Upper limit on the two-photon emission branch for the $0_2^+ \rightarrow 0_1^+$ transition in ^{98}Mo

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Background: Two-photon emission, while well known in atomic physics, is a rare second-order process in nuclear physics with only three cases where a two-photon branch is measured. The limited knowledge stems from the experimental difficulty in resolving two-photon emission from dominant single-photon emission, restricting practical cases for study to $0^+ \rightarrow 0^+$ ($E0$) transitions, since single-photon emission is forbidden. In practical terms, this limits the range of easily accessible cases to even-even nuclei with the unusual property of a first excited state with spin/parity of 0^+.
Purpose: Two-photon branches are measured for the closed-shell nuclei, ^{16}O, ^{40}Ca, and ^{90}Zr. The intention of the present work was to obtain data for a case which was not a closed-shell nucleus. Of the possible nuclei relevant to such a study, ^{98}Mo was chosen as its first-excited state is 0^+ and lies below 1 MeV, meaning that internal pair transitions are not allowed.
Method: The first excited state ($J^\pi = 0^+$) in ^{98}Mo was excited in resonant inelastic proton scattering using a 6.7-MeV proton beam. The population of the state was selected using an annular double-sided silicon strip detector (DSSD). The decay of the state by conversion electrons was observed using the same DSSD, while Gammasphere was used to detect possible two-photon events.
Results: An upper limit on the two-photon branch obtained was 1×10^{-4} at the 95% confidence level (CL).
Conclusions The upper limit obtained is smaller than any other previously obtained two-photon branch. Phase space considerations suggest that the actual value of the branching ratio in this case may be significantly smaller than the upper limit obtained.

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Two-photon emission is a common process in atomic physics, first discussed by Göppert-Mayer over seventy years ago [1]. Two photons are spontaneously emitted with energies which individually form a continuum distribution but whose sum corresponds to the transition energy. Aside from the intrinsic interest in this second-order process, it has also been suggested to contribute to the continuum radiation associated with planetary nebulae [2]. Two-photon emission was first discussed in a nuclear context by Oppenheimer and Schwinger in terms of the decay of an excited $J^\pi = 0^+$ state in ^{16}O [3]. In principle, such higher-order processes may compete with any first-order decay process such as single-photon emission or internal conversion. As a second-order quantum mechanical process, the branch should be around 10^{-4}, being proportional to α^2, where α is the fine-structure constant. This is not such a small branch but, in practice, the detection of such higher-order processes is extremely challenging as two distinct photons are difficult to distinguish from a single photon which has undergone Compton scattering. Accordingly, our knowledge of this process is limited. In fact, our knowledge is confined only to cases where the first-order photon transition is forbidden. This condition is satisfied in the case of a $0^+ \rightarrow 0^+$ transition as the intrinsic spin, $S = 1$, of the photon will not allow it to mediate a transition between two states with $J^\pi = 0^+$. Such a transition can only proceed by internal conversion, or in cases where the transition energy is above 1.022 MeV by internal pair production. This situation therefore constitutes an excellent field for searching for two-photon emission, or in principle, transitions mediated by two conversion electrons, or the combination of one photon and one conversion electron. Only the former process has been observed experimentally and specifically, in the special case of nuclei where the first excited state has $J^\pi = 0^+$. This is a highly unusual situation given that the first excited state in even-even nuclei nearly always has $J^\pi = 2^+$. In fact, the occurrence of a $J^\pi = 0^+$ first excited state may only arise where the underlying nuclear structure changes or where there are notably large shell closures.

