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Precision mass measurements of ^{67}Fe and ^{69,70}Co: nuclear structure towards $N = 40$ and impact on r-process reaction rates

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Accurate mass measurements of neutron-rich iron and cobalt isotopes, ^{67}Fe and ^{69,70}Co, have been realized with the JYFLTRAP double Penning trap mass spectrometer. With novel ion-manipulation techniques, the masses of the ^{69,70}Co ground states and the $1/2^-$ isomer in ^{69}Co have been extracted for the first time. The measurements remove ambiguities in the previous mass values and yield a smoother trend on the mass surface, extending it beyond $N = 40$. The moderate $N = 40$ subshell gap has been found to weaken below ^{68}Ni, region known for shape coexistence and increased collectivity. The excitation energy for the $1/2^-$ intruder state in ^{69}Co has been determined for the first time and is compared to large-scale shell-model calculations. The new mass values also reduce significantly mass-related uncertainties for the astrophysical rapid neutron capture process calculations.

Nuclear mass is an intrinsic property of atomic nuclei. It provides a way to determine nuclear binding energy, which holds the nucleons together and reflects subtle changes in the inner structure of nuclei (see e.g. [1]). As such, binding energies yield information on the nuclear shell model [2], such as evolution and magnitude of shell closures at magic neutron (N) and/or proton (Z) numbers (see e.g. [3–6]). Nucleons can also arrange themselves to form long-living excited states known as isomers. The binding energies of isomers, i.e. their excitation energies, can provide relevant data for nuclear structure and shape coexistence as well. Resolving isomers from the ground states is essential not only for studying nuclear structure far from stability but accurate mass values are also the main nuclear inputs for the rapid neutron capture process (r-process) [7–9] which produces around half of the elements heavier than iron. Sensitivity studies [10] have shown that the neutron capture rates ^{67}Fe$(n,\gamma)^{68}$Fe and ^{68}Co$(n,\gamma)^{69}$Co have a particularly strong impact on the calculated abundances in the weak r-process producing lighter r-process elements most likely at several astrophysical sites [11–16]. These rates, and in particular their inverse photodissociation rates, depend sensitively on the reaction Q-value, i.e. on nuclear masses.

In this Letter, we report precision mass measurements of neutron-rich iron (Z=26) and cobalt (Z=27) isotopes. The studied nuclei lie below ^{68}Ni ($Z = 28$, $N = 40$), which exhibits typical doubly-magic characteristics: the first 2^+ state has a high excitation energy of 2033 keV [17] and a low reduced transition probability of $B(E2; 0^+_1 \rightarrow 2^+_1) = 260(50)\ e^2fm^4$ [18, 19]. The observed values, however, have not been interpreted as evidence for a strong $N = 40$ closure [20, 21]. So far, precision mass measurements on nickel, copper and gallium isotopes have indicated only a localized weak subshell closure at $N = 40$ [22, 23]. Below nickel, cobalt and iron isotopes up to ^{69}Co [24, 25] and ^{66}Fe [24, 26] have been studied at the LEBIT Penning trap but the overall trend after $N = 40$ has remained unclear, partly due to long-living isomers.

The $N = 40$ region is known for shape coexistence [27]. Several low-lying 0^+ states with different shapes have been observed in ^{68}Ni [28–32] and ^{70}Ni [32–34]. These states and many isomers in the region stem from particle(p)-hole(h) excitations across the $Z = 28$ and/or $N = 40$ shell closures. The presence of neutrons in the $g_{9/2}$ orbital makes proton excitations more likely due to the tensor force which decreases the gap between the $1f_{7/2}$ and $1f_{5/2}$ proton orbitals [35–37]. These so-called intruder states can even become the ground state when strong quadrupole correlations drive them lower in energy, and an "island of inversion" can occur [38].

