

Isomerism in Ba<sup>129</sup>†

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Isomers of half-lives  $2.13 \pm 0.06$  and  $2.20 \pm 0.15$  h have been identified in Ba<sup>129</sup>. The relative ordering of the isomeric states, which most probably have assignments  $\frac{3}{2}^-$  and  $\frac{3}{2}^+$ , has not been established. Their energy separation is less than 700 keV. No evidence has been found for an isomeric transition. These data are in qualitative accord with the Kumar-Baranger prediction of oblate-equilibrium nuclear shapes for odd-neutron nuclei in this mass region. However, the weight of evidence strongly suggests that the nuclides thus far studied lie in a collective transitional region of predominantly vibrational rather than rotational character.

## INTRODUCTION

CURRENT nuclear collective models share the prediction of well-defined static-aspherical-equilibrium nuclear shapes at neutron and proton numbers well removed from shell closures. Thus far these predictions have received extensive experimental confirmation at  $A \cong 8$ ,  $19 \lesssim A \lesssim 33$ ,  $150 \leq A \leq 190$  and  $A \gtrsim 220$ . In the first two regions the neutron-proton shells are filling in phase, whereas in the last two, an effectively accidental overlap of different shell fillings is responsible for the effect. The models specifically include prediction of at least two further islands of strong deformation in highly neutron-deficient species in the lanthanide and the actinide regions of the nuclide charts (where  $N \approx Z$ ). The nucleon shells in these regions are expected to again fill in phase as in the case of the two lowest mass regions of deformation.

Fragmentary experimental evidence supporting this prediction in the lanthanide region has already been reported.<sup>1-3</sup> For example, it has been demonstrated that the excitation energy of the first 2+ state in even-even barium isotopes rapidly decreases with decreasing neutron number, reaching the Alaga critical value, making the possible onset of permanent deformation at  $A=128$ . Evidence was also adduced for the existence of relatively well-developed rotational bands based on the ground states. However, subsequent measurements in this laboratory<sup>4</sup> indicate considerable ambiguities in the interpretation of some of the data, and in particular, in that leading to the

suggestion of the rotational bands. Measurements via  $(\alpha, 4n\gamma)$  and  $(\alpha, 2n\gamma)$  reactions have been carried out in the xenon isotopes in the range  $124 \leq A \leq 132$ .<sup>3</sup> Again, it has been found that although the excitation energy of corresponding states decreases smoothly with decreasing neutron number, the relative excitations,  $E(J^\pi)$ , in any given nuclide appear more characteristic of vibrational motion than of rotation. While the excitations of the 2+ and 4+ states decrease systematically with the decreasing neutron number, the ratio  $E(4+)/E(2+)$  lies in the range 2 to 2.5 rather than 3.3, hence suggestive of vibrational rather than rotational character. The second 2+ states, where located, have been found in the immediate vicinity of the lowest 4+ state. The conclusion from these data would appear to be that although the nuclides thus far studied are in a transition region, it will be necessary to go to more neutron-deficient species to find strong static deformations.

Recently Kumar and Baranger<sup>5</sup> have utilized a Hartree-Bogolyubov treatment of a pairing-plus-quadrupole nuclear-force model appropriate to the region to examine the equilibrium nuclear shapes. It was found that the odd-neutron nuclei were expected to have oblate equilibrium shapes whereas odd-proton nuclei were predicted to be almost spherical or, if aspherical, slightly prolate.<sup>6</sup>

In view of the intrinsic interest in establishing the existence of static deformations in this region, and if possible, the sign of these deformations, we have carried out a series of measurements utilizing heavy-ion bombardment to create the highly neutron-deficient species.

Reference to the appropriate section of a Nilsson eigenvalue plot<sup>7</sup> reproduced in Fig. 1 will illustrate a possible approach to the determination of the deformation sign. For the mass region under study, the odd-

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<sup>1</sup> R. K. Sheline, T. Sikkeland, and R. Chanda, *Phys. Rev. Letters* **7**, 446 (1961); R. Chanda, J. Clarkson, and R. K. Sheline, *Proceedings of the Third Conference on Reactions between Complex Nuclei, Asilomar, California*, 1963 (University of California Press, Berkeley, California, 1963).

<sup>2</sup> C. Gerschel, M. Pautrat, R. A. Ricci, J. Teillac, and J. Vanhorenbeeck, *Comptes Rendus du Congrès International de Physique Nucléaire*, edited by P. Gugenberger (Centre National de la Recherche Scientifique, Paris, 1964), paper 3b (1)/C270.

<sup>3</sup> N. L. Lark and H. Morinaga (to be published); H. Morinaga, (private communications).

<sup>4</sup> A. C. Li, I. L. Preiss, P. M. Strudler, and D. A. Bromley, preceding paper, *Phys. Rev.* **141**, 1089 (1965).

<sup>5</sup> K. Kumar and M. Baranger, *Phys. Rev. Letters* **12**, 73 (1964).

<sup>6</sup> The only prior experimental evidence for oblate shapes was obtained in the region of Si<sup>28</sup> [D. A. Bromley, H. E. Gove, and A. E. Litherland, *Can. J. Phys.* **35**, 1057 (1957); C. Broude, L. L. Green, J. C. Willmott, and J. J. Singh, *Physics* **22**, 1139 (1956), and recently predicted theoretically in that region by Bar-Touv and Kelson-A. Bar-Touv and I. Kelson, *Phys. Rev.* **138**, B1035, (1965), and I. Kelson (private communication)] in a model similar to that of Kumar and Baranger (Ref. 5).

<sup>7</sup> S. G. Nilsson, *Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd.* **29**, No. 16 (1955).

