Masses of ^{103,104,105}In and ^{72,73}Br

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A mass-spectrometric technique, employing the Chalk River On-Line Isotope Separator at the Chalk River TASCC Facility, has been used to determine the mass ratios between some neutron-deficient isotopes. Two mass doublets ^{103}In-^{104}In and ^{104}In-^{105}In and the mass triplet ^{103}In-^{104}In-^{105}In have been studied. We have also measured the mass ratio between ^{72}Br and ^{73}Br to a higher precision than that reported in our earlier work. When combined with the known masses of ^{103}In, ^{105}In, and ^{73}Br our data yield the atomic mass of ^{104}In and improve our earlier value for ^{72}Br. The measured mass ratios are in good agreement with systematics. Our results for the ^{104}In and the ^{72}Br masses are 103 918 367 (126) and 71 936 430(294) μu, respectively.

I. INTRODUCTION

The mass of a nuclide is one of its most fundamental properties. A systematic knowledge of atomic masses provides an abundance of nuclear-structure information. Various techniques have been used to measure atomic masses. One of the most reliable and precise methods has been direct mass measurements made with high-resolution, double-focusing mass spectrometers. Unfortunately this technique is only readily applicable to naturally occurring nuclides. The masses of radioactive species have traditionally been determined through reaction Q-value measurements or through measurements of the energy released in α and β decays. For nuclei far from stability, mass information has been obtained predominantly with decay-energy measurement techniques, which require detailed knowledge of the decay schemes of the nuclides investigated. However, recent technical advances have opened up the possibility of routine direct mass-spectrometric determination of the masses of such exotic atoms. We describe here the results obtained with one such method, based on the high-resolution Chalk River On-Line Isotope Separator.

Considerable interest has been shown recently in the masses of nuclides near the doubly magic ($N=Z=50$), ^{100}Sn. Information about the behavior of the nuclear mass surface in this region makes it possible to test critically the theories of nuclear masses and to delineate nuclear structure in the region of a double shell closure. In one case of particular interest, Wouters *et al.* [1] have reported large discrepancies between their results for ^{103,104,105}In and values arrived at from systematics and mass formulas [2]. Subsequently, the β spectra from all three isotopes were remeasured [3,4]. The results of these recent measurements combined with level-scheme information yield Q_β values for ^{103}In and ^{105}In, and, in a less direct manner, one for ^{104}In as well. They disagree with the earlier measurement and follow the expected systematic behavior.

To settle the disagreement between these two sets of experiments, we have directly measured the atomic mass ratios for ^{103,104,105}In, using mass-spectrometric techniques, with the Chalk River On-Line Isotope Separator. Our results, which are not dependent on the details of available level schemes for the decays of these isotopes, are in good agreement with both systematics and mass formulas, and are consistent with the recent results of Bom *et al.* [4]. A preliminary report on our work has been presented earlier [5]. The values presented here are consistent with, but supersede those given in that report.

One of the first measurements carried out by us with the isotope separator [6] was the determination of the mass ratio ^{72}Br/^{73}Br. Because we have made changes to our experimental apparatus and to our measurement procedure we have remeasured this ratio. The increased precision of the new result clearly demonstrates the effects of the improvements to our technique. Additionally we have also attempted the direct measurement of the excitation energy of the 101-keV isomeric state in ^{72}Br.

II. EXPERIMENTAL PROCEDURE

A detailed description of our mass-measurement apparatus has already been published [6]. In the present work, the indium radioactivities of interest were produced by the bombardment of an enriched ^{92}Mo target, placed near the entrance window of a FEBIAD ion source, with a 100-MeV ^{16}O beam. The bromine activities were produced with a 63-MeV ^{16}O beam and a natural nickel target. The reaction products recoil out of the target, through a few heat shields and are implanted into a low-density graphite catcher inside the ion source. They subsequently diffuse out of the catcher and are ionized in the plasma region of the source. An ion beam is extracted and formed by the extraction potential (nominally 40 kV) applied to the source. The ion beam is then

analyzed by the separator magnet and brought to a focus at an image slit. With an ion-source exit orifice of 0.5 mm and an image-slit width of 0.25 mm, a resolving power ($M/\delta m$, where δm is the FWHM of the mass peak corresponding to the mass M) of 7000 was achieved for the measurements described here. Ions that pass through this slit are guided through an electrostatic beam-transport line into a low-background area where they are implanted into an aluminized Mylar type of a tape-transport station. At the end of a preset interval, the activity collected on the tape is moved in front of a HPGe detector where the γ-ray spectrum of the sample is recorded.

