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Experimental Studies of the Neutron-Deficient Gadolinium Isotopes. II. Gd^{145m}

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The N=81 isomer Gd^{145m} is characterized as having a half-life of 85 ± 3 sec and an M4 isomeric transition of 721.4 ± 0.4 keV. It also has a direct β^+/ϵ branch to the $h_{11/2}$ state at 716.1 keV in Eu¹⁴⁵. The intensity of this branch is 4.7% of the decay, implying a log*ft* of 6.2. The M4 transition probability is calculated and compared with the trends among other isomeric transitions in this region.

I. INTRODUCTION

Gadolinium isotopes cover a wide range of nuclear types, extending from permanently deformed nuclei to spherical single closed-shell nuclei at N = 82. As a result, systematic studies of their decay properties and structures should prove quite rewarding, for here is one of the few regions in the nuclidic chart where one can follow trends in nuclear states when moving from one extreme nuclear type to another. We have embarked recently on such a systematic study. The first paper in this series described the electron-capture decay of Gd¹⁴⁹, a nucleus that lies midway between the spherical and spheroidal regions.¹ On the neutron-deficient side of N = 82 the Gd isotopes have not been very well characterized until quite recently, although their decays present some interesting

anomalies, such as the peculiar ground-state decay of Gd¹⁴⁵ into what appear to be three-quasiparticle states in its Eu¹⁴⁵ daughter, the subject of which will form the third paper in this series.² On the neutron-deficient side of N = 82 a systematic study of the odd-mass isotones also appears well worthwhile because of the appearance of long series of nuclear isomers having quite different and distinct decay properties. The longest series of these isomers, in the N = 81 nuclei, extended from Te¹³³ to Sm¹⁴³, and it seemed reasonable that Gd¹⁴⁵, as the next nucleus in line, should also exhibit isomeric states.

We subsequently observed the metastable state in Gd¹⁴⁵ and its isomeric transition. The energy of the transition was found to be 721.4±0.4 keV and the half-life of the state, 85 ± 3 sec. These values were consistent with our predictions based on the systematics of the other N = 81 isotones, and they were first reported in November 1968.³ Since that time, Jansen, Morinaga, and Signorini⁴ have published results in very good agreement with our γ -ray energy and half-life values. Since our first preliminary report we have also observed the conversion electrons from the isomeric transition, clearly identifying it to be of M4 multipolarity, and we have observed a β^+/ϵ branch from Gd^{145m} directly to states in Eu¹⁴⁵.

Even by 1951, some 77 nuclear isomers had

been classified by Goldhaber and Sunyar.⁵ Since then, of course, isomers have been one of the prominent nuclear properties used to test the validity of nuclear models. In particular, *M*4 transitions are of interest for testing the extreme singleparticle model. If such a thing as a "pure" singleparticle transition exists, these transitions are good candidates for that distinction. The *M*4 transitions observed in the N = 81 nuclei are thought to proceed from $h_{11/2}$ to $d_{3/2}$ states, and the $h_{11/2}$ state should be particularly pure owing to its being the only odd-parity high-spin state at low excitations. We discuss the properties of these *M*4 transitions in Sec. III.

II. EXPERIMENTAL

We produced Gd^{145m} in this laboratory by both the Sm¹⁴⁴(τ , 2n)Gd^{145m} and the Sm¹⁴⁴(α , 3n)Gd^{145m} reactions. The calculated Q values for these reactions were -10.4 and -30.9 MeV, respectively.⁶ For all of these experiments, separated isotope Sm¹⁴⁴ (95.10%, obtained from Oak Ridge National Laboratory) in the form of Sm₂O₃ was used as the target material. The τ and α beams, typically 20 and 40 MeV, respectively, were furnished by the Michigan State University (MSU) sector-focused cyclotron. Excitation functions were run to determine the energy for maximum Gd^{145m} yield in each

FIG. 1. Singles γ -ray spectrum from 85-sec Gd^{145m}. The 721.4-keV peak is the M4 isomeric transition, while the 386.6- and 329.5-keV peaks come after a direct β^+/ϵ branch from Gd^{145m} to the $h_{11/2}$ state in Eu¹⁴⁵. The unlabeled peaks result from the decay of 21.8-min Gd^{145g}.



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case. Most of our experiments were performed with the τ beam, and typically a 10-mg or smaller target would be bombarded with a $0.5-\mu A$ beam for 1 min. Because of the short half-life of Gd^{145m}, no chemical separations could be carried out. Fortunately, they proved to be unnecessary, owing to the cleanness of the reactions. After most bombardments it took less than 2 min to retrieve the target and transport it to the counting area.

