Precise Atomic Mass Values near ^{132}Sn: The Resolution of a Puzzle

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The total β^- decay energies of 14 nuclides in the vicinity of ^{132}Sn have been measured using high resolution spectroscopic methods. The present results derived for the atomic masses in this region are significantly more precise than the previously accepted values, and differ significantly from these in some cases. The precision of the new mass values surpasses that expected from the systems currently proposed for direct mass measurements of nuclei far from stability. The present results resolve a recently observed and highly puzzling discrepancy between the experimental mass values in the ^{132}Sn region and theoretical systematics. [S0031-9007(99)08609-3]

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The mass data in the vicinity of doubly closed shells (DCS) for nucleons give highly valuable structure information. This has a particular significance for the far-from-stability regions where new phenomena may arise due to low binding energies. Precise binding energies, derived from the atomic masses, are also of critical importance for the modeling of the astrophysical r process, especially in the regions bordering the doubly closed shell nuclei of ^{78}Ni and ^{132}Sn. Consequently, it was of particular concern that the accepted masses of the neutron rich $N = 82$ isotones in the vicinity of ^{132}Sn have been recently questioned [1] on the grounds of a "serious inconsistency" with predictions based on a shell model reduction technique. The authors compare a mass "window" W calculated using a specific combination of the $N = 82$ ground state masses, to the value of W extracted via a simple formula involving the experimental excited state energies in ^{134}Te and ^{135}I. A significant difference of almost 500 keV has been noted [1] between $W = -3570$ keV from level spectroscopy and $W = -3080\ (150)$ keV from the $N = 82$ masses [2]. Since such a comparison gives an agreement to within a few keV for the $N = 126$ isotopes at ^{208}Pb, the authors conclude [1] that the $N = 82$ mass values could be inaccurate by considerably more than the estimated errors. However, this particular conclusion was later challenged in the first theoretical work [3] employing a realistic effective interaction for shell model calculations of the $N = 82$ isotones. The authors of Ref. [3] claim that some approximations used in the derivation of W, as well as configuration admixtures in the states involved, can have a significant influence on the values obtained from the shell model reduction method. Other theoretical arguments, presented later in the present paper, suggest that the reduction method cannot in all cases yield a precise agreement with the experimental mass data, but nevertheless it should be applicable to the $N = 82$ and $N = 126$ nuclides of interest in the comparisons of Zhang *et al.* [1]. Quite clearly, there is an obvious need for an experimental clarification of the mass data in close vicinity to ^{132}Sn for the dual purpose of understanding the inconsistency between experimental data and a seemingly well-founded empirical systematics, and to establish a very firm basis for tests of the increasingly more accurate full scale shell model calculations in this neutron rich region.

We have therefore undertaken a major reinvestigation of the total β-decay energies (Q_β values) in the isobaric chains involving ^{132}Sn and the nearest $N = 82$ isotones. The atomic masses of far-from-stability nuclides are obtained by adding the Q_β energies to the known mass values of nuclides closer to the stability line. This standard procedure was used in our previous study [2] of the mass data near ^{132}Sn. We note that the values derived that way depend critically on the accuracy of the accepted mass data on nuclei closer to the stability line, as well as on the correctness of existing decay scheme information. A substantial part of the present work was devoted to $\gamma\gamma$-coincidence measurements in order to verify some of the decay schemes of importance to the Q_β determinations. The investigation has covered several nuclides placed relatively close to the stability line, which have been studied earlier but perhaps with insufficient precision for the requirements of the present work. In particular, some of these decays were studied only on a single occasion in a relatively distant past when the experimental techniques or equipment were not sufficiently refined. The $\gamma\gamma$-coincidence experiment also provided a means for a critical examination of the possible γ-ray impurities in the spectra projected from gates selected in the Q_β measurements. That proved to be of vital importance in one of the cases studied. Another important factor in the current investigation has been the use of a high resolution Si(Li) detector for very accurate measurements of low-energy β transitions.

