while Kalos[12] obtained a value of −9.47 MeV. In comparing with our value of the upper bound, there is a difference of about 0.3 MeV.[13] Normally, a difference of this magnitude will not be considered as substantial, but since we consider the methods of both Baker *et al.* and Kalos as quite accurate, the fact that such a difference exists is somewhat astonishing. It is possible that the numerical accuracy of the result of Baker *et al.* may be impaired to a certain degree by their use of a rather large mesh spacing. In their numerical calculation, they used a mesh spacing of about 0.1 F, while we use a much smaller spacing of 0.005 F. In our code, such a small spacing is admissible, since double-precision arithmetic is employed whenever necessary to avoid error by truncation.

[12] M. H. Kalos, Phys. Rev. **128**, 1791 (1962).

[13] It is interesting to point out that already for a simpler trial wave function

$$\psi = \prod_{i<j=1}^{3} [\exp(-\alpha r_{ij}^2) + C\exp(-\beta r_{ij}^2)]$$

with three variational parameters, the upper bound is −9.63 ±0.04 MeV with 50 000 estimates.

IV. CONCLUSION

This investigation shows that the type of trial wave function used here is capable of yielding very accurate results. For both types of two-body potential considered, the gap between the upper and the lower bound is so small as to allow us to make a good estimate of the eigenvalue. Also, it is quite easy to employ this wave function in a numerical calculation. Although it may sometimes contain as many as eight parameters, at least four of them, namely, the separation distances d and the energy parameters e, can be assigned good starting values and need very little subsequent variation.

At present, we are using this type of trial wave function to investigate the binding energies of the alpha particle, the hypernuclei and the helium molecules. From the closeness of the upper and lower bound found in this calculation, we believe that reliable estimates of the binding energies will be obtained in all these cases.

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Mössbauer Effect in Tm^{169} and Total Internal Conversion of the 8.42-keV Transition*

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The absolute yield of the Mössbauer absorption of the 8.42-keV transition in Tm^{169} was determined for a thulium oxide and a thulium metal absorber. The 8.42-keV gamma ray was resolved from the L x rays of erbium by means of a flat lithium fluoride crystal diffraction spectrometer. From the observed Mössbauer absorption effect the total conversion coefficient $\alpha_{tot}=325\pm35$ and the magnetic transition rate of $B(M1, \frac{3}{2}\rightarrow\frac{1}{2})=5.1\times10^{-2}$ $(e\hbar/2Mc)^2$ was derived.

INTRODUCTION

THE total cross section for nuclear resonance scattering and, in particular, for Mössbauer scattering or absorption depends directly on the internal conversion coefficient of the gamma transition involved. It is important to have a knowledge of this coefficient if the aim of an experiment is to determine the Debye-Waller factors or to find the optimum conditions for a Mössbauer experiment. Conversely, the conversion coefficient can be deduced from a Mössbauer experiment if all the other conditions are known.

The present work deals with the determination of the conversion coefficient from studies of the Mössbauer effect in Tm^{169}. The 8.42-keV transition from the $\frac{3}{2}$ state to the $\frac{1}{2}$ ground state has been employed in several Mössbauer experiments.[1] This transition has predominantly magnetic-dipole character, and the state from which it originates is well understood[2] from the point of view of nuclear structure being a member of a rotational band. Its energy, however, is so low that a theoretical conversion coefficient can only be estimated from extreme extrapolations of Rose's[3] tables. Such an estimate is probably good to a factor of 2 only.

There is, however, an indirect way of estimating the value of the conversion coefficient. The lifetime of the

* This work was performed under the auspices of the U. S. Atomic Energy Commission.

[1] M. Kalvius, W. Wiedemann, R. Koch, P. Kienle, and H. Eicher, Z. Physik **170**, 267 (1962); M. Kalvius, P. Kienle, H. Eicher, and W. Wiedemann, *ibid.* **172**, 231 (1963); R. G. Barnes, E. Kankeleit, R. L. Mössbauer, and J. M. Poindexter, Phys. Rev. Letters **11**, 253 (1963); R. L. Cohen, Phys. Letters **5**, 177 (1963).

