Shape evolution in heavy sulfur isotopes and erosion of the $N=28$ shell closure

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The structure of neutron-rich ^{40,42,44}S nuclei has been investigated through in-beam γ-ray spectroscopy using the fragmentation reaction of a 60 MeV A ^{48}Ca beam on a thin Be target. E_γ, I_γ, $\gamma\gamma$-coincidence, and γ-ray angular distributions were measured for each produced fragment. The level schemes previously containing only a single γ transition were extended, and spin values were proposed for the new states. The experimental results were interpreted by use of microscopic collective-model and large-scale shell-model calculations. The results of the model calculations are consistent with each other, and give a reasonable description of the experimental results. Both models predict an erosion of the $N=28$ shell closure at $Z=16$ and suggest a deformed ground state for ^{40,42}S and a spherical-deformed mixed configuration for ^{44}S.

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I. INTRODUCTION

A major breakthrough in nuclear physics was the discovery of the shell structure in nuclei and the explanation of magic numbers in the framework of the shell model. Nuclei at shell closures have gaps in their single-particle energy of the order of 4–5 MeV and are classified as magic analogous to noble gas configurations in atoms. The magic numbers expected for a harmonic oscillator mean field potential (2,8,20,40,70 . . .) are perturbed in the nucleus by a strong spin-orbit interaction, which pushes the highest spin state out of a major oscillator shell towards the lower ones. As a result, new magic numbers such as 28, 50, and 82 do appear, while some of the harmonic oscillator magic numbers (e.g.,

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70) completely disappear. These magic numbers are well established close to the valley of stability.

In the case of neutron-rich sulfur isotopes, the spin-orbit interaction is expected to lower the $f_{7/2}$ neutron orbit just into the middle of the gap between the sd and fp oscillator shells, resulting in a magic number at $N=28$. Even a relatively small change in the shape of the mean field may push the $\nu f_{7/2}$ orbit up or down slightly and in this way may reduce the gap energy above or below the $f_{7/2}$ subshell. If the energy gap above the $f_{7/2}$ subshell is reduced to some extent, nuclei with 28 neutrons may become midshell nuclei with a possible deformed shape. In the case of heavy sulfur isotopes a weakening of the spin-orbit interaction is also expected [1], which may further reduce the $N=28$ neutron gap.

From β-decay studies of $N=28$ nuclei ($^{43}_{15}$P, $^{44}_{16}$S, and $^{45}_{17}$Cl) an onset of the quadrupole deformation below ^{48}Ca was suggested [2]. This idea was soon confirmed by the determination of $B(E2)$ values for the heavy sulfur isotopes using the Coulomb-excitation method, where a large deformation of $\beta_2 \sim 0.3$ was deduced for these nuclei [3,4]. In a recent measurement at GANIL a low-energy isomeric state was found in ^{43}S [5], with a lifetime suggesting a spherical-deformed shape coexistence in this nucleus. All these measurements suggest an erosion of the $N=28$ shell closure.

Large-scale shell-model calculations indicate deformed ground states for ^{40,42}S and a strongly correlated one for the closed-shell nucleus ^{44}S [4,6,7], indicating that the $N=28$ shell closure is somewhat eroded at a large neutron excess. According to these calculations, the increase of $B(E2)$ values in heavy sulfur isotopes may arise from an enhancement of the proton collectivity induced by the decrease of the $\pi d_{3/2}$-$\pi s_{1/2}$ energy difference [6,8].

Relativistic mean field calculations [5,9,10], as well as Hartree-Fock-Bogolyubov calculations with different Skyrme forces [11,12], and with Gogny interaction [13–15], predicted both prolate and oblate deformed minima in the potential-energy surface (PES) calculated for the $N=28$ ^{44}S isotope. Reinhard *et al.* [12] have shown that only the effective interactions predicting a low (2–3 MeV) $f_{7/2}$-$p_{3/2}$ energy difference result in well pronounced prolate and oblate deformations. Including the dynamical part of the collective Hamiltonian, nuclei with both prolate and oblate minima may have γ-soft, triaxial, or coexisting prolate and oblate shapes depending on the depths of the two minima [13]. Energies of higher-lying states, as well as their γ branching ratios, may provide information on which of the above possibilities have been realized.