The method employed for the Q_β determination was $\beta\gamma$-coincidence spectroscopy, where the end-point energies of selected β transitions were measured using solid state spectrometers. The key ingredient in the data analysis is a transformation of the observed pulse-height distribution to the β energy spectrum. A precise knowledge of the spectrometer response to monoenergetic electrons of different energies is absolutely necessary in order to provide highly accurate end-point energies. Two detectors have been used. The response of the HPGe spectrometer employed in most of our far-from-stability end-point measurements at ^{132}Sn [2] was carefully determined with the BILL electron spectrometer [4] (previously in operation at ILL in Grenoble) which provided monoenergetic electrons in the energy range 1–8 MeV. The HPGe spectrometer is particularly suitable in the case of relatively high β energies, well exceeding 1 MeV. The measurements of β transitions having energies below about 2 MeV (which included a few cases with a high total Q_β decay energy, but having intense low-energy β branches) were all performed using a high resolution Si(Li) diode. The response of this spectrometer was determined using conversion electrons from thin sources of ^{137}Cs and ^{207}Bi. Since the thickness of the Si(Li) diode was 2 mm, the upper range of β end-point energies measured with this spectrometer was restricted to about 2 MeV. As in the previous Q_β determinations [2], the spectrometers were operated at modest counting rates to reduce pulse pile-up effects, and the electronic system included active pile-up rejection circuitry.

The current reinvestigation of the Q_β energies was performed for the decays of Sn, Sb, Te, I, and Xe in the mass range $A = 131-135$. All nuclides were obtained as mass separated fission products at the OSIRIS ISOL facility [5] at Studsvik, Sweden. The low-energy radioactive ion beam was collected on a thin movable tape which was used, when necessary, to remove the long-lived daughter products. The Si(Li) detector was placed inside the vacuum chamber facing the beam deposition spot at an angle of about 45° to the surface of the tape. The HPGe β spectrometer, on the other hand, was separated from the vacuum by the foils of 0.08 mm Al and 0.25 mm Be. The energy loss for electrons crossing these foils has been measured as a function of the electron energy with an uncertainty of less than about 5 to 10 keV, depending on the electron energy. This uncertainty is in fact one of the main contributions to the total uncertainty of the β end-point energies for the electron energies up to about 4–6 MeV. At higher energies, the total uncertainty is larger due to an incomplete knowledge of the response function, and becomes about 30 keV at an electron energy of 10 MeV. The HPGe detector was energy calibrated by γ rays from standard sources, including the 6.129 MeV line in ^{16}O. The Si(Li) detector was calibrated on-line using known conversion electron lines in the fission product nuclei, and also off-line using a ^{207}Bi source placed at the location of the beam spot. A possible uncertainty from a distortion of the Si(Li) spectra, due to coincidence summing of a β-particle event with events from conversion electrons or x rays, was investigated by a computer simulation. The maximum distortion of the end-point energies was found to be less than 3 keV in all cases of interest here.

The data analysis was performed by gating individual γ rays and projecting (background subtracted) β distributions from the $\beta\gamma$-coincidence matrix. These spectra were subsequently transformed into the β energy spectra by using the empirical response functions, and then converted into Fermi-Kurie distributions for the end-point energy determinations by a least squares fit method (see Fig. 1). Whenever possible, results from several different γ-ray gates have been averaged to yield the final Q_β value.

The analysis of the $\gamma\gamma$-coincidence data served to examine the purity of γ-ray peaks selected as gates for β energy spectra. Severe problems were found in the case of the ^{134}I decay, where the 1136.2 keV γ ray used as an important gate in the previous Q_β analysis [2] was found to be a doublet, while the other gates included

