How Robust is the N = 34 Subshell Closure? First Spectroscopy of ⁵²Ar

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⁵³K(p, 2p) one-proton removal reaction at ~210 MeV/u at the RIBF facility. The 2⁺₁ excitation energy is found at 1656(18) keV, the highest among the Ar isotopes with N > 20. This result is the first experimental signature of the persistence of the N = 34 subshell closure beyond ⁵⁴Ca, i.e., below the magic proton

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number Z = 20. Shell-model calculations with phenomenological and chiral-effective-field-theory interactions both reproduce the measured 2_1^+ systematics of neutron-rich Ar isotopes, and support a N = 34 subshell closure in ⁵²Ar.

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In the shell-model description of atomic nuclei, magic numbers of nucleons correspond to fully occupied energy shells below the Fermi surface [1], and present the backbone of our understanding of nuclei. Scientific advances over the past decades have shown that the sequence of magic numbers established for stable nuclei is not universal across the nuclear landscape [2]. A few prominent examples are the breakdown of the conventional N = 20, 28 magic numbers [3–5] and the emergence of a new N = 16 magic number [6,7] in neutron-rich nuclei. Considerable efforts have been spent to unfold the driving forces behind such shell evolution [8–11].

For rare isotopes, the first 2^+ excitation energy $[E(2_1^+)]$ in even-even nuclei is often the first observable accessible to experiment to characterize shell effects. In a simplified shell model picture, a high $E(2_1^+)$ is interpreted as resulting from nucleon excitations across a large shell gap [12].

Recently, neutron-rich *pf*-shell nuclei have received much attention on both experimental and theoretical fronts with the possible appearance of new subshell closures at N = 32 and 34. A sizable N = 32 subshell closure has been reported in the region from Ar to Cr isotopes based on $E(2_1^+)$ [13–16], reduced transition probabilities $B(E2; 0_1^+ \rightarrow 2_1^+)$ [17,18], and mass [19–21] measurements, although some ambiguity remains due to the newly measured large charge radii of ^{49,51,52}Ca [22], masses of ^{51–55}Ti [23], and the low $E(2_1^+)$ in ⁵⁰Ar [16]. On the other hand, the N = 34 subshell closure has been so far suggested only in ${}^{54}Ca$ [24,25]. In the Ti and Cr isotopes, the systematics of $E(2_1^+)$ [26,27] and $B(E2; 0^+_1 \rightarrow 2^+_1)$ [17,18] show no local maximum and minimum at N = 34. The measured low-lying structure of ⁵⁵Sc [28] indicated a rapid weakening of the N = 34 subshell closure in Z > 20 nuclei. The measured $E(2_1^+)$ of ⁵⁴Ca was 2043(19) keV, ~0.5 MeV lower than ⁵²Ca [24]. Despite this lower $E(2_1^+)$, ⁵⁴Ca was concluded to be a doubly magic nucleus from a phenomenological shell-model interpretation [24], whereas ab initio coupled-cluster calculations indicated a weak N = 34 subshell closure [29]. Very recently, the mass measurements of ${}^{55-57}$ Ca [25] confirmed the N = 34 subshell closure in ⁵⁴Ca. Until now, it is still an open question how the N = 34 subshell evolves below Z = 20 towards more neutron-rich systems, such as ⁵²Ar.

The heaviest Ar isotope with known spectroscopic information so far is ⁵⁰Ar [16]. Phenomenological shell-model calculations [16,30] reproducing the available $E(2_1^+)$ data for neutron-rich Ar isotopes predict a relatively high-lying 2_1^+ state in ⁵²Ar, and suggest that the N = 34 subshell closure in ⁵²Ar is stronger than the one reported for ⁵⁴Ca. In the present Letter, we report on the first spectroscopy of ⁵²Ar, the most neutron-rich even-even N = 34 isotone accessible today and possibly for the next decades. A clear enhancement of $E(2_1^+)$ at N = 34 is found, supporting the persistence of the N = 34magic number in Z < 20 nuclei.

