Mössbauer Effect in Two Excited States of a Rotational Band in Yb^{171}†

G. M. KALVIUS AND J. K. TISON
Argonne National Laboratory, Argonne, Illinois
(Received 1 July 1966; revised manuscript received 22 August 1966)

Mössbauer measurements on the ratio of the quadrupole splitting of the first and second excited states in Yb^{171} show that the intrinsic quadrupole moment is constant within the experimental error of 3% for the first two members of the $K=\frac{1}{2}$ rotational band. The width of the resonance line gives a lifetime for the 75.9-keV state in Yb^{171} of $\tau=(1.5\pm0.3)$ nsec. This is 40% shorter than the electronically measured value. The electric field gradient for the C_2 site in Yb_2O_3 is found to be $(2.83\pm0.06)\times10^{18}$ V/cm^2.

I. INTRODUCTION

SEVERAL strongly deformed nuclei have more than one excited state low enough in energy to allow the observation of the Mössbauer effect. The resonance of higher excited states has been successfully investigated in Eu^{153} [1] and Dy^{161}.[2] Several attempts in Pt^{195} have not given a definitive result.[3] The method of using more than one resonance in the same nucleus allows one to keep the solid-state parameter of a hyperfine interaction (for instance, the electric-field gradient) constant, thus giving easier access to the determination of the nuclear parameters. In this manner, ratios of the parameters of different levels can be obtained with very high precision. In Yb^{171}, in contrast to Eu^{153}, and Dy^{161}, both the first and the second excited states belong to the same rotational band. This is a $K=\frac{1}{2}$ band, which has been studied by nuclear spectroscopy up to its $\frac{7}{2}^-$ member at 230.6 keV.[4] The special feature of staying within a given band will allow an accurate check on the constancy of properties which belong to the band as a whole, while the nucleus is in different states of excitation. A typical property of this kind is the intrinsic nuclear quadrupole moment Q_0. However, from the strongly deformed nuclei available, all even-even nuclei cannot be used for such a Mössbauer experiment since their second excited state (the 4^+ state) will not decay directly into the ground state ($E4$-transition). The odd A nuclei are more favorable candidates for the observation of the Mössbauer resonance of two consecutive excited levels. Among these nuclei those having a $K=\frac{1}{2}$ band are especially favorable, since their rotational spectrum is anomalous because of the decoupling of the spin momentum from the rotational motion. A disadvantage is that the ground state ($I=\frac{1}{2}$) will not show a quadrupole moment, but the observation of the splitting of two excited states makes it possible to use the inherently high accuracy of the Mössbauer effect for a proof of the constancy of Q_0.

The positive value[5] of the decoupling parameter in Yb^{171} causes a large separation of the first excited state (66.7 keV) from the ground state and a close spacing of the second excited state (75.9 keV) to the first, making this nucleus an ideal candidate for this kind of investigation.

II. EXPERIMENTAL

The recoil-free resonance absorption of the 66.7-keV gamma ray of Yb^{171} has been investigated previously.[6] The present paper deals with the observation of the Mössbauer effect, in particular with the quadrupole splitting of the 75.9 keV second excited state. This measurement required the use of a different source activity, since the β^- parent Tm^{171} ($T_{1/2}=2y$), which was previously employed in the investigation of the 66.7-keV state, does not decay via the 75.9-keV level.[7] Therefore, we had to use the K-capture source Lu^{171} ($T_{1/2}=8$, $4d$). It was produced by two reactions, either via Yb^{171} (p,n) Lu^{171} or via $Tm^{169}(\alpha,2n)Lu^{171}$. The latter reaction offers the advantage that natural Tm (with Tm^{169} 100% abundant) is the starting material, and metal and alloy sources are thus easier to obtain. A 1-mil Tm foil of $\frac{3}{8}$ in. diam was irradiated for 20 μA h with a 25-MeV-alpha beam of the 60 in. ANL cyclotron. This source also provides a solution to the difficulty in the Mössbauer spectroscopy of the rare earths (RE) of finding a single-line emitter. Most often cubic Laves phase intermetallic compounds, for instance, $(RE)Al_2$, are used.[8] In Yb, the divalent ion has a filled $4f$ shell and thus a $^1S_0$ ground state. Yb metal contains only the divalent ion; its main disadvantage is its low Debye temperature of 100°K. Wagner *et al.*[9] used Tm metal as a single-line source for the investigation of the 84 keV state in Yb^{170} with a reasonable recoil-free fraction. Our source of Lu^{171} in

† Based on work performed under the auspices of the U. S. Atomic Energy Commission.