Two-photon emission has been observed in the closed-shell nuclei ^{16}O, ^{40}Ca, and ^{90}Zr. In each case, the first excited 0^+ state was populated either by inelastic scattering or beta decay. Despite the very different decay energies and the strong variance in nuclear structure, the two-photon branching ratio $\Gamma_{\gamma\gamma}/\Gamma_{\rm tot}$ is found to vary by less than a factor of 4 (6.6×10^{-4} for ^{16}O [4], 4.5×10^{-4} for ^{40}Ca, and 1.8×10^{-4} [5] or 2.2×10^{-4} [6] for ^{90}Zr). Surprisingly, in all cases, the γ-ray angular correlation between the two photons was found to be asymmetric about 90 degrees. This was interpreted as evidence for an interference between $2E1$ and $2M1$ contributions to the transition, which were found to have similar order.

An approximation to the two-photon width is given by

$$\Gamma_{\gamma\gamma} = \omega_0^7 \left(\alpha_{E1}^2 + \chi^2 + \omega_0^4 \frac{\alpha_{E2}^2}{4752} \right), \quad (1)$$

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where ω_0 is the transition energy, χ is magnetic dipole transition susceptibility, and α_{E1} and α_{E2} are the electric dipole- and quadrupole-transition polarizabilities [4], respectively. Recoil corrections are neglected as they are expected to be small for heavy nuclei. In measured cases, it has been found that the $2E2$ contribution is negligible. The two-photon width is dominated by the phase-space factor, but the situation is somewhat more complicated. On simple phase-space considerations, for example, the two photon branch for ^{90}Zr should be more than 90 times less than that for ^{40}Ca, but in fact is only around 2 times less. The difference is attributable to the nuclear structure and comprises a coherent sum of contributions from $2E1$ and $2M1$ transitions, which probes the electric and magnetic properties of the nucleus.

Given the limited information on two-photon emission, it is necessary to find other examples, particularly in cases not concerning closed-shell nuclei and cases where deformation is believed to play a role. There are three other stable isotopes in which the first excited state is 0^+, namely ^{96}Zr, ^{72}Ge, and ^{98}Mo. Of these stable isotopes, the latter two are particularly favorable for a two-photon emission search from the experimental perspective since the relevant transition energies are below 1 MeV and so internal pair production is not allowed, meaning that the competition is only between internal conversion and two-photon emission. Of ^{72}Ge and ^{98}Mo, the latter was chosen as it is believed to be a good example of nuclear shape coexistence [7]. The transition connecting the excited 0^+ state and 0^+ ground state is strong $[\rho^2(E0) \times 10^{-3} = 27(5)]$ compared to $\rho^2(E0) \times 10^{-3} = 3.3(17)$ for ^{90}Zr [8]. In their review of $E0$ transitions, Wood *et al.* [8] describe the strong $E0$ transition as arising from coexistence of different shapes, i.e., spherical ground state and deformed excited state. From analysis of a Coulomb excitation of ^{98}Mo, Zielinska *et al.* [9] suggest that the ground state is triaxial and the excited state is prolate deformed, but unusually the magnitude of the deformation of both states is the same.

In the present work, the excited 0^+ state in ^{98}Mo was populated via inelastic proton scattering with a 6.7-MeV proton beam from the ATLAS accelerator at Argonne National Laboratory. The beam energy was chosen to coincide with a known strong resonance for excitation of the 0^+ state [10]. The beam was incident on a 3 mg/cm^2 thick ^{98}Mo target of 96.87% purity. The beam was delivered in bunches every 82.47 ns, derived from the intrinsic RF of the accelerator. The purpose of the beam pulsing was to exploit the 22-ns half-life of the excited 0^+ state to separate it cleanly from excitation of promptly decaying states.

Scattered protons were selected using an annular silicon detector of 500 μm thickness (the S2 type from Micron Semiconductor [11]) mounted at backwards angles and covering the angular range 138.67° to 164.6° in the laboratory frame. This detector could also serve to detect $E0$ conversion electrons from the decay of the excited 0^+ state, providing a direct and sensitive normalisation for the two-photon decay measurement. Gamma rays were detected using the Gammasphere array of 100 high-purity Compton-suppressed germanium detectors, affording an efficiency of 9% at 1.33 MeV. The trigger for the experiment was the detection of a particle in the annular Si detector.

