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Improved measurement of the $0_2^+ \rightarrow 0_1^+$ $E0$ transition strength for ^{72}Se using the SPICE spectrometer

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The selenium isotopes lie at the heart of a tumultuous region of the nuclear chart where shape coexistence effects grapple with neutron-proton pairing correlations, triaxiality, and the impending proton drip line. In this work, a study of ^{72}Se by internal conversion electron and γ -ray spectroscopy was undertaken with the SPICE and TIGRESS arrays. New measurements of the branching ratio and lifetime of the 0_2^+ state were performed, yielding a determination of $\rho^2(E0; 0_2^+ \rightarrow 0_1^+) = 29(3)$ milliumits. Two-state mixing calculations were performed that highlighted the importance of interpretation of such $E0$ strength values in the context of shape coexistence.

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I. INTRODUCTION

The nucleus ^{72}Se sits in the heart of a region of rapidly evolving nuclear shapes and shape coexistence, in which higher mass isotopes show prolate ground states while the more neutron-deficient nuclei are predicted to exhibit rarer oblate deformed ground states [1–7]. Predicting the point of this apparent shape transition provides an extremely sensitive test of nuclear models. It was recently confirmed by Coulomb excitation that the ground state of ^{72}Se is dominated by prolate

deformation [6], while the isotopic and isobaric even-even neighbors ^{70}Se [7] and ^{72}Kr [8,9] both show indications of oblate ground states. However, the situation is complicated by substantial mixing between coexisting configurations and the additional consideration of triaxiality, which has been clearly observed in ^{72}Ge [10] and ^{76}Se [11], the presence of which undermines simpler experimental interpretations of deformation. Only with sufficient experimental data to accurately disentangle the mixing of coexisting configurations can we comment on the underlying structure.

Due to quantum mechanical constraints, the nonsphericity of a 0^+ nuclear state cannot be directly observed. Deformation can be inferred, either from the $B(E2; 0^+ \rightarrow 2^+)$ reduced transition strength, or by measurement of the diagonal matrix element of the associated 2^+ state. However, this relies on the model assumption, typically the axially symmetric rotor model. Quadrupole deformation parameters may be determined by applications of model-independent sum rules [12]. Unfortunately these require the measurement of many linking matrix elements, which can in principal be obtained through low-energy Coulomb excitation, but is particularly experimentally challenging when dealing with excited 0^+ states. Due to the simplicity of the monopole operator, $\rho^2(E0; 0_i^+ \rightarrow 0_f^+)$ values can be directly equated to the difference in mean

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square charge radii and the degree of mixing [13], and do not rely on any assumption of axial symmetry.

In this paper, we report on new measurements of both the lifetime and branching ratio of the 0_2^+ state in ^{72}Se , which in turn yield new values for the $\rho^2(E0)$ between the 0_2^+ and ground state and the $B(E2)$ from the 0_2^+ state to the 2_1^+ state. The new values are consistent with previous measurements and indicate a high degree of state mixing.

II. EXPERIMENT

A beam of ^{36}Ar ions was produced by a microwave ion source [15] and delivered at 120 MeV (3.33 AMeV) by the TRIUMF-ISAC-II linear accelerator chain [16,17] to the TIGRESS spectrometer [18] with an average intensity of 1 pA. The beam was incident on a target of 0.5 mg/cm^2 natural calcium. The target was backed by a 0.2-mg/cm^2 gold “adhesive” layer and a 15.7-mg/cm^2 silver beam stopper. The upstream side of the target was sealed by a 0.3-mg/cm^2 layer of gold to prevent oxidization of the calcium during transfer to the target chamber. Additionally, the chamber was flushed with xenon gas prior to target mounting. Despite these precautions, contaminant reactions from the ^{36}Ar beam on oxygen were observable in the resultant data.