We also produced Gd^{145m} in a set of confirming experiments performed at the Yale University heavy-ion accelerator. C¹² beams ranging between 70 and 120 MeV were used, and the reactions of interest were Nd¹⁴²(C¹², $\alpha 5n$)Gd^{145m} and Sm¹⁴⁴-(C¹², $2\alpha 3n$)Gd^{145m}. The latter was discovered quite by accident and has an unexpectedly large cross section. It must proceed by a combination of cluster stripping and compound-nucleus formation.

The γ -ray energies were determined by simultaneous counting with the standards listed in Table I. A γ -ray singles spectrum is shown in Fig. 1. The peaks appearing in this spectrum without energy assignments listed come from the decay of the ground state of Gd¹⁴⁵.² The detector used was a five-sided trapezoidal Ge(Li) detector fabricated in this laboratory. It has an active volume of \approx 7 cm³ and a resolution of 2.9 keV full width at half maximum (FWHM) for the Co⁶⁰ 1.333-MeV γ ray. The γ rays associated with the decay of Gd^{145m} had energies of 721.4±0.4, 386.6±0.3, and 329.5±0.3 keV, as determined from the averages of a number of experiments.

The Gd^{145m} half-life was determined with the help of a computer code called GEORGE.⁷ This code allows us to accumulate data through an 8192-channel analog-to-digital converter interfaced to the MSU Cyclotron Laboratory Sigma-7 computer. We can dump segments of the spectrum in successive time intervals: Counting can be stopped, the spectrum segment dumped onto a rapid access disk

ΓABLE	I.	γ -ray	energy	stand	lard	s
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Nuclide	γ-ray energies (keV)	Reference		
Ce ¹⁴¹	145.43 ± 0.02	a		
Cm^{243}	209.85 ± 0.06	b		
	228.28 ± 0.08	b		
	$\boldsymbol{277.64 \pm 0.02}$	b		
Co^{56}	846.4 ± 0.5	С		
	1038.9 ± 1.0	с		
	1238.2 ± 0.5	с		

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TABLE II. Transition data summary for Gd^{145m} .

	This work	Jansen, Morinaga, and Signorini (Ref. 4)
$E_{\gamma}(M4)$ $E_{\gamma}(M2)^{a}$ $E_{\gamma}(M1)^{a}$ $t_{1/2}$	$721.4 \pm 0.4 \text{ keV}$ $386.6 \pm 0.3 \text{ keV}$ $329.5 \pm 0.3 \text{ keV}$ $85 \pm 3 \text{ sec}$	$721.3 \pm 0.7 \text{ keV}$ $85 \pm 7 \text{ sec}$

^aThese transitions occur in Eu¹⁴⁵ and result from the direct feeding of the $h_{1/2}$ state in that nucleus by Gd^{145m} The multipolarities are assumed from the properties of the states as determined from Gd^{145g} decay and scattering. Cf. Refs. 2 and 12.

(for punching on cards at a later time), the memory erased, and the counting resumed, all in considerably less than one second of elapsed time. In the present case the spectra were dumped at 15sec intervals. A pulser peak was included in each spectrum for later determination of the proper dead-time corrections. The net peak areas, corrected for dead time, then yielded the half-life information in the usual manner. (A set of these spectra, together with the half-life curves can be found in Eppley.⁸) A listing of our E_{γ} and half-life values is given in Table II, where they are compared with those of Jansen, Morinaga, and Signorini.

The electron spectra were obtained by use of a $1000-\mu$ -thick Si(Li) surface-barrier detector, cooled to methanol-dry-ice temperature and operating with a bias of +200 V. The resolution of this detector was typically 5 keV FWHM in the 600-keV region, the electrons having passed through a 0.25-mil Havar window. A resulting electron spectrum is shown in Fig. 2.

In order to arrive at a value for the conversion coefficient of the isomeric transition, the γ -ray and electron spectra were measured simultaneous-



FIG. 2. Electron spectrum showing conversion lines from the 721.4-keV M4 isomeric transition in Gd¹⁴⁵. This spectrum was taken with a 1000- μ Si(Li) detector mounted in a dipstick cryostat with a 0.1-mil Havar window.

ly from the same source, which was placed in a fixed, reproducible geometry. Again, owing to the short half-life, "mass-free" sources could not be made. However, as can be seen from the electron spectrum, our "thick" sources led to a minimum of straggling. For calibrating the detector efficiencies and the geometry corrections, a Cs¹³⁷ source was used as a standard. A value of 0.094 was used for the α_K of its 661.6-keV transition; this is an average of published values.^{9, 10}

Two separate experiments made at widely differing times were performed to determine the Gd^{145m} K- and L-internal-conversion coefficients. The results, compared with theoretical values for various multipolarities, are shown in Table III. The logarithms of the theoretical values were interpolated from a quadratic least-squares fit to the tabulated values of Hager and Seltzer.¹¹ The experimental α_K value definitely shows the isomeric transition to be *M*4 in character. The measured K/L ratio places it as being either *M*3 or *M*4. The former is a more sensitive test, however, and an *M*4 assignment fits in quite well with the systematics of transition probabilities in the N = 81 isotones, as we shall see in the next section.