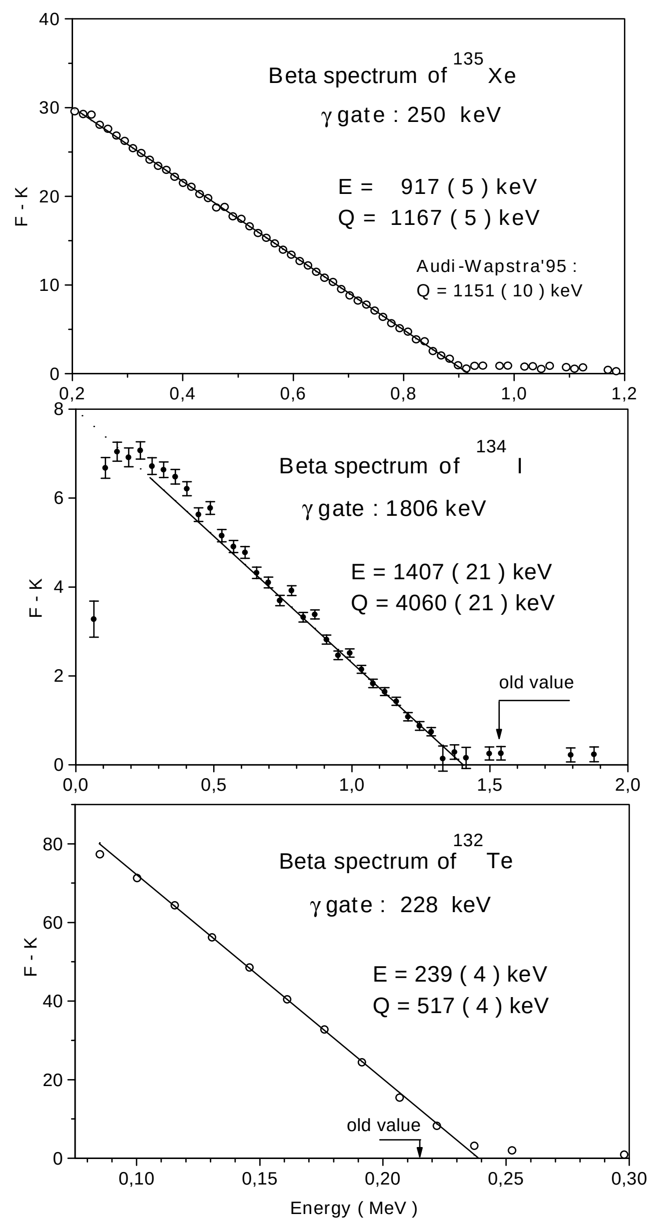

FIG. 1. Fermi-Kurie plot for β energy spectra observed in the Si(Li) detector and gated by selected γ rays of ^{135}Xe, ^{134}I, and ^{132}Te. The solid lines indicate fits to the data. The listed uncertainties for the β end-point energy (E) and the Q_β value (Q) are only statistical ones.

strong true-coincidence summing effects from a cascade of lower lying transitions. The previous data [2], taken with a 5 mm thick Si(Li) detector, could not be recovered for reexamination by a different choice of γ-ray gates. Consequently, new measurements were performed using the 2 mm Si(Li) placed at different distances from the ^{134}I source. The new data provide conclusive evidence that for some gating transitions, and at a solid angle exceeding a few percent, the true-coincidence summing of β particles and Compton events does distort the β energy spectrum by shifting it towards higher energies. The fact that Compton events, rather than photoevents, were involved made the sum spectra practically indistinguishable from those due to pure β particles. The final analysis of the β decay energy of ^{134}I (see Table I and Fig. 2) was performed using the data taken at a solid angle of less than about 3%, where the summing contribution was negligible.

The new β-decay energies and mass excess values in the ^{132}Sn region are summarized in Table II. The reported energies were measured with the Si(Li) diode, which provided results consistent with those from the HPGe spectrometer, but with higher accuracy. We report new results on eight nuclides studied previously by us [2] and on six nuclides placed closer to the stability line. New values are also reported for the decays of ^{131}In, ^{132}In, ^{134}Sb, and ^{134}Sn, not studied here, but subject to significant shifts in the mass excess values. The notable discrepancy with the previous value for ^{134}I can be understood from the preceding discussion. Another significant discrepancy was found for the decay energy of ^{132}Te (see Fig. 1), for which only one measurement was previously made [6].

The precision in the new mass excess values has been strongly improved and is now of the order of $\delta m/m \sim 3 \times 10^{-7}$ even for the most exotic $N = 82$ isotone considered here, ^{131}In. The uncertainties are thus about a factor of 2 lower than those expected [8] from direct mass measurements using a rf spectrometer. Consequently, the results listed in Table II provide benchmark values which can be used for future comparison with the results from direct mass spectrometry on very exotic nuclides. The highly interesting recent developments of the Penning trap technology allow [9] for a precision of $\delta m/m \sim 3 \times 10^{-8}$, corresponding to a few keV near $A = 132$, to be attained for unstable nuclei. Although not applicable to very short-lived ($T_{1/2}$ less than about 10^1 s) nuclides, the method could possibly lead to even further improvement of some of the mass data covered in Table II.

