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Q values for neutron-rich rubidium and lanthanum isotopes

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Beta-ray end-point energies for Rb and La fission products were measured on an on-line mass separator using a hyperpure Ge β spectrometer. Coincidence measurements were used to establish feeding relationships and to verify level schemes in daughter nuclides. Q_β values are reported for ^{88,94,96,98}Rb and ^{146,148}La. Our results are compared with those from other experiments and with predictions of mass formulae.

[RADIOACTIVITY ^{88,94,96,98}Rb, ^{146,148}La [from ^{235}U(n,f)]; measured E_β, E_γ, $\beta\gamma$, $\gamma\gamma$ coin; deduced Q_β mass excess. On-line mass separation, HpGe, Ge(Li) detectors.]

I. INTRODUCTION

The determination of accurate atomic masses in neutron-rich nuclei is necessary for further refinement of mass formulae which are used to predict properties of unknown nuclides far from stability. Improvement in the accuracy of these equations is important since they are used in calculations of both fundamental and practical significance. For example, improved accuracy in mass equations would aid in refining reactor decay-heat predictions, guide searches for delayed neutron emitters, and provide a basis for improving calculations of the competition between neutron capture and β decay on rapid time scales. The first two items are important for efficient and safe design of future fission reactors[1]; the third relates to theories of astrophysical significance.[2,3]

Masses of unstable neutron-rich nuclides traditionally have been determined by β-ray end-point measurements. If the excitation energy of the populated level or levels is known, then the mass difference between the two members of the isobaric chain is determined. Normally, plastic scintillators are used as β detectors and the β-ray singles spectrum and/or beta spectra in coincidence with γ deexcitation of daughter nuclei are recorded. Recently, hyperpure Ge(HpGe) detectors have been introduced. Because of their inherent superior energy resolution, ease and accuracy of calibration, linearity of response, and stability, they offer the potential for determining end-point energies with higher precision than is possible with scintillation detectors, thus permitting more precise mass determinations for nuclides which undergo β decay.[4,5]

For stable isotopes high resolution mass spectroscopy has been used to directly measure mass differences. Recently, Epherre *et al.*[6] used an adaptation of this technique to study radioactive nuclides far from stability. Using the on-line mass separator ISOLDE at CERN they made precision direct mass measurements of $^{74-79}$Rb, $^{90-99}$Rb, and numerous Cs isotopes. In some instances their results were found to be at variance with those determined by Q_β measurements.[7–9] In the case of Cs isotopes the discrepancies between the two types of measurements were found to result from a significant error in the value of a reference mass used in calibrating the direct mass experiments.[10] No such systematic error affected the Rb direct mass measurements.

The recent Q_β work on very neutron-rich Rb isotopes comes from three groups. Keyser *et al.*[11,12] determined Q_β values for $^{92-98}$Rb using a plastic scintillation counter-telescope/Ge(Li) spectrometer at the mass separators LOHENGRIN and OSTIS located at the high flux reactor of the Institute Laue-Langevin (ILL). Decker *et al.*[9] also reported Q_β values from OSTIS measured using a HpGe β detector. The results of Peuser *et al.*[8] were obtained using a plastic counter telescope at the Mainz reactor. There are major discrepancies among the various Q_β measurements (>1 MeV in some cases) as well as between some of the Q_β results and the direct mass measurements.

In the neutron-rich La region there exist fewer spectral data and no direct mass measurements. The Q_β results are from two groups working at the Institute Laue-Langevin. Stippler *et al.*[13] reported Q_β values for ^{144,145,147}La obtained from β-ray spectra measured using a plastic scintillation counter-telescope/Ge(Li) spectrometer at the LOHENGRIN separator. In a subsequent paper Keyser *et al.*[14] reported results for ^{146}La. Very recently Decker *et al.*[15] also reported a Q_β value for ^{146}La obtained at OSTIS from a γ-gated β spectrum measured with a HpGe/Ge(Li) β-γ spectrometer.

At TRISTAN we chose to begin our program of Q_β measurements in the Rb region. Since the ^{88}Rb Q_β value is known with extremely high precision and because there exists a large body of data for heavier Rb isotopes this seemed like a good region in which to refine our experimental and analytic techniques. Once this was accomplished we sought to extend our measurements as far from stability as technically feasible, in this instance to ^{98}Rb. Q_β values based on beta-ray end-point measurements require accurate decay schemes. Since a major recent effort at TRISTAN has been the study of decay of very neutron-rich La isotopes, we chose to look next in this region.

II. EXPERIMENTAL METHODS

A. The TRISTAN facility

The TRISTAN on-line mass separator at the High Flux Beam Reactor, Brookhaven National Laboratory, became operational late in 1980. Detailed information about the capabilities of the separator and associated data acquisition/analysis systems can be found in Refs. 16 and 17 and citations therein. Initial operations have used a positive ion surface ionization source[18] which contains ~ 5 g of enriched ^{235}U in a graphite cloth matrix. The source is positioned in a neutron beam flux of $\sim 1.5\times10^{10}$ n/cm^2 s external to the reactor shield. Primary beams of alkali metals (Rb, Cs) and alkaline earths (Sr, Ba) are extracted, mass separated by a 90° magnetic sector, and deposited on a movable tape. A tape transport mechanism permits timed movements of the source deposit relative to detectors positioned at the point of deposit (parent port) or at a secondary station (daughter port). Proper choice of time sequencing permits selective enhancement of various members of an isobaric decay chain. Because of massive shielding of the ion source and primary separator components a low background is maintained at the counting stations.