To measure the mass ratio between two species, the ion beams corresponding to the selected masses are swept across the image slit by changes of the extraction potential in a series of preprogrammed steps. At each of these steps a γ-ray spectrum is taken for the ions transmitted to the tape station and the corresponding extraction voltages are recorded. In this manner, each of up to three mass peaks may be scanned over. The duration of each step is dictated by the half-lives of the activities involved.

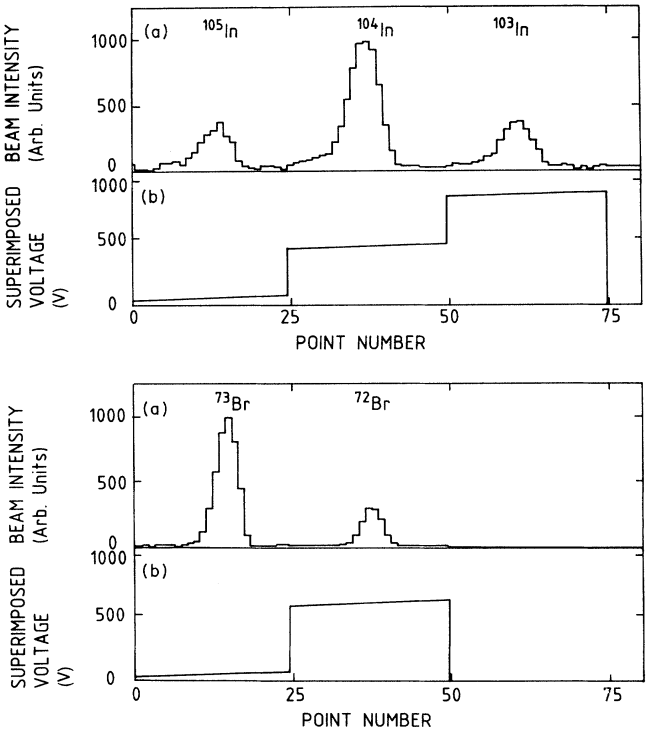

FIG. 1. Mass spectral peaks obtained during a single mass scan. The upper part of the figure shows data obtained in one mass scan of the In triplet measurement. The part labeled *A* shows the intensity of γ rays specific to ^{105}In, ^{104}In, and ^{103}In as a function of point number. Part *B* shows the extraction voltage, minus a common base value, for these data points. For clarity, the point numbers are shown in order of increasing voltage and not in the order of their accumulation. The lower part of the figure shows the analogous data obtained for a bromine mass scan.

The raw data consists of numerous γ-ray spectra and their corresponding values of the extraction potential. We generated a mass spectrum by plotting the area of selected γ-ray peaks against the applied voltages. Examples of two such spectra are shown in Fig. 1. Because of the γ-ray tagging, it is possible to generate mass spectra that are free of contamination from unwanted isobars or isomers even when the resolving power of the separator is insufficient to resolve the untagged mass-spectral peaks.

The centroid voltages of the generated mass-spectral peaks are used to calculate the mass ratio between the two species through Bleakney's theorem, which states that two ions of masses M_1 and M_2 will follow identical trajectories through the separator if all magnetic fields are kept constant and all electric potentials applied to the instrument are changed so that

$$M_1 V_1 = M_2 V_2 \ . \tag{1}$$

Therefore, from a knowledge of the centroidal voltages, V_1 and V_2, and one of the masses, it is possible to calculate the unknown mass.

