transition (with the same experimental accuracy as in Method 1). As this result is about 78% of the previously determined value, it suggests that nonradiative transitions plus transitions to the $^4I_{15/2}$ and $^4I_{13/2}$ multiplets make up about one-fifth of the total drain from the $^4F_{3/2}$ levels. The radiative laser-transition lifetime corresponding to the above cross section is 758 μsec.

Either of the above two techniques can also be used with other four-level systems. The second method, while undoubtedly more accurate, requires that a reasonable percentage of the upper-state population decay directly to the ground state (or to some state which has a reasonable Boltzmann population).

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Mössbauer Effect Following Coulomb Excitation in the Even-Even Isotopes of Ytterbium*

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Mössbauer levels in Yb^{172}, Yb^{174}, and Yb^{176} have been observed following the population of the first excited states by Coulomb excitation. Using oxide and metallic targets and absorbers, the quadrupole energy splittings and relative quadrupole moments were determined for each isotope. Coulomb-excitation cross-section data were used to determine the electric-field gradient at the nucleus for Yb^{3+} in Yb_2O_3.

I. INTRODUCTION

RECENT experiments have shown that Mössbauer effects can be observed when suitable nuclear levels are populated by Coulomb excitation[1–3] and by direct nuclear reactions.[4,5] The successful observation of the Mössbauer effect by accelerator-produced reactions significantly enlarges the list of possible nuclear levels in which the Mössbauer effect can be observed. The requirement that a nuclear level be populated in the decay of a radioactive parent with a suitably long half-life need no longer apply.

The rare-earth region contains many stable isotopes with suitable Mössbauer levels which are not accessible by β decay. Figure 1 shows seven stable ytterbium isotopes from Yb^{168} to Yb^{176} with eight possible levels for recoilless emission. Only a few of these states[6–8] have been studied by the conventional method using a radioactive parent. Of these seven isotopes, Yb^{171}, Yb^{172}, Yb^{174}, and Yb^{176} are sufficiently abundant in the naturally occurring ytterbium compounds to make use of enriched absorbers unnecessary. All of the possible ytterbium Mössbauer levels shown in Fig. 1 are collective, and most of them possess large Coulomb-excitation cross sections.[9]

Of special interest is the variation of the nuclear quadrupole moment and gyromagnetic ratio as one proceeds from one isotope to another of the same element. The present article deals with the observation of Mössbauer hyperfine spectra in the even-even isotopes Yb^{172}, Yb^{174}, and Yb^{176}, and the variation of the electric quadrupole moment among these isotopes. These data were compared with the Mössbauer spectrum for Yb^{170} which was obtained following β decay of Tm^{170}. Both oxide and metallic targets and absorbers were used.

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* Work supported in part by the U. S. Atomic Energy Commission.

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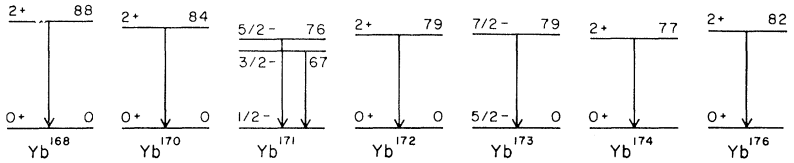

FIG. 1. Low-lying excited levels in the family of ytterbium isotopes.

Yb_2O_3 is paramagnetic, but owing to the short spin-lattice relaxation time of the Yb^{3+} ion, it exhibits only electric quadrupole hyperfine interaction. Ytterbium metal, being divalent and thus having an electronic ground state $^1S_0$, and crystallizing in a cubic lattice, exhibits no hyperfine interaction.[6] Therefore, an oxide target (absorber) and a metal absorber (target) make suitable combinations for observing the electric quadrupole splitting of ytterbium isotopes in Yb_2O_3. All the Mössbauer effects observed involved a $2^+ \rightarrow 0^+$ transition with no quadrupole splitting for the ground state. Each of the isotopes was studied with an oxide target and metallic absorber, or the reverse. Hence, the relative values of the energy splittings among isotopes, expressed as ratios, yield directly the ratio of the moments of the first excited states. The values obtained from earlier Coulomb-excitation cross-section data[10] for the intrinsic quadrupole moments are derived from the $E2$ transition rates and depend upon the validity of the collective model.[9] The ratios derived in this work are obtained directly from the Mössbauer hyperfine spectra, and therefore are model independent. Because only an energy splitting can be measured using the Mössbauer effect, independent information about the electric field gradient is necessary in order to extract an absolute measurement of the quadrupole moment. The difficulty in calculating the electric field gradient at the Yb nucleus for Yb_2O_3 prevents giving an absolute measurement of the quadrupole moment. Using the values of the intrinsic quadrupole moment derived from Coulomb-excitation cross-section data using the rotational model, a best value of the electric field gradient at the Yb nucleus in Yb_2O_3 was determined.