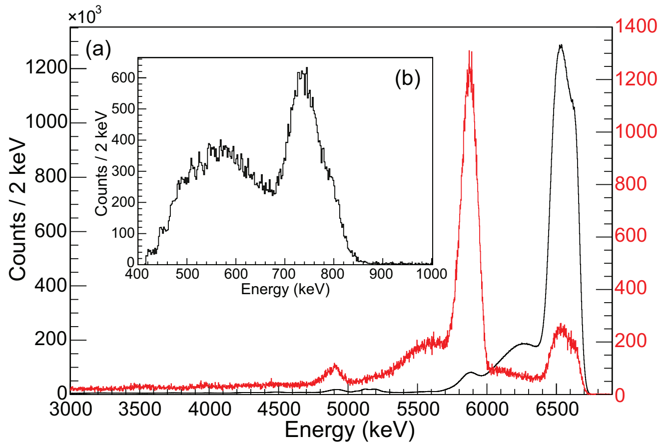

FIG. 1. (Color online) (a) Spectra from the double-sided silicon strip detector (DSSD). The black data correspond to the ungated proton spectrum which is dominated by a peak from elastic scattering. The red data are the same data but demand a coincident conversion electron; the spectrum is dominated by the peak corresponding to inelastic scattering to the excited 0^+ state. (b) The low-energy region of the DSSD spectrum corresponds to conversion electrons from the 0^+ state.

The silicon spectrum exhibits peaks from elastic and inelastic scattering to a number of excited states including, prominently, the first excited state (0^+) at 735 keV and the second excited state (2^+) at 787 keV [see Fig. 1(a)]. In addition, a peak is visible corresponding to the deexcitation of the first excited state by internal conversion [Fig. 1(b)]. The energy resolution of the silicon detector is not sufficient to completely resolve the excitation of the first and second excited states as they are only 50 keV apart. The peak corresponding to excitation of the 0^+ state is, however, clearly identified by requiring the coincident detection of a conversion electron (see Fig. 1).

Two separate approaches were pursued to search for the two-photon branch of the 735-keV 0^+ state. The first approach was to search for the decay following direct population of the 735-keV state via inelastic scattering. The second approach was to exploit the fact that the 735-keV state is also populated relatively strongly via a 1024-keV γ ray which depopulates the 2^+ state at 1758 keV; this state also being excited by inelastic scattering. Since the two approaches described are effectively simultaneous and independent measurements of the same phenomenon, they have been analyzed separately.

In the first approach, a gate was set on the proton spectrum with a width and centroid corresponding to that of the inelastic peak associated with the 0^+ state, as identified through gating on the conversion electron. The half-life of the 0^+ state is 21.8(9) ns, while the separation of beam bursts is 82.47 ns. Accordingly, the majority of the events of interest are located between the beam bursts. The analysis was made for events which were delayed with respect to the pulse, which had a width of 2 ns. To ensure that the selection of the time window excluded any contamination from the prompt beam pulse, a series of delayed time selections were made for the ranges 5.5–58 ns, 11–58 ns, and 22–58 ns. The 58-ns limit for the

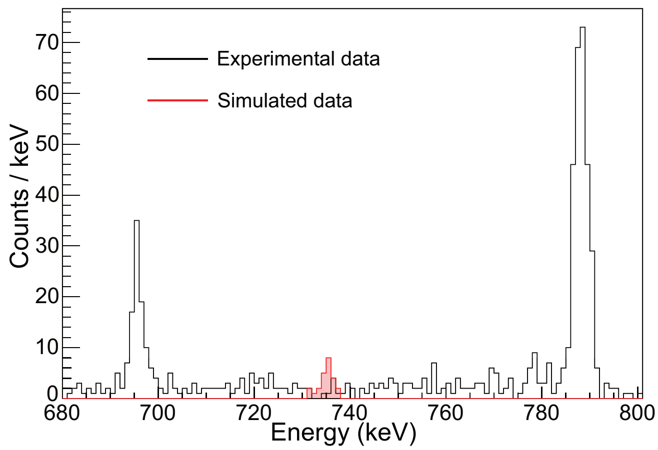

FIG. 2. (Color online) Black data: Gamma sum spectrum for twofold events which are delayed by $5.5 < t < 58$ ns with respect to the beam burst. Red data: Simulated Gaussian distribution corresponding to the lowest detected intensity that would be consistent with a branching ratio of 5×10^{-4} in the 95% confidence level (CL).