Shape coexistence and isomeric states have been observed in neutron-rich cobalt isotopes [39–41]. The $(7/2^-)$ ground state in ^{67}Co has been described as a pro-

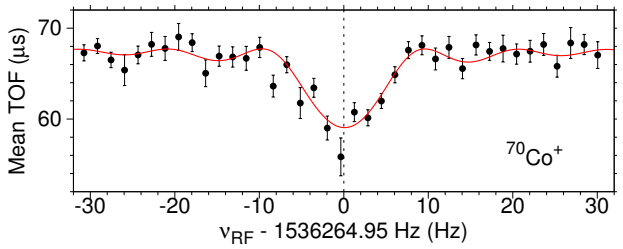


FIG. 1. Time-of-flight spectrum for ^{70}Co collected with 100 ms quadrupolar excitation for the longer, 513 ms measurement cycle. The solid red line is a fit of the data (black points) to the theoretical line shape [54].

ton hole coupled to the spherical ^{68}Ni ground state [41], and the $(1/2^-)$ isomeric state at 491.6(10) keV with a half-life of 496(33) ms [39] as a prolate $[321]1/2^-$ proton intruder state where one proton from the $f_{7/2}$ shell has been excited across $Z = 28$ [39]. Two long-living states have also been observed in ^{69}Co [42] but the energy of the deformed $1/2^-$ state has remained unknown. It has been estimated to be less than 467 keV based on the unobserved $1/2^- \rightarrow 7/2^-$ $M3$ transition [42]. The shorter-living $(7/2^-)$ state in ^{69}Co ($T_{1/2}$ = 180(29) ms [42]) strongly feeds $(5/2^-)$ states in ^{69}Ni, and has been observed in several studies [42–46] whereas only Ref. [42] reports on a longer-living 750(250) ms state, based on a fit on the total decay curve of ^{69}Fe. Further evidence for the existence of two long-living states are the γ-transitions at 1128, 1319, 1343, 1545, and 1642 keV, which were unplaced in Ref. [44] but found to be much more populated via beta decay of ^{69}Fe $(1/2^-)$ than via prompt ^{69}Co production in [42]. Therefore, the longer-living state is likely $1/2^-$, similarly to the proton-intruder states at 491.6(10) keV in ^{67}Co [39] and at 1095.0 keV in ^{65}Co [40]. In this Letter, we employ Penning trap mass spectrometry to determine the location of the $(1/2^-)$ state beyond the $N = 40$ subshell closure.

In ^{70}Co, the ground state has been predicted to be the shorter-living high-spin state with spin and parity $(6^-, 7^-)$ and $T_{1/2}$ = 112(7) ms, studied in many fragmentation experiments [33, 47–49]. Recently, a beta-decay study of ^{70}Fe [50] and Monte Carlo shell-model calculations based on the A3DA Hamiltonian [51], suggest the ground state of ^{70}Co to be a $(1^+, 2^+)$ state with $T_{1/2}$=508(7) ms. The long half-life is explained by the stabilizing effect of the type II shell evolution [37]. The minimum of the potential energy surface of ^{70}Co has been assigned to correspond to deformed single-particle orbitals analogous to Nilsson $\pi[321]1/2^-$ and $\nu[301]1/2^-$ orbitals [50]. The $1/2^-$ isomer in ^{67}Fe has also been interpreted as $\nu[301]1/2^-$, however, the isomers in ^{67}Fe [52, 53] are too short-living (sub-ms) for Penning-trap mass spectrometry.

In this Letter, we investigated long-living ground and isomeric states in the $N = 40$ region via Penning-trap mass spectrometry. The neutron-rich iron and cobalt isotopes were produced by 35-MeV protons impinging on a 15 mg/cm^2-thick ^{nat}U target at the IGISOL facility [55]. The reaction products were thermalized in helium gas, extracted out from the gas cell, transported using a sextupole ion guide [56] and accelerated to 30 keV before mass separation with a dipole magnet. A radiofrequency cooler and buncher (RFQ) [57] was used to convert the continuous mass separator beam to short ion bunches that are injected into the double Penning trap mass spectrometer JYFLTRAP [58]. There the ions were first purified via the buffer gas cooling technique [59] before the transfer to the precision trap for high-precision mass measurements. The ion's cyclotron resonance frequency $\nu_c = \frac{1}{2\pi}\frac{q}{m}B$, where q and m are the charge and mass of the ion, respectively, was measured using the time-of-flight ion cyclotron resonance (TOF-ICR) technique [54, 60] (see Fig. 1). The magnetic field strength B was determined with ^{84}Kr$^+$ ions as a reference. For ^{69,70}Co, quadrupolar excitation times of 50 ms and 100 ms were employed. The ^{67}Fe data were collected using Ramsey's method of time-separated oscillatory fields [61–63] with an excitation pattern of 25-50-25 ms (On-Off-On). Systematic errors due to temporal fluctuations in the magnetic field, $\sigma_B(\nu_{c,ref})/\nu_{c,ref}$=$(8.18(19)\times 10^{-12}/\text{min}) \times \Delta t$ [64], and the mass-dependent uncertainty $\sigma_m(r)/r$=$(2.2(6) \times 10^{-10}/u) \times \Delta m$ [65], were quadratically added to the statistical uncertainties.