II. EXPERIMENTAL PROCEDURE

The first excited states of Yb^{172}, Yb^{174}, and Yb^{176} were populated by Coulomb excitation using a beam of 3-MeV protons obtained from the Johns Hopkins University 3-MeV Van de Graaff accelerator. Although the cross section is larger at 3 MeV for α particles than for protons, the gamma-ray yield for a thick target is reduced for α particles due to the large dE/dx losses of the α particle.[9] For the case of the oxide targets, a 0.2-μA beam was used. The gamma-ray spectrum for a Yb^{174} oxide target is shown in Fig. 2. A 1:1 signal-to-noise ratio was obtained in the counting window with a total counting rate of 600 counts/sec for a solid angle of approximately 5%. As can be seen from Fig. 2, the main source of noise in the energy-selection window for gamma rays is the Yb K x rays. For the case of the Yb^{174} metallic target, the counting rate in the gamma-ray window was 1500 counts/sec for a 0.1 μA beam when the detector subtended a 5% solid angle.

Since the energies of the first excited states for the even-even Yb isotopes lie between 76.5 and 84.3 keV,[11] it was necessary to cool both target and absorber to liquid-helium temperature in order to observe an appreciable Mössbauer effect. The apparatus used to achieve this is shown schematically in Fig. 3 and is described in detail elsewhere.[12] The velocity drive is the conventional drive described by Cohen,[13] and was operated with a constant-acceleration input. The velocity scale was calibrated using the six-line spectrum of Fe^{57}, taken with a Fe metal absorber versus a Co^{57} source in

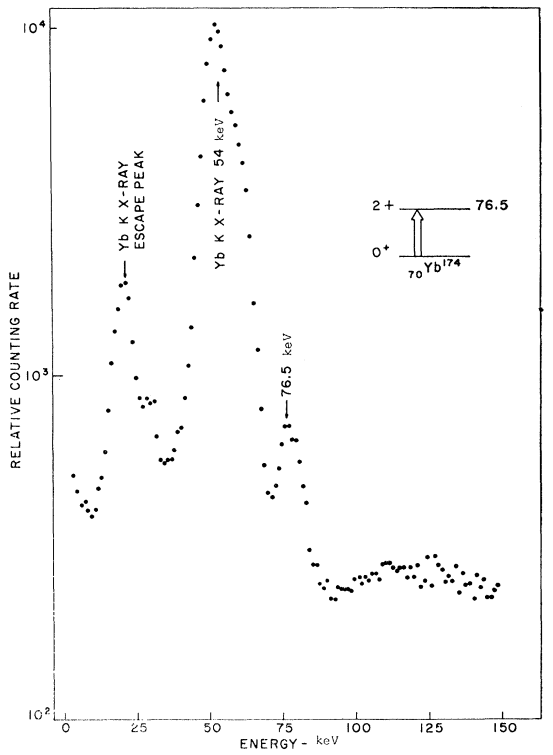

FIG. 2. Coulomb-excitation gamma spectrum for Yb^{174} using an Yb_2O_3 97.3% enriched in Yb^{174} target.

[10] B. Elbek, M. C. Olesen, and O. Skolbreid, Nucl. Phys. **19**, 523 (1960).