time windows was to ensure that there was no population from the subsequent beam pulse. In each case, twofold γ-ray events were examined where the γ rays were in prompt coincidence with each other but fell within the delayed time window with respect to the beam pulse.

Figure 2 shows the γ-ray sum spectrum for twofold events which are delayed by 5.5–58 ns with respect to the beam burst. In this spectrum, two peaks are visible. The one at 695 keV from coincident detection of the 160- and 535-keV γ rays from the isomeric decay of the 0^+ state in ^{100}Mo, and a weak 787-keV peak resulting from Compton scattering of the 787-keV $2^+ \rightarrow 0^+$ transition. The latter is presumably attributable to prompt production through the small dark current of protons between beam bursts. No clear evidence is seen for a sum peak corresponding to 735 keV, which would be expected for the two-photon decay of the 0^+ state in ^{98}Mo. This implies that the two-photon branch must be smaller than that obtained in the previous examples of ^{16}O, ^{40}Ca, and ^{90}Zr. A simulated peak is shown in red in Fig. 2 for a branching ratio of 5×10^{-4} and it is clear that such a peak, if it existed in the data, would be easily visible. The simulated peak was created using a randomized Gaussian from the ROOT software libraries [12], with a FWHM corresponding to that of the 787-keV peak. The intensity of the peak was calculated as being the lowest integer such that the data would lie within the 95% confidence limit of a branching ratio of 5×10^{-4}.

Since no obvious peak could be found corresponding to two-photon emission, a procedure was followed whereby the γ-ray sum spectra in the region between 720 and 750 keV was fitted for the different delayed time ranges considered (5.5–58 ns, 11–58 ns, and 22–58 ns) as shown in Fig. 3. A constant background was fitted over the energy range, with a variance of typically <10% and a Gaussian peak shape with centroid of 735 keV. An upper limit on the mean signal, μ, was extracted using the method outlined by Feldman and Cousins [13]. The extracted values were then normalized to account for efficiency. In the second approach, events were