Three major changes have taken place since our initial measurements with this system [6]: (1) The separator has been moved to a new location and the temperature stability of the cooling-water system has been improved. The resulting improvement in the stability of our main magnetic field has greatly reduced the contributions from this source to the uncertainties associated with our measurements. (2) The data acquisition and computer control of the apparatus are now carried out through a CAMAC-based system. This has greatly simplified the overview and control of the experiment and improved the reliability of our system. (3) The order in which the data points are accumulated during a mass scan has been changed. This was done to reduce the sensitivity of the technique to possible instrumental drifts.

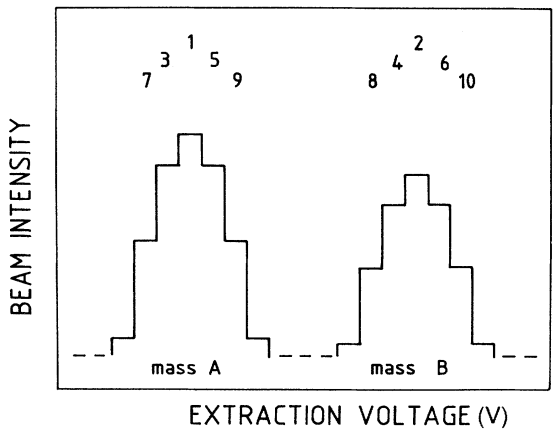

FIG. 2. Schematic representation of the order of data collection in a mass scan that covers two ion beams, one with mass *A* and the other with mass *B*. The numbers above the peaks indicate the order in which data points are taken. The first voltage point selected corresponds to the maximum transmission of the ion beam corresponding to mass *A* and the second to the maximum for ion beam *B*.

Previously, the mass scans were conducted in a sequential manner; i.e., the data points were accumulated in order of increasing (or decreasing) voltage. Thus, first one mass and then another would be scanned across the slit and detected. A slow drift in any one of several crucial components of our system would therefore have had a sizable impact because of the relatively long time spent completing the scan over one mass before data collection began on the next. In order to average out the effect of such drifts, this sequential method has been replaced by interleaved scans. Now data collection alternates between points corresponding to each of the two (or three) different masses being scanned. In addition to interleaving the scans over the various peaks, the sequence also alternates between either side of each individual peak. The procedure is illustrated in Fig. 2. All peaks are therefore observed quasisimultaneously, greatly reducing the likelihood of systematic errors arising from slow drifts.

The reduction in possible systematic errors with the interleaved scanning technique has been investigated for a number of scenarios. As an illustration of a particular case, Fig. 3 shows the shift in the centroid of a peak that would occur if the intensity of the radioactive beam, produced by the separator, declined linearly with time to 50% of its initial value over the course of one scan. A decline in the accelerator beam current or a malfunctioning separator ion source could give rise to such a situation. The figure shows that a drift in production rate of this magnitude would cause (in the worst case) a shift in the absolute position of the centroid of 4 ppm for a sequential scan, but that the worst error for an interleaved scan would be 10 times smaller. Furthermore, in an interleaved scan of two mass peaks, both centroids would be affected in nearly the same manner by the instrumental drifts. The error in their difference, which is the important quantity in a mass determination, is thus further reduced. This would contribute, at worst, a bias of a few keV in the measured mass of an $A=100$ ion beam. The effects of other drifts and instabilities are similarly reduced to a level far smaller than the accuracy with which our centroids are located for a single scan.

