## Evolution of Collectivity in <sup>72</sup>Kr: Evidence for Rapid Shape Transition

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The transition rates from the yrast  $2^+$  and  $4^+$  states in the self-conjugate  $^{72}$ Kr nucleus were studied via lifetime measurements employing the GRETINA array with a novel application of the recoil-distance method. The large collectivity observed for the  $4^+ \rightarrow 2^+$  transition suggests a prolate character of the excited states. The reduced collectivity previously reported for the  $2^+ \rightarrow 0^+$  transition was confirmed. The irregular behavior of collectivity points to the occurrence of a rapid oblate-prolate shape transition in  $^{72}$ Kr, providing stringent tests for advanced theories to describe the shape coexistence and its evolution.

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Shape coexistence is a prominent phenomenon manifested in atomic nuclei. In contrast to the shapes of molecules, which are governed by the arrangements of the bonding electron pairs, nuclear shapes are determined by a delicate interplay of quantum many-body correlations such as shell effects, pairing, and quadrupole deformation, which can result in nearly degenerate states with different shapes in a single nucleus. This competition of shapes is known to occur widely across the nuclear chart [1], particularly at and near closed shells, where spherical configurations compete with deformed ones that involve particle-hole excitations across shell gaps.

A remarkable feature is that the energy relationship of two coexisting states can evolve drastically within isotopic chains. If the level energies are close enough, the coexistence between the two different configurations can result in eigenstates having strongly mixed configurations. Of particular importance is the identification of such critical states, which play a defining role in characterizing the evolution of nuclear shapes and thus provide stringent benchmarks to test theoretical models.

The increasing availability of rare isotope beams and emerging advanced gamma-ray tracking arrays [2,3] opens new capabilities to investigate shape coexistence of atomic nuclei far from stability. Previous studies have shown that the even-even neutron-deficient Kr isotopes represent one of the unique regions where the ground-state deformation appears to change from prolate (cigarlike) to oblate (pancakelike) shapes as the self-conjugate nucleus  ${}^{72}_{36}$ Kr<sub>36</sub> is approached [4–8]. The occurrence of the oblate ground state in  $^{72}$ Kr has attracted much attention [4,5] in relation to the well-known open question about the predominance of prolate deformation in well-deformed nuclei [9,10]. A variety of nuclear shapes are expected in this mass region due to the complexity of the Nilsson diagram which shows pronounced subshell gaps at nucleon numbers 34 and 36 (oblate), 34 and 38 (prolate), and 40 (spherical) [11,12]. However, a parabolic trend in the excitation energies of the second  $0_2^+$  states in the Kr isotopes [4] suggests that the prolate-oblate shape transition is induced smoothly as a function of neutron number and the inversion occurs at <sup>74</sup>Kr. The enhanced monopole transition strength peaked at <sup>74</sup>Kr supports this picture [4,6], suggesting that the prolate and oblate configurations are significantly mixed in the <sup>74</sup>Kr ground state. For the low-lying excited states, a multiple Coulomb excitation study has determined that the ground-state bands of <sup>74,76</sup>Kr have a prolate character [7,8]. As for <sup>72</sup>Kr at N = Z, the reduced transition probability  $B(E2; 0^+ \rightarrow 2^+)$  was deduced in the intermediate-energy Coulomb excitation study [5]. The result is consistent with various theoretical calculations [11-16] that predict a weakly deformed oblate ground state. However, earlier studies of high-spin states in this nucleus have suggested that a robust prolate band is developed in  $^{72}$ Kr above the medium-spin  $(4-6\hbar)$  yrast states [13,17-19]. These contrasting characteristics point to the occurrence of another type of the oblate-prolate shape transition induced by nuclear rotations at low frequencies [20]. As a crucial step toward a complete picture of this phenomenon, in this work, we study the evolution of collectivity in  $^{72}$ Kr to comprehend the roles of angular momentum and nucleon number on shape coexistence. As discussed later, this nucleus turns out to be located at the critical spot in the drastic prolate-oblate shape phase transition as a function of spin and isospin.