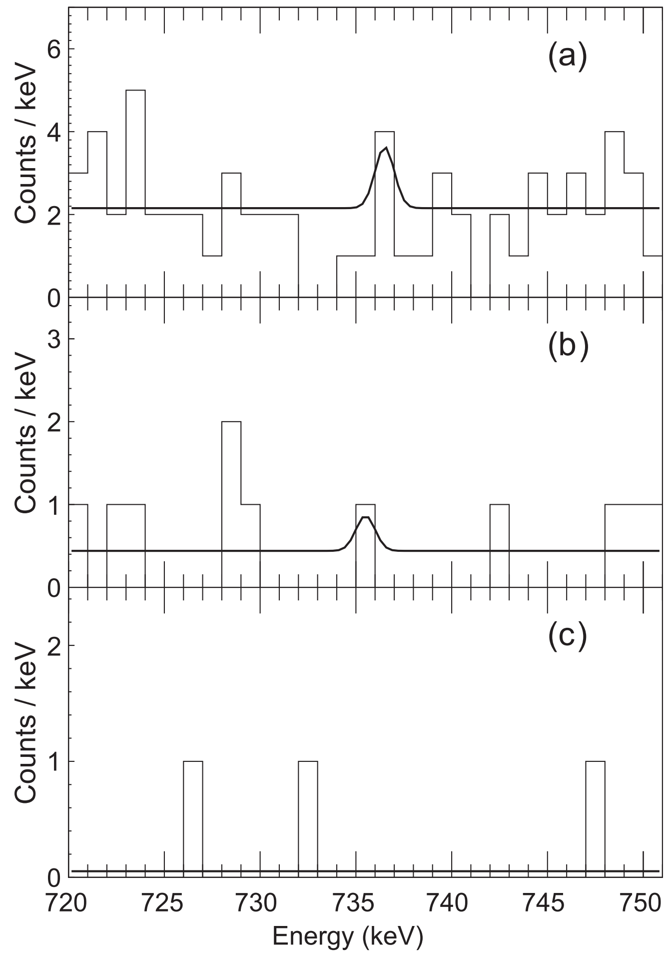

FIG. 3. γ-ray summed spectra for the following conditions: (a) is proton gated with a $5.5 < t < 58$ ns requirement, (b) is proton gated with a $20 < t < 58$ ns requirement and (c) is gated on, and in prompt coincidence with, the 1024-keV γ ray.

selected where a 1024-keV γ ray was detected promptly with the beam pulse. Again, a sum spectrum is produced [see Fig. 3(c)] and a maximum likelihood peak was obtained in the same manner as described previously.

In order to determine the two-photon branch, the upper limit on the observed number of events in each case was corrected both for the fraction of the decay curve of the 0^+ state which was sampled, and the efficiency for detecting the two-photon decay. The efficiency was assumed to be 4% based on a Gaussian distribution of individual energies, with the most probable energy distribution being an equal split of the energy between the two γ rays, folded with the efficiency of the Gammasphere array. Fortuitously, the Gammasphere efficiency is near maximum for an equal energy sharing. The number of conversion electrons in coincidence with the inelastic proton was also obtained. A simulation showed that only 30% of the conversion electrons are fully stopped in the DSSD. The procedure then was to fit the number of counts observed in the fully stopped peak and then to correct this for the fraction expected to be stopped in addition to the solid angle coverage of the DSSD. No correction for angular

TABLE I. Gating method: Gate used and time window after the trigger for which $\gamma\gamma$ events are accepted. $N_{\gamma\gamma\mathrm{det}}$: Signal above background for $\gamma\gamma$ events in the expected location of the 735-keV peak. $N_{\gamma\gamma\mathrm{corr}}$: Signal above background for $\gamma\gamma$ events, corrected for efficiencies and the time window used. $N_{e^-\mathrm{det}}$: Number of electrons completely stopped in the DSSD. $N_{e^-\mathrm{corr}}$: Number of electrons, corrected for efficiency. $\Gamma_{\gamma\gamma}/\Gamma$: The extracted branching ratio. All values are quoted to a 1σ confidence level.

Gating method	Time window	$N_{\gamma\gamma\mathrm{det}}$	$N_{\gamma\gamma\mathrm{corr}}$	$N_{e^-\mathrm{det}}$	$N_{e^-\mathrm{corr}}$	$\Gamma_{\gamma\gamma}/\Gamma$
0^+ proton	$5.5 < t < 58$ ns	$\leqslant 5.15$	$\leqslant 212$	$6.1(1) \times 10^4$	$2.0(1) \times 10^6$	$\leqslant 1.07 \times 10^{-4}$
0^+ proton	$11 < t < 58$ ns	$\leqslant 4.02$	$\leqslant 209$	$6.1(1) \times 10^4$	$2.0(1) \times 10^6$	$\leqslant 1.06 \times 10^{-4}$
0^+ proton	$22 < t < 58$ ns	$\leqslant 3.02$	$\leqslant 251$	$6.1(1) \times 10^4$	$2.0(1) \times 10^6$	$\leqslant 1.27 \times 10^{-4}$
1024-keV γ ray	prompt	$\leqslant 3.07$	$\leqslant 85$	$1.0(1) \times 10^4$	$3.3(1) \times 10^5$	$\leqslant 2.58 \times 10^{-4}$

distribution is made as the $E0$ conversion electron emission should be isotropic. The data obtained from the different analysis strategies are tabulated in Table I.

