# Coulomb Excitation of Tc<sup>99</sup> and Sm<sup>147</sup>

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Coulomb excitation gamma-ray spectra from  $Tc^{99}$  and  $Sm^{147}$  have been investigated by means of 2-MeV protons. A NaI(Tl) scintillation spectrometer was used to measure the energy and intensity of the gamma rays. Gamma rays of energy  $120\pm3.2$  keV and  $199\pm4.1$  keV were observed from the excited nucleus  $Sm^{147}$ . In the case of  $Tc^{99}$ , gamma rays of energy  $140\pm3.5$  keV and  $178\pm4.0$  keV were observed. Total reduced transition probabilities for these transitions are of the same order of magnitude as the single-particle estimate.

#### 1. INTRODUCTION

THE method of Coulomb excitation has been used extensively to explore the properties of the lowlying excited states throughout the mass table.<sup>1</sup> Coulomb excitation studies have provided a striking confirmation of the collective nuclear model of Bohr and Mottelson.<sup>2</sup> This model has been successfully used to describe the properties of nuclei in regions between closed nuclear shells.

In the present work we report on the Coulomb excitation of two nuclei situated near closed shells:  $Tc^{99}$  and  $Sm^{147}$ . The nucleus  $Tc^{99}$  contains 56 neutrons and is near the closed shell at N=50;  $Sm^{147}$  contains 85 neutrons and is near the closed shell at N=82. The method of Coulomb excitation has not been used to excite  $Tc^{99}$ previous to this investigation, while  $Sm^{147}$  has been studied by Heydenburg and Temmer<sup>3</sup> who observed no gamma rays resulting from the Coulomb excitation of this nucleus. Since the transition probabilities and energy levels of the low-lying energy level structure of  $Ta^{181}$  are well known, this nucleus was included in the study as a check on the techniques used.

## 2. EXPERIMENTAL PROCEDURE

The source of projectiles was the University of North Carolina 2-MeV Van de Graaff generator. A technetium target was obtained from the Oak Ridge National Laboratory. It consisted of about 5  $\mu$ Ci of Tc<sup>99</sup> on a nickel backing. Samarium was obtained as an oxide powder 97.8% enriched in Sm<sup>147</sup>, and a nickel-backed target was prepared by the method of Huus *et al.*<sup>4</sup> The target was observed at an angle of 125°, where the value of the Legendre polynomial  $P_2$  in the gamma-ray angular distribution is near zero. After a Tc<sup>99</sup> or Sm<sup>147</sup> target was exposed to the beam, the target holder was rotated to expose a tin foil, and the bremsstrahlung spectrum from this foil was subtracted from the accumulated data for an equal exposure of integrated beam current.

Gamma rays had to pass through a 5-mil aluminum window and an x-ray shield before they entered the detector, a  $1\frac{1}{2}$ -in.×2-in. NaI(Tl) crystal mounted on a DuMont 6292 photomultiplier tube. The average separation between target and detector was  $1\frac{1}{2}$  in. The detector was shielded from machine background by 2 in. of lead on all sides. Pulses from the photomultiplier were preamplified and sorted by a RCL 128-channel pulseheight analyzer.

The energy response of the spectrometer was calibrated with the following gamma-ray sources: Co<sup>57</sup>, Na<sup>22</sup>, Cs<sup>137</sup>, and Mn<sup>54</sup>. The spectrometer was calibrated for absolute gamma-ray detection efficiency by means



FIG. 1. Gamma-ray spectrum from Tc<sup>99</sup> with spectrum. from NaCl shown for comparison.

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<sup>&</sup>lt;sup>1</sup> K. Alder, A. Bohr, B. Mottelson, and A. Winther, Rev. Mod. Phys. 28, 432 (1956).

<sup>&</sup>lt;sup>2</sup> A. Bohr and B. Mottelson, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. **27**, No. 16 (1953).

<sup>&</sup>lt;sup>8</sup> N. P. Heydenburg and G. M. Temmer, Phys. Rev. 100, 150 (1955).

<sup>&</sup>lt;sup>4</sup> T. Huus, J. H. Bjerregaard, and B. Elbek, Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd. 30, No. 17 (1956).



of a  $Co^{57}$  standard source prepared by the ChemTrac Corporation. The error in the intensity measurements due to source calibration is estimated to be 10%.