B. Beta-ray end-point measurements

We have developed a system for Q-value measurements similar in design to that reported by Wünsch *et al.*[4,5] in which a HpGe detector is used to measure β spectra. Our detector, which has 250 mm^2 surface area, 10 mm active thickness, and moderate resolution for γ rays (~ 2.0 keV full width at half maximum at 1332 keV), is mounted in a cryostat equipped with a 12 μm titanium entrance window which serves to protect detector integrity. The detector assembly is integrally mounted into the vacuum system of the TRISTAN moving tape collector so that the source-to-detector distance is 15 mm. Determination of energy loss for β rays in the detector window and dead layer was made using a ^{207}Bi conversion electron source. The γ-ray sensitivity of the β detector is employed as a means for energy calibration using high energy neutron-capture γ rays and the many well known γ rays of ^{90}Rb measured on-line at the separator.[19] To minimize systematic errors in β-ray end-point energies due to accidental summing, pulse pileup rejection circuitry was normally used and counting rates were kept below 3 kHz.

Additional information is provided by measuring γ-ray spectra in coincidence with β rays using a 20% Ge(Li) detector located on the opposite side of the source. β-γ coincidence measurements are necessary when there is more than a single β branch of significant intensity; when Q_β for decay of the daughter nuclide is comparable to or greater than Q_β for the nuclide of interest; or when β decay does not occur between ground states. We record event-mode coincidence data which is sorted off-line into spectra for various β branches. This provides data on alternative paths for checking derived Q_β values. It also provides a consistency check on β feeding determined by γ spectroscopy.

Pileup rejection circuitry and a constant-fraction timing coincidence system are interfaced with a digitally-stabilized data acquisition system based on a PDP-11/20 computer. A fast microprogrammed branch driver controls the flow of data from up to eight separate CAMAC channels to the central processor or directly to external devices. Coincidence data are event-mode recorded (β-γ-t) on magnetic tape. An off-line PDP-11/34 computer is used for tape scanning, plotting, and data analysis. Of particular interest for these studies is an interactive computer code, BDK, for analysis of β-ray spectra, which has been described in detail by Rehfield.[20] In most cases reported here this code was used to linearize data in the high energy region of the β spectrum, thus permitting accurate determination of the end point. The uncertainties assigned to Q values reported here include statistical error calculated for the fit to the experimental β spectrum (see Ref. 20 for a detailed discussion) and systematic error, which is somewhat subjective, and difficult to evaluate. The systematic error reflects our knowledge of experimental conditions and when possible, the results of multiple experiments. Finally, in those instances where the decay scheme based on γ-ray intensity balances suggests the existence of more than a single intense β branch, a computer code MULTBR (Ref. 20) was used to simulate the composite β spectrum based on these branchings. Agreement between experimental and generated β spectra serves as a consistency check on the decay scheme.

III. Rb ISOTOPES: RESULTS AND DISCUSSION

Experimental Q_β values for some neutron-rich Rb isotopes are found in Table I. In addition to the results of the present study we include for comparison values reported by others using HpGe detectors,[9,10,21] and plastic scintillation counter telescopes.[8,12] In each category we show only the most recent results reported by the various experimental groups, assuming that these are meant to supersede earlier results. The mass spectrometer results together with mass excess values derived from Q_β measurements are shown in Table III. Each of the Rb isotopes will be discussed in turn.

A. ^{88}Rb

In Fig. 1 we show experimental data and a Fermi-Kurie fit to the end-point region of the ^{88}Rb β spectrum as measured at TRISTAN using a HpGe detector. The first forbidden unique shape factor for the ^{88}Rb ground state to the ^{88}Sr ground state transition was incorporated into the Fermi-Kurie analysis. We determine an end-point energy of 5313$\pm$5 keV, in excellent agreement with the results of Rehfield *et al.*[21] at McGill and Wollnik

TABLE I. Q_β values for Rb isotopes (keV).

	^{88}Rb	^{94}Rb	^{96}Rb	^{98}Rb
TRISTAN (HpGe)[a]	5313$\pm$ 5	10353$\pm$100	11547$\pm$100	12343$\pm$150
McGill (HpGe)[b]	5310$\pm$10			
OSTIS (HpGe)	5317$\pm$ 3[c]	10304$\pm$ 30[d]	$>$11303$\pm$250[d]	
OSTIS (plastic)[e]		10130$\pm$ 90	11355$\pm$130	12440$\pm$110
		(10400$\pm$ 90)	(11850$\pm$ 90)	(12645$\pm$145)
Mainz (plastic)[f]			10800$\pm$220	11200$\pm$110

[a]This work.
[b]Reference 21.
[c]Reference 10.
[d]Reference 9.
[e]Reference 12, isomeric values shown in parentheses.
[f]Reference 8.

