Penning trap mass spectrometry of neutron-rich Fe and Co isotopes around $N = 40$ with the LEBIT mass spectrometer

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Penning trap mass spectrometry is presented as a complementary tool to nuclear spectroscopy experiments for the study of nuclear structure in the vicinity of $N = 40$, $Z = 28$. High-precision mass measurements of the $^{63-66}$Fe and $^{64-67}$Co isotopes have been carried out with the Low Energy Beam and Ion Trap (LEBIT) Penning trap mass spectrometer. The newly obtained mass values for ^{66}Fe and ^{67}Co are presented, together with the previously reported LEBIT mass measurements in this region. In the case of ^{65}Fe the existence of a new isomer is reported, and an isomer recently discovered by decay spectroscopy in ^{67}Co is confirmed. Relative mass uncertainties as low as 4×10^{-8} are obtained. All mass values are found to be in good agreement with previous experimental results with the exception of ^{64}Co, where a 5σ deviation is observed. Using these data the two neutron separation energies S_{2n} are calculated. However, the large error bars in the mass values of the neighbor Fe and Co isotopes with $N > 40$ complicate the validation of a weak subshell closure at $N = 40$ for the Co isotopes or the possible reduction in the neutron shell gap in the case of the Fe isotopes, in accordance with the theoretical predictions of an onset of deformation in the region.

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I. INTRODUCTION

Most of the detailed present knowledge of nuclear structure is based on the study of nuclei on, or close to, the β-stability line. Here, in the frame of the nuclear shell model, a quantity like the magic numbers suggested by M. G. Mayer [1] and, independently, by O. Haxel *et al.* [2] in 1949 presently describes the shell structure in terms of a harmonic oscillator potential with spin-orbit coupling. With the advent of radioactive beam facilities [3], an extension of the limits of nuclear existence became feasible. Far from the valley of β stability it is observed that new magic numbers have appeared for both protons and neutrons, for example, $Z = 14$ [4], $N = 16$ [5], and $N = 32$ [6], whereas some of the known magic numbers have disappeared, for example, $N = 8$ [5] and $N = 20$ [7]. In a fashion similar to the island of inversion near ^{31}Na [8,9], a particular shell evolution in the vicinity of $N = 40$ is expected for isotones between Ca and Ni [10].

The observation of a high 2^+ excitation energy $E(2^+)$ and a low transition probability $B(E2)$ between the ground state 0^+ and the excited state 2^+ in ^{68}Ni [11–13] has been discussed as an indication of a subshell closure at $N = 40$ caused by a sufficiently large gap between the $\nu g_{9/2}$ and the $\nu(pf)$ shell. Subsequent theoretical predictions [14] concluded that the small experimental $B(E2)$ value does not provide sufficient evidence for a doubly magic character of ^{68}Ni. Furthermore, trends in the two neutron separation energies S_{2n} from recent mass measurements in this region do not support a large shell gap [15,16], and results from a number of other experiments [10,17–19] show that the signatures of double magicity observed in ^{68}Ni vanish rapidly when moving away from this nucleus. The explanation for this rapid disappearance is that the attractive monopole contribution of the tensor force acting between proton and neutron orbitals is weakened as protons are removed from the $\pi f_{7/2}$ orbital, when moving from nickel toward calcium [9,20]. As a result, the gap between the $\nu g_{9/2}$ and the $\nu(pf)$ shell, already small in ^{68}Ni, is reduced further, inducing neutron pair scattering to the intruder $\nu g_{9/2}$. The lowering of the $\nu g_{9/2}$ orbital for $Z < 28$ nuclei makes the nonspherical orbitals more likely to be occupied as the neutron number increases beyond $N = 36$, thus generating increased deformation at $N \approx 40$ [17].

In this paper we present Penning trap mass spectrometry [21] as a complementary method to γ or β spectroscopy for the study of nuclear structure in neutron-rich isotopes far beyond the valley of β stability. We discuss the existence of an unexpected isomer in ^{67}Co, discovered by β spectroscopy [22] and now confirmed at the Low Energy Beam and Ion Trap

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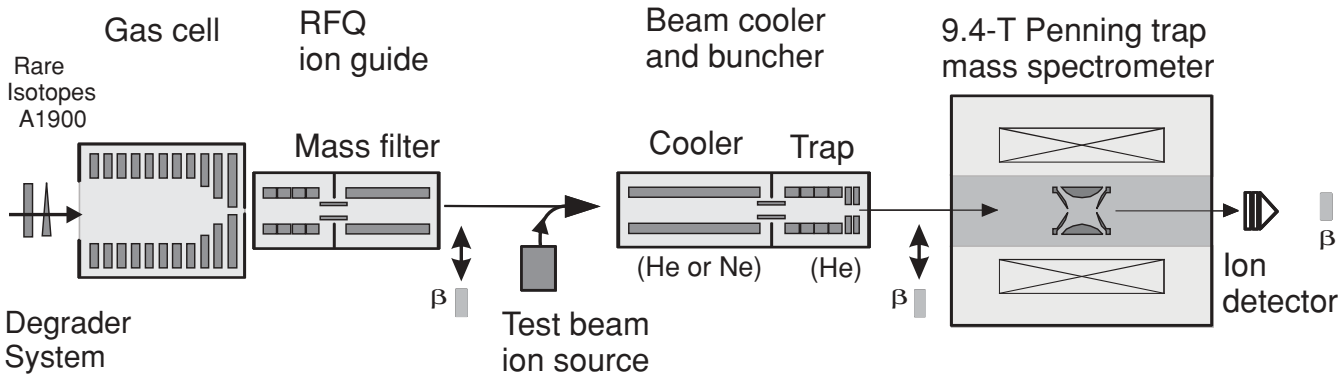

FIG. 1. Schematic overview of the LEBIT facility.

(LEBIT) facility, as well as the first discovery, reported in Refs. [23] and [24], of a nuclear isomer by Penning trap mass spectrometry.

The combination of short half-life species and available low production rates make measurements in this region difficult, and hence the masses of many neutron-rich rare isotopes still either are experimentally unknown or have large uncertainties [25]. Most of the current mass values in the $N = 40$, $Z = 28$, region have been obtained by time-of-flight (TOF) mass measurements [26–28] that can reach far from the valley of stability but, unfortunately, achieve a precision not better than 100 keV. Typical mass resolving powers of 10^4 are normally insufficient to resolve isomeric and ground states, which can lead to large systematic errors in the determination of the ground-state mass as discussed, for example, in Ref. [27].

Penning trap mass spectrometry has proven on several occasions [29–32] to be able to successfully resolve known isomers and ground states, leading to an unambiguous and very accurate determination of ground-state masses. Penning trap mass measurements close to $N = 40$ had only been performed for $Z \geqslant 28$ on neutron-rich Ni, Cu, and Ga isotopes [15,16], as mentioned earlier. In this paper we summarize the first high-precision mass measurements in the region of $Z < 28$, namely the isotopes $^{63-65}$Fe and $^{64-66}$Co, reported in Refs. [23] and [24], together with more recent results on measurement of ^{66}Fe and ^{67}Co. With these new measurements we extend the high-precision mass values up to $N = 40$.

