First spectroscopy of ^{66}Se and ^{65}As: investigating shape coexistence beyond the $N = Z$ line.

A. Obertelli[a], T. Baugher[b], D. Bazin[b], S. Boissinot[a], J.-P. Delaroche[d], A. Dijon[e], F. Flavigny[a], A. Gade[b,c], M. Girod[d], T. Glasmacher[b,c], G. F. Grinyer[b], W. Korten[a], J. Ljungvall[a], S. McDaniel[b,c], A. Ratkiewicz[b,c], B. Sulignano[a], P. Van Isacker[e], D. Weisshaar[b]

[a] *CEA, Centre de Saclay, IRFU/Service de Physique Nucléaire, F-91191 Gif-sur-Yvette, France*
[b] *National Superconducting Cyclotron Laboratory, East Lansing, USA*
[c] *Michigan State University, East Lansing, USA*
[d] *CEA, DAM, DIF, F-91297 Arpajon, France*
[e] *Grand Accélérateur National d'Ions Lourds, CEA/DSM–CNRS/IN2P3, BP 55027, F-14076 Caen Cedex 5, France*

Abstract

We report on the first γ spectroscopy of ^{66}Se and ^{65}As from two-neutron removal at intermediate beam energies. The deduced excitation energies for the first-excited states in ^{66}Se and ^{65}As are compared to mean-field-based predictions within a collective Hamiltonian formalism using the Gogny D1S effective interaction and to state-of-the-art shell-model calculations restricted to the $pf_{5/2}g_{9/2}$ valence space. The obtained Coulomb-energy differences for the first excited states in ^{66}Se and ^{65}As are discussed within the shell-model formalism to assess the shape-coexistence picture for both nuclei. Our results support a favored oblate ground-state deformation in ^{66}Se and ^{65}As. A shape transition for the ground state of even-odd As isotopes from oblate in ^{65}As to prolate in ^{67,69,71}As is suggested.

Keywords: Spectroscopy, Coulomb-energy differences

Shape coexistence in atomic nuclei is a delicate quantum phenomenon which involves two intrinsic configurations of different deformation that coexist within a few hundreds of keV [1, 2]. The region of krypton and selenium isotopes in the vicinity of self-conjugate nuclei is one of the very few where oblate and prolate deformed states are expected to compete at low excitation energy [3]. Shape coexistence and a shape transition from a prolate ground state in ^{78,76,74}Kr to an oblate one in ^{72}Kr have been established experimentally [4, 5]. The case of the selenium isotopes is less clear. Low-lying excited 0^+ states, characteristic of shape coexistence, are predicted at about 1 MeV in light selenium isotopes [6, 7, 8, 9] but have not been observed so far in $T_z = 0$ and $T_z = 1$ isotopes, in contrast to the krypton case. Shape coexistence is found in ^{72}Se with a low-lying 0^+ state at 937 keV and a strong mixing of intrinsic configurations at low spin [10]. The low-energy Coulomb excitation of ^{70}Se(2^+) [11] together with lifetime measurements of its low-lying states [12] concluded an oblate shape for the ground state. The transition probability $B(\mathrm{E2}; 0^+_1 \rightarrow 2^+_1)$ in ^{68}Se, recently measured from intermediate-energy Coulomb excitation, is in agreement with model predictions of an oblate ground state in the self-conjugate selenium isotope with a strong triaxial component [13]. Several of the above-mentioned studies have been successfully analyzed within a Collective-Hamiltonian formalism considering all quadrupole degrees of freedom [14] and the Gogny D1S interaction [15, 16]. Recently, mirror energy differences between high-spin states in ^{67}Se and ^{67}As were correctly reproduced [17] within a shell-model approach based on the JUN45 shell model interaction [9] in the $pf_{5/2}g_{9/2}$ valence space. A broad and detailed comparison of predictions regarding shapes and shell occupancies from these two different approaches in this mass region is still lacking.