[1] E. Steichele, S. Hüfner, and P. Kienle, Phys. Letters **14**, 321 (1965).

[2] V. V. Skylarevskii, B. N. Samoilov, and E. P. Stepanov, Zh. Eksperim. i Teor. Fiz. **40**, 1874 (1961) [English transl.: Soviet Phys.—JETP **13**, 1316 (1961)].

[3] A. B. Buym and L. Grodzins, Phys. Letters **21**, 389 (1966).

[4] G. Kaye, Nucl. Phys. **86**, 241 (1966).

[5] B. Elbeck, K. O. Nielsen, and M. C. Olesen, Phys. Rev. **107**, 406 (1957).

[6] G. M. Kalvius, Phys. Rev. **137**, B1441 (1965); C. Gunther and E. Kankeleit, Phys. Letters **22**, 443 (1966); W. Hennig, P. Kienle, E. Steichele, and F. Wagner, *ibid* **22**, 446 (1966).

[7] B. L. Shanna, Nucl. Phys. **25**, 175 (1961).

[8] R. L. Cohen and J. H. Wernick, Phys. Rev. **134**, B503 (1964).

[9] F. E. Wagner, F. W. Stanek, P. Kienle, and H. Eicher, Z. Physik **166**, 1 (1962).

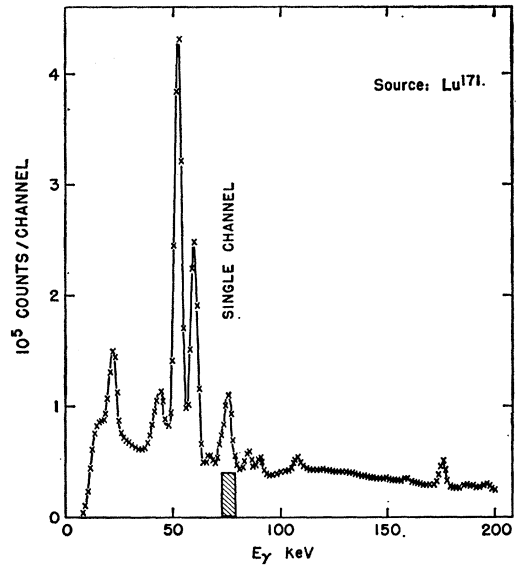

FIG. 1. Low-energy gamma-ray spectrum of Lu^{171} as obtained with a lithium-drifted germanium detector. The vertical bars indicate the window of the single-channel analyzer set on the 75.9-keV line.

Tm metal gave similar results, which proves that the highly diluted Yb is in the divalent state inside the Tm^{3+} lattice; and, despite the fact that Tm metal is ferrimagnetic at low temperatures,[10] there exists no (or only a very small) magnetic field at the Yb nucleus.

Measurements were performed at 4.2°K in standard-transmission geometry. Both source and absorber were mounted in the helium container of a glass cryostat. The high energy of the gamma ray does not require any special windows, and the beam had to pass through 2 mm of glass. The velocity spectrometer was an electromechanical feedback system with sinusoidal motion. The spectrum was stored in a 400-channel anaylzer operated in time mode. The signal from the 200 bistable of the analyzer was used after some shaping to phase lock an Hewlett-Packard 3300A function generator. Its sinusoidal output was used as the reference signal .The feedback loop consisted of a Sanborn 8875A dc amplifier and a Hewlett-Packard 467A power amplifier in a circuit similar to the one described by Kankeleit.[11] The transducer used a loudspeaker type motor and a 6LV1 velocity transducer as a sensing element. It was mounted on top of the cryostat, feeding the motion via a stainless-steel tube to the absorber.[12]

