Identification of the $\frac{9}{2}^+$ to $\frac{5}{2}^-$ transition in ⁶⁹Se

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Gamma rays from ⁶⁹Se have been investigated using the reaction ⁴⁰Ca(³²S,2pn γ)⁶⁹Se between 80 and 110 MeV beam energy. Isotopic identification was based on excitation functions, n- γ and n- γ - γ coincidences, as well as genetic relationships. An isomeric state ($T_{1/2} = 853 \pm 78$ ns) was found at 575.0 \pm 0.4 keV decaying to the first excited state which was located at 40.2 \pm 0.3 keV. The isomeric state lifetime reveals the same degree of inhibition as in analogous M2 ($\frac{9}{2}^+ \rightarrow \frac{5}{2}^-$) transitions and the excitation energy displays the systematic trend of the first $\frac{9}{2}^+$ state in the N = 35 and N = 37 isotones. The observed γ decay properties are consistent with $J^{\pi} = \frac{3}{2}^-$ for the ⁶⁹Se ground state, in agreement with the indications given by previous beta decay studies. A first level scheme, including four levels in ⁶⁹Se, is proposed.

The $T_z = \frac{1}{2}$ even-odd nuclei for $A \ge 32$ are known to be precursors of delayed proton emission and their decay properties have been studied up to ⁸⁵Mo. Recently the knowledge of excited levels for these nuclei has been extended to 65 Ge.¹ Around A = 70, the study of even-odd nuclei is important to understand this transitional mass region where rapid changes in the shell effects can be observed.² For N=33, 35, and 37 isotones close to stability a consistent pattern is found for the ordering of the low lying levels. The first states, of negative parity, illustrate the narrow spacing of the $2p\frac{1}{2}$, $1f\frac{5}{2}$, and $2p\frac{3}{2}$ subshells. The first positive parity states are identified either as the single particle state resulting from the odd neutron moving in the $\frac{9}{2}^+$ orbit or as configurations obtained by coupling the same single particle state to vibrational states in the neighboring even-even nuclei.^{3,4}

Prior to this work, information about ⁶⁹Se was given by the study of its β^+ decay ($T_{1/2}=27.4$ s) which has been investigated in detail by γ and delayed proton spectroscopy.⁵ In this paper, we describe an in-beam experiment on ⁶⁹Se designed to study the first excited states. A brief report has been given previously.⁶ Information obtained about an isomeric state is compared to the properties of the $\frac{9}{2}^+$ states in N=35 isotones and systematic trends are discussed.

I. EXPERIMENTAL METHODS AND RESULTS

Experiments were performed at the Centre de Recherches Nucléaires tandem Van de Graaff facility. In our earlier studies of the radioactive decay of ⁶⁹Se with the helium-jet recoil transfer technique,⁷ this nucleus was efficiently produced using the ⁴⁰Ca(³²S,2pn γ)⁶⁹Se reaction at about 100 MeV incident energy. The same reaction was chosen for the in-beam study. As the fusion cross section is spread over different masses corresponding mainly to charged particle evaporation, an efficient neutron detector was used to select the neutron reaction channels. The in-beam investigation of ⁶⁹Se consisted of (i) γ -ray excitation functions in coincidence with neutrons (ii) γ - γ -n coincidence measurements with special care to low energy transitions, and (iii) lifetime determinations using the delayed coincidence technique. In the first case, targets consisted of 200 μ g/cm² thick natural calcium supported on a tantalum backing while for the other experiments a layer of 600 μ g/cm² evaporated on a lead sheet (60 mg/cm^2) was mounted on the tantalum backing. Figure 1 shows a schematic view of the target-detector geometry. A Ge(Li) detector (G1) of 2.7% efficiency [relative to a 7.6 cm \times 7.6 cm NaI(T1) detector] and with a resolution of 2 keV at 1332 keV was positioned at 135° to the beam axis for the detection of low energy gamma rays. A Ge(Li) γ -ray detector (G2) of 22% relative efficiency and a resolution of 2.6 keV was also placed close to the target at 90° to the beam axis. For energy and efficiency calibrations, ¹³³Ba, ¹⁸²Ta, and ⁵⁶Co sources were used. Neutrons were detected in a large scintillation counter which consists of six identical and independent cells filled with NE213. Each cell has a polyhedral shape, the distance between two edges of the hexagonal section being equal to the thickness of the cell (16 cm). In this experiment, the total active volume for neutron detection was 22.5 l. The distance between the center of each cell and the target was about 16 cm. A 1 cm thick lead wall is set in front of the neutron counters and also between two adjacent ones. Each cell is viewed by an XP 2040 phototube. The neutron events were identified by pulse shape discrimination. Data for events in which at least one neutron detector was fired in fast ($\Delta t < 100$ ns) or delayed ($\Delta t < 2000$ ns) coincidence with one (or two) gamma counter(s), were recorded. The neutron multiplicity (M) was also measured.