^{72}Se nuclei were produced in the fusion-evaporation reaction $^{40}\text{Ca}(^{36}\text{Ar}, 4p)^{72}\text{Se}$. Recoiling selenium nuclei were stopped in the silver backing with a stopping time of approximately 0.2 ps. ^{72}Se was also populated indirectly via β decay of ^{72}Br ($t_{1/2} = 78.6\text{ s}$ [19]) produced in the $^{40}\text{Ca}(^{36}\text{Ar}, 3pn)^{72}\text{Br}$ reaction, with a cross section approximately 10% that of the former reaction (PACE4). Evaporated protons were detected by a Micron S3 silicon detector of $140\text{ }\mu\text{m}$ thickness located downstream of the target to aid in selection of the reaction channel of interest.

Twelve of the TIGRESS high-purity germanium (HPGe) clover detectors were positioned around the target location to detect γ rays. Four clovers were located at 45° with respect to the beam axis and eight at 90° . Each clover was Compton suppressed and positioned at a target-to-detector distance of 14.5 cm in order to optimize the peak-to-total configuration of the TIGRESS spectrometer [18].

The Spectrometer for Internal Conversion Electrons (SPICE) [14,20–22] was used to detect internal conversion electrons. SPICE utilizes a 6.1-mm-thick lithium-drifted silicon [Si(Li)] detector located upstream from the reaction target, and shielded from direct sight by a photon shield. A magnetic lens formed of rare-earth permanent magnets collects and directs internal conversion electrons around the photon shield to the Si(Li) detector. A diagram of the setup is shown in Fig. 1.

The detector signals were processed by the TIGRESS data acquisition system [23]. Data were recorded to disk for every event in which a Si(Li) trigger was detected. Additional events were recorded for a γ - γ trigger, these events were down-scaled by four or eight, manually selected dependant on the data rate.

The absolute γ -ray efficiency of TIGRESS was obtained from standard radioactive sources of ^{133}Ba , ^{152}Eu , ^{207}Bi , and ^{56}Co .



FIG. 1. SPICE setup inside the TIGRESS array, with a single downstream recoil detector. The ^{36}Ar beam enters from the left and passes through openings in both the SPICE Si(Li) detector and photon shield before impinging on the silver-backed natural calcium target.

A. SPICE efficiency

As fusion-evaporation products were implanted into a thick target backing, additional straggling of electrons, especially at low energies, reduced the efficiency compared to that determined from using “open” offline radioactive sources, which have minimal scattering material covering the activity.

The relative in-beam electron detection efficiency curve of SPICE was obtained from a detailed GEANT4 simulation of the setup [20,24]. This curve was normalized to measurements of internal conversion coefficients (ICCs) for known transitions for which the calculated ICCs are reliable [25,26]. It was possible to use 24 γ -ray gated known $E2$ and $M1$ transitions from in-beam products $^{107,109}\text{Ag}$, ^{72}Se , ^{73}Br , and ^{70}Ge . Additional data points were determined from the ratio of $0_2^+ \rightarrow 0_1^+$ $E0$ ICEs to other ($E2$) transitions. In the case of ^{70}Ge a competing $0_2^+ \rightarrow 2_1^+$ transition allowed an easy comparison to the well-measured branching ratio. For ^{72}Ge , the 0_2^+ is the lowest excited state with no competing branch. However, ^{72}Ge was entirely populated in β decay, having two additional neutrons with respect to the compound nucleus ^{76}Sr .¹ Consequently the ratio of the $0_2^+ \rightarrow 0_1^+$ $E0$ peak, which has no competing

¹As natural calcium was used in the target, the 0.647% ^{42}Ca and 2.09% ^{44}Ca would allow the $^{42}\text{Ca}(^{36}\text{Ar}, 6p)^{72}\text{Ge}$ and $^{44}\text{Ca}(^{36}\text{Ar}, 4p\alpha)^{72}\text{Ge}$ population channels; however, temporal gating with the beam-RF confirmed these prompt contributions are imperceptible at the sensitivity level of this work.