The experiment was performed at the Radioactive Isotope Beam Factory operated by the RIKEN Nishina Center and the Center for Nuclear Study of the University of Tokyo. Radioactive nuclei were produced by fragmentation of a 345 MeV/u ⁷⁰Zn primary beam with an average beam intensity of 240 pnA on a 10-mm-thick rotating Be target. The secondary cocktail beam, magnetically centered on ⁵³K, was identified using the $B\rho$ - ΔE -TOF method [31] in the BigRIPS two-stage fragment separator [32]. The average intensity and purity of the ⁵³K beam were 1.0 particle per second and 0.53%, respectively.

The secondary beam impinged on a 151(1)-mm-thick liquid hydrogen (LH₂) target operated at 18.14 K with a density of 73 kg/m³ to induce one-proton knockout reactions. Two multiwire drift chambers (MDCs) [33], located upstream of the LH₂ target, were used to measure the trajectories of the incoming projectiles. The kinetic energy of 53 K at the entrance of the target was ~245 MeV/u. Its energy loss in the LH_2 target was approximately 70 MeV/u. The LH₂ target was surrounded by a 300-mm-long time projection chamber (TPC), constituting the MINOS device [34]. The tracks of outgoing protons were recorded by the TPC to reconstruct reaction vertices [35]. The measured efficiency to detect at least one of the two protons is 92(3)%with an estimated vertex resolution of 4 mm (FWHM) [35]. Knowledge of reaction vertices allowed precise Doppler correction of deexcitation γ rays measured by the DALI2+ [36] γ -ray spectrometer, which surrounded the MINOS device.

DALI2+ consisted of 226 NaI(Tl) crystals covering polar angles from 15° to 118° with respect to the center of the LH₂ target. Thresholds of NaI(Tl) crystals were set to about 50 keV. Addback was applied when the centers of hit detectors were less than 20 cm apart. For 1 MeV γ rays emitted from nuclei moving at 60% of the velocity of light, the photopeak efficiency and energy resolution with addback were 30% and 11% (FWHM), respectively. DALI2+ was calibrated using ¹³³Ba, ¹³⁷Cs, ⁶⁰Co, and ⁸⁸Y sources yielding a calibration error of 4 keV and a good linearity from 356 to 1836 keV.

Downstream from the LH_2 target, reaction residues were transported to the SAMURAI spectrometer [33] and



FIG. 1. Particle identification plot of reaction residues with the selection of 53 K identified at BigRIPS.

identified with the $B\rho$ - ΔE -TOF method. The $B\rho$ of charged fragments passing through the SAMURAI magnet with a central magnetic field of 2.7 T was reconstructed using two MDCs placed upstream and downstream the magnet [33]. The ΔE and TOF information were provided by a 24-element plastic scintillator hodoscope. Figure 1 shows the particle identification of fragments with the selection of ⁵³K identified at BigRIPS. For Ar isotopes, a 6.6σ separation in Z and a 9.1 σ separation in A were achieved. Over the data taking of seven days, 438 counts of ⁵²Ar were accumulated from ${}^{53}\text{K}(p,2p){}^{52}\text{Ar}$ reactions, in which the kinematics of protons measured by MINOS supported a quasifree scattering reaction mechanism. The reaction loss of ⁵³K in materials along the beam and fragment trajectories was determined by measuring the unreacted ⁵³K. The measured inclusive cross section was 1.9(1) mb.

The Doppler-shift corrected γ -ray energy spectrum of ⁵²Ar is presented in Fig. 2. A clear peak is present in the



FIG. 2. Doppler-shift corrected γ -ray energy spectrum of ⁵²Ar following the ⁵³K(p, 2p) reaction. The fit function to the spectrum (black solid line) includes simulated response functions for the observed transitions (red dotted line) and a double-exponential background (blue dashed line). The significance level (S.L.) is given for the observed transitions. The inset shows the deduced experimental level scheme.