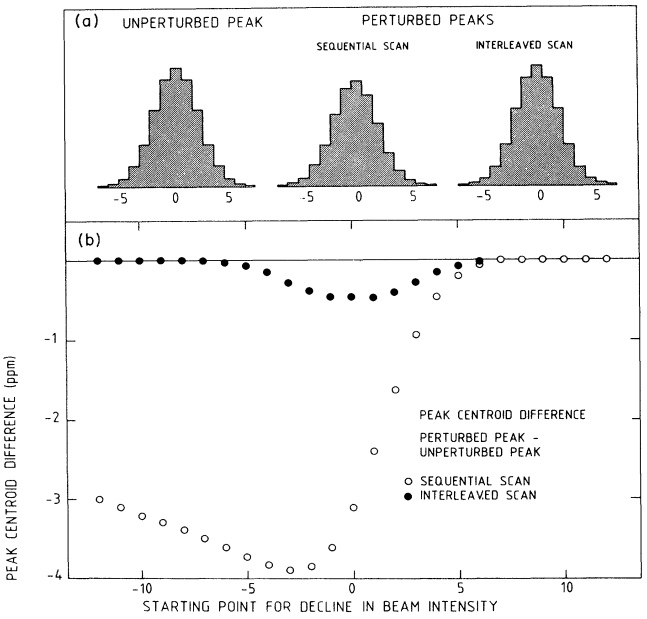

FIG. 3. Simulation of the errors in peak centroid determination resulting from a linear decline with time in the separated radioactive-beam intensity. Typical separator operating conditions have been assumed. The mass peak is centered at an extraction potential of 40 000 V, the resolving power is 6000, and the mass scan is 25 points (30 V) wide. (a) shows our reference peak, as well as two perturbed peaks resulting from a beam-intensity decline commencing in point -2 and continuing to the end of the scan where the intensity has dropped to 50% of its initial value. The effect of this intensity change is clearly visible in the mass peak obtained with a sequential scan but is hardly noticeable for that obtained with an interleaved scan. (b) shows the shift in the centroid, declined as the difference in the centroids of the reference and perturbed peaks divided by the reference peaks's centroid value. The shift is shown as a function of the point number where the intensity decline begins.

III. RESULTS AND ANALYSIS

The γ-ray spectra, recorded during the scans over the mass numbers 103, 104, 105, 72, and 73, are shown in Fig. 4. Only the most intense γ-ray peaks associated with each nuclide [7–11] were used to generate the mass spectra and these are identified in Fig. 4.

In the experiment on indium activities, it is possible in principle that isobaric tin activities could be produced and transmitted through the apparatus, subsequently decaying to their indium daughters. If some of those daughter nuclides then decayed while they were in front of the HPGe detector, these events would be mistakenly labeled as part of the indium peak and the recorded mass spectrum would thus require a correction for this effect. Accordingly, we have carefully examined the mass-specific γ-ray spectra (Fig. 4) for signatures of tin activities and concluded that any biases from such a contamination are negligible.

In the case of the spectra at $A=104$ from the indium experiment no peak was visible at an energy [12] of 132.7 keV which would correspond to the ^{104}Sn activity. Our estimates for the possible contribution of this activity to our mass spectra are based on limits arrived at from the background levels present at this energy in our γ-ray spectra. From such an analysis, we conclude that the production rate of ^{104}Sn was less than 0.5% of the observed rate for ^{104}In. Such a contribution would bias the measured ^{104}In mass upward by, at worst, 25 μu. Since the timing cycle used for the mass measurements did not favor the observation of the relatively short-lived tin activity ($t_{1/2}=21$ s), a separate measurement of the relative production rates was carried out with a timing cycle optimized for the ^{104}Sn activity. Again, no evidence of the 132.7-keV γ ray was seen and the results of this independent measurement place an upper limit of 1.25 μu on the required correction, which is negligible when compared to our error limits.

A similar analysis for the $A=105$ mass spectrum,

based on the 309.5-keV γ ray assigned to ^{105}Sn [13], provides an upper limit on the production rate for this nuclide of 0.13% relative to ^{105}In. This contribution would shift the measured mass of ^{104}In downward by less than 6 μu. No information on γ rays specifically assigned to ^{103}Sn is currently available. However, on the basis of our experience at $A=104$ and 105 we believe that the possibility of significant contributions to the mass spectra at $A=103$ from ^{103}Sn is remote as well.

The possibility of contamination of the bromine mass spectra as a result of the presence of isobaric krypton activities was also investigated. No photopeaks were found at the energies of 178 keV (^{73}Kr) [11] and 415 keV (^{72}Kr) [10], indicating that no significant contamination was present. Estimates were made, based on the background level, of an upper limit for any corrections to the measured mass ratios from this possible source and were found to be much smaller than the uncertainty for our final result.