We present a measurement of the lifetimes of the yrast  $2^+$ and  $4^{+}$  states in the self-conjugate nucleus <sup>72</sup>Kr to deduce the reduced transition probabilities  $B(E2; 2^+ \rightarrow 0^+)$  and  $B(E2; 4^+ \rightarrow 2^+)$ . The energy ratio  $E(4^+)/E(2^+)$  of 1.86 observed for  ${}^{72}$ Kr  $[E(2^+) = 709.7 \text{ keV} \text{ and } E(4^+) =$ 1321.4 keV [21]] deviates from the vibrational (rotational) limit of 2.0 (3.3), suggesting an irregular behavior of collective properties of this nucleus. The aim of this work is to provide new insight for the prolate-oblate shape competition in the vicinity of <sup>72</sup>Kr based on the evolution of collectivity in low-spin states. For even-even nuclei along N = Z,  $B(E2; 2^+ \rightarrow 0^+)$  has been measured up to <sup>76</sup>Sr (except for <sup>60</sup>Zn) [21,22], while no B(E2) data are available for the  $4^+ \rightarrow 2^+$  transitions in self-conjugate nuclei above <sup>56</sup>Ni. In the experiment, we employ the Gamma-Ray Energy Tracking In-beam Nuclear Array (GRETINA) [2] and apply a novel implementation of the recoil-distance Doppler-shift technique based on the use of multilayer foils by taking advantage of the excellent position and energy resolution from GRETINA. The present measurement significantly extends the sensitive range of lifetimes that can be covered with a single setup and thus enables us to study several states simultaneously. To demonstrate the method, data are also taken on <sup>74</sup>Kr in the same setup and compared to the precise measurements available for this nucleus [8].

The experiment was performed at the Coupled Cyclotron Facility of the National Superconducting Cyclotron Laboratory (NSCL), Michigan State University. A secondary beam of <sup>74</sup>Kr was produced by fragmentation of a primary <sup>78</sup>Kr beam at 150 AMeV incident on a <sup>9</sup>Be target. The A1900 fragment separator [23] was used to collect and separate the fragments. The <sup>74</sup>Kr beam had a typical intensity of  $1 \times 10^5$  particles per second with a purity of about 40%. Reactions of <sup>9</sup>Be(<sup>74</sup>Kr, <sup>74</sup>Kr $\gamma$ ) and <sup>9</sup>Be(<sup>74</sup>Kr, <sup>72</sup>Kr $\gamma$ ) were used to populate the yrast 2<sup>+</sup> and 4<sup>+</sup> states in <sup>74</sup>Kr and <sup>72</sup>Kr, respectively.

The secondary beam was directed onto a new plunger device [24,25], the TRIple PLunger for EXotic beams (TRIPLEX), which was mounted at the target position of the S800 spectrograph [26]. The TRIPLEX was recently developed by a Cologne-NSCL collaboration [25] and can accommodate up to three target and degrader foils to facilitate advanced techniques in lifetime measurements, including the differential recoil-distance Doppler-shift method [24]. In this study, a 750  $\mu$ m Be target, a 125  $\mu$ m Ta degrader, and a 90  $\mu$ m Ta degrader were used. The target-1st degrader and 1st degrader-2nd degrader

separations were both set to 1 mm to cover lifetimes in the ranges of 1–100 ps. Data at a large target-degrader distance were also taken to evaluate possible background contributions due to reactions on the degraders. The degrader(s)-to-target yield ratios were found to be 10%– 20% for <sup>72,74</sup>Kr. The <sup>72,74</sup>Kr recoil velocities behind each foil were estimated to be  $v/c \approx 0.37$  (target), 0.33 (1st degrader), and 0.29 (2nd degrader), respectively. Reaction products were identified based on time-of-flight and energy-loss measurements using the focal-plane detection system of the S800 [26].

Deexcitation  $\gamma$  rays were measured using seven GRETINA detector modules [2] which consist of 28 Ge crystals in total. Each crystal is divided into 36 segments and provides a total of 37 signals from the segments and the central contact. Doppler-shift corrections for  $\gamma$  rays emitted from recoils in flight were made based on the position information reconstructed from the digitized signals and the subsequent signal decomposition procedure [2]. The tracking information for outgoing particles measured by the S800 was incorporated event by event in the Doppler-shift corrections. The plunger target was placed about 12 cm upstream of the center of GRETINA in order to achieve a good balance between  $\gamma$ -ray detection efficiencies and the varying degrees of Doppler shifts caused by the recoil velocity changes through the plunger device, where the latter is essential for the present method. The four forwardmost detectors covered the laboratory angles of  $20^{\circ}$ -50° with respect to the beam axis and were mainly used to determine lifetimes. The remaining three detectors were placed at around 70°, which provided additional information to identify possible feeding transitions populating the states of interest.

