Define

$$I(\lambda)=\int dc\int dc'|f_c{}^{\mu *}f_{c'}{}^{\mu}+\lambda f_c{}^{\nu *}f_{c'}{}^{\nu}|^2,$$

where λ is an arbitrary real parameter. Since $I(\lambda)$ is non-negative for all values of λ, the result follows immediately that

$$\int dc\int dc' f_c{}^{\mu *}f_c{}^{\nu}f_{c'}{}^{\mu}f_{c'}{}^{\nu *}\leq\left(\int dc|f_c{}^{\nu}|^2\right)\left(\int dc'|f_{c'}{}^{\mu}|^2\right).$$

This result immediately gives

$$|\langle X_\mu|\hat{T}_I|X_\nu\rangle|^2\leq\left(\int dc|f_c{}^{\nu}|^2\right)\left(\int dc'|f_{c'}{}^{\mu}|^2\right).$$

When Eq. (11) is used the inequality takes the form

$$|\langle X_\mu|\hat{T}_I|X_\nu\rangle|^2\leq\Gamma_\mu(E_\mu)\Gamma_\nu(E_\mu)/4.$$

It is important to note that the width Γ_ν is evaluated at E_μ because the shift is being evaluated at the resonance corresponding to the state X_μ, i. e., at E_μ.

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Mössbauer-Effect in Yb^{171}†

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Recoil-free nuclear resonance absorption of the 67-keV γ ray has been observed. Tm^{171} in the form of the sesquioxide was used as a source, and Yb_2O_3 and Yb metal as absorbers. Source and absorber were held at the same temperatures of 50, 4.2, and 1.6°K. Due to the field gradient present in the oxide crystal, the 67-keV state ($I=\frac{3}{2}$) splits into two substates with a separation of 2.4×10^{-6} eV extrapolated to 0°K. The Yb metal forms a cubic crystal lattice and shows no splitting. The observed linewidth gives a lower limit of 7×10^{-10} sec for the lifetime of the 67-keV state. Comparison with previous data of other authors on the hyperfine splitting of the 84-keV level in Yb^{170} in Yb_2O_3 gives a value of 1.04 for the ratio $Q_0(171)/Q_0(170)$ of the intrinsic quadrupole moments, which agrees well with the values obtained by Coulomb excitation.

INTRODUCTION

THE observation of the hyperfine structure of nuclear levels in heavy rare-earth isotopes has recently become a powerful method for the determination of their dipole and quadrupole moments.[1–4] The information so gained is especially valuable because the theory of strongly deformed nuclei[5] is highly developed and allows quite accurate predictions of the moments of these nuclei.

It has recently been shown[6] that in the β^- decay of Tm^{171} the 66.7-keV state of Yb^{171} is weakly excited. This level has a spin of $\frac{3}{2}$ and is the first excited state of a $K=\frac{1}{2}$ rotational band. The γ transition to the ground state ($I=\frac{1}{2}$) should be suitable for Mössbauer experiments; it would be of interest because the weak γ-ray intensity makes it experimentally difficult to get information about the 66.7-keV state by other methods, as for instance coincidence spectroscopy.

EXPERIMENTAL

The experimental methods used were the standard techniques of Mössbauer spectroscopy in transmission geometry. The source was produced by a two-week irradiation of 20 mg of 87% enriched $Er_2{}^{170}O_3$ with a neutron flux 2×10^{14} n/cm^2 sec in the Oak Ridge Research Reactor. Neutron capture produces 7.5-h Er^{171} which undergoes a β^- decay into Tm^{171}. After two months, only the Tm^{171} activity together with a small contamination of 127-day Tm^{170} was present. The source was not further processed except for a heat treatment of 24 h at 1000°C in air to make sure that only the trivalent oxide is present. The oxide powder was uniformly spread over an area of 1 cm^2 and glued with epoxy between two 10-mil copper plates. The absorbers used were 80 mg/cm^2 of Yb_2O_3 powder enriched to 80% in Yb^{171} and 170 mg/cm^2 of natural Yb metal foil. Both the oxide powder as well as the metal sheet were glued with epoxy between two 10-mil copper sheets. Source and absorber were mounted in the exchange-gas chamber of a helium cryostat[7] as shown in Fig. 1. The exchange gas (1 Torr He) assures thermal equilibrium of source and absorbers. The temperature was measured by a Cu-Au/Co thermocouple. The source was moved