The upper limit on the branching ratio is extracted using the proton gating method is 1.07×10^{-4} for a 5.5–58 ns window. This upper limit corresponds to a confidence level of 39.4% in the case of the γ-gated data. Combining these two statistically independent measurements gives an upper limit on the branching ratio of 1.07×10^{-4} at the 97% (2.17σ) confidence level, corresponding to an upper limit of 1×10^{-4} at the 95% confidence level.

This decay branch is smaller than those previous observed in the decays of the first 0^+ excited states in ^{16}O, ^{40}Ca, and ^{90}Zr. A comparison to ^{90}Zr since it is the most similar measured case to the present example. In ^{98}Mo, ω_0 is 735 keV compared to 1761 keV in ^{90}Zr. On the assumption that α_{E1} and χ are unchanged, then $\Gamma_{\gamma\gamma}$ for ^{98}Mo would be 450 times smaller. However, Γ_{tot} should be about 3 times larger for ^{98}Mo as its half-life is about 3 times shorter than ^{90}Zr. This would imply $\Gamma_{\gamma\gamma}/\Gamma_{\mathrm{tot}}$ for ^{98}Mo of $\sim 10^{-7}$ which is significantly below the sensitivity of the present measurement. There are good grounds, however, to suspect that this is a significant underestimate; this arises from the deformed nature of ^{98}Mo compared to ^{90}Zr.

The contribution to Eq. (1) from the magnetic dipole transition susceptibility, χ, is a sum of paramagnetic and diamagnetic components, both of which might expected to be larger in ^{98}Mo than ^{90}Zr. The paramagnetic component is given by a sum over the contributions of individual states [14]:

$$\chi_P^{12} = -\frac{4}{9}\pi \times 2 \sum_n \frac{\langle 0_1^+ || M(M1) || 1_n^+ \rangle \langle 1_n^+ || M(M1) || 0_2^+ \rangle}{E_n - \frac{1}{2}\Delta E_{12}}. \tag{2}$$

The lower the energy of a given state, the larger its contribution is to the sum. In this respect, it is notable that there is significant $M1$ strength in ^{98}Mo as low as 3–4 MeV, with cumulative $M1$ strength of $B(M1)\uparrow$ of $0.8\mu_N^2$ by 3.8 MeV [15]. In ^{90}Zr, this level of cumulative $M1$ strength is not observed until above 8 MeV [16]. In fact, the sum strength is $4.17(56)\mu_N^2$ with centroid of 9 MeV. The summed $M1$ strength should be similar for deformed ^{98}Mo but, clearly, significant parts of it are shifted to much lower energies.

The diamagnetic part can be approximated by [4]

$$\chi_D^{12} = -\frac{e^2}{6m}\langle 0_1^+ | \bar{r}^2 | 0_2^+ \rangle. \tag{3}$$

Given that $\rho^2(E0)$ is known to be around 10 times larger for ^{98}Mo than ^{90}Zr, then χ_D^{12} should be around 3–4 times larger for ^{98}Mo.

The contribution from the electric-dipole transition polarizability, α_{E1}^{12}, is somewhat harder to evaluate. Formally, it is given by [14]

$$\alpha_{E1}^{12} = \frac{4}{9}\pi \times 2 \sum_n \frac{\langle 0_1^+ || iM(E1) || 1_n^- \rangle \langle 1_n^- || iM(E1) || 0_2^+ \rangle}{E_n - \frac{1}{2}\Delta E_{12}}. \tag{4}$$