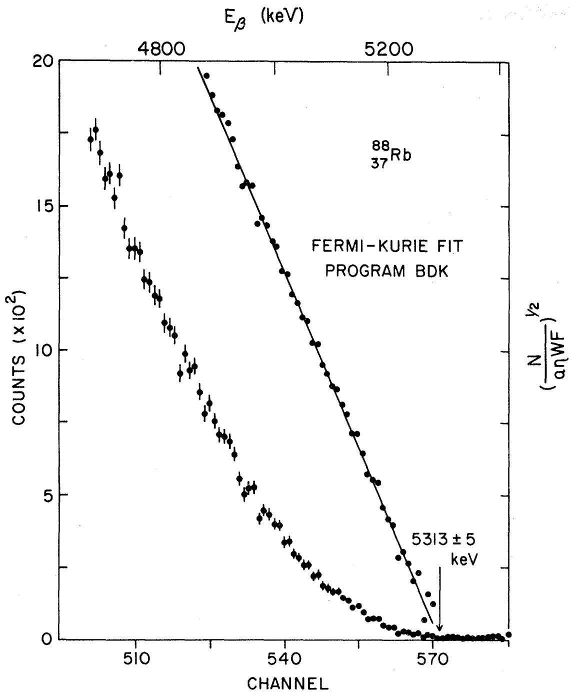

FIG. 1. The β spectrum and Fermi-Kurie fit in the end-point region for ^{88}Rb decay.

et al.[10] at OSTIS (Table I). It should be noted that the very high precision of Ge β-detector measurements for ^{88}Rb is made possible by a number of favorable circumstances including a high fission yield, a convenient half-life, a dominant ground state β branch to stable ^{88}Sr, and the availability of well-known calibration rays from ^{90}Rb which bracket the end-point region.

B. ^{94}Rb

There exist discrepancies among Q_β values reported for decay of ^{94}Rb which may be due to the existence of an as yet uncharacterized isomeric state in ^{94}Rb. In their 1979 paper Epherre *et al.*[6] reported a variation in the mass of ^{94}Rb with temperature of the ionizing surface in their on-line mass spectrometer at ISOLDE. At that time they attributed this behavior to a ^{94}Rb isomer approximately 200 keV less bound than the ground state and with half-life somewhat shorter than 2.7 s. No spectroscopic evidence for this isomer has been reported and, very recently, the ISOLDE group withdrew the isomeric mass assignment. They now believe that the anomaly was due to an unknown contaminant in the ^{94}Rb beam.[22]

Keyser *et al.*[11,12] have reported Q_β values derived from β spectrum end-point measurements obtained with a scintillation counter telescope at the LOHENGRIN and OSTIS separators. Analysis of the γ-gated beta spectra yielded end-point energies from which Q_β values were derived. In their more recent study[12] these Q_β values appeared to fall into two groupings with means of $10\,130\pm90$ and $10\,400\pm90$ keV, which they assigned to ground- and isomeric-state decays, respectively.

Decker *et al.*[9] have also reported a Q_β value for ^{94}Rb measured at OSTIS using a HpGe detector. They derived a Q_β value of $10\,304\pm30$ keV from their end-point energy measurements.

In our experiments at TRISTAN using a HpGe β-ray/Ge(Li) γ-ray coincidence spectrometer, ^{94}Rb activity ($t_{1/2}=2.7$ s) was collected for a period of 5.0 s at the parent port during which data acquisition was operational, followed by a 0.5 s interval in which the ion beam was deflected; counting was inhibited and the tape was moved so that the activity spot was in a shielded location. This duty cycle served to enhance ^{94}Rb activity recording relative to longer-lived daughter products.

Analysis of our β-γ coincidence spectra confirms the result of others[9,23] that the most energetic β branch feeds a level at 2414 keV in ^{94}Sr. We derive an end-point energy of 7939 ± 100 keV which implies $Q_\beta=10\,353\pm100$ keV. Our result for ^{94}Rb is in excellent agreement with Decker *et al.*[9] but falls outside combined errors of the ground state reported by Keyser *et al.*[12] In Table III we list mass excess values as determined by direct mass measurements and as derived from Q_β measurements using the most recent set of evaluated mass data[24] for the daughter isotopes. Here we see that our result is in agreement with the mass spectrometer measurement for ^{94}Rb.

It is important to address the question of isomerism in ^{94}Rb and other Rb isotopes under consideration here. To our knowledge there is no direct evidence for the existence of isomerism in ^{94}Rb and ^{96}Rb. Keyser *et al.*,[12] perhaps because of an earlier suggestion of isomerism in ^{94}Rb by the ISOLDE group[6] (later withdrawn),[22] analyzed their β spectra assuming the presence of two isomers. From γ-gated β spectra they extracted end-point energies from which they derived Q_β values. These Q_β values appeared to fall into two groups which yielded mean values which differed by 270 ± 127 keV. It was assumed that this energy difference represented the excitation energy of ^{94}Rb^m.

We feel that it is important to note the formidable difficulties inherent in this method, which relies on the accurate determination of β spectrum end-point energies measured with a plastic scintillator. It must be remembered that there are numerous factors which can introduce error in the experimental determination of end-point energies from continuous spectra, for example, unknown shape factors, uncertainties in β branching ratios to levels in the daughter nuclide (owing to unobserved γ transitions), pulse pileup, and random coincidence events, to name several. If the uncertainty assigned to the end-point energies of individual spectra is underestimated, then fortuitous groupings of experimental Q_β values might be expected irrespective of the existence of an isomeric decay mode. Given the difficulty in performing end-point measurements and the lack of direct evidence for isomeric states in ^{94}Rb and ^{96}Rb, it is our opinion that the question of isomerization and associated masses remains unresolved for these nuclides.

C. ^{96}Rb

In a similar fashion we have determined Q_β for ^{96}Rb. Because of shorter half-lives, a 1.0 s (collect, count)/0.3 s (tape move) cycle was used to enhance ^{96}Rb ($t_{1/2}=0.20$ s) data collection relative to daughter activities. The end-point energy for the β branch to the 1628 keV level in ^{96}Sr was determined to be 9919 ± 100 keV yielding a Q_β value of $11\,547\pm100$ keV. This is in agreement within stated errors with the most recent ground-state value reported by Keyser *et al.*[12] but is outside combined uncertainties for their isomeric result. The derived mass excess is in agreement with the ISOLDE result (Table III). Peuser *et al.*[8] in work done at Mainz, report $Q_\beta=10\,800\pm220$ keV, substantially below all other results. The reader is referred to the discussion on isomerism in the previous section which is also applicable to ^{96}Rb.