As negative parity levels, resulting from the near degeneracy of f and p states, are expected to be located at low energy in ⁶⁹Se like in the other N=35 isotones, an additional experiment has been made for the observation, with optimum conditions, of low energy lines between 8 and 800 keV. A target chamber with thin walls was used and an intrinsic Ge low energy photon spectrometer (G3) took the place of the γ detector G1. In this case, the G2 and G3 counters were placed at equal



FIG. 1. Schematic diagram of the experimental setup.

angles (135°) symmetrically to the beam axis. For the G3 detector, the resolution was 2.1 keV at 122 keV. A copper absorber was set in front of G3 to reduce the intense x-ray yield ($E_x \leq 9$ keV) induced in the target by the ³²S beam. One element of the neutron counter, shielded by 1 cm lead, was located 5 cm downstream from the target at 0°. To study the low energy gamma rays, gamma direct and gamma-n spectra, as well as prompt and delayed $\gamma \cdot \gamma$ coincidences, were registered.

A. Excitation functions and relative yields

Direct and γ -n spectra have been recorded with $E(^{32}S) = 83$ to 110 MeV by 3 MeV steps, the energy loss in the target being about 2 MeV at 100 MeV. The relative normalization is obtained from the integrated beam charge at each energy. Singles γ -ray spectra are found to be dominated by lines assigned to nuclei produced in the xp channels, $^{69}As(3p)$ and $^{68}Ge(4p)$, and in the $2p\alpha$ channel, ^{66}Ge . The γ -ray excitation functions are qualitatively consistent with predictions of the statistical model computer code ALICE.⁸ In particular the yields for the 3p and 4p outgoing channels peak at an energy within 3 MeV of the predicted value.

With the neutron coincidence requirement, the intensity of the lines of ⁶⁹As and ⁶⁸Ge in the n-gated spectra indicate that the breakthrough of non-neutron events is at the 0.8% level. The spectra are then dominated by the 3p1n channel and the comparison of the ⁶⁸As lines in direct and n-gated spectra yields a neutron detection efficiency of ~30%.

The excitation function for ⁶⁸As (1n3p channel), deduced from the observation of the 158 keV γ ray, peaks at 110 MeV in good agreement with the predicted value [Fig. 2(a)]. Using the n-gated spectra, we find similar shapes, peaking around 98 MeV, for the relative yield of a number of transitions. An example is shown in Fig. 2(b) which displays the yield as a function of bombarding energy for the 676, 714, and 1079 keV γ rays, measured in the n-gated spectra and for the 535 keV line, observed in the singles. The coincidence measurements, described hereafter, have established that these transitions belong to the same level scheme. Furthermore, these lines are enhanced in the n-gated γ spectrum $(n \ge 1)$ with respect to the (0n) lines by a factor of about

20 [Fig. 2(c)] which corresponds to the factor achieved for known isotopes produced in the 1nxp channels (e.g., ⁶⁸As). From the 1nxp predictions, reported in Fig. 2(a), it appears that the 1n2p outgoing channel is the only one with a maximum yield under 100 MeV incident energy. Another possibility would be that the lines discussed before, result from α pn or α 2pn evaporation leading, respectively, to ⁶⁶As or to ⁶⁵Ge. These unidentified lines (535, 676, 714, and 1079 keV) cannot be assigned to ⁶⁵Ge as there is no overlap with the published level scheme.¹ It is more difficult to exclude ⁶⁶As as there is no information on the electromagnetic spectrum and as the maximum of the yield curve is predicted at 100 MeV incident energy. Nevertheless the cross section calculated with the statistical code is four times weaker for the α pn channel than for the 2pn one. Therefore the ⁶⁹Se is the most likely source of this group of γ rays.