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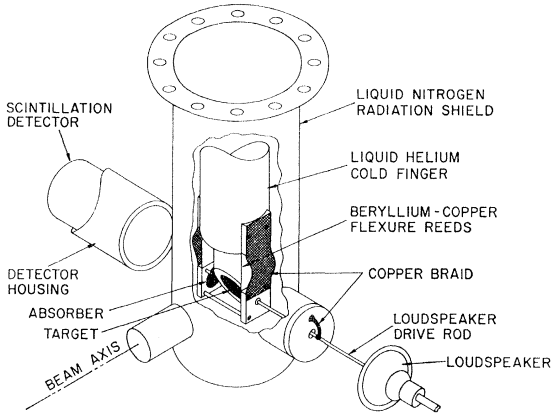

FIG. 3. Schematic drawing of liquid-helium system. Drawing does not show outer vacuum jacket.

stainless steel, and the calibration is accurate to 1%. The same velocity scale was used for each run. The gamma rays were detected by a 2-in.$\times\frac{1}{4}$-in. NaI(Tl) scintillation crystal, and the counts were stored in a 256-channel analyzer operating in the time mode. The helium Dewar was a standard commerical Dewar mounted on a 16-in. stainless-steel box which had entry ports and flanges on all six faces. For a 0.2-μA beam of 3-MeV protons, the helium consumption rate was 0.75 liter/h.

The oxide targets were prepared by placing a thin layer of epoxy resin on the aluminum cold-finger cap, pressing the enriched oxide into the epoxy, and then curing for 20 min at 300°F. Aluminum caps were used to mount the targets to the copper cold finger in order to eliminate the gamma-ray peaks at 54 and 115 keV from the reaction $Cu^{65}(p,n)Zn^{65}$. The enriched metallic target was prepared by fastening a 3-mil Yb metal foil (95.8% enriched in Yb^{174}) to an aluminum cold-finger cap with an epoxy-resin mixture of 60% adhesive, 40% activator, and then curing for 3 h at 150°F. This procedure produces a bond sufficiently durable to withstand the thermal stresses which arise when the target is cooled to liquid-helium temperature. It is estimated that there is less than a 2°K temperature drop across the epoxy layer between target and cold finger with a 0.2-μA beam incident on the target. For the runs employing oxide targets, a metallic absorber was used. The metallic absorber was a 10-mil thick piece of natural Yb metal with a 15-mil backing of copper to reduce the intensity of Yb K x rays incident on the detector. For the Yb^{174} run using the metallic target, the absorber consisted of 45 mg/cm^2 of Yb_2O_3 (95% enriched in Yb^{174}) with a 15-mil Cu backing. The Yb^{170} spectrum was taken at liquid-helium temperature using an 8-mc source of Tm^{170} in Tm metal. The absorber was a 15-mg/cm^2 Yb_2O_3 absorber (65% enriched in Yb^{170}).

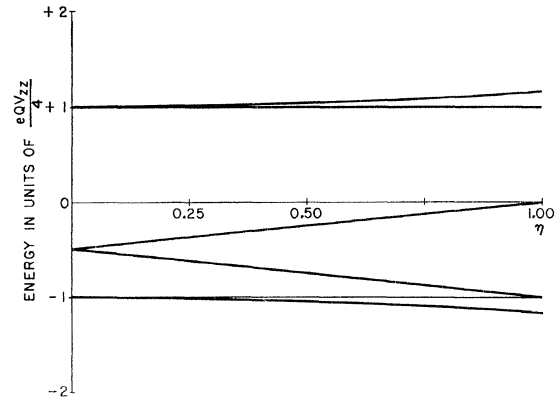

FIG. 4. Quadrupole energy splitting versus asymmetry parameter for a nuclear state of spin $I=2$.