We can now return to the issue of the Q_β puzzle at the $N = 82$ isotones [1] and examine the mass relations expressed as mass "windows" by Zhang *et al*. One should point out that, although these authors employ a diagonal shell model approach to relate the excitation energy of the fully aligned and practically pure $\pi g_{7/2}^3$ $15/2^+$ state to the energies of simpler configurations involving zero, one, or two $\pi g_{7/2}$ particles, they do not perform a shell model calculation of the state energies in ^{134}Te. Instead, they adopt a suitably weighted average of the *experimental* energies of the $(\pi g_{7/2}^2)$ 4^+ and 6^+ states of this nucleus, in place of the corresponding differences of pair matrix elements, which automatically include the bulk of the two-particle correlations. However, such a procedure is not generally applicable since it does not take into account other admixtures. For example, in ^{209}Pb there is a large admixture of the two-particle one-hole $\{3_1^- \otimes j_{15/2}\}_{9/2^+}$ configuration to the $g_{9/2}$ single-particle state caused by the non-spin-flip matrix element of $\langle j_{15/2} \| Y_3 \| g_{9/2} \rangle$.

TABLE I. End-point energies and the total β-decay energy values (Q_β) measured for individual γ-ray-gated β spectra in ^{134}I. The final Q_β value deduced for ^{134}I is given in Table II.

Gating γ-ray (keV)	End-point energy (keV)	Q_β (keV)
1806	1407 (21)	4060 (21)
1040	1407 (25)	4060 (25)
1741	1479 (22)	4067 (22)
974	1454 (14)	4042 (14)
857	1458 (13)	4046 (13)
235	1480 (29)	4068 (29)

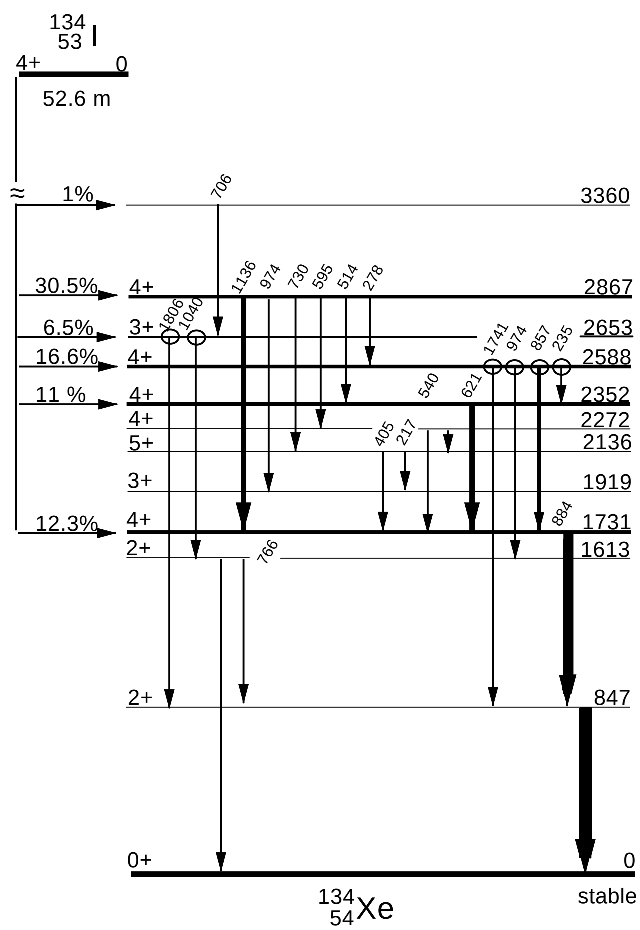

FIG. 2. A partial decay scheme of ^{134}I. Open circles label those γ transitions which were selected as gates for projecting β energy spectra subsequently used to extract the end-point energies.

TABLE II. Experimental Q_β energies and deduced mass excess (ME) values from this work compared to the atomic mass compilation of Ref. [7]. The DME value represents an observed difference between the ME value deduced in this work and the one given in Ref. [7].

