Shape of ⁴⁴Ar: Onset of deformation in neutron-rich nuclei near ⁴⁸Ca

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The development of deformation and shape coexistence in the vicinity of doubly magic ⁴⁸Ca, related to the weakening of the N = 28 shell closure, was addressed in a low-energy Coulomb excitation experiment using a radioactive ⁴⁴Ar beam from the SPIRAL facility at GANIL. The 2_1^+ and 2_2^+ states in ⁴⁴Ar were excited on ²⁰⁸Pb and ¹⁰⁹Ag targets at two different beam energies. B(E2) values between all observed states and the spectroscopic quadrupole moment of the 2_1^+ state were extracted from the differential Coulomb excitation cross sections, indicating a prolate shape of the ⁴⁴Ar nucleus and giving evidence of an onset of deformation already two protons and two neutrons away from doubly magic ⁴⁸Ca. New Hartree-Fock-Bogoliubov based configuration mixing calculations have been performed with the Gogny D1S interaction for ⁴⁴Ar and neighboring nuclei using two different approaches: the angular momentum projected generator coordinate method considering axial quadrupole deformations and a five-dimensional approach including the triaxial degree of freedom. The experimental values and new calculations are furthermore compared to shell-model calculations and to relativistic mean-field calculations. The new results give insight into the weakening of the N = 28 shell closure and the development of deformation in this neutron-rich region of the nuclear chart.

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I. INTRODUCTION

The shape of the atomic nucleus reflects very sensitively the underlying shell structure of the nucleons. Nuclei in the vicinity of closed shells are generally spherical. The presence of only few valence particles (or holes) is usually not sufficient to polarize the stiff "magic" core and induce deformation. Away from closed shells, on the other hand, the valence nucleons can scatter into various orbitals near the Fermi surface. Because the energies of the orbitals change with deformation, in particular for intruder orbitals from the next major shell, the nucleus can minimize its energy by assuming a deformed shape.

Following this argumentation, nuclei in the vicinity of doubly magic ⁴⁸Ca should be spherical. However, it is known that the shell structure changes when moving away from the valley of stability: The large energy gaps at the magic numbers present in stable nuclei can get washed out, and new shell gaps at other nucleon numbers may appear. The fundamental question of how the nuclear shell structure evolves with proton or neutron excess is one of the main motivations to study nuclei far from stability. The study of the shape of such exotic nuclei

can reveal changes in the shell structure and is a particularly sensitive test of nuclear structure models and their predictive power.

The disappearance of a shell closure in neutron-rich nuclei was first discovered in the so-called "island of inversion" at N = 20, where the ground states of neutron-rich Na, Mg, and Ne isotopes are dominated by deformed intruder configurations [1]. While the N = 20 shell closure disappears rather abruptly between ³⁴Si and ³²Mg, evidence has been accumulated recently for a more gradual erosion of the neutron shell closure in the proton-deficient N = 28 isotones below ⁴⁸Ca. The shell closure at N = 28 is the first one driven by the spin-orbit interaction and, unlike N = 20, separates orbitals of the same parity. Neutron single-particle energies have been determined via the $d({}^{46}\text{Ar}, {}^{47}\text{Ar})p$ reaction, indicating a moderate reduction of the N = 28 gap for ⁴⁶Ar [2]. A rather low-lying and collective 2_1^+ state in ⁴⁴S [3] and the presence of an isomeric 0^+_2 state at low excitation energy [4] suggest the presence of deformation and shape coexistence in ⁴⁴S. The g-factor measurement of a low-lying isomeric state in ⁴³S established a $v f_{7/2}$ configuration of this state and an intruder character of the ground state [5]. Finally, the very low-lying 2_1^+ state at only 770 keV in 42 Si [6] provides evidence for a substantial deformation and the complete collapse of the N = 28 shell closure at Z = 14. This result is at variance with an earlier interpretation based on the measurement of the two

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proton knock-out cross section, which concluded that a strong Z = 14 subshell closure will result in a spherical "magic" nucleus [7].

Large-scale shell-model calculations with the full proton sd and neutron pf shells as valence space originally predicted the persistence of the N = 28 shell closure and a doubly magic character of ⁴²Si [8]. As more experimental data became available, the effective interaction was adjusted, and shellmodel calculations are now reproducing the experimentally observed collapse of the shell closure [9]. The erosion of the shell closure and the development of deformation for the proton-deficient N = 28 isotones was also predicted by Skyrme Hartree-Fock and relativistic mean-field (RMF) calculations [10,11]. Fully self-consistent relativistic Hartree-Bogoliubov calculations predicted a staggering of the groundstate deformation between oblate in ⁴⁶Ar and ⁴²Si and prolate in ⁴⁴S and ⁴⁰Mg, while the absolute deformation was found to increase with decreasing proton number [12]. Spectroscopic properties of N = 28 isotones were calculated using the angular momentum projected generator coordinate method with the axial quadrupole moment as generator coordinate and the Gogny D1S interaction [13]. These calculations predicted the onset of deformation for ⁴⁶Ar and the coexistence of welldeformed oblate and prolate shapes in the lighter isotones, with oblate shapes favored in ⁴⁴S and ⁴²Si and prolate deformation dominating in ⁴⁰Mg.

The nuclear shape changes rapidly not only with the proton but also with the neutron number in this region of the nuclear chart. Spectroscopic results in the neutron-rich sulfur isotopes, for example, are consistent with a transition from prolate deformation in ⁴⁰S via γ softness in ⁴²S to prolate-oblate shape coexistence in ⁴⁴S [14]. This interpretation is supported by Hartree-Fock-Bogoliubov based configuration mixing calculations using a five-dimensional collective Hamiltonian and the Gogny D1S interaction [14,15]. These calculations predict that oblate shapes become favored in the N = 30 isotope ⁴⁶S.

The spectroscopy of proton-deficient nuclei with $N \approx 28$ is very challenging and at the very limits of feasibility with present-day techniques. B(E2) values have been measured via intermediate-energy Coulomb excitation for ⁴⁴Ar [16], ⁴⁶Ar [16,17], $^{38-42}$ S [16], and 44 S [3]. Only the excitation energy of the 2_1^+ state is known in ⁴²Si [6], while a measurement of the B(E2) value for this nucleus is presently out of reach. The dripline nucleus ⁴⁰Mg, finally, was recently observed for the first time by identifying three events in a fragmentation experiment [18]. The measurement of the energy and excitation probability of the first excited state gives a first indication of the collectivity of a nucleus. The measurement of quadrupole moments and the excitation probability of non-yrast states provides more profound nuclear structure information. Such measurements require radioactive ion beams of high quality and intensity at energies well below the Coulomb barrier and have so far never been attempted in this region of the nuclear chart. In this work we report on the measurement of the spectroscopic quadrupole moment and the B(E2) values to the 2^+_1 and 2^+_2 states in ⁴⁴Ar. The precise measurement of these observables via low-energy Coulomb excitation and the reorientation effect gives insight into the development of deformation in this region

of the nuclear chart and serves as a stringent test for both microscopic collective and shell models.