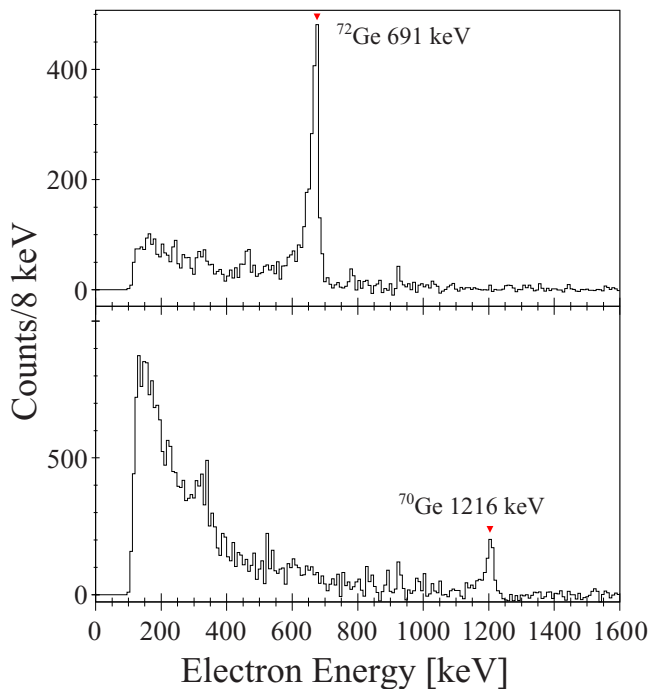


FIG. 2. SPICE data showing internal conversion electron peaks associated with $E0$ transition calibration points from ^{70}Ge (bottom) and ^{72}Ge (top). Each are selected in coincidence with feeding γ -ray transitions detected in TIGRESS.

branch, can be taken with respect to the $2_1^+ \rightarrow 0_1^+$ transition using the well-measured decay scheme of ^{72}As [19]. The associated electron peaks for the $^{70,72}\text{Ge}$ $E0$ calibration points are shown in Fig. 2.

Additional data were obtained from an offline ^{207}Bi source measurement; the source data points were adjusted for the change in position between the source and target, and the depth of implantation, using a GEANT4 simulation. A systematic uncertainty was included in each ^{207}Bi point, equal to 30% of the adjustment; this was found to be sufficient to ensure a reduced χ^2 of less than 1 between the ^{207}Bi points and the final efficiency curve. The validity of the simulation was further qualified by comparing the spectra from an unshielded ^{207}Bi source and that obtained when placing an aluminium foil of known thickness obscuring the source as a surrogate for implantation.

Finally, the measured points were spanned by the GEANT4 simulations to give the resultant efficiency curve and uncertainty shown in Fig. 3. The in-beam efficiency calculated is proportional to $F_{\gamma\gamma}$, the down-scaling factor of the $\gamma\gamma$ coincidence data. Due to changes to the down-scaling factor of the $\gamma\gamma$ trigger condition during the data collection $F_{\gamma\gamma}$ cannot be accurately determined; however, as the factor cancels out in experimental measurements the value is not required.

III. RESULTS

To determine precise values for the $\rho^2(E0)$ and $B(E2)$ strengths of transition depopulation, the 0_2^+ state in ^{72}Se , both a precise measurement of the state lifetime and of the transition branching ratio was performed.

Energy gated time difference spectra were produced for γ -ray transitions populating and internal conversion electrons depopulating the 937 keV 0_2^+ state. The internal conversion electron peak corresponding to the $E0$ transition to the ground state, shown in Fig. 4, provided an unambiguous gate leading