The results are summarized in Table I. For ^{69}Co, it was not possible to separate the two long-living states from each other using the TOF-ICR technique. Since the half-lives of the two states are significantly different, the composition of the ion bunches was manipulated by changing the waiting time, t_{wait}, from the moment the ion beam accumulation in the cooler was stopped to the extraction toward JYFLTRAP. The two measurement sets for ^{69}Co had otherwise identical measurement schemes, except for t_{wait} = 500 ms in the long cycle, which lasted in total 726 ms after the ion beam production had stopped. There was no waiting time in the short, 226 ms cycle. Due to the much longer half-life of the $(1/2^-)$ state, it is likely to dominate the measurements using the long cycle. Based on the ratio of the average number of ions in the short- and long-cycle measurements, $R = N_{short}/N_{long}$ = 2.6(4), the fraction of the longer-living state in the beam (f_l) was determined at the moment the production stopped (f_l = 33(10)%), as well as 226 ms (f_l = 49(13)%) and 726 ms (f_l = 81(9)%) after it, assuming the shorter-living state contributes $f_s = 1 - f_l$ of the beam. The mass-excess values for the longer- and shorter-living states ($\Delta_{l,s}$), were determined from the measured mass-excess values ($\Delta_{meas}(t$ = 226 ms) = −50296(15) keV and $\Delta_{meas}(t$ = 726 ms) = −50238(20) keV) using $\Delta_{meas}(t) = [1 - f_l(t)]\Delta_s + f_l(t)\Delta_l$. The determined mass excess value for ^{69}Co, −50383(44) keV, agrees well with

TABLE I. The half-lives, spins and parities for the ions of interest based on [66], measured frequency ratios $r = \nu_{ref}/\nu$, and mass-excess values Δ in comparison with the literature values from [66, 67]. '#' denotes a value based on extrapolations or systematics. Singly-charged ions of ^{84}Kr ($m = 83.911497729(4)$ u [67]) were used as a reference for all studied cases.

Nuclide	$T_{1/2}$ (ms)	I^π	r	Δ_{JYFL} (keV)	Δ_{lit} (keV)	Diff. (keV)
^{67}Fe	394(9)	$(1/2^-)$	0.797874190(8)	-45709.1(3.8)	-45610(270)	-99(270)
^{69}Co	180(29)	$7/2^-$#	0.821649141(428)[a]	-50383(44)	-50280(140)	-103(147)
^{69}Co^m	750(250)	$1/2^-$#	0.821651504(291)[a]	-50207(36)	-49780(240)#	-430(240)#
^{70}Co[b]	508(7) [50]	$(1^+, 2^+)$[50]	0.833615937(21)	-46525(11)	-46430(360)#	-95(360)#

[a] Calculated based on the isomeric fractions f_l for the longer-living state and the frequency ratios determined from the files using the 226-ms cycle ($f_l = 49(13)$ %, $r = 0.821650299(36)$) and the 726-ms cycle ($f_l = 81(9)$ %, $r = 0.821651055(92)$), see text for details.
[b] Assigned as the ground state in [50]. Considered as a 3^+# isomer 200(200)# keV above a $(6^-, 7^-)$, $T_{1/2}$=112(7) ms state in [66].

the most recent Atomic Mass Evaluation (AME16) [67] value based on measurements using the TOFI spectrometer [68, 69], $B\rho$-TOF method [70, 71] and isochronous mass spectrometry [72]. The obtained mass excess value for the isomer ^{69}Co^m, $-50207(36)$ keV, is in perfect agreement with the ground-state value, $-50214(14)$ keV [25], reported recently from the LEBIT Penning trap, suggesting they have actually measured the isomer.

We have determined the excitation energy, $E_x = 176(57)$ keV, for the longer-living $(1/2^-)$ state in ^{69}Co for the first time. The $(1/2^-)$ state is interpreted as deformed $1p-2h$ proton intruder state for which the role of neutron excitations across $N = 40$ is essential as they increase correlation energies to overcome the $Z = 28$ shell gap. Shell-model calculations employing the Lenzi-Nowacki-Poves-Sieja (LNPS) interaction with minor modifications in the *pf-sdg* valence space [73] predict that the $1/2^-$ intruder state becomes the ground state in ^{69}Co instead of the spherical $7/2^-$ proton-hole state (see Fig. 2). Here we confirm the proximity of the coexisting deformed intruder and spherical states in ^{69}Co, and the decreasing trend from $N = 40$ to $N = 42$, similarly to the intruder states observed in nickel and copper isotones (see Fig. 2).