range of 1500-1800 keV, while three structures are visible in the range of 600-900, 1000-1300, and 2000-2500 keV. In order to quantify the significance level of these peaks, we performed the likelihood ratio test by fitting the spectrum of ⁵²Ar using the GEANT4 [37] simulated response functions on top of a double-exponential background. Given the low statistics of the γ -ray spectrum of ⁵²Ar, a Poisson distribution was adopted to describe the fluctuations of each bin, and the double-exponential background line shape was extracted from the ${}^{51}K(p, 2p){}^{50}Ar$ reaction, leaving the magnitude of the background as a free parameter in the fitting. To estimate the systematic uncertainties caused by this background assumption, the spectrum of ⁵²Ar was also fitted using a free double-exponential background, as well as background line shapes extracted from ${}^{54}Ca(p, pn){}^{53}Ca$ and ${}^{55}V(p, 2pn){}^{53}Ca$ reactions. Note that ${}^{53}Ca$ has similar transitions and neutron separation energy (S_n) as ⁵²Ar. As a result, a significance level of 5σ was obtained for the 1656 (18) keV transition. The 2295(39) keV γ line was found to have a significance of 3σ , while the other two structures in the range of 600-900 and 1000-1300 keV both had a significance level of less than 1σ and are therefore not considered in the following analysis. Note that errors of the deduced γ -ray energies shown above include both statistical and systematic uncertainties. The former dominate and the latter mainly originate from the energy calibration uncertainty. Lifetime (τ_{γ}) effects of the excited states on the deduced γ -ray energies are negligible, since Raman's global systematics [38] suggests $\tau_{\gamma} < 2$ ps for the observed two states.

Based on measured γ -ray intensities, the 1656 keV transition is attributed to a direct decay to the ground state. The low statistics do not allow us to conclude any (non) coincidence between the 1656 and 2295 keV transitions from γ - γ correlations. However, the cascade scenario is very unlikely due to the expected low S_n of ⁵²Ar that is more exotic than ⁵⁴Ca. The measured S_n of ⁵⁴Ca is 3840 (70) keV [19]. The 2016 Atomic Mass Evaluation [39] gives an estimated S_n of 2660(850) keV for ⁵²Ar, and excludes the coincidence scenario. The proposed energy level scheme of ⁵²Ar is presented in the inset of Fig. 2. The measured partial cross sections to the 1656 and 2295 keV states are 0.9(2) and 0.4(1) mb, respectively. Assuming no population to other excited states, the cross section to the ground state is deduced to be 0.7(3) mb via subtraction from the inclusive cross section. The quoted uncertainties are dominated by statistical errors, while systematic uncertainties mainly arise from the estimation of MINOS efficiency and the background assumption. All experimental results are summarized in Table I. The 1656 keV state with the higher population is assigned as 2^+_1 . The 2295 keV state decaying directly to the ground state is assigned as 2^+_2 . Further discussions about these spin-parity assignments are given later.

TABLE I. Experimental excitation energies (E_{exp}) and cross sections (σ_{exp}) from the ⁵³K $(p, 2p)^{52}$ Ar reaction in comparison with theoretical calculations. Predicted excitation energies (E_x) , J^{π} , and spectroscopic factors (C^2S_{th}) associated with the removed protons from different orbits (l_j) were obtained using the VS-IMSRG method predicting a ⁵³K $(3/2^+)$ ground state. Theoretical partial cross sections (σ_{th}) were computed using the C^2S_{th} values and beam-energy-weighted average single-particle cross sections $(\langle \sigma_{sp} \rangle)$.

Experiment		Theory					
E _{exp} (keV)	σ_{exp} (mb)	E_x (keV)	J^{π}	l_j	$C^2 S_{\rm th}$	$\langle \sigma_{ m sp} angle$ (mb)	$\sigma_{\rm th}$ (mb)
0	$0.7(3)^{a}$	0	0_{1}^{+}	$d_{3/2}$	0.28	3.03	0.86
1656(18)	0.9(2)	1849	2_{1}^{+}	$s_{1/2}$	0.10	0.92	1.13
			1	$d_{3/2}$	0.33	2.94	
				$d_{5/2}$	0.02	4.82	
		1974	0_{2}^{+}	$d_{3/2}$	0.01	2.93	0.04
2295(39)	0.4(1)	2367	2^{-}_{2}	$s_{1/2}$	0.13	0.92	0.30
			-	$d_{3/2}$	0.05	2.91	
				$d_{5/2}$	0.01	4.76	
Inclusive	1.9(1)			7			2.32

^aDeduced by assuming no population to other excited states except the 1656- and 2295-keV state as described in text.