† Supported in part by the U. S. Atomic Energy Commission.
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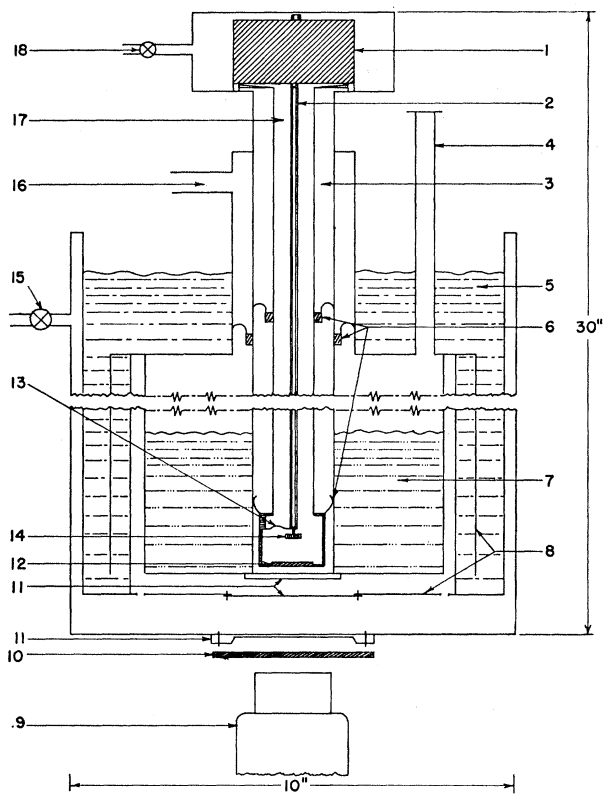

FIG. 1. Schematic of experimental setup: 1. Double loudspeaker. 2. Driving tube. 3. Exchange-gas chamber. 4. Liquid-helium fill. 5. Liquid-nitrogen shield. 6. Heat contacts. 7. Liquid-helium vessel. 8. Conduction shield. 9. Scintillation counter. 10. Copper absorber. 11. Windows for γ rays (Be or Cu). 12. Resonance absorber. 13. Leafspring for support of source. 14. Source. 15. Dewar pumpout connection. 16. Helium pump line. 17. Support tube. 18. Exchange-gas inlet.

with constant acceleration via a stainless-steel tube by a double loudspeaker drive with high negative feedback, very similar to systems already described.[8,9] The Mössbauer spectrum was displayed in 200 channels of a TMC-401 analyzer used in multiscalar operation. The γ rays were detected by a scintillation spectrometer.

Figure 2 shows the pulse-height spectrum obtained behind the resonance absorber. A 500-mg/cm^2 copper absorber was used to weaken the K x-ray intensity. The 67-keV line is still only poorly resolved from the x ray, but clearly visible. The 84-keV is due to a contamination of our source with Tm^{170}. The setting of the single channel used is also indicated in Fig. 2. The velocity spectrometer was calibrated with the known hyperfine spectrum of a Co^{57} source in metallic iron and an iron metal absorber[10] which extends to velocities of ± 1.07 cm/sec and also gives the precise location of the zero-velocity channel. The calibration should be accurate within $\pm 3\%$.

RESULTS

The recoil-free resonance absorption of the 67-keV γ line of Yb^{171} was readily observed. Some typical data obtained are shown in Figs. 3 and 4. Figure 3A shows the transmission spectrum of an oxide absorber at 50°K. This temperature was achieved by filling the helium container of the cryostat with liquid nitrogen and lowering its vapor pressure by pumping with a forepump of 1500-liters/min capacity. The transmission spectrum indicates a poorly resolved hyperfine splitting. At 4.2°K the form of the spectrum is principally the same but the maximum resonance absorption increases from 7 to 10%. At 1.6°K, which was achieved by lowering the vapor pressure of the He bath, the spectrum shown as curve B was obtained. It obviously is no longer symmetric with respect to zero velocity, extends to a wider velocity range, and the maximum resonance absorption decreases somewhat, whereas the area under the curve stays constant within 3%. Furthermore no