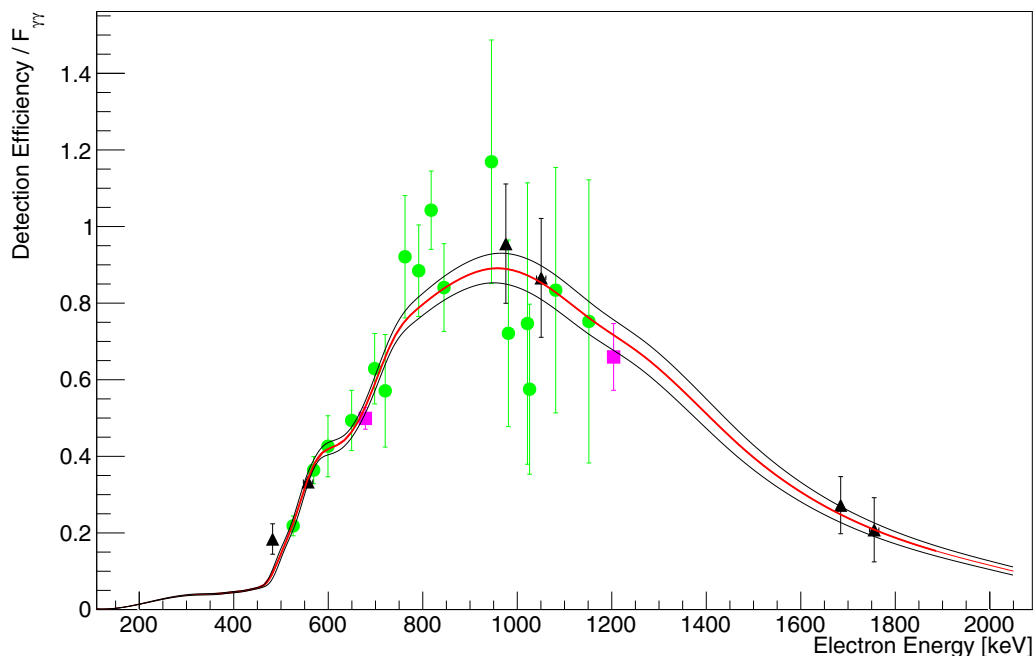


FIG. 3. Efficiency curve of the SPICE detector (red line) determined from GEANT4 simulations [14], scaled to in-beam $E2$ and $M1$ data (green circles), $E0$ data (pink squares), and offline ^{207}Bi data (black triangles). See text for details. One σ uncertainty bands resulting from the scaling (black lines) are shown.

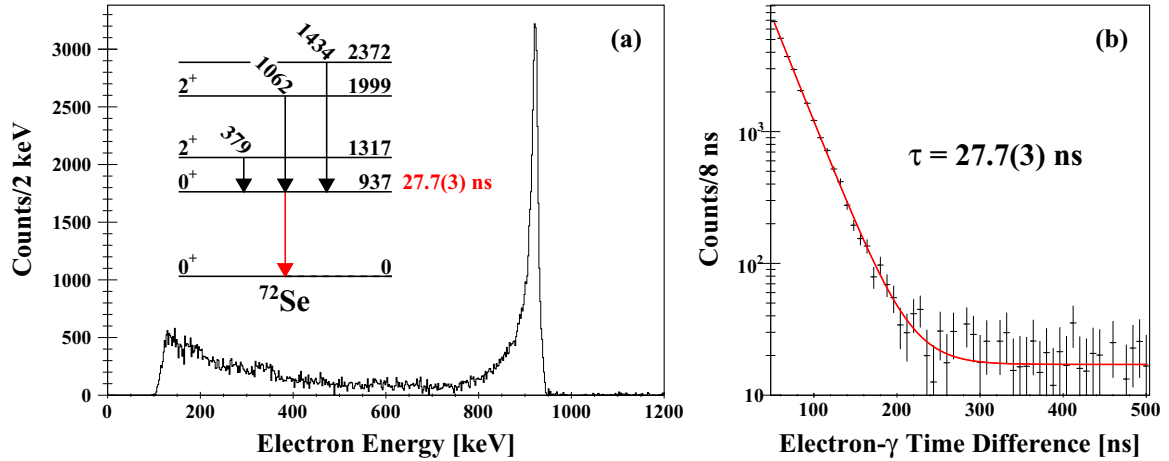


FIG. 4. (a) Electron spectrum produced from a sum of three γ -ray gates, 379, 1062, and 1434 keV, showing the $0_2^+ \rightarrow 0_1^+$ $E0$ transition of ^{72}Se cleanly isolated. (b) Electron- γ time difference spectrum between transitions populating and depopulating the 0_2^+ state in ^{72}Se , produced from a sum of the same three populating γ -ray gates as in panel (a). A fit to the data yields a lifetime measurement of 27.7(3) ns.

to a clean and distinct time difference lifetime spectrum. The internal conversion peaks of K-, L- and higher shell electrons for selenium cannot be resolved within the energy resolution of SPICE obtained in this beam time and a single energy gate was used.