[2] E. N. Hatch, F. Boehm, P. Marmier, and J. W. M. DuMond, Phys. Rev. **104**, 745 (1956); P. Alexander and F. Boehm, Nucl. Phys. **46**, 108 (1963).

[3] M. E. Rose, *Internal Conversion Coefficients* (North-Holland Publishing Company, Amsterdam, 1958).

8.42-keV state has been determined accurately in two recent experiments.[4,5] The magnetic gamma transition rate $B(M1)$ can be derived with help of the rotational model from the properties[6] of the rotational spectrum. The total conversion coefficient $\alpha_{\text{tot}}=\alpha_M+\alpha_N+\cdots$ follows directly from these quantities and turns out to be 300. There is, on the other hand, an experimental value $\alpha_M+\alpha_N=106\pm6$ obtained by Charpak and Suzor[7] with a proportional counter coincidence technique. The disagreement between this experimental value and the estimate is rather puzzling and, if true, might shed some light on the validity of the predictions of the magnetic properties by the rotational model. The present investigation was made to help clarify this question. It was found that the conversion coefficient agrees well with the prediction of the rotational magnetic transition rate.

FORMULATION OF THE PROBLEM

The determination of the total conversion coefficient α_{tot} is based on the measurement of the maximum resonant-absorption cross section

$$\sigma_0(\text{barns})=\frac{2.45\times10^9}{E_0^2(\text{keV})}\frac{2I_B+1}{2I_A+1}\frac{1}{1+\alpha_{\text{tot}}},$$

which appears in the cross-section formula

$$\sigma(E)=(\Gamma/2)^2\frac{\sigma_0}{(E-E_0)^2+(\Gamma/2)^2}. \tag{1}$$

E_0 is the energy of the transition, I_B, I_A are the spins of excited and ground states, and Γ is the total width of the excited level.

In a transmission-type Mössbauer experiment the observed absorption effect is

$$\mathcal{E}(v)=[N(\infty)-N(v)]/N(\infty),$$

$N(v)$ and $N(\infty)$ being, respectively, the counting rate at velocity v between source and absorber and the rate at a high velocity where resonance absorption cannot occur. This absorption effect can be expressed as follows:

$$\mathcal{E}(v)=\beta f_S\int_{-\infty}^{+\infty}\left(1-\exp\frac{(\Gamma/2)^2\sigma_0 n_A f_A}{(E-E_0)^2+(\Gamma/2)^2}\right)\times\frac{\Gamma}{2\pi}\frac{1}{[E-E_0-E_0(v/c)]^2+(\Gamma/2)^2}dE. \tag{2}$$

f_S and f_A are the Debye-Waller factors of source and absorber, respectively, n_A is the number of nuclei of the particular isotope per cm^2, and β is the fraction of the gamma rays due to the transition in question from all the detected events in the discriminator window of the amplifier.

If due to disturbances of the lattice or other reasons the nuclei do not possess equivalent electromagnetic surroundings the composite action of all source and absorber nuclei can give rise to a broadening effect of the emission and absorption lines. This broadening effect does not change the energy integrals over these lines. If we make the assumption that both lines are broadened by a factor κ so that the Lorentzian shape remains, we have to replace in Eq. (2) Γ by $\kappa\Gamma$. Since $\int_{-\infty}^{+\infty}\sigma(E)dE=\sigma_0\pi\Gamma/2$, the quantity $t=\sigma_0 n_A f_A$ in the exponent has to be divided by κ. With these modifications and using calculations of Bykov and Hien[8] we get for the absorption effect at zero velocity for a thin absorber $t=\sigma_0 n_A f_A\ll1$:

$$\mathcal{E}(0)=\tfrac{1}{2}\beta f_S\frac{t}{\kappa}\left[1-\frac{3}{8}\frac{t}{\kappa}+\frac{5}{48}\left(\frac{t}{\kappa}\right)^2-\cdots\right] \tag{3}$$

and for the area A under the absorption curve

$$A=\frac{\pi}{2}\frac{\Gamma c}{E_0}\beta f_S t\left[1-\frac{1}{4}\frac{t}{\kappa}+\frac{1}{16}\left(\frac{t}{\kappa}\right)^2-\cdots\right]. \tag{4}$$

