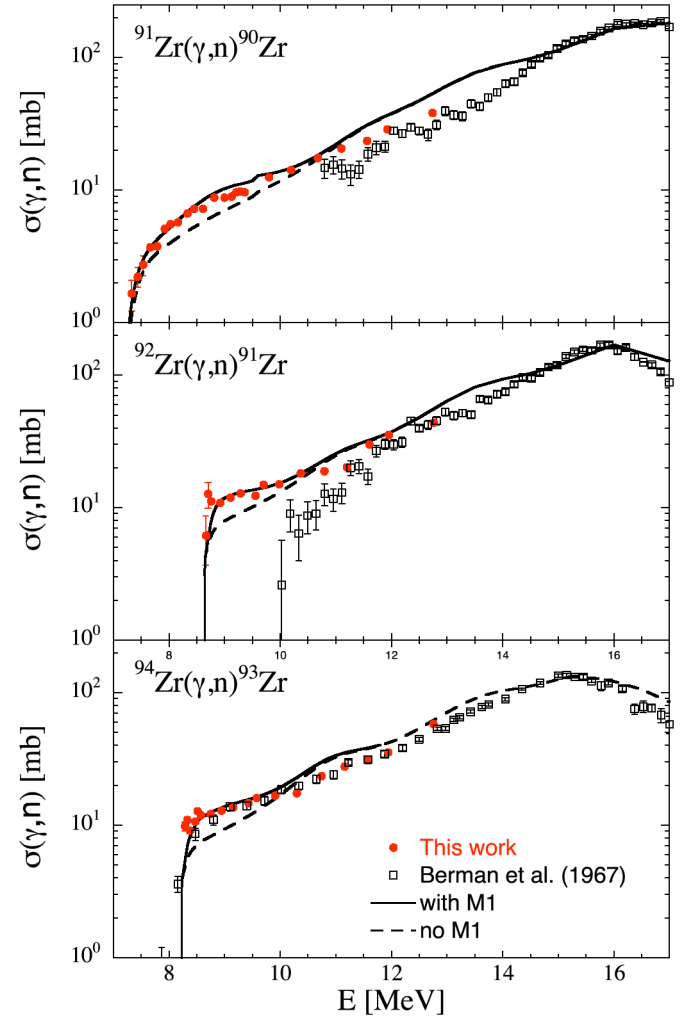


FIG. 3 (color online). Comparison of experimental and theoretical photoneutron capture cross section. In all panels, the solid line corresponds to the calculation with an $M1$ strength renormalized as explained in the text, and the dashed line without any contribution of the $M1$. In all cases, the $E1$ strength from the HFB + QRPA calculation of [4] is adopted.

we also show by the dashed line the cross sections obtained omitting any $M1$ contribution. In both cases, the $M1$ is seen to have an important impact on the cross section. In comparison with the generalized Lorentzian model, the HFB + QRPA model predicts more strength above some 11 MeV photon energy, so that a narrow $M1$ resonance can bring the missing magnetic-dipole γ strength function above the threshold in the photoreaction and at the same time below the threshold in radiative neutron capture (Figs. 2 and 3). Here again, this conclusion holds independently of the uncertainties affecting the nuclear level densities.

With such an $M1$ resonance strength tuned on the $^{92}\text{Zr}(\gamma, n)^{91}\text{Zr}$ reaction, the calculation of the photoneutron cross section was repeated for ^{91}Zr and ^{94}Zr . The results shown in Fig. 3 (adopting the $E1$ strength from the HFB + QRPA calculation of [4]) are seen to be in excellent agreement with the present experimental data, especially around the $M1$ centroid energy at 9 MeV. At higher energy, some deviations from the data of [14] are obtained, and these are due to some additional $E1$ strength predicted by the global HFB + QRPA model. The coherent description of the photoreactions for these three Zr isotopes is an additional confirmation of the presence of this strong $M1$ resonance.

We remark here that an equivalently good fit to the $^{92}\text{Zr}(\gamma, n)^{91}\text{Zr}$ and $^{91}\text{Zr}(n, \gamma)^{92}\text{Zr}$ cross section could have been obtained if we would assume a localized $E1$ resonance of strength $\sigma_0^{E1} = 3$ mb at 9 MeV with a width of 2.5 MeV. The best hint that the corresponding extra strength is of $M1$ origin comes not only from the inelastic scattering data [9], but also from a variety of theoretical calculations [23,24]. To confirm the presence of the dipole strength in the 9–10 MeV region, new HFB + QRPA calculations were performed for ^{92}Zr using another effective interaction, namely, the Gogny D1S force [25]. Like the Skyrme HFB + QRPA calculation, no extra low-lying isovector $E1$ strength is predicted below some 12 MeV. In contrast, a significant spin-flip $M1$ strength is found in ^{92}Zr at the energy of 10.2 MeV, exhausting about 70% of the total cross section, in agreement with the previous work done on ^{90}Zr [23,24]. A collective spin-flip neutron excitation from the $g_{9/2}$ to the $g_{7/2}$ shells may be the origin of the $M1$ strength for the zirconium isotopes. The spin parity ($5/2^+$) of the ground state in ^{91}Zr and ^{93}Zr shows that the $2d_{5/2}$ shell is occupied by the excess neutron(s), leaving the $g_{7/2}$ shell fully unoccupied for $^{91,92,94}\text{Zr}$. Thus, the same $M1$ strength can be explained for the three zirconium nuclei.