The frontiers of this region of shape coexistence have not yet been determined since, up to now, no experimental information has been collected beyond the $N = Z$ line. In fact, if charge symmetry were to hold exactly, spectra of mirror nuclei with N and Z interchanged should be identical. The question whether charge symmetry holds for the *shape* of mirror nuclei still has to be addressed in detail [18]. Coulomb energy differences (CEDs, defined below) between analog states in mirror nuclei turn out to be extremely sensitive to nuclear structure, as is known mostly for nuclei in the *sd* and low-mass *pf* shells [19]. The recent study of CEDs between states in ^{67}Se and ^{67}As showed indeed a strong sensitivity to the occupation of relevant single-particle orbitals [17]. CEDs bring constrains to theoretical wave-functions and therefore, in the specific case of $N = Z \sim 35$ nuclei, are expected to be related to their shape.

Two-nucleon knockout has been shown in recent years to be a powerful reaction mechanism to populate low-lying states in exotic nuclei. In the case of neutron-deficient nuclei, it has been used to study nuclei in the *sd* shell [20, 21]. In this Letter we show that intermediate-energy beams in the region of light selenium isotopes can be produced with sufficient intensity to investigate the low-lying spectroscopy of the most exotic species. We report on the first γ spectroscopy of ^{66}Se and ^{65}As produced by this technique.

The experiment was performed at the National Superconducting Cyclotron Laboratory (NSCL), at Michigan State University. A cocktail beam containing ^{68}Se (1.5%) and ^{67}As (20%) was produced by fragmentation of a ^{78}Kr primary beam at 150 MeV/nucleon on a 329 mg/cm^2-thick ^{9}Be production target and purified in the A1900 separator [22]. The secondary beam impinged on a 376 mg/cm^2-thick ^{9}Be target for two-nucleon removal. The mid-target energy of ^{68}Se was 78 MeV/nucleon. The target was positioned at the

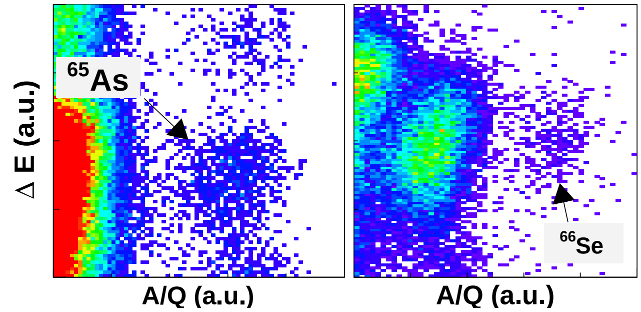

Figure 1: (Color online) Identification of ^{65}As (left) and ^{66}Se (right) in the S800 focal plane in coincidence with incoming ^{67}As and ^{68}Se particles, respectively.

pivot point of the S800 magnetic spectrometer [23] and inside the SeGA segmented HPGe array [24]. For this experiment, SeGA was composed of 17 detectors positioned at 20 cm from the target and arranged in a configuration consisting of two rings with cylindrical symmetry around the beam axis: 7 detectors at 37° in the forward direction and 10 detectors at 90° relative to the beam direction. The energy thresholds of all detectors were set to values below 100 keV. The SeGA photopeak efficiency was 2.49(2) % for a 1-MeV transition emitted at rest in the laboratory frame from the target position. Energy calibrations were performed with ^{152}Eu, ^{60}Co, ^{57}Co and ^{241}Am sources. The identification of incoming particles was performed on an event-by-event basis by a time-of-flight (TOF) measurement between two plastic scintillators located at the image point of the A1900 and at the object point of the S800. A clear separation of incoming particles was performed by correlating the TOF measurement to a dispersion measurement from two position-sensitive parallel-plate avalanche counters (PPACs) at the mid-plane of the S800 analysis beam line. Downstream from the secondary target, reaction products were transmitted to the S800 spectrograph focal plane. The magnetic rigidity of the spectrograph was set to optimize the transmission of the two-neutron knockout residues. Transmitted particles were identified via time of flight and energy loss (see Fig. 1).