We shall see later that the 386.6- and 329.5-keV γ peaks fit between known^{2, 12} states and imply that 4.7% of the decay of Gd^{145m} goes via a direct β^+/ϵ branch to an $h_{11/2}$ state in Eu¹⁴⁵, the other 95.3% going via the isomeric transition. The 4.7% branch was determined by correcting the 386.6-keV/721.4-keV photon ratio (found to be 0.048) for conversion, again using the conversion coefficients of Hager and Seltzer and assuming the 386.6-keV transition to be a pure M2 transition. The 329.5-keV γ rays result also from the decay of the ground state of Gd¹⁴⁵, so no quantitative information about their intensity could be obtained from these experiments.

III. $Gd^{145 m}$ AND N = 81 M4 ISOMERS

Figure 3 shows our decay scheme for Gd^{145m} . While the other known N = 81 odd-mass isotones have been assigned $d_{3/2}$ ground-state configurations, there is some evidence² that in Gd^{145} the $s_{1/2}$ state has replaced the $d_{3/2}$ state as the ground state, which may account partly for the peculiar

TABLE III. Conversion coefficients for the isomeric transition in Gd^{145} .

		Theoretical (Ref. 11)			
	Experimental	E3	E4	M3	M4
$\alpha_K \\ \alpha_K / \alpha_L$	0.12 ± 0.2 5.4 ± 0.7	$\begin{array}{c} 0.011\\ 3.50\end{array}$	0.024 3.53	0.054 5.77	0.118 4.88



ground-state decay of Gd¹⁴⁵. Newman *et al.*,¹² also came to this conclusion when interpreting their scattering data from the Sm¹⁴⁴(τ , d)Eu¹⁴⁵ reaction in conjunction with their Gd^{145g} decay data. If the $s_{1/2}$ assignment is correct, the *M*4 isomeric transition in Gd¹⁴⁵ does not proceed directly to the ground state but to a state ΔE in energy above the ground state. This excited $d_{3/2}$ state would then decay to the ground state via a predominantly *M*1 transition.

In a search for a low-energy M1 transition, we have utilized a Si(Li) x-ray detector, which is useful in the 5–100-keV energy region for γ rays and is also sensitive to charged particles. No peaks that are in evidence in any of our spectra in the 10-100-keV range can be attributed to the decay of Gd^{145m}. The high positron flux, primarily from Gd^{145g} decay, contributed to a high background problem below 10 keV and ruled out observing a transition in that region. Assuming such a lowenergy transition to be present, most likely M1 in character, it would be converted primarily in the L_I shell, with α_{L_I} having a value of 4.5 at 46 keV and increasing to 246 at 9.38 keV.¹¹ Consequently, assuming the transition to be low in energy, say, 20 keV or less, would preclude observing the photons. However, we should have been able to see the L_I conversion line if the energy were above 10 keV.

The radial matrix elements for the M4 transitions in the N = 81 odd-mass isotones were calculated using Moszkowski's¹³ approximations for a single neutron hole,

$$T_{(ML)}^{SP} = \frac{2(L+1)}{L[(2L+1)!!]^2} \omega \frac{e^2}{\hbar c} \left(\frac{R_0}{c}\right)^{2L} \left(\frac{\hbar}{mcR_0}\right)^2 (\mu_N L)^2 \times |M|^2 S(j_i, L, j_f),$$

where L is the multipolarity of the transition, R_0 is the effective nuclear radius, $S(j_i, L, j_f)$ is a statistical factor (i.e., angular momentum portion of the matrix element), which for $\frac{11}{2} - \frac{3}{2}$ transitions has the value $\frac{15}{11}$, and μ_N is the dipole moment of the neutron. Symbolically, $|M|^2$ has the form,

$$|M|^2 = \left[\int_0^\infty R_f \left(\frac{r}{R_0}\right)^{L-1} R_i r^2 dr\right]^2.$$

Our results are plotted in Fig. 4, where we also show the differences in energy between the $h_{11/2}$ and $d_{3/2}$ states.

