Direct Measurement of the 4.6 MeV Isomer in Stored Bare ^{133}Sb Ions

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Abstract

The core-excited isomer in fully-ionized ^{133}Sb has been directly studied for the first time by applying the novel technique of isochronous mass spectrometry at GSI. The neutron-rich nuclides in high charge states were produced by projectile fission of 411 MeV·A ^{238}U ions, separated in flight by the fragment separator (FRS) and stored in the isochronous storage ring (ESR). The measured excitation energy is 4.56(10) MeV. The neutral-atom half-life is known to be 17 μs. This is the shortest-lived isomer measured directly with mass

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spectrometry techniques. The extended in-flight half-life of the bare ions in the ESR, which is due to the exclusion of the strong internal conversion, demonstrates that there should be another nuclear level above that identified from isomer-decay spectroscopy, in support of shell-model calculations. This measurement opens up a new half-life domain for storage-ring measurements.

Keywords: nuclear mass, isomer, nuclear half-live, shell model
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The region around the doubly-magic ^{132}Sn nucleus is currently a subject of great theoretical and experimental interest [1, 2]. It bridges the gap between the lighter doubly closed-shell systems (^{16}O, ^{40}Ca, ^{48}Ca) and ^{208}Pb, and therefore plays an essential role in developing models with universal nucleon-nucleon interactions. In modern calculations, such data allow the extraction of the properties of the residual nucleon-nucleon interactions, thus giving a new impetus to predict nuclear structure of more exotic systems. In particular, it is important to study this region for answering one of the major questions in nuclear physics, i.e. whether the traditional shell-gap properties of $Z = 50$ and $N = 82$ remain when going toward the neutron dripline [3, 4, 5]. In the realm of nuclear astrophysics, the stability of the $N = 82$ shell gap has important consequences for understanding the r-process abundance peak at $A = 130$ [6].

One such test case is ^{133}Sb, which has a single proton outside the doubly-magic ^{132}Sn core. It can give direct information on the low-energy single-proton excitations for the ^{132}Sn region, and above 4 MeV on the coupling of the valence proton to the exited core. Investigations of this nuclide thus allow stringent tests of the shell model in this region. In this work we report on the first direct measurement of the 4.6 MeV isomeric state in ^{133}Sb nuclei. A low half-life limit on the γ-deexcitation channel, i.e. 58^{+47}_{-18} μs (1σ), has been obtained due to the disabled internal conversion decay mode in fully-ionized nuclei. We like to note that among numerous applications, such investigations of pure decay modes and related life-time modifications are of basic interest for understanding decay phenomena.

Shortly after the first experimental information about ^{133}Sb in 1973 [7], an isomer with a half-life of 17 μs and spin $I \geq 13/2$ was reported [8]. All the low-lying single-proton excitations in ^{133}Sb were subsequently identified [9, 10]. More recent detailed studies with the LOHENGRIN spectrometer [11] and the EUROGAM2 gamma detector array [12] confirmed the existence

of the 17 μs isomer and interpreted it as the $I^\pi = 21/2^+$ member of the $\pi g_{7/2} \otimes \nu(f_{7/2}h_{11/2}^{-1})$ multiplet based on shell-model calculations. However, no direct information on the isomeric decay transition was obtained. It was concluded that the isomer decays by a highly converted $E2$ transition with an estimated energy of less than 20 keV [11]. A more extensive work [13] further clarified the high-energy structure populated in the decay of the 17 μs isomer, but left open the question of the direct decay of the isomer itself. It has remained, therefore, a considerable experimental challenge to demonstrate that the calculated $I^\pi = 21/2^+$ shell-model state is in fact correctly predicted.