Kramp et al. [4] show that this transition polarizability is typically one to two orders of magnitude smaller than the electric-dipole polarizability, α_{E1}^{11}, but the evaluation of α_{E1}^{12} is not straightforward. Bertsch [17] and Kramp et al. [4] show schematically how to attempt this as a mixing betwen a spherical ground state and deformed excited state. A detailed calculation for ^{16}O is presented by Hayes et al. [18]. The mixing of the two 0^+ states in ^{98}Mo has been inferred by Rusev et al. [19] by populating excited 1^+ states around 3–4 MeV through inelastic photon scattering and observing the branching ratio to the two lowest 0^+ states. The nuclear structure of ^{98}Mo is, however, considerably more complicated than the closed-shell nuclei where two-photon emission was earlier observed; both the relevant states appear deformed and the ground state is suggested to be triaxial [9]. Naively, these strong admixtures of multiparticle multihole states might enhance α_{E1}^{12} compared to ^{90}Zr, but a full calculation is beyond the scope of the present work.

In conclusion, a search for two-photon emission for the $0_2^+ \to 0_1^+$ transition in ^{98}Mo has led to the establishment of an upper limit for the branch of 1×10^{-4} at the 95% CL. If ^{98}Mo had the same structure as ^{90}Zr in all respects other than the energy difference between the 0^+ states, then the branch would be expected to be around 10^{-7}. The presence of deformation and/or shape coexistence in ^{98}Mo is likely to enhance this estimate by one or two orders of magnitude. Further theoretical work is warranted to better predict the branching ratio. On the experimental side, it would be difficult to enhance the sensitivity of the present measurement. The most obvious avenue would be to improve the γ-ray detection efficiency. This would make a good case for study with a γ-ray tracking array or with an array of next generation scintillators such as lanthanum bromide. Ideally, angular distribution measurements should be used to disentangle the $2E1$ and $2M1$ components.

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[1] M. Göppert-Mayer, Ann. Phys. (Leipzig) **401**, 273 (1931).
[2] L. Spitzer, Jr. and J. L. Greenstein, Astrophys J. **114**, 407 (1951).
[3] J. R. Oppenheimer and J. Schwinger, Phys. Rev. **56**, 1066 (1939).
[4] J. Kramp, D. Habs, R. Kroth, M. Music, J. Schirmer, D. Schwalm, and C. Broude, Nucl. Phys. A **474**, 412 (1987).
[5] J. Schirmer, D. Habs, R. Kroth, N. Kwong, D. Schwalm, M. Zirnbauer, and C. Broude, Phys. Rev. Lett. **53**, 1897 (1984).
[6] E. R. Mucciolo and O. Helene, Phys. Rev. C **40**, 2403 (1989).
[7] T. Thomas *et al.*, Phys. Rev. C **88**, 044305 (2013).
[8] J. L. Wood, E. F. Zganjar, C. de Coster, and K. Heyde, Nucl. Phys. A **651**, 323 (1999).
[9] M. Zielińska *et al.*, Acta. Phys. Pol. B **33**, 515 (2002).
[10] W. J. Courtney and C. F. Moore, Phys. Lett. B **31**, 131 (1970).
[11] Micron Semiconductor Catalogue (2013).
[12] R. Brun and F. Rademakers, Nucl. Instrum. Methods A **389**, 81 (1997).
[13] G. J. Feldman and R. D. Cousins, Phys. Rev. D **57**, 3873 (1998).
[14] D. Habs, J. Schirmer, R. Kroth, N. Kwong, D. Schwalm, M. Zirnbauer, and C. Broude, Ann. Isr. Phys. Soc. **7**, 245 (1984).
[15] G. Rusev *et al.*, Phys. Rev. C **73**, 044308 (2006).
[16] G. Rusev *et al.*, Phys. Rev. Lett. **110**, 022503 (2013).
[17] G. F. Bertsch, Part. Nucl. **4**, 237 (1972).
[18] A. C. Hayes, J. L. Friar, and D. Strottman, Phys. Rev. C **41**, 1727 (1990).
[19] G. Rusev *et al.*, Phys. Rev. Lett. **95**, 062501 (2005).