The in-beam γ spectroscopy of fragmented projectilelike nuclei has been shown to be an efficient method for studying excited states in light-neutron-rich nuclei [16,17]. In the present case, we apply this method to explore the structure of neutron-rich nuclei close to the $N=28$ shell closure, and report on results obtained for the ^{40,42,44}S nuclei.

II. EXPERIMENTAL METHODS

A ^{9}Be target of 2.76 mg/cm^2 thickness was bombarded by a ^{48}Ca^{19+} beam of 60.3 MeV A energy and 15 en A intensity delivered by the accelerator complex at GANIL. The emerging fragments were detected and identified at the focal plane of the SPEG spectrometer operated in a dispersive mode. At the focal plane a plastic scintillator was used to determine the total energy of the fragments, while ionization and drift chambers provided information on their energy loss and position. The time of flight was derived from the timing signal of the plastic scintillator with respect to the cyclotron radio frequency. It was corrected for the position of the fragments in the focal plane of SPEG in order to obtain a better time resolution and subsequently a better fragment identification. γ rays, emitted in flight by the excited fragments, were detected in an array of 74 BaF_2 crystals and 3 segmented Ge clover detectors. The BaF_2 detectors were mounted symmetrically above and below the target at a mean distance of 16 cm to cover about 80% of the total solid angle giving a full energy peak efficiency of 25% at 1.33 MeV. The segmented clover Ge detectors with an efficiency of 0.4% at 1.33 MeV were located at 15 cm from the target at angles of 85°, 122°, and 136° with respect to the beam direction. The time signal from the plastic scintillator was used as the trigger. A total of about 2.5×10^7 fragments was detected in four days of beam time.

Due to the large fragment velocity ($v/c=0.34$), the γ spectra had to be corrected for Doppler shift. Doppler broadening, arising mainly from the finite solid angle of the detectors, was decreased by exploiting the segmentation of the clover detectors. Consequently, the full width at half maximum for a γ ray of 1577 keV energy was $\sim$35 keV. The analysis of the γ yields from fragments with known level schemes showed that the ratio of the total γ intensity feeding the ground state to the number of the fragments produced is nearly constant and independent of the reaction channel. The placement of transitions in level schemes using the BaF_2 coincidence spectra was found to be possible only for the strongest transitions of the most abundant fragments. For weaker transitions, an alternative method based on γ-ray multiplicity was used. It consists of the comparison of γ-ray spectra gated on low and high γ multiplicities, respectively. Transitions with increased intensities in the high multiplicity spectra are expected to be connected to the ground state through other transitions, while those stronger in the low multiplicity spectra are assigned to feed the ground state directly [18].

Nonisotropic angular distributions were observed for some of the strongest γ rays indicating an angular momentum orientation in the fragmentation reaction at an intermediate energy. Since the clover detectors were placed at three different angles, we could deduce γ-ray intensity ratios for two pairs of angles. These ratios were normalized to that of the 1577 keV stretched $E2$ transition of ^{46}Ar [3,19]. Transitions from all the fragments were analyzed and the obtained anisotropy ratios were found to form two groups. All known stretched $E2$ transitions cluster within one group, while some transitions in odd fragments have anisotropy ratios within a second group. We interpret this as a distinction between stretched quadrupole and stretched dipole transitions. The calculated theoretical values of the anisotropy ratios for stretched quadrupole and dipole transitions have been found