D. ^{98}Rb

In the case of ^{98}Rb decay there is an important disagreement between the decay scheme of Peuser *et al.*[8] and that of Jung.[25] Peuser *et al.* reported β feeding to a level at 2606 keV in ^{98}Sr which deexcited by a 2172-289-145 keV cascade to the ground state. Jung, however, placed the 2172 keV γ ray as deexciting a level at 2316 keV via a 2316-145 keV cascade (Fig. 2). Since the most energetic β branch is seen in coincidence with the 2172 keV γ ray it

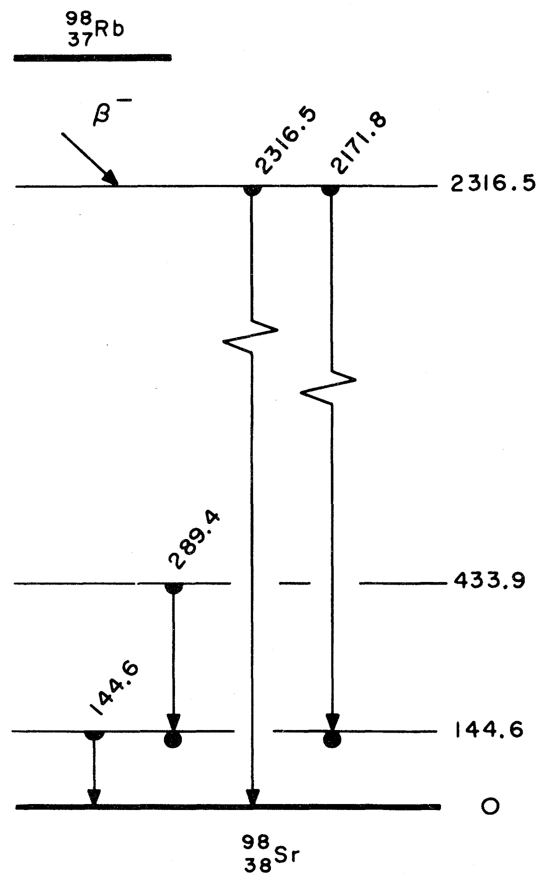

FIG. 2. Partial decay scheme for ^{98}Rb.

was necessary to determine which of the above schemes is correct. γ-γ coincidence spectra recorded simultaneously with our β-γ measurements clearly confirm the scheme reported by Jung.

Schussler *et al.*[26] recently reported evidence for isomerism in ^{98}Rb. They measured γ transitions in ^{98}Sr and found characteristic half-lives of 96 and 114 ms which were explained as arising from β decay of a pair of isomers in ^{98}Rb. The energy, spin, and parity of the excited isomeric state could not be determined, unfortunately.

Figure 3 shows the end-point region of the ^{98}Rb β spectrum. At very high β energies uncertainties in our detector response function prevented accurate Fermi-Kurie analysis using the computer code BDK. Fortunately, a simple semilogarithmic plot served to linearize the data in the end-point region and permit the extraction of the end point. We arrive at an end-point energy of $10\,026\pm150$ keV, yielding $Q_\beta=12\,343\pm150$ keV. Our result is in excellent agreement with the direct mass measurement and the ground state value of Keyser *et al.* Again the Mainz value falls far lower than all others. Because

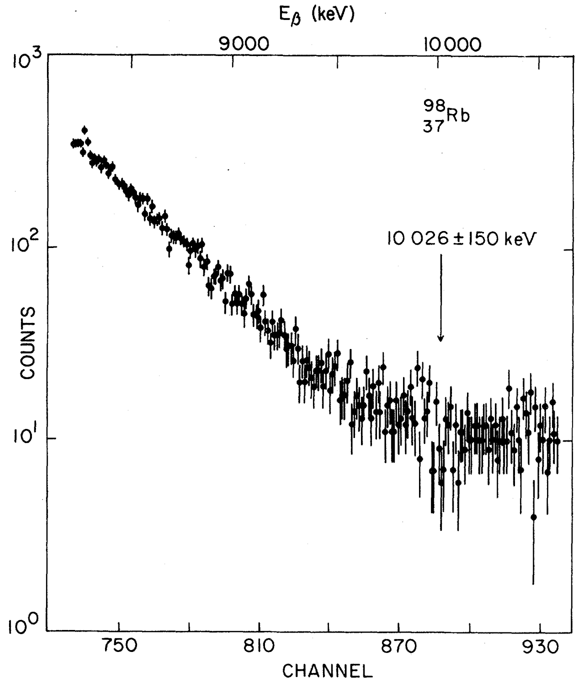

FIG. 3. The β spectrum end-point region for ^{98}Rb decay.

of the uncertainty about the isomer situation in ^{98}Rb we have no way of knowing whether our data correspond to a measurement of the ground state mass, the isomeric state mass, or some weighted mean value.

IV. La ISOTOPES: RESULTS AND DISCUSSION

In Table II we list Q_β values for some neutron-rich La isotopes measured at TRISTAN and elsewhere. La isotopes are obtained at TRISTAN from β decay of their isobaric precursors, Cs and Ba, both of which are directly extracted from the ion source. Because precursors often have higher β end-point energies than the isotope of interest one must be very careful to choose sequencing times for the tape collector so that contributions from these precursors are suppressed. Fortunately the half-lives of the $A=146$ and 148 precursors are shorter than the corresponding La chain member and suitable timing conditions could be chosen. Because of this fortunate circumstance β singles spectra as well as coincidence end-point measurements were useful in establishing Q_β values.

