MEASUREMENTS OF THE NUCLEAR GAMMA RAYS IN MUONIC ATOMS OF SEVERAL DEFORMED NUCLEI*

S. Bernow, S. Devons, I. Duerdoth, D. Hitlin, J. W. Kast,
W. Y. Lee, E. R. Macagno, J. Rainwater, and C. S. Wu
Columbia University, New York, New York
(Received 10 June 1968)

The energy shifts of the nuclear gamma rays from the first rotational states of Nd^{150}, Sm^{152}, Gd^{154}, Er^{166}, W^{182}, W^{184}, and W^{186} due to the presence of a muon in a 1s state have been measured. The shifts are related to changes in the nuclear mean-square charge radius, and are compared with Mössbauer isomer-effect measurements.

In May 1967 we reported[1] the observation of an increase of the order of 1 keV in the energy of the first rotational gamma-ray transition of Sm^{152} (121.8 keV) due to the presence of a muon in the $1s$ state. The excitation of a strongly deformed nucleus to its low-lying rotational levels in a muonic atom is produced by the dynamic quadrupole interaction between the muon and the nucleus, as first predicted theoretically by Wilets[2] and Jacobsohn.[3] The probability that the nucleus will be left in the $I=2^+$ state by the dynamic $E2$ excitation depends on the interaction energy, $2p$ fine structure, and the rotational energy: For example, it is 0.30 for Nd^{150} and 0.56 in U^{238}. The subsequent de-excitation of the nuclear state, which has a lifetime of about 10^{-9} sec, takes place in the presence of the muon, since the muon capture lifetime from the $1s$ state is about 10^{-7} sec in the rare-earth region.

In our previous Letter,[1] the observed shift in the nuclear γ-ray energy in Sm^{152} was interpreted as a change in the binding energy of the muon for the 0^+ and 2^+ nuclear states, arising from a difference between the nuclear charge distribution of these two states. This difference has been observed by Mössbauer techniques and is called the "isomer effect." For the case of Sm^{152}, the difference in the mean-square charge radii of 0^+ and 2^+ states obtained from the γ-ray shift in the muonic atom was comparable with that deduced from Mössbauer measurements.

Following this first measurement an improved energy comparison system was devised, and we report here the energy shifts for seven deformed nuclei. The first three nuclei, $_{60}Nd^{150}$, $_{62}Sm^{152}$, and $_{64}Gd^{154}$, all of which have 90 neutrons, are considered to be permanently deformed, and to behave like "soft" rotators. These three nuclei all exhibit approximately equal positive energy shifts. If the shift is due only to the difference of the mean-square charge radius of the two nuclear states, then the positive energy shift implies that the excited state is larger than the ground state, as expected from centrifugal stretching.[4] It was therefore surprising when negative energy shifts were observed in the muonic atoms of Er^{166}, W^{182}, W^{184}, and W^{186}, particularly since small but positive isomer shifts had been reported[5,6] in the W nuclei by Mössbauer techniques.

The small energy shifts of the nuclear gamma rays due to the presence of a $1s$ muon were obtained by directly and simultaneously comparing the energy of the nuclear gamma ray in coincidence with a stopped muon with that of the unshifted gamma ray following beta decay from a $Z\pm 1$ nucleus (see Fig. 1). The general principle of the method was described in a previous Letter.[1] In the case of Nd^{150}, where no radioactive source for direct comparison is available, the de-excitation line in the muonic atom was measured with respect to several well-known calibration lines. The unshifted line, produced by Coulomb excitation, was subsequently measured using the same calibration lines.

Several improvements to the previous system were introduced. A new windowless Ge(Li) detector was fabricated by removing both n- and p-type regions from the front and back of a conventional planar detector and re-establishing new n- and p-type regions on the top and bottom edges of the crystal. The dimensions of this detector were 7 cm^2 area by 0.4 cm depth. The resolution (full width at half-maximum) was 1.3 keV at 122 keV and 2.3 keV at 1.33 MeV. The detector was placed outside of the muon beam. Two thin targets of 6 cm $\times$ 6 cm with surface density of ~0.5 g/cm^2 were placed at 45° to the beam and detector axes. A thin (1.5-mm) anticoincidence plastic scintillator was placed in front of the Ge(Li) detector to reject events caused by scattered particles entering the detector.