III. DATA ANALYSIS

The analysis of the oxide-versus-metal Mössbauer spectrum for Yb isotopes is greatly simplified due to the presence of electric quadrupole interaction only. The quadrupole-interaction Hamiltonian of the nucleus with the external electric field gradient is given by $H=\mathbf{Q}\cdot\nabla E$.[14] Here, $\mathbf{Q}$ is the quadrupole-moment tensor of the nucleus, and, since the nucleus is in a state of definite angular momentum, the charge has cylindrical symmetry. Because of this fact, it can be shown that only a single parameter eQ is necessary to specify the nine components of $\mathbf{Q}$.[15] The electric-field-gradient tensor ∇E is symmetric by definition. A symmetric tensor is specified by six parameters, but, because the potential must satisfy Laplace's equation, this number is reduced to five. If the electric field gradient is referred

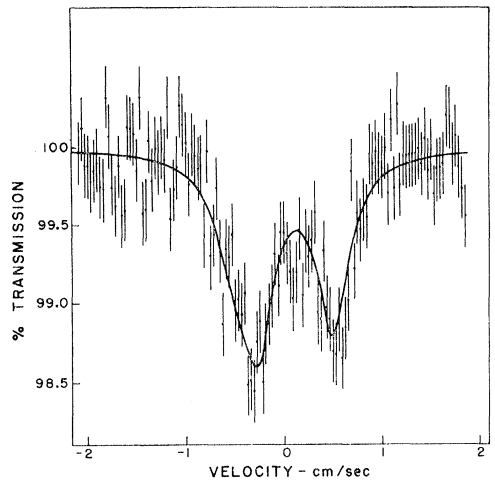

FIG. 5. Mössbauer absorption spectrum following Coulomb excitation. Yb^{174} metallic target versus Yb^{174} oxide absorber.

[14] C. P. Slichter, *Principles of Magnetic Resonance* (Harper and Row, New York, 1963), p. 163.
[15] J. D. Jackson, *Classical Electrodynamics* (John Wiley & Sons, Inc., New York, 1962), p. 102.

to principal axes, the number of parameters necessary to specify ∇E is reduced to two. These two parameters are usually taken to be V_{zz}, the electric field gradient, and $\eta=(V_{xx}-V_{yy})/V_{zz}$, the asymmetry parameter. By choosing $V_{zz}\geqslant V_{xx}\geqslant V_{yy}$, this restricts η to the range $0\leq\eta\leq 1$. If H_Q is now diagonalized for a nuclear state $I=2$, the energy eigenvalues are given by

$$W_{1,-1}=(\tfrac{1}{4}eQ)V_{zz}(-\tfrac{1}{2}\pm\tfrac{1}{2}\eta),$$
$$W_{0,2,-2}=\pm(\tfrac{1}{4}eQ)V_{zz}(1+\eta^{2/3})^{1/2},\quad W_{2,-2}=eQ(\tfrac{1}{4}V_{zz}). \tag{1}$$

The subscripts on the energy eigenvalues indicate which unperturbed magnetic substates are mixed by the quadrupole-interaction Hamiltonian. The quadrupole energy splittings as a function of asymmetry parameter are shown in Fig. 4.

Yb_2O_3 is a member of crystal class T_h^7, in which the Yb ion occupies two different lattice sites.[6] One-fourth of the rare-earth ions occupy sites with C_{3i} symmetry, while remaining three-fourths occupy sites of C_2 symmetry.[16] Previous work[17,18] has indicated that the Mössbauer spectra of the rare-earth oxides can be sufficiently well explained by considering only the C_2 sites. Because of the low symmetry of the C_2 sites, the asymmetry parameter is not negligible and is necessary along

TABLE I. Experimental results for even-even ytterbium isotopes

Atomic no.	γ-ray energy (keV)	$T_{1/2}$ (nsec)	Width[a] (eV$\times10^{-6}$)	$eQV_{zz}/4$ (eV$\times10^{-6}$)	Q^A/Q^{170}	η
170	84.3	1.58	0.917±0.028	1.62±0.05	1.0	0.2 ±0.2
172	78.7	1.6	0.975±0.021	1.65±0.027	1.01 ±0.02	0.38±0.2
174	75.7	1.9	0.655±0.018	1.66±0.05	1.04 ±0.02	0.04±0.4
176	82.2	1.8	0.770±0.022	1.70±0.05	1.045±0.02	0.3 ±0.5

[a] Full width at half-maximum.

with V_{zz} to specify the electric field gradient at the Yb nucleus. η is not well determined in these experiments because of the poorly resolved spectra and the rather weak dependence of the energy eigenvalues on η.