The resulting transition probabilities are consistently smaller than the approximation of a constant wave function,

$$|M|^2 = \left(\frac{3}{L+2}\right)^2 (\mu_N L)^2 = 14.6,$$

but this fact should not concern us particularly, for M4 transitions are customarily retarded over such estimates and one needs much more detailed information about the nuclear wave functions in order to make detailed comparisons meaningful. What is of more importance is the fact that the values of $|M|^2$ are not constant but show a definite trend in this series of isotones. [It is unusual for $|M|^2$ not to be constant over such a series. For example, in the odd-mass neutron-deficient lead isotopes. $|M|^2$ was constant to the point that an apparent 15% discrepancy at Pb²⁰³ suggested that an unobserved transition was competing with the M4 isomeric transition, and this competing transition was subsequently discovered.¹⁴ Examinations of more of these series of isomers can be found in Ref. 8.

The complete answer as to why the discrepancy in magnitude and in trend exists between the experimental and theoretical values is still not forthcoming. Kotajima¹⁵ summarizes several effects that could contribute to these deviations: (1) cancellation of the magnetic moments because of the mesonic effect, (2) configuration mixing or spin polarization, or (3) the use of too crude an approximation for the radial wave functions. In the same paper he explores the usefulness of applying a more realistic function for these radial wave functions, but the matrix elements were remarkably insensitive to changes in these wave functions. This effect, then, by itself could not explain even the deviations in magnitude, much less the "bowing" of the curve as seen in Fig. 4. More recently, Jansen, Morinaga, and Signorini⁴



FIG. 4. Plots of the $h_{11/2}$ - $d_{3/2}$ energy differences and of the squares of the radial matrix elements for the M4 isomeric transitions that connect these states in the N=81 odd-mass isotones.

have also sought to better the experimental-theoretical agreement. In a calculation similar to that used by Kotajima, they recalculated the radial matrix elements using a potential function consisting of a Woods-Saxon part and a spin-orbit part. In this manner they recalculated the radial part of the Schrödinger equation for these nuclei. In addition to this refinement, they took account of configuration mixing by utilizing the results of Horie and Oda.¹⁶ In this manner, much of the Z dependence was removed. However, the experimental values are still lower than the theoretical ones for all reasonable values of R_0 . For an R_0 of 1.22 F, the experimental-to-theoretical ratio hovers around 0.5.

An alternate attack on this problem might prove to be useful. Recasting the problem in a quasiparticle framework would allow a simplified picture, with the transition probabilities depending only on the particle-hole character of the states involved. Stripping and pickup reactions are just now beginning to be carried out on nuclei in this region, so occupation numbers may very well become available from these before long. As soon as such information is available, this type of formulation can be explored more fully.

Finally, it should be noted that as Z increases there is an increase in the direct β^+/ϵ decay to the $h_{11/2}$ states in the daughter nuclei. On the neutron-deficient side of the N = 81 odd-mass isotones, Ce^{139m} does not have sufficient decay energy to populate such a state in La¹³⁹, but the decay is energetically possible for Nd^{141m}, Sm^{143m}, and Gd^{145m}. We have made a careful search for direct

population of the $h_{11/2}$ state in Pr¹⁴¹ by Nd^{141m} decay^{8, 17} and have been able to set an upper limit of 0.01% on any such population. This results in a $\log ft$ greater than 7. From Sm^{143m} decay, however, Feldsteiner and Rosner¹⁸ were able to detect a direct branch of 0.2% to the state in Pm¹⁴³, implying a $\log ft$ of 6.7. And we find a direct branch of 4.7% from Gd^{145m} decay, resulting in a log ft of only 6.2. We consider this to be an indication of the increased occupation of the $h_{11/2}$ orbit by proton pairs as one moves up the series. A more detailed consideration of this phenomenon,

however, is beyond the scope of this paper and will be the subject of another paper to be forthcoming shortly.¹⁹

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Higher-Order Deformations and Electric Monopole Transitions in Deformed Nuclei*

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Calculation of electric monopole X ratios in deformed nuclei using the higher-order shape deformations Y_4 and Y_6 yields much better agreement with measured values than is obtained when only pure quadrupole surfaces are assumed. Use was made of recently measured values of β_4 , and β_6 .

I. INTRODUCTION

For about a decade now it has been known that the axially symmetric rotor model with quadrupole (β) vibrations is unable to predict the ratio of the

strengths of the electric monopole (E0) to electric quadrupole transitions depopulating the β bands in rare-earth nuclei.¹ More recent and complete discussions of E0 transitions in this region have shown that both the axially symmetric model² and

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