Mass Measurements Demonstrate a Strong N = 28 Shell Gap in Argon

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We present results from recent time-of-flight nuclear mass measurements at the National Super-conducting Cyclotron Laboratory at Michigan State University. We report the first mass measurements of 48 Ar and 49 Ar and find atomic mass excesses of -22.28(31) MeV and -17.8(1.1) MeV, respectively. These masses provide strong evidence for the closed shell nature of neutron number N=28 in argon, which is therefore the lowest even-Z element exhibiting the N=28 closed shell. The resulting trend in binding-energy differences, which probes the strength of the N=28 shell, compares favorably with shell-model calculations in the sd-pf shell using SDPF-U and SDPF-MU Hamiltonians.

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The "magic" numbers of protons and neutrons, which enhance nuclear binding for isotopes near the valley of β stability, can evolve for more neutron-rich or neutrondeficient nuclei [1–3]. The neutron magic number N=28has been the subject of extensive recent experimental and theoretical investigations [4–8]. Since neutron-rich N=28nuclei are within experimental reach and are computationally tractable for shell-model calculations, they are ideal candidates for illuminating the fundamental forces at work in exotic nuclei. It is known that the N=28 shell gap, which stabilizes doubly magic ${}^{48}_{20}$ Ca₂₈, is absent in the Z = 14 and Z = 16 isotopic chains at ${}^{42}_{14}Si_{28}$ [9–12] and $^{44}_{16}S_{28}$ [13–17]. Experimental information on the structure of ⁴⁰Mg₂₈ suggests it has a prolate deformed ground state [18], which would be consistent with the absence of a neutron shell gap.

The existence of the N=28 shell gap for argon is a matter of some controversy. Several previous experimental studies have assessed the shell structure of neutron-rich argon [19–29]. Investigation of the energy of the lowest excited states of $^{45}_{18}\mathrm{Ar}_{27}$ via β -decay spectroscopy of $^{45}_{17}\mathrm{Cl}_{28}$ suggested a weakened, but still present, N=28 shell closure for argon [21]. The first 2^+ state energies $E(2^+_1)$ along the argon isotopic chain [25,26,30] and information on neutron single-particle structure from transfer [23,24] and knockout [22] reactions are consistent with the

presence of an N=28 shell gap in $_{18}^{46}$ Ar₂₈. Though disagreement exists as to the inferred nuclear structure from measurements of the ${}^{46}_{18}\mathrm{Ar}_{28}$ quadrupole excitation strength, $B(E2, 0_1^+ \rightarrow 2_1^+)$, written as B(E2) hereafter for brevity. Three projectile Coulomb excitation measurements, two at intermediate energies [19,20] and one at Coulomb-barrier beam energy [29], deduce a low B(E2), corresponding to a reduced quadrupole collectivity. In this case quadrupole collectivity reflects a propensity for neutrons to be excited across the N=28 shell gap, and thus a low B(E2) may be expected for a semimagic nucleus. State-of-the-art shellmodel calculations that properly account for the breakdown of the N=28 magic number in silicon and sulfur isotopes predict a markedly higher B(E2) for ⁴⁶Ar [28]. A lowstatistics lifetime measurement of the 2⁺₁ state of ⁴⁶Ar deduced a high B(E2) value in agreement with theory [27], but at odds with the three consistent, independent Coulomb excitation measurements [19,20,29].

However, B(E2) measurements are not necessarily unambiguous probes of neutron shell structure, since they are sensitive to proton degrees of freedom and protonneutron interactions. In contrast, mass measurements, and the neutron separation energies derived from them, directly probe the neutron shell gap in a model-independent way.

We report here results from the first [31] mass measurements of ⁴⁸Ar and ⁴⁹Ar, which provide robust evidence for