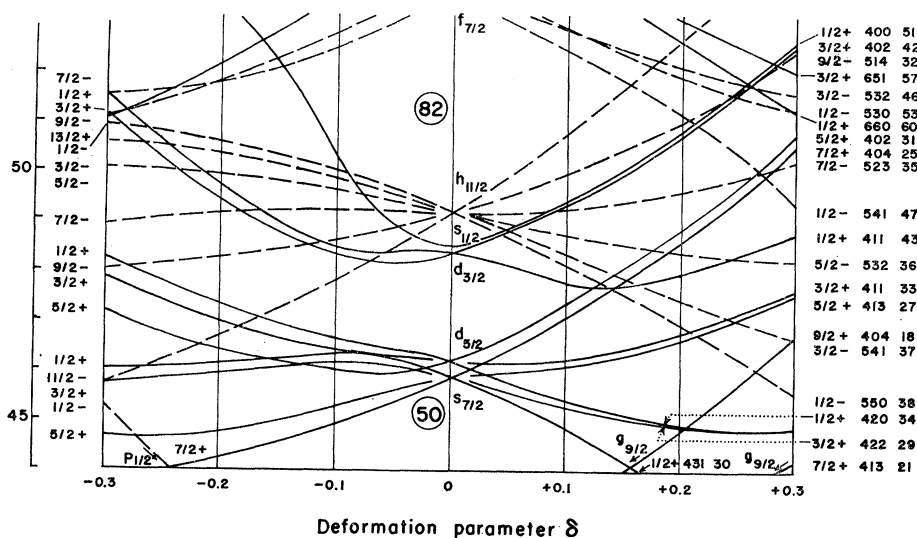


FIG. 1. Excerpt from the Nilsson eigenvalue plot for odd-nucleon number in the range 50 to 82 as given by Ref. 22. The entries at the right of the diagram list in sequence, the spin and parity, the asymptotic quantum numbers and the Nilsson numbers of the orbits involved. The ordinate is in units of  $E \times (A/100)^{1/3}$  MeV.

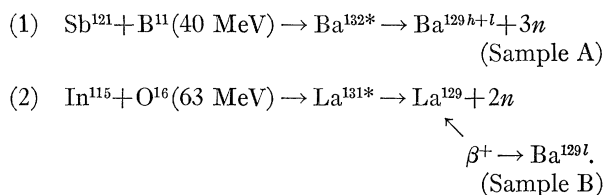
neutron nuclei fall on the portion of the diagram where the  $s_{1/2}$ ,  $d_{3/2}$ , and  $h_{11/2}$  orbits are juxtaposed. Examination of this diagram shows that for  $N < 73$ , isomerism is likely to occur for oblate but not for prolate deformations. Isomeric states have previously been observed for  $Ba^{135}$ ,<sup>8</sup>  $Ba^{133}$ ,<sup>8</sup> and  $Ba^{131}$ ,<sup>9</sup> whose neutron numbers are, respectively, 79, 77, and 75. If  $Ba^{128}$  indeed marks the onset of a region of static oblate deformation as has been proposed,<sup>1</sup> this trend of isomerism may be expected to persist through the more neutron-deficient species. The work of Arbman and Haller<sup>10</sup> alludes to the existence of an isomer of  $Ba^{129}$ , although no firm conclusion can be derived from works published to date.<sup>10-13</sup> In this paper, we report on a successful search for isomerism in  $Ba^{129}$ . We suggest, however, that evidence for well-defined static deformation in this region is not yet compelling. Additional studies must be carried to the still more neutron-deficient species.

#### EXPERIMENTAL PROCEDURE

We have utilized high-energy  $B^{11}$  and  $O^{16}$  beams from the Yale Heavy Ion Accelerator in these studies. Use of heavy projectiles conveys two particular advantages. First, because all conveniently available targets in this mass region have  $N \geq Z + 20$ , only reactions of the form (HI,  $xn$ ) populate states in nuclides far enough removed from the valley of stability to be of interest. Second, as has long been recognized, the heavy pro-

jectiles transfer much higher angular momenta to the reacting system and thus can populate much higher spin residual states than would otherwise be accessible. It has already been demonstrated<sup>14</sup> that this may lead to enhanced population of residual high-spin isomeric states in the final nucleus. In the present studies this characteristic has been used to advantage in separating isomeric activities for study.

$Ba^{129}$  sources were made in the following reactions<sup>15</sup>:



In the first of these reactions, an average of 10-15 units of angular momentum<sup>16</sup> are deposited in the compound nucleus. The product nucleus is therefore preferentially formed in a high-spin isomeric state, although a low-spin state is not thereby excluded. In the second reaction, reflecting the beta-decay selection rules, population of only one of the two spin states is anticipated. We have designated this state as low-spin for reasons which subsequent sections will make clear. The primary objective was to establish that both samples were predominantly  $Ba^{129}$ . In both instances beam energies were selected to optimize the yield of the desired  $x$  values in the (HI,  $xn$ ) reaction. Similarly, bombardment durations, chemical preparation, timing, etc. were all chosen (on the basis of available information and

<sup>8</sup> *Nuclear Data Tables* edited by K. Way (Printing and Publishing Office, National Academy of Sciences—National Research Council Washington, 25, D. C., 1963).

<sup>9</sup> R. S. Tilsbury and L. Yaffe, *Phys. Rev.* **129**, 1709 (1963).

<sup>10</sup> E. Arbman and I. B. Halber, *Nucl. Phys.* **22**, 341 (1961).

<sup>11</sup> W. Henkes, *Phys. Rev.* **113**, 803 (1959).

<sup>12</sup> R. W. Fink and D. H. Templeton, *J. Am. Chem. Soc.* **72**, 2818 (1950), R. W. Fink and E. O. Wiig, *Phys. Rev.* **95**, 708 (1954).

<sup>13</sup> D. J. Horen, W. H. Kelly and L. Yaffe, *Phys. Rev.* **129**, 1712 (1963).

<sup>14</sup> For example, see N. Dudev, doctoral dissertation, Clark University, 1963 (unpublished).

<sup>15</sup> The notations  $h$  and  $l$  refer to high- and low-spin states, respectively. The reason for such designation is given in the text which follows.

<sup>16</sup> T. D. Thomas, *Phys. Rev.* **116**, 703 (1959).

systematic measurements carried out in this laboratory) to minimize the yield of other barium isotopes.

In the following subsections the procedures utilized in preparing the two samples of Ba<sup>129</sup> for study are presented in outline.

#### Sample A

Thick, water-cooled, targets of natural antimony powder were bombarded with 40-MeV B<sup>11</sup> ions from the Heavy Ion Linear Accelerator for periods of two hours. The bombardment energy was selected on the assumption that the excitation functions measured by Black<sup>17</sup> for the (B<sup>11</sup>,  $xn$ ) reaction on a Cd<sup>116</sup> target would be essentially identical to those on the antimony target.