Isomer investigations were performed recently with stored ions in storage rings and Penning traps, e.g. with excitation energies of 103(12) keV ($T_{1/2} > 1$ s) in ^{125}Ce [14], and of 402(5) keV ($T_{1/2} > 150$ ms) in ^{65}Fe [15]. The present work applies a novel method to measure mass-resolved nuclear isomers with lifetime down to a few tens of μs. The ions of interest have been stored in the storage ring (ESR) [16] tuned to isochronous ion-optical mode [17]. Compared to our former measurements, the isochronous mass spectrometry (IMS) method has been significantly improved to achieve a higher mass resolving power [18]. The first direct mass measurement of the 4.6 MeV isomer in ^{133}Sb ($T_{1/2} = 17$ μs in the neutral atom) has now been performed. This result marks the shortest-lived isomer investigated with direct mass spectrometry techniques. Both the ground state and the isomeric state in ^{133}Sb have been produced. Due to different rest masses, they were well resolved in the experiment via the difference in their revolution times. In this way the excitation energy of the isomer has been determined directly. Simultaneously, we obtained a low limit for the half-life of the isomer, as a bare ion, which usefully constrains the nuclear structure properties.

The IMS technique was developed based on the unique combination of the in-flight fragment separator (FRS) [19] and the ESR, which was used as a high-resolution multi-turn time-of-flight spectrometer. The ESR was operated in the isochronous mode, which provides first-order orbital-frequency compensation for different velocities, leading to an unambiguous relationship between the revolution times of the circulating ions and their mass-to-charge ratios [17]. The IMS technique is presently suitable for mass measurements of exotic nuclides with lifetimes longer than about a few tens of μs. It is only limited by the flight time through the FRS and the necessity to complete about 100 revolutions in the ESR for high-resolution measurements. Recently, the technique was improved significantly by employing an additional magnetic-rigidity determination [20], and the first large-scale mass-

measurement program for neutron-rich uranium fission fragments was successfully carried out [18]. The masses of 35 nuclides, including 8 cases for the first time, were directly measured. This present work is part of that program.

An important feature of the mass-measurement program with the FRS-ESR facilities is that the stored relativistic ions are highly charged. Under this condition the observed half-lives of nuclear isomeric states can be much longer if the electron conversion branch is suppressed [21]. In principle, this presents the possibility to observe bare isomers with lifetimes that would be even shorter than μs in the corresponding neutral atoms.

For the present measurement, neutron-rich exotic nuclei were produced by projectile fission of 411 MeV·A ^{238}U ions in a 1 g/cm^2 Be target placed at the entrance of the FRS. The energy of the primary beam was chosen such that the energy of ^{133}Sn^{50+} fragments corresponded to the relativistic Lorentz factor γ=1.41 which matches exactly the isochronous condition of the ESR. The intensity of the primary beam was about $2 \cdot 10^9$ particles per spill. The fission fragments emerged from the target mainly as bare ions. The uranium fission products were then separated in flight according to their magnetic rigidity and injected into the ESR. The magnetic-rigidity of the fission fragments has been determined at the focal plane S2 of the FRS, which defines precisely the velocity spread ($\Delta v/v$) of fragments to about 10^{-4} [20].

A fast timing detector has been used to record time stamps of each ion circulating in the ESR. The detector consists of a thin (17 μg/cm^2) carbon foil coated with 10 μg/cm^2 CsI on both sides, and was inserted into the ring aperture [22]. The ions penetrated the foil at every revolution thus releasing secondary electrons. These electrons were guided by electric and magnetic fields to two sets of micro channel detectors. The signals were recorded and then used to determine the revolution times of the ions. Each measurement cycle was 1 ms long which corresponded to about 2000 revolutions of the ions in the ESR. On average there was about one stored ion per injection. More than 13000 independent injections were made over four days.

The extracted revolution times of different nuclides were accumulated in a spectrum, in which more than 70 different peaks were unambiguously identified as highly-charged ions. A portion of the spectrum is shown in Fig. 1, where the resolving power is $m/\Delta m$(FWHM) $\approx$ 60000. The ground-state ^{133}Sb ions were observed in two charge states, 121 times as the fully ionized state and 15 times as hydrogen-like ions. Five ions were unambiguously identified as bare ^{133}Sb ions in the isomeric state. No other fragments in different