Spectroscopic data on ⁴⁴Ar had been obtained previously using various techniques to populate excited states. The resulting level schemes above the 2_1^+ state at 1158 keV, however, are inconsistent. A sequence comprising a $2^+, 4^+$, and 6⁺ state at 1158, 2746, and 3439 keV was proposed based on the γ rays observed following deep-inelastic reactions [19]. On the other hand, a direct transition from a state at 2748 keV to the ground state was observed following the β decay of ⁴⁴Cl [20], casting doubt on the (tentative) assignment of spin-parity 4^+ for the state observed after deep-inelastic collisions. A second 2^+ state was reported at an excitation energy of 1.78 MeV in an experiment using nucleon removal reactions at relativistic energies [21]. Other experiments do not confirm the presence of a 2^{+} state at this energy. The 2^{+}_{2} state was reported at an excitation energy of 2011 keV in the β -decay study [20]. Finally, the $B(E2; 0^+_1 \rightarrow 2^+_1)$ value for ⁴⁴Ar has been measured using Coulomb excitation at intermediate energy [16], and the density distribution has been probed using inelastic proton scattering [22].

Low-energy Coulomb excitation can provide valuable complementary information to the existing data for the lowlying states in ⁴⁴Ar. The technique is in particular sensitive to diagonal matrix elements via second-order effects, which allow extracting quadrupole moments including their sign. In this way the deformation and shape associated with a given state can be measured. Low-energy Coulomb excitation is the only experimental technique that can distinguish between prolate and oblate shape of the nucleus in a short-lived excited state. Contrary to intermediate-energy Coulomb excitation, it is furthermore possible to excite higher-lying states in multiple steps. The measurement of the differential excitation cross section for each state as a function of scattering angle yields a complete set of matrix elements connecting all states populated in the experiment. Also the spin-parity of the excited states can usually be inferred from the observed excitation probabilities. In this work we report on the results of a projectile Coulomb excitation experiment with radioactive ⁴⁴Ar beam at "safe" energy below the Coulomb barrier.

The article is organized as follows: Experimental details are presented in Sec. II, followed by a description of the data analysis procedure and the presentation of the results in Sec. III. The results are discussed and compared with shell-model, mean-field, and beyond-mean-field calculations in Sec. IV. Finally, the work is summarized and conclusions are drawn in Sec. V.

II. EXPERIMENTAL DETAILS

A radioactive ⁴⁴Ar beam was produced at the SPIRAL facility [23] at GANIL using the Isotope Separation On-Line (ISOL) technique. An intense ⁴⁸Ca primary beam was accelerated subsequently by the two large sector-separated cyclotrons of GANIL to an energy of 60A MeV with an average beam current of $3.5 \ e \ \mu$ A. The ⁴⁸Ca projectiles were fragmented on the carbon production target of SPIRAL, which was operated at high temperature to facilitate the migration of the radioactive atoms to the surface. From there the reaction products diffused into a Nanogan-3 [24] Electron Cyclotron Resonance (ECR) ion source via a cold transfer tube, which largely prevented nongaseous elements from reaching the plasma of the source. Radioactive ⁴⁴Ar atoms ($T_{1/2} = 11.9$ min) were ionized to charge state 8+ and transported to the CIME cyclotron for reacceleration. The cyclotron itself provides at the same time a high mass resolution resulting in a quasipure secondary beam.

To exploit the strong Z dependence of the Coulomb excitation cross section, the experiment was split into two parts during which the reaccelerated ⁴⁴Ar projectiles were impinging on ¹⁰⁹Ag and ²⁰⁸Pb secondary targets at energies of 2.68A and 3.68A MeV, respectively. The secondary beam energies were chosen such that the distance of closest approach d fulfilled the condition

$$d \ge 1.25 \left(A_p^{1/3} + A_t^{1/3} \right) + 5 \text{ fm}, \tag{1}$$

which ensures a purely electromagnetic excitation [25]. While Coulomb excitation of the doubly magic nucleus ²⁰⁸Pb is negligible under these conditions, the ¹⁰⁹Ag target nuclei are easily excited, which can be used for normalization purposes. The ¹⁰⁹Ag and ²⁰⁸Pb targets had thicknesses of 1.0 and 0.9 mg/cm², respectively. The average intensity of the secondary beam was 2×10^5 ions per second at the target position.

The γ rays from the Coulomb excited states were detected in the EXOGAM array [26], which comprised 10 large escapesuppressed germanium clover detectors for this experiment. Each clover detector consists of four HPGe crystals, the outer contacts of which are electrically segmented into four segments, giving precise spacial information on the energy deposit in the detectors. The scattered projectiles and the recoiling target nuclei were detected in an annular doublesided silicon detector, divided into 16 concentric rings on the front and 96 azimuthal sectors on the back side. The active surface of the detector had inner and outer radii of 9 and 41 mm, respectively, and it was placed 25 mm downstream from the target. The energy resolution of the particle detector was sufficient to distinguish between scattered projectiles and target recoils, thereby allowing the kinematic reconstruction of the Coulomb excitation events. The segmentation of both the silicon and the germanium detectors resulted in a precise measurement of the emission angle of the γ rays with respect to the velocity vector of the scattered projectile. A resolution of 10 keV was obtained for a γ ray of 1158 keV after Doppler correction. The coincidence requirement between γ rays detected in EXOGAM and scattered particles detected in the silicon detector helped to suppress the otherwise overwhelming background from the β decay of the beam.

Both projectile and target nuclei can be excited in a collision between ⁴⁴Ar and ¹⁰⁹Ag. The well-known transition strengths in ¹⁰⁹Ag [27,28] can thus be used to normalize the excitation probability in ⁴⁴Ar. The Doppler-corrected γ -ray spectra in coincidence with scattered ⁴⁴Ar ions and ¹⁰⁹Ag recoils are shown in Fig. 1. Projectile and target detection correspond to small and large scattering angles in the center-of-mass system, respectively. The dependence of the excitation process on the scattering angle is clearly visible, for example, the enhancement for large scattering angles of the 9/2⁻ \rightarrow 5/2⁻



FIG. 1. Total γ -ray spectra after Coulomb excitation of ⁴⁴Ar on a ¹⁰⁹Ag target in coincidence with ⁴⁴Ar ejectiles (left) and ¹⁰⁹Ag recoils (right), corresponding to small and large center-of-mass scattering angles, respectively. The spectra are Doppler corrected using the velocity vector of the ⁴⁴Ar projectiles.

transition at 676 keV in 109 Ag, which requires a multistep excitation.