A. ^{146}La

Several groups of experimenters have presented evidence for isomerism in ^{146}La. Skarnemark *et al.*[27] in a chemical separation study report finding two isomers with half-lives of 8.5 s and 4.5 min. Recent studies at on-line mass separators report 6.2 and 10.0 s isomers.[28] At TRISTAN ^{146}La is produced only through beta decay of the ^{146}Ba 0^+ ground state. Because of this "spin filter" we measure the decay of a single isomeric state; in this case the 6.2 s species which is thought to decay to the ground state and the 258 keV 2_1^+ state in ^{146}Ce, and therefore, must have low spin (likely 2^-).[29] In order to enhance ^{146}La relative to other members of the decay chain, ^{146}Cs and ^{146}Ba activities were collected for a 10 s interval, followed by a 4 s delay, and a 10 s counting period. In Figs. 4 and 5 we show experimental data and Fermi-Kurie fits to the end-point regions of the ^{146}La singles and 258 keV $(2_1^+ \rightarrow 0_1^+)$ γ-gated β spectra as measured at TRISTAN. From these we determine end-point energies of 6348 ± 50 keV for the singles spectrum and 6120 ± 30 keV for the coincidence gate, establishing Q_β as 6380 ± 30 keV for the low-spin isomer.

Our results are to be compared with those obtained by two groups at the ILL. Keyser *et al.*[14] working at the LOHENGRIN recoil spectrometer report Q_β values for both the 6.2 and 10.0 s iso-

TABLE II. Q_β values for La isotopes (keV).

	^{145}La	^{146}La[a]	^{147}La	^{148}La
TRISTAN (HpGe)[b]		6380 ± 30		$\geq 5862 \pm 100$
OSTIS (HpGe)[c]		6175 ± 100		
LOHENGRIN	4110 ± 100[d]	6850 ± 100[e]	4750 ± 120[d]	
(plastic, HpGe)		(6660 ± 120)[f]		

[a] $t_{1/2} = 6.2$ s.
[b] This work.
[c] Reference 15.
[d] Reference 13.
[e] Reference 14.
[f] Q_β reported for decay of 10.0 s ^{146}La.

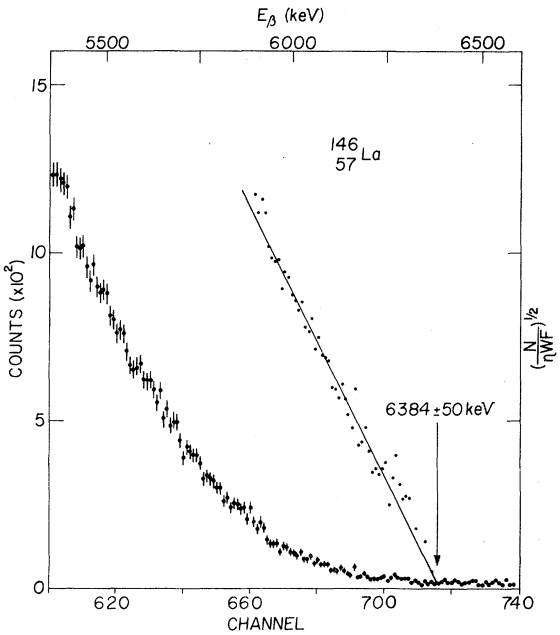

FIG. 4. The β spectrum and Fermi-Kurie fit in the end-point region for decay of 6.2 s ^{146}La.

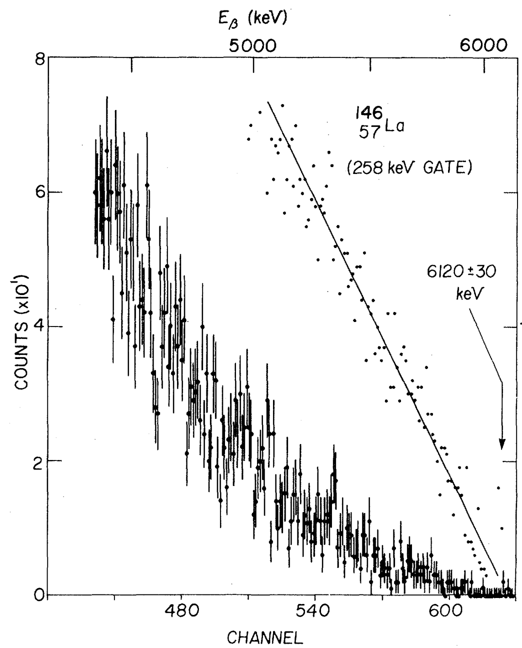

FIG. 5. The β spectrum in coincidence with the 258 keV γ ray ($2_1^+ \rightarrow 0_1^+$) in ^{146}Ce following decay of 6.2 s ^{146}La. The Fermi-Kurie fit to the data is also shown.

mers. Their coincidence data were collected using a plastic/Ge β-γ spectrometer. A singles β spectrum measured using a HpGe detector was also reported for the 6.2 s isomer. The Q_β values deduced, 6850 ± 100 keV for the 6.2 s isomer and 6660 ± 120 keV for the 10.0 s species, led them to conclude that the latter is the ground state of ^{146}La.