II. EXPERIMENT

The LEBIT facility [33] at the National Superconducting Cyclotron Laboratory is at present the only facility capable of performing high-precision mass measurements of rare isotopes produced by projectile fragmentation, providing access to isotopes of elements not available from other production techniques [34–36]. Figure 1 shows a schematic layout of the main components of the LEBIT facility. In the gas cell [37] the ions are stopped and thermalized by collisions with high-purity He gas. A subsequent set of radio-frequency quadrupole (RFQ) structures [38] guides the ions through different pressure regimes and selects species by their mass-to-charge ratio A/Q by operating the last RFQ section as a mass filter [38]. A beam cooler and buncher [39] is used to decelerate, cool, and bunch the ions for delivery to the 9.4-T Penning trap mass spectrometer [40]. In the trap, the superposition of a homogeneous magnetic field with an electrostatic quadrupolar potential is employed to spatially confine the ions in three dimensions [41–43]. Finally, the mass of the trapped ions is investigated by means of the TOF ion cyclotron resonance (TOF-ICR) detection technique [44–46].

In the experiments reported here the radioactive Fe and Co isotopes were produced by fragmentation of a ^{76}Ge primary beam at an energy of 130 MeV/nucleon. The A1900 fragment separator [47] delivered a secondary beam with 86 MeV/nucleon energy containing a cocktail of Fe and Co isotones. The stopping and extraction of the desired isotone were optimized by adjusting the angle of the glass degrader in conjunction with an insertable Si detector placed in the stopping path of the ions within the gas cell.

To determine the chemical form of the radioactive ions extracted from the gas cell, the amplitude of the radio-frequency (RF) field applied to the mass filter was scanned, which provided a typical resolving power of $R \approx 70$. The transmitted activity was then measured with another insertable Si detector at the exit of the mass filter. As an example, such activity scans for ^{64}Fe and ^{66}Co are shown in Fig. 2.

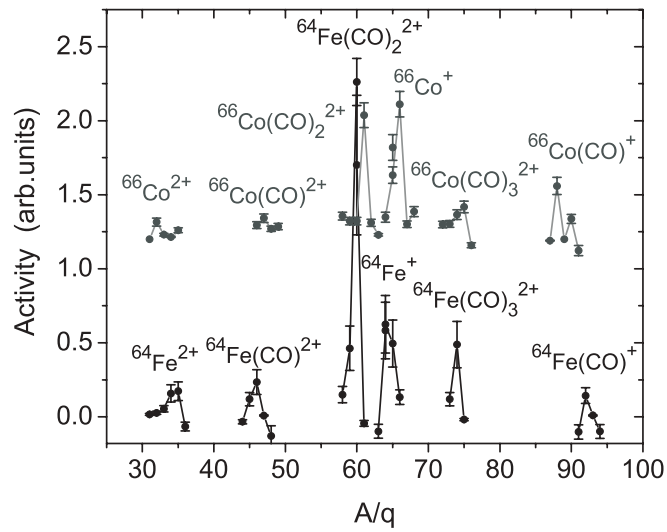

FIG. 2. Detected activity of ions extracted from the gas cell as a function of the mass-to-charge ratio A/Q scanned in the RFQ mass filter. For clarity an offset has been added to the ^{66}Co data.

In the case of ^{66}Co, activity was detected at $A/Q = 33$, 47, 61, 66, 75, and 94. The activity at $A/Q = 33$ and 66 was identified in the Penning trap as singly and doubly charged ^{66}Co ions. The remaining peaks are consistent with singly and doubly charged molecular ions of the form $[^{66}\mathrm{Co(CO)_m}]^{n+}$, where $n, m = 1, 2$. The Fe isotopes exhibited a similar behavior. For mass measurement of a given isotope the mass filter was set to the A/Q value for which the higher ratio of the number of rare isotope ions to the number of contaminant ions was observed in the Penning trap.

For example, in the case of ^{66}Co, $[^{66}\mathrm{Co(CO)_2}]^{2+}$ with $A/Q = 61$ was chosen. In the beam cooler and buncher the CO ligands were stripped off by collision-induced dissociation with He as buffer gas at a pressure in the 10^{-2} mbar range, providing $^{66}\mathrm{Co}^{+}$ for the actual measurement. Ion bunches about 100 ns in length were extracted and purified on their way to the trap by a TOF separator, with a typical resolving power of $R \approx 300$, allowing only ions of the proper A/Q to be captured. For optimum injection of the ions into the Penning trap, a Lorentz steerer [48] was used for fast and controlled placement of the ions in the trap. Remaining unwanted isobars in the trap were removed by excitation of their modified cyclotron motion with an azimuthal dipole RF field, to avoid frequency shifts owing to ion-ion interactions of different species [29].

The trapped ions were then exposed to a quadrupolar excitation with an azimuthal quadrupolar RF field of frequency ν_{RF} close to the cyclotron frequency, $\nu_c = q\mathrm{B}/(2\pi m)$, of the ions, where q and m are the charge and mass of the ion, and B is the strength of the magnetic field. Subsequently, the ions were extracted and their TOF to a multichannel plate (MCP) detector in the Daly configuration [50], located outside the homogeneous B field, was measured. To obtain the ion's cyclotron frequency ν_c, this cycle of trapping, excitation, ejection, and TOF measurement was repeated while scanning ν_{RF} in frequency steps around the expected cyclotron frequency ν_c.

In resonance, $\nu_{\mathrm{RF}} = \nu_c$, the ions gain maximum radial energy during the RF excitation of their motion. This radial energy is then converted into axial energy when the ions pass the gradient of the B field on their way toward the MCP detector. As a result of this energy conversion, a reduced TOF is observed for the ions in resonance [29,44]. As an example, in Fig. 3 a typical resonance for $^{65}\mathrm{Co}^{2+}$ is shown. The fit of the theoretical line shape [45] to the experimental data provides the center frequency of the resonance curve, corresponding to the cyclotron frequency ν_c of the ions being investigated.

The data presented in this paper were taken in two separate experiments. In the first run the masses of $^{63-65}$Fe and $^{64-66}$Co were measured [23,24]. In the second run the mass measurements in this region were extended up to the $N = 40$ nuclei ^{66}Fe and ^{67}Co. In this second experiment the masses of ^{63}Fe and ^{64}Co were remeasured and found to be in perfect agreement with the values of the first experiment.

In the first five columns in Table I the ion species investigated, half-life, number of measurements, total number of collected ions, and rate of detected ions summarized in this work are given.