The $T_z = -1/2$ nucleus ^{65}As has recently been

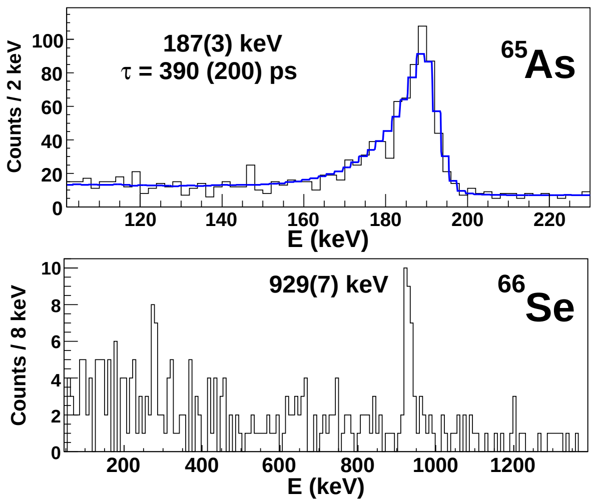

Figure 2: Doppler-effect corrected γ spectra of ^{65}As (top) and ^{66}Se (bottom). In the case of ^{65}As, the best-fit simulated line shape (blue line) that corresponds to a lifetime τ=390 ps is superimposed to the experimental spectrum.

suggested to be particle unbound with a low proton-separation energy of -90(85) keV [25] with still a long-lived ground-state (lifetime of 128(16) ms) due to the Coulomb barrier that slows down the proton decay. It could therefore be located beyond the proton dripline. We observed, for the first time in this nucleus, a prompt γ transition at 187(3) keV (top panel of Fig. 2). The 3-keV error results from the quadratic sum of uncertainties on the energy calibration and the peak-maximum determination in the measured γ spectrum. If the unbound character of ^{65}As is confirmed, this gamma-ray would correspond to a transition between two particle-unbound states. The mirror nucleus ^{65}Ge has a ground state with spin-parity $3/2^-$ and a first-excited $5/2^-$ state [26] at 111 keV. Since ^{65}As is unbound (or very weakly bound), the observed 187-keV line most likely corresponds to a $5/2^-_1 \rightarrow 3/2^-_{gs}$ transition. In this mass region low-energy $3/2^-_1$ and $5/2^-_1$ states compete to be the ground state of even-odd nuclei. In the arsenic isotopes, the inferred ordering in ^{65}As (*i.e.*, a $3/2^-_1$ ground state) represents an inversion compared to $^{67-71}$As (see Fig. 5). This inversion of the ground-state spin has already been noticed from ^{65}Ge to ^{67}As by M. Hasegawa *et al.* and could not been explained in their formalism [27]. Note that the line shape of the measured transition with its low-energy tail can be reproduced by Monte-Carlo simulations assuming a lifetime of 390±200 ps (χ^2 minimization), as shown by the simulated line shape in Fig. 2.

The γ spectrum of ^{66}Se, produced by two-neutron removal from ^{68}Se, is shown in the bottom panel of Fig. 2. Its proton-separation energy is estimated to be 2.0(4) MeV from systematics [29]. One new transition is observed at 929(7) keV. Its full width at half maximum (FWHM) energy is 25 keV, consistent with the expected energy resolution. The 7-keV uncertainty results from the quadratic sum of a 6-keV uncertainty from the choice of the β-velocity for Doppler correction due to low statistics, corresponding to a β uncertainty of 0.005 c, and a 4-keV systematics uncertainty on the transition-energy determination. The latter uncertainty has been obtained from the comparison of extracted energies and known values for a set of transitions in well-known nuclei produced by fragmentation with high statistics in this experiment. Since the mirror nucleus ^{66}Ge has a 2^+_1 level at 957 keV, the 929-keV transition in ^{66}Se can be most likely associated with $2^+_1 \rightarrow 0^+_1$.

In addition to the 929-keV transition, the reader may notice a sizable amount of counts measured at 273(5) keV. The observed "peak" has a FWHM of $\sim$ 10 keV, as expected for a prompt transition of this energy. It contains 15 counts over a background of about 2 counts per 8-keV bin. As in ^{65}As, the two-nucleon removal channel is very clean and no sizable background from random coincidences is measured. What we call "background" here most likely corresponds to Compton events from transitions in ^{66}Se. We performed a statistical analysis of the low-energy part of the gamma spectrum of ^{66}Se. The hypothesis of a linear background from 50 to 500 keV has been tested. The deviation of the measured spectrum to the linear-background hypothesis is calculated assuming a Poisson distribution law. We obtain a deviation of 2.7 σ to the linear-background hypothesis, which corresponds to a 0.7% chance to be a statistical fluctuation. Although this deviation appears significant, due to a potentially more complicated structure of the background and the