Consequently we have

$$\frac{A}{\mathcal{E}(0)}=\pi\frac{\Gamma c}{E_0}\kappa\left[1+\frac{1}{8}\frac{t}{\kappa}+\frac{1}{192}\left(\frac{t}{\kappa}\right)^2-\cdots\right]. \tag{5}$$

Since (5) is rapidly converging one gets from a measurement of $A/\mathcal{E}(0)$ a good determination of κ if Γ is known and if t can be estimated to be sufficiently small. With the value of κ, Eq. (4) can be used to derive t and consequently α.

If one replaces in the expansions of (3) to (5) t/κ by $t/2\kappa$, these equations are approximately valid for equally split source and absorber patterns as in our measurements with the oxide source and oxide absorber.

The present experiment consists of two parts. In the first, the ratio β was determined with the help of a crystal spectrometer arrangement. In the second part, the area A was found from a Mössbauer experiment.

EXPERIMENTAL ARRANGEMENT

In most experiments on the Mössbauer effect in Tm^{169}, gas proportional counters have been used. In these counters the 8.42-keV gamma ray can not be resolved from the fluorescent L x rays of erbium which occur between 6.9 and 9.1 keV. Since in the present experiment the knowledge of the size of the Mössbauer effect is relevant, it is necessary to resort to a technique which enables us to resolve the gamma line from the L

[4] T. Sundström, P. Sparrman, J. O. Lindström, and J. Lindskog, Phys. Letters **6**, 56 (1963).
[5] R. E. McAdams, G. W. Eakins, and E. N. Hatch, Phys. Letters **6**, 219 (1963).
[6] F. Boehm, J. de Boer, and D. Bowman, Conference on Perturbed Angular Correlations, Uppsala, 1963 (unpublished).
[7] G. Charpak and F. Suzor, J. Phys. Radium **20**, 33 (1959).
[8] G. A. Bykov and P. Z. Hien, Zh. Eksperim. i Teor. Fiz. **43**, 909 (1962) [English transl.: Soviet Phys.—JETP **16**, 646 (1963)].

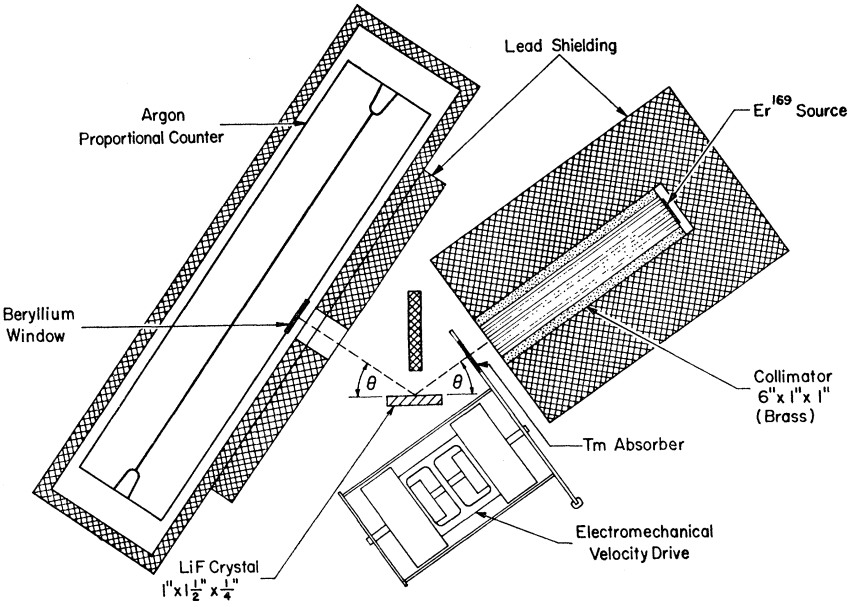

FIG. 1. Experimental arrangement.