Since Lu^{171} has a rather complex gamma-ray spectrum with several lines between 50 and 100 keV,[13] and since the separation of the 66.7- and 75.9-keV line had to be complete, a high-resolution detector had to be used. Figure 1 shows the gamma-ray spectrum through the resonance absorber as obtained with a 20-mm-diam by 10-mm-thick lithium-drifted germanium crystal.[14] The resolution is 3.5 keV full width at half maximum (FWHM) at count rates of 5×10^4 cps employing a tube preamplifier and a TC-200 main amplifier. As one sees from Fig. 1, the resolution of 75.9-keV gamma ray from the 66.7-keV ray is complete; however, a 72.3-keV ray, is only partially resolved. This gamma ray originates from a transition between a 167.3-keV state to a 95.1-keV state, both belonging to a $K=\frac{7}{2}$ band.[4] Therefore, this gamma ray will only contribute to the background in the single channel, whose setting is also indicated in Fig. 1. The oxide absorber was made from 95% enriched $Yb^{171}{}_2O_3$. The oxide was mixed with Lucite powder and pressed into a disc of $1\frac{1}{4}$ in. diam at 250°F.

III. RESULTS

The velocity spectrum obtained with the metal source and a Yb_2O_3 absorber at 4.2°K is shown in Fig. 1. Its structure reflects the quadrupole splitting of the $I=\frac{5}{2}$ second excited state into three hyperfine levels due to the noncubic surrounding of the Yb ions in Yb_2O_3. Actually, the rare-earth ions occupy two different lattice sites in the sesquioxides, with one quarter of the ions having a C_{3i} symmetry, the rest a C_2 symmetry. However, it has been shown in several cases (as, for instance, in Tm^{169})[15] that the quadrupole spectrum in the sesquioxides can be explained satisfactorily by considering the C_2 sites only. This would mean that the field gradients at the two sites are indistinguishable with the limited resolution obtainable with these rather short-lived nuclear states. However, a broadening of the resonance line may well result.

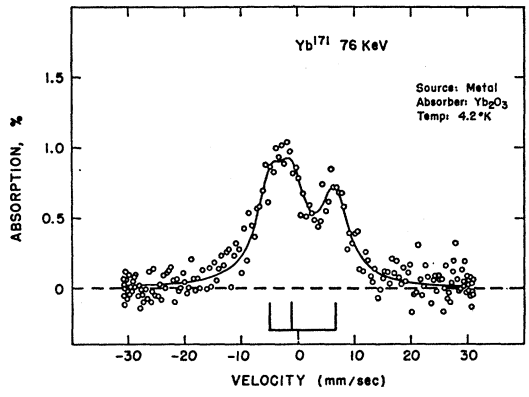

FIG. 2. Mössbauer spectrum for Yb_2O_3 at 4.2°K. The source is Yb in Tm metal. The bars indicate the positions and intensities of the three partially resolved hyperfine lines due to the quadrupole splitting of the $\frac{5}{2}$ excited state.

[10] W. C. Koehler, J. W. Cable, E. O. Wollan, and M. K. Wilkinson, Phys. Rev. **126**, 1672 (1962).
[11] E. Kankeleit, *Mössbauer Effect Methodology*, edited by I. Grouverman (Plenum Press, Inc., New York, 1966), Vol. I.
[12] M. Kalvius, *Mössbauer Effect Methodology*, edited by I. Grouverman (Plenum Press, Inc., New York, 1966), Vol. I.
[13] B. Harmatz, T. H. Handley, and J. W. Mihelich, Phys. Rev. **119**, 1345 (1960).
[14] Geli 50, made by H. Mann, Electronics Division, Argonne National Laboratory.
[15] R. G. Barnes, R. L. Mössbauer, E. Kankeleit, and J. B. Poindexter, Phys. Rev. **136**, A1751 (1964).

TABLE I. Results of Mössbauer spectroscopy of the 75.9 and 66.7-keV gamma rays in Yb^{171}. Source and absorber are both at 4.2°K.