Following bombardment, the antimony powder was dissolved in aqua regia and the barium produced was precipitated as Ba(NO<sub>3</sub>)<sub>2</sub> by addition of fuming nitric acid to the solution in the presence of 10 mg of barium carrier. The nitrate precipitate was centrifuged and redissolved in a minimum of water. The resultant solution was made basic and antimony, coprecipitated in the process, was separated as a mixed oxide; this last step was repeated until no detectable Sb precipitate formed. The barium sample for study was prepared from the final solution through precipitation and filtering of barium sulphate.

Both because targets of natural antimony were in use and since the (B<sup>11</sup>,  $xn$ ) reaction only gives relative enhancement of a given  $x$  value, the sulphate sample thus produced inevitably was contaminated with barium isotopes other than Ba<sup>129</sup>. The following precautions were taken to enhance the Ba<sup>129</sup> yield:

(a) As low a B<sup>11</sup> energy as possible, while still inducing reasonable yield of the (B<sup>11</sup>,  $3n$ ) reaction, was used to minimize formation of the more neutron-deficient species through (B<sup>11</sup>,  $xn$ ) reactions with  $x=4$  or larger.

(b) Bombardment was limited to two hours in order to keep the specific activity of 2.4-day Ba<sup>128</sup> low in the sample.

(c) Following bombardment, but prior to chemical separation, a waiting period of one hour was used to permit the decay of much of the 11-min Ba<sup>127</sup>.

(d) The 42.75% relative abundance of Sb<sup>123</sup> in the source, and the selection of the (B<sup>11</sup>,  $3n$ ) reaction inevitably led to high yield of Ba<sup>131</sup>.

Fortunately the relatively long decay half-life of this isotope, 11.6 days, resulted in very low specific activity in the sample as noted under (b). Similarly the 14.6-min isomeric decay was considerably diminished as noted under (c).

The final sample was counted as a precipitate on its filter paper substrate. Chemical processing occupied a total time of 30 min excluding the 1-h waiting period.

#### Sample B

Water-cooled, 0.002-in. natural indium foils were bombarded with 63-MeV O<sup>16</sup> ions from the heavy-ion

accelerator for bombardment periods of 10 minutes. Following bombardment a waiting period of 10 min was observed to permit the decay of short-lived lanthanum isotopes.

The bombarded foils were first dissolved in hot concentrated HCl; 10 mg of lanthanum and barium carriers were added and initial separation of lanthanum was accomplished by introduction of 5 ml of saturated NaF to the solution. The La<sup>129</sup> parent was allowed to decay to Ba<sup>129</sup> for a period of 25 minutes, following which the initial precipitate was dissolved in a 4:1, HNO<sub>3</sub>:H<sub>3</sub>BO<sub>3</sub> solution. Residual lanthanum was precipitated from this solution as the hydroxide and discarded. Finally the barium was precipitated as the nitrate and redissolved. The final sample was reprecipitated as BaSO<sub>4</sub>.

Since the individual final samples had relatively low specific activities, in most of the work reported herein, as many as 6 separate bombardments and separations were carried out and combined to form the B sample.

The chemical separation following a given bombardment occupied 30 min exclusive of waiting time.

In marked contrast to the situation pertaining to sample A, maximizing of the Ba<sup>129</sup> isotopic yield in sample B was much more difficult. First, in the evaporation reactions used, the (O<sup>16</sup>,  $2n$ ) reaction has small intrinsic yield. Second, the half-lives of other La activities are much closer to that of the La<sup>129</sup> precursor. For example, the half-life of La<sup>129</sup> is only about twice that of La<sup>127</sup> and of La<sup>128</sup>. Although Ba<sup>128</sup> has a long half-life as noted above, both the 11 min Ba<sup>127</sup> and its 6.2-h Cs<sup>127</sup> daughter presented a problem. Ba<sup>131</sup> posed no problem since no significant La<sup>131</sup> was produced in its ground state in the reaction used.

#### MASS ASSIGNMENT

In order to obtain a mass assignment, and thus identify the barium isotope involved in samples A and B, a chemical milking procedure was invoked. In such a procedure, activity attributable to the radioactive daughter of an initially pure radioactive parent is separated from the parent activity at successive, equally spaced time intervals. The decrease in the initial activity of the daughter components thus obtained in each fraction provides a direct measure of the half-life of the parent radionuclide.

In separate experiments, with both the O<sup>16</sup>-In and B<sup>11</sup>-Sb systems, Ba(NO<sub>3</sub>)<sub>2</sub> was isolated as described previously. The sample was dissolved in a minimum of H<sub>2</sub>O, and at 90-min intervals, Cs<sub>2</sub>PtCl<sub>6</sub> was precipitated by addition of 1 ml of CsCl (10 mg/ml Cs<sup>+</sup>), 500 μl of 10% H<sub>2</sub>PtCl<sub>6</sub> solution, and a few drops of 90% ethyl alcohol. The Cs<sub>2</sub>PtCl<sub>6</sub> was centrifuged, filtered, dried, weighed, and counted. Chemical yields between 95 and 100% were obtained.

The decay of the activity, identifiable as Cs<sup>129</sup>

<sup>17</sup> R. Black, doctoral dissertation, MIT, 1964 (unpublished).