Time differences were determined using the digital data acquisition system. Event times for γ rays detected in TIGRESS were determined online within the constant fraction discrimination algorithm of the TIG10 digitizer firmware. The γ -ray timing distribution suffered from significant tailing due to charge collection times. Wave forms from SPICE were recorded at 100 MHz and fit offline to determine accurate times for electron detection. This fitting procedure for SPICE ensured there was no tailing due to electron time measurement and subsequently the electron- γ timing distribution remained Gaussian on the side of the timing distribution on which the lifetime was to be measured. The combined timing resolution for electron- γ coincidence events was determined to be $\sigma_t \approx 14$ ns.

A sum of three different 0_2^+ state feeding γ -ray gates (379, 1062, 1434 keV) was used to produce the spectra shown in Fig. 4. The shape of the detector response for the HPGe-Si(Li) time difference was determined experimentally from prompt (\sim ps lifetime) transitions and incorporated into the fit.

To ensure the contribution of the energy-dependant γ -ray time distributions were well understood, time spectra were simulated for the individual γ -ray gates, based on the experimentally determined prompt time spectra and a variable lifetime. These simulated spectra were fit to the individually gated data and minimized to extract a lifetime. The results for each individual gate agreed with that of the combined fit.

A value of $\tau = 27.7(3)$ ns was determined for the lifetime of the 0_2^+ state. This agrees with the previous measurement [29] of $\tau = 27.8(6)$ ns. However, the value of $\tau = 22.8(14)$ ns reported in Ref. [30] is significantly discrepant and so the normalized residual method (NRM) was used to determine a weighted average [31,32]. A value of $\tau(0_2^+) = 27.7(4)$ ns is adopted.

In order to determine the $E0/E2$ branching ratio for depopulation of the 937-keV state, $\gamma\gamma$ and electron- γ data were selected by coincidence with the 379- and 1062-keV γ -ray feeding transitions. As the competing $0_2^+ \rightarrow 2_1^+$ $E2$ transition is only 75.06(20) keV [27], it could not be easily resolved in the data and the efficiency of the TIGRESS array at this energy is poorly constrained. Instead, the area of the subsequent 862-keV $2_1^+ \rightarrow 0_1^+$ transition was used, which due to direct feeding can be equated to the intensity of the depopulating $E2$ branch. Branch intensities are given by

$$I(E2) = \frac{N_\gamma(1 + \alpha)}{\epsilon_\gamma \epsilon_{\gamma\gamma} F_{\gamma\gamma} \Delta t_{\gamma\gamma}}, \quad (1)$$

where N_γ , α , and ϵ_γ are the measured counts, conversion coefficient, and γ -ray detection efficiency for the 862-keV transition, and by

$$I(E0) = \frac{N_e \Omega_{\text{Tot}}}{\epsilon_e \epsilon_{\gamma\gamma} F_{\gamma\gamma} \Delta t_{e\gamma} \Omega_e}, \quad (2)$$

where N_e , Ω , and ϵ_e are the measured counts, electronic factors, and electron detection efficiency for the $E0$ transitions. As previously discussed, the experimental electron efficiency $\epsilon_e = \epsilon_{\text{Abs}}/F_{\gamma\gamma}$. The efficiency for the gating transition, $\epsilon_{\gamma\gamma}$ and the $\gamma\gamma$ down-scaling factor, $F_{\gamma\gamma}$, need not be evaluated as these cancel when the ratio is taken with $I(E0)$. The coincidence time fraction Δt must be calculated as it is different for $\gamma\gamma$ and electron- γ coincidences.