For the mass determination the magnetic field B was calibrated by measuring the cyclotron frequency $\nu_{c,\mathrm{ref}}$ of a well-known reference ion (see column 6 in Table I) before and after measurement of the ion of interest. A linear interpolation can then be used to obtain the magnetic field at the moment of measurement of the radioactive ion. These calibration measurements were typically performed within 2 h of each other. This time interval was found sufficient to neglect short-time fluctuations owing to a stabilization of the magnetic field B of the LEBIT's superconducting solenoid by an automatic regulation system of the pressure in the He dewar and to a compensation of its natural decay by the superposition of an additional longitudinal magnetic field along the trap's axis.

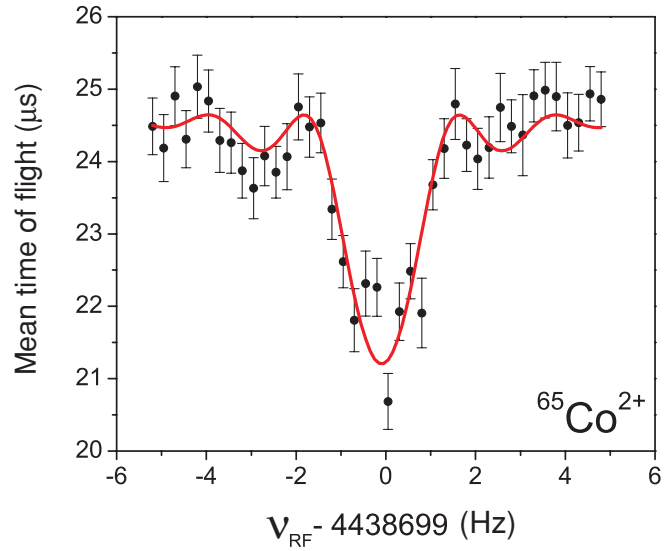

FIG. 3. (Color online) Cyclotron resonance for $^{65}\mathrm{Co}^{2+}$. The solid curve is the fit of the theoretical line shape to the data points.

III. DATA ANALYSIS

The value of the cyclotron frequency ν_c obtained from the fit of the theoretical line shape to the data points and the linearly interpolated cyclotron frequency $\nu_{c,\mathrm{ref}}^{\mathrm{int}}$ of the reference ion with a well-known mass are combined in a frequency ratio,

$$r = \frac{\nu_c}{\nu_{c,\mathrm{ref}}^{\mathrm{int}}} = \frac{Q}{Q_{\mathrm{ref}}} \frac{m_{\mathrm{ref}} - Q_{\mathrm{ref}} m_e}{m - Q m_e}, \tag{1}$$

where Q_{ref}, m_{ref}, Q, and m are the charge state and mass of the reference ion and of the ion under investigation, respectively, and m_e is the electron's mass. For the masses of the reference ions m_{ref}, we have used the values given in the last Atomic Mass Evaluation (AME'03) with the exception of the more recent mass values for ^{19}F and ^{28}Si, which were taken from Refs. [51] and [52], respectively. Note that the use of these new values might slightly change the results of the mass values given in Refs. [23] and [24] from those given later here. A weighted average $\bar{r}$ of the frequency ratios was determined provided that several resonances were taken for the same ion species in each case.

A major part of the data analysis consisted of evaluating the possible sources of uncertainty related to the frequency ratio.

TABLE I. Summary of the results presented in this work. Ion species investigated, half-life [49], number of measurements, and total number of ions, as well as the average particles per minute registered on the MCP detector, are given in the first five columns. The ion species used as the reference for each measurement is listed in the sixth column. The weighted mean frequency ratios $\bar{r}$ with their uncertainties obtained from the N_{meas} individual measurements and the total uncertainties reached are listed in the last two columns.

Ion	$T_{1/2}$ (s)	N_{meas}	N_{ions}	Avg./min on MCP	Ref. ion	$\bar{r}$	Total uncertainty
$^{63}Fe^{+}$	6.1(6)	5	4879	30	$HC_2F_2^+$	1.00102246(16)	1.6×10^{-7}
$^{63}Fe^{+}$[a]		1	572	10	CSF^+	1.00047988(16)	1.6×10^{-7}
$^{64}Fe^{+}$	2.0(2)	5	1466	10	$HC_2F_2^+$	0.985355803(83)	8.4×10^{-8}
$^{65g}Fe^{2+}$	1.3(3)	2	1191	11	O_2^+	0.98513559(11)	1.1×10^{-7}
$^{65m}Fe^{2+}$	>0.15	4	1237			0.98512904(13)	1.3×10^{-7}
$^{66}Fe^{+}$	0.44(4)	3	165	0.9	SiF_2^+	1.000416750(67)	6.7×10^{-8}
$^{64}Co^{+}$	0.30(3)	5	4266	65	$HC_2F_2^+$	0.985433836(72)	7.2×10^{-8}
$^{64}Co^{+}$[a]		1	235	5	SO_2^+	1.00040626(19)	1.9×10^{-7}
$^{65}Co^{2+}$	1.20(6)	4	2145	20	O_2^+	0.985265293(35)	3.6×10^{-8}
$^{66}Co^{+}$	0.194(17)	4	3133	17	COF_2^+	1.00079282(23)	2.3×10^{-7}
$^{67g}Co^{+}$	0.425(20)	6	1163	3.3	SiF_2^+	0.98555609(12)	1.2×10^{-7}
$^{67m}Co^{+}$	0.496(33)	4	601			0.98554829(19)	1.9×10^{-7}

[a]Ion species remeasured in the second experiment.

The evaluation of the statistical uncertainties is summarized as follows.

(i) The fit of the theoretical line shape to the experimental data, either for the ion of interest ν_c or for the reference species $\nu_{c,\text{ref}}$, provides the value of the central frequency with its statistical uncertainty.
(ii) In the case of the reference ion the two measurements bracketing the measurement of the ion of interest were used to calculate by linear interpolation the cyclotron frequency $\nu_{c,\text{ref}}^{\text{int}}$ and its uncertainty at the time of the actual measurement.
(iii) These data were used to calculate a frequency ratio $r = \nu_c/\nu_{c,\text{ref}}^{\text{int}}$ and its statistical uncertainty.
(iv) The weighted mean values of the frequency ratios $\bar{r}$ for a given nuclide were calculated. Internal versus external uncertainty consistencies were compared. Their ratio was found to be close to unity in all cases, indicating that only statistical fluctuations were present in the experimental data [53]. The larger value of the two was taken as the final uncertainty. In this way the uncertainties are always given conservatively.