Care was taken to avoid any possible systematic shifts between the two spectra by making the

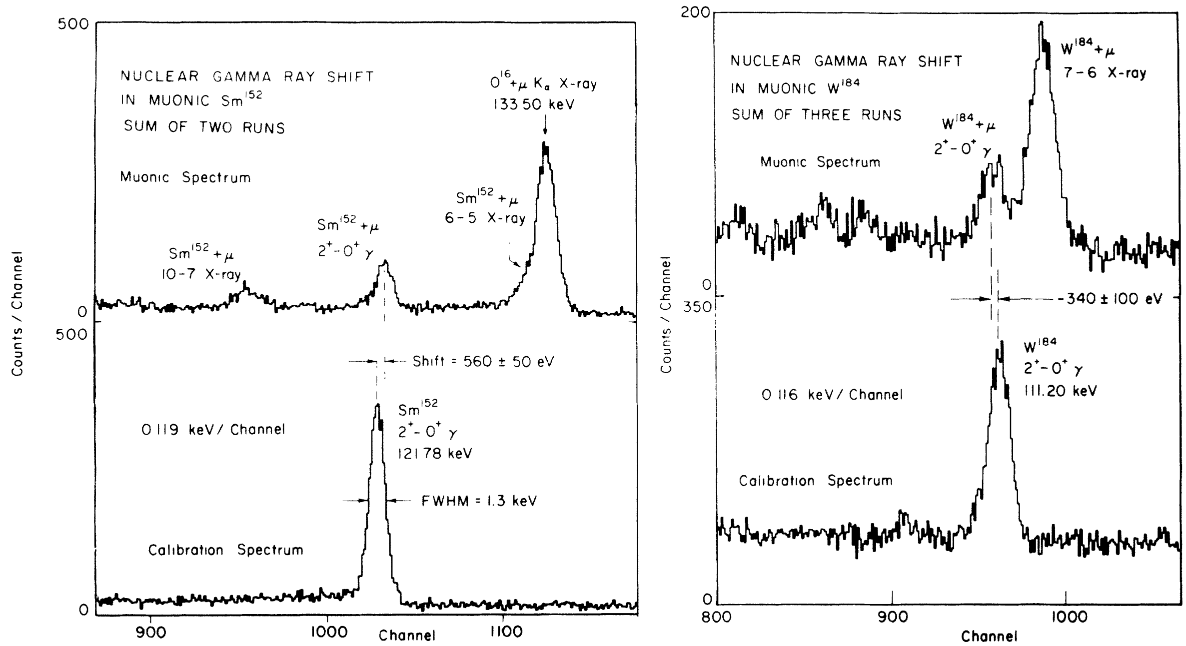

FIG. 1. The shifts in the energies of the first rotational nuclear gamma rays of Sm^{152} and W^{184} due to the 1s muon. The upper spectra show the de-excitation gamma rays $(2^+ \rightarrow 0^+)$ in the presence of the muon. The lower spectra exhibit the corresponding $(2^+ \rightarrow 0^+)$ gamma rays from radioactive sources.

time relation between the analog pulse and the pulse which enabled the linear gate independent of the type of event involved (muonic, calibration, or stabilization), and by positioning the calibration source close to the target.[7] Any possible shifts were repeatedly checked for by comparing the atomic K x rays of Pb from a Pb target in the muonic spectrum with the K x rays of Pb from Bi^{207} (K capture) in the calibration spectrum. No shifts were found within the experimental error (±20 eV).

Examples of our data are shown in Fig. 1. The two spectra on the left show a positive energy shift of +560 eV in Sm^{152} and the two on the right show a negative shift of −340 eV in W^{184}. The positions of the lines were determined by finding the best least-squares fit of a Gaussian plus background. In the case of overlapping peaks several Gaussians were fitted simultaneously. The sources used for the determination of the shift are listed in column III of Table I. The energy shifts obtained are listed in column IV. The uncertainties include both statistical and systematic errors. The latter arise from three sources which contribute approximately ±20 eV each: the difference in geometry and position of the target and the calibration source, the possible shifts in timing between the two spectra mentioned above, and the presence of weak unresolved muonic or pionic x-ray lines.