The phase-imaging ion-cyclotron-resonance technique (PI-ICR) [76], recently commissioned at JYFLTRAP [77] was used to determine the composition of the ^{70}Co beam. Unfortunately, there was no sign of another long-lived state at a statistically significant level. The production rates and determined mass-excess values for ^{70}Co changed only moderately when the measurement cycle was increased from 232 ms to 513 ms, supporting that the measured state was the 508(7) ms, $(1^+, 2^+)$ state [50]. This is consistent with the previous ^{70}Co experiment [78] employing the same production method, where the longer-living low-spin state was favoured as well. Using the PI-ICR data, an upper limit for the $(6^-,7^-)$ state contribution could be set. This was $\leq 17\%$ for the short-cycle measurements and $\leq 5\%$ for the long cycles. For the final result, the value determined with the long cycle was adopted.

Our mass-excess value for ^{70}Co agrees with the extrapolation given in AME16 [67] and is 295(280) keV above the only previous experimental value [70] which has been rejected in AME16 as an anomalous point since it e.g. introduces a kink on the S_{2n} values. A much smoother trend is obtained with our new value as shown in Fig. 3 giving further support that the $(1^+, 2^+)$ is the ground state as suggested in Ref. [50]. If we had measured the isomer, then the S_{2n} value for ^{70}Co would be larger, introducing a kink at $N = 43$. We also noticed that using the value from [25] for ^{68}Co introduces an anomaly from $N = 41$ to 43 (the black dashed line in Fig. 3) suggesting that the longer-living isomeric state was measured both for ^{68}Co and ^{69}Co in Ref. [25]. This is further supported by a measurement done at the ESR storage ring for an unknown mixture of ^{68}Co states [79] that falls 195(96) keV below the value reported for the ground state in Ref. [25].

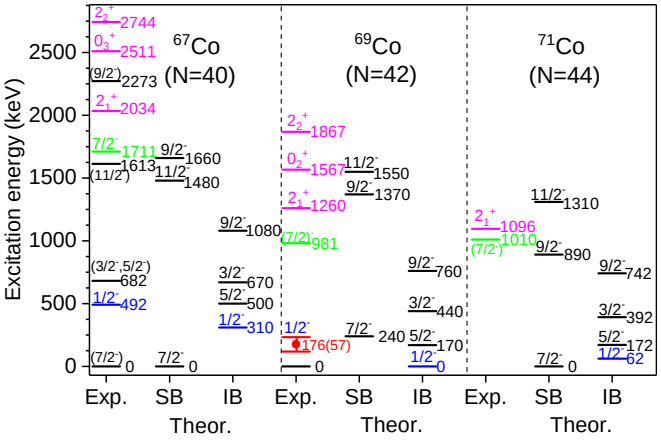


FIG. 2. Experimental level schemes for ^{67}Co [39, 41] and ^{69}Co in comparison with the shell-model calculations for the spherical (SB) and $1/2^-$ intruder (IB) bands. The $1/2^-$ states in Co (in blue, or red from this work) follow a similar trend as the 2^+ and prolate 0^+ [31, 33, 34, 74] intruder states in Ni (in magenta) and $7/2^-$ [75] states in Cu isotones (in green).

The present data confirm that the $N = 40$ subshell closure gets weaker below nickel. Two-neutron separation energies in the studied iron and cobalt isotopic