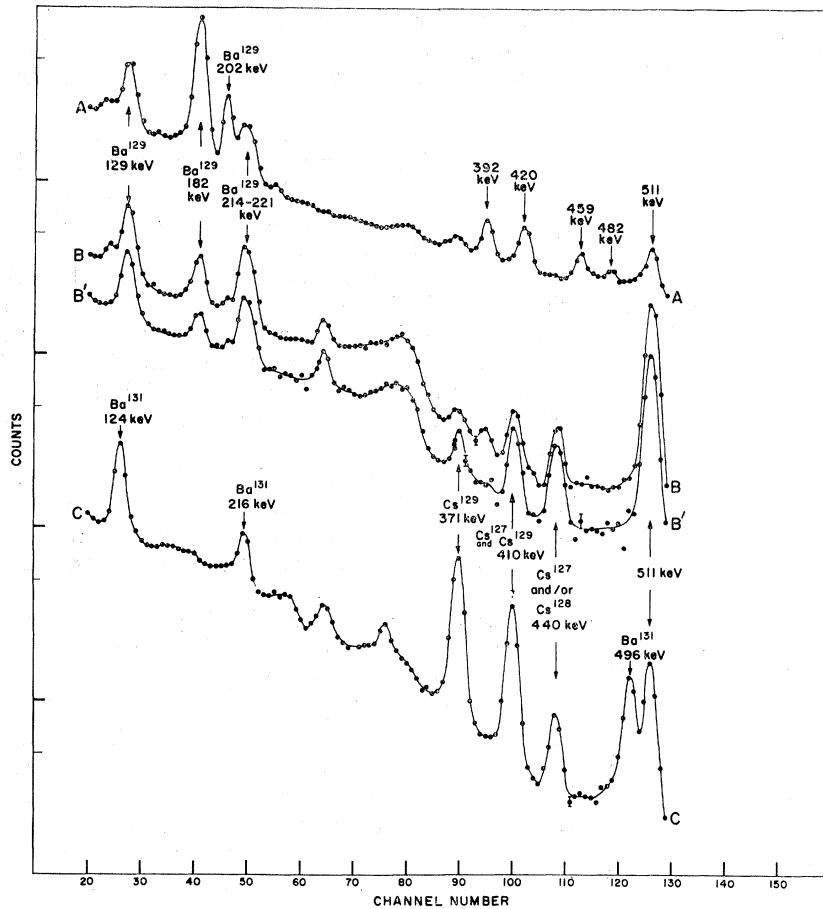


FIG. 2. Representative gamma radiation spectra from the decay of  $Ba^{129}$ . The germanium detector used had dimensions of  $10 \times 20 \times 2$  mm. Detailed description of these spectra are given in the text.

through the well-established 370- and 410-keV transitions associated with its decay, was followed using a  $3 \times 3$ -in. NaI spectrometer and standard associated electronic instrumentation. The  $Cs^{129}$  half-life, as determined from these measurements, was always within 5% of its reported value of 30.7 h.

The half-life of the barium activity milked from the  $B^{11}$ -Sb sample was  $2.1 \pm 0.2$  h, whereas when milked from the  $O^{16}$ -In sample it was  $2.0 \pm 0.3$  hours. Both of these values were in complete accord with the results obtained by direct observations on the  $Ba^{129}$  gamma radiation to be described in the next sections.

These measurements unambiguously establish that samples A and B were indeed both predominantly  $Ba^{129}$ .

#### GAMMA-RADIATION STUDIES

Studies were carried out on the  $Ba^{129}$  gamma radiation from both samples utilizing both standard NaI scintillators and lithium-drifted germanium detectors fabricated in this laboratory. Representative spectra obtained with germanium detectors are shown in Fig. 2.

Spectrum A was obtained with sample A about 3 h after the end of bombardment. As was anticipated from the conversion-electron work of Arbman and Haller,<sup>10</sup>

a large number of  $\gamma$  rays were detected. Each line reported by these authors having  $K$ -conversion line intensity greater than 0.4 of that of the 182-keV line was observed in the present work, in addition to several of lesser intensity. A point to be noted is that annihilation radiation is not present to any large extent.

Spectrum B (taken with sample B), in contrast to spectrum A, exhibited a very strong 511-keV peak. The 129-, 182-, and 214-221-keV lines are clearly still present, but now have very different relative intensities from those of spectrum A. In the work of Henkes,<sup>11</sup> reporting a study of triple coincidences of 511-511-keV radiation with  $\gamma$  rays, 127- and 210-keV lines were found to be in coincidence with positrons. It might be argued that this latter experiment was essentially a sorting out of the present "sample B" component from a composite source.

The spectrum labeled B' was taken about  $2\frac{1}{2}$  hours after the recording of spectrum B. Those  $\gamma$  rays identified with  $Ba^{129}$  have decayed appropriately while the various Cs lines have not. The growth of the  $Cs^{129}$  371-keV line is clearly seen. This is, again, evidence in support of the assignment of the initial activity in the sample as being predominantly due to  $Ba^{129}$ . The ob-

servation of the  $Cs^{129}$  410-keV line is obscured by the presence of the 410-keV  $Cs^{127}$   $\gamma$  ray.

The decay of sample A was followed for two days. Spectrum C shows the residual  $\gamma$  rays in sample A 30 h after spectrum A was taken. These  $\gamma$  rays in terms of their respective energies and half-lives, may be assigned to various contaminant and residual isotopes. As we have already pointed out, little  $Cs^{127}$  was expected and none was detected. From the intensities<sup>18</sup> of the  $\gamma$  rays identified and from the proposed decay schemes of the various isotopes, the relative disintegration rates of  $Ba^{128} - Cs^{128} : Cs^{129} : Ba^{131}$  were found to be approximately in the ratio 1:12:2. From the 12:2 ratio it might be concluded that the initial amounts of  $Ba^{129}$  and  $Ba^{131}$  in sample A were roughly equal, assuming that all the  $Ba^{129}$  had decayed into  $Cs^{129}$  by the time spectrum C was taken. This appears entirely reasonable, in view of the fact that the Ba isotopes were produced mainly by the  $(B^{11}, 3n)$  reaction on natural Sb, in which there is approximately 57%  $Sb^{121}$ , and 43%  $Sb^{123}$ .

We shall defer more complete discussion of these and other decay spectra to our consideration of the  $Cs^{129}$  level structure.<sup>19</sup> It is possible however, to draw several conclusions from even a cursory examination of these data.

It appears that sample A contains a significant admixture of  $Ba^{129}$  and  $Ba^{129m}$ . Whichever is the more abundant decays predominantly via electron capture and gives rise to strong 182- and 202-keV gamma transitions. Since the sample B resulted entirely from beta decay of a precursor it can contain either  $Ba^{129}$  or  $Ba^{129m}$  but not both. Whichever this may be, it decays with a relatively large positron component, and, excepting the associated 511-keV annihilation radiation, the strongest gamma line associated with this activity is a composite at 214–221 keV.