Nuclide	Q_β (MeV) this work	ME (MeV) this work	ME (MeV) Ref. [7]	DME (keV)
^{135}I $7/2^+$	2.627 (6)[a]	−83.793 (8)	−83.788 (23)	5
^{135}Xe $3/2^+$	1.167 (5)[a]	−86.420 (6)	−86.436 (10)	−16
^{135}Cs $7/2^+$	...	...	−87.587 (3)	...
^{134}Sn 0^+	7.370 (90)[b]	−66.799 (101)	−66.640 (100)	159
^{134}Sb 0^-	8.390 (45)[b]	−74.169 (46)	−74.010 (50)	159
^{134}Te 0^+	1.513 (7)[a]	−82.559 (11)	−82.400 (30)	159
^{134}I 4^+	4.052 (8)[a]	−84.072 (8)	−83.949 (15)	123
^{134}Xe 0^+	...	...	−88.124 (1)	...
^{133}Sn $(7/2^-)$	7.990 (25)[b]	−70.961 (38)	−70.970 (80)	−9
^{133}Sb $(7/2^+)$	4.002 (7)[c]	−78.951 (28)	−78.960 (80)	−9
^{133}Te $(3/2^+)$	2.942 (24)[c]	−82.953 (27)	−82.960 (80)	−7
^{133}I $7/2^+$	1.757 (4)[a]	−85.895 (12)	−85.878 (26)	17
^{133}Xe $3/2^+$	0.424 (11)[a]	−87.652 (11)	−87.648 (4)	4
^{133}Cs $7/2^+$	...	...	−88.076 (3)	...
^{132}In (7^-)	14.135 (60)[b]	−62.442 (65)	−62.490 (70)	−48
^{132}Sn 0^+	3.115 (10)[c]	−76.577 (24)	−76.621 (26)	−44
^{132}Sb (4^+)	5.491 (20)[c]	−79.692 (22)	−79.724 (23)	−32
^{132}Te 0^+	0.517 (4)[a]	−85.183 (8)	−85.210 (11)	−27
^{132}I 4^+	3.580 (7)[a]	−85.700 (7)	−85.703 (11)	−3
^{132}Xe 0^+	...	...	−89.280 (1)	...
^{131}In $(9/2^+)$	9.174 (22)[b]	−68.149 (37)	−68.220 (80)	−71
^{131}Sn $(3/2^+)$	4.688 (14)[c]	−77.323 (30)	−77.390 (70)	−67
^{131}Sb $(7/2^+)$	3.200 (26)[c]	−82.011 (26)	−82.020 (70)	−9
^{131}Te $3/2^+$	...	...	−85.211 (2)	...

[a]From this work, measured with the Si(Li) detector.
[b]From Ref. [2], measured with the HPGe detector.
[c]Average of the results from this work [using a Si(Li) detector] and Ref. [2].

The mixed character of this single-particle state leads to four-particle one-hole admixtures in ^{211}Pb. Consequently, if one takes, for example, the 639 keV seniority three $11/2^+$ state in ^{211}Pb, one obtains a disagreement between the mass window W [1] and the spectroscopic prediction of the order of 100 keV. However, the possible $\{3_1^- \otimes h_{11/2}\}_{7/2^+}$ and $\{3_1^- \otimes i_{13/2}\}_{9/2^-}$ admixtures to the $g_{7/2}$ and $h_{9/2}$ states in ^{133}Sb and ^{209}Bi, respectively, are small due to the spin-flip character of corresponding matrix elements. In these cases, one expects good agreement between the values of the mass windows and the decomposition technique.

Using the new mass excesses of ^{132}Sn, ^{133}Sb, ^{134}Te, and ^{135}I, we deduce the empirical mass window for these $N = 82$ isotones as $W = -3608(94)$ keV, which is in agreement within the 1σ limit with $W = -3570$ keV obtained [1] from the decomposition of the energy of the $15/2^+$ state of ^{135}I. Such close agreement can be taken as support both for the authors' [1] interpretation of their experimental data and for the applicability of the shell model reduction technique (subject to the provisions given above). In particular, one should not expect excellent agreement if the reduction technique is applied to ^{135}Sn located along the $Z = 50$ isotopes.

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