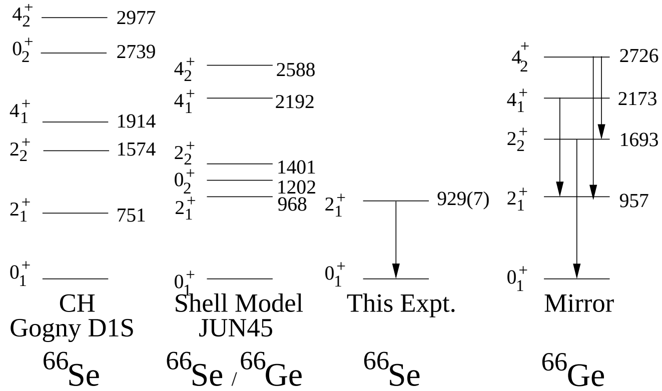

Figure 3: Experimental low-lying spectroscopy of ^{66}Se deduced from this work, compared to shell-model predictions with the JUN45 interaction [9] restricted to the $pf_{5/2}g_{9/2}$ valence space and to CH calculations with the Gogny D1S interaction. Experimental low-lying levels for ^{66}Ge are from [35]. Energies are given in keV. For clarity, only the 2 first 0^+, 2^+ and 4^+, when possible, are shown.

low statistics obtained in this experiment, the observed peak cannot entirely be confirmed as a transition and further experimental evidence has to remain a challenge for future experiments.

We now compare the spectroscopic information obtained for ^{66}Se with beyond-mean-field calculations using the Gogny D1S interaction in a 5-dimensional Collective Hamiltonian (CH) formalism that takes into account quadrupole degrees of freedom. This approach has already given satisfactory results regarding the question of deformation and shape coexistence in Kr and heavier Se isotopes [30]. We further compare our results to shell-model calculations using the JUN45 interaction which was developed to reproduce correctly the spectroscopy of nuclei in the $pf_{5/2}g_{9/2}$ valence space [9]. Both formalisms predict an oblate ground state, with spectroscopic quadrupole moments Q = +0.5 efm^2 for the 2_1^+ state. They also agree on predicting Q = -7 efm^2 for the second 2^+ level. Similar properties are found for the mirror states in ^{66}Ge. The excitation energies of the low-lying $2_{1,2}^+$ and 4_1^+ states are reproduced within 250 keV in both calculations. However, the two formalisms are at variance with respect to the excitation energies predicted for the first excited 0^+ state. Only the shell model calculations is predicting a 0_2^+ level below the 2_2^+ state, suggesting shape coexistence in ^{66}Se. So far, the CH calculations were shown successful in the interpretation of spectroscopic measurements in the region of light Kr and Se isotopes except for isotones in the vicinity of the subshell closure N = 40 where the calculated 0_2^+ energies are too high [34]. In ^{66}Se the 0_2^+ state is predicted at 2.9 MeV, far above that for the 2_2^+ level. This feature is not compatible with a coexistence picture for ^{66}Se, but consistent with the calculated potential energy surface (see Fig. 4) which displays no minima at either prolate and oblate deformation. If a low-lying 0_2^+ level were to be predicted, this would require an extension of the present beyond mean-field approach to include two quasiparticle degrees of freedom [31].

Table 1: Energies and quadrupole moments of low-lying states in ^{66}Se and ^{65}As, calculated in the shell model (SM) with the JUN45 interaction and modified single-particle energies (see text), and (for ^{66}Se) in a collective Hamiltonian (CH) with the Gogny D1S interaction (see text for details).