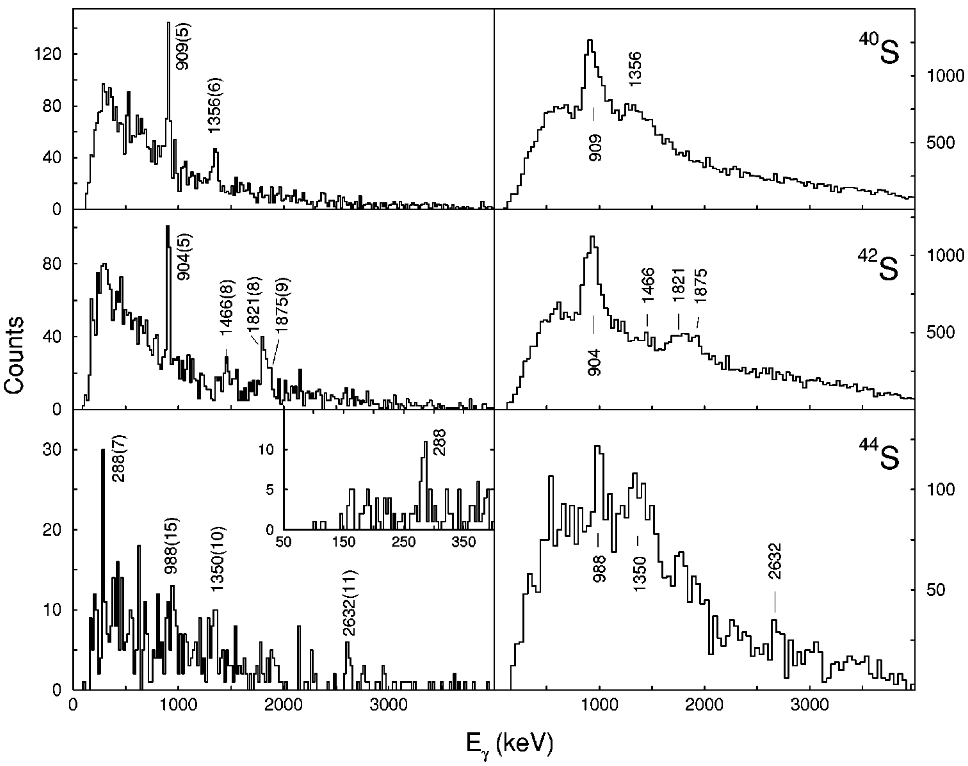

FIG. 1. Ge and BaF_2 γ-ray spectra of $^{40,42,44}S$ obtained by in-beam gamma spectroscopy using the fragmentation of a ^{48}Ca beam on a ^{9}Be target. The 288 keV transition of ^{44}S having a small width because of small Doppler broadening is also shown in a different binning as an inset in the Ge spectrum.

to be in agreement with the measured ones assuming an orientation of the order of 30%–70%.

III. RESULTS

A. The γ transitions

The γ-ray spectra obtained for $^{40,42,44}S$ are shown in Fig. 1. The spectra obtained from the Ge detectors are on the left hand side, and those from the BaF_2 detectors are shown on the right hand side. In order to confirm the presence of the low-energy 288 keV line, for which the Doppler broadening is small, the low-energy Ge spectrum of ^{44}S is also shown in an inset with a smaller binning. All but the 288 keV transition detected by the Ge detectors have their counterparts in the BaF_2 spectra. The 288 keV transition is missing in the BaF_2 spectra due to a higher-energy threshold.

We assigned two γ rays to ^{40}S, as shown in Fig. 1. The 909 keV transition has also been observed in the Coulomb-excitation experiment [3], and both transitions were seen in the β-decay study of ^{40}P [20].

In the γ-ray spectra of ^{42}S, four γ lines were observed. Only the 904 keV γ ray is previously known [3]. Both from linewidths and line shapes with different multiplicity conditions, it appears that a doublet of two transitions (1821 and 1875 keV) is involved in the γ decay of ^{42}S. A weaker γ ray with an energy of 1466 keV is also visible in the spectra of ^{42}S.

^{44}S is the most neutron-rich fragment observed in this experiment and consequently the corresponding γ-ray spectra have lower statistics. However, in addition to the previously known $2^+ \rightarrow 0^+$ 1350 keV transition [4], two other γ lines (988 and 2632 keV) are seen, though barely, both in the Ge and BaF_2 spectra. Furthermore, a low-energy (288 keV) γ ray is observed in the Ge spectrum.