Since the two samples decay with markedly different characteristics, and since the production mechanism favors population of high-spin states in sample A, for purposes of discussion we shall associate sample A with the high-spin isomer and sample B with the low-spin isomer. The half-lives of the two states are therefore obtained by following the decay of the 182–202 keV and 214–221 keV radiations, respectively. The results are shown in Fig. 3. As will be discussed further in a later section, it is not possible *ab initio* to establish the energy ordering of the low- and high-spin members of the  $Ba^{129}$  isomeric configuration.

The 182-keV line and the 202-keV line both decayed with the same half-life of  $2.13 \pm 0.06$  h, a value which agrees very well with the measurements of Arbman and Haller. However, that the decay of the 214–221-keV line should show a half-life all but indistinguishable from

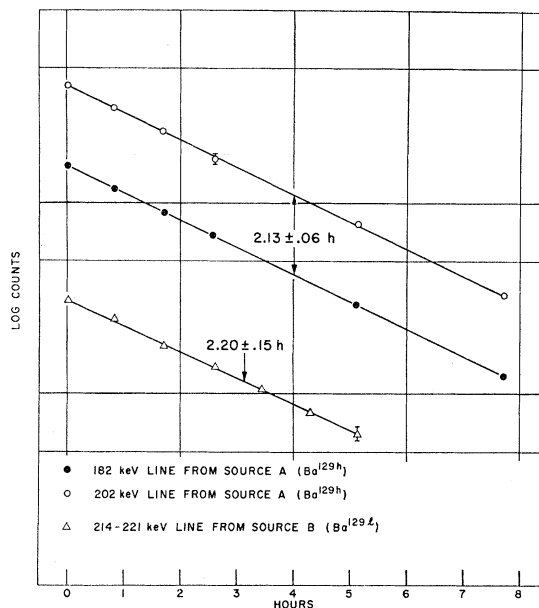


FIG. 3. Half-life measurements of the  $Ba^{129}$  isomers as determined from gamma radiations.

213 h is surprising, since this implies that  $Ba^{129m}$  and  $Ba^{129}$  have effectively the same half-life. This is clearly a most unusual situation. However, since the evidence which we have presented herein defines an isomeric situation, we must conclude that this effective identity of lifetimes is simply a statistical accident.

A best fit to the points of the 214–222-keV decay gives a half-life of  $2.20 \pm 0.15$  h. The uncertainties quoted for the two half-lives have included statistical as well as estimated systematic errors. Our attempt at half-life determination from annihilation radiation did not give a conclusive result because our samples contained small, but not insignificant, amounts of  $Ba^{127}$  and  $Ba^{128}$ . Both of these latter isotopes have positron-emitting daughters, and  $Ba^{127}$  is itself a positron emitter. The lifetime measurements based on the 214–221-keV line has the advantage that there is no subtraction of other decay components involved.

Inasmuch as the 214–221-keV transition has been associated herein with the state exhibiting positron decay it would be anticipated that the lifetime should be that measured directly on the positron decay by Arbman and Haller and with that measured by Henkes on the associated annihilation radiation. Our value of  $2.20 \pm 0.02$  h does not agree with the  $2.61 \pm 0.02$  h and  $2.45 \pm 0.05$  h quoted by the above authors within their stated errors. Since, as indicated by Fig. 3, the present measurement appears relatively unambiguous we believe that the half-life of  $2.20 \pm 0.15$  h which we quote is a realistic one. By reason of the way sample B was prepared, it can contain no  $Ba^{131}$ . (Hence the 216-keV line seen in the residual activity of sample A does not interfere with our measurement.) Furthermore, even

<sup>18</sup> The efficiency of the Li-Ge detector was determined by calibration against a  $3 \times 3$ -in.-NaI crystal using standard gamma-ray sources.

<sup>19</sup> A. C. Li, I. L. Preiss, P. M. Strudler, and D. A. Bromley (to be published).

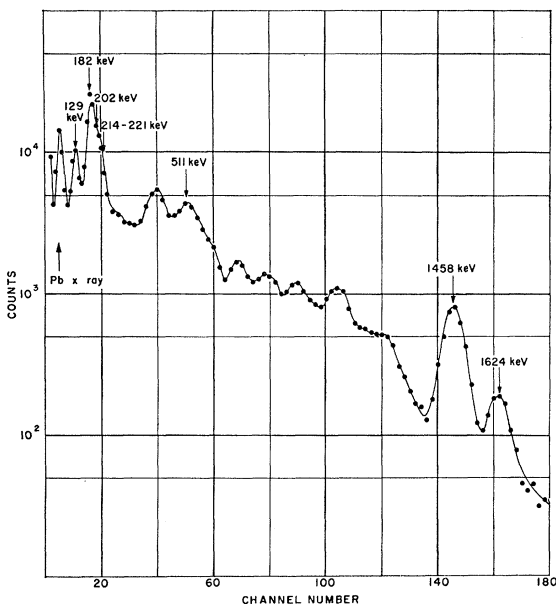


FIG. 4. Representative gamma-radiative spectrum obtained using a 3×3-in.-NaI(Tl) spectrometer. The spectrum characteristics are discussed in the text.

if there had been some long-lived interfering gamma ray whose presence was overlooked, the discrepancy would lie in the opposite direction from that which would bring us into agreement with previous measurements. We do not believe that any essential disagreement is involved concerning this lifetime, since the limits of error placed on the lifetime in the earlier work may be slightly overstated in view of the inherent spread of values for individual measurements quoted.

The basis for comparison with the half-life measurements of Fink and Templeton<sup>12</sup> and Yaffe *et al.*<sup>20</sup> is not so clear. These works do not either implicitly or explicitly distinguish between two isomeric components in their sources. Therefore it is simply noted that Fink and Templeton gave a value of 1.9 h for the half-life, and Yaffe *et al.* gave a value of  $2.22 \pm 0.05$  h.