The statistics collected during \sim 50 h of data taking with the ¹⁰⁹Ag target allowed us to subdivide the data into four subsets corresponding to different ranges of scattering angles as shown in Table I. The ranges A and B correspond to the detection of scattered ⁴⁴Ar projectiles, while the ranges C and D correspond to detection of the recoiling target nuclei. The overlap between the different ranges is due to a misalignment of the beam with respect to the particle detector. This displacement was estimated to be 2.8(2) mm from the energy of scattered beam particles measured by individual azimuthal sectors for each

TABLE I. Measured γ -ray yields (without efficiency correction) for scattering on ¹⁰⁹Ag and ²⁰⁸Pb targets, respectively. The ranges of scattering angles are given in the center of mass frame.

Data set	Nucleus	Transition	Counts	Error
A	⁴⁴ Ar	$2^+_1 \rightarrow 0^+_1$	493	34
[22.1°, 50.0°]	¹⁰⁹ Ag	$3/2_1^- \rightarrow 1/2_1^-$ $5/2_1^- \rightarrow 1/2_1^-$	6280 6690	110 110
В	⁴⁴ Ar	$2^+_1 \rightarrow 0^+_1$	1154	58
[37.9°, 78.6°]	¹⁰⁹ Ag	$3/2_1^- \rightarrow 1/2_1^-$ $5/2_1^- \rightarrow 1/2_1^-$	13100 13740	250 220
C [61.8°, 105.0°]	⁴⁴ Ar	$\begin{array}{c} 2^+_1 \rightarrow 0^+_1 \\ 2^+_2 \rightarrow 2^+_1 \end{array}$	1575 48	58 20
	¹⁰⁹ Ag	$3/2_1^- \to 1/2_1^-$ $5/2_1^- \to 1/2_1^-$	12430 13240	230 200
D [92.1°,148.3°]	⁴⁴ Ar	$\begin{array}{c} 2^+_1 \rightarrow 0^+_1 \\ 2^+_2 \rightarrow 2^+_1 \end{array}$	1274 54	54 26
	¹⁰⁹ Ag	$3/2_1^- \rightarrow 1/2_1^-$ $5/2_1^- \rightarrow 1/2_1^-$	11050 11530	220 210
E [62.8°, 153.4°]	⁴⁴ Ar	$\begin{array}{c} 2^+_1 \to 0^+_1 \\ 2^+_2 \to 2^+_1 \\ 2^+_2 \to 0^+_1 \end{array}$	2243 113 39	73 24 11
	²⁰⁸ Pb	-	-	_

of the rings. This misalignment was taken into account in the Doppler correction procedure and the Coulomb excitation analysis.

The influence of the quadrupole moment of the 2_1^+ state in ⁴⁴Ar on its excitation probability varies significantly with the scattering angle. It is hence possible to extract both the transitional and diagonal matrix elements involved in the excitation process from a differential measurement of the excitation cross section. The division of the data into angular ranges was a compromise between the number of independent data points for the γ -ray yields and the minimum level of statistics obtained for the individual ranges. It was decided to use wider ranges for large scattering angles, where the influence of the quadrupole moment on the population of the 2_1^+ state is strongest, to determine the corresponding γ -ray yields with higher accuracy.

In the second part of the experiment data was collected for ~100 h with a ²⁰⁸Pb target at a beam energy of 3.68*A* MeV. The higher *Z* of the target material resulted in larger cross sections, so that higher-lying states in ⁴⁴Ar became accessible. In addition to the 2_1^+ state, the level at 2011 keV, which was already observed after β decay of ⁴⁴Cl and tentatively assigned as 2_2^+ [20], was populated and its deexcitation via two γ rays to the 2_1^+ and the ground state was observed. The $2_2^+ \rightarrow 2_1^+$ transition is partly overlapping with the Compton edge of the $2_1^+ \rightarrow 0_1^+$ transition, hampering the precise determination of its intensity. The decay scheme of the states that were populated by Coulomb excitation on the ²⁰⁸Pb target is shown in Fig. 2 together with the Doppler-corrected, background-subtracted γ -ray spectrum in coincidence with ²⁰⁸Pb recoils. The resulting γ -ray yields are given in Table I (range E).

During this part of the experiment a beam halo of poorly focused ⁴⁴Ar ions of different charge state was present because of charge-exchange reactions with the residual gas in the beam line. Direct hits of these particles on the inner part of the silicon detector (of the order of 1000/s) could not be cleanly distinguished from scattered beam particles and lead to an increased background of random coincidences with the decay

of the highly radioactive beam. To exclude systematic errors originating from this background, small scattering angles were not considered in the analysis of the data taken with the ²⁰⁸Pb target.

III. COULOMB EXCITATION ANALYSIS AND RESULTS

The Coulomb excitation least-squares fitting code GOSIA [29] was used to determine transitional and diagonal E2 matrix elements. A χ^2 function was constructed from the observed γ -ray yields and those calculated from a set of matrix elements between all relevant states. The excitation probabilities were calculated in a semiclassical way [30] from a given set of matrix elements by integrating over the different ranges of scattering angles and the range of incident projectile energies resulting from their energy loss in the target foil. To calculate the γ -ray yields from the excitation probabilities, the geometry and relative efficiency of the detectors were taken into account. The γ -ray intensities were corrected for internal conversion, and the angular correlation of the γ rays was corrected for relativistic effects and the attenuation caused by the deorientation effect during recoil into vacuum. The latter was caused by the interaction of the recoiling nucleus with the fluctuating hyperfine fields of the atomic electrons, which was treated in a simplifying phenomenological model [31,32].

In a first step the influence of the quadrupole moment of the 2_1^+ state on its excitation probability was investigated at small scattering angles. For this purpose a GOSIA calculation was performed with the input data from the ¹⁰⁹Ag target restricted to the angular range A (see Table I) and with the B(E2) fixed to the experimental value from intermediate-energy Coulomb excitation, $B(E2; 0_1^+ \rightarrow 2_1^+) =$ $345(41) e^2$ fm⁴ [16]. Changing the value of the quadrupole moment had only a negligible effect on the excitation probability compared to the statistical uncertainty of the γ -ray yield. In an analogous way it could be shown that higher-lying states have no significant influence on the excitation probability of the 2_1^+ state at the small scattering angles of range A.