x rays. The well-known method of x-ray diffraction from a single crystal has therefore been employed in the present experiment. A lithium fluoride crystal was cut along the cleavage plane and a 1-×1½-in. flat piece was used as a Bragg crystal. Reflection from the 200 planes having a lattice constant of $2d=4.01$ Å gave good efficiency for the wavelengths in question. However, the diffraction linewidth was much too narrow for the present application. To increase the intensity of the reflected beam, a large surface mosaic structure was introduced artificially by sandblasting the crystal face. This widened the diffraction linewidth to about 0.5 deg. The source was placed behind a parallel-plate collimator, 6 in. long. This collimator had a angular definition of about 0.5 deg matching the crystal mosaic. The diffracted radiation was observed at the Bragg angle in an argon-methane proportional counter. The setup is depicted in Fig. 1. With a fixed single-channel window for the proportional counter pulses equivalent to gamma-ray energies from 5 to 10 keV the diffraction curve was measured for a Er^{169} source.[9] Figure 2A shows the result of such a run.

For the analysis we have used a pure erbium L x-ray spectrum as shown in Fig. 2B. This spectrum was obtained by irradiating the source which gave rise to the curve 2A with a powerful x-ray beam from a 100-keV x-ray tube. The x-ray tube could be approached close to the source so that the geometry of Fig. 1 was unchanged. The fluorescent x-ray intensity produced in this fashion was much stronger than the x-ray intensity due to radioactivity as shown in the respective ordinate scales of Figs. 2A and 2B. After normalization to the Er L_β peak, the intensity of the 8.42-keV gamma ray could be derived easily by decomposition (lower curve in Fig. 2A).

A small correction had to be applied for the presence of Tm^{170} in our source. During the 3-week irradiation cycle of the Er^{168} sample in the reactor an appreciable amount of Tm^{169} was formed which due to its large capture cross section gave rise to a noticeable Tm^{170} radioactivity. A study was made of this contribution by measuring with a 1-mm-thick NaI crystal the x-ray and gamma-ray spectrum of the source. The time decay of the peak at 8 keV normalized to the peak at 84 keV due to the Tm^{170} gamma line revealed a 9% contribution by the Yb L x rays to the 8-keV peak at the time of the oxide measurement. The Yb L_β lines just coincide in energy with the 8.4-keV gamma line of Er^{169}. The Yb L_α lines were not resolved in the experiment (Fig. 2A) but are expected to occur in the valley between the strong Er x-ray groups.

In the experiment illustrated in Fig. 2A, a 1.15-mg/cm² thulium oxide Mössbauer absorber was placed in the beam between collimator and crystal. A 8.5-mg/cm² thulium oxide absorber and a thulium metal absorber (2.03 mg/cm²) were also used. The absorbers were prepared in the following way:

The 1.15-mg/cm² thulium oxide absorber was obtained by vacuum evaporation of thulium oxide onto a 1-mm beryllium disk. Use was made of a device for evaporation of high melting point material by electron bombardment described by de Boer *et al.*[10] The 8.5-mg/cm² absorber consisted of a mixture of thulium

[9] The source was obtained by neutron capture of 100-mg erbium oxide enriched in Er^{168} and had a gamma-ray strength of about 10 mCi. An effective reduction of the number of fluorescent x rays was achieved by diluting the erbium oxide with about 400 mg of wax. The erbium oxide-wax mixture was deposited evenly on a 1-in.-diam beryllium disk.

[10] Y. Dar, H. M. Loebenstein, and J. de Boer, Nucl. Instr. Methods (to be published).