For a determination of the ratios of quadrupole moments, the less than perfect resolution of the five lines was not particularly serious. By measuring the energy difference between the centroids of the observed two peaks in a spectrum such as the one shown in Fig. 5, one obtains an energy proportional to the quadrupole moment. From such measurements for each of the isotopes, we are able to determine the ratios Q^A/Q^{170}. As these centroids can be determined much more reliably than the five line fits, the ratios are more accurate than the quoted values of $eQV_{zz}/4$.

The data obtained in the present work were fitted with a least-squares-fitting routine[19] by five Lorentzian lines

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[17] R. G. Barnes, R. L. Mössbauer, E. Kankeleit, and J. M. Poindexter, Phys. Rev. **136**, 175 (1964).
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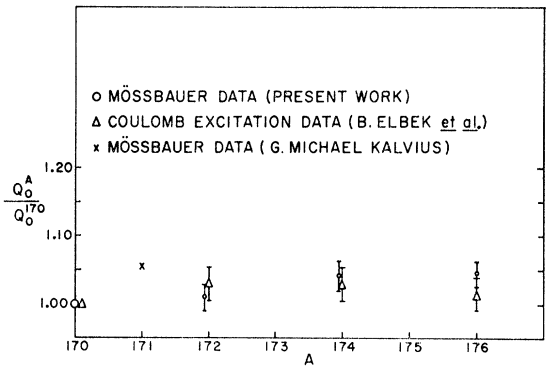

FIG. 6. Relative values of quadrupole moments of Yb isotopes versus atomic number.

having the same width and depth and whose positions were given by Eq. (1). The width, depth, $eQV_{zz}/4$, η, and the level of nonresonant transmission were varied to obtain the best fit. These values are listed in Table I along with the ratio of quadrupole moments Q_0^A/Q_0^{170}. The results are plotted in Fig. 6, where they are compared with Coulomb-excitation cross-section results. The uncertainty assigned to the above-determined values of $eQV_{zz}/4$ is the statistical error and reflects the uncertainty in fitting the data using the above-mentioned least-squares-fitting routine. This error may be a bit conservative, in light of the widths of the fitted lines (see Sec. V).

IV. EXPERIMENTAL RESULTS

Data taken using the Yb^{174} metallic target and the natural metallic absorber are shown in Fig. 7. The single line of width 1.8 mm/sec, which is 1.3 times the natural linewidth, and the undiminished recoil fraction (within experimental error), indicate that there are no detectable radiation-damage effects present when an Yb

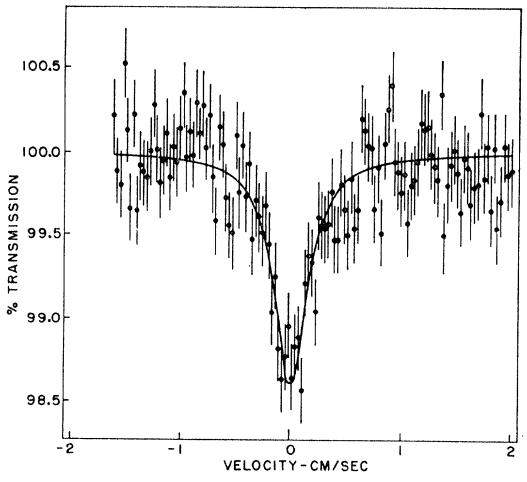

FIG. 7. Mössbauer absorption spectrum following Coulomb excitation. Yb^{174} metallic target versus natural Yb metallic absorber.