	J^π	E (keV)			Q (efm^2)	
		Expt	SM	CH	SM	CH
^{66}Se	2_1^+	929(7)	937	751	+5	+5
	2_2^+	—	1378	1574	−7	−7
^{65}As	$\frac{5}{2}{}_1^-$	187	0	691	−39	−53
	$\frac{3}{2}{}_1^-$	0	138	0	+29	+33

In contrast, shell-model calculations with the JUN45 interaction predict a low-lying 0_2^+ at 1202 keV, 234 keV above the 2_1^+ state, both in ^{66}Se and ^{66}Ge. Note that several nuclei in this mass region with $N \gtrsim Z$, such as ^{74,76}Kr or ^{72}Se, have a 0_2^+ state decaying to 2_1^+ by E2. Their transition probabilities obtained from the lifetime of these known 0^+ states lead to lifetimes of 10-400 ps for a ∼250 keV 0_2^+-to-2_1^+. If confirmed, the 273-keV transition could correspond to the decay of such a 0_2^+ state. Is it possible that the mirror 0_2^+ state has not been previously observed in the rather well-known ^{66}Ge? This nucleus has been studied via (i) two-proton transfer from stable ^{64}Zn [32] and (ii)

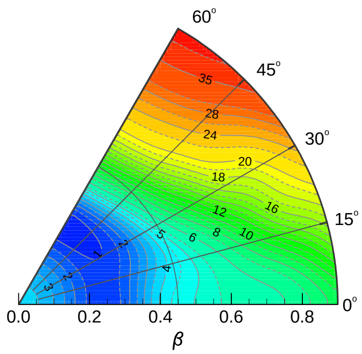

Figure 4: (Color online) Potential-energy surface (MeV) for ^{66}Se from HFB calculations with the Gogny D1S force as a function of quadrupole degrees of freedom β and γ.

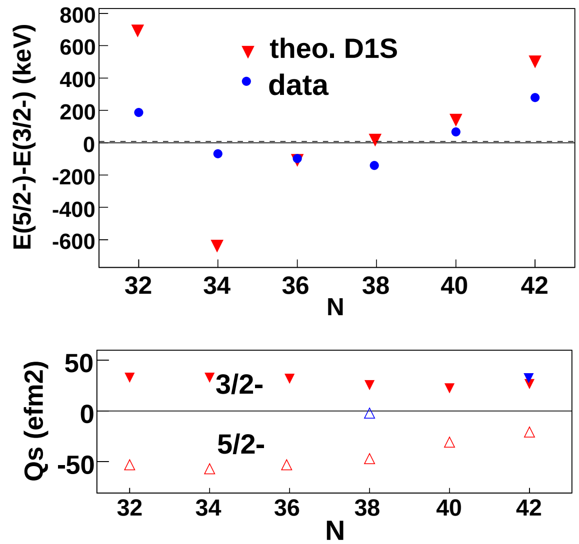

Figure 5: (Color online) (Top) Excitation energy of the $5/2^-_1$ state relative to the first $3/2^-_1$ state in even-odd arsenic isotopes from the dripline nucleus ^{65}As to the stable ^{75}As. (Bottom) Spectroscopic quadrupole moments for blocked-quasiparticle states at $K^\pi = 3/2^-$ (open triangles) and $K^\pi = 5/2^-$ (filled triangles) that minimize the total energy of ^{65}As constrained to axial symmetry.

fusion-evaporation from heavy-ion collisions [33]. The 2^+_2 state has not been observed in two-proton transfer, indicating that the overlap between the ground state of ^{64}Zn and states of an (assumed) 0^+_2 band of ^{66}Ge is small. In fusion-evaporation reactions, the 2^+_2 is populated from higher-spin band members but, assuming the 0^+_2 to lie below 2^+_2, the $2^+_2 \to 0^+_2$ transition may be hindered by the competition with the decay to the 2^+_1 and 0^+_1 states. In other terms, the existing data do not show any low-lying excited 0^+ state in ^{66}Ge and ^{66}Se, and more generally in other neutron deficient Selenium isotopes. We believe that there is still a significant probability that they exist but have not been observed. Further searches for a low-lying 0^+_2 state in ^{66,68}Se and ^{66}Ge, for example via two neutron transfer (p,t), are necessary to clarify the shape coexistence phenomenon in the light Selenium isotopes.

