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Mössbauer-Effect Observations of Th^{232} following Coulomb Excitation*

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Resonant absorption following Coulomb excitation has been observed for γ rays from the first excited state of Th^{232} in Th metal and ThC targets with ThO_2, Th metal, ThN, and ThC_2 absorbers. Most spectra were obtained with targets at 30 °K and absorbers at 25 °K using 1-μA beams of 6-MeV He^{4+} ions. Absorber and target recoilless fractions (f_a and f_s) and zero-thickness absorber linewidth (Γ_0) were determined from a series of measurements of the width (half-width at half-maximum) of absorption spectra versus absorber thickness. These spectra were obtained with Th metal targets and ThO_2 absorbers. We find that $f_a = 0.35 \pm 0.04$ and $f_s = 0.31 \pm 0.06$, which correspond to a Debye temperature of (121 ± 13) °K. The zero-thickness linewidth Γ_0, uncorrected for target-absorber geometry, was found to be 11.35 ± 0.26 mm/sec. The value of Γ_0, corrected for broadening due to the geometry of the experimental configuration, was estimated to be 9.6 ± 0.4 mm/sec.

I. INTRODUCTION

For several years it has been realized that, in principle, the Mössbauer effect could be used to study the actinide elements and their γ transitions from the low-lying levels.[1] With the exception of Np^{237},[2,3] Am^{243},[4] and U^{238},[5] such studies have not been carried out because of the lack of suitable radioactive sources. Oleson *et al.*[6] partially overcame this difficulty for U^{238} by using Coulomb excitation to excite the 2^+ level. However, data accumulation was slow because of the large internal-conversion coefficient of U^{238} (approximately 600). Since large internal-conversion coefficients characterize nearly all actinide nuclei, Mössbauer-effect experiments utilizing resonant absorption following Coulomb excitation (RACE) inevitably encounter the same difficulty with count rates. Th^{232} is suitable for RACE experiments because the ground state has an extremely long half-life (1.4×10^{10} yr) and the 49.8-keV $E2$ transition from the first excited state is relatively far from the characteristic K and L x rays.

The first Mössbauer-effect measurements of Th^{232} were made by Hershkowitz *et al.*[7] following Coulomb excitation by a 1.5-μA beam of 4.5-MeV He^{4+} ions. The absorber (ThO_2) and target (Th metal) were cooled to near liquid-nitrogen temperatures. The half-width at half-maximum (HWHM) of the observed Mössbauer line was found to be 8.35 mm/sec. We have reevaluated the uncertainty of this measurement using an improved statistical procedure and also taking into account some systematic errors and find that the HWHM was 8.4 ± 1.4 mm/sec which corresponds to a half-life of 0.33 ± 0.06 nsec. This result agrees with earlier electronic measurements of 0.345 ± 0.015 nsec.[8]

The primary difficulties encountered by Hershkowitz *et al.* were low count rate (about 120/sec) and a small percent effect (about 1.2%) which could be attributed, respectively, to substantial internal conversion (the internal-conversion coefficient for Th is approximately 255)[9] and to small recoilless fraction at liquid-nitrogen temperature. The con-

ditions necessitated using the University of Iowa's Van de Graaff accelerator continuously for about a week to obtain only *one* RACE spectrum.

In order to improve on the preliminary experiment previously reported, we have increased beam energies and detector area to increase count rates and used lower target and absorber temperatures to increase recoilless fractions. Th metal and ThC have been used for targets and ThO_2, Th metal, ThN, and ThC_2 have been used for absorbers. The linewidth dependence of ThO_2 absorption lines as a function of thickness has been measured to determine recoilless fractions.

II. EXPERIMENTAL CONDITIONS

A. General Considerations

Th^{232} nuclei in Th metal and ThO_2 targets were excited with 6.0-MeV beams of He^{4+}. A typical pulse-height spectrum taken with a Ge(Li) detector is given in Fig. 1.

Preliminary experiments by Hershkowitz *et al.*[7] and experiments with ThO_2 and ThC targets and with ThO_2, ThN, and ThC_2 absorbers made use of the apparatus described by Jacobs and Hershkowitz.[10] Targets and absorber temperatures were close to liquid-nitrogen temperature (77.3 °K). Most other experiments were carried out at temperatures near 25 °K using the apparatus described by Wender *et al.*[11] Details of target and absorber preparation were similar to those already described by Jacobs and Hershkowitz.[10]

B. Targets and Absorbers

Both Th metal and ThO_2 are convenient to use in RACE experiments because they are chemically stable materials with cubic structures and relatively high recoilless fractions, and are readily available. The other compounds used in these experiments are not stable in air. Debye-Scherrer x-ray powder patterns were made of all compounds used in these experiments.