Values of $I_{E0}/I_{E2} = 0.47(6)$ and $q_K^2(E0/E2) = 0.71(9)$ were determined, which may be related by [33]

$$q_K^2(E0/E2) = \frac{I(E0) \Omega_K (1 + \alpha)}{I(E2) \Omega_{\text{Tot}} \alpha_K}, \quad (3)$$

where the conversion coefficients are for the $0_2^+ \rightarrow 2_1^+$ transition. Values of $\alpha = 2.42(4)$ and $\alpha_K = 2.042(35)$ are taken from BrIccs v2.3d [26]. Electronic factors $\Omega_{\text{Tot}} = 3.46(17) \times 10^8$ and $\Omega_K = 3.09(15) \times 10^8$ are from the latest mass-corrected relativistic calculations [34,35].

Taking a weighted average of I_{E0}/I_{E2} with previously reported values 0.38(20) [36], 0.41(8) [27] and 0.32(4)

TABLE I. γ -ray branching ratios for the ^{72}Se 1317-keV 2_2^+ state, $B_{\gamma,\text{expt}}$, from the present work, alongside McCutchan *et al.* B_{McC} [27], Mukherjee *et al.* B_{Muk} [28], and the 2009 evaluation values B_{lit} [19].

$E_{\gamma,\text{expt}}$ (keV)	$B_{\gamma,\text{expt}}$	B_{McC}	B_{Muk}	B_{lit}
1316.5(4)	100(3)	100(5)	100(10)	100(6)
454.6(5)	79(2)	77(4)	167(20)	76(5)
379.3(2)	16(2)	15(1)	22(3)	35(2)

(converted from measured q_K^2) [37,38] yields a final value of $I_{E0}/I_{E2} = 0.38(4)$, with a reduced $\chi^2 = 1.70$ confirming reasonable agreement of all data.²

Combining the weighted branching ratio with the lifetime determined above, an $E0$ strength $\rho^2(E0; 2_2^+ \rightarrow 0_1^+) = 29(3)$ milliunits is extracted, using the relation

$$\rho^2(E0) = \frac{1}{\tau \left(1 + \frac{1}{I_{E0}/I_{E2}}\right) \Omega_{\text{Tot}}}. \quad (4)$$

This updates the previous value of 30(5) milliunits [13] with an increase in precision. A value of $B(E2; 0_2^+ \rightarrow 2_1^+) = 148(5)$ W.u. is also determined, which is in agreement with previous values.

In addition, branching ratios for γ rays depopulating the 1317-keV 2_2^+ state in ^{72}Se were measured. The values are shown in Table I. These are consistent with those reported by McCutchan *et al.* [27] confirming a smaller feeding of the 0_2^+ state than determined in previous literature evaluation [19]. It appears that in the evaluation the 455-keV branch intensity is taken from ^{72}Br ε decay, while the 379-keV branch is taken from in-beam reactions, as the 379-keV γ ray was multiply placed in the decay data. However, the 379-keV intensity from ε decay ($I_\gamma \leq 21$) agrees reasonably with the value reported here. A recent measurement by Mukherjee *et al.* [28] reports discrepant branching ratios, which are excluded from our discussion.

IV. DISCUSSION

Previous studies of ^{72}Se [27,59] suggested that the low-lying states of the nucleus can be partially described if one assumes there is a crossing of a rotational band built on a prolate 0_2^+ state, with states of a near-spherical vibrator band built on the ground state, in which mixing is limited to the 2^+ and 4^+ states. The remarkable agreement in the energy of the 0_2^+ with the yrast $J > 4$ states when fit with a polynomial of $I(I+1)$ reported in Ref. [59] would seem to agree with this characterization. However, this assignment is based on the assumption of minimal mixing between the 0^+ states, which is not supported by the moderate $\rho^2(E0)$ strength. When one introduces mixing of the 0^+ states, the unperturbed energy of the initially pure 0_{rotor}^+ will be reduced in energy, relative to

the observed 0_2^+ state and have poorer agreement with the described $I(I+1)$ fit.

In the previous works [27,59], small 0^+ mixing was suggested to explain the overprediction of the $2_2^+ \rightarrow 0_1^+$ strength. However, the authors wrongly conclude that this mixing would produce destructive interference in the $2_2^+ \rightarrow 0_1^+$ transition and constructive interference in the $2_1^+ \rightarrow 0_2^+$ transition. In the model used, the interference terms in these transitions would be of the same sign.