the persistence of the N=28 shell gap for argon. These results were obtained with the time-of-flight (TOF) technique at the National Superconducting Cyclotron Laboratory [33–35]. Neutron-rich isotopes of silicon to zinc were produced by fragmentation of a 140 MeV/u 82 Se primary beam impinging on a beryllium target. A target thickness of 517 mg/cm² was used to produce less neutron-rich nuclei, required for calibration, whereas a target thickness of 658 mg/cm² was used to produce the more neutron-rich fragments of interest. The fragments were transmitted through the A1900 fragment separator [36] to the focal plane of the S800 spectrograph [37]. A 7.5 mg/cm² Kapton wedge degrader was used in the A1900 to remove the high flux of low-Z nuclei that would otherwise complicate fragment identification. The thick and thin targets were used alternately, while the magnetic rigidity $B\rho$ of the A1900 beam line and the S800 were left unchanged. This allowed us to measure the TOF for nuclei with a broader range of mass-to-charge ratios m/q. By design, the lower m/q isotopes observed generally had well-known masses and could be used to calibrate the relationship between m/q and TOF, whereas the higher m/q nuclei observed generally had unknown masses. TOF was measured over a 60.6 m flight path using fast timing scintillators located at the A1900 and S800 focal planes. A typical TOF was ≈500 ns. The finite momentum spread of the beam, limited to $\delta p/p = \pm 0.5\%$ by slits in the A1900, made a precise measurement of $B\rho = p/q$ necessary for each nucleus produced. $B\rho$ was measured by detecting the position of each ion at a dispersive focus at the S800 target position. Position measurements were performed by collecting electrons emitted from a gold foil due to passing beam particles on a position sensitive microchannel plate detector [38]. The energy loss measurement obtained from the ionization chamber in the S800 focal plane combined with TOF provided fragment identification.

In principle, the simultaneous measurement of an ion's TOF, charge q, and $B\rho$ through a magnetic system of a known flight path L_{path} directly yields its mass, $m_{\text{rest}} =$ $(TOF/L_{path})(q(B\rho)/\gamma)$, where γ is the Lorentz factor. However, in practice neither L_{path} nor the ion optical dispersion used to determine $B\rho$ are known with sufficient precision. Furthermore, only a measurement of $B\rho$ relative to the central ion optical axis is performed. Therefore, the $(m_{\rm rest}/q)$ (TOF) relationship is determined empirically using reference nuclei with well-known masses [35]. The TOF distributions for reference nuclei and ^{48,49}Ar are shown in Fig. 1. Twenty reference nuclei with masses known to better than 100 keV and no known isomeric states longer lived than 100 ns [32,39,40] were fitted with a 7-parameter calibration function of second order in TOF, first order in TOF*Z, and containing first, second, and fourth order Z terms. The calibration function represents a minimal set of terms that minimized the overall fit residual to literature masses and resulted in no detectable systematic

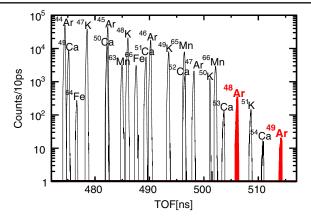


FIG. 1 (color online). Rigidity-corrected time-of-flight distributions for reference nuclei (unfilled histograms) used to calibrate the $(m_{\rm rest}/q)({\rm TOF})$ relationship to obtain masses from TOFs of $^{48}{\rm Ar}$ and $^{49}{\rm Ar}$ (red-filled histograms).

biases [33], as seen in Fig. 2. Note that the apparently deviant point ⁵⁴Ca in Fig. 2 does not significantly impact the results of the mass fit, due to its large statistical uncertainty. A systematic uncertainty of 9.0 keV/q was included as described in [33] to normalize the χ^2 per degree of freedom of the mass fit to one. Two additional uncertainties related to the extrapolation were added to the final mass uncertainties, one to reflect the uncertainties in the TOFs of reference nuclei, which leads to an uncertainty in the fit coefficients of the (m_{rest}/q) (TOF) relation, and one to reflect the uncertainty inherent in choosing a particular calibration function over another which has a comparable goodness of fit. The latter was determined by investigating the robustness of the results to adding additional terms to the calibration function. The total mass uncertainty is a sum in quadrature of statistical,

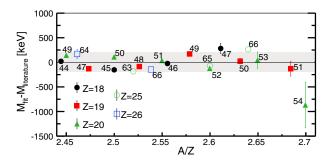


FIG. 2 (color online). Residuals of the fit to the time-of-flight of calibration nuclei (discussed further in the text) as a function of the mass number to nuclear charge ratio A/Z. Isotopes are labeled with their mass number and symbols indicate the elements (solid circle for argon, solid square for potassium, solid triangle for calcium, open circle for manganese, and open square for iron). Calibration masses were fit to within $9 \ \text{keV}/q$ without any systematic trends. The gray band shows the average systematic mass uncertainty included for reference nuclei as described in [33].