The 2n evaporation leads to the unreported nuclei ⁶⁸Se (2n2p channel) and ⁶⁷As (2n3p channel). From the statistical model, the production yield is predicted to peak at 115 MeV (⁶⁸Se) and 130 MeV (⁶⁷As). Candidate transitions in ⁶⁸Se can be found in our γ spectrum taken in coincidence with the multiplicity M=2 neutrons and distinguished from ⁶⁹Se candidates. Unlike the latter, these transitions do not appear in the direct spectrum and therefore are not reported in Fig. 2(c).

In the experiment achieved for the low energy gamma identification, spectra taken with a 300 μ g/cm² thick ⁴⁰Ca target reveal a line ($E_{\gamma} = 40.2 \pm 0.3$ keV) strongly enhanced in the n-gated mode (Fig. 3) and displaying a similar excitation function as the other ⁶⁹Se candidates in the studied energy range.

In order to confirm the assignment of the lines to transitions between levels in ⁶⁹Se, we have measured the yield of γ rays subsequent to ⁶⁹Se beta decay. With a 200 μ g/cm² thick target, the intensity of the 98 keV line (deexcitation of the first excited state in the daughter nucleus ⁶⁹As) (Ref. 9) is found to peak around 96 MeV bombarding energy in good agreement with the excitation function for the candidate transitions in ⁶⁹Se. The ratio between the intensity of the ⁶⁹Se beta decay γ rays and the intensity of the 535 keV line has been measured using a chopped beam. The 535 keV intensity has been determined during the in-beam time interval (175 s) and the 98 keV in the following off-beam period (100 s). The ratio between the intensity of the 535 keV line and the beta decay strength is found equal to 0.43 ± 0.07 . The factor of 2 larger cross section observed for the ⁶⁹Se activity suggests that other decay modes than that involving the 535 keV transition occur in the deexcitation of the ⁶⁹Se yrast levels.

We thus conclude that the different lines which are found in coincidence in the γ - γ -n measurement, enhanced by a factor corresponding to the 1n channel in the n- γ spectrum and with a relative yield peaking around 98 MeV in the fusion reaction originate from transitions in ⁶⁹Se.

B. Coincidence measurements

Neutron- γ - γ coincidence measurements were performed at $E(^{32}S) = 103$ MeV with the 600 μ g/cm² thick

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target. Gamma (G1 and G2) and neutron counters were located as described in Sec. I. A total of about $1\,800\,000$ events was registered. In the analysis of the prompt and delayed events, gates were set on the lines selected by the

 $n-\gamma$ excitation curve. The dispersion of the time spectrum was 0.26 ns, in the prompt, and 5 ns per channel, in the delayed, measurement. An example of the results obtained with the delayed coincidences is shown in



FIG. 2. (a) Calculated excitation functions for the (1nxp) exit channels. (b) Excitation function for the 535, 676, 714, and 1079 keV rays observed in the 40 Ca(32 S,2pn γ) 69 Se reaction. The experimental points are connected by lines to guide the eye. (c) Ratio of the yields of lines, observed in singles, to those of the lines observed in coincidence with one (or more) neutron detector(s).



FIG. 3. Partial low energy γ spectrum recorded with the intrinsic Ge photon spectrometer. (a) Direct spectrum; (b) gated by a liquid scintillator neutron counter (0° to beam). The energies of the γ -ray peaks are given in keV. The transitions without labels are either unidentified or known contaminants.

Fig. 4 which displays a portion of the energy spectrum measured at 135°, gated by the neutrons and by the transitions between 0.1 and 3.0 MeV detected at 90° in a 2 μ s time interval. The spectrum is found to be dominated by the 535 keV line which is the most intense γ ray which satisfies the 1n2p channel requirements. This en-

ergy is clearly separated from the 537 line (β decay of ⁶⁶Ge), present in the direct spectrum, but suppressed by the neutron coincidence.