In conclusion, an extra γ strength was systematically identified for ^{91}Zr , ^{92}Zr , and ^{94}Zr in the photoneutron channel on top of the $E1$ γ strength function estimated by the HFB + QRPA model calculation. By attributing all the remaining strength to $M1$ (as suggested by QRPA calculations), we have identified the $M1$ resonance at $E_0 = 9$ MeV with a width $\Gamma = 2.5$ MeV in ^{92}Zr through a

coherent analysis of $^{92}\text{Zr}(\gamma, n)^{91}\text{Zr}$ and $^{91}\text{Zr}(n, \gamma)^{92}\text{Zr}$ cross sections. The total $M1$ strength required was about 75% larger than the strength predicted by the systematics. The photoneutron cross section data for ^{91}Zr and ^{94}Zr also exhibit the same $M1$ strength. The energy domain near neutron threshold constitutes a rich research field for investigating the γ strength function with a variety of multipolarity. Further experimental investigations leading to a direct detection of the $M1$ nature along with a thorough examination of the nuclear structure are desirable.

This work is supported by the Japan Private School Promotion Foundation and the Konan-ULB convention. S. G. acknowledges the FNRS support.

-
- [1] M. Arnould and S. Goriely, *Phys. Rep.* **384**, 1 (2003).
 - [2] H. Utsunomiya *et al.*, *Phys. Rev. C* **67**, 015807 (2003).
 - [3] S. Goriely and E. Khan, *Nucl. Phys.* **A706**, 217 (2002).
 - [4] S. Goriely, E. Khan, and M. Samyn, *Nucl. Phys.* **A739**, 331 (2004).
 - [5] H. Utsunomiya, P. Mohr, A. Zilges, and M. Rayet, *Nucl. Phys.* **A777**, 459 (2006).
 - [6] D. Meuer *et al.*, *Nucl. Phys.* **A349**, 309 (1980).
 - [7] N. Anantaraman *et al.*, *Phys. Rev. Lett.* **46**, 1318 (1981).
 - [8] F.E. Bertrand *et al.*, *Phys. Lett.* **103B**, 326 (1981).
 - [9] G.M. Crawley *et al.*, *Phys. Rev. C* **26**, 87 (1982).
 - [10] S.K. Nanda *et al.*, *Phys. Rev. Lett.* **51**, 1526 (1983).
 - [11] R.M. Laszewski, R. Alarcon, and S.D. Hoblit, *Phys. Rev. Lett.* **59**, 431 (1987).
 - [12] D.E. Bainum *et al.*, *Phys. Rev. Lett.* **44**, 1751 (1980).
 - [13] H. Utsunomiya *et al.*, *Phys. Rev. C* **74**, 025806 (2006).
 - [14] B.L. Berman *et al.*, *Phys. Rev.* **162**, 1098 (1967).
 - [15] E.P. Wigner, *Phys. Rev.* **73**, 1002 (1948).
 - [16] T. Shizuma *et al.*, *Phys. Rev. C* **72**, 025808 (2005).
 - [17] A.J. Koning, S. Hilaire, and M.C. Duijvestijn, in *Proceedings of the International Conference on Nuclear Data for Science and Technology*, edited by C. Haight *et al.* [AIP Conf. Proc. 769, 1154 (2005)].
 - [18] P. Axel, *Phys. Rev.* **126**, 671 (1962).
 - [19] T. Begya *et al.*, *Handbook for Calculations of Nuclear Reaction Data, RIPL-2* (IAEA-Tecdoc-1506, 2006).
 - [20] J. Kopecky and M. Uhl, *Phys. Rev. C* **41**, 1941 (1990).
 - [21] A.G. Bohr and B.R. Mottelson, *Nuclear Structure* (Benjamin, London, 1975), Vol. II, p. 636.
 - [22] S. Hilaire and S. Goriely, *Nucl. Phys.* **A779**, 63 (2006).
 - [23] A.I. Vdovin, V.V. Voronov, V. Yu Ponomarev, and C. Stoyanov, *Sov. J. Nucl. Phys.* **30**, 479 (1979).
 - [24] D. Cha, B. Schwesinger, J. Wambach, and J. Speth, *Nucl. Phys.* **A430**, 321 (1984).
 - [25] S. Péru and H. Goutte, *Phys. Rev. C* (to be published).
 - [26] K. Ohgama, M. Igashira, and T. Ohsaki, *J. Nucl. Sci. Technol.* **42**, 333 (2005).
 - [27] A.R. Del Musgrove, J.W. Boldemann, B.J. Allen, J.A. Harvey, and R.L. Macklin, *Aust. J. Phys.* **30**, 391 (1977).
 - [28] S.P. Kapchigashev, *At. Energ.* **19**, 294 (1965).