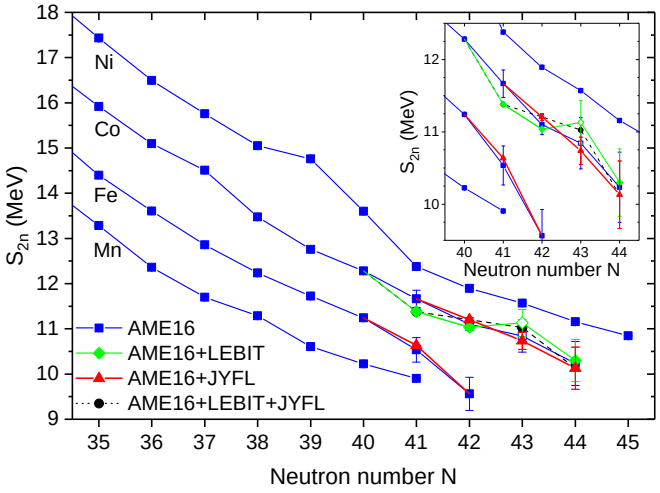


FIG. 3. Two-neutron separation energies based on experimental values from AME16 [67] (in blue) and including the results from this work (in red). The recent ^{68,69}Co measurements at LEBIT [25] (in green) introduce a kink, the same is true if only the result for ^{68}Co from [25] is included, pointing that it is likely to belong to the isomer ^{68}Co^{m}. For ^{70}Co, AME16 is based on extrapolations (indicated with an open symbol) and our value is for the $(1^+, 2^+)$ state.

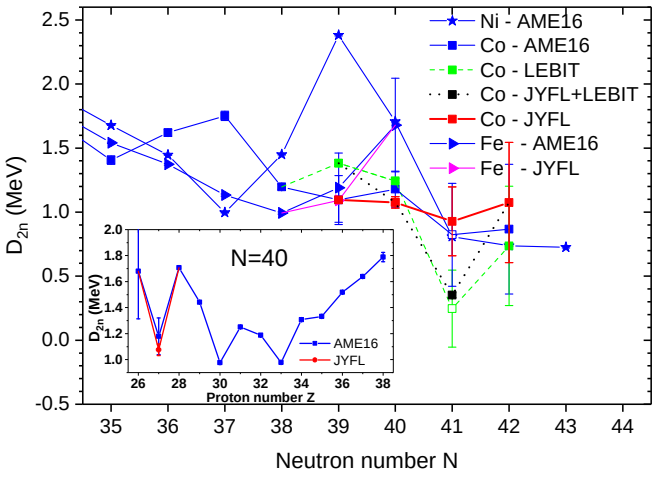


FIG. 4. Two-neutron shell gap parameter $D_{2n}(Z,N) = S_{2n}(Z,N) - S_{2n}(Z,N+2)$ based on AME16 [67] (in blue) and this work (red/magenta). Including ^{68,69}Co from LEBIT [25] (in green), or only ^{68}Co (in black), results in a kink at $N = 40$, pointing toward an isomeric state measurement. The inset shows D_{2n} for $N = 40$.

chains do not drop significantly after $N = 40$, and the empirical two-neutron shell-gap energy, determined as $D_{2n}(Z,N) = S_{2n}(Z,N) - S_{2n}(Z,N+2)$, is almost 0.7 MeV lower for $N = 40$ at ^{67}Co than at ^{68}Ni (see Figs. 3-4). This is consistent with the earlier spectroscopic studies [80–85] and the recent mass measurements of $^{58-63}$Cr [86] indicating increased collectivity below nickel. The gain in the correlation energy flattens the two-neutron separation energy trend, and the D_{2n} parameter decreases. Our measurements bring the D_{2n} values for ^{65}Fe and ^{67}Co 100 keV lower than in AME16 [67] but the values are still at around 1 MeV. This is higher than $D_{2n} \approx 0.7$ MeV observed for the most neutron-rich Cr and Mn isotopes [67, 86, 87]. Moreover, the decreasing trend in the D_{2n} values seems to stop at $N \approx 38$ for the Fe and Co chains. Santamaria et al. [84] have observed that also the 2^+ and 4^+ excitation energies in the Fe isotopic chain decrease until $N = 40$ after which a plateau is reached. This has been interpreted as an extension of the N=40 'island of inversion' toward N=50 [84] that could lead to a disappearance of the magicity of $N = 50$ below ^{78}Ni.