In the experiment for search of low lying states, prompt ($\Delta t = 100$ ns) and delayed ($\Delta t = 2 \mu$ s) gammagamma coincidences were registered. From a careful



FIG. 4. Portion of a 4096-channel spectrum in delayed coincidences gated with neutrons and with γ rays between 0.1 and 3.0 MeV. The time window begins 130 ns after the prompt peak and is open during 2 μ s. In the inset, the decay curve of the 535 keV γ ray is reported, with the background subtracted. In that case, each channel corresponds to 60 ns.

analysis of the γ - γ data, evidence has been found for a 40 keV transition in delayed coincidence with the 535 keV line (Fig. 5). Taking into account the relative time conditions between the two γ counters (G2 and G3) operating in coincidence, the 40 keV transition is found to follow the 535 keV one. Therefore the lowest lying excited state in ⁶⁹Se is located at 40.2 ± 0.3 keV. The established $n-\gamma-\gamma$ coincidence relationships are summarized in Table I and a list of γ rays assigned to transitions in ⁶⁹Se is given in Table II. The relative intensities I_{γ} are in arbitrary units. They have been deduced from n-gated gamma spectra taken with the 135° Ge(Li) detector at 103 MeV incident energy. The intensity of the 535 keV transition in the n-coincidence measurement is reduced, due to the lifetime value. In that case the intensity is taken from the direct spectrum and corrected by the neutron efficiency (0.30) for comparison with the n-coincidence data. The error quoted for the intensity for the 535 keV transition results from the statistics and from the neutron efficiency factor. Although both dipole and quadrupole transitions should be present in the decay scheme, no corrections were applied for the γ -ray angular distribution for the data measured at 135°. The intensity of the 40 keV transition has been deduced from the n-gated spectrum taken at 103 MeV and normalized through the 535 keV line with the other intensities. The 57 keV line which can be distinguished from the 56.1



FIG. 5. Portion of $\gamma \cdot \gamma$ coincidence spectrum recorded at $E({}^{32}S) = 103$ MeV, gated by (a) the 535 keV transition in ${}^{69}Se$, (b) the corresponding background, and (c) the 535 keV line after background subtraction. The ${}^{66}Ga$ transition of 44 keV observed on backgrounds is due to the ${}^{66}Ge$ beta decay. The 40 keV assignment (${}^{60}Se$) is discussed in the text.

TABLE I. Coincidence data at 103 MeV. All these transitions are found enhanced in the γ -n spectrum.

E_{γ} (keV)	Coincident transitions (keV)
40 ^a	535
535ª	676,775,1079
676	535ª
715	1079
(772+775)	57,412,676
1079	715

^aDelayed coincidences.

keV γ ray assigned to ⁶⁸As (214.7 \rightarrow 158.6 keV) and from tantalum x rays on the basis of the coincidence relationships with the other candidates (Table I) has been attributed to ⁶⁹Se. As no coincidences have been found between the 56.8 keV line and the decay of the low lying states reported in Fig. 6, the 56.8 keV transition could not be placed in the level scheme.

From the analysis of the coincidence data, a simple level scheme has been constructed (Fig. 6). Owing to the number of levels populated in this reaction, there are several possible orderings of the γ transitions and thus of the level positions above 2 MeV. Thereby this first level scheme of ⁶⁹Se is limited to four excited levels, unambiguously established from our data.

C. Half-life measurements

The half-life of the isomeric state at 575 keV has been inferred from the delayed coincidences between the 135° (G1) and the 90° (G2) detectors. The timing of the delayed γ rays is taken relative to the other γ detector (G2) which gives the prompt signal, but for each delayed coincidence the detection of one neutron is required. To take into account the neutron time of flight, a 500 ns time window was set for γ (G2) and neutron coin-

TABLE II. Energy and relative intensity of γ rays assigned to transitions in ⁶⁹Se. Relative intensities, in arbitrary units, are deduced from γ spectra taken with the Ge(Li) detector at 135° in coincidence (excepted for the 40 keV and 535 keV γ rays) with the neutron counters surrounding the target (see text).