FIG. 2. Total Doppler-corrected γ -ray spectrum after Coulomb excitation of ⁴⁴Ar on a ²⁰⁸Pb target in coincidence with ²⁰⁸Pb recoils. The decay scheme of the populated states is shown on the right. The transitions are labeled with their energies (in keV), and the width of the arrows is proportional to the deduced *B*(*E*2) values.

TABLE II. Reduced E2 matrix elements in ⁴⁴Ar obtained in the present work and resulting B(E2) values and spectroscopic quadrupole moments.

I_1	I_2	$\langle I_2 \ E2 \ I_1 \rangle$ (e b)	$B(E2; I_1 \to I_2)$ $(e^2 \text{ fm}^4)$	Q_s (e fm ²)
0_{1}^{+}	2_{1}^{+}	$0.194^{+0.009}_{-0.015}$	378^{+34}_{-55}	
0_{1}^{+}	2^{+}_{2}	$0.048\substack{+0.004\\-0.004}$	23^{+2}_{-2}	
2_{1}^{+}	2^{+}_{2}	$0.58\substack{+0.06\\-0.04}$	680^{+150}_{-90}	
2_1^+	2_{1}^{+}	$-0.11\substack{+0.04\\-0.04}$		$-8.3^{+3.0}_{-3.0}$

The γ -ray yield of set A for the 2_1^+ state depends therefore only on the transitional matrix element $\langle 0_1^+ || \mathcal{M}(E2) || 2_1^+ \rangle$, which can consequently be obtained from data set A alone by normalizing to the excitation probability of states in the ¹⁰⁹Ag target nucleus. The uncertainty of the resulting B(E2) value is dominated by the statistical error of the measured yields and the uncertainties of the matrix elements for the target nucleus ¹⁰⁹Ag [27].

In a second step, the remaining data sets (B-E) from both the ¹⁰⁹Ag and ²⁰⁸Pb targets were analyzed simultaneously with the GOSIA code using the standard procedure of χ^2 minimization. The $B(E2; 0_1^+ \rightarrow 2_1^+)$ value determined from data set A and the branching ratio $I_{\gamma}(2^+_2 \rightarrow 0^+_1)/I_{\gamma}(2^+_2 \rightarrow 2^+_1)$ known from β -decay studies [20,33] together with their experimental uncertainties were included as additional data points in the fit. The γ -ray yields measured for ¹⁰⁹Ag were used to determine the normalization factors related to the Rutherford cross-section and the efficiency of the particle detection for the subsets of data collected on the ¹⁰⁹Ag target. These normalization factors were then used when fitting matrix elements in ⁴⁴Ar to the observed γ -ray yields. The use of the B(E2) value that was derived independently from data set A as additional input data enhanced the sensitivity of the remaining data to the diagonal matrix element $\langle 2_1^+ || \mathcal{M}(E2) || 2_1^+ \rangle$ and those related to the 2^+_2 state. The two-step analysis was found to reduce the uncertainties of the matrix elements compared to a one-step analysis, where the data from all angular ranges were fitted simultaneously. The resulting transitional and diagonal E2 matrix elements and the deduced B(E2) values and the spectroscopic quadrupole moment are presented in Table II.

The reduced transition probability, $B(E2; 0_1^+ \rightarrow 2_1^+) = 378_{-55}^{+34} e^2 \text{ fm}^4$, is in agreement with the value of $345(41) e^2 \text{ fm}^4$, which was found in intermediate-energy Coulomb excitation [16]. Alternative subdivisions of the data into two, five, six, and eight angular ranges were tested and analyzed with GOSIA. The resulting sets of matrix elements were consistent, but the adopted division into four ranges was found to be the best compromise between the number of data points and the level of statistics for each data set, resulting in the highest accuracy for the quadrupole moment $Q_s(2_1^+)$. The measured value of $Q_s(2_1^+) = -8(3) e \text{ fm}^2$ represents experimental proof of the prolate shape of the ⁴⁴Ar nucleus in the 2_1^+ state.

In the rotational model the spectroscopic quadrupole moment Q_s and the B(E2) value are related to the intrinsic

quadrupole moment Q_0 as

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

$$B(E2; I_1 \to I_2) = \frac{5}{16\pi} Q_0^2 |\langle I_1 K 20 | I_2 K \rangle|^2.$$
(3)

For a purely vibrational nucleus, however, it is $Q_s = 0$. The intrinsic quadrupole moment obtained from the $B(E2; 0_1^+ \rightarrow 2_1^+)$ value in the rotational model is $Q_0 = +62_{-4}^{+3} e \text{ fm}^2$, which corresponds to $Q_s = -17.6_{-1.3}^{+0.8} e \text{ fm}^2$ and a quadrupole deformation of $\beta_2 = +0.23$. The measured spectroscopic quadrupole moment of $Q_s = -8(3) e \text{ fm}^2$ lies hence in between a rotational and a vibrational value. This argumentation, however, is only valid for a nucleus with axial symmetry. A potential triaxial deformation, which is difficult to prove experimentally, would also result in a lower spectroscopic quadrupole moment compared to axial symmetric deformation.

The spectroscopic quadrupole moment is also smaller than one would expect from the deformation parameter $|\beta_2| =$ 0.31(5) that was extracted from the inelastic proton scattering experiment [22]. While Coulomb excitation probes only the proton distribution, inelastic proton scattering at intermediate energy is also sensitive to the neutron distribution. The difference between the two results could therefore be explained by a stronger neutron deformation. One should keep in mind, however, that the extraction of a deformation parameter from inelastic proton scattering is model dependent. While the absolute magnitude of deformation extracted from inelastic proton scattering should be taken with care, a consistent picture emerges when comparing the relevant quantities with those of ³⁶Ar. The shape of ³⁶Ar was found to be oblate with $Q_s(2_1^+) = +11(6) e \text{ fm}^2$ [34]. The proton scattering experiment, which is not sensitive to the sign of the deformation, found an absolute deformation parameter of $|\beta_2| = 0.36(4)$ for ³⁶Ar [22], slightly larger than that for ⁴⁴Ar and consistent with the larger quadrupole moment. The smaller B(E2)value of 300(30) e^2 fm⁴ and larger spectroscopic quadrupole moment suggest that ³⁶Ar has a more rotational character than ⁴⁴Ar.

The E2 matrix element linking the 2_2^+ and the 2_1^+ states is found to be large. The low level of statistics does not permit extraction of the M1 matrix element for this transition. While the uncertainty due to a possible M1 contribution is included in the value for the E2 matrix element given in Table II, the uncertainty due to unknown couplings to higher-lying states is not included. A strong coupling of the 2_2^+ state to higher-lying states could reduce the $\langle 2_2^+ || E2 || 2_1^+ \rangle$ matrix element by up to 20%, even if the corresponding transitions are unobserved. Without further constraints on the feeding of the 2_2^+ state it was also impossible to determine its diagonal matrix element.