## 3. RESULTS

#### A. The Nucleus Tc<sup>99</sup>

A Na<sup>23</sup> impurity proved troublesome in this target. With 2-MeV protons incident on the target, a backscatter peak from the 445-keV gamma ray due to a  $Na^{23}(p,p')Na^{23}$  reaction obscured whatever gamma rays from Coulomb excitation one might hope to observe. By varying the incident proton energy on a target of fused NaCl, the resonance structure of the 445-keV gamma-ray yield curve was explored. Because of resonances in the yield curve, it was found that by changing the incident proton energy we could reduce the gamma-ray peak from the Na<sup>23</sup> reaction by a factor of 2 or 3 relative to the gamma-ray peak resulting from the Coulomb excitation of the Tc<sup>99</sup> nucleus. The resulting gamma-ray spectrum when 1.8-MeV protons are used is shown in Fig. 1. The line shape when sodium alone is bombarded is shown for comparison. A  $140 \pm 3.5$ -keV gamma-ray peak is clearly seen, and there is a possibility that there is a small 178±4.0-keV gamma-ray peak rising above the backscatter peak. The excitation function for the 140-keV peak was found to follow the theoretical prediction for E2 Coulomb excitation. The excitation curve for the 178-keV peak was not measured.

There is a question of whether the observed 140-keV gamma ray is to be associated with a transition from the 140-keV level  $(\frac{7}{2}+)$  or the 142-keV level  $(\frac{1}{2}-)$  to the ground state  $(\frac{9}{2}+)$ . The energy-level diagram<sup>5</sup> for Tc<sup>99</sup> is reproduced in Fig. 2. The scintillation spectrometer does not have sufficient energy resolution to distinguish between the two resulting gamma rays. However, the multipolarity of the 142-keV excitation has to be M4 or E5 whereas the 140-keV level may be reached by an E2 excitation. The observed gamma ray can thus be safely

<sup>5</sup> Landolt-Börnstein Tables, Supplementary Volume on Nuclear Physics (Springer-Verlag, Berlin, 1961), 6th ed. assigned to a transition from the 140-keV level to the ground state. The observed 178-keV gamma ray may probably be assigned as a transition from the 181-keV level to the ground state.

#### B. The Nucleus Sm<sup>147</sup>

Gamma rays from Coulomb excitation of Sm<sup>147</sup> were of such low intensity that the bremsstrahlung background was roughly 4 times greater than the observed gamma-ray intensity. This necessitated a careful subtraction of the background from the experimental data. The gamma-ray spectrum from Sm<sup>147</sup> reduced in intensity by a 1.5-mm copper absorber is shown in Fig. 3. The yield of the  $120\pm3.2$ -keV peak follows the theoretical E2 Coulomb excitation function. An excitation curve was not measured for the  $199\pm4.1$ -keV peak.

The observed yields from oxide targets have been found to be less than from pure metal targets of the same element. Heydenburg and Temmer<sup>3</sup> found an empirical conversion factor of  $\frac{1}{2}$  for rare-earth oxides of the form  $X_2O_3$ . Because of this conversion factor, the observed yield from the samarium oxide target has been multiplied by 2.

By comparison of the energy level scheme, Fig. 4, and the usual selection rules, it is seen that E2 excitation is possible for both the first and second exicted states of the nucleus Sm<sup>147</sup>. The observed 120- and 199-keV



Nucleus	Transition energy (keV)	Yield of $\gamma$ per 10 <sup>10</sup> protons (2 MeV)	$\begin{array}{c} \text{Cross section} \\ \sigma(\gamma) \\ (\mu\text{b}) \end{array}$	${B(E2)/e^2\over (10^{-48}~{ m cm}^4)}$	${B_{ m sp}(E2)/e^2\over (10^{-48}~{ m cm^4})}$	$B(E2)/B_{\rm sp}(E2)$
Ta <sup>181</sup>	137 303	232 8.38	630 57	1.71 0.44	0.0303 0.0303	56 14.5
Sm <sup>147</sup>	120	10.3	27.5	0.034 <sup>b</sup> 0.038°	0.0232	1.5 <sup>ь</sup> 1.6°
	199	3.17	12.1	0.016	0.0232	0.73
Tc99	140 178	18.5ª 5.6ª	46 11.3	$0.035 \\ 0.021$	$0.0136 \\ 0.0136$	2.5 1.5

TABLE I. Summary of results.

These data were taken at 1.8 MeV.
<sup>b</sup> These data were computed in the limit of pure M1 radiation.
<sup>c</sup> These data were computed in the limit of pure E2 radiation.

gamma rays may clearly be assigned to a transition from the first and second excited states, respectively, to the ground state on the energy level diagram.