Source material	Source activity	Absorber material	Absorber thickness mg/cm² Yb^{171}	Resonance Line keV	Spectrum	Linewidth measured mm/sec	Linewidth corrected mm/sec	Quadrupole splitting $eQ\,V_{zz}$ mm/sec
Yb in Tm metal	Lu^{171}	Yb_2O_3	30 mg/cm²	75.9	Partially resolved three-line pattern	6.0±0.6	2.3 ±0.3	25.6±0.4
Yb in Tm metal	Lu^{171}	Yb metal	25 mg/cm²	75.9	Single-line pattern	3.8±0.8	1.8 ±0.4	0
Yb in Tm_2O_3	Lu^{171}	Yb_2O_3	30 mg/cm²	75.9	Unresolved seven-line pattern	a	a	a
Yb in Er_2O	Tm^{171}	Yb_2O_3	30 mg/cm²	66.7	Partially resolved three-line pattern[b]	1.0±0.05	0.44±0.05	20.7±0.4

[a] The lack of structure does not allow evaluation of these parameters.

[b] See also Ref. 6.

The solid curve through the experimental points in the figure is a least-squares fit, assuming three hyperfine lines of equal intensity and width. The fitting was performed by a computer program described elsewhere.[16] The goodness-of-fit parameter χ^2 is normalized to one per degree of freedom. The illustrated curve has 190 data points; its $\chi^2=180$ indicates that a three-line fit leads to a good representation of the experimental results. Data obtained for several source and absorber combinations are summarized in Table I.

IV. DISCUSSION

From the results of the present investigation as summarized in Table I we may draw the following conclusions:

(a) *Line width:* The single line Yb metal absorber gives somewhat sharper lines than the split-oxide absorber. The accuracy is limited here because the observed resonance absorption in Yb metal is only 0.4%, because of its low Debye temperature ($\Theta\approx 100$°K).[17]

The experimental width of the resonance was corrected for finite absorber thickness.[18] From

$$\Gamma=1.8\pm0.4 \text{ mm/sec}=(4.5\pm0.1)\times10^{-7} \text{ eV},$$

we find

$$\tau_{MB}=1.5\pm0.3 \text{ nsec}.$$

This number is a lower limit, since a residual hyperfine interaction may broaden the line. A very recent electronic measurement[4] of this lifetime gave

$$\tau_{EL}=(2.5\pm0.4) \text{ nsec}.$$

The reduced transition probability as determined by Coulomb excitation[5]

$$B_{1/2-5/2}(E2)=3.7\times10^{-48}\ e^2 \text{ cm}^4,$$

and a conversion coefficient

$$\alpha_{tot}\approx\alpha_K+\alpha_L=7.25$$

from the tables of Sliv and Band[19] leads to a lifetime

$$\tau_{CE}=3.0 \text{ nsec}.$$

The difference between τ_{MB} and τ_{EL} has its origin probably in a broadening of our source line due to the ferrimagnetism in the host material thulium. The longer lifetime τ_{CE} shows that roughly 30% of the decay uses the additional $M1$-channel via the 66.7-keV level. The intensity of the thus generated 9.2-keV transition is hard to detect since its energy is very close to the L x rays ($L_\alpha=7.4$ keV, $L_\beta=8.6$ keV) of Yb.

(b). *Quadrupole interaction:* The C_2 symmetry of the Yb^{3+} ion in Yb_2O_3 gives rise to a nonaxially symmetric-field gradient. However, the eigenvalues change only very little with increasing asymmetry parameter η.[20] This makes an independent determination of V_{zz} and η impossible with the resolution available. Indeed, our spectrum could be fitted with only little variation in χ^2 for $0\leqslant\eta\leqslant0.3$. Since the splitting, a $2+$ is more sensitive to the value of η, and we may take the results of Wagner *et al.*[9] on the 84-keV level in Yb^{170}. A re-evaluation from the line position as given in Fig. 1 of Ref. 9 leads to

$$\eta=0.25\pm0.5.$$

This gives us

$$eQ_{5/2}V_{zz}=25.6\pm0.4 \text{ mm/sec}=(6.5\pm0.1)\times10^{-6} \text{ eV}.$$

The intrinsic quadrupole moment of Yb^{171} has been given as 8.0 b[5]; this leads to an electric-field gradient at the Yb nucleus in Yb_2O_3 of $V_{zz}=(2.83\pm0.06)\times10^{18}$ V/cm². This value is in good agreement with $V_{zz}=2.9\times10^{18}$ V/cm² as obtained from the splitting of the 2^+ state in Yb^{170}.[21]

[16] See, for further references, S. L. Ruby, G. M. Kalvius, R. E. Snyder, and G. B. Beard, Phys. Rev. **148**, 176 (1966).