The melting point of ThO_2 (3300 °C) is the highest of all oxides. High melting points of ThO_2, ThN, and ThC indicate stiff lattices and the possibility of observing the Mössbauer effect of these compounds at liquid-nitrogen temperature. Since the lattice of ThO_2 is cubic (CaF_2 type), with the thorium atoms forming a face-centered structure, one would not expect an electric field gradient at the thorium lattice site. A Debye-Scherrer powder pattern of ThO_2 gives a lattice parameter equal to 5.582 ± 0.010 Å in agreement with previous measurements. ThN slowly decomposes into ThO_2. Thorium carbide (ThC) and dicarbide (ThC_2) are extremely reactive to oxygen and nitrogen.[12] Therefore, a sample of ThC was prepared from ThO_2 *in vacuo* and a target made in a helium atmosphere. ThC was converted from ThO_2 using the reaction

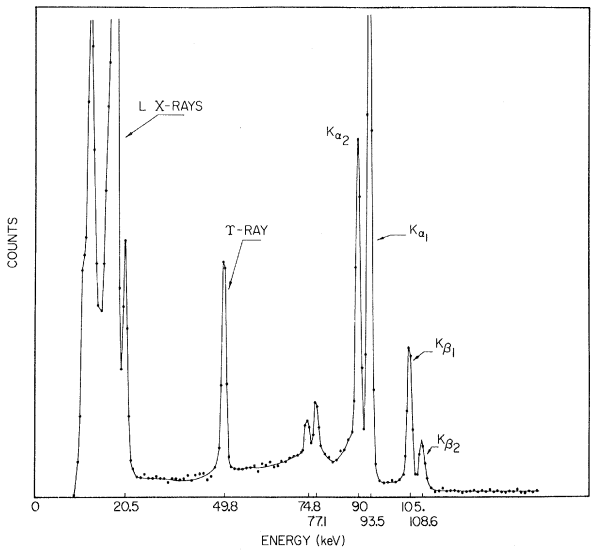

FIG. 1. Thorium pulse-height spectra taken with a Ge(Li) detector when 6.0-MeV α particles are used as bombarding beam.

$ThO_2 + 3C \rightarrow ThC + 2CO$ in an induction-heated carbon crucible located in an evacuated quartz condenser.

C. Detectors

Two detectors were used in the present experiment, a nuclear diodes Ge(Li) detector model LGP 7.0-7T with a 7-cm^2 crystal, and a Harshaw sodium iodide detector, type 8s.5Q-X, with a $\frac{1}{8}$-in. -thick 2-in. -diam NaI(Tl) scintillation crystal. The thorium Mössbauer-effect observations of Hershkowitz *et al.*[7] were made with a $\frac{1}{32}$-in. -thick 1-in. -diam Harshaw NaI(Tl) detector. The much higher count rate from the sodium iodide detector [usually about four times greater than the nuclear diodes Ge(Li)], which can be attributed to the larger detecting area, initially appeared to more than compensate for the smaller signal-to-noise ratio in comparison to the nuclear diodes detector. However, the percent absorption was inevitably greater with the Ge(Li) detector due to the superior resolution, and the data accumulation time needed for comparable statistics was about the same.

D. Velocity Calibration

Simultaneous Fe^{57} Mössbauer spectra [using a Co^{57} (Pd) source and an Fe^{57} metal absorber] were simultaneously taken with Th^{232} RACE spectra for velocity calibration as has previously been described.[10,11] The velocity reference triangle was also accurately scaled down from thorium velocities (± 5 cm/sec) to iron velocities (± 1 cm/sec) to improve velocity resolution. An absolute velocity calibration using a Michelson-type interferometer gave agreement to the calibration to better than 1%.

TABLE I. List of the thorium experiments, the distinguishing experimental conditions, and the observed HWHM.

Experiment No.	Absorber, thickness (mg/cm^2)	Temperature (°K) Target	Absorber	Detector	Count rate[a] at 1-μA beam (counts/sec)	HWHM (mm/sec)	Variance ratio
1	ThO_2 14.3	30.0±1.5[b]	25.5±1.0	Ge (Li)	425	11.26±0.67	1.14
2	28.2			NaI (Tl)	1000	11.95±0.30	1.31
3	73.9			Ge (Li)	180	13.34±0.21	1.28
4	73.9			NaI (Tl)	940		
5	136			NaI (Tl)	500	15.01±0.27	1.27
6	170			Ge (Li)	115	15.41±0.36	1.09
7	272			Ge (Li)	40	18.39±0.89	0.96
8[c]	ThO_2 73.9	30.0±1.0	25.0±1.0