While $B(E2)$ values have received much attention, in the present work the somewhat neglected interpretation of $\rho^2(E0)$ strength is considered, along with the recently obtained experimental spectroscopic quadrupole moment $Q_s(2_1^+)$ [6]. Table II shows results of a two-state mixing calculation. This includes the spherical-prolate results from Ref. [27] alongside new calculations for coexisting rotor bands, which use the same basis as Refs. [27,59], taken from Ref. [60]. The ratio of unperturbed level spacing between the intrinsic rotor bands was given by the rigid-body moment of inertia as

$$\frac{E(2_a^+) - E(0_a^+)}{E(2_b^+) - E(0_b^+)} = \frac{1 + 0.315\beta_b + 0.249\beta_b^2}{1 + 0.315\beta_a + 0.249\beta_a^2}, \quad (5)$$

in which a negative value for β_a/β_b is adopted for an oblate shape of band a/b . With the exception of the imposed mixing matrix elements, the two structures are assumed to be independent and to have negligible interband matrix elements. Only mixing of 0^+ and 2^+ states was considered; constraints were not imposed based on high-lying yrast states.

Electric monopole transition strengths between mixed states are calculated as [61]

$$\rho^2(E0) = a_0^2 b_0^2 \left(\frac{3}{4\pi}\right)^2 Z^2 (\beta_a^2 - \beta_b^2)^2, \quad (6)$$

where a_0 and b_0 are the mixing coefficients of the 0^+ states. The intrinsic quadrupole moments Q_0 of the unmixed bands are taken as a function of β_2 under a sharp-surface approximation using

$$Q_0 = \frac{3}{\sqrt{5\pi}} Z R_0^2 \beta (1 + 0.16\beta). \quad (7)$$

The mixed state quadrupole moments were taken as

$$Q_0 = a^2 Q_a + b^2 Q_b, \quad (8)$$

under the stated independence assumption, i.e., $\langle 2_b | \hat{Q}_2^0 | 2_a \rangle \simeq 0$. This was related to the observed spectroscopic quadrupole moment by the usual axial-rotor expression

$$Q_s = \frac{3K^2 - I(I+1)}{(I+1)(2I+3)} Q_0. \quad (9)$$

Within the limitations of the twin-rotor model, in order to reproduce the significantly different $E(2^+) - E(0^+)$ energy spacing of the observed bands, a significantly greater mixing of the 0^+ than 2^+ states is required. A prolate-oblate shape coexistence description, with equal 0^+ and 2^+ mixing matrix elements, can only be achieved with unphysically large deformation parameters ($\beta > 1$). Reproducing the observed $\rho^2(E0)$ then strongly limits the magnitude difference of deformation of the two bands, per Eq. (6). Consequently, it is

²As the reduced χ^2 value (1.61) of the weighted average is less than the 95% critical value (2.60), the data are not deemed to be discrepant [39]; however, ‘‘external error,’’ inflated by $\sqrt{\chi_r^2}$, is used.