In interpreting the observed shifts ΔE_{obs}, we list the physical effects which can contribute significantly:

$$\Delta E_{obs} = \Delta E_{pol} + \Delta E_{c.g.} + \Delta E_{isomer}.$$

These are considered in turn.

(1) ΔE_{pol} is the difference in nuclear polarization between the 2^+ and 0^+ states. Chen[8] has shown that in the simple rotational model the nuclear polarization corrections to the 2^+ and 0^+ states due to excitations to intermediate states in the same rotational band or neighboring excited K bands are nearly identical. The muonic intermediate states of importance are d states in the continuum, with energies of 10 MeV or higher; therefore, a change of 100 keV in the energy denominator results in a negligible effect. However, this conclusion applies only to the rotational model, which, in general, is not expected to hold to better than a few percent. Since the total nuclear polarization contribution in deformed nuclei is estimated to be about 10 keV for the 1s state, a difference of a few percent of this value could result in energy shifts comparable with those we have observed. In the absence of any reliable estimates, we have neglected this possible contribution to the observed shifts; this should be borne in mind in comparing values of

Table I. A comparison of $\Delta\langle r^2\rangle/\langle r^2\rangle$ as obtained with muonic atoms and Mössbauer techniques.

I Isotope	II $2^+ \to 0^+$ energy (keV)	III Comparison source	IV Exp. energy shift ΔE_{obs} (eV)	V Center-of-gravity shift[a] $\Delta E_{c.g.}$ (eV)	VI ΔEisomer (eV)	VII $\frac{\Delta\langle r^2\rangle}{\langle r^2\rangle}\times 10^4$ muonic	VIII $\frac{\Delta\langle r^2\rangle}{\langle r^2\rangle}\times 10^4$ Mössbauer
$_{60}Nd^{150}$	130.17	Coulomb excitation[b]	$+570 \pm 120$	-270	$+840 \pm 120$	$+5.8 \pm 0.8$	No measurement
$_{62}Sm^{152}$	121.78	$Eu^{152}(\beta^+)$ (EC)	$+560 \pm 60$	-360 -240	$+920 \pm 70$ $+800 \pm 70$	$+5.9 \pm 0.4$ $+5.1 \pm 0.4$	$+3.7$[c] $+4.8$[d]
$_{64}Gd^{154}$	123.07	$Eu^{154}(\beta^-)$	$+670 \pm 150$	-310	$+980 \pm 150$	$+5.9 \pm 0.8$	No measurement
$_{66}Er^{166}$	80.56	$Ho^{166}(\beta^-)$	-350 ± 150	-320	-30 ± 150	-0.16 ± 0.8	No measurement
$_{74}W^{182}$	100.10	$Ta^{182}(\beta^-)$	-320 ± 100	-290 -240	-30 ± 100 -80 ± 100	-0.13 ± 0.5 -0.33 ± 0.5	$+0.15$[c] $+1.2$[e]
$_{74}W^{184}$	111.20	Re^{184}(EC)	-340 ± 100	-315 -250	-25 ± 100 -90 ± 100	-0.11 ± 0.5 -0.40 ± 0.5	$+0.8$
$_{74}W^{186}$	122.57	Re^{186}(EC)	-350 ± 100	-340 -250	-10 ± 100 -100 ± 100	-0.04 ± 0.5 -0.50 ± 0.5	No measurement

[a]The values of the center-of-gravity shift for Sm^{152}, Er^{166}, W^{182}, W^{184}, and W^{186} are taken from A. Gal, L. Grodzins, and J. Hüfner, preceding Letter [Phys. Rev. Letters 21, 453 (1968)]. Those for Nd^{150} and Gd^{154} are calculated by Hüfner (private communication) using the value 0.2 for the feeding factor. The uncertainty is estimated to be ±10%.

[b]The Coulomb excitation on Nd^{150} was carried out in the 8-MeV He^{++} beam of the Pegram Van de Graaff accelerator by E. Greenbaum, F. Hsu, Z. Y. Chow, and R. Howes. The calibration lines used were the 137.16- and 122.6-keV lines of Re^{186}.

[c]Ref. 6.