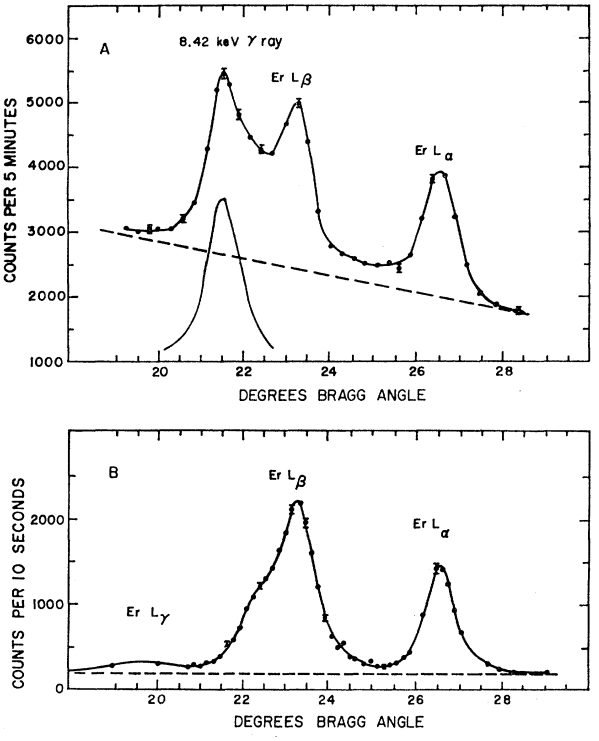

FIG. 2. A—Diffraction spectrum of Er^{169}. B—Diffraction spectrum of Er x rays.

oxide, diamond powder, and wax shaped into a cylindrical disk with a pill press. The metal absorber was a vacuum-evaporated thulium deposit on beryllium.

For the measurement of the Mössbauer absorption cross section in the crystal diffraction spectrometer the absorbers were mounted on to a frame which could be moved by an electromechanical drive system. This system has been described in detail.[11] It consists essentially of two mechanically coupled loudspeaker systems which are electrically connected by a feedback circuit. One is driving, the other picking up a velocity proportional signal which is compared with a reference signal. The amplified error signal actuates the driving system.

In synchronization with a triangular wave form of the reference signal a multichannel analyzer operating in the multiscaler mode is scanning through the channels of the analyzer and counting the pulses received from a single-channel analyzer. A velocity spectrum is represented by a diagram of number of counts per channel versus channel number. Each channel number corresponds to a certain velocity interval.

RESULTS

Measurement with 1.15-mg/cm² Tm_2O_3 Absorber

The crystal diffraction spectrum shown in Fig. 2 gives easily the ratio β, the intensity of the 8.42-keV line to the total intensity of gamma ray and x rays for each setting of the monochromator. In particular, if the monochromator was set on the peak of the 8.42-keV line this ratio was $\beta=0.40$. The number of counts in this monochromator setting was then measured for zero velocity and for a sine motion of the 1.15-mg Tm_2O_3 absorber. A correction was applied taking into account the averaging effect of the sine wave motion. The absorption effect with this correction applied was $\mathcal{E}(0)=2.8\%$.

Next, the entire velocity spectrum was measured for the same source and absorber in a conventional Mössbauer transmission geometry. The spectrum obtained is shown in Fig. 3. The value of $A/\mathcal{E}(0)=5.5$ cm/sec was derived. If we insert the total width Γ as given by the measured half-life of 4.0 nsec (see discussion) and the Debye-Waller factor $f_S=f_A=0.925$ ($\theta=240°$K),[12] we find the conversion coefficient $\alpha_{tot}=340\pm40$.

An experiment with an 8.1-mg/cm² thulium oxide absorber was performed in the same fashion as described above. The conversion coefficient turned out to be considerably larger than the above value. However, a measurement of the grain size of the oxide powder showed that the diameter of the grains gave rise to local absorber thicknesses greater than 8.1 mg/cm² and that $t/\kappa>1$ meaning that the absorber was very inhomogeneous. These measurements were therefore considered inconclusive.