More recently Decker *et al.*[15] working at OSTIS reported results of an end point measurement for the 6.2 s isomer made using a HpGe/Ge(Li) β-γ coincidence apparatus. The Q_β value based solely on a 258 keV γ-gated spectrum is 6175 ± 100 keV.

The present situation for ^{146}La is somewhat unsettled. The reported Q_β values for the 6.2 s low-spin isomer differ by as much as 675 keV with each measurement falling outside stated uncertainties of the other members of the set. We quite naturally prefer our measurement, noting the consistency between the Q_β values derived from our singles and 258 keV γ-gated spectra, the high quality of our data as illustrated by the results of our Fermi-Kurie analysis, and the high precision in energy calibration, inherent with HpGe β detectors.

Finally, we feel the assignment of the ^{146}La 10.0 s isomer as the ground state, while reasonable on the basis of the Gallagher-Moskowski rules,[30] is nevertheless premature, because the Q_β values for the isomers reported by Keyser *et al.* have overlapping uncertainties. Furthermore, the apparent dispersion among the Q_β results for the 6.2 s isomer, as determined by three different groups of experimenters, suggests that estimates of uncertainties may often be too small.

B. ^{148}La

Gill *et al.*[31] recently reported results of a study of the ^{148}La decay scheme at TRISTAN. Their intensity balances for individual levels (with appropriate awareness for the concerns of pandemonium[32]) suggest the existence of a significant β branch to the 2_1^+ state in ^{148}Ce at 158 keV. This seems quite reasonable if one considers the orbital occupancy of the neighboring odd-A nuclides. According to Schussler *et al.*[33] the ^{147}La ground state is likely $\frac{5}{2}^+$ [413]p while ^{147}Ba is $\frac{3}{2}^-$ [532]n. The ^{149}Ce ground state is unknown but is probably either $\frac{3}{2}^-$ [532]n or $\frac{3}{2}^+$ [651]n similar to its neighboring higher-A isotones. Coupling these states suggests $(1 \leq I \leq 4)^\pm$ for ^{148}La with the lower spin states favored for the ground state.[30] Recently extensive studies of ^{148}Ba decay have been conducted at TRISTAN. A preliminary analysis of the data suggests that the ^{148}La ground state is either 2^- (most likely) or 1^-.[34] In either case the ^{148}La should β decay to the 2_1^+ level of ^{148}Ce and, also, possibly to the ground state.

The ^{148}La β end-point energy has been deter-

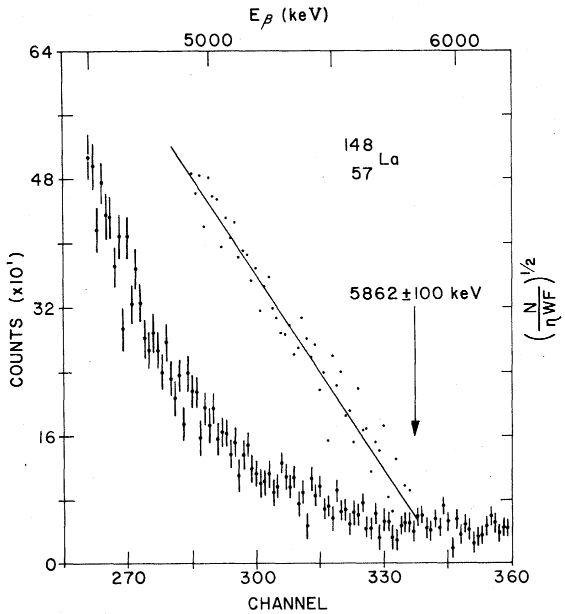

FIG. 6. The β spectrum and Fermi-Kurie fit in the end-point region for decay of ^{148}La.

mined at TRISTAN using our HpGe detector. Mass 148 activity (primarily Ba) was collected for a period of 2.5 s. After a delay of 0.5 s during which the ion beam was deflected to permit decay of ^{148}Ba, data acquisition was initiated and continued for a period of 3.0 s. This sequence enhanced ^{148}La activity relative to its daughter activities, eliminated contributions from ^{148}Cs ($t_{1/2}$=0.11 s), and suppressed contributions from ^{148}Ba ($t_{1/2}$=0.59 s). Singles and β-γ coincidence data were collected although the latter proved too sparse to permit accurate determination of an end-point energy. A singles β spectrum and Fermi-Kurie fit to the data are shown in Fig. 6. We determine an end-point energy of 5862±100 keV. Because of poor statistics the β spectrum in coincidence with the 158 keV ($2_1^+ \rightarrow 0_1^+$) γ ray in ^{148}Ce was inconclusive. If we reverse our analysis and assemble the γ-ray spectrum that appears in coincidence with high-energy β rays, the 158 keV γ ray is the only ^{148}Ce γ ray detected. While this is weak evidence, it nevertheless is consistent with the assumption of β feeding to the 2_1^+ state from a 1^- or 2^- state in ^{148}La. It is not possible to ascertain from existing data whether the end-point energy determined from our singles measurements corresponds to a β branch to the ^{148}Ce ground state or to the 158 keV level. In reporting a Q_β value of $\geq$5862±100 keV (Table II) we allow for the existence of a ^{148}La$\rightarrow$^{148}Ce ground state-to-ground state transition. A mass excess derived from our Q_β determination and the evaluated data for ^{148}Ce (Ref. 24) is found in Table III. No experimental data are available for comparison with our result.

C. Comparisons with mass equations

In Table III we compare experimental mass excess values determined by Q_β and direct mass measurements for Rb and La isotopes. Mass excess values from Q_β measurements were computed using

TABLE III. Mass excess values for Rb and La isotopes (keV)[a].