In addition, a careful evaluation of the systematic uncertainties was carried out. The uncertainty caused by temporal magnetic field changes was found to be negligible owing to a field stability of 5×10^{-9}/h and a short time interval, typically less than 2 h, between measurements of the reference ion. To minimize mass dependent uncertainties, reference ions were selected with an A/Q ratio as close as possible to that of the ion under investigation (see Table I). A mass-dependent uncertainty of $5(5) \times 10^{-10}/u$ was established earlier for LEBIT [35] and turned out to be negligible for the data presented in this work. The possibility of frequency shifts owing to the presence of contaminant ions not removed completely by the in-trap isobar cleaning method was avoided by performing measurements with less than one ion per cycle. The frequency shift owing to the presence of background ions was evaluated in cases where a large dispersion in the TOF for a given frequency step was observed. Frequency shifts were found to be negligible in all cases except for one of the ^{66}Fe measurements, where a relative uncertainty of 6.4×10^{-8} was added in quadrature to its statistical uncertainty to account for the shift. Frequency shifts originating from relativistic effects were evaluated, found to be of the order of 3×10^{-10}, and thus far below the statistical uncertainty of the data in this experiment, and consequently neglected.

Once all sources of uncertainty were included in the frequency ratio, the weighted mean $\bar{r}$ (see Table I) was used to finally obtain the atomic mass of the ions under investigation

$$m = \frac{(m_{\text{ref}} - m_e)}{\bar{r}} + m_e. \tag{2}$$

Note that in Eq. (2) we have assumed the particular case of singly charged ions $Q = Q_{\text{ref}} = 1$.

IV. RESULTS

The experiments performed in this work provide high-precision mass values in the vicinity of $Z < 28$ and $N \leqslant 40$. Highlights are as follows: a new long-lived isomeric state was discovered by Penning trap mass spectrometry in ^{65}Fe [23,24], the masses of the $N = 40$ nuclides ^{66}Fe and ^{67}Co were obtained, and a long-lived isomeric state in ^{67}Co, recently observed in β-decay studies [22], was confirmed. In the second experiment, which was dedicated to extending mass measurements in this region, the masses of ^{63}Fe and ^{64}Co were remeasured, providing a mass excess of ME = −55 637.5(9.5) keV and ME = −59 584.4(11.4) keV, respectively, both in very good agreement with the results obtained in the first experiment [23,24]. The weighted mean of the results acquired for these isotopes in the two experiments are used as the final value in the mass excess; see Table II.

TABLE II. Comparison of mass excess values, ME = [m(amu) − A], between the data obtained in this work and the previous experimental data from AME'03 [25]. An excitation energy E_{ex} = 402(10) keV for ^{65m}Fe and E_{ex} = 493(14) keV for ^{67m}Co is deduced from the mass difference between the ground state and the corresponding isomer.

Ion	$ME_{AME'03}$ (keV)	ME_{LEBIT} (keV)	Difference (keV)
$^{63}Fe^{+}$	−55 545(168)	−55 634.1(6.7)[a]	89(168)
$^{64}Fe^{+}$	−54 772(280)	−54 969.5(5.0)	198(279)
$^{65g}Fe^{2+}$	−50 878(242)	−51 221.3(6.8)[b]	343(242)
$^{65m}Fe^{2+}$	–	−50 819.4(8.0)[b]	–
$^{66}Fe^{+}$	−49 574(298)	−50 067.7(4.1)	494(298)
$^{64}Co^{+}$	−59 793(20)	−59 685.7(4.1)[a]	−107(20)
$^{65}Co^{2+}$	−59 168(20)	−59 185.1(2.1)	17(20)
$^{66}Co^{+}$	−56 113(252)	−56 408.1(14.1)	295(252)
$^{67g}Co^{+}$	−55 061(317)	−55 322.2(7.3)	261(317)
$^{67m}Co^{+}$	–	−54 828.8(12.0)	–

[a]Weighted mean values of the results obtained for these species in the two experiments.
[b]Note that the uncertainties of the mass excess for the ground state and the isomer in ^{65}Fe have been corrected here from the erroneous values given in Ref. [23].

Figure 4 shows a comparison between the mass excess values obtained in this work and those listed in AME'03 [25] (see also Table II). A detailed discussion of the results obtained for the investigated nuclides is given here.

A. ^{64}Co

The ^{64}Ni(t,^{3}He) reaction was used at the Los Alamos Van de Graaff facility to obtain a mass excess ME = −59 803(20) keV for the ^{64}Co ground state [54]. In AME'03 a mass excess of ME = −59 793(20) keV is given after the inclusion of more recent values for the reaction members. The latter value disagrees with our value of ME = −59 685.7(4.1) keV by 5 standard deviations. Trying to find an explanation for the large disagreement observed between the two measurements is difficult, owing to the scarcity of information on this isotope available in the literature. A close look at the mass surface in this region seems to give a slight preference for a higher mass of ^{64}Co [55], favoring the result obtained in this work. We must note, though, that with a discrepancy of 107 keV in the mass values, the mass surface cannot be used as a strong argument to decide for either value.

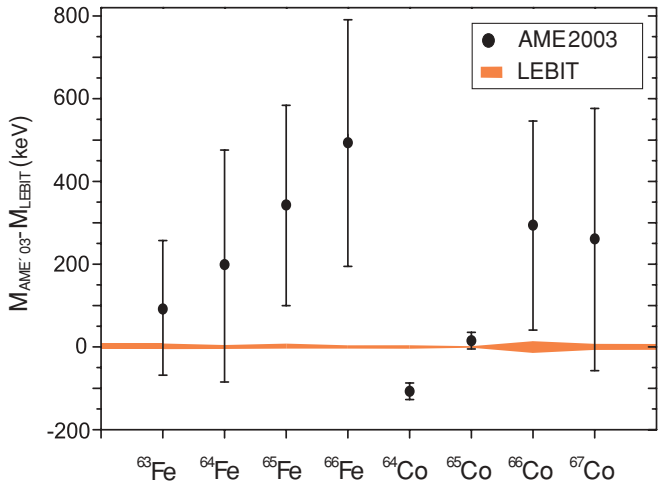

FIG. 4. (Color online) Comparison of the LEBIT results with previous literature values from AME'03.

A possible way to reconcile the results of the two high-precision mass measurements would be to assume the existence of a yet unobserved isomer in ^{64}Co with an excitation energy of E_{ex} = 107(20) keV. We could tentatively assign to the isomer a spin-parity $J^{\pi} = 5^{+}$, in a similar fashion to the known even-neighbor isotopes ^{62}Co (E_{ex} = 22 keV, $J^{\pi} = 5^{+}$) and ^{66}Co (E_{ex} = 175 keV, $J^{\pi} = 5^{+}$) [55].

Let us assume that the situation in our experiment was such that the production of the unknown 5^{+} isomeric state was favored in a much larger proportion than the production of the 1^{+} ground state, which could be justified by the particular mechanism of production of isotopes by projectile fragmentation. We could then consider a situation without precedents in which we measured a mostly pure sample of the isomeric state. The possibility of having an admixture with similar proportions of the two states in the trap can be neglected, as a closer look at the ^{64}Co data does not show any indication of resonance broadening, which otherwise would be easily observed even in the case of an admixture of states with proportions as low as 20% to 80% between the two states. Taking into account the mentioned assumptions, the Los Alamos measurement would be considered as the ground-state mass, and our value, obtained repeatedly in two different experiments, would correspond to the mass of the yet-to-be-observed isomer. A rough lower limit of the half-life could be given by taking into account the time interval that the ions spend in the system, which, in the case of ^{64}Co, is $T_{1/2} > 280$ ms (30 ms of cooling-bunching plus 250 ms of RF quadrupolar excitation), as the observed resonance is very clean and an almost-pure sample must be considered.