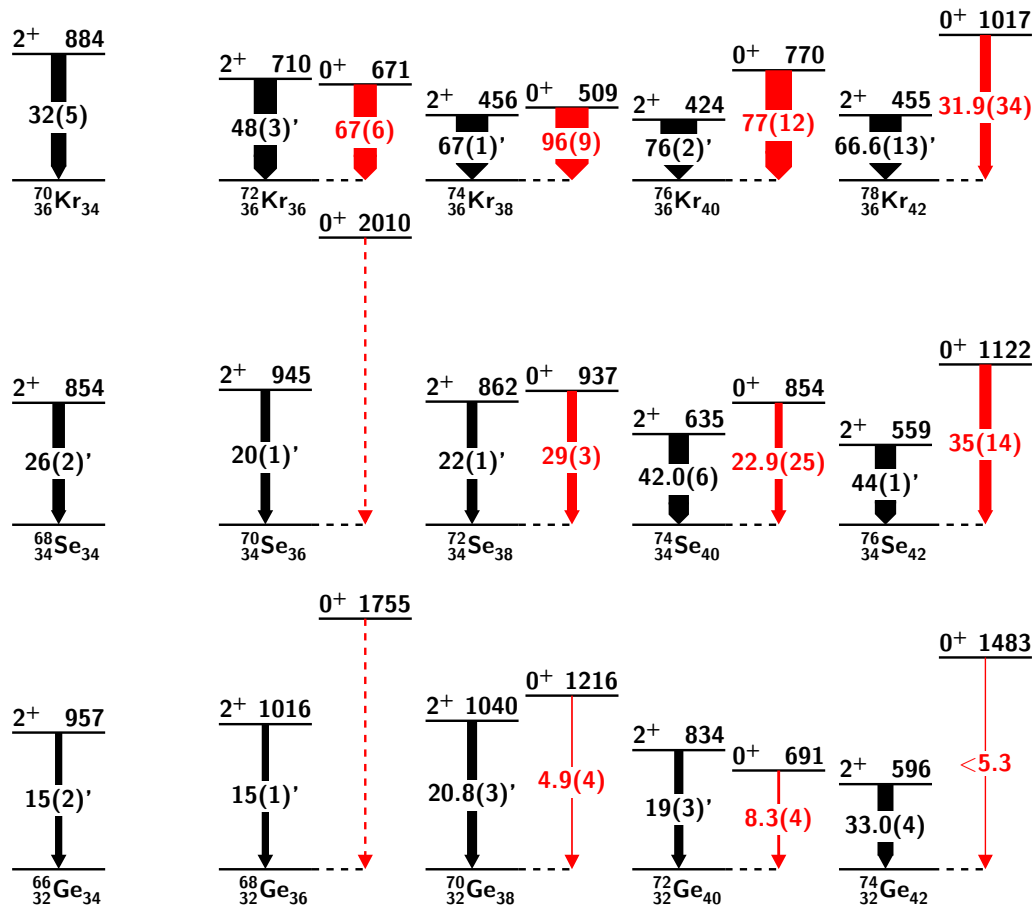


FIG. 6. Figure showing the lowest known excited 2^+ and 0^+ states in Kr, Se, and Ge isotopes. Known $B(E2)$ transition strengths in W.u. and $\rho^2(E0)$ values in milliu. are given and are indicated by the relative line width of transition arrows. A dashed line denotes a possible $E0$ transition which has not been directly observed. $\rho^2(E0)$ values are from Ref. [13], except ^{72}Se . $B(E2)$ values with apostrophes are weighted averages between published data tables [19,40–45] and more recent data [7,8,10,11,46–58] (not evaluated values).

great in the selenium isotopes if the magnitude difference of the coexisting shapes is smaller. To obtain a more complete picture of the structure of these nuclei requires the measurement of both $E0$ transition strength and quadrupole moments of other low-lying states.

Simple two-state mixing models fail to describe ^{72}Se adequately and there is significant evidence that any description based on mixing of distinct independent structures will not adequately capture the nature of the nucleus. Robust mean-field calculations which can construct states in a shared potential of multiple minima are called for.

V. CONCLUSION

Utilizing the SPICE and TIGRESS arrays, an independent measurement of the branching ratios and lifetime of the 0_2^+ state in ^{72}Se was performed by direct observation of internal conversion electron and γ rays. Combining these experimental observables, a new evaluation of the $E0$ and $E2$ strength between the 0_2^+ state and the ground state was performed. Values of $\rho^2(E0; 0_2^+ \rightarrow 0_1^+) = 29(3)$ milliu. and $B(E2; 0_2^+ \rightarrow 2_1^+) = 148(5)$ W.u. were determined,

improving on the precision of the previous values. Mixing axial-rotor model calculations were performed to explore the significance of the observed $\rho^2(E0)$ in combinations with the recently measured spectroscopic quadrupole moment. Within the confines of the model it was demonstrated that the deformation of the ground state band should be prolate in nature and that mixing between intrinsic 0^+ states in ^{72}Se may be significant and should not be neglected in calculations. However, it is concluded that independent two-state mixing does not describe the nucleus well overall.

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