	TRISTAN[b] (HpGe)	ILL[c] (HpGe)	ILL[d] (plastic)	Mainz[e] (plastic)	ISOLDE[f] (direct)
^{94}Rb	−68 484±100	−68 533± 31	−68 707± 90 (−68 437± 90)		−68 610±140
^{96}Rb	−61 359±113		−61 551±140 (−61 056±104)	−62 106±226	−61 210± 80
^{98}Rb	−54 196±174		−54 099±141 (−53 894±170)	−55 339±141	−54 250±130
^{146}La[g]	−69 393± 87	−69 598±129	−68 923±129 (−69 113±145)[h]		
^{148}La	$\leq$ −64 547±173				

[a]For columns 2−5 computed from Q_β results and data from Ref. 24.
[b]This work.
[c]References 9 and 15.
[d]References 12, 13, and 14.
[e]Reference 8.
[f]Reference 22.
[g]6.2 s ^{146}La.
[h]10.0 s ^{146}La.

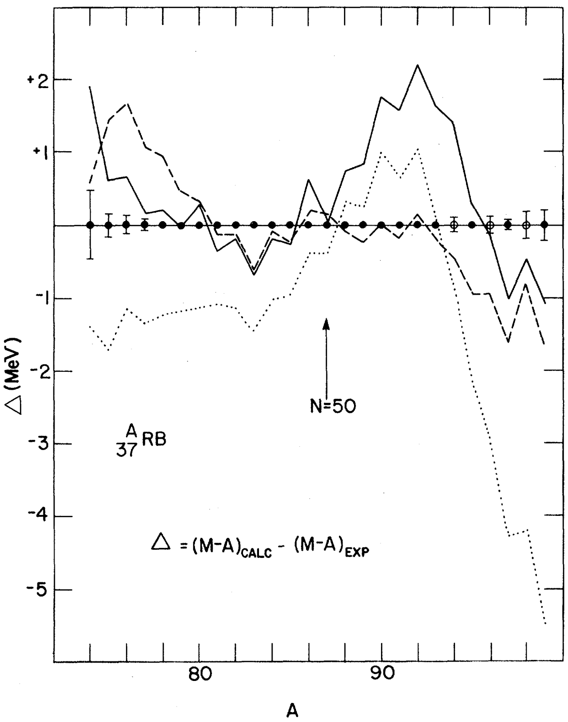

FIG. 7. Comparison of experimental and calculated mass excesses for Rb isotopes. The dotted, dashed, and solid curves are the predictions of Myers (Ref. 35), Liran and Zeldes (Ref. 36), and Möller and Nix (Ref. 37), respectively. Our results are shown as open circles.

the evaluated data set of daughter mass excess recently compiled by Wapstra and Bos.[24] There appears to be substantial agreement among experimenters for mass excess values for very neutron rich Rb isotopes and to a somewhat lesser degree for neutron-rich La nuclides. In general, Q_β measurements strongly support the mass excess results of Epherre *et al.*[6] for the Rb isotopes out through ^{98}Rb, the most neutron-rich species studied to date by β decay. Thus the Rb isotopes provide a well-founded basis for comparison with predictions of mass formulae. In Fig. 7 the differences between calculated and experimental mass excess values for Rb isotopes are plotted. The calculated results are those of Myers,[35] Liran and Zeldes,[36] and Möller and Nix[37] and represent three different types of mass formulae.

The Myers formulation is based on the droplet model corrected for shell effects. The 16 coefficients of the model were obtained by fitting atomic masses and fission barriers of heavy nuclei. Liran and Zeldes have developed a semiempirical shell model formula containing 178 independent parameters which are determined by adjustment to experimental mass data. The formula due to Möller and Nix uses a Yukawa-plus-exponential macroscopic model and a folded-Yukawa single-particle potential to calculate ground state masses. Theirs is a very recent endeavor which includes several previously neglected physical effects and only five adjusted constants.

It appears from Fig. 7 that the best global fit to the Rb masses is obtained from the Liran-Zeldes formula while that of Myers has the largest deviations. Several general trends are discernable: (1) agreement usually worsens at the extremes; (2) all three equations and, indeed, all mass equations we have examined overestimate the stability of very neutron-rich Rb isotopes; (3) the effect of pairing appears to be treated incorrectly resulting in an odd-even staggering; and (4) the Myers formula, and to an even greater degree the Möller-Nix equation, underestimates the binding in the shape transition region beyond the $N=50$ shell closure.

In Fig. 8 we see that there are fewer data for the La isotopes. On the neutron-rich side the most distant isotope for which a mass value exists, ^{148}La, is five nuclides removed from the bottom of the valley of stability as compared to ^{99}Rb which is seven from the minimum. A pattern of deviations which is similar but less drastic than that found for Rb is apparent. Again the best agreement is found for the Liran-Zeldes description while the Möller-Nix calculations have the largest fluctuations with a particularly large excursion at the $N=82$ shell closure. In the transition region beyond the shell the heavier

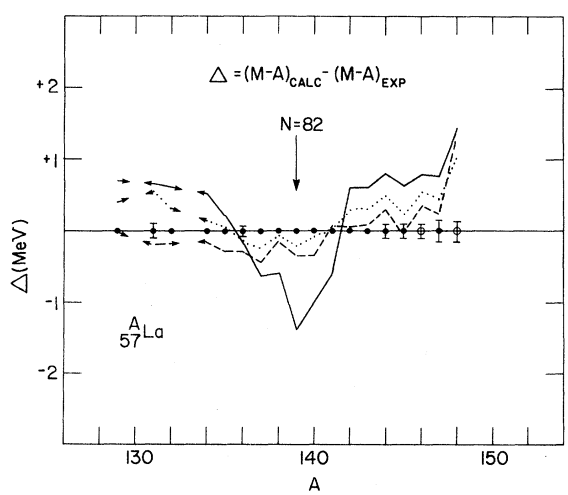

FIG. 8. Comparison of experimental and calculated mass excesses for La isotopes. The theoretical curves are as identified in the caption to Fig. 7. Our results are shown as open circles.