B. ^{65}Co

The Q value of the five-nucleon pickup reaction ^{40}Zn(^{3}He,^{8}B)^{65}Co was measured to obtain a mass excess ME = −59 168(20) keV for ^{65}Co [56]. The present value, with a 10-fold smaller uncertainty, is found to be in good agreement with this value.

C. ^{66,67}Co and $^{63-66}$Fe

These isotopes were produced at Los Alamos by proton-induced fragmentation and fission reactions. Their masses were measured using the TOF isochronous (TOFI) recoil spectrometer. In AME'03 an adjusted mass value is provided from the input data of several experiments [26–28]. Within their uncertainty of a few hundred kilo–electron volts, some of the TOFI measurements agree fully with ours. Known isomers with microsecond half-lives in this region [57] could have affected the measurements, as the TOFI mass resolution was insufficient to resolve ground and isomeric states. Should

such an admixture be present, the measurement would lead, in general, to a less bound mass determination, which would agree with the observed trend. Another explanation for the discrepancy observed in these isotopes could be an undiscovered systematic error in the TOFI mass calibration.

D. Isomers

As mentioned in the Introduction, the possibility of resolving ground and isomeric states gives Penning trap mass spectrometry the ability to assign correct ground-state mass values unambiguously. This fact is a consequence of the high resolving power achievable, which is defined as [45]

$$R = \frac{\nu_c}{\Delta\nu_c} \approx \nu_c T_{\mathrm{RF}}, \tag{3}$$

where T_{RF} is the time duration of the quadrupolar RF excitation applied to the trapped ions. The resolving power R is then mainly limited by the half-life of the species being investigated. For this reason, fast preparation of the ions for their mass determination is of utmost importance when trying to reach a high resolving power or, similarly, a high mass resolution. The LEBIT facility is designed to minimize the time necessary for efficient purification of the ion beam and subsequent injection into the Penning trap. The use of a fast TOF electrostatic deflector and the Lorentz steerer are two examples of such optimization.

Recent results of β-decay studies performed at LISOL [22] reported the discovery of an isomeric state in ^{67}Co with a half-life of 496(33) ms. Proposed spin and parity suggest a spherical $(7/2^-)$ ^{67}Co ground state and a deformed first excited $(1/2^-)$ state at 492 keV, interpreted as a proton $1p$-$2h$ prolate intruder state.

The mean TOF of the ions as a function of the applied frequency obtained in our experiment for $^{67}Co^+$ is shown in Fig. 5. In the resonance, two minima are observed, indicating the presence of two nuclear states differing in mass. After a cautious check for the presence of any possible combination of radioactive or stable contaminants in the trap that would have such a mass, we concluded that the observed additional cyclotron resonance resulted from the presence of the long-lived isomer reported earlier by LISOL. A fit of the theoretical line shape results in an excitation energy of 493(14) keV, which is in excellent agreement with the spectroscopic result.

A fit of the theoretical line shape to the resolved ground and isomeric state resonance resulted in an excitation energy of 402(10) keV for the unknown isomer in ^{65}Fe (see Refs. [23] and [24]). Note the corrected error bars of the ground and isomeric states of ^{65}Fe listed in Table II. As reported in Refs. [23] and [24], from the total time of the ions in the apparatus before detection, a rough lower limit of 150 ms for the half-life was determined. This long half-life for the new 402-keV isomer makes the $9/2^+$ state the most probable candidate, as a high spin is required to create a high γ-ray multipolarity. This assignment agrees well with the level systematics of decreasing $9/2^+$ energies until $N = 40$, also supported by shell-model calculations [58]. As a result of this new $9/2^+$ state, the spin-parity assignment given in the literature for the low-lying levels had to be modified to match the properties of the observed γ rays in ^{65,67}Fe. Using the information from the γ rays and the similarities of the known levels in the neighbor isotopes [57–60], the most likely transition multipolarities can be derived and used to infer a plausible level scheme (see Fig. 6) for the odd-A Fe isotopes with $N = 35$–41 (see Refs. [23] and [24] for a detailed explanation of the spin and parity assignments). These new assignments are kept tentative, as a slight change in deformation would change the complete picture.

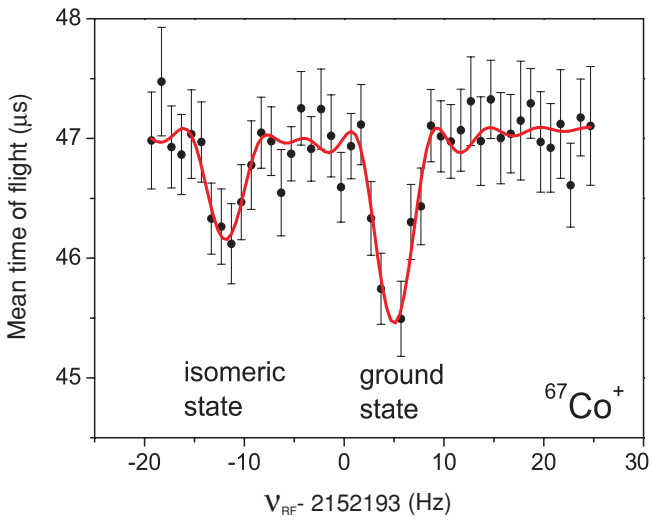

FIG. 5. (Color online) Cyclotron resonance curve for $^{67}Co^+$ ions. The two minima visible in the resonance correspond to the long-lived nuclear isomer and the ground state. The solid curve represents the fit of the theoretical line shape to the data points, which results in an excitation energy of $E_{\mathrm{ex}} = 493(14)$ keV.

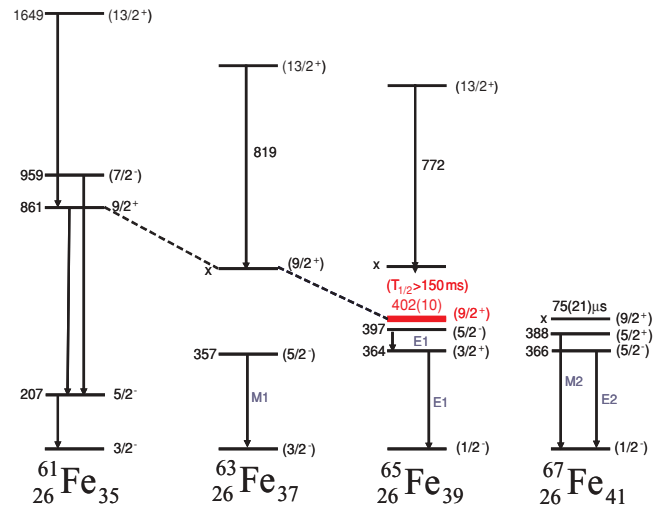

FIG. 6. (Color online) Level scheme of the odd-A $^{61-67}$Fe isotopes based on the data from Refs. [23,24], and [57–60]. Known energies are given as kilo–electron volts. Tentative spin-parity assignments are in parentheses, and unknown energies are marked with an x. The new 402-keV isomer in ^{65}Fe is highlighted by the thickest (red) horizontal bar. The multipolarity labels of the 364-keV transition in ^{65}Fe and the 366-keV transition in ^{67}Fe are corrected from the erroneous values given in Refs. [23] and [24].