IV. COMPARISON WITH THEORY AND DISCUSSION

The present results represent the first direct measurement of the nuclear shape for an excited state in this neutron-rich region of the nuclear chart, and for the first time it was possible to extract matrix elements beyond the first 2^+ state in this region.

TABLE III. Excitation energies (MeV), spectroscopic quadrupole moments ($e \text{ fm}^2$), and $B(E2; 0_1^+ \rightarrow 2_1^+)$ values ($e^2 \text{ fm}^4$) for the 2_1^+ states in ${}^{42}\text{Ar}$, ${}^{44}\text{Ar}$, ${}^{46}\text{Ar}$, ${}^{42}\text{S}$, ${}^{44}\text{S}$, and ${}^{42}\text{Si}$. For ${}^{44}\text{Ar}$ also the results for the 2_2^+ state are given. The experimental values are compared to the five-dimensional GCM(GOA) calculations, to angular momentum projected GCM calculations restricted to axial shapes (AMPGCM), and to shell-model calculations. Results for which no reference is given were obtained in the present work.

		Experiment	GCM(GOA)	AMPGCM	Shell model
⁴² Ar	$E(2_{1}^{+})$	1.208(1) [40]	2.076	3.17	1.155
	$Q_s(2_1^+)$	_	-10.2	-2.2	+5.7
	$B(E2; 0^+_1 \to 2^+_1)$	430(100) [40]	374	257	238
⁴⁴ Ar	$E(2_{1}^{+})$	1.158(1)	1.758	2.10	1.087
	$E(2_{2}^{+})$	2.011(1)	3.460	4.19	1.776
	$Q_s(2_1^+)$	-8(3)	-7.3	+2.7	-2.5
	$Q_s(2_2^+)$	-	+5.3	-0.5	+4.3
	$B(E2;0^+_1 \rightarrow 2^+_1)$	378^{+34}_{-55}	434	266	233
	$B(E2;0^+_1 \rightarrow 2^+_2)$	23(2)	1.77	1.51	100
	$B(E2; 2_1^+ \to 2_2^+)$	680^{+150}_{-90}	139	_	88
⁴⁶ Ar	$E(2_{1}^{+})$	1.555(9) [17]	1.911	1.73 [13]	1.593
	$Q_s(2_1^+)$	-	+9.8	+16.7 [13]	+17.5
	$B(E2;0^+_1 \rightarrow 2^+_1)$	209(24) [16,17]	334	326 [13]	365
⁴² S	$E(2_{1}^{+})$	0.904(5) [14]	1.16	1.54 [13]	0.998
	$Q_s(2_1^+)$	-	-15.7	-16.0 [13]	-17
	$B(E2;0^+_1\to 2^+_1)$	397(63) [16]	427	459 [13]	374
⁴⁴ S	$E(2_{1}^{+})$	1.334(7) [3,14,41]	1.38	1.40 [13]	1.17
	$Q_s(2_1^+)$	-	-5.9	+9.6 [13]	-16
	$B(E2;0^+_1 \rightarrow 2^+_1)$	314(88) [3]	414	377 [13]	365
⁴² Si	$E(2_{1}^{+})$	0.770(19) [6]	1.269	0.83 [13]	0.82 [9]
	$Q_s(2_1^+)$	-	+11.5	+19.2 [13]	+20 [9]
	$B(E2;0^+_1\to 2^+_1)$	_	325	478 [13]	430 [9]

In the following the experimental results are compared to calculations using different theoretical approaches and discussed in the wider context of the development of deformation in nuclei with $N \approx 28$ below ⁴⁸Ca. Experimental and theoretical values for excitation energies, B(E2) values, and quadrupole moments for several nuclei in the region are summarized in Table III and presented in Fig. 3.

A. Shell-model calculations

The new experimental results for ⁴⁴Ar, only two protons and two neutrons away from doubly magic ⁴⁸Ca, call for a comparison with shell-model calculations. Extensive calculations have been performed by Retamosa *et al.* [8] for the neutron-rich nuclei in the region of interest using the full *sd* and *pf* shells for protons and neutrons, respectively. A new effective interaction was derived in this work, and its parameters were fitted to known experimental data in the region. The results were in good agreement with the experimental excitation energies of the 2⁺₁ states in the Ar isotopes [8]. However, large discrepancies appeared for the proton-deficient N = 28 isotones, in particular for ⁴²Si. Retamosa *et al.* [8] had predicted that the N = 28 shell closure persists in the proton-deficient isotones and that ⁴²Si is a doubly magic nucleus. The same conclusion was also supported by shell-model calculations using a truncated valence space [7].

As more experimental data became available in this mass region, the monopole component of the SDPF interaction was modified [35], and new calculations were performed for the N = 28 isotones [36]. With the modified interaction, SDPF-NR, the shell-model calculations found an increase of the 2p-2h intruder configuration for the ground-state wave functions in the N = 28 isotones. The intruder configuration was predicted to become dominant in the ground state of ⁴⁰Mg. However, the calculations still found a rather high excitation energy of 1.49 MeV for the 2_1^+ state in ⁴²Si [36]. When the excitation energy of this state was experimentally established to be only 770 keV [6], the monopole interaction was again modified to reproduce this result [9]. Using this latest version of the interaction, SDPF-U, and effective charges $\epsilon_{\pi} = 1.35 \ e$ and $\epsilon_{\nu} = 0.35 \ e$, the spectroscopic properties of neutron-rich Ar and S isotopes were calculated with the code ANTOINE [37–39]. The results are included in Table III and Fig. 3.

With the new SDPF-U interaction the shell-model calculations reproduce the energies of the 2_1^+ states very well throughout the region. The $B(E2; 0_1^+ \rightarrow 2_1^+)$ values predicted for ⁴²Ar and ⁴⁴Ar are too small, while the one for ⁴⁶Ar is too large. The shell-model calculations predict an increase of the B(E2) values from ⁴⁴Ar to ⁴⁶Ar in contrast to the



FIG. 3. (Color online) Graphical representation of the experimental and theoretical values from Table III showing the excitation energy of the 2_1^+ state (bottom), the $B(E2; 0_1^+ \rightarrow 2_1^+)$ value (center), and the spectroscopic quadrupole moment of the 2_1^+ state (top) for the neutron-rich argon isotopes ^{42–46}Ar and along the chain of N = 28isotones from ⁴⁶Ar to ⁴²Si.

sharp drop that is observed experimentally. This increase of the B(E2) values in the calculations coincides with a shape change from a very small prolate deformation in ⁴⁴Ar to a relatively large oblate deformation in ⁴⁶Ar. The predicted spectroscopic quadrupole moment $Q_s(2_1^+)$ in ⁴⁴Ar is somewhat smaller than the experimental value, but has the correct sign. The shell model predicts alternating oblate (⁴⁶Ar, ⁴²Si) and prolate (⁴⁴S, ⁴⁰Mg) shapes for the 2_1^+ states in the N = 28 isotones with relatively large deformation.