[d]L. Grodzins et al., Phys. Rev. Letters 18, 791 (1967). Recently S. Hüfner has measured the 5f shielding effect in solids by observing the isotope shifts of Eu^{151} and Eu^{153} in CaF_2 and found an increase of the electron density by a factor of 2.09 compared with the free atom. Hence $\Delta\langle r^2\rangle/\langle r^2\rangle = +10\times10^{-4}/2.09 = +4.8\times10^{-4}$.

[e]Ref. 5.

$\Delta\langle r^2\rangle$ derived from muonic and Mössbauer measurements.

(2) The second mechanism for obtaining an energy shift due to the magnetic hfs is described by the term $\Delta E_{c.g.}$. The 2^+ nuclear state splits into an unresolved hyperfine doublet ($F=\frac{5}{2},\frac{3}{2}$) when coupled to the $1s_{1/2}$ muon. The $\frac{5}{2}$ state lies several hundred eV above the $\frac{3}{2}$ state. If these states were statistically populated, the center of gravity of the unresolved doublet would not be shifted. The theoretical analysis, however, indicates that because of the dynamic interaction in the muonic $2p$ states, the magnetic sublevels are not statistically populated, which would result in a slight upward shift of the center of gravity. However, Gal, Grodzins, and Hüfner[9] have shown that the $\frac{5}{2}$ state decays mainly to the $\frac{3}{2}$ state via a strong Auger transition, and not directly to the ground state ($F=\frac{1}{2}$). This intense $M1$ intradoublet transition has the effect of lowering the observed energy, since most of the radiative de-excitation takes place from the $F=\frac{3}{2}$ substate. The resulting energy shift is comparable with the observed negative energy shifts in Er^{166} and the W isotopes.

(3) Since the $1s$ muon wave function is not constant over the nuclear volume in high-Z atoms, the binding energy of the $1s$ muon depends not only on the nuclear mean square radius ($\langle r^2\rangle$), as in electronic atoms, but on finer details of the charge distribution. ΔE_{isomer} is then the difference of the binding energies due to the difference of the two distributions. We have represented the charge distribution in each state by a three-parameter deformed Fermi distribution, and have assumed that the difference between these distributions may be described by a change of a single parameter.

The value of the ΔE_{isomer} (Table I, column VI) is obtained by correcting the observed energy shift ΔE_{obs} for the center-of-gravity shift $\Delta E_{c.g.}$ due to the intense $M1$ intradoublet transition, using the estimates of Gal, Grodzins, and Hüfner, neglecting ΔE_{pol}. The Dirac equation is

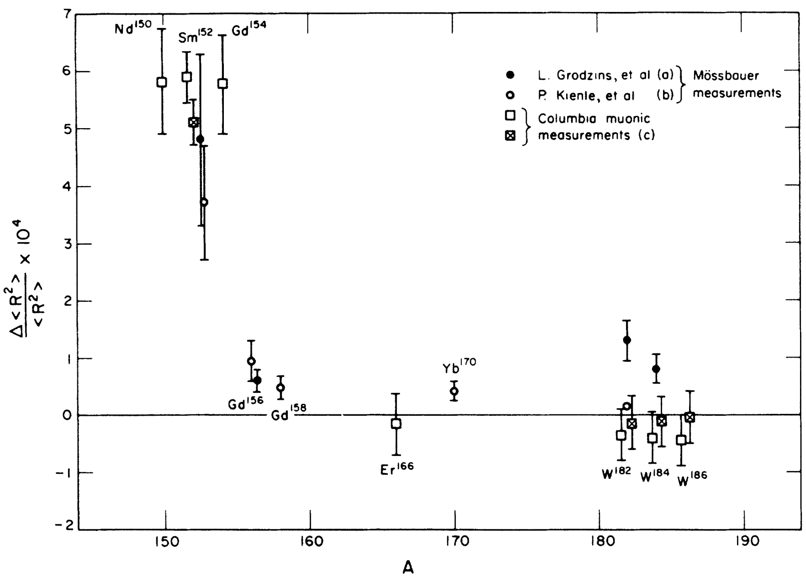

FIG. 2. $\Delta\langle r^2\rangle/\langle r^2\rangle$ plotted against mass number A for several even-even nuclei in the rare-earth region. (a) Table I and Refs. 4 and 5; (b) Ref. 6; (c) the two values of $\Delta\langle r^2\rangle/\langle r^2\rangle$ shown for Sm^{152} represent two values of $\Delta E_{\text{c.g.}}$ corresponding to two different measurements of the g factor of the 2^+ state. The two values of $\Delta\langle r^2\rangle/\langle r^2\rangle$ shown for each of the W isotopes represent two values of $\Delta E_{\text{c.g.}}$ corresponding to N_{I} or O_{I} electron shell conversion (Ref. 9).