Measurement with 2.03-mg/cm² Tm Metal Absorber

A further measurement was performed with a metal absorber of 2.03 mg/cm² produced by evaporation of the metal onto a beryllium disk. In this case the absorber had a broadened single line. Formulas (3) and (5) can not be applied but (4) is still correct.

The crystal diffraction spectrum with the absorber in the beam looks similar to Fig. 2A. A value $\beta=0.31$ was evaluated from this measurement. The area in this case is determined from the velocity spectrum (Fig. 4) which

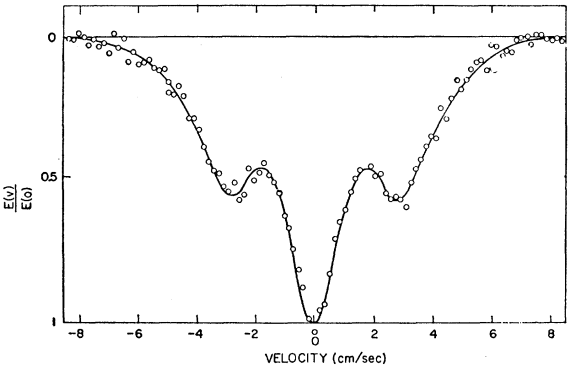

FIG. 3. Velocity spectrum with Tm_2O_3 absorber.

[11] E. Kankeleit, Rev. Sci. Instr. **35**, 194 (1964).

[12] Quoted in R. L. Cohen, thesis, California Institute of Technology, 1962 (unpublished).

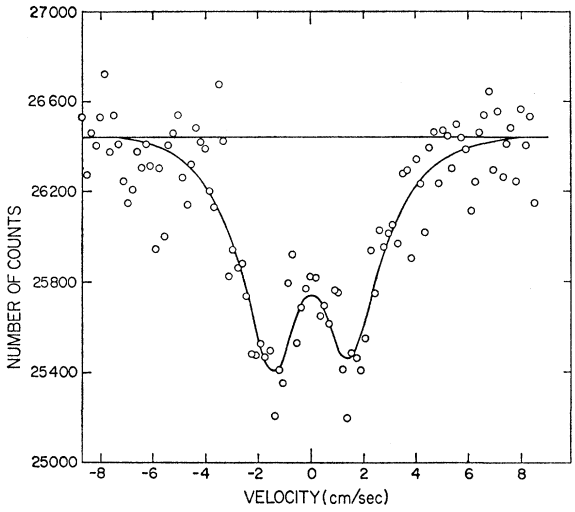

FIG. 4. Velocity spectrum with Tm metal absorber measured in the diffraction spectrometer.

had been measured for a period of 3 days in the crystal spectrometer. The diffraction angle was set on the peak position of the 8.4-keV line. The solid line drawn was obtained from a conventional direct beam Mössbauer experiment with good statistics and adjusted to the experimental points by varying the abscissa scale and the base line for a best fit. The area deduced this way was $A=0.23$ cm/sec. Using a Debye-Waller factor for the absorber $f_A=0.83$ ($\theta=160°$K).[12] The conversion coefficient finally is $\alpha_{tot}=300\pm50$.

Summarizing the results with oxide and metal absorbers we present a final result for the total internal conversion coefficient of the 8.42-keV line:

$$\alpha_{tot}=325\pm35.$$

DISCUSSION

The conversion coefficient obtained in the present experiment $\alpha_{tot}=325$ can be compared with the measurement of Charpak and Suzor[7] giving $\alpha_M+\alpha_N=106\pm6$. Unfortunately we can not offer an explanation of this serious disagreement.