The above discussion illustrates a limitation of shellmodel calculations for exotic nuclei: The matrix elements of the effective interaction must be adjusted to experimental observables within the region of interest. If experimental data are scarce, extrapolations to exotic nuclei can lead to significant deviations, as can be seen in the case of 42 Si. A satisfactory description of the spectroscopic properties throughout the region of interest was only possible after experimental data became accessible. In calculations based on the mean-field approach, however, the effective interaction is derived not locally, but globally. Such models have consequently a better chance to describe at least the global trends in the structure of exotic nuclei. Calculations based on the mean-field approach and shell-model calculations therefore generally provide complementary nuclear structure information.

B. Relativistic mean-field calculations

The region of proton-deficient $N \approx 28$ nuclei has been studied by Werner et al. [10,11] using Skyrme Hartree-Fock and relativistic mean-field (RMF) calculations. Albeit differing in some details, both approaches predict the onset of deformation in this mass region. Another detailed analysis of shape effects in the framework of RMF theory has been presented by Lalazissis et al. [12]. In the latter work the authors calculated ground-state properties such as the quadrupole deformation, proton and neutron radii, and two-neutron separation energies using a unified relativistic Hartree-Bogoliubov (RH + B) description of mean-field and pairing correlations with the NL3 effective interaction for the RMF Lagrangian and the pairing term of the Gogny D1S interaction. It was found that the spherical N = 28 shell closure is strongly suppressed resulting in deformed ground states and shape coexistence. The groundstate deformation of the N = 28 isotones was predicted to alternate between oblate in ⁴⁶Ar and ⁴²Si and prolate in ⁴⁴S and ⁴⁰Mg, in agreement with the predictions of the shell model. The importance of the correct description of pairing correlations was investigated by comparing the fully self-consistent RH+B calculations with results from a simplified RMF approach with BCS pairing and constant pairing gap (RMF+BCS). The most pronounced difference between the two calculations was found for the ground-state deformation of ⁴⁴Ar, which was predicted to be oblate in the RH+B, but prolate in the RMF+BCS calculation [12]. The prolate experimental quadrupole moment for the 2_1^+ state in ⁴⁴Ar seems to support the RMF+BCS calculation. One should, however, be careful with such a general conclusion about the pairing treatment from this result. The potential energy for ⁴⁴Ar is rather soft with respect to deformation, and the ground-state shape cannot be easily inferred from the spectroscopic quadrupole moment of the 2_1^+ state.

The RMF calculations describe only ground-state properties, and no angular momentum or particle number projection was performed. To compare calculations with spectroscopic observables related to excited states, the wave functions must be transformed into the laboratory frame of reference. The fact that both prolate and oblate shapes are found nearly degenerate in energy furthermore suggests that shape coexistence may occur. To account for configuration mixing, correlations beyond the mean-field description must be considered. Another deficiency of the mean-field calculations mentioned above is the fact that they do not correct for the zero point rotational energy.

C. Angular momentum projected generator coordinate method

The above limitations of the mean-field approach can be overcome by calculations using the angular momentum projected generator coordinate method (AMPGCM) with the axial quadrupole moment as generator coordinate [13,42]. The starting point in this approach is a set of axially symmetric wave functions from Hartree-Fock-Bogoliubov (HFB) calculations using the Gogny D1S interaction [43,44] with constraints on the quadrupole moment q_{20} . The superposition amplitudes of the constrained wave functions are found after projection onto angular momentum in a variational approach by solving the Hill-Wheeler-Griffin equation [45,46]. The basis states are restricted to reflection-symmetric states, limiting the configuration mixing calculations to positive-parity states with even spin. Different states of the same spin and parity correspond either to different quadrupole deformation or to quadrupole-vibrational states. The AMPGCM allows the direct computing of spectroscopic observables such as B(E2) values and spectroscopic quadrupole moments. The only parameters entering in the calculations are those of the Gogny D1S interaction, which have been universally adjusted and are valid for the entire nuclear chart. Details of the method are discussed in Ref. [42] and results for the N = 28 isotones can be found in Ref. [13]. New AMPGCM calculations for ⁴²Ar and ⁴⁴Ar have been performed in the present work. The results are included in Table III and Fig. 3.

The AMPGCM calculations predict the coexistence of oblate and prolate shapes throughout the region of protondeficient $N \approx 28$ nuclei. The excitation energies found in the AMPGCM calculations for the N = 28 isotones are in good agreement with the experimental values [13]. In particular the very low-lying 2_1^+ energy in ⁴²Si was correctly predicted. However, the energy spectra calculated for the lighter Ar isotopes are much too stretched, and the excitation energies calculated for the 2_1^+ states in 42 Ar and 44 Ar are found significantly too high. One could remark that a model that uses the quadrupole deformation to characterize the states is not well adapted to describe near-spherical nuclei with a soft potential. The B(E2) values found in the AMPGCM calculations are similar to those found by the shell model. The $B(E2; 0_1^+ \rightarrow 2_1^+)$ values for ⁴²Ar and ⁴⁴Ar are smaller than the experimental values, while the one for ⁴⁶Ar is too large. The small B(E2) values for ⁴²Ar and ⁴⁴Ar are due to a strong mixing of prolate and oblate components in the wave functions of the 2_1^+ states. The 2_1^+ state in 46 Ar, on the other hand, is found with a rather pure oblate character [13]. The calculations reproduce the experimentally observed increase of the B(E2) values from ⁴⁶Ar to ⁴⁴S, but the systematic trend along the chain of Ar isotopes is not correctly described.

The AMPGCM calculations predict oblate quadrupole moments for the 2_1^+ states in the N = 28 isotones (except for 40 Mg). Pronounced shape coexistence is found in 44 S with similar energies for oblate and prolate states, resulting in strong mixing and a smaller quadrupole moment for the 2_1^+ state [13]. Strong mixing is also found for the wave functions in 42 Ar and 44 Ar. However, the deformations are smaller and the spectroscopic quadrupole moments for the 2_1^+ states are consequently found close to zero, slightly prolate for 42 Ar and slightly oblate for 44 Ar, contrary to the sign of the experimental value.