E_{γ} (keV)	I_{γ}	$E_i \; (\text{keV}) \rightarrow E_f \; (\text{keV})$
40.2±0.3	168.8±50.7	$40.2\pm0.3\rightarrow0$
56.8±0.5	< 10 ^a	
402.7±0.5	37.8±2.6	1653.8±0.4→1250.8±0.5
411.7±0.5	33.8±2.7	
534.8±0.3	257.5±17.0	575.0±0.4→ 40.2±0.3
675.8±0.3	123.1±6.7	1250.8±0.5→ 575.0±0.4
714.5±0.5	38.1±3.0	
772.1±0.7	73.6±5.6	
$775.2{\pm}0.7$	42.9±4.3	
1078.9±0.3	138.3 ± 7.8	1653.8±0.4→ 575.0±0.4

^aMultiplet with the ⁶⁸As 56.1 keV line and the intense Ta x rays. The intensity limit is estimated from the γ -n data obtained with the intrinsic Ge detector.



FIG. 6. Level scheme for ⁶⁹Se including the first excited states populated in the ⁴⁰Ca(³²S,2pn)⁶⁹Se reaction. The γ intensities are roughly proportional to the width of the arrows.

cidences. The decay curve spectrum was obtained with a gate on the 535 keV line (G1) and the n-gated spectrum for (G2), as all transitions feeding the 575 keV level have been found in the prompt coincidences. The rate of the delayed coincidences, summed up in time bins of 60 ns/channel is presented in the inset of Fig. 4. A weighted least squares fit of the data, after background subtraction, yields the value $T_{1/2} = 853 \pm 78$ ns.

A 854 keV γ ray is apparent in the portion of spectrum displayed in Fig. 4. This line is assigned to the transition $2160 \rightarrow 1306$ keV in ${}^{68}As.{}^{10}$ This isotope is produced in the experiment through the 3p1n channel, open at 103 MeV incident energy. The lifetime of the isomeric state at 2160 keV has been determined in the same $\gamma - \gamma$ n-gated experiment. No other isomeric state has been found, in our measurements, at higher excitation energy which would populate the level at 2160 keV. Therefore the time spectrum obtained with a gate on 854 keV (G1) and on all transitions (G2) feeding the 2160 keV level yields the half-life $T_{1/2} = 34.3 \pm 7.1$ ns in agreement with a recent measurement reported by Raghavan et al.,¹¹ who give $T_{1/2} = 37$ ns. For the 40 keV level, limits for the half-life, 100 ns $< T_{1/2} < 1 \mu$ s, can be deduced from the comparison of the 40 keV intensity in prompt and delayed coincidences between the 535 and 40 keV transitions.

II. DISCUSSION

The ground state of ⁶⁹Se, connected by allowed β^+ transitions to the low lying states of ⁶⁹As, is assumed to be of negative parity with a J value restricted to $(\frac{1}{2}, \frac{3}{2}, \frac{5}{2})$ from the analysis of the beta decay.⁵ Taking into account the known properties of all neighboring nuclei and shell model estimates, one expects $J = \frac{9}{2}$ for the first positive parity level of ⁶⁹Se. Production of ⁶⁹Se via a heavy ion reaction, such as ³²S + ⁴⁰Ca enhances the population of high spin states and provides favorable conditions for observing the $\frac{9}{2}^+$ state in the decay of the yrast levels.

Our experimental results have revealed an isomeric level at 575 keV in ⁶⁹Se. Our measured value for its lifetime can only be analyzed in terms of an M2 or M2/E3 transition (575 \rightarrow 40 keV). A pure M2 transition presents a strength of 0.048±0.005 W.u. (Weisskopf units). The most likely explanation for the isomeric transition is then the $\frac{9}{2}^+ \rightarrow \frac{5}{2}^-$ decay.