Figure 3 displays the measured $E(2_1^+)$ in ⁵²Ar alongside values for lighter Ar isotopes [40]. Notably, the measured $E(2_1^+) = 1656(18)$ keV in ⁵²Ar is the highest among the Ar isotopes with N > 20. It is larger than the $E(2_1^+) =$ 1554(1) keV [41] in ⁴⁶Ar which reflects the conventional N = 28 shell closure. Moreover, the measured systematics of $E(2_1^+)$ along the Ar isotopic chain is characterized by a pronounced enhancement at N = 34 relative to its N = 32even-even neighbor, unlike the trend observed for Ca, Ti, and Cr isotopes in which a decrease is seen from N = 32 to 34. Our results offer the first experimental signature of the N = 34 subshell closure in ⁵²Ar.

To gain further insight into the structure of ⁵²Ar, we compare the results to state-of-the-art nuclear structure calculations. Here, two advanced *ab initio* approaches are adopted: the valence-space in-medium similarity renormalization group (VS-IMSRG) [48-51] (for calculational details, see in particular Refs. [42,51]) and coupled-cluster theory [52,53], employing two sets of two- (NN) and threenucleon (3N) interactions derived from chiral effective field theory [54,55]: 1.8/2.0(EM) [42–44] and N²LO_{sat} [45]. The coupled-cluster method is well suited for closed (sub-)shell nuclei and their neighbors. By employing a double-charge exchange equation-of-motion (DCE-EOM) coupled-cluster technique, the $E(2_1^+)$ in ^{40,48,52}Ar are obtained from generalized excitations of the ground states of the closed (sub-) shell nuclei 40,48,52 Ca, while the $E(2^+_1)$ in 44 Ar is obtained from excitations of the ⁴⁴S ground state, respectively.



FIG. 3. Experimental 2_1^+ energies for even-even Ar isotopes compared with theory: VS-IMSRG with the chiral interaction 1.8/2.0(EM) [42–44] and N²LO_{sat} [45], coupled-cluster calculations (CC) using the DCE-EOM and 2PR-EOM methods with the 1.8/2.0(EM) interaction, and large-scale shell model (LSSM) calculations with the SDPF-U [46] and the so-called *original* [47] and *modified* [30] SDPF-MU interactions. The hatched regions represent the theoretical uncertainties in coupled-cluster calculations. Experimental data are taken from Refs. [40,41] and this work. Note the broken *y*-axis scale between 2.2 and 2.7 MeV.

The $E(2_1^+)$ in ^{46,50,52}Ar are also computed using the twoparticle-removed equation-of-motion (2PR-EOM) coupledcluster method [56]. The measured $E(2_1^+)$ in ⁵²Ar offers a rather unique case to compare these two coupled-cluster methods.

In this work, we employ the DCE-EOM coupled-cluster calculations with particle-hole excitations truncated at the singles, doubles, and approximate triples level (CCSDT-3) [57], while the 2PR-EOM coupled-cluster calculations are truncated at the three-hole–one-particle excitation level using CCSD and CCSDT-3 for the ground states of ^{48,52,54}Ca. Theoretical uncertainties in coupled-cluster calculations are estimated by comparing results with and without triples excitations. In addition, we also compare our results to large-scale shell model calculations with the SDPF-U [46] and SDPF-MU [47] interactions. Note that the original SDPF-MU Hamiltonian was modified using recent experimental data on exotic Ca [24] and K [58] isotopes and details of the modifications are given in Ref. [30].