Anisotropy ratios obtained for the two strongest transitions of 909 and 904 keV in ^{40}S and ^{42}S, respectively, are shown in Fig. 2. These anisotropy ratios are consistent with the stretched $E2$ nature deduced from the Coulomb-excitation experiment [3]. In the case of ^{42}S, the anisotropy ratios for each transition of the 1821–1875 keV doublet could not be determined separately. However, if one of the transitions would have a stretched dipole nature, it would shift the average anisotropy ratio towards the dipole area, as

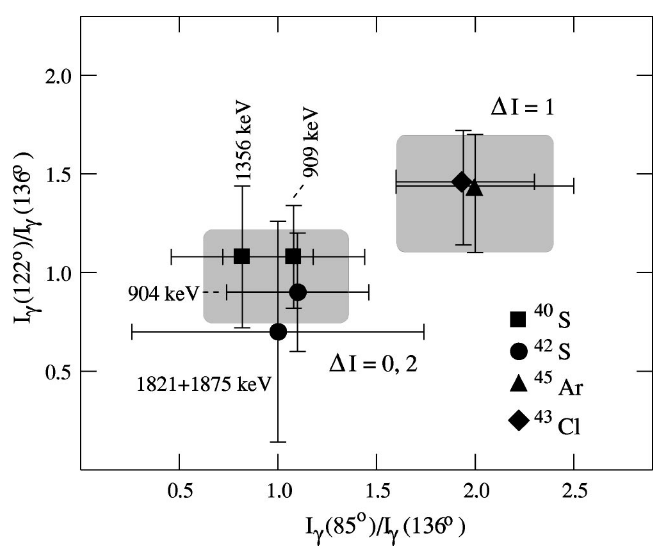

FIG. 2. Anisotropy ratios deduced for the transitions of $^{40,42}S$ as well as for some $\Delta I=1$ transitions. The hatched areas show typical uncertainties observed for the strongest $\Delta I=2$ and $\Delta I=1$ transitions in the reaction.

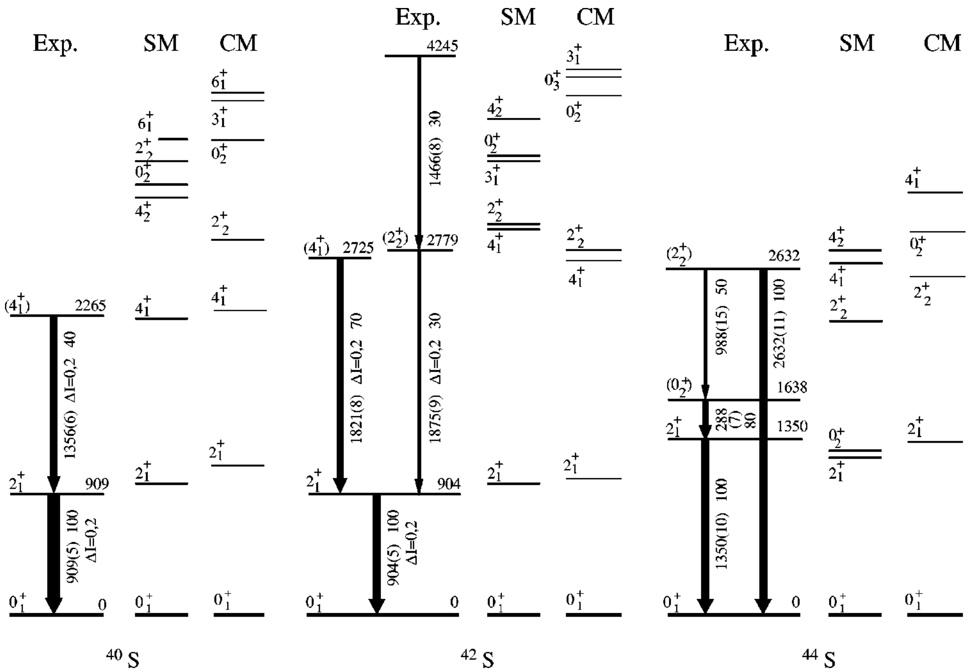

FIG. 3. Proposed level schemes of ^{40,42,44}S from the present experiment in comparison with the results of shell-model (SM) and microscopic collective-model (CM) calculations. γ-ray energies (with uncertainties), multipolarities, and relative intensities are given.

the two transitions have comparable intensities. Since the angular distributions for the stretched quadrupole transitions $\Delta I=2$ and the nonstretched dipole transitions $\Delta I=1$ are similar, we cannot distinguish between them. Thus, from the anisotropy measurement we can conclude that the 1356 keV transition of ^{40}S, as well as the 1821 and 1875 keV transitions of ^{42}S connect states having a spin difference of 2 or $0\hbar$.