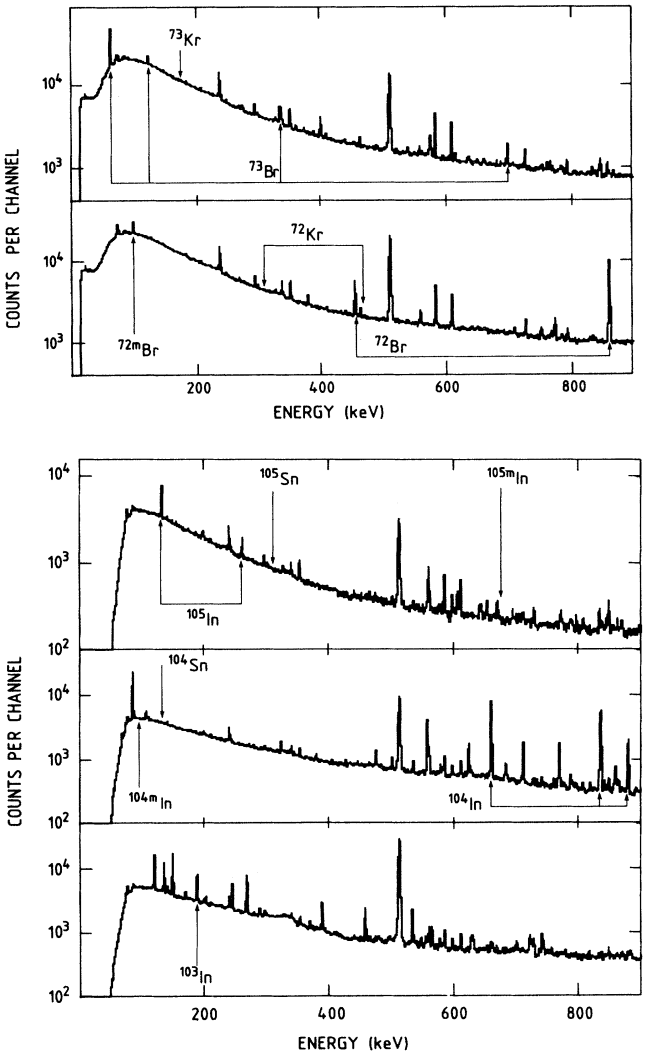

FIG. 4. Mass-specific γ-ray spectra recorded during the indium and bromine measurements. For the bromine case the spectra shown are the result of summing the individual spectra obtained over all the mass scans of these isotopes. For the indium case, the spectra represents sums over spectra obtained during the two triplet scans only. The peaks marked for ^{73}Br, ^{72}Br, ^{72}Br^m, ^{105}In, ^{104}In, and ^{103}In were used in determining the mass spectra. All of the other labeled arrows indicate peaks that were sought but not observed (with the exception of ^{105}In^m); see text for discussion.

A similar bias in our measured masses could also arise from the presence of long-lived isomeric states. Such isomers are known to exist for ^{104}In, ^{105}In, and ^{72}Br. In the case of ^{104}In, the 93.5-keV photopeak associated with the isomer [14] was not observed. We estimate an upper limit for the production rate of ^{104}In^m($t_{1/2}=16$ s, $E_x=93.5$ keV) to be 1% of the rate for the ground state. The resultant correction to the measured mass of ^{104}In would be less than 1 μu, insignificant in comparison with the uncertainties quoted. At $A=105$ the production of ^{105}In^m activity ($t_{1/2}=55$ s, $E_x=674.1$ keV) was observed and its rate was measured to be 0.4% relative to that of the ground state. The presence of this activity causes an increase in our observed ^{104}In mass by 2.8 μu. We have corrected the measured mass of ^{105}In downward by this amount.

A long-lived isomer also exists in the case of ^{72}Br ($t_{1/2}=10$ s, $E_x=101.2$ keV). In this case, the production rate for the isomer relative to that of the ground state was determined to be 33% on the basis of the areas of the 101-keV photopeak, associated uniquely with the isomer, and the 454.7-keV peak, associated with both the isomer and ground-state decays [10]. Therefore a correction of -48 ± 10 μu was applied to the measured mass of ^{72}Br even though this amount is well within the uncertainty of our measurement.