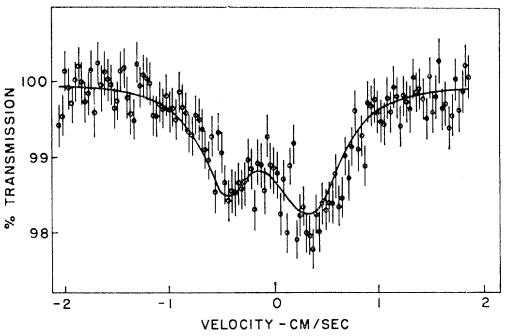

FIG. 8. Mössbauer absorption spectrum following Coulomb excitation. Yb^{172} oxide target versus natural Yb metallic absorber.

metal target is bombarded with 3-MeV protons. The recoilless fraction for the 76.5-keV level of Yb^{174} in Yb metal ($\Theta_D=118.1°K$ at 4°K)[20] is calculated to be 0.07. The recoilless fraction for the target measured from the spectrum in Fig. 7 using the method of areas[21] is 0.05 ± 0.03. The large error attributed to this measurement does not only arise from experimental uncertainty, but is, in part, due to the uncertainty in the value of the internal conversion coefficient used to calculate the Mössbauer absorption cross section. Boyle and Hall[22] list a value of 8 for the internal conversion coefficient for the first excited state of Yb^{174}, while Alder *et al.*[9] list a value of 9. The absence of radiation-damage effects when a metal is bombarded with 3-MeV protons is in agreement with earlier Fe^{57} results.[23] Using the same target, but an Yb_2O_3 absorber (90% enriched in Yb^{174}), the Mössbauer spectrum exhibiting quadrupole splitting was taken for Yb^{174} and is shown in Fig. 5. Since oxide targets were to be used for the other isotopes, a spectrum using an Yb_2O_3 target (95% enriched in Yb^{174}) and the natural Yb metallic absorber was taken in order to observe whether any radiation-damage effects are present when the oxide is bombarded with 3-MeV protons. No spurious peaks which could be attributed to radiation damage were observed. Although no recoilless fraction was determined from the Yb^{174} oxide target versus Yb metal absorber and the Yb^{174} metallic target versus Yb_2O_3 absorber experimental data, the percent absorption was the same within 10% for these two cases after subtracting the nonresonant background and taking into account the line broadening due to finite absorber thickness in each case. Radiation-damage effects were anticipated on the basis of earlier results on Fe_2O_3,[23] but none were observed. The Yb^{172} and Yb^{176} spectra were taken using only oxide targets and the metallic absorber. The spectrum obtained for Yb^{172} is shown in Fig. 8.

The spectrum for Yb^{176} was completely similar to that for Yb^{172} and is not shown.

V. DISCUSSION OF RESULTS

As can be seen from Fig. 6, the relative values for the quadrupole moments determined by Mössbauer spectroscopy for the even-even Yb isotopes agree with those calculated on the basis of the collective model using Coulomb-excitation cross-section data. Despite the close agreement of the two results, the small variation of quadrupole moment as one proceeds from Yb^{170} to Yb^{176}, and the lack of an absolute measurement of the intrinsic quadrupole moment for the even-even Yb isotopes, precludes drawing firm conclusions concerning the validity of the rotational model of deformed nuclei from the relative values of the quadrupole moments determined in the present work. If the values for the intrinsic quadrupole moments determined by Elbek *et al.*[10] are used in order to determine the best value of V_{zz} for Yb^{3+} in Yb_2O_3, a value of $(3.03\pm0.15)\times10^{18}$ V/cm^2 is obtained, where the values of V_{zz} determined for each isotope have been averaged to determine the best value. The best value of η obtained by a weighted average of the values listed in Table I is $\eta=0.2\pm0.1$. The observed linewidths in the present work vary from 3–4 times the natural linewidth. The reason for such broad linewidths is not clearly understood, but is, in part, attributed to neglecting the effects of the Yb ions located at the C_{3i} symmetry sites in the data analysis. Although they constitute only one-fourth of the Yb^{3+} ions in the Yb_2O_3 crystal, they can still make a significant contribution to the observed linewidth.

This is the first reporting of observation of Mössbauer quadrupole spectra for the first excited states of Yb^{172}, Yb^{174} and Yb^{176}. The use of Coulomb excitation to populate the relevant Mössbauer levels makes possible the observation of Mössbauer effects from levels not accessible by radioactive decay. It has come to our attention that Mössbauer effects have recently been observed[24] in Yb^{172} and Yb^{174} using radioactive decay of Lu^{172} and Lu^{174}. These experiments were performed using Yb $Cl_3\cdot6H_2O$ absorbers, and yielded information about the magnetic moments of these states.

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