B. The level schemes

The level schemes of ^{40,42,44}S deduced from the present data are shown in Fig. 3. The 909 and 1356 keV transitions in ^{40}S were observed to be in coincidence. Since the 909 keV γ line has a higher intensity, we have placed the 1356 keV transition on top of it, in accordance with the results of the Coulomb-excitation [3] and the β-decay studies [20]. Both from the previous [16–18] and the present experiments we have noticed that the fragmentation reaction favors yrast states. Hence spin parity $I^{\pi}=(4^+)$ was assigned to the state at 2.265 MeV in ^{40}S, although 2^+ and 4^+ values are allowed by the anisotropy ratio obtained for the 1356 keV transition.

A level scheme of ^{40}S has recently been published by Winger *et al.* [20] where for the lowest three states a 2_1^+ state was indicated at 904 keV, a (4_1^+) at 1917 keV, and a (2_2^+) state at 2255 keV. One can find two discrepancies between that and the present level schemes: First, in the present case the 1013 keV γ line, which deexcites the 1917 keV level, was not observed. Our experience shows that we should have seen the 1917 keV level, if it would have been the yrast 4^+ state. Second, we have observed the 2.265 ± 0.011 MeV level, which corresponds to the level at 2255 keV in Ref. [20], but we rather assigned a 4^+ spin to it. To solve the first discrepancy, we suggest an alternative to the level scheme of Winger *et al.* Transitions with 1013 keV and 2808 keV decay from and to the 1917 keV state, respectively, in the decay scheme. Since they have the same intensities, it is possible to exchange the order of these transitions. This would result in a new state at 3712 keV instead of 1917 keV. For the second point, although we cannot rule out the spin assignment of the 2_2^+ state, we would, however, be in favor of the 4^+ assignment because of the yrast argument. This assignment does not contradict the results obtained in the β-decay study, and is also supported by the fact that the direct decay of the 2255 keV state to the ground state is not observed in any of the two experiments.

In ^{42}S, the high intensity of the 904 keV line implies that it feeds the ground state, in agreement with the previous Coulomb-excitation results [3]. The 1821–1875 keV doublet was seen in coincidence with the 904 keV transition, suggesting that they both feed the 904 keV state. The intensity of the 1875 and 1466 keV γ lines are enhanced in the high multiplicity γ-gated spectra, indicating that they are members of a longer cascade. Because the 1466 keV transition's intensity is lower, it has been placed on top of the 1875 keV transition. The analysis of the anisotropy ratios suggests that both the 1821 and 1875 keV transitions connect states with 0 or $2\hbar$ spin differences. Since these states are connected to a 2^+ state, spins 0^+, 2^+, or 4^+ are therefore expected. From the asymmetry of the 1821–1875 keV doublet it is seen that the 1821 keV line is somewhat stronger than the 1875 keV one. Keeping in mind that yrast states are favored in a fragmentation reaction, we assign the higher spin (4^+) to the state at 2725 keV (which decays through the stronger 1821 keV line), and a lower spin (2^+) to the state at 2779 keV.

In ^{44}S, the placement of the 1350 keV transition is fixed by the Coulomb-excitation experiment [4]. In contradiction to the case in ^{40,42}S, the intensity of this transition does not carry all the feeding intensity estimated from the number of nuclei produced. The missing intensity has to be found in an additional transition, which decays directly from an excited state to the ground state. A good candidate for that is the 2632 keV transition, which has the right intensity. This transition has not been seen in the Coulomb excitation, possibly because of the lower probability to excite the 2632 keV level. The 288 and 988 keV transitions were not seen either, suggesting that they are not directly connected to the ground state. The energies of the 288, 988, and 1350 keV transitions add up to 2626 keV, which overlaps the energy of the 2632 keV transition. This indicates that the 2632 keV γ ray may be a crossover transition within the uncertainties in the γ energies. Both the 288 and 988 keV lines have stronger intensities in the high multiplicity spectra, which means that they are members of longer γ cascades. These facts support the placement of the three lower-energy transitions on top of each other. The ordering of the transitions is determined by their intensities. The spin of the 2632 keV state is constrained to 1^+, 2^+, and less likely 3^- by the fact that it is directly connected to the ground state. The relatively low energy of this state and the missing transition to the first 2^+ state makes the spin/parity assignment 3^- very unlikely. A spin 2 has been chosen on the basis of the yrast argumentation. The systematics of even-even nuclei suggests spin 0 or 4 for a low-lying second excited state. According to the feeding pattern, the 1638 keV second excited state is unlikely to be yrast. Thus, we have tentatively assigned spin (2^+) and (0^+) to the 2632 and 1638 keV states, respectively.