Using our formulas (1) to (5) we can also calculate the gamma transition strength, $T_\gamma(M1)$ for the 8.4-keV line. However, having derived the conversion coefficient it is simpler to give $T_\gamma(M1)$ in terms of α. There are two recent determinations of the half-life of the 8.4-keV level by Sundström *et al.*[4] and by McAdams *et al.*[5] giving $\tau_{1/2}=3.45\pm0.25$ and $\tau_{1/2}=4.36\pm0.17$ nsec, respectively. Although the two results are in poor agreement we can tentatively expand the error limits and take a consolidated value of $\tau_{1/2}=4.0$ nsec. Since the conversion coefficient $\alpha_{tot}=[\alpha_{M1}+\delta^2\alpha_{E2}]\gg1$, we find

$$T_\gamma(M1)=\ln2/\tau_{1/2}[\alpha_{M1}+\delta^2\alpha_{E2}]=5.3\times10^5\ \text{sec}^{-1}$$

and

$$B(M1,\tfrac{3}{2}\rightarrow\tfrac{1}{2})=5.1\times10^{-2}\ \text{in units}\ (e\hbar/2Mc)^2.$$

A comparison with the prediction of the rotational model is now possible. In this model we have

$$B(M1,\tfrac{3}{2}\rightarrow\tfrac{1}{2})=0.02(g_K-g_R)^2(1+b_0)^2=5.47\times10^{-2},$$

where the intrinsic and the rotational g factors are taken to be $g_K=-1.57$, $g_R=0.406$ and the decoupling parameter $b_0=-0.16$ as derived from other experimental data in this rotational band and quoted in Ref. 6. The agreement between the experimental $B(M1)$ and the estimate from the model is satisfactory, indicating that the rotational model predicts the magnetic transition rate within the accuracy of present experiments.

An estimate of the branching ratio δ^2 can be obtained in the following way. The $B(E2)$ for electric quadrupole excitation of the $\frac{1}{2}\rightarrow\frac{5}{2}$ transition has been measured by Elbek.[13] We know that the intrinsic quadrupole moment is the same for the states in a rotational band and can, therefore, derive $B(E2,\frac{3}{2}\rightarrow\frac{1}{2})=1.12$. This yields for the branching ratio

$$\delta^2=T(E2)/T(M1)=0.9\times10^{-3}.$$

Independently this branching ratio can be obtained from the observed M-subshell conversion ratios. Experimental data by Hatch and Boehm[14] and by Shliagin *et al.*[15] give about 10^{-3} for δ^2 if one resorts to an extreme extrapolation of Rose's[3] M-conversion ratios. In this extrapolation the total absolute conversion coefficient is about 500, which is higher than the present experimental value.

The following possible criticism should be mentioned. Our measurement includes the assumption that in the velocity spectrum, $N(\infty)$ is the counting rate *outside* the range of recoil-free absorption. In other words, we assume that the overlap of the two hyperfine spectra gives just the spectrum as shown in Figs. 3 and 4 and that this spectrum is not superimposed on a broad resonant-background spectrum. Since the *integral* over the velocity curve is relevant in yield measurements, even a small background can be significant if it extends to high velocities. Possible causes for such a resonant background could be high-field distortions at grain boundaries or distortions due to the preceding beta decay. In the case of beta decay the charge exchange in the atomic shells may not be completed when the gamma ray is emitted. It is also conceivable that due to the recoil following beta decay the atom is lifted into one of its higher excited crystalline field levels. But all these effects seem to us rather unlikely to occur because the times involved are too short and the hyperfine splittings are probably not large enough. None of these effects have been observed so far. If background effects of this sort were present, the appropriate correction would tend to lower the reported conversion coefficient.

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[13] M. C. Olesen and B. Elbek, Nucl. Phys. **15**, 134 (1960).
[14] E. N. Hatch and F. Boehm, Bull. Am. Phys. Soc. **1**, 390 (1956).
[15] K. N. Shliagin and P. S. Samoilov, Zh. Eksperim. i Teor. Fiz. **34**, 29 (1958) [English transl.: Soviet Phys.—JETP **7**, 20 (1958)].