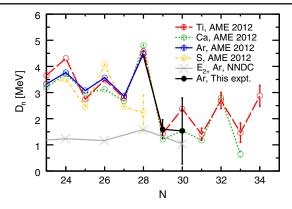


FIG. 3 (color online). D_n [42] as a function of neutron number N near N=28 for sulfur (dot-dashed line), argon (solid line), calcium (dotted line), and titanium (dashed line). The previously known [32] argon trend (solid line, open circles) is shown along with results from this experiment (solid line, solid circles). $E(2_1^+)$ energies [25,26,30] are shown for comparison (crosses). The peak at N=28 followed by a reduction in D_n for N>28 as compared to N<28 indicates the presence of a closed shell. From shell-model calculations we conclude the transition from $D_n\approx 3$ MeV for N<28 to $D_n\approx 1.5$ MeV for N>28 corresponds to the transition from filling the $f_{7/2}$ orbit to filling the $p_{3/2}$ orbit.

systematic, and two extrapolation uncertainties. The relative contribution of the extrapolation uncertainties becomes larger as the distance in m/q and Z from reference nuclei increases.

The atomic mass excesses obtained for ⁴⁸Ar and ⁴⁹Ar were -22.28(31) MeV and -17.8(1.1) MeV, respectively. This corresponds to a measurement precision of $\delta m/m \approx 10^{-5}$. These masses can now be used as a probe of shell structure [41]. Typically, binding-energy differences of neutron-rich nuclei are examined for this purpose in order to isolate the impact of adding neutrons. One such probe that is frequently used is the two-neutron separation energy S_{2n} . $S_{2n}(Z,A) = 2 \times ME_{neutron} + ME(Z,A-2)$ ME(Z,A), where ME is the mass excess, represents the energy required to remove two neutrons from a nucleus with Z protons and A-Z neutrons. Along an isotopic chain, S_{2n} generally declines with increasing N due to the liquid-drop aspect of nuclear binding that penalizes a large neutron-proton asymmetry. This decline is markedly increased following a nucleus that exhibits a magic neutron number. However, the change in slope that indicates a shell closure is not always easy to interpret. A recently introduced quantity D_n [42], where $D_n(Z,A) = (-1)^{N+1} \times$ $[S_n(Z, A+1) - S_n(Z, A)]$, provides a more readily recognizable signature of a shell closure. In a given mass region, D_n indicates the number of orbital angular momentum projection "m" states that participate in pairing for a given nucleus. A peak in D_n at a certain neutron number along with a change in the D_n level before and after that neutron number indicates a shell gap [42]. The change in the D_n

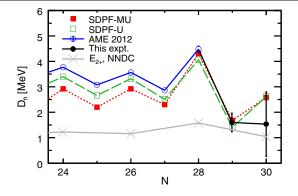


FIG. 4 (color online). The D_n [42] trend near N=28 for argon from currently known masses [32] (open circles) and this experiment (solid circles) is shown along with shell-model calculations employing the SPDF-MU Hamiltonian [7] (solid squares) and the SDPF-U Hamiltonian [5] (open squares). $E(2_1^+)$ energies [25,26,30] are shown for comparison (crosses).

level is a crucial element since it indicates a transition from filling one "m" state to filling another.

The D_n values for argon isotopes from this work show a clear signature for an N=28 shell closure (Fig. 3). With the new mass excesses for 48,49 Ar, it is apparent that neutron-rich argon displays the same systematics in D_n as calcium and titanium, which are known to exhibit an N=28 shell gap [4]. As seen in Fig. 3, sulfur does not peak at N=28 [14], which is consistent with prior conclusions that sulfur does not exhibit the N=28 closed shell [13]. Based on our experimental data we can therefore conclude that argon is the lowest even-Z element with a closed neutron shell for N=28.

We compare the experimental D_n trend to the D_n trends for local mass predictions obtained from shell-model calculations using the SDPF-U [5] and SDPF-MU [7] interactions in Fig. 4. In both cases there is excellent agreement between experiment and theory. This indicates current shell-model calculations adequately describe the interaction between core and valence neutrons around N=28 for argon.

In summary, we performed the first mass measurements of 48 Ar and 49 Ar via the time-of-flight technique. We find the N=28 closed shell is present for argon, which makes argon the lowest even-Z element that exhibits an N=28 shell gap. Based on this result we can conclude that the problems of shell model calculations in describing electromagnetic observables in argon isotopes near N=28 are not related to the neutron shell gap, but instead point to issues with the interaction of valence neutrons and core protons.