IV. DISCUSSION

Simple nuclear models can be used to give a qualitative interpretation of the level schemes. For instance, the collective behavior of these nuclei can be deduced from the energies of the low-lying 2_1^+, 2_2^+, and 4_1^+ states.

In ^{40}S the $E_{4_1^+}/E_{2_1^+}$ ratio was found to be ~ 2.5, which qualitatively suggests a transitional or γ-soft rotor nature for this nucleus. In ^{42}S, the value of the $E_{4_1^+}/E_{2_1^+}$ ratio is ~ 3.0, which is quite close to the rigid rotor value. The 4_1^+ (at 2725 keV) and the 2_2^+ (at 2779 keV) excited states are almost degenerate. Furthermore, the $2_2^+ \to 0_1^+$ transition is unobserved due to its very weak intensity ($<20\%$ relative intensity). These characteristics are in accordance with the predictions of the γ-unstable rotor model, according to which the 3^+ member of the γ band should be at an excitation energy of $E_{3^+} = 4.5 \times E_{2_1^+} = 4068$ keV. The excited state at 4245 keV decaying to the 2_2^+ state may be a candidate for the 3_1^+ γ-vibrational state.

The experimental γ deformation in the case of ^{42}S can be estimated by use of the triaxial rotor model in which the $E_{2_2^+}/E_{2_1^+} = 3.1$ energy ratio corresponds to a triaxial shape with $\gamma = 23^{\circ}$ deformation.

In the case of ^{44}S the branching ratio from the (2_2^+) state at 2632 keV suggests that the $2_2^+ \to 0_1^+$ transition has a relatively low probability, as the observed intensity of the 2632 keV transition is mainly due to the E_γ^5 energy factor in the γ-decay intensity. This suggests that the excited (0_2^+) state at 1638 keV and the (2_2^+) state at 2632 keV have similar structures, which is different from that of the ground state and the 2_1^+ state.

Two different models have been applied to interpret the level schemes of the neutron-rich sulfur isotopes: the microscopic collective model (CM) and the shell model.

For the description of the collective states, the microscopic CM calculations [21], with configuration mixing included in the space of the Hartree-Fock-Bogolyubov states, (HFB) has been applied. As a first step, the potential-energy surfaces (PESs) are determined by the constrained HFB method [13], which gives qualitative information on the structure of these nuclei. Figure 4 shows the obtained PESs for ^{40,42,44}S in the (β-γ) plane. The ^{40}S surface shows a prolate minimum, while the ^{44}S surface displays two minima of similar energy at prolate and oblate deformations separated by a barrier of about 2 MeV. The ^{42}S surface exhibits an intermediate, γ-soft situation. Similar conclusions were obtained in Refs. [12,15]. In addition to the potential-energy surfaces, other collective-model inputs are the vibrational inertia parameters and the moments of inertia. The model assumes that moments of inertia, calculated at zero rotational frequency, do not change with increasing frequency. This assumption is verified in heavy rotorlike nuclei at low spin. It is, however, not the case in the sulfur isotopes. To check this, we have performed, for each of the three sulfur isotopes, a self-consistent cranked HFB [14] calculation at the minimum of the PES of the various excited states. For ^{40}S, the moment of inertia changed by a factor of 1.4 from spin 0 to spin 2 and by 2.3 from spin 0 to spin 4. These factors are 1.13 and 1.4, respectively, for ^{42}S, and 1.05 and 1.3, respectively, for ^{44}S. For that reason, the collective Hamiltonian has been solved by increasing all moments of inertia by average factors (2 for ^{40}S, 1.25 for ^{42}S, and 1.1 for ^{44}S). It is interesting to note that the strong variation of these moments of inertia is a consequence of the softness of these nuclei against β and γ deformations. The results of the calculations are compared with the experimental results in Fig. 3. It can be seen that the agreement with the experiment is fairly good except for the (0_2^+) state in ^{44}S. The energy of the 2_2^+ state in the ^{42}S nucleus, interpreted as the band head of the γ band in the CM is found to be close to the experimental value. In the full CM calculation, triaxial shapes are obtained for the ^{40,42,44}S nuclei with a mean value of the γ deformation for the ground states of 20°, 22°, and 26°, respectively. For the 0_2^+ states, these γ deformations are 18°, 12°, and 14°, respectively, while the 0_3^+ states are on the oblate side of the PES with $\gamma \sim 36°$.