solved to obtain the 1s muon binding energy for several sets of values of the half-density radius (c), skin thickness (t), and deformation (β). From the values of ΔE_{isomer}, we then find the corresponding changes in each of the parameters c, t, or β while the other two are held constant. Identical values of $\Delta\langle r^2\rangle$ (within the experimental uncertainty) are derived from any one of the parameter changes obtained in this manner. The values of $\Delta\langle r^2\rangle/\langle r^2\rangle$ obtained in this way are listed in column VII of Table I. Values of $\Delta\langle r^2\rangle/\langle r^2\rangle$ obtained by Mössbauer methods are shown in column VIII.

The $\Delta\langle r^2\rangle/\langle r^2\rangle$ from both methods have been plotted versus mass number in Fig. 2. It is interesting to see that the agreement between them is good. The three nuclei Nd^{150}, Sm^{152}, and Gd^{154}, which occur at the onset of large nuclear deformation at neutron number 90, show approximately equally large increases in mean-square charge radius. As these nuclei are considered "soft" rotators, this large centrifugal stretching may be anticipated. E^{166} and the three W isotopes are examples of "rigid" rotators; their values of $\Delta\langle r^2\rangle/\langle r^2\rangle$ are an order of magnitude less than that of the soft rotators. The observed values of $\Delta\langle r^2\rangle/\langle r^2\rangle$ are only a small fraction of the value ($\sim 4\times 10^{-4}$) estimated in the framework of the rotation-vibration-coupling model.[10]

When the weakening of pairing correlations and the increase of deformation due to rotation are taken into account in calculating the isomer effect,[4,11] the estimated $\Delta\langle r^2\rangle$ are only in good accord with the observed values for the soft rotators, but greatly overestimated again for the rigid rotators.

We wish to thank Dr. G. Rogosa for his constant interest and his help in procuring the enriched isotopes. We are greatly indebted to Dr. J. Hüfner and Professor L. Grodzins for communicating their results to us prior to publication. The valuable assistance of Dr. R. C. Barrett and Dr. M. Y. Chen with some of the theoretical considerations was most helpful.

*Work partially supported by the U. S. Atomic Energy Commission and National Science Foundation.

[1]S. Bernow, S. Devons, I. Duerdoth, D. Hitlin, J. W. Kast, E. R. Macagno, J. Rainwater, K. Runge, and C. S. Wu, Phys. Rev. Letters 18, 787 (1967).

[2]L. Wilets, Kgl. Danske. Vindenskab. Selskab, Mat.-Fys. 29, No. 3 (1954).

[3]B. A. Jacobsohn, Phys. Rev. 96, 1637 (1954).

[4]E. R. Marshalek, Phys. Rev. Letters 20, 214 (1968).

[5]D. Yeboah-Amankwah, L. Grodzins, and R. B. Frankel, Bull. Am. Phys. Soc. 12, 580 (1967).

[6]P. Kienle, W. Heming. G. Kaindl, H. J. Körner, H. Schaller, and F. Wagner, J. Phys. Soc. Japan 24, 207 (1968).

[7]This type of systematic shift would account for the discrepancy between the present measurement in Sm^{152}

and that reported in Ref. 1.
[8]M. Y. Chen, private communication; and thesis, Princeton University (unpublished).
[9]A. Gal, L. Grodzins, and Jörg Hüfner, preceding Letter [Phys. Rev. Letters 21, 453 (1968)].
[10]A. Fassler and W. Greiner, Z. Physik 168, 425 (1960), and 170, 105 (1962), and 177, 190 (1964).
[11]J. Krumlinde and S. G. Nilsson, J. Phys. Soc. Japan 24, 641 (1968). Also see Hyperfine Structure and Nuclear Radiations, edited by E. Matthias and D. A. Shirley (North-Holland Publishing Company, Amsterdam, The Netherlands, 1968).