We now turn to the calculation of CEDs defined as $E_< - E_>$ where $E_<$ is the excitation energy of a state in the $T_z < 0$ nucleus and $E_>$ the excitation energy of the corresponding state in the mirror nucleus with $T_z > 0$. They have been obtained in the shell model (with the code ANTOINE [36]) by adding to the proton-proton matrix element of the JUN45 interaction, a Coulomb contribution $V_{\rm c}$ (without screening), calculated in a harmonic-oscillator basis. In a first calculation, identical single-particle energies were assumed for neutrons and protons, as in the JUN45 parametrization. Small CEDs of a few keV were found (see Table 2 under '$V_{\rm c}$'), leading to the conclusion that, in this mass region and for the wave functions as obtained with JUN45, the two-body Coulomb matrix elements play a minor role in the CEDs. In a second calculation, different single-particle energies were assumed for neutrons and protons. In principle, these should be taken from the spectroscopy of ^{57}Ni and ^{57}Cu. The single-particle energies thus determined, however, would have been inconsistent with the JUN45 interaction where they are obtained from a fit [9]. We adopt here the strategy to take the differences $\Delta\epsilon_j \equiv \epsilon^\nu_j - \epsilon^\pi_j$ from the spectroscopy of ^{57}Ni and ^{57}Cu and to adjust the averages $(\epsilon^\nu_j + \epsilon^\pi_j)/2$ to those used by Honma *et al.* [9]. We remark that the differences thus extracted are close (differing by a few 10s of keV) to those of Trache *et al.* [37]. The difference $\Delta\epsilon_{g_{9/2}}$ is not known experimentally but can be estimated to be small and negative following the procedure of Trache *et al.* [37]. The results shown here are obtained with $\Delta\epsilon_{g_{9/2}} = -100$ keV and are not significantly altered by reasonable variations around

Table 2: Coulomb-energy differences (CEDs) for the first-excited states in the mirror pairs ^{66}Se – ^{66}Ge and ^{65}As – ^{65}Ge. The two calculations labeled 'V_c' and '$V_c + E_{sp}$' are explained in the text. For ^{65}As – ^{65}Ge, CEDs are calculated for the $5/2^-_1$ state relative to the $3/2^-_1$ state.

Mirror pair	J^π	CED (keV)		
		Expt	Theory	
			V_c	$V_c + E_{sp}$
^{66}Se – ^{66}Ge	2^+_1	−28(7)	+6	−31
^{65}As – ^{65}Ge	$5/2^-_1$	+76(3)	−3	+60

this value. The single-particle energies adopted for the $p_{3/2}$, $f_{5/2}$, $p_{1/2}$ and $g_{9/2}$ orbitals are 0, 989, 1992 and 3516 keV for the neutrons and 0, 1249, 1986 and 3616 keV for the protons. The resulting CEDs are shown in Table 2 (under '$V_c + E_{sp}$'). For ^{66}Se – ^{66}Ge, the calculated CED is of the same sign and magnitude as the measured value. The CED between the analog 2^+ states in ^{66}As and ^{66}Ge is calculated to be small, −5 keV, close to its experimental value of +7 keV [38]. The calculated excitation energies of the 2^+ states with $T = 1$ in the ^{66}Se – ^{66}As – ^{66}Ge triplet are 938, 964 and 969 keV, respectively, close to the experimental values 929(7), 964 and 957 keV. The calculated ^{65}As – ^{65}Ge CED of 60 keV is strongly dependent on the difference $\epsilon_{f_{5/2}} - \epsilon_{p_{3/2}}$ and, as a consequence, any agreement with the data is rather fortuitous. In other words, our shell-model calculation has no predictive power for the CED between the odd-mass nuclei. In contrast, the calculated $T = 1$ CEDs are more stable against reasonable variations around the single-particle energies deduced from ^{57}Ni and ^{57}Cu. Contrary to $f_{7/2}$-shell nuclei [39], no $J = 2$ anomaly is needed to reproduce these CEDs with our shell-model calculation, without appreciable shape changes among the different analog states. On the other hand, the shape change between mirror nuclei is the basis of the explanation of the CEDs between ^{70}Br and ^{70}Se as proposed by Singh *et al.* [18] and further measurements of CEDs in this mass region are thus needed to clarify the situation. The shell model predicts a prolate $5/2^-$ ground state and a low-lying oblate $3/2^-$ state in ^{65}As and ^{65}Ge. The closeness of two states with different deformation is an indication of shape coexistence but the ordering is reversed from experiment, assuming that the proposed level scheme based on the spectroscopy of ^{65}Ge is correct. The very small $B(\mathrm{M1}; 5/2^-_1 \to 3/2^-_1)$ value of $2.10^{-2}\ \mu_N^2$ calculated by the shell model yields a lifetime of 417 ps to be compared to the 390(200) ps, determined from the measured line shape.