Theoretical (p, 2p) cross sections to different final states of ⁵²Ar are computed with spectroscopic factors calculated with the VS-IMSRG method using the 1.8/2.0 (EM) interaction and single-particle cross sections (σ_{sp}) calculated using the Glauber theory as described in Ref. [59]. The input of the σ_{sp} calculations are the nucleon-nucleon cross sections, using the parametrization from Ref. [60], and the nuclear ground-state densities deduced from a mean-field Hartree-Fock-Bogoliubov calculation using the SLy4 interaction. The involved single-particle states are calculated using a Woods-Saxon potential including the Coulomb and spin-orbit terms with parameters chosen to reproduce the proton separation energies. The range of the Woods-Saxon potential was taken as $R = r_0(A - 1)^{1/3}$ fm with $r_0 = 1.25$ fm, and the diffuseness is chosen as 0.65 fm. The strength of the spin-orbit potential is set to -6 MeV. Since the reaction vertices were reconstructed with MINOS, the energy dependence of the cross section was considered by taking the average of $\sigma_{\rm sp}$ at different incident energies weighted by observed statistics ($\langle \sigma_{\rm sp} \rangle$). As an illustration, σ_{sp} for the removal of a $d_{3/2}$ proton in ⁵³K to the ground state of ⁵²Ar varies from 2.38 mb at 180 MeV/u to 3.64 mb at 246 MeV/u. Table I lists theoretical results for states lying below the extrapolated S_n of ⁵²Ar [39]. As shown in Table I, the measured cross sections to the 1656 and 2295 keV states in ⁵²Ar agree well with the predictions for the 2_1^+ state at 1849 keV and the 2_2^+ state at 2367 keV, respectively. The ratio of the experimental cross section to the theoretical prediction is in line with the systematic reduction factor reported from (e, e'p)measurements on stable targets [61] and from (p, 2p)reactions on oxygen isotopes [62,63]. The good agreement between experiment and theory not only supports the spinparity assignments, but also indicates that the VS-IMSRG approach with the 1.8/2.0 (EM) interaction provides a satisfactory description of the structure of ⁵²Ar.

We now discuss the systematics of $E(2_1^+)$ in Ar isotopes. As seen in Fig. 3, phenomenological shell-model calculations with the SDPF-U and SDPF-MU interactions as well as the VS-IMSRG calculations with the 1.8/2.0 (EM) interaction reproduce the steep rise of $E(2_1^+)$ from ⁵⁰Ar to ⁵²Ar. The SDPF-U calculations and the modified SDPF-MU calculations provide the best overall description of the experimental data along the Ar isotopic chain including the $E(2_1^+)$ of ⁵²Ar. The VS-IMSRG approach using the 1.8/2.0 (EM) interaction reasonably reproduces the measured $E(2_1^+)$ in neutron-rich Ar isotopes, though an overprediction is seen between N = 28 and 34. The dependence of the ab initio calculations on the initial NN and 3N forces is illustrated by the VS-IMSRG calculations with the N²LO_{sat} Hamiltonian. Compared to results with the 1.8/2.0 (EM) interaction, the N²LO_{sat} Hamiltonian systematically underpredicts the data, despite a better agreement at N = 28 and 30. The DCE-EOM calculations with the 1.8/2.0 (EM) interaction reproduce the $E(2_1^+)$ in ^{44,48}Ar within the estimated uncertainties, but underestimate the $E(2_1^+)$ in ⁵²Ar by ~600 keV. The 2PR-EOM result for ⁵²Ar is consistent with the DCE-EOM calculation, but fails to reproduce the steep increase of $E(2_1^+)$ from ⁵⁰Ar to ⁵²Ar. We note that 2PR-EOM gives a $E(2_1^+)$ at 3.0 MeV for ⁴⁶Ar, consistent with the N =28 shell closure but almost twice the experimental value. For ⁴⁰Ar, which is characterized by deformation and shape coexistence [64], all considered calculations underestimate its $E(2_1^+)$.