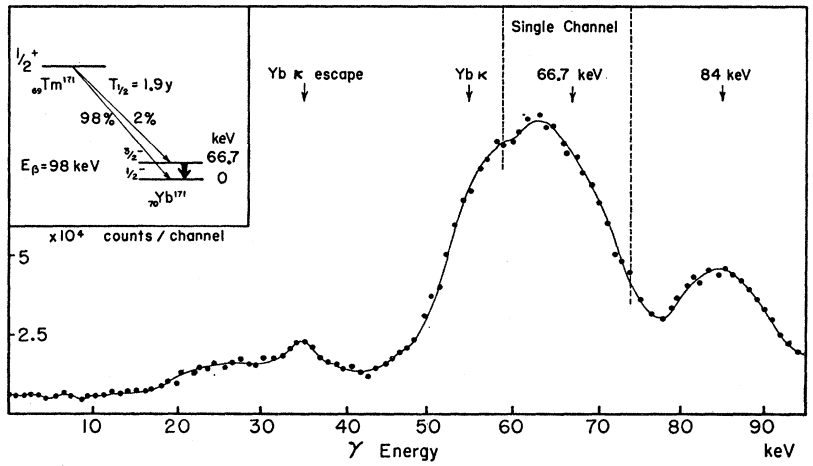

FIG. 2. Decay scheme and scintillation spectrum of Tm^{171}.

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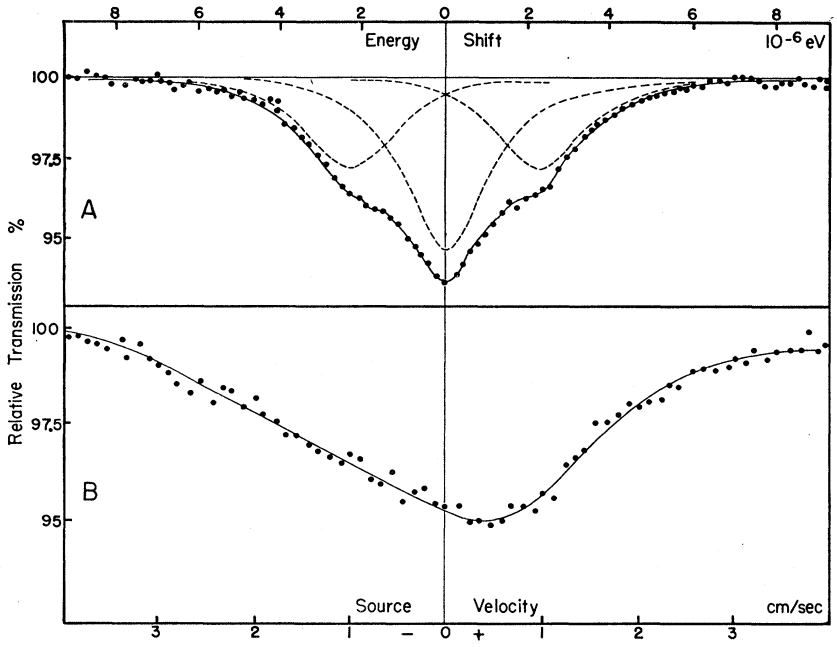

FIG. 3. Resonance absorption pattern in Yb^{171} in Yb_2O_3. The source is Tm_2O_3. Source and absorber are at the same temperatures: $A=50°K$; $B=1.6°K$.

structure can be resolved. The spectrum of Fig. 4 was taken with the metal absorber at 4.2°K and shows two fairly well resolved hyperfine lines. Within our limits of error there is no shift of the center of gravity of the spectrum compared to the spectra of the oxide absorber. Due to the fact that enriched metal absorbers are not easily available and that the Debye temperature of ytterbium metal is only[11] around 100° compared to a value around 350° for the sesquioxide, the resonance absorption is rather weak and measurements at higher temperatures are not possible.