Most recently properties of sulfur isotopes around $N = 28$ have been calculated in the framework of a similar model, where the angular momentum projected generator coordinate method was used with the axial quadrupole moment applied as collective coordinate and the Gogny force, with the $D1S$ parametrization as the effective interaction [15]. Although, as a result of the angular momentum projection, a larger (3–4 MeV) barrier develops between the prolate and oblate minima of the potential-energy surfaces, the dynamical part of the collective Hamiltonian mixes the two shapes at low spin values in ^{40,42}S nuclei in this description. Thus,

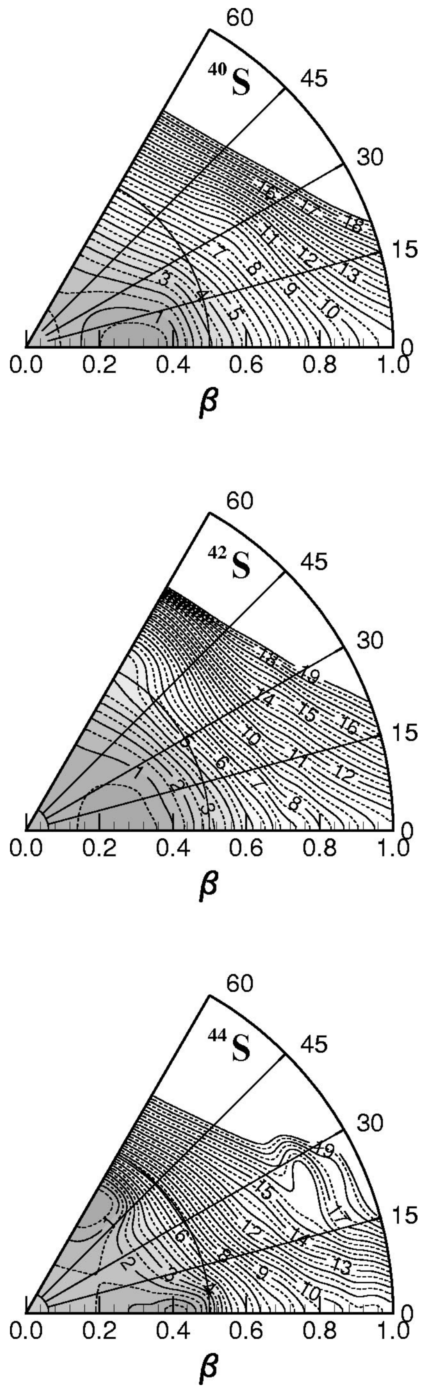

FIG. 4. HFB potential-energy surfaces of ^{40,42,44}S in the β-γ plane.

for these nuclei qualitative conclusions were drawn similar to the ones discussed above. For ^{44}S this model predicts a clear shape coexistence with an oblate ground state, which is to be compared with our mixed shape ground and low-spin states.