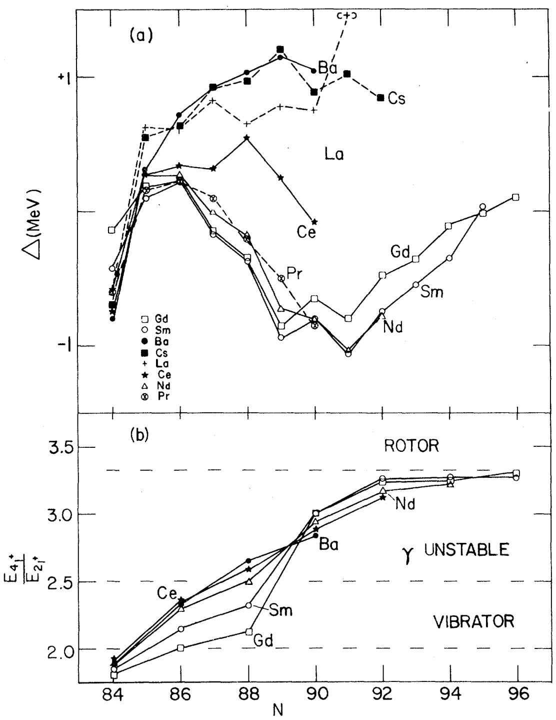

FIG. 9. (a) Comparison of experimental and calculated mass excesses for the shape transition region beyond the $N=82$ shell closure. The experimental data are from Wapstra and Bos (Ref. 24) supplemented by results from this work. The calculated values are from Möller and Nix (Ref. 37). (b) Systematics of $E_{4_1}^{+}/E_{2_1}^{+}$ in the $A\sim150$ region.

isotopes are predicted to be too unstable while those closer to stability have their binding overestimated.

The situation in the transition region with $N>82$ can be examined in greater detail in Fig. 9(a) where we show deviations between experiment and the predictions of Möller and Nix for Ba through Gd. At a glance two features are evident. In the regions $N<86$ and $N>94$ the deviations are systematic and have the same value for a given N within an envelope of about ±250 keV. In the intervening region $86<N<94$, however, there is an apparent separation of the curves with the lower Z elements generally showing positive deviations and the higher Z ones, negative values of Δ.

Möller and Nix[37] noted that systematic discrepancies appeared slightly beyond doubly magic nuclei. They attempted to account for these deviations by introducing an ϵ_3 (octupolar) degree of freedom into their model. The introduction of this asymmetric degree of freedom was successful in removing much of the discrepancy in the region above ^{208}Pb but had no appreciable effect near lower shells.[37]

In a recent Letter Casten *et al.*[38] examined the systematics of the onset of deformation for even-even nuclides in the $A\sim150$ region in light of the recently discovered subshell closure at $Z=64$.[39] They illustrated the changing character of nuclides in this region in terms of a plot of the energy ratio $E_{4_1}^{+}/E_{2_1}^{+}$ as a function of N [Fig. 9(b)] and provided a qualitative microscopic explanation of the origin of deformation for these nuclides in terms of the strong n-p interaction that arises when valence nucleons occupy spin-orbit-partner orbits. Their understanding of the complex behavior in this region has very recently received quantitative support from IBA-2 calculations.[40] Comparing Figs. 9(a) and (b), one is struck by the similarity of the spread and trends toward recombination of the systematic features beginning at $N=86$. It is tempting to speculate whether we are observing another manifestation, this time affecting ground-state masses, of the n-p interaction among valence nucleons and the $Z=64$ subshell. Furthermore, Casten *et al.*[38] put forth an interesting idea which suggests a possible approach for improving mass formulae. They noted that the $E_{4_1}^{+}/E_{2_1}^{+}$ plot displays a tendency for nuclear axial asymmetry in Nd and Sm with $N=88$, 90. Thus better agreement between calculated and experimental masses might be achieved by permitting a softness toward triaxial shapes in those mass formulae, such as the Möller-Nix equation, which involve a minimization of total potential energy with respect to shape coordinates. Indeed, Newton[41] has shown that the preferred shape for some sets of particles in the region between the 28 and 50 closed shells is not necessarily axially symmetric and that the total energy lowering accompanying a change from axial symmetry to a triaxial shape may be as large as $10-15\%$ $\hbar\omega_0$, comparable in magnitude to the dispersion found in Fig. 9(a).

Finally we note the extreme importance of extending our knowledge of masses of other isobaric chains far away from stability, especially on the neutron-rich side. The systematic deviations which exist for very neutron-rich Rb and La nuclides suggest that further extrapolations of mass equations to unknown nuclides may be very unreliable. Calculations which rely on predictions of mass formulae for very neutron-rich nuclides far from stability may also be unreliable. For example, Avignone and Moore[42] have pointed out that extrapolation techniques fail far from stability and that experimental Q_β values are needed in order to improve the calculation of the fission antineutrino spectrum at high energies (>8 MeV).

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