DISCUSSION

The rare-earth sesquioxides form crystals of the class T_h^7. In this crystal the rare-earth ion occupies two different lattice sites. One quarter of the rare-earth ions occupy places with a C_{3i} symmetry, the rest places with a C_2 symmetry. It has been shown[12,13] in the case of Tm^{169} that the hyperfine spectra of rare-earth oxides could be sufficiently well explained by considering only the rare-earth ions on C_2 lattice places. Because of the low symmetry of the surroundings of the Yb^{+++} ion we expect the existence of a large electric-field gradient at the nucleus, causing the excited state ($I=\frac{3}{2}$) to split into two sublevels. So our source will emit two lines of equal intensity separated by ΔE_Q. The same holds for the Yb_2O_3 absorber. In our transmission experiment we shift the two emitter lines over the two absorber lines, thus producing a three-line pattern with a line separation of ΔE_Q, an intensity ratio of 1:2:1, and symmetry with respect to zero velocity. In Fig. 3A the hyperfine spectrum is analyzed according to these considerations. Because of the temperature dependence of the quadrupole splitting in rare-earth ions,[13,14] the separation of the lines increases slightly with lower temperatures.

Yb metal crystallizes in a cubic lattice. Therefore, no quadrupole interaction is to be expected for the metal absorber. The data of Fig. 4 show only two hyperfine lines representing the quadrupole splitting of the $\frac{3}{2}$ level in the source. No isomer shift has been observed so far for a transition between two nuclear levels of the same rotational band, because there is no change in the

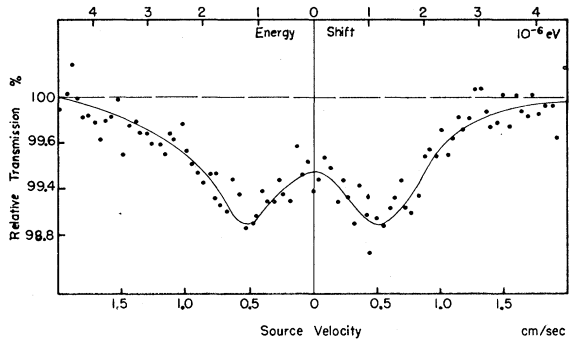

FIG. 4. Resonance absorption pattern in Yb^{171} in metallic Yb. The source is Tm_2O_3. Both source and absorber are held at 4.2°K.

[11] K. A. Gschneidner, Jr., *Rare Earth Alloys* (D. Van Nostrand and Company, New York, 1961), p. 38.

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TABLE I. Results of measurements on the hyperfine splitting in Yb^{171} for different absorbers and temperatures. The source is always Tm_2O_3, and is kept at the same temperature as the absorber.

Absorber	Thickness of Yb^{171} (mg/cm²)	Temp. (°K)	Quadrupole splitting (cm/sec)	Isomer shift (cm/sec)	Experimental linewidth (cm/sec)	Corrected linewidth (cm/sec)
Yb_2O_3	55	50	1.02	0.0	0.99	0.42
Yb_2O_3	55	4.2	1.10	0.0	1.07	0.46
Yb	25	4.2	1.05	0.0	0.90	0.45

nuclear charge radius involved if the transition is purely rotational.

Table I shows the results of the analysis of the spectra. Averaging the different runs and extrapolating to 0°K gives a value for the quadrupole splitting of the 67-keV state of Yb^{171} in Yb_2O_3 of

$$\Delta E_Q = 1.07 \pm 0.10 \text{ cm/sec} = (2.4 \pm 0.2) \times 10^{-6} \text{ eV}.$$

In Table I the experimental widths of the hyperfine lines are also listed together with the correction for finite absorber thickness.[15] The mean value for the corrected line width from the different measurements is

$$\Gamma_e = 0.44 \text{ cm/sec}.$$

Considering that a broadening of the resonance line in a Mössbauer experiment is always possible, by using the uncertainty principle

$$\tau = \Gamma/\hbar,$$

we can set the lower limit for the lifetime of the 66.7-keV level to

$$\tau \geq 0.7 \text{ nsec}.$$

The broadening of the hyperfine spectra in Yb_2O_3 at very low temperatures (Fig. 3B) is not yet understood. The asymmetry of the absorption spectrum indicates that the hyperfine splitting in source and absorber is now different. This is in agreement with an observation of a blurring of the resonance spectrum in Yb^{170} taken with metal source and an Yb_2O_3 absorber.[16] It is probably due to magnetic ordering in Yb_2O_3 at temperatures around 2°K.