When comparing the AMPGCM calculations with other predictions in Fig. 3, one is inclined to suspect that the model is biased toward oblate shapes. This might be related to the fact that the model is restricted to axial deformations and vibrations, whereas in reality the nucleus has other degrees of freedom. In particular in a nucleus with a soft potential like ⁴⁴Ar one might expect γ vibrations to be a low-lying mode of excitation. As the γ degree of freedom lies outside the model space, the AMPGCM calculations cannot account for such excitations and seem to be biased toward axially symmetric oblate shapes because of a lack of other available excitation modes. To draw such a conclusion, however, it would be necessary to measure more quadrupole moments in neighboring nuclei. It is interesting to note that a similar bias toward oblate shapes was also observed in another region of prolate-oblate shape coexistence: axial GCM calculations using the same technique with a Skyrme interaction found the ground states of the neutron-deficient krypton isotopes to be dominated by oblate shapes [47], contrary to experimental evidence [48]. Configuration mixing calculations that included the triaxial degree of freedom, however, were able to reproduce the correct ordering of prolate and oblate states in this region of shape coexistence [48,49].

D. Triaxial configuration mixing calculations

To extend the GCM calculations from axial to triaxal shapes represents a considerable task, because rotations about the three Euler angles also have to be considered, resulting in a five-dimensional collective coordinate. A formally exact solution of the GCM problem including the triaxial degree of freedom has only very recently been attempted for the light nucleus ²⁴Mg [50]. Equivalent calculations for heavier nuclei are at present prohibitively time consuming. The task can be simplified by introducing the so-called Gaussian overlap approximation (GOA), which assumes the overlap of the GCM basis states to be Gaussian. The starting point for such calculations is again a set of wave functions from HFB calculations with the Gogny D1S interaction, but this time constrained to both axial and triaxial quadrupole parameters (q_{20}, q_{22}) . The superposition amplitude of the correlated states is then again found as the solution of the Hill-Wheeler-Griffin equation [45,46]. However, the use of the GOA transforms this integro-differential equation into a Schrödinger-like equation with a collective Bohr-like Hamiltonian.

The moments of inertia are calculated self-consistently [51] at rotational frequency close to zero ($\hbar \omega = 2$ keV). The collective masses are first calculated in the cranking approximation [52,53] and then renormalized to fulfill their relationships with the moments of inertia along symmetry axes [54]. As a result the well-known deficiencies of collective masses calculated in the Inglis-Belyaev approximation are alleviated. The renormalized masses are higher than before, in keeping with expectations based on the Thouless-Valatin prescription [51]. The implementation of this renormalization in the present GCM(GOA) approach, which remains parameter free, decreases the excitation energy of 0^+_2 states throughout the chart of nuclei and significantly reduces B(E2) values in particular for medium and light-mass nuclei, which are soft against β and γ deformation. The new prescription and its implication for collective properties at low spin and

FIG. 4. (Color online) Potential energy surfaces over the (β, γ) plane from constrained HFB calculations with the Gogny D1S interaction and including a zero-point energy correction for (from left to right) ⁴²Ar, ⁴⁴Ar, ⁴⁶Ar, ⁴⁴S, and ⁴²Si.

excitation energy will be described in detail in a separate publication [55]. The method and the procedure to calculate excitation energies, transition probabilities, and spectroscopic quadrupole moments can be found in Ref. [56].

New HFB-based configuration mixing calculations using the GCM(GOA) approach with the finite-range, densitydependent Gogny D1S effective interaction have been performed for ⁴⁴Ar and several neighboring even-even nuclei. Similar calculations had already been performed for the chain of neutron-rich sulfur isotopes [14]. Potential energy surfaces from the calculations for the argon isotopes and N = 28isotones are displayed in Fig. 4. The isotopes ⁴²Ar and ⁴⁴Ar are found to be relatively soft. A well-localized oblate minimum is developing in ⁴⁶Ar in addition to a spherical minimum. For ⁴⁴S, the coexistence of prolate and oblate shape is predicted with two distinct minima separated by a triaxial barrier. Finally, the prolate minimum vanishes again for ⁴²Si, which is predicted to be a well-deformed oblate rotor. The calculated values for excitation energies, B(E2) values, and spectroscopic quadrupole moments are included in Table III and Fig. 3.

The excitation energies of the 2^+_1 states found in the GCM(GOA) calculations are systematically too large for the nuclei in this region, with the exception of ⁴⁴S. Also the higher-lying states are generally predicted at too high excitation energies, as can be seen for the 2^+_2 state in ⁴⁴Ar. On the other hand, it seems that the triaxial calculations are less affected by the softness of the potential in ⁴²Ar and ⁴⁴Ar than the axial AMPGCM calculations. The systematic trend of the 2_1^+ energies is well reproduced, in particular the increase of $E(2_1^+)$ from ⁴⁴Ar to ⁴⁶Ar and the decrease along the chain of N = 28 isotones from ⁴⁶Ar to ⁴²Si. The calculations thus confirm the erosion of the shell closure, but it is obvious that the moments of inertia are not correctly described. The moments of inertia are calculated near rotational frequency zero, and the model assumes that they do not change with increasing frequency. This is usually a valid assumption for heavier well-deformed nuclei at low spin. For nuclei in the region of the present investigation, and in particular for the rather soft argon isotopes, this assumption is inappropriate, as was already concluded from the energy spectra of the ${}^{40-44}$ S isotopes [14].

The new procedure to renormalize the collective masses has improved the agreement of the calculated B(E2) values with experiment, despite the manifest discrepancies for the moments of inertia. The $B(E2; 0_1^+ \rightarrow 2_1^+)$ values for ${}^{42}\text{Ar}$ and ${}^{44}\text{Ar}$, for example, have been reduced by approximately 30%. Although the absolute values are still slightly too large, the experimentally observed decrease from ${}^{44}\text{Ar}$ to ${}^{46}\text{Ar}$ and the increase from ${}^{46}\text{Ar}$ to ${}^{44}\text{S}$ are well reproduced. A difference between the axial and triaxial configuration mixing calculations is observed not only for the Ar isotopes but also for the $B(E2; 0_1^+ \rightarrow 2_1^+)$ value in ${}^{42}\text{Si}$. Unfortunately, an experimental measurement of this value is presently out of reach.

The principal new result obtained from the experiment is the negative spectroscopic quadrupole moment for the 2^+_1 state in ⁴⁴Ar of $Q_s(2_1^+) = -8(3) e \text{ fm}^2$, indicating a prolate shape. The GCM(GOA) calculation predicted $Q_s(2_1^+) = -7.3 \ e \ fm^2$ in agreement with the experimental value. Compared to the results of the axial GCM calculation, the quadrupole moments from the triaxial calculation are systematically shifted toward spherical and prolate shapes, resulting in alternating prolate and oblate shapes for ⁴⁴Ar, ⁴⁶Ar, ⁴⁴S, and ⁴²Si, respectively. Given the softness of the potential for some nuclei and the occurrence of prolate-oblate shape coexistence for others, such a shift seems at least plausible. With only one experimental data point, however, it is difficult to draw firm conclusions. The measurement of spectroscopic quadrupole moments in neighboring nuclei would shed more light on the validity of the different theoretical approaches.