A total of 13 scans over the ^{104}In-^{105}In mass doublet, 4 over the ^{103}In-^{104}In mass doublet, and 16 over the ^{72}Br-^{73}Br mass doublet were completed. Each of these provides an independent value for the corresponding mass ratio. In addition to these mass-doublet scans, two scans were conducted over the mass-triplet ^{103}In-^{104}In-^{105}In. The results produced by the triplet scans were consistent with those produced by the doublet scans and were used to provide two additional values for the ^{104}In/^{105}In and ^{103}In/^{104}In mass ratios. The final results, quoted in Table I, represent the weighted average of all available scans for a particular mass ratio. Because of the difficulties in establishing a representative value for our possible systematic errors—a common problem in mass-

TABLE I. Results of the mass measurements. The uncertainties are shown in parentheses. All uncertainties have been inflated by the reduced χ^2.

Mass ratio	Number of scans	Result	Reduced χ^2
^{103}In/^{104}In	6	0.990 389 00(222)	1.75
^{104}In/^{105}In	15	0.990 502 93(139)	2.36
^{72}Br/^{73}Br	16	0.986 353 12(227)[a]	2.74

[a]See text for a recommended value for this quantity.

spectrometric measurements—only statistical errors were used in the χ^2 evaluation. Consequently, the reduced χ^2 is greater than 1, indicating the presence of systematic errors at a moderate level. In previous mass-ratio determinations on nuclides with known masses we have demonstrated that there is no systematic bias in our procedure [6]. Accordingly, we have taken account of systematic errors by multiplying our uncertainties by the square root of the reduced χ^2. No other corrections have been applied.

Isobaric activities with $Z < 49$ were also produced during the scans over the indium activities. In particular, the production rates of ^{103,104}Ag and ^{103,104}Cd were found to be high enough to allow the determination of mass ratios involving these activities with a precision of approximately 150 keV. We have observed that mass ratios involving different elements (e.g., Cd/In or Ag/In) systematically differ from accepted values whereas those involving different isotopes of the same element agree with accepted values. A difference between the energies of the extracted Cd and Ag ion beams and the In ion beam of about 0.15 eV would account for the observed effect. For this reason we have restricted our quoted results so far to doublets or triplets involving only a single element. We are investigating the nature of these offsets and will present a more detailed report elsewhere [15].

IV. CONCLUSION

The mass ratios summarized in Table I have been combined with the auxiliary data shown in Table II to yield values for the atomic masses of ^{104}In and ^{72}Br. The atomic mass of ^{104}In, calculated as the weighted average of the results from the 6 independent values for the ^{103}In/^{104}In mass ratio and the 15 independent values for the ^{104}In/^{105}In mass ratio, is 103 918 369(126) μu. This value is in excellent agreement with the value of 103 918 328(215) μu, which is based only on systematics [16]. Our result may also be combined with the known mass of ^{104}Cd to yield a Q_{ec} value for ^{104}In of 7938(140) keV. This value disagrees substantially with the earlier result by Wouters *et al.* [1] of 7260(250) keV but is in good agreement with the value of 7800(250) keV recently determined, via an indirect route, by Bom *et al.* [3]. Bom *et al.* have already pointed out that their β end-point measurement agrees with Wouters *et al.* and that the discrepancy arises because Wouters used an erroneous decay scheme for ^{104}In.

Our measured mass ratios for ^{103}In/^{104}In and ^{104}In/^{105}In can also be used to deduce a mass ratio for ^{103}In/^{105}In of 0.980 983 21(260). This result when combined with the mass [16] of ^{105}In yields a value of 102 919 521(273) μu for the mass of ^{103}In. The precision of this result is primarily limited by the uncertainty in the measured mass ratio for ^{103}In/^{104}In which is based on only six scans and thus more susceptible to systematic errors. It is instructive to compare our deduced mass for ^{103}In with the value given by Wapstra *et al.* [16] of 102 919 906.9(27.2) μu. In this way, our result can be used to check the consistency of the chain of Q_β and reaction Q-value measurements that determine the relative masses of ^{103,105}In as given by Ref. [16]. The two values are in general agreement with each other (to within 1.4σ) indicating that no serious inconsistencies are present.