Energy spectra of  $\gamma$  rays obtained for <sup>74</sup>Kr and <sup>72</sup>Kr are shown in Figs. 1(a) and 1(b), respectively. The peaks associated with the  $2^+ \rightarrow 0^+$  and  $4^+ \rightarrow 2^+$  transitions are clearly observed for both nuclei together with characteristic spectral shapes due to the different components (fast, reduced, and slow) depending on the recoil velocities at the time when  $\gamma$  decays occur. Doppler-shift corrections were made by assuming that all  $\gamma$  decays occur just behind the first degrader and, hence, optimized for the reduced peaks which correspond to the known transition energies [21]. If the lifetime ( $\tau$ ) of the state is short, the  $\gamma$  decays occur inside the target. In this case, the Doppler shifts for the fast component reflect the velocity distribution of the decaying ions, which results in either a broad peak ( $\tau < 1$  ps) or an asymmetric spectral shape (1 ps  $< \tau < 10$  ps) depending on the range of the lifetime. For  $^{74}$ Kr, 71(9)% of the 2<sup>+</sup> state population was made through the transitions originating from the higher-lying  $4^+$ ,  $6^+$ , and the second  $2^+_2$ states, where the fast components of the latter two transitions can be seen in the inset of Fig. 1(a). On the other hand, most of the  $2^+$  population in  $^{72}$ Kr is accounted for by the feeding from the  $4^+$  [45(10)%] and tentatively assigned  $3^{-}$  [50(11)%] states [Fig. 1(b) inset].



FIG. 1 (color online). Doppler-shift corrected  $\gamma$ -ray spectra for (a) <sup>74</sup>Kr and (b) <sup>72</sup>Kr compared to the simulated ones (solid curves). The arrows indicate the different components (*f* is fast, *r* is reduced, and *s* is slow) in lifetime measurements. In (b), a reference spectrum simulated with  $\tau(2^+) = 5.6$  ps and  $\tau(4^+) = 0$  ps is also shown (the dotted curve) for <sup>72</sup>Kr.

We first study the case of <sup>74</sup>Kr to test the performance of the present experimental method. Lifetimes of the excited states were determined based on the comparison between the measured and simulated spectra as shown in Fig. 1(a). The simulation is based on our existing GEANT4 code used in previous works [22,27] with modifications to incorporate GRETINA in the present configuration. The least-squares method was employed in the fitting procedure where the variable parameters are the lifetimes of the states, the amplitudes of the simulated spectra, and an exponential background. For the higher-lying states, the adopted lifetime ( $\tau = 0.96$  ps) [8] was assumed for the 6<sup>+</sup> decay, while the lifetime of the  $2^+_2$  state was estimated based on the measured E2 transition strength [7,21]. In the analysis, the lifetime of the  $4^+$  state was first determined and, for the  $2^+$ state, the lifetime was obtained by taking into account the feeding lifetimes of all observed states including the 4<sup>+</sup> state. The sensitivity to the lifetime is afforded by the yield ratios among the three different Doppler-shift components as clearly observed for the  $2^+ \rightarrow 0^+$  peak. For the  $4^+$  state, the slow component is less pronounced; however, the spectral shapes of the fast and reduced components provide the means to determine shorter lifetimes in the range of a few ps. This feature is shown in more detail in Fig. 2(a),



FIG. 2 (color online). The results of the line-shape analysis are shown for the  $4^+ \rightarrow 2^+$  transitions in (a) <sup>74</sup>Kr and (b) <sup>72</sup>Kr. Data are compared to simulated spectra where the  $4^+$  lifetime of <sup>74</sup>Kr (<sup>72</sup>Kr) is set as 4.1, 5.9, and 7.7 ps (1.4, 3.5, and 5.6 ps).

where the sensitivity to different lifetimes is evident in the line shape on the higher-energy side of the fast component as well as the reduced-to-fast peak ratio. The present results are thus determined to be  $\tau(2^+) = 32.2(22)$  ps and  $\tau(4^+) = 5.9(6)$  ps as summarized in Table I. The errors include the statistical and systematic uncertainties, where the latter mainly stems from ambiguities in the feeding effects and the background contributions from the reactions in the degraders. The present results are in excellent agreement with the adopted values of  $\tau(2^+) = 33.8(6)$  ps and  $\tau(4^+) = 5.2(2)$  ps [8].

Figure 1(b) shows the results of the line-shape fit (the solid curve) to the data for  $^{72}$ Kr. The analysis procedure is similar to that applied to  $^{74}$ Kr, while the lifetime of  $2^{+1}_{-0.5}$  ps was determined for the  $(3^{-})$  state [Fig. 1(b) inset] and taken into account. The lifetimes are obtained as 5.6(10) ps and 3.5(7) ps for the  $2^+$  and  $4^+$  states, respectively (Table I). The present result confirms the  $B(E2; 0^+ \rightarrow 2^+)$  value previously determined by intermediate-energy Coulomb excitation [5]. The error for the  $2^+$  lifetime includes the ambiguities of the feeding effects from the  $4^+$  and  $(3^-)$ states. For the  $4^+$  state, a clear absence of the reduced and slow components as well as the asymmetric shape of the fast peak constrained the result as shown in Fig. 2(b). As an independent test, a fit to the total spectrum including the 2<sup>+</sup> and  $4^+$  peaks was also performed to determine both the lifetimes simultaneously. The results were found to be consistent with the above quoted values.