Masses of neutron-rich nuclei close to $N = 40$ are also relevant for astrophysics. Neutron-capture rates $N_A\langle\sigma v\rangle$ for ^{67}Fe$(n,\gamma)^{68}$Fe and ^{68}Co$(n,\gamma)^{69}$Co have been highlighted as ones of the most influential for the r-process calculations, with impact factors $F = 15.8$ and $F = 11.6$, respectively, on the abundances when the rates were varied by a factor of 100 [10]. In this work, we determined the mass of ^{67}Fe (see Table I) with a 70 times higher precision than before. It was found to be around 100 keV lower than the recommended value [67] based on less precise methods [68–71, 88, 89]. The mass of ^{69}Co was also found to be around 100 keV lower and three times more precise than in AME16 [67]. The mass values affect the calculated neutron-capture rates, and even more importantly, their inverse photodissociation rates $\lambda_{\gamma,n}$ which depend exponentially on the reaction Q-value: $\lambda_{\gamma,n} \propto N_A\langle\sigma v\rangle exp[-Q/(kT)]$ for temperature T. Figure 5 shows a comparison of photodissociation rates and mass-related uncertainties for ^{68}Fe$(\gamma,n)^{67}$Fe and ^{69}Co$(\gamma,n)^{68}$Co from this work and from REACLIB v1.0 [90] used for the sensitivity study in Ref. [10]. The new rate calculated with $Q = 5.85(37)$ MeV, determined from the masses of ^{67}Fe from this work and ^{68}Fe from AME16 [67], is significantly higher than the rate obtained with REACLIB v1.0, which relies on NON-SMOKER [91, 92] neutron-capture rates with $Q = 6.86(82)$ MeV, based on experimental AME95 value [93] for ^{67}Fe and theoretical FRDM1995 value [94] for ^{68}Fe. At 1.5 GK, the mass-related uncertainties have been reduced by a factor of ≈30 and the photodissociation rate is around 2500 times higher. The Q-value for ^{68}Co$(n,\gamma)^{69}$Co based on our work for ^{69}Co and ^{68}Co from [67], $Q = 6.52(20)$ MeV, is also much lower than the value used in REACLIB v1.0, $Q = 7.29(50)$ MeV [93]. As a result, the photodissocation rate is much higher than before (see Fig. 5). If the isomeric-state mass had been used for ^{69}Co, like obtained in Ref. [25], the rate had been four times higher at 1.5 GK. This highlights the importance of precise knowledge of the masses.

In conclusion, we have performed the first precision mass measurements of ^{67}Fe and ^{70}Co. With novel ion-manipulation techniques, we were able to identify the measured states in ^{69,70}Co, and to provide accurate mass

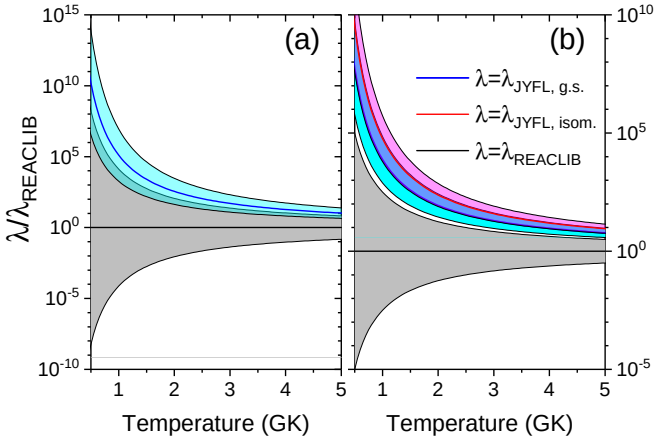


FIG. 5. Rates λ and their mass-related uncertainties for (a) ^{68}Fe$(\gamma,n)^{67}$Fe and (b) ^{69}Co$(\gamma,n)^{68}$Co based on this work (JYFL, g.s.) and REACLIB v1.0 [90]. For comparison, the rate calculated using the isomeric-state mass value of ^{69}Co from this work is also plotted (JYFL, isom.).

data relevant for nuclear structure and astrophysics. The position of the $(1/2^-)$ proton intruder state in ^{69}Co was determined for the first time, and the decreasing trend of the $1/2^-$ intruder states up to $N = 42$ was confirmed, in agreement with the large-scale shell-model calculations predicting increased collectivity and close-lying spherical and deformed states for ^{69}Co. No strong $N = 40$ subshell closure is observed below nickel and the S_{2n} values follow a smooth trend, favoring spherical $7/2^-$ orbital as the ground state also for ^{69}Co. On the other hand, the increased collectivity drives the deformed $1/2^-$ intruder state lower in energy and it could become the ground state. In future, spectroscopy on ^{69}Co states would be welcome to firmly confirm the spins of the isomeric states. In addition, mass measurements of ^{68}Co, ^{68}Fe, and ^{71}Co would be highly desirable to further extent our knowledge of the evolution of isomeric states in this fascinating region exhibiting shape coexistence, and to provide accurate Q-values for the weak r-process.

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