A plot of S_{2n} values for nuclei in this region is shown in Fig. 5. The curves based on the results of Wouters *et al.* [1] show a pronounced downward break at $N = 58$. When these results were published they represented the nuclides with known mass closest to the doubly magic nuclide, ^{100}Sn, and their odd behavior stimulated further research into the atomic masses in this region. With the inclusion of the new values from this work and the work of Bom *et al.* [3,4] no evidence of any unusual behavior in the systematics of atomic masses for these nuclides is observed.

Our new value for the ^{72}Br/^{73}Br mass ratio, 0.986 353 12(227), is seen to be in excellent agreement with our earlier result [6] of 0.986 352 43(453) and represents a substantial improvement in its precision. We recommend that the value of 0.986 352 98(202), the weighted average of our two results, be adopted for general use. This result may be combined with the known mass [15] of ^{73}Br to yield a mass of 71 936 429(284) μu. It should be noted that the uncertainty in the mass of ^{73}Br, 243 μu, is the dominant contribution to the final uncertainty in this case. Our value for this mass is in good

TABLE II. Auxiliary data.

Value	Reference
^{103}In 102 919 906.9(27.2) μu	[16]
^{105}In 104 914 661.3(17.5) μu	[16]
^{104}Cd 103 909 847.4(10.2) μu	[16]
^{73}Br 72 931 720(243) μu	[16]
1 u 931 494.32(28) keV	[17]

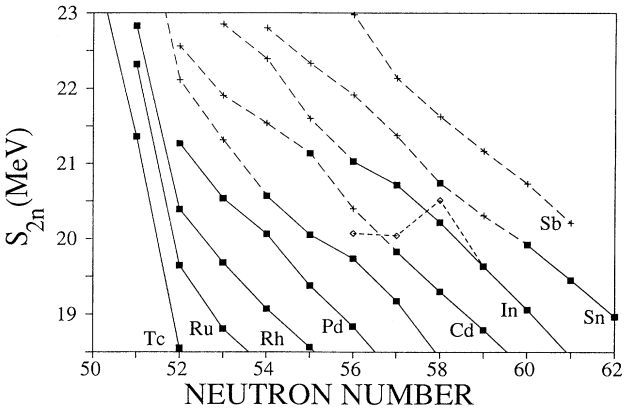

FIG. 5. S_{2n} values in the region $51 < N < 62$ and $43 < Z < 51$ plotted as a function of neutron number. Solid lines connect the results from experimental data (represented by squares) while the dashed line shows those arrived at from systematics (crosses) [16]. The new mass for ^{104}In determined in this work, together with the recent results from Bom *et al.* [3,4] have been used. The results of Wouters *et al.* [1] are also shown (diamonds connected by short dashes). Error bars have been omitted for clarity.

agreement with the value of 71 936 630(215) μu predicted by systematics [16].

The mass ratio for ^{72}Br/^{73}Br was determined despite heavy (33%) contamination from an isomeric state which illustrates how well this technique discriminates against contaminant activities. The analysis that corrects the mass difference for the effects of the isomeric state also yields a rough estimate of the excitation energy of the isomer. It was possible to compare the centroid of the mass peak tagged by the 101-keV photopeak (associated purely with the 10.1-s isomeric state [10] of ^{72}Br) with the centroid of the mass peak associated with both the isomeric and ground-state decays. An excitation energy for the isomeric state of 94(207) keV, which is in agreement with the accepted value of 101.1(0.2) keV, is obtained from the separation of these two mass peaks.

The results described here represent the first to be accomplished after the move of our separator to its new location in the TASCC Facility. Recent improvements of the separator stability and changes in our mass-measurement procedures have led to an increased precision. We have directly determined the mass of ^{104}In to an accuracy of 1.2 ppm and find that our value agrees with and substantially improves upon the recent result from Bom *et al.* [3]. We have also remeasured the mass of ^{72}Br with improved precision.

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