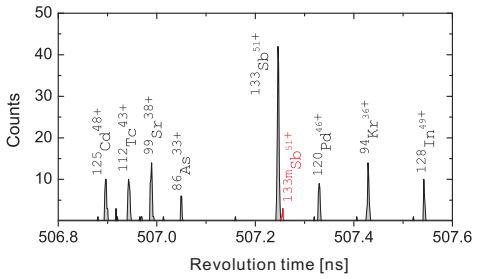

Figure 1: (Color online) Part of the revolution-time spectrum including the ^{133}Sb ions in the ground and isomeric states. The identified ions with well-known masses are indicated in the spectrum.

charge states can account for this revolution time. Taking into account the identical transmission through the FRS and the injection into the ESR for both ground and isomeric states, it is evident that the isomeric state accounts for about 4% of the fission yield of ^{133}Sb.

In the off-line data analysis, any slow drift of the revolution times common for the entire spectrum, e.g., due to instabilities of the magnet power supplies, was automatically corrected by using the correlation matrix method [23, 18]. In this way the mass resolving power was further improved and amounted to $m/\Delta m(\mathrm{FWHM}) \approx 2 \cdot 10^5$ as illustrated in Fig. 2.

As already mentioned in Ref. [18], the lack of accurately known reference masses in the region of neutron-rich fission fragments can limit the accuracy achieved in the final mass evaluation. An attractive characteristic of our analysis, however, is that any new reference mass can be easily added to re-calibrate the entire dataset. In the present analysis, the new results from the Penning-trap measurements [24] have been used for calibration, in addition to those listed in Ref. [18]. Consequently, the typical mass uncertainty has been decreased to about 100 keV. Except for the different calibrations, the data analysis is the same as that described in detail in Ref. [18]. The excitation energy of the ^{133}Sb isomeric state is thus determined to be 4560(100) keV, which agrees well with the estimate of $4526 < E^* < 4546$ keV in Ref. [11]. As a consistency check, the 15 counts of fully ionized ^{133}Sn (see Fig. 2) provide a mass difference between ^{133}Sn and ^{133g}Sb of 8130(100) keV, which is in good agreement with the literature value of 8096(34) keV [25, 4].

In the IMS technique, presently most of the stored ions are lost in the ring during the 1 ms measurement. This can be understood as mainly arising from the non-radioactive losses, in particular the energy-loss and charge-exchange

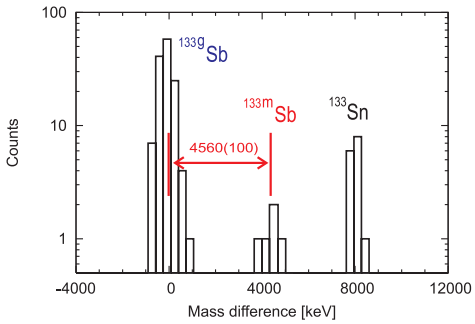

Figure 2: (Color online) Mass distribution for $A = 133$ isobars relative to the ^{133}Sb ground state from the final data evaluation. The isomeric state in ^{133}Sb with an excitation energy $E^* = 4560(100)$ keV is clearly resolved from the corresponding ground state. Note that the ^{133}Sn events were observed with charge state $q = 50$.

processes during multiple passages through the detector foil at each turn and the interaction with the residual gas atoms in the ring. The disappearance of ions can be quantified by measuring the "survival time" between the injection into the ESR and the time stamp of the last passage through the detector.

No evidence for β^- decay or isomeric transitions was observed in the time range of the present measurement. This is not surprising, though, considering that the recording time is much less than the half-lives of the radionuclides investigated, at least for the ground-state decays.

The distribution of the survival times in the ring for both ground and isomeric states in ^{133}Sb are displayed in Fig. 3. An exponential-like decay is observed, and we fitted the ground-state distribution with the equation:

$$\frac{\Delta N(t)}{\Delta t} = N_0 \cdot \lambda_{loss} \cdot e^{-\lambda_{loss} t} \ , \tag{1}$$

where N_0 is the number of ions at the time of injection ($t = 0$) and the decay constant λ_{loss} depends on both the radioactive and non-radioactive losses of ions in the ring. The least-squares fitted value of $\lambda_{loss} = 0.0073(6)\ \mu s^{-1}$ corresponds to a mean survival time of 137(11) μs in the laboratory frame, or a survival half-life of $T_{loss} = 68(6)$ μs in the rest frame (using the Lorentz factor of 1.41) which is much less than nuclear-decay half-life of 2.5(1) m [25]. Therefore, $T_{loss} = 68$ μs should be related solely to the non-radioactive loss process in the ring rather than the real nuclear decay.