The systematic behavior of the excitation energies of the yrast  $2^+$  and  $4^+$  states and the reduced transition probabilities B(E2) for their decays is illustrated in Fig. 3. The level structures of <sup>74,76,78</sup>Kr are similar to each other [the solid lines in Fig. 3(a)] while both the  $2^+$  and  $4^+$  excitation energies suddenly increase at <sup>72</sup>Kr, which has been ascribed to the lowering of the (unperturbed) oblate  $0^+$  state with respect to the prolate  $0^+$  state which pushes down the ground-state energy [4]. A similar behavior of the  $2^+$  states has been observed in <sup>70,72</sup>Se at N = 36 and 38 [Fig. 3(a)] [28]. As for the  $B(E2; 2^+ \rightarrow 0^+)$  value [Fig. 3(b)], a gradual decrease is evident when moving toward <sup>72</sup>Kr, suggesting increased migration of the weakly

TABLE I. Summary of the lifetimes in <sup>74</sup>Kr and <sup>72</sup>Kr measured in the present work. The transition rates B(E2) are given in units of  $e^2$ fm<sup>4</sup> for the decays of the 2<sup>+</sup> and 4<sup>+</sup> states and compared to the previous results [5,8]. The transition energies are taken from Ref. [21].

A	Transition	$E_{\gamma}$ (keV)	$\tau_{\rm exp}$ (ps)	B(E2) present	B(E2) previous
<sup>74</sup> Kr	$2^+ \rightarrow 0^+$	455.6	32.2(22)	1290(90)	1223(22) [8]
	$4^+ \rightarrow 2^+$	557.7	5.9(6)	2560(260)	2895(111) [8]
<sup>72</sup> Kr	$2^+ \rightarrow 0^+$	709.7	5.6(10)	810(150)	999(129) [5]
	$4^+ \rightarrow 2^+$	611.7	3.5(7)	2720(550)	

deformed oblate shape into the ground states [4,5]. A striking feature is that the present result for  $B(E2; 4^+ \rightarrow 2^+)$  of <sup>72</sup>Kr exhibits a large degree of collectivity similar to that of the prolate-deformed <sup>74,76</sup>Kr. As corroborated from the constant energy spacings between the 2<sup>+</sup> and 4<sup>+</sup> states in the Kr isotopes including <sup>72</sup>Kr, the strong transition rate for the 4<sup>+</sup>  $\rightarrow$  2<sup>+</sup> transition is indicative of the persistence of the prolate character in the low-spin yrast states of <sup>72</sup>Kr. This is in marked contrast to the previously assigned oblate shape associated with the ground state [4].

The measured  $B(E2; 4^+ \rightarrow 2^+)/B(E2; 2^+ \rightarrow 0^+)$  ratio of 3.4 in <sup>72</sup>Kr deviates significantly from typical values ranging from 1.43 (rotor) to 2.0 (vibrator) and cannot be explained by canonical collective models that assume the ground-state shape as a single basis to describe low-lying excitation properties. Therefore, the importance of the shape evolution in the yrast states in <sup>72</sup>Kr should be emphasized, although the proof of this picture represents an experimental challenge because the (intrinsic)  $0^+$  ground-state shape is essentially not accessible in the laboratory frame. In this study, we determined the E2 transition strengths for both the  $2^+ \rightarrow 0^+$  and  $4^+ \rightarrow 2^+$  transitions to investigate the mismatch in quadrupole deformations among the yrast states. The  $B(E2; 2^+ \rightarrow 0^+)$  is found to be significantly reduced relative to the one for the  $4^+ \rightarrow 2^+$  transition which clearly shows that the overlap between the  $0^+$  and  $2^+$  wave functions is small. To put this interpretation on a simple but more quantitative basis, a two-level mixing model [7] is



FIG. 3 (color online). Experimental values of (a) excitation energies of the yrast  $2^+$  and  $4^+$  states in  $^{72-78}$ Kr with N = 36-42 and (b) B(E2) for their decays. The filled (open) squares denote the present results obtained for the  $2^+$  ( $4^+$ ) states in  $^{72,74}$ Kr, while previous data (circles) are taken from Refs. [5,8,21]. The level scheme for the Se isotopes [21] is shown in (a) for comparison.