Despite being rooted in the same chiral effective interaction, namely, 1.8/2.0 (EM), the coupled-cluster and VS-IMSRG approaches predict very different behaviors about the change of $E(2_1^+)$ from ⁵⁰Ar to ⁵²Ar. However, for closed (sub-)shell Ca isotopes, theses two calculations give consistent results. The differences in calculated $E(2_1^+)$ in neutron-rich Ar isotopes indicate that the total theoretical uncertainties might be larger than the estimated error bars shown in Fig. 3. The observed steep rise of the $E(2_1^+)$ from ⁵⁰Ar to ⁵²Ar serves as an important benchmark to understand the uncertainties of the employed many-body methods.

It is worth noting that the modified SDPF-MU shell model calculations and the VS-IMSRG approach using the 1.8/2.0 (EM) interaction have both been used along the N = 34isotonic chain to investigate the shell evolution. Both calculations suggest that the N = 34 shell gap persists from ⁵⁴Ca towards more exotic N = 34 isotones, which is consistent with the measured high-lying 2_1^+ state in ${}^{52}Ar$ presented here. However, we would like to emphasize that there is no direct correlation between the measured $E(2_1^+)$ and the size of the shell gap which is defined as the difference between the effective single-particle energies (ESPEs), since the latter is not an observable. Indeed, calculations predicting similar $E(2_1^+)$ might give different magnitudes of shell gaps. As is the case here, the ESPEs extracted by the modified SDPF-MU calculations using the method described in Ref. [65] indicate that the N = 34 shell gap in ⁵²Ar (\sim 3.1 MeV) exceeds that in ⁵⁴Ca (\sim 2.6 MeV) [16]. Conversely, the ESPEs calculated by the VS-IMSRG approach using the method of Ref. [66] suggest the N =34 shell gap in 52 Ar (~2.6 MeV) is smaller than that in 54 Ca (~3.2 MeV). In addition, the VS-IMSRG approach also provides the orbital occupancies of the 0_1^+ and 2_1^+ states in 52 Ar and 54 Ca. It reveals that in the 2^+_1 excitation of 52 Ar only ~0.5 neutrons are excited from $p_{1/2}$ to $f_{5/2}$ and proton excitations also contribute due to the open proton shell, whereas in the case of ${}^{54}Ca$, ~0.9 neutrons are excited across the N = 34 shell gap. This is consistent with the observed decrease in $E(2_1^+)$ between ⁵⁴Ca and ⁵²Ar. Nevertheless, both calculations predict ⁴⁸Si as a new doubly magic nucleus. The $E(2_1^+)$ of ⁴⁸Si in SDPF-MU [30] and VS-IMSRG [67] calculations lies at 2.85 and 3.13 MeV, respectively. However, it is not yet known whether the ⁴⁸Si ground state and its 2^+_1 state are stable against neutron emission. Mass models that reproduce well the observed limits of existence in the *pf*-shell region [68] tend to predict 48 Si as a drip-line nucleus in which continuum effects might also play an important role in the structure of ⁴⁸Si.

To summarize, the low-lying structure of 52 Ar was investigated using the 53 K(p, 2p) 52 Ar reaction at ~210 MeV/u. The measured 2^+_1 state lies at 1656(18) keV, the highest among the Ar isotopes with N > 20. The measured (p, 2p)cross sections to different final states of 52 Ar agree with calculations and support the proposed spin-parity assignments. Shell-model calculations with phenomenological and the chiral interaction 1.8/2.0 (EM) both reproduce the measured 2_1^+ systematics of the neutron-rich Ar isotopes, and suggest a N = 34 subshell closure in 52 Ar. However, coupled-cluster calculations based on the same chiral interaction underestimate the 2_1^+ excitations in 52 Ar. The measured $E(2_1^+)$ of 52 Ar serves as an important benchmark to understand the uncertainties of the employed many-body methods. Our results offer the first experimental signature of the persistence of the N = 34 subshell closure below Z = 20, and agree with shell-model calculations predicting 48 Si as a new doubly magic nucleus far from stability.

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