Considering now the 5 measured ions in the isomeric state, the survival times range up to 220 μs (in the laboratory frame) following the initial 50

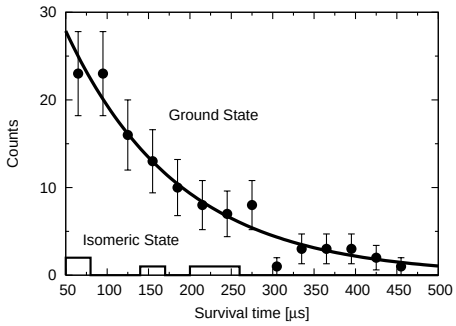

Figure 3: Survival-time distributions in the ESR for the fully-ionized ground state and isomeric state in ^{133}Sb. The solid line shows a fit to the ground-state data with an exponential decay function. The bin width in the histogram is 30 μs.

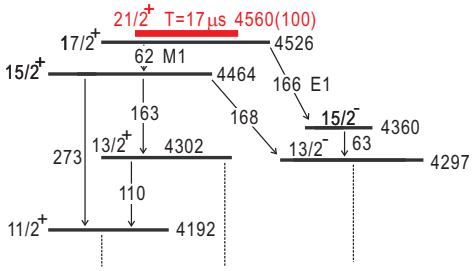

Figure 4: (Color online) Level scheme of ^{133}Sb above 4 MeV. Excited levels and γ transitions are labeled with energies given in keV. The levels and transitions below the isomer are from Refs. [11, 13]

μs used for identification. The mean survival time is 118 μs, yielding a survival half-life in the centre-of-mass frame of 58^{+47}_{-18} μs (1σ) [26]. This is in agreement with the 68(6) μs obtained for the ground state, and there is no evidence for nuclear decay of the isomeric state during the observation period in the storage ring. This result is pivotal in establishing experimentally the existence of the $I^\pi = 21/2^+$ isomer, as distinct from the highest known level of $I^\pi = 17/2^+$ to which it has been assumed to decay, guided by shell-model predictions.

As already pointed out, direct gamma radiation from the assumed $I^\pi = 21/2^+$ isomer has not been observed in other experiments, and the limit placed on the decay energy is less than 20 keV [11]. The possibility is now addressed that the isomeric state might in fact be the $I^\pi = 17/2^+$, 4526 keV state itself [8](See Fig. 4). That state is known [13] to decay by about 98%

to the $15/2^+$ level via a 62 keV $M1$ transition [8, 11] with a total electron-conversion coefficient of $\alpha_T = 2.5$ [27], and 2% to the $15/2^-$ level via a 166 keV $E1$ transition. If these transitions were to account for the neutral-atom half-life of 17 μs, then the corresponding bare-ion half-life would be $T_{nuclear} = 56$ μs. To estimate the center-of-mass survival half-life T for the isomer in the ESR, this should be combined with the $T_{loss} = 68(6)$ μs survival half-life via $T = 1/(1/T_{nuclear}+1/T_{loss})$, thus yielding an expected overall survival half-life of 31(1) μs. This is not consistent with the measured isomer survival half-life of 58^{+47}_{-18} μs (1σ). For our low statistics measurement, the survival half-life at the 2σ confidence level is 58^{+495}_{-27} [26] enabling the above hypothesis to be rejected. Therefore, there is another state that leads to the experimental isomer properties. Indeed, there is no problem in understanding the long bare-ion survival time on this basis. The experimental upper limit of 20 keV for the $E2$ decay of the $I^\pi = 21/2^+$ state gives a calculated $\alpha_T = 991$. This leads to a bare-ion half-life of about 17 ms (rather than the 17 μs of the neutral atom) which is consistent with the lack of evidence for nuclear decay during the 1 ms observation time.