#### ASSIGNMENTS OF STATES

The energy of the low-spin state of Ba<sup>129</sup> relative to the ground state of Cs<sup>129</sup> is directly determinable from the end point of the most energetic positron branch assuming that a ground-state to ground-state transition occurs. Comparison of the intensity of the annihilation radiation with that of all other gamma radiation in spectra *B* and *B'* of Fig. 2 indicates that indeed a strong positron branch to the Cs<sup>129</sup> ground state is required. An experimental study of the positron spectra using a 3-mm-deep lithium-drifted silicon detector, and requiring coincidences with the associated annihilation radiation, gave results entirely consistent with the more

<sup>20</sup> L. Yaffe, J. M. Alexander, and E. K. Hyde, *Can. J. Chem.* **41**, 1951 (1963).

accurate spectrometer measurements of Arbman and Haller.<sup>10</sup> We therefore defer to the greater precision of their measurement and adopt their end-point value of 1425 keV, placing the parent Ba<sup>129</sup> state 2.447 MeV above the Cs<sup>129</sup> daughter ground state.<sup>21</sup>

From the intensity of the annihilation radiation, it can be ascertained that positrons of 2-h half-life are at least twice as numerous as all the gamma rays associated with the Ba<sup>129</sup> decay taken together. Therefore, a limit may be established on the  $\log ft$  value of the ground-state to the ground-state decay branch, even in the absence of complete knowledge of the decay scheme. The limit of  $\log ft \leq 5$  indicates, at most, a first-forbidden character for this decay branch. Since the ground state of Cs<sup>129</sup> is known to have a spin of  $\frac{1}{2}$ , the Ba<sup>129</sup> parent state must have a spin of  $\frac{1}{2}$  or  $\frac{3}{2}$  and our assignment of this isomeric configuration as low spin is thereby unambiguously confirmed.

The energy of the high-spin state is not so simply determined. The most direct determination would, of course, be via a determination of the energy of the isomeric transition linking the two states. In gamma-ray measurements, the identity of lifetimes just noted makes it essentially impossible to detect the occurrence of an isomeric transition. That such a transition does *not*, in fact, occur is definitely indicated by the high-resolution measurements of Arbman and Haller,<sup>10</sup> using a double focusing spectrometer to study conversion electrons. In consequence, we are forced to more indirect determinations.

The annihilation radiation appearing in spectrum *A* of Fig. 2 can be completely accounted for by postulating an admixture of low-spin isomeric state in Sample A consistent with the intensity of the 214–221 composite line of spectrum *A*. These data are entirely consistent with completely positron-free decay of the high-spin isomer to high-spin high excitation states, in the daughter Cs<sup>129</sup>. The high-excitation specification follows from the observation of some 100 gamma transitions both in spectrum *A* and in the work of Arbman and Haller. These transitions are currently being analyzed for information on the Cs<sup>129</sup> level structure. However this analysis is extremely complicated and is far from completed as yet. Certain restricted aspects of this decay, pertinent to the present discussion are illustrated in Fig. 4, which is a NaI spectrum corresponding to spectrum *A*, but measured at lower gain to display a number of more energetic transitions. A very strong line at 1.46 MeV is in evidence together with a line some 20% as intense at 1.62 MeV. Gamma-gamma coincidence studies have shown that both of these transitions participate in cascades involving the 182-keV level in Cs<sup>129</sup>.

<sup>21</sup> It should be noted that Cameron's semiempirical mass equations [Chalk River report CPR-690, AECL-433, 1957 (unpublished)], predict a value at 3.559 MeV for this separation, providing some measure of the uncertainty in the mass predictions in this region and, more particularly, evidence precluding use of such mass equations in any arguments concerning level ordering in these nuclei.

Therefore, the parent state is clearly higher than 1.80 MeV in excitation.

An upper limit to the energy of the high-spin state, relative to the Cs<sup>129</sup> ground state, may also be estimated from a determination of a limit for the ratio of electron capture (1.62 MeV) to positron decay, corresponding to the decay which populates the level from which the strong 1.46-MeV transition in Cs<sup>129</sup> originates. This was accomplished through examination of the spectrum of gamma-radiation coincident with the photopeak of the 1.46-MeV transition. The absence of annihilation radiation in this spectrum gives a limit for the  $K/\beta^+$  ratio of  $\geq 120$ . On the assumption that the most pronounced branch decay is allowed (which appears entirely reasonable on the grounds that the decaying nucleus has a high level density from which to choose a daughter state) a positron end-point energy of less than 0.5 MeV is obtained. Correspondingly, this limits the separation of the isomeric states in Ba<sup>129</sup> to 700 keV.

Since, as noted above, the low-spin isomer member in Ba<sup>129</sup> decays via an apparently allowed positron branch to the  $\frac{1}{2}^+$  ground state of Cs<sup>129</sup>, its assignment is  $(\frac{1}{2}^+, \frac{3}{2}^+)$ . The high-spin member does not decay by the energetically open channel to low-lying states of Cs<sup>129</sup>, indicating that such decays are precluded on angular-momentum grounds.

It may be useful, for orientation purposes, to examine the variation for the gamma-ray transition lifetime as a function of the transition energy, for several possible multipolarities. On the basis of any reasonable model, the transition in question almost certainly must involve substrates of the  $h_{11/2}$  and  $s_{1/2}$  or  $d_{3/2}$  orbits. Hence a parity change is expected. Figure 5 is a plot of Weisskopf estimates,<sup>22</sup> appropriately corrected for internal conversion, for E3, M4, and E5 multipoles, together with the experimentally determined Ba<sup>129m</sup> half-life.

Since the half-lives of the isomeric states are so nearly identical, it makes little difference to the argument that is not known which is the ground and which the metastable state. As shown above, the separation of the isomeric states has a limit of 700 keV. It appears reasonable to assume that if the branching ratio for an isomeric transition is as much as  $\frac{1}{10}$  in the depopulation of Ba<sup>129</sup> the transition would have been seen. The non-observation of an isomeric transition therefore implies that the partial half-life for this process is  $> 8 \times 10^4$  sec. Barring a very extraordinary retardation for E3, the transition in question is most probably of the type  $\Delta J \geq 4$ , yes. This argues for an assignment of  $(\frac{1}{2}^+, \frac{3}{2}^+)$  and  $(\frac{3}{2}^-, \frac{1}{2}^-)$  to the isomeric configurations.

## DISCUSSION

As indicated previously, the identification of an isomeric configuration in Ba<sup>129</sup> is in accord with qualitative

<sup>22</sup> G. J. Nijgh, A. H. Wapstra, and R. Van Lieshout, *Nuclear Spectroscopy Tables* (North-Holland Publishing Company, Amsterdam, 1959).