## 4. ANALYSIS OF DATA

From the measured value of the thick-target yield, one may calculate the E2 cross section by means of the equations given in Ref. 1. A thick-target correction has to be made to allow for changes in the yield of gamma rays as the charged particles become degraded in energy while traversing the target material. For the proton stopping power the data of Stelson and McGowan<sup>6</sup> have been used. An effective target thickness is given graphically<sup>1</sup> to an accuracy of about 5% and represents the ratio of the observed yield to that which would result if the excitation cross section and the stopping power were independent of the energy of the projectile and had values corresponding to the initial energy of the projectile.

The semiclassical theory of Coulomb excitation may be used to relate the E2 cross section to the partial reduced transition probability  $\epsilon [B(E2)/e^2]$ . The decay fraction  $\epsilon$  depends on the mixing ratio, the internal conversion coefficients, and the branching ratio. Values for the internal conversion coefficients were obtained from tables given by Rose.<sup>7</sup> The branching ratio equals unity



<sup>6</sup> P. H. Stelson and F. K. McGowan, Phys. Rev. 110, 489 (1958). <sup>7</sup> M. E. Rose, Internal Conversion Coefficients (North-Holland Publishing Company, Amsterdam, 1958).

for the first excited state. For Tc<sup>99</sup> the number of 181keV transitions has been found<sup>8</sup> to be about equal to the number of 41-keV transitions; i.e.,  $N_{181} \simeq N_{41}$ . For Ta<sup>181</sup> the ratio  $N_{303}$ :  $N_{165}$  has been taken<sup>1</sup> to be 1:3. The ratio  $N_{198}: N_{57}$  has been assumed to be unity for the nucleus Sm<sup>147</sup> in the calculations of the present work. It is known<sup>9</sup> that in the case of  $Tc^{99}$  the K/L ratios and conversion coefficients are consistent with the assignment of the 140-keV gamma ray as M1 radiation, and the 181-keV gamma ray as E2 radiation. This assignment determines the mixing ratio. The 121-keV gamma ray from the first excited state of Sm147 has been identified10 as some mixture of M1+E2 radiation, while the 198-keV gamma ray is E2 radiation.

Once the decay fraction  $\epsilon$  is known, the total reduced transition probability  $B(E2)/e^2$  may be found. The total reduced transition probability is estimated to have limits of error of 20% for Tc<sup>99</sup> and Sm<sup>147</sup>.

# 5. CONCLUSION

A summary of experimental and derived results is presented in Table I. This table contains the cross section and the total reduced transition probability derived from Coulomb excitation theory. Heydenburg and Temmer<sup>3</sup> observed no gamma rays from Sm<sup>147</sup> even though they had available an enriched sample of the rare earth. They set an upper limit for the intensity of any gamma rays from the Coulomb excitation of Sm<sup>147</sup> as being about 2% of the intensity of the gamma rays they observed in the even Sm isotopes. This is consistent with the value of the reduced transition probability for the first excited state of Sm<sup>147</sup> which is reported here; our value being less than 2% of the values Temmer and Heydenburg report for the even Sm isotopes. Where two values are given in the table the first corresponds to the

<sup>&</sup>lt;sup>8</sup> J. Ravier, P. Marguin, and A. Moussa, J. Phys. Radium 22, 249 (1960).

<sup>9</sup> H. Médicus, D. Maeder, and H. Schneider, Helv. Phys. Acta

<sup>&</sup>lt;sup>10</sup> N. M. Anton'eva, A. A. Bashilov, B. S. Dzhelepov, K. G. Kaun, A. F. A. Meyer, and V. B. Smirnov, Zh. Eksperim. i Teor. Fiz. 40, 23 (1961) [translation: Soviet Phys.—JETP 13, 15 (1961)].

assumption of pure *M*1 radiation and the second to pure E2 radiation for the gamma rays.

The half-life for the decay of the second excited state of Tc<sup>99</sup> has been measured<sup>11</sup> to be  $(3.57 \pm 0.05) \times 10^{-9}$  sec. By means of the equation

$$(\tau_{1/2})^{-1} = 1.4T_{\gamma}(E2)(1+\alpha),$$

it is possible to relate the observed lifetime to the transition probability for gamma-ray emission  $T_{\gamma}(E2)$ . For the conversion coefficient  $\alpha$ , an experimental value of  $0.13 \pm 0.03$  has been found.<sup>8</sup> The transition probability thus obtained may be converted to a reduced transition probability by a simple calculation.<sup>1</sup> The cross section for Coulomb excitation and the corresponding radiative lifetime thus provide two independent measurements of B(E2). From the radiative lifetime, a value of 0.045  $\times 10^{-48}$  cm<sup>4</sup> is obtained for  $B(E2)/e^2$ , while the Coulomb excitation data reported here result in a value of  $0.021 \times 10^{-48}$  cm<sup>4</sup>. The Coulomb excitation reduced transition probability is smaller by a factor of 2 which

<sup>11</sup> E. Bodenstedt, E. Matthias, and H. J. Körner, Z. Physik 153, 423 (1959).

reflects the limited accuracy of the yield determination for the radiation from the second excited state.