Our measurements of the quadrupole splitting reveal only the product of the quadrupole moment and the electric-field gradient. It is not possible to give a good estimate of the field gradient in the rare-earth oxide where only limited optical data are available. So a direct determination of the quadrupole moment is not possible. On the other hand a measurement of the quadrupole splitting of the 2^+ level in Yb^{170} in Yb_2O_3 is available.[17] This allows a determination of the ratio of the intrinsic quadrupole moments $Q_0(171)/Q_0(170)$ of the two ytterbium isotopes. Because of the low symmetry (C_2) in the oxide crystal, the electric-field gradient is not rotationally symmetric and therefore has to be described by two parameters. A convenient choice is

$$q = V_{zz}/e,$$

and the asymmetry parameter

$$\eta = (V_{xx} - V_{yy})/V_{zz}$$

with

$$V_{xx} = \partial^2 V/\partial x^2, \quad \text{etc.}$$

The general matrix element for quadrupole interaction is

$$\langle M|H|M'\rangle = (-)^{I-M'}(\tfrac{1}{5}\sqrt{\pi})eQV_{M-M'}{}^{(2)} \times C(I,2,I;M',M-M',M)/C(I,2,I;I,0,I)$$

with $M = -I, -I+1, \cdots, +I$ and

$$V_0{}^{(2)} = \tfrac{1}{4}e(5/\pi)^{1/2}q,$$
$$V_{\pm 2}{}^{(2)} = \tfrac{1}{4}e(5/6\pi)^{1/2}q\eta.$$

The secular determinants have the following roots:

for $I = \frac{3}{2}$

$$W_{1,2} = \pm\tfrac{1}{4}e^2qQ(1+\tfrac{1}{3}\eta^2)^{1/2}$$

for $I = 2$

$$W_{1,2} = \pm\tfrac{1}{4}e^2qQ(1+\tfrac{1}{3}\eta^2)^{1/2}, \quad (M = 0, 2, -2)$$
$$W_3 = \tfrac{1}{4}e^2qQ, \quad (M = 2, -2)$$
$$W_{4,5} = \tfrac{1}{4}e^2qQ(-\tfrac{1}{2} \pm \tfrac{1}{2}\eta), \quad (M = 1, -1).$$

So a $I = \frac{3}{2}$ state will always split into two sublevels, but for $\eta = 0$ we expect a $I = 2$ level to split into five substates. The data of Wagner *et al.*[17] obtained on the 2^+ level of Yb^{170} in Yb_2O_3 are consistent with a splitting calculated for $\eta = 4$. It has to be noted that the only assumption about the choice of the quantization axis z in the calculation above is, that the x, y, z coordinate system is a system of principal axes. It is customary to use a coordinate system such that

$$|V_{zz}| \leq |V_{yy}| \leq |V_{xx}|,$$

which implies that η has the property $0 \leq \eta \leq 1$, but Wagner *et al.*[17] used for the representation of their results a coordinate system in which V_{zz} is the smallest component of the field-gradient tensor. In this case the asymmetry parameter may become larger than unity. However, the comparison of the quadrupole moments of Yb^{170} and Yb^{171} is independent of the value of η if both quadrupole couplings are measured in the same crystal. From the calculation above we see that the solutions $W_{1,2}$ are equal for $I = \frac{3}{2}$ and $I = 2$. In the case of $I = \frac{3}{2}$,

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the separation of the two hyperfine lines is directly equal to $2W_{1,2}$. In the case of $I=2$ the separation of the two outermost hyperfine lines is always equal to $2W_{1,2}$ (see Fig. 2 in Ref. 17).