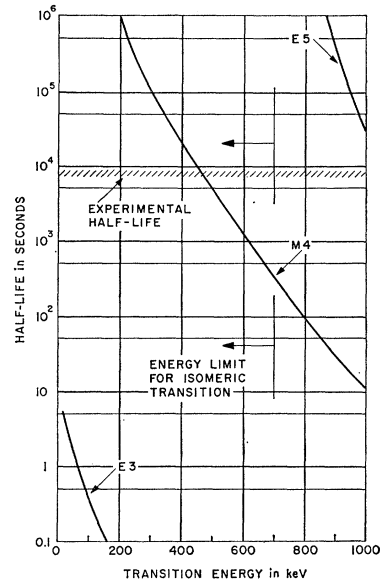


Fig. 5. Graphical representation of Weisskopf lifetimes estimates for selected gamma radiation multipolarities as functions of the transition energies. The lifetimes have been corrected appropriately for internal conversion contributions to the decays.

expectations based on the Nilsson model<sup>7</sup> level ordering. However, this model ordering does not reflect pairing and other refinements which have been treated in the above-mentioned calculations by Kumar and Baranger.<sup>5</sup>

The harmonic-oscillator eigenvalues which were utilized in these calculations were essentially those of Mottelson and Nilsson<sup>23</sup> for the protons and those of Nilsson<sup>7</sup> for the neutrons. Figure 6 reproduces the eigenvalue plots given by Kumar and Baranger for the  $50 \geq N, Z \geq 82$  odd-neutron and odd-proton cases, respectively. The striking difference between these two sets of eigenvalues is the lowering of the  $h_{11/2}$  shell for protons from its original value which had resulted from the Mottelson-Nilsson choice of adjustable parameters to reproduce the then available data. This effect presumably reflects Cohen's<sup>24</sup> discovery of a relatively strong average interaction between nucleon shells which have the same total angular momentum. Thus when the neutron  $h_{11/2}$  shell is filling, the corresponding proton shell remains open, while when the proton shell is filling, the neutron shell is already filled and the mutual interaction increases the binding of the  $h_{11/2}$  protons. Applying this modified model of Kumar and Baranger to the mass region in question, both odd-neutron and odd-proton nuclei are expected to exhibit isomerism for oblate deformations. However, as demonstrated by these authors, the behavior of the total-binding energy precludes such a deformation for the odd-proton cases.

<sup>23</sup> B. R. Mottelson and S. G. Nilsson, *Kgl. Danske Videnskab. Selskab, Mat. Fys. Skrifter* **1**, No. 8 (1959).

<sup>24</sup> B. L. Cohen, R. Patell, A. Prakash, and E. J. Schneid, *Phys. Rev.* **135**, B383 (1964).

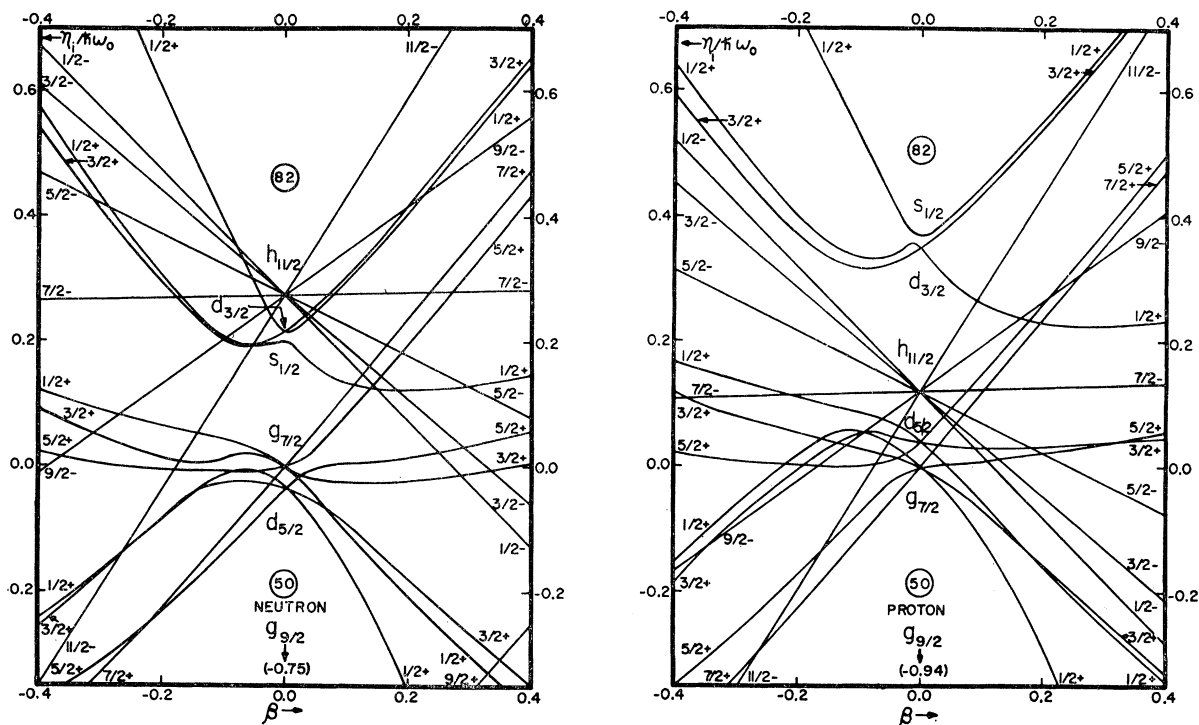


FIG. 6. Eigenvalues of the deformed nuclear Hamiltonian plotted as functions of the deformation parameter  $\beta$  for the nucleon number range 50 to 82. The figures on the left and right are appropriate to neutrons and protons, respectively. These figures are taken from the work of Kumar and Baranger.