A more sensitive test of the single-particle aspects of the structure of these nuclei can be obtained by comparing the experimental results with those of the shell-model calculations. In previous studies, where the $f_{7/2}$-$p_{3/2}$ energy difference in ^{35}Si could not be fixed unambiguously, shell-model calculations showed the persistence of the $N=28$ shell closure. Taking into account the new data on ^{35}Si [22], the $f_{7/2}$-$p_{3/2}$ effective single-particle energy differences [23] are now known as 4.35, 3.75, and 3.25 MeV for ^{48}Ca, ^{46}Ar, and ^{44}S, respectively [24]. These numbers are in reasonable aggreement with those obtained in the CM calculations (4.7, 4.2, and 3.9 MeV, respectively) [13], as well as those obtained by Lalazissis *et al.* from relativistic mean field calculations [10] (3.8 MeV for ^{46}Ar and 3.6 MeV for ^{44}S). These calculated values for the $N=28$ gap show an erosion, which is not large enough to wash out the effects of the spherical shell closure completely. As a result, a remnant of the $N=28$ gap may be found in ^{44}S, which possibly exhibits a mixed spherical-deformed shape.

The results of the new large-scale shell-model calculation for ^{40,42,44}S are compared with our experimental data in Fig. 3. It can be seen that the energies of the first 2^+ and 4^+ states characterizing the collective properties of the ground state are well described in all three nuclei. The existence of a γ band, a $\Delta I=1$ sequence of states, is also predicted by the model.

In ^{44}S, the second excited state is calculated within the shell model to have an energy close to the experimental one, and it is predicted to be a 0^+ state. The calculated energy difference between the 0_2^+ and 2_2^+ states is also close to the experimental value. According to this calculation, the 2_1^+ state, which is connected by a transition of a large $B(E2)$ value to the ground state, has a large negative quadrupole moment ($Q=-17.4$ efm^2) corresponding to a prolate ground state deformation, while the 2_2^+ state connected by a transition with a large $B(E2)$ value to the 0_2^+ state has a small positive quadrupole moment ($Q=2.6$ efm^2), which implies a close-to-spherical, slightly oblate shape of the 0_2^+ state. In spite of the differences in the quadrupole moments, the members of the two sets of states are also connected by transitions with substantial $B(E2)$ values. The shell model therefore predicts a sizeable mixing of states with different shapes, in accordance with the collective-model calculations.

In the shell model, the deformation driving force is the pseudo-SU(3) symmetry of the proton $\pi s_{1/2}$-$\pi d_{3/2}$ orbits [6], which are approximately degenerate in the neutron-rich sulfur isotopes. In this framework, the ground state of ^{44}S, which has two protons in the $l=1$ pseudospin orbit, obeys a prolate deformation. ^{46}Ar, which contains four protons (two proton holes) in it, would be slightly oblate. Excitations of the two core protons of ^{44}S to the pseudo-SU(3) orbit would result in an ^{46}Ar-like, slightly oblate excited 0_2^+ state. Thus, the differences between the quadrupole fields associated with the proton configurations may be responsible for the development of two shapes in ^{44}S. In the large-scale calculations these shapes become mixed by the effective interaction.

V. CONCLUSIONS

In-beam γ spectroscopy with fragmentation reactions at intermediate energies made possible the extension of the level schemes in neutron-rich sulfur isotopes ($A=40$, 42, 44) to higher-lying excited states. Collective-model and shell-model calculations are in very good agreement with the experimental level schemes. It is found that ^{40}S and ^{42}S are deformed, γ-soft nuclei, while ^{44}S exhibits a shape mixing in the low-energy states, which suggests the erosion of the $N=28$ spherical closed shell. The microscopic model calculations performed show that although the $N=28$ shell gap has decreased with decreasing Z, it has not disappeared at $Z=16$. On the other hand, according to the shell model, the $Z=16$ subshell effect present at $N=20$ is completely lost when adding neutrons up to $N=28$. As a result of the near degeneracy between the $\pi s_{1/2}$ and $\pi d_{3/2}$ orbitals, the quadrupole field of the pseudo-SU(3) configuration could polarize the neutron core to such an extent that a permanent quadrupole deformation evolved at $N=24$–28.

From these models it is expected that the $N=28$ shell gap still decreases at $Z=14$. However, the proton configuration of the Si isotopes is more rigid against quadrupole deformations compared to the S isotopes due to the $Z=14$ subshell closure. As a result, a mixed shape configuration is expected for the ground state of ^{42}Si [24]. Straightforward and more precise information on the $N=28$ shell gap could be obtained from transfer reactions, where the single-particle energies can be deduced from the energies and spectroscopic factors of the excited states.

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