[17] K. A. Gschneider, Jr., *Rare Earth Alloys* (D. Van Nostrand Company, Inc., New York, 1961), p. 38.

[18] S. Margulies and T. R. Ehrman, Nucl. Instr. Methods **12**, 131 (1961).

[19] K. Siegbahn, *Alpha-, Beta-, and Gamma-Ray Spectroscopy* (North-Holland Publishing Company, Amsterdam, 1965), p. 1639 ff.

[20] M. H. Cohen, Phys. Rev. **96**, 1278 (1954).

[21] The number given by Wagner *et al.* is for a system of coordinates which does not obey the usual condition $|V_{zz}|\geq|V_{yy}|\geq|V_{xx}|$. The transofrmation of the value for V_{zz} given in Ref. 9 to the generally used system of coordinates leads to the number quoted.

(c). *Constancy of the intrinsic quadrupole moment with rotational excitation:* The quadrupole interaction of the $\frac{3}{2}$ first excited state in Yb^{171} in Yb_2O_3 is also given in Table I. The number is within the limits of error the same as found earlier.[22]

$$eQ_{3/2}V_{zz}=20.7\pm0.4 \text{ mm/sec}=(4.60\pm0.08)\times10^{-6} \text{ eV}.$$

If the concept of a constant intrinsic quadrupole moment is truly valid in a rotational band, the ratio of $Q_{5/2}$ to $Q_{3/2}$ should only depend on the spin of these two levels. We can write, with $I_1=\frac{3}{2}$ and $I_2=\frac{5}{2}$:

$$\left(\frac{eQ_{5/2}V_{zz}}{eQ_{3/2}V_{zz}}\right)\Big/\frac{[3K^2-I_2(I_2+1)][(I_1+1)(2I_1+3)]}{[(I_2+1)(2I_2+3)][3K^2-I_1(I_1+1)]}$$

$$=\frac{(6.5\pm0.1)\times7}{(4.60\pm0.08)\times10}=0.99\pm0.03.$$

This ratio, being equal to one within the limits of error, verifies that the intrinsic quadrupole moment, and thus the static deformation of the nucleus,[23] are indeed not changed by a pure rotational excitation.

Another technique to show that the intrinsic quadrupole moment remains unaltered within a given rotational band is to determine the ratio of the reduced transition probabilities $B(E2)$ of the first and second rotational state in odd-A nuclei.[23] The experimental accuracy, however, is generally around 10–20%.[5] In the case of Yb^{171}, the experimental determination of the ratio $B_2(E2)/B_1(E2)$ is somewhat more difficult since, owing to the closeness of the two excitation energies, the lines of inelastically scattered particles cannot totally be resolved.[5]

Note added in proof. The magnetic splitting of the 75.9-keV level has very recently been observed in a Mössbauer experiment by Hennig *et al.*[24]

ACKNOWLEDGMENTS

We wish to thank M. C. Oselka of the Argonne National Laboratory Cyclotron group for his help and cooperation in producing the source activities. We benefited also from valuable discussions with several of our colleagues, especially with J. R. Gabriel, S. L. Ruby, and A. E. Blaugrund. One of us (M.K.) is also thankful to P. Kienle (T. H. Munich) for a useful discussion.

[22] The number 2.4×10^6 eV is equal to $\frac{1}{2}e^2qQ(1+\eta^2/3)$ rather than $\frac{1}{4}e^2qQ(1+\eta^2/3)^{1/2}$, as erroneously stated in Ref. 6a.

[23] K. Alder, A. Bohr, T. Huus, B. Mottleson, and M. Winther, Rev. Mod. Phys. **28**, 432 (1956).

[24] W. Hennig, P. Kienle, and H. J. Körner, Z. Physik (to be published).