The Coulomb excitation of ⁴⁴Ar resulted in the first experimental measurement of B(E2) values beyond the 2^+_1 state in this region of the nuclear chart. A surprisingly large matrix element was found between the first and second 2⁺ states, which seems difficult to explain. Because of the low level of statistics it was impossible to measure the quadrupole moment of the 2^+_2 state. The GCM(GOA) calculation finds significantly lower $B(E2; 2_1^+ \rightarrow 2_2^+)$ and $B(E2; 0_1^+ \rightarrow 2_2^+)$ values. The calculation predicts a positive spectroscopic quadrupole moment of $Q_s(2^+_2) = +5.3 \ e \ \text{fm}^2$. This value is consistent either with an oblate intrinsic shape and K = 0or with a γ -vibrational K = 2 state based on a prolate intrinsic shape. The calculations clearly support the latter interpretation and find the K = 2 component in the wave function of the 2^+_2 state to be 80%. For the yrast sequence, however, the calculation finds an almost pure K = 0 character. The quadrupole moment is predicted to increase to -14 and



FIG. 5. (Color online) Mean-square charge radii (top) and isotope shifts relative to ³⁸Ar (bottom) for the even-mass Ar isotopes. The experimental values are taken from Blaum *et al.* [57] and compared to the GCM(GOA) and to spherical Skyrme SGII Hartree-Fock calculations [58]. The error bars on the experimental data represent the statistical errors only; systematic errors are indicated by the shaded areas.

 $-19 \ e \ \text{fm}^2$ for the 4_1^+ and 6_1^+ states, respectively, showing the development of a sizable prolate deformation and rotational character with higher spin in the ground-state band. It is interesting to note that the axial AMPGCM calculations find a similar behavior with $Q_s(4_1^+) = -6.2 \ e \ \text{fm}^2$ and $Q_s(6_1^+) = -15.4 \ e \ \text{fm}^2$, pointing to a situation in which there is strong mixing of oblate and prolate configurations near the ground state, while prolate deformed states become dominant with increasing spin values along the yrast band.

The onset of deformation in the argon isotopes is also reflected in the recently measured mean-square charge radii obtained in collinear laser spectroscopy [57], which are shown in Fig. 5. The experimental radii show a parabolic trend superimposed on a monotonic increase, which was attributed to deformation effects. This behavior is consistent with the fact that spherical Skyrme Hartree-Fock calculations [58] only reproduce the monotonic increase with neutron number, but not the deformation-related decrease from ⁴⁴Ar to ⁴⁶Ar. The GCM(GOA) calculations, however, are in good agreement with the experimental data, both on an absolute scale and relative to ³⁸Ar. They reproduce in particular the decrease in radius from ⁴⁴Ar to ⁴⁶Ar.

Proton radii for the argon isotopes were also calculated by Lalazissis *et al.* [12]. Only the RMF calculation using BCS pairing reproduces the decrease in the charge radius from ⁴⁴Ar to ⁴⁶Ar, whereas the fully self-consistent RH+B calculation finds a smooth behavior without any pronounced shape effects.

One should again be careful not to draw wrong conclusions about the validity of the description of pairing from this result. Because the RMF+BCS model predicts a change from prolate ground state in ⁴⁴Ar to oblate in ⁴⁶Ar, it is only consistent that this shape transition is also reflected by the radii. Because the RH+B calculation predicts both ⁴⁴Ar and ⁴⁶Ar to be oblate, one would not expect a discontinuity in the radius between these two isotopes. The different behavior of the charge radii in the RMF+BCS and RH+B models is most likely related to the softness of the potential in ⁴⁴Ar than to the treatment of pairing.

V. SUMMARY AND CONCLUSIONS

The development of deformation in neutron-rich $N \approx 28$ nuclei was investigated in a low-energy Coulomb excitation experiment using a radioactive ⁴⁴Ar beam from the SPIRAL facility at GANIL. The Coulomb excitation cross section to the first and second 2^+ state was measured as a function of scattering angle and for two different target materials $(^{109}\text{Ag and }^{208}\text{Pb})$. The three transitional E2 matrix elements connecting the $0_1^+, 2_1^+$, and 2_2^+ states were extracted from the observed γ -ray yields together with the diagonal matrix element for the 2^+_1 state using the coupled-channel leastsquares fitting code GOSIA. The $B(E2; 0^+_1 \rightarrow 2^+_1)$ value is in agreement with the value found in an intermediate-energy Coulomb excitation experiment. The shape connected to the 2_1^+ state is found to be prolate. These results represent the first direct measurement of a quadrupole moment and the first measurement of B(E2) values beyond the 2^+_1 state in this region of neutron-rich nuclei.

The experimental results are compared to shell-model and relativistic mean-field calculations and to both axial and triaxial configuration mixing calculations using the generator coordinate method with the Gogny D1S interaction. Using the latest effective interaction for calculations in the *sd-pf* valence space, SDPF-U, the shell model is in very good agreement with the energies of the 2^+_1 states throughout the region, but cannot reproduce the systematic trend of B(E2) values for the chain of Ar isotopes. Relativistic mean-field calculations describe the collapse of the N = 28 shell closure, but produce ambiguous results concerning the shape and charge radius of ⁴⁴Ar. Configuration mixing calculations including correlations beyond the mean field allow a direct comparison with spectroscopic observables. The angular momentum projected generator coordinate method with the axial quadrupole deformation as generator coordinate and the Gogny D1S interaction describes the properties of the N = 28 isotones well, but finds too large excitation energies and too small B(E2) values for the soft nuclei ⁴²Ar and ⁴⁴Ar. Without taking into account the triaxial degree of freedom, the AMPGCM calculations seem to be biased toward oblate shapes. Five-dimensional configuration mixing calculations using the GCM approach with Gaussian overlap approximation and the Gogny D1S interaction are in better agreement with the experimental excitation energies, but the spectra are still too dilated for the Ar isotopes. The systematic trends of the B(E2) values are reproduced both for the Ar isotopes and for the N = 28 isotones, and the calculations are also able to reproduce the measured quadrupole moment in ⁴⁴Ar. They suggest furthermore a γ -vibrational character of the 2^+_2 state in ⁴⁴Ar. More experimental data on transition strengths and quadrupole moments in neighboring nuclei are clearly needed to draw further conclusions. Their measurement remains a challenging goal for future experiments.

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