The spectroscopy of even-odd As isotopes has been calculated with a 1-dimension collective Hamiltonian [40], restricted to axial symmetry, with particles blocked successively on states with quantum numbers $K^\pi = 3/2^-$ and $J^\pi = 5/2^-$, K being the projection of the angular momentum on the symmetry axis and π the parity. We assume in the following that the energy difference between the states with blocked K^π can be compared to the experimental energy difference between $3/2^-_1$ and $5/2^-_1$ states. Results are plotted in Fig. 5. The ordering of the states is well reproduced from $N = 32$ (^{65}) to $N = 40$ (^{73}As), including the inversion of ordering from ^{67}As to ^{65}As. Interestingly, along the isotopic chain, the $K^\pi = 3/2^-_1$ states are all calculated to be of oblate deformation, with a spectroscopic quadrupole moment close to 35 efm^2, whereas the $K^\pi = 5/2^-_1$ states are calculated as prolate deformed with $Q \sim -50$ efm^2. This assignment of a prolate deformation to $5/2^-_1$ states and an oblate deformation to $3/2^-_1$ states in even-odd As nuclei is supported by the quadrupole moments $Q = +30(5)$ efm^2 measured for ^{75}As($3/2^-$) and $Q = -1.7(10)$ efm^2 measured for ^{71}As($5/2^-$) [41]. The change in the ordering of the two states signals a shape transition from a prolate ground state in ^{67}As to an oblate ground state in ^{65}As which indicates a dominance of oblate ground states beyond the $N = Z$ line in this mass region.

In summary, we performed the first spectroscopy of the neutron-deficient ^{65}As and ^{66}Se nuclei, populated via two-neutron knockout at intermediate energies. We measured a transition in ^{66}Se at 929(7) keV, tentatively assigned to $2^+_1 \to 0^+_1$, and a transition in ^{65}As at 187(3) keV with a lifetime of 390±200 ps estimated from the gamma-photopeak line shape. Both beyond-mean-field calculations within a collective-Hamiltonian formalism using the D1S Gogny interaction and shell-model calculations using the recent JUN45 interaction in the $pf_{5/2}g_{9/2}$ valence space predict a favored oblate deformation for the ground state of ^{66}Se. The isotope ^{66}Se is the first $T_z = -1$ nucleus in the $A \sim 70$ region for which CEDs have been measured and calculated CEDs, based on the JUN45 interaction, agree with experiment. This agreement strenghtens the reliability of shell-model wave functions for this nucleus and may therefore indirectly support shell-model predictions for an extension shape coexistence beyond the $N = Z$ line in this region. The CH approach, on the other hand does not predict shape coexistence in ^{66}Se with no distinct energy minima in the corresponding potential energy surface, and a high-lying second 0^+ state, well above the shell-model prediction by more than 1 MeV. Based on the accepted spin assignment in the mirror nucleus ^{65}Ge, the ground state of ^{65}As is assumed to be $3/2^-$ and the measured transition is assigned as $5/2^-_1 \rightarrow 3/2^-_{\rm gs}$. This ordering is at variance with the heavier isotopes ^{67,69,71}As. This inversion is reproduced by beyond-mean-field calculations restricted to axial symmetry and interpreted as a shape transition in the ground state of As isotopes from prolate in ^{67,69,71}As to oblate in ^{65}As. The shell model does not predict the correct ordering in ^{65}As but the quadrupole moments of $3/2^-$ and $5/2^-$ are in agreement with the D1S-interaction predictions. The shell-model B(M1) transition-probability prediction between these two states is also in agreement with the measured lifetime. Finally, further experimental investigations are needed to assess the existence, or not, of a 0^+_2 state below the 2^+_2 state. (p,t) transfer reactions in inverse kinematics, as a well known method to populate and identify 0^+ states, could be used to detail the low-lying spectroscopy not only of ^{66}Se but also of ^{68}Se and ^{66}Ge and therefore clarify the shape coexistence phenomenon in this mass region.

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