The level at $E_x = 40$ keV is identified as the first excited state in ⁶⁹Se $\hat{J}^{\pi} = \frac{5}{2}^{-}$. The range for the half-life (100 $ns \le T_{1/2} \le 1 \mu s$ indicates that either a quadrupole or a dipole transition $(40 \rightarrow 0)$ can occur. An upper limit of the internal conversion ratio has been deduced from the comparison of the 535 keV and 40 keV line intensities. This value ($\alpha \leq 1$) implies a dipole for the 40 \rightarrow 0 transition. As only negative parity states related to $p_{\frac{3}{2}}$, $p_{\frac{1}{2}}$, and $f_{\frac{5}{2}}$ orbits are observed at low energy in odd nuclei throughout this mass region, we expect no parity change between the low lying states and thus an M1 transition for the $40 \rightarrow 0$ decay. The corresponding spin and parity assignments for the ⁶⁹Se ground state, $J^{\pi} = \frac{3}{2}^{-}$, is in agreement with previous beta decay results⁵ which support $(\frac{1}{2}, \frac{3}{2}, \frac{5}{2})$ with a slight preference for $\frac{3}{2}$. The $\frac{1}{2}^{-}$ state which has not been observed in our experiment is expected to be only weakly populated as nonyrast.

The well-established pattern of similar transitions in neighboring nuclei reveals that the observed properties of ⁶⁹Se are consistent with the systematics for N=35 iso-



FIG. 7. Lower part: low-lying level systematics of the N=35 isotones. Dotted lines are drawn to guide the eye. The data are taken from ⁶³Ni, ⁶⁵Zn, ⁶⁷Ge (Ref. 13), ⁶⁹Se (present study), and ⁷¹Kr (Ref. 16). Upper part: effective M2 coupling constants $g^{\text{eff}}/g^{\text{s.p.}}$ for single quasiparticle transitions $(g\frac{9}{2} \rightarrow f\frac{5}{2})$. The data are taken from Ref. 17 (⁶³Ni), Ref. 18 (⁶⁵Zn), Ref. 12 (⁶⁷Zn), Ref. 3 (⁶⁷Ge), Ref. 9 (⁶⁹Ge), (⁶⁹Se, this work), Ref. 19 (⁷¹Ge, ⁷¹As), Ref. 20 (⁶⁹As), Ref. 21 (⁷³As,⁷⁷As), Ref. 22 (⁷⁵As), Ref. 23 (⁸¹Br), and Ref. 24 (⁸⁵Rb).

tones (Fig. 7). The survey of the N=37 isotones indicates the same trend for the level structure at low energy between ${}^{65}Ni$ and ${}^{71}Se$. The reduced M2 transition probability for ${}^{69}Se$ [$B(M2)=49\times10^{-3}$ W.u.] is close to the value of the corresponding transition in ${}^{67}Ge$ [B(M2) $=79\times10^{-3}$ W.u.].³ We note that the reduced transition probabilities for M=35 are within an order of magnitude equal to those for M2 ($\frac{9}{2}^+ \rightarrow \frac{5}{2}^-$) transitions in N=37: ${}^{67}Zn$ [$B(M2)=70\times10^{-3}$ W.u.] (Ref. 12) and ${}^{69}Ge$ [$B(M2)=66\times10^{-3}$ W.u.].⁹ It has been first pointed out by Murphy *et al.*¹³ that the *E*2 transition probabilities between the low lying negative parity states ($\frac{1}{2}^- \rightarrow \frac{5}{2}^-$) decreased by an order of magnitude with the addition of two neutrons in the cases of ${}^{63,65}Ni$, ${}^{65,67}Zn$, and ${}^{67,69}Ge$. With the available data, we do not note a similar inhibition for the M2 transitions when N increases form 35 to 37.

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The effective coupling constant $g^{\text{eff}}/g^{\text{s.p.}}$ can be calculated for the $g_2^9 \rightarrow f_2^5$ simple quasiparticle transitions, using the relations¹⁴

$$B(M2,J_1 \rightarrow J_2) = (3\mu_0/\pi) \langle r \rangle^2 (g^{\text{eff}})^2 \langle J_2 \frac{1}{2} 20 | J_1 \frac{1}{2} \rangle^2$$

where $\langle r \rangle \sim 0.75 r_0 A^{1/3}$ with $r_0 = 1.2$ fm and $g^{s.p.} = g_s - \frac{2}{3}g_1$.

The value obtained for ⁶⁹Se $(g^{\text{eff}}/g^{\text{s.p.}}=0.122\pm0.005)$ is strongly reduced with respect to the single particle estimates and comparable with previous determinations in the other odd isotopes (Fig. 7). Using a pairing model calculation, Ejiri and Shibata¹⁵ have explained this reduction as a uniform effect of core polarization, independent of the mass number.

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