employed by assuming two regular rotational bands. The quadrupople transition moments of +3.6 eb and -1.5 ebwere used for the prolate and oblate configurations, respectively, to reproduce the measured  $B(E2; 4^+ \rightarrow 2^+)$  values of <sup>72,74</sup>Kr. The above moments are consistent with the values extracted in the previous work for <sup>74</sup>Kr [7]. The experimental mixing amplitudes of 0.10 and 0.48 [4] were used for the prolate configuration in the ground states of <sup>72</sup>Kr and <sup>74</sup>Kr. respectively, while the prolate dominant amplitude of 0.82, as determined for the 2<sup>+</sup> state of <sup>74</sup>Kr [7], was assumed for all the 2<sup>+</sup> and 4<sup>+</sup> excited states of <sup>72,74</sup>Kr. With these parameters, the  $B(E2; 2^+ \rightarrow 0^+)$  [ $B(E2; 4^+ \rightarrow 2^+)$ ] values are obtained as 530  $e^2$  fm<sup>4</sup> (2960  $e^2$  fm<sup>4</sup>) for <sup>72</sup>Kr and 1480  $e^2$ fm<sup>4</sup> (2960  $e^2$ fm<sup>4</sup>) for <sup>74</sup>Kr. This calculation reproduces the trend of the measured B(E2) values well and corroborates the presence of the two coexisting shapes in the vrast states of <sup>72</sup>Kr.

A variety of advanced theoretical studies have recently been performed for <sup>72</sup>Kr in terms of the self-consistent mean-field theory and shell model [12–16,20,29–32], enabling a direct comparison of spectroscopic observables. In Fig. 4, the measured B(E2) values for the yrast states of <sup>72</sup>Kr are compared to the theoretical predictions available to date including the excited Vampir variational approach (EV) [13], beyond mean-field calculations (BMF) [12], Hartree-Fock-Bogoliubov-based configuration mixing



FIG. 4 (color online). The experimental and theoretical values of  $B(E2; 2^+ \rightarrow 0^+)$  (filled symbols) and  $B(E2; 4^+ \rightarrow 2^+)$  (open symbols) are compared for <sup>72</sup>Kr (see text). The gray bar for the  $2^+ \rightarrow 0^+$  transition rate indicates the average of the present and previous [5] data.

calculations (HFB) [14], constrained HFB + the five-dimensional collective Hamiltonian (CHFB+ 5DCH) [29], CHFB + local quasiparticle random phase approximation (LQRPA) [15], and beyond relativistic mean-field studies with the PC-PK1 force (PC-PK1) [16]. These theories consistently predict an oblate shape for the ground state, while calculations for the lowest prolate band [EV(pro), BMF(pro), and LORPA(pro)] are also shown in Fig. 4. The comparison is not trivial due to the large variation of the predicted values, particularly for the  $4^+ \rightarrow 2^+$  transition. However, two remarks can be made. First, when both the ground and yrast  $2^+$  states are predicted to have a robust oblate character, the calculations tend to exhibit smaller collectivity in both transitions as shown by BMF(obl) [12] and HFB [14]. Second, the theoretical results appear to be more consistent with data when the oblate configuration is predicted to mix with the prolate configuration preferentially at the lowest  $0^+$  state in the band [BMF(pro) [12], LORPA(pro) [15], and PC-PK1 [16]]. In the latter case, the band structure evolves into a prolate character at higher spins, yielding the enhanced  $B(E2; 4^+ \rightarrow 2^+)$  values. The above comparison further supports that the evolution of collectivity presently observed for <sup>72</sup>Kr is indeed a signature of the rapid shape transition among the low-spin yrast states.

In summary, an advanced technique of the recoil-distance Doppler-shift method has been developed to measure the absolute transition rates of the low-lying yrast states of <sup>72,74</sup>Kr. The sensitivity in lifetime measurements with fast rare isotope beams has been vastly improved by GRETINA, realizing the first measurements of the lifetimes of the yrast  $2^+$ and 4<sup>+</sup> states in <sup>72</sup>Kr at N = Z. The rapid increase of collectivity in the  $4^+ \rightarrow 2^+$  transition relative to that for the  $2^+ \rightarrow 0^+$  transition suggests that the onset of the oblateprolate shape transition occurs at low spin in <sup>72</sup>Kr, presenting an extreme example of the shape transition in atomic nuclei. This result, together with previous evidence [4-8, 17-19], demonstrates the extreme fragility of the oblate shape in the <sup>72</sup>Kr ground state against adding angular momentum and extra nucleons, thus highlighting that this self-conjugate nucleus is located in a region of rapidly evolving nuclear shapes as a function of both spin and isospin.

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