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- [1] B. Brown, Prog. Part. Nucl. Phys. 47, 517 (2001).
- [2] O. Sorlin and M.-G. Porquet, Prog. Part. Nucl. Phys. 61, 602 (2008).
- [3] R. Cakirli and R. Casten, Int. J. Mass Spectrom. 349–350, 187 (2013).
- [4] O. Sorlin and M.-G. Porquet, Phys. Scr. **T152**, 014003 (2013).
- [5] F. Nowacki and A. Poves, Phys. Rev. C 79, 014310 (2009).
- [6] L. Gaudefroy, Phys. Rev. C 81, 064329 (2010).
- [7] Y. Utsuno, T. Otsuka, B. A. Brown, M. Honma, T. Mizusaki, and N. Shimizu, Phys. Rev. C 86, 051301(R) (2012).
- [8] E. Caurier, F. Nowacki, and A. Poves, Phys. Rev. C 90, 014302 (2014).
- [9] B. Bastin et al., Phys. Rev. Lett. 99, 022503 (2007).
- [10] C. M. Campbell et al., Phys. Rev. Lett. 97, 112501 (2006).
- [11] S. Takeuchi et al., Phys. Rev. Lett. 109, 182501 (2012).
- [12] S. R. Stroberg et al., Phys. Rev. C 90, 034301 (2014).
- [13] T. Glasmacher et al., Phys. Lett. B 395, 163 (1997).
- [14] F. Sarazin et al., Phys. Rev. Lett. 84, 5062 (2000).
- [15] L. Gaudefroy et al., Phys. Rev. Lett. 102, 092501 (2009).
- [16] C. Force et al., Phys. Rev. Lett. 105, 102501 (2010).
- [17] D. Santiago-Gonzalez et al., Phys. Rev. C 83, 061305(R) (2011).
- [18] H. L. Crawford et al., Phys. Rev. C 89, 041303(R) (2014).
- [19] H. Scheit et al., Phys. Rev. Lett. 77, 3967 (1996).
- [20] A. Gade et al., Phys. Rev. C 68, 014302 (2003).
- [21] S. Grévy et al., Nucl. Phys. A722, C424 (2003).
- [22] A. Gade et al., Phys. Rev. C 71, 051301(R) (2005).
- [23] L. Gaudefroy et al., Phys. Rev. Lett. 97, 092501 (2006).
- [24] L. Gaudefroy et al., Phys. Rev. C 78, 034307 (2008).

- [25] S. Bhattacharyya et al., Phys. Rev. Lett. 101, 032501 (2008).
- [26] A. Gade et al., Phys. Rev. Lett. 102, 182502 (2009).
- [27] D. Mengoni et al., Phys. Rev. C 82, 024308 (2010).
- [28] R. Winkler et al., Phys. Rev. Lett. 108, 182501 (2012).
- [29] S. Calinescu et al., Acta Phys. Pol. B 45, 199 (2014).
- [30] S. Raman, C. W. Nestor, Jr., and P. Tikkanen, At. Data Nucl. Data Tables 78, 1 (2001).
- [31] The review of shell structure in the N=28 region [4] lists the mass of ${}^{48}_{18}\text{Ar}_{30}$ as measured, though the link they cite pointing to an AME version from 2011 is no longer in operation. The more recent 2012 Atomic Mass Evaluation [32] lists the mass of ${}^{48}_{18}\text{Ar}_{30}$ as unmeasured and we are unable to find experimental data for this mass after a detailed search of the literature.
- [32] G. Audi, M. Wang, A.H. Wapstra, F.G. Kondev, M. MacCormick, X. Xu, and B. Pfeiffer, Chin. Phys. C 36, 1287 (2012).
- [33] M. Matoš et al., Nucl. Instrum. Methods Phys. Res., Sect. A **696**, 171 (2012).
- [34] A. Estradé et al., Phys. Rev. Lett. 107, 172503 (2011).
- [35] Z. Meisel and S. George, Int. J. Mass Spectrom. 349-350, 145 (2013).
- [36] D. Morrissey, B. Sherrill, M. Steiner, A. Stolz, and I. Wiedenhoever, Nucl. Instrum. Methods Phys. Res., Sect. B **204**, 90 (2003).
- [37] D. Bazin, J. Caggiano, B. Sherrill, J. Yurkon, and A. Zeller, Nucl. Instrum. Methods Phys. Res., Sect. B 204, 629 (2003).
- [38] D. Shapira, T. Lewis, and L. Hulett, Nucl. Instrum. Methods Phys. Res., Sect. B 454, 409 (2000).
- [39] F. Wienholtz et al., Nature (London) 498, 346 (2013).
- [40] G. Audi, F. Kondev, M. Wang, B. Pfeiffer, X. Sun, J. Blachot, and M. MacCormick, Chin. Phys. C 36, 1157 (2012).
- [41] D. Lunney, J. M. Pearson, and C. Thibault, Rev. Mod. Phys. **75**, 1021 (2003).
- [42] B. A. Brown, Phys. Rev. Lett. 111, 162502 (2013).