From the data of Wagner *et al.*[17] we take the separation of the two outermost lines ($M=0, 2, -2$) in the hyperfine spectrum of Yb^{170} in Yb_2O_3 and get

$$\tfrac{1}{4}e^2qQ_2{}^{170}(1+\tfrac{1}{3}\eta^2)^{1/2}=3.3\times10^{-6}\text{ eV},$$

and from the present measurement

$$\tfrac{1}{4}e^2qQ_{3/2}{}^{171}(1+\tfrac{1}{3}\eta^2)^{1/2}=2.4\times10^{-6}\text{ eV}.$$

By using the expression

$$Q_0=\frac{(I+1)(2I+3)}{3K^2-I(I+1)}Q_I,$$

one obtains a ratio of the intrinsic quadrupole moments:

$$Q_0{}^{171}/Q_0{}^{170}=2.4\times5\times2\times10^{-6}/3.3\times7\times10^{-6}=1.04.$$

By Coulomb excitation[2] in Yb^{170} and Yb^{171}, the quadrupole moments were determined to be

$$Q_0{}^{171}=8.0\text{ b},^{18}$$

$$Q_0{}^{170}=7.5\text{ b},^{19}$$

leading to a ratio of

$$Q_0{}^{171}/Q_0{}^{170}=8.0/7.5=1.07,$$

which agrees very well with our data.

ACKNOWLEDGMENTS

The author would like to thank Dr. S. Hüfner (Technische Hochschule Darmstadt) and Dr. B. Chi for enlightening discussions, and B. W. Murray for his help during the course of the experiment.

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Gamma Rays from the Low-Lying Levels of F^{18}†

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A 5-mm-thick lithium-drifted germanium counter was used to detect the low-energy gamma rays ($E\gamma<1.8$ MeV) emitted at 90° to the beam following bombardment of an O^{16} gas target with He^3 ions with a mean energy of 1.8 MeV. The excitation energies of the first seven levels of F^{18} were determined to ±2 keV or better from energy measurements of the de-excitation gamma rays from these levels. The decay of the F^{18} 2.10-MeV level to the 0.94- and 1.08-MeV levels was confirmed, and an upper limit of 6% was placed on the branch to the 1.045-MeV level.

INTRODUCTION

THE sixth excited state of F^{18} at an excitation energy of 2.10 MeV has been reported[1] to have a mean lifetime of $(0.7\pm0.2)\times10^{-12}$ sec and decays to the F^{18} ground state and first excited state at 0.94 MeV with branching ratios of 38±3% and 28±5%, respectively.[2,3] The remaining 34% is made up of branches to one or both of the 1.045- and 1.082-MeV levels so that gamma rays with energies of 0.94, 1.02, 1.045, 1.055, 1.082, and 1.16 MeV are possible from the decay of the 2.10-MeV level. Both of the previous investigations[2,3] of the decay of the F^{18} 2.10-MeV level were done with NaI (Tl) gamma-ray detectors which do not have adequate resolution to separate these gamma-ray lines. The work of Poletti and Warburton[3] indicated that the branch to the 1.082-MeV level was more intense than that to the 1.045-MeV level, but this was not absolutely certain and no sharp limit was placed on the intensity of the 2.10 → 1.045 transition.

The F^{18} 0.94- and 1.045-MeV levels have $J^\pi=3^+$ and 0^+, respectively; while the 1.082-MeV level has $J=0$, 1, or 2 with 0 preferred.[3] The F^{18} 2.10-MeV level has $J=1$ or 2.[3] Thus, either the 2.10 → 0.94 or the 2.10 → 1.045 transition must be quadrupole and the 2.10 → 1.082 transition may be also. Combining the lifetime measurement for the 2.10-MeV level with the branching ratios gives a strength of 54±18 Weisskopf units for the 2.10 → 0.94 transition if it is $E2$ and 128±42 Weisskopf units to be divided between the 2.10 → 1.082 and 2.10 → 1.045 transitions if they are both $E2$.[3] These transition strengths are strong enough to be quite startling and so it is of interest to check on the decay modes of the 2.10-MeV level and in particular to find the rela-

† Work performed under the auspices of the U. S. Atomic Energy Commission.

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