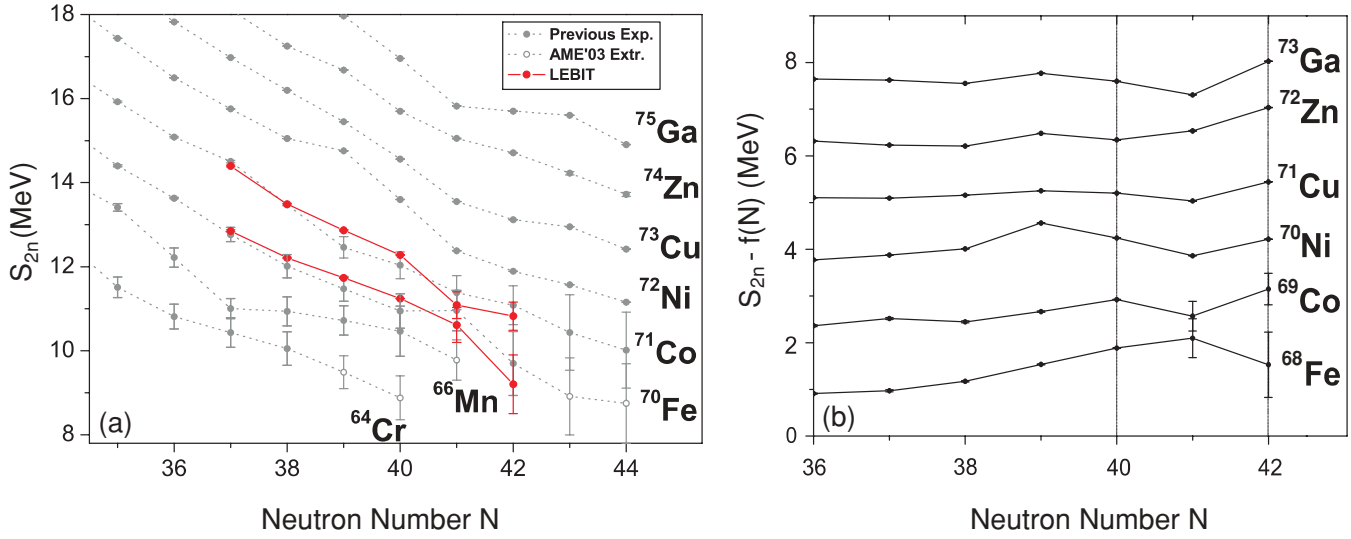

FIG. 7. (Color online) (a) Two-neutron separation energy S_{2n} as a function of the neutron number N for isotopes from chromium to gallium. Darker (red) filled circles connected by solid lines correspond to the data investigated in this work. Experimental data from AME'03 [25] and more recent data from Ref. [16] are plotted as lighter (gray) filled circles, whereas open circles correspond to extrapolations based on systematic trends [25]. (b) To visualize better the discontinuities of the mass surface at $N = 40$, S_{2n} values with a linear function of N subtracted are shown.

E. Mass surface at $N = 40$, $Z = 28$

The two-neutron separation energy S_{2n} of a given nucleus is a measure of the binding energy of the last two valence neutrons and is a useful tool to visualize effects owing to shell structure and nuclear deformation in the mass surface. S_{2n} values as a function of the neutron number N around ^{68}Ni are shown in Fig. 7(a). Lighter (gray) filled circles in the figure represent previous experimental data from AME'03, with the exception of those corresponding to $^{70-72}$Ni and ^{73}Cu, which were recently measured at JYFLTRAP [16]. Note here that the data from Ref. [15], although published in 2007, were included in AME'03. Open circles represent data extrapolated in AME'03 from the systematic trends in this mass region. The darker (red) filled circles are the S_{2n} values affected by the data obtained in this work. For the Fe and Co isotopes up to $N = 40$ our more precise data indicate a slight trend toward enhanced binding. For the heavier isotopes ($N > 40$) the uncertainties in the S_{2n} values are dominated or completely determined by the large uncertainties in the TOFI data. The use of these data are necessary to calculate the S_{2n} values beyond $N = 40$, which consequently have a larger error than the $N < 40$ isotopes, and they turn out to be less bound than the previous experimental values. Considering the trend to less bound mass excess values observed in the TOFI measurement (see Fig. 4), one would expect a slightly smaller effect on the slope between $N = 40$ and $N = 41$, especially for the Fe isotopes.

Figure 7(b) shows the S_{2n} values after subtraction of a linear function $f(N)$ of the neutron number N, to better visualize the departure from a linear trend typically observed. Here, the function $f(N)$ is obtained from a linear fit of the Ni isotopes from $N = 35$ to $N = 44$. A deviation from a linear trend can indicate a subshell closure or other nuclear structure effects. Unfortunately, more precise mass measurements of ^{67,68}Fe and ^{68,69}Co are required to reduce the error bars in the S_{2n} values for $N = 41$ and $N = 42$ and, thus, to be able to draw a firm conclusion about the indication of a weak subshell closure at $N = 40$, as observed in nickel, copper, and gallium isotopes. For iron no big change in the slope is foreseen, as observed in zinc, owing to an expected reduction in the shell gap. This reduction, resulting from the lowering of the energy of the $g_{9/2}$ orbital, would be the cause of an onset of deformation [17] that has been estimated to reach $\beta_2 \approx 0.3$ for $^{65-67}$Fe and slightly lower values for the isotopes $^{70-72}$Zn [61]. Moreover, the existence of this onset of deformation has also been correlated with the low 2^+ energies measured in the Fe and Zn isotopes near $N = 40$ [10,62].

V. CONCLUSIONS

High-precision mass measurements of $^{63-66}$Fe and $^{64-67}$Co have been carried out with the LEBIT Penning trap mass spectrometer in two different experiments (see Refs. [23] and [24]). A comparison of the LEBIT data with previous experimental results shows a discrepancy of 5 standard deviations for ^{64}Co. The rest of the mass values investigated are found to be in agreement ($<2\sigma$). Given the small uncertainty of both the new and the previous ^{64}Co mass measurements, we consider the possibility of an unknown isomer of ^{64}Co produced and observed in our experiment rather than the ground state. Under this assumption, the LEBIT mass value for ^{64}Co would correspond to that of this newly found isomeric state with an excitation energy of 107(20) keV ($J^\pi = 5^+$). All mass uncertainties of the nuclides studied have been significantly reduced: for most of the cases by up to 2 orders of magnitude.