We can thus conclude that at the 95% confidence level the ^{133}Sb isomer with a neutral-atom half-life of 17 μs is located above the $I^\pi = 17/2^+$, 4526 keV state, giving direct experimental evidence for the shell-model prediction of an energetically favored $I^\pi = 21/2^+$ state [11, 12, 13]. The above 4 MeV level scheme is updated in Fig. 4.

In summary, the novel IMS technique has been employed to investigate the 17 μs isomer in ^{133}Sb. The excitation energy and survival time of the isomer have been determined based on precise revolution time measurements of a few individually stored ions. These results give direct experimental support for modern shell-model predictions. Furthermore, by employing the IMS technique it is possible to gain access to rare nuclear isomers, including those associated with highly converted decay transitions, which can be very difficult or impossible to measure by γ-ray and electron spectroscopy. This achievement has opened the way to accurate mass measurements of short-lived exotic isomeric states, which have a great potential for nuclear structure and nuclear astrophysics investigations at the future FAIR facility [28]. We would like to emphasize that such measurements are extremely sensitive since they require only a few ions in total and therefore can be applied to the rarest nuclear species.

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[1] L. Coraggio *et al.*, Prog. Part. Nucl. Phys. 62 (2009) 135 .

[2] J.A. Pinston and J. Genevey, J. Phys. G 30 (2004) R57.

[3] A. Jungclaus *et al.*, Phys. Rev. Lett. 99 (2007) 132501.

[4] M. Dworschak *et al.*, Phys. Rev. Lett. 100 (2008) 072501.

[5] M. Górska *et al.*, Phys. Lett. B 672 (2009) 313.

[6] H. Grawe, K. Langanke, and G. Martinez-Pinedo, Rep. Prog. Phys. 70 (2007) 1525.

[7] S. Borg *et al.*, Nucl. Phys. A 212 (1973) 197.

[8] K. Sistemich *et al.*, Z. Phys. A 285 (1978) 305.

[9] J. Blomqvist *et al.*, Z. Phys. A 314 (1983) 199.

[10] M. Sanchez-Vega *et al.*, Phys. Rev. Lett. 80 (1998) 5504.

[11] J. Genevey *et al.*, Eur. Phys. J. A 7 (2000) 463.

[12] W. Urban *et al.*, Phys. Rev. C 62 (2000) 027301.

[13] W. Urban *et al.*, Phys. Rev. C 79 (2009) 037304.

[14] B. Sun *et al.*, Eur. Phys. J. A 31 (2007) 393.

[15] M. Block *et al.*, Phys. Rev. Lett. 100 (2008) 132501.

[16] B. Franzke, Nucl. Instr. Meth. B 24/25 (1987) 18.

[17] M. Hausmann *et al.*, Nucl. Instr. Meth. A 446 (2000) 569.

[18] B. Sun *et al.*, Nucl. Phys. A 812 (2008) 1.

[19] H. Geissel *et al.*, Nucl. Instr. Meth. B 70 (1992) 286.

[20] H. Geissel *et al.*, Hyperfine Interact. 173 (2006) 49.

[21] Yu.A. Litvinonv *et al.*, Phys. Lett. B 573 (2003) 80 .

[22] J. Trötscher *et al.*, Nucl. Instr. Meth. B 70 (1992) 455.

[23] T. Radon *et al.*, Nucl. Phys. A 677 (2000) 75 .

[24] J. Hakala *et al.*, Phys. Rev. Lett. 101 (2008) 052502.

[25] G. Audi *et al.*, Nucl. Phys. A 729 (2003) 337.

[26] K.-H. Schmidt et al., Z. Phys. A 316 (1984) 19.

[27] T. Kibédi *et al.*, Nucl. Instr. Meth. A 589 (2008) 202.

[28] C. Scheidenberger *et al.*, Hyperfine Interact. 173 (2006) 61.