The Weisskopf single-particle estimate<sup>12</sup> for the reduced transition probability is given by

$$B_{\rm sp}(E\lambda)/e^2 = rac{S}{4\pi} \left(rac{3}{3+\lambda}
ight)^2 R_0^{2\lambda}$$
,

with  $R_0 = 1.2A^{1/3}$  F, and S a statistical factor which depends on the spins of the initial and final states involved and the multipole order  $\lambda$ . In the present work S has been somewhat arbitrarily taken<sup>1</sup> to be  $(2\lambda+1)$ .

The nucleus Ta<sup>181</sup> is typical of strongly deformed nuclei for which one finds reduced transition probabilities of 10-100 times the Weisskopf single-particle estimate. The reduced transition probabilities from Sm<sup>147</sup> and Tc<sup>99</sup> are comparable in magnitude to the single-particle estimate.

The authors wish to thank Dr. E. Merzbacher for helpful comments

<sup>12</sup> J. M. Blatt and V. F. Weisskopf, Theoretical Nuclear Physics (John Wiley & Sons, Inc., New York, 1955).

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# Positron Decay of Co<sup>58</sup><sup>+\*</sup>

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The allowed positron transition from the  $Co^{58}$  ground state (2+) to the Fe<sup>58</sup> first excited state (2+, 810 keV) was studied using a  $4\pi$  positron-scintillation spectrometer. The experimental shape factor exhibits a small systematic deviation from that theoretically predicted for an allowed transition and corresponds to an excess of low-energy positrons. The experimental shape factor can be fit by a curve proportional to (1+b/W) with b=0.3. The Fermi-Kurie plot is linearized by this correction factor and yields an end-point energy of 474±5 keV. Similar discrepancies between the experimental and theoretically predicted shapes of Gamow-Teller beta transitions for negatrons (In<sup>114</sup>, P<sup>82</sup>, and Y<sup>90</sup>) and positrons (Na<sup>22</sup>) have been reported. In these cases it was found that this same shape factor with 0.2 < b < 0.4 would linearize the theoreticallycorrected Fermi-Kurie plots. No satisfactory explanation for this effect has been offered. As previously suggested by other investigators, the shape factor (1+b/W) must, for the present, be regarded as a purely empirical correction. The end-point energy from the present investigation combined with the recent highprecision measurement of the gamma-ray transition energy from the first excited state to the ground state of Fe<sup>58</sup> yields a Co<sup>58</sup>-Fe<sup>58</sup> mass difference of 2306±5 keV.

## INTRODUCTION

**C**MALL discrepancies between the experimental and  $\mathbf{\mathfrak{I}}$  theoretical shapes of certain beta spectra have been reported by various investigators.<sup>1</sup> Johnson, Johnson,

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<sup>1</sup> For a comprehensive review and discussion of the measure-

and Langer<sup>2</sup> have reported small systematic deviations from linearity in the theoretically-corrected Fermi-Kurie (F-K) plots for the Gamow-Teller (G-T) electron transitions in In<sup>114</sup>, P<sup>32</sup>, and Y<sup>90</sup>. In each case, the F-K plot deviated from linearity in a manner corresponding to an excess of low-energy electrons, and it could be linearized

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<sup>\*</sup> This report is based in part on a thesis submitted by J. I. Rhode to the faculty of Purdue University in partial fulfillment of the requirements for the degree of Ph.D. in Physics. See also, O. E. Johnson and J. I. Rhode, Bull. Am. Phys. Soc. 8, 74 (1962). \* National Science Foundation Cooperative Fellow, September

ment of beta spectra, in general, and these deviations, in particular, see L. M. Langer, in *Proceedings of Rehoroth Conference on Nuclear Structure*, (North-Holland Publishing Company, Amsterdam, 1958), p. 437. <sup>2</sup> O. E. Johnson, R. G. Johnson, and L. M. Langer, Phys. Rev. **112**, 2004 (1958). See also, O. E. Johnson, Ph.D. thesis, Indiana University, 1956 (L. C.:Mic. 56-3064, University Microfilms, Inc., Ann Arbor, Michigan).