A long-lived isomer in ^{67}Co, recently discovered at LISOL [22], has been confirmed by LEBIT. The excitation energy of 493(14) keV obtained from the measured mass difference

between the isomeric and the ground state is in full agreement with the value of 492 keV obtained in β-decay studies.

A new long-lived nuclear isomer [23,24] was discovered in ^{65}Fe for the first time by Penning trap mass spectrometry. An excitation energy of 402(10) keV was determined from the measured mass difference between the isomeric and the ground states.

With the LEBIT data the two neutron separation energies S_{2n} for Fe and Co isotopes around $Z = 28$, $N = 40$, have been considerably improved. The observed small discontinuities in the S_{2n} values reported previously in neighboring chains of the elements Ni, Cu, and Ga [15,16] cannot be confirmed in the Co isotopes owing to the large error bars in the TOFI measurements affecting the $N > 40$ data. The same problem is found in the Fe isotopes, where an expected reduction of the neutron shell gap consistent with the strong deformation predicted in this region cannot yet be verified.

Penning trap mass measurements of the ^{67,68}Fe and ^{68,69}Co isotopes would be required to reduce the large uncertainties of the S_{2n} values. Only then will we be able to provide a more forceful argument on the peculiar shell evolution in this region.

ACKNOWLEDGMENTS

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[1] M. G. Mayer, Phys. Rev. **75**, 1969 (1949).
[2] O. Haxel *et al.*, Phys. Rev. **75**, 1766 (1949).
[3] W. Nazarewicz and R. F. Casten, Nucl. Phys. A **682**, 295c (2001).
[4] J. Fridmann, I. Wiedenhöver, A. Gade, L. T. Baby, D. Bazin, B. A. Brown, C. M. Campbell, J. M. Cook, P. D. Cottle, E. Diffenderfer *et al.*, Nature **435**, 922 (2005).
[5] A. Ozawa, T. Kobayashi, T. Suzuki, K. Yoshida, and I. Tanihata, Phys. Rev. Lett. **84**, 5493 (2000).
[6] S. N. Liddick, P. F. Mantica, R. Broda, B. A. Brown, M. P. Carpenter, A. D. Davies, B. Fornal, T. Glasmacher, D. E. Groh, M. Honma *et al.*, Phys. Rev. C **70**, 064303 (2004).
[7] M. Kowalska, Hyperfine Interact. **171**, 167 (2006).
[8] C. Thibault, R. Klapisch, C. Rigaud, A. M. Poskanzer, R. Prieels, L. Lessard, and W. Reisdorf, Phys. Rev. C **12**, 644 (1975).
[9] Y. Utsuno, T. Otsuka, T. Glasmacher, T. Mizusaki, and M. Honma, Phys. Rev. C **70**, 044307 (2004).
[10] P. Adrich, A. M. Amthor, D. Bazin, M. D. Bowen, B. A. Brown, C. M. Campbell, J. M. Cook, A. Gade, D. Galaviz, T. Glasmacher, S. McDaniel, D. Miller, A. Obertelli, Y. Shimbara, K. P. Siwek, J. A. Tostevin, and D. Weisshaar, Phys. Rev. C **77**, 054306 (2008).
[11] M. Bernas, P. Dessagne, M. Langevin, J. Payet, F. Pougheon, and P. Roussel, Phys. Lett. B **113**, 279 (1982).
[12] R. Broda, B. Fornal, W. Kròlas, T. Pawlat, D. Bazzacco, S. Lunardi, C. Rossi-Alvarez, R. Menegazzo, G. de Angelis, P. Bednarczyk *et al.*, Phys. Rev. Lett. **74**, 868 (1995).
[13] O. Sorlin, S. Leenhardt, C. Donzaud, D. Duprat, F. Azaiez, F. Nowacki, H. Grawe, Z. Dombrádi, F. Amorini, A. Astier *et al.*, Phys. Rev. Lett. **88**, 092501 (2002).
[14] K. Langanke, J. Terasaki, F. Nowacki, D. J. Dean, and W. Nazarewicz, Phys. Rev. C **67**, 044314 (2003).
[15] C. Guénaut, G. Audi, D. Beck, K. Blaum, G. Bollen, P. Delahaye, F. Herfurth, A. Kellerbauer, H.-J. Kluge, J. Libert *et al.*, Phys. Rev. C **75**, 044303 (2007).
[16] S. Rahaman, J. Hakala, V.-V. Elomaa, T. Eronen, U. Hager, A. Jokinen, A. Kankainen, I. D. Moore, H. Penttilä, S. Rinta-Antila *et al.*, Eur. J. Phys. A **34**, 5 (2007).
[17] M. Hannawald *et al.* (ISOLDE Collaboration), Phys. Rev. Lett. **82**, 1391 (1999).
[18] O. Sorlin, C. Donzaud, F. Nowacki, J. Angélique, F. Azaiez, C. Bourgeois, V. Chiste, Z. Dlouhy, D. G.-M. S. Grévy, F. Ibrahim *et al.*, Eur. Phys. J. A **16**, 55 (2003).
[19] N. Aoi, H. Suzuki, E. Takeshita, S. Takeuchi, S. Ota, H. Baba, S. Bishop, T. Fukui, Y. Hashimoto, H. Ong *et al.*, Nucl. Phys. A **805**, 400c (2008).
[20] T. Otsuka, R. Fujimoto, Y. Utsuno, B. A. Brown, M. Honma, and T. Mizusaki, Phys. Rev. Lett. **87**, 082502 (2001).
[21] K. Blaum, Phys. Rep. **425**, 1 (2006).
[22] D. Pauwels, O. Ivanov, N. Bree, J. Büscher, T. E. Cocolios, J. Gentens, M. Huyse, A. Korgul, Y. Kudryavtsev, R. Raabe *et al.*, Phys. Rev. C **78**, 041307(R) (2008).
[23] M. Block, C. Bachelet, G. Bollen, M. Facina, C. M. Folden III, C. Guénaut, A. A. Kwiatkowski, D. J. Morrissey, G. K. Pang, A. Prinke *et al.*, Phys. Rev. Lett. **100**, 132501 (2008).
[24] M. Block, C. Bachelet, G. Bollen, M. Facina, C. M. Folden III, C. Guénaut, A. A. Kwiatkowski, D. J. Morrissey, G. K. Pang, A. Prinke *et al.*, Phys. Rev. Lett. **101**, 059901(E) (2008).
[25] G. Audi and A. H. Wapstra, Nucl. Phys. A **729**, 337 (2003).
[26] X. L. Tu, X. G. Zhou, D. J. Vieira, J. M. Wouters, Z. Y. Zhou, H. L. Seifert, and V. G. Lind, Z. Phys. A **337**, 361 (1990).
[27] H. L. Seifert, J. M. Wouters, D. J. Vieira, H. Wollnik, X. G. Zhou, X. L. Tu, Z. Y. Zhou, and G. W. Butler, Z. Phys. A **349**, 25 (1994).
[28] Y. Bai, D. J. Vieira, H. L. Seifert, and J. M. Wouters, AIP Conf. Proc. **455**, 90 (1998).
[29] G. Bollen, H.-J. Kluge, M. König, T. Otto, G. Savard, H. Stolzenberg, R. B. Moore, G. Rouleau, and G. Audi, Phys. Rev. C **46**, 2140(R) (1992).
[30] S. Schwarz, F. Ames, G. Audi, D. Beck, G. Bollen, J. Dilling, F. Herfurth, H.-J. Kluge, A. Kellerbauer, A. Kohl *et al.*, Hyperfine Interact. **132**, 335 (2001).
[31] G. Bollen, C. Bachelet, M. Block, D. Davies, M. Facina, C. Folden, C. Guénaut, J. Huikari, E. Kwan, A. Kwiatowski *et al.*, Eur. J. Phys. Special Topics **150**, 337 (2007).
[32] C. Rauth, D. Ackermann, G. Audi, M. Block, A. Chaudhuri, S. Eliseev, F. Herfurth, F. Hessberger, S. Hofmann, H.-J. Kluge *et al.*, Eur. J. Phys. Special Topics **150**, 329 (2007).
[33] R. Ringle, P. Schury, T. Sun, G. Bollen, D. Davies, J. Huikari, E. Kwan, D. Morrissey, A. Prinke, J. Savory *et al.*, Int. J. Mass Spectrom. **251**, 300 (2006).
[34] G. Bollen, D. Davies, M. Facina, J. Huikari, E. Kwan, P. A. Lofy, D. J. Morrissey, A. Prinke, R. Ringle, J. Savory *et al.*, Phys. Rev. Lett. **96**, 152501 (2006).

[35] R. Ringle, T. Sun, G. Bollen, D. Davies, M. Facina, J. Huikari, E. Kwan, P. A. Lofy, D. J. Morrissey, A. Prinke *et al.*, Phys. Rev. C **75**, 055503 (2007).
[36] P. Schury, C. Bachelet, M. Block, G. Bollen, D. A. Davies, M. Facina, C. M. Folden III, C. Guénaut, J. Huikari, E. Kwan *et al.*, Phys. Rev. C **75**, 055801 (2007).
[37] L. Weissman, P. Lofy, D. Davies, D. Morrissey, P. Schury, S. Schwarz, T. Sun, and G. Bollen, Nucl. Phys. A **746**, 655c (2004).
[38] P. Schury, G. Bollen, M. Block, D. J. Morrissey, R. Ringle, A. Prinke, J. Savory, S. Schwarz, and T. Sun, Hyperfine Interact. **173**, 165 (2006).
[39] S. Schwarz, G. Bollen, D. Lawton, A. Neudert, R. Ringle, P. Schury, and T. Sun, Nucl. Instrum. Methods Phys. Res. B **204**, 474 (2003).
[40] R. Ringle, G. Bollen, A. Prinke, J. Savory, P. Schury, S. Schwarz, and T. Sun, Nucl. Instrum. Methods Phys. Res. A **604**, 536 (2009).
[41] L. S. Brown and G. Gabrielse, Rev. Mod. Phys. **58**, 233 (1986).
[42] G. Bollen, *The Euroschool Lectures on Physics with Exotic Beams*, Vol. I (Springer Berlin/Heidelberg, 2004).
[43] F. G. Major, V. N. Gheorghe, and G. Werth, *Charged Particle Traps* (Springer, New York, 2004).
[44] G. Gräff, H. Kalinowsky, and J. Traut, Z. Phys. A **297**, 35 (1980).
[45] M. König, G. Bollen, H.-J. Kluge, T. Otto, and J. Szerypo, Int. J. Mass Spectrom. Ion Processes **142**, 95 (1995).
[46] G. Bollen, H.-J. Kluge, T. Otto, G. Savard, L. Schweikhard, H. Stolzenberg, G. Audi, R. Moore, and G. Rouleau, J. Mod. Opt. **39**, 257 (1992).
[47] D. J. Morrissey, B. M. Sherrill, M. Steiner, A. Stolz, and I. Wiedenhoever, Nucl. Instrum. Methods Phys. Res. B **204**, 90 (2003).
[48] R. Ringle, G. Bollen, A. Prinke, J. Savory, P. Schury, S. Schwarz, and T. Sun, Int. J. Mass Spectrom. **263**, 38 (2006).
[49] G. Audi, O. Bersillon, J. Blachot, and A. H. Wapstra, Nucl. Phys. A **729**, 3 (2003).
[50] N. R. Daly, Rev. Sci. Instrum. **31**, 264 (1960).
[51] M. Redshaw, B. J. Mount, and E. G. Myers, Phys. Rev. A **79**, 012507 (2009).
[52] M. Redshaw, J. McDaniel, and E. G. Myers, Phys. Rev. Lett. **100**, 093002 (2008).
[53] R. Birge, Phys. Rev. **40**, 207 (1932).
[54] E. Flynn and J. Garret, Phys. Lett. B **42**, 49 (1972).
[55] G. Audi and W. Meng (private communication, 2009).
[56] R. T. Kouzes and D. Mueller, Nucl. Phys. A **307**, 71 (1978).
[57] R. Grzywacz, C. B. R. Béraud, A. Emsallem, M. Glogowski, H. Grawe, D. Guillemaud-Mueller, M. Hjorth-Jensen, M. Houry, M. Lewitowicz, A. C. Mueller *et al.*, Phys. Rev. Lett. **81**, 766 (1998).
[58] S. Lunardi, S. M. Lenzi, F. D. Vedova, E. Farnea, A. Gadea, N. Mărginean, D. Bazzacco, S. Beghini, P. G. Bizzeti, A. M. Bizzeti-Sona *et al.*, Phys. Rev. C **76**, 034303 (2007).
[59] J. M. Daugas, M. Sawicka, M. Pfützner, I. Matea, H. Grawe, R. Grzywacz, N. L. Achouri, J. C. Angélique, D. Baiborodin, F. Becker *et al.*, AIP Conf. Proc. **831**, 427 (2006).
[60] N. Vermeulen, S. K. Chamoli, J. M. Daugas, M. Hass, D. L. Balabanski, J. P. Delaroche, F. de Oliveira-Santos, G. Georgiev, M. Girod, G. Goldring *et al.*, Phys. Rev. C **75**, 051302(R) (2007).
[61] Y. Aboussir, J. M. Pearson, A. K. Dutta, and F. Tondeur, ADNDT **61**, 127 (1995).
[62] S. Raman, C. W. Nestor, S. Kahane, and K. H. Bhatt, Phys. Rev. C **43**, 556 (1991).