Turning to the odd-neutron cases, in the modified model, the ordering of the  $d_{3/2}$  and  $s_{1/2}$  orbits is interchanged from the Nilsson ordering and the  $h_{11/2}$  orbit is slightly lowered.<sup>25</sup> Isomeric configurations with the experimentally indicated angular-momentum difference of  $\geq 4$  still occur only for oblate deformations, and then only for very small deformations ( $\beta \sim -0.05$ ). cursory inspection of the figure would suggest that the isomeric states in  $\text{Ba}^{129}$  might be identified with Nilsson configurations No. 51,  $|400, \frac{1}{2}^+\rangle$ , and No. 32,  $|514, \frac{9}{2}^-\rangle$ , and thus provide support for the Kumar-Baranger predictions of oblate shape. The ground-state assignment of  $\frac{1}{2}^+$  in the case of  $\text{Cs}^{129}$  suggests a prolate shape with  $0.05 \leq \beta \leq 0.2$ . Consistent with the anticipated approach to spherical shape with increasing neutron number,  $\text{Cs}^{131}$  has an assignment  $\frac{5}{2}^+$  while  $\text{Cs}^{133}$ ,  $\text{Cs}^{135}$ , and  $\text{Cs}^{137}$  have assignments<sup>8</sup> of  $\frac{7}{2}^+$  as required by the model. It must be noted, however, that the indicated values of  $\beta$  are not adequately large to insure a statically deformed nuclear configuration. This and the above-mentioned level structure of the barium and xenon isotopes both lend credence to the con-

clusion that the nuclides thus far studied fall in a collective transitional region of predominantly vibrational character.

Additional support for this contention may be seen in Table I. The available  $E2$  reduced transition probabilities in this mass region are compared with the well-known samarium transitional region, where the vibrational rotational character of  $\text{Sm}^{148}$  and  $\text{Sm}^{154}$ ,

TABLE I. Comparison of reduced transition probabilities, in single-particle units, in the barium-cesium region with those in the samarium transitional region.

Nucleus	Transition (energy in keV)	$E2$ Enhancement <sup>a</sup>
$\text{Sm}^{148}$	2 (551) 0	38 <sup>b</sup>
$\text{Sm}^{150}$	2 (334) 0	56 <sup>b</sup>
$\text{Sm}^{152}$	2 (122) 0	142 <sup>b</sup>
$\text{Sm}^{154}$	2 (82) 0	190 <sup>b</sup>
$\text{Ba}^{130}$	2 (359) 0	37 <sup>b</sup>
$\text{Ba}^{132}$	2 (470) 0	35 <sup>b</sup>
$\text{Cs}^{131}$	? (124) $\frac{5}{2}$	65 <sup>c</sup>
	? (55) $\frac{7}{2}$	51 <sup>c</sup>

<sup>a</sup>  $E2$  Enhancement is defined as the ratio of the experimental  $B(E2)$  to the  $B(E2)$  calculated from the Weisskopf formula for single-particle transition as given in J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley & Sons, Inc., New York, 1952), p. 627.

<sup>b</sup> Calculated from half-lives taken from *Nuclear Data Sheets*, compiled by K. Way *et al.* (Printing and Publishing Office, National Academy of Sciences—National Research Council, Washington, 25, D. C.).

<sup>c</sup> Taken from D. J. Horen, J. M. Hollander, and R. L. Graham, *Phys. Rev.* **135**, B301 (1964).

<sup>25</sup> In the absence of sufficient experimental data against which comparisons can be made, discrepancies such as these suggest that further relative adjustment of various orbital states may not be precluded. Therefore even though the following discussion takes a somewhat literal view of the model, certain reservations should be understood as implied.



respectively, are quite well established. Again these data suggest that the barium and cesium isotopes studied thus far fall in a transitional region.

Having concluded that the evidence for a static deformation is not compelling, it then becomes of interest to examine the degree to which the new nuclear information presented herein may be compatible with a pure vibrational interpretation based on a spherical equilibrium configuration.

The nuclides under discussion here, are just on the edge of the projected region of validity of the calculations which have been carried out by Kisslinger and Sorensen (KS).<sup>26</sup> These calculations apply simple residual forces to predict the vibrational behavior of spherical and near-spherical nuclei. Kumar and Baranger<sup>5</sup> have noted that if they insert Kisslinger and Sorensen rather than Nilsson eigenfunctions in their calculations of the equilibrium deformation no significant changes result. There are however, significant differences in the predicted eigenvalues corresponding to the valence nucleons in the two models. The models agree with one another and with experiment in predicting  $J = \frac{1}{2}^+$  for Cs<sup>12</sup>, and no low-lying isomeric structure. The (KS) calculations have not been carried below  $A = 131$  for the barium isotopes. However, extrapolation of the indicated trends would predict a  $\frac{1}{2}^-, \frac{3}{2}^-, \frac{5}{2}^-, \frac{7}{2}^-$  sequence at lower excitation, precluding existence of a significant isomeric lifetime in the model. The  $\frac{9}{2}^-$  state which appears to play the isomeric role in Ba<sup>129</sup> falls at much higher excitation ( $> 1$  MeV) in the (KS) calculations.

It would thus appear that although the (KS) vibrational and Nilsson rotational eigenfunctions give comparable equilibrium-deformation predictions, the latter are somewhat favored in the light of the observed isomeric behavior.

<sup>26</sup> L. S. Kisslinger and R. A. Sorensen, Rev. Mod. Phys. 35, 853 (1963).

## CONCLUSION

Unambiguous evidence has been found for isomerism in Ba<sup>129</sup>. While in the light of the Kumar-Baranger model calculations this may be suggestive of oblate deformation, the weight of evidence in this region strongly implies that all nuclides studied to date lie in a collective transitional region of predominantly vibrational character.

Indications are that data on more highly neutron-deficient species are required before any definitive statement can be made concerning the detailed characteristics of the anticipated new islands of deformation near the  $N=Z$  line. The present studies, and others to be reported from this laboratory, suggest that extension of measurements to the required species will pose formidable experimental problems. For example, the rapid decrease of the isotope half-lives may eventually preclude the chemical separations necessary for unambiguous isotopic identification of activities. This same shortness of the lifetimes also puts severe restrictions on the types of measurements which can be performed. More elaborate mass-identification techniques may well prove very fruitful; on-line mass spectroscopy of recoiling radionuclides or fast off-line mass spectroscopy of bombarded targets appear to represent promising approaches to the problem.

Present indications are that the modified Nilsson model continues to provide a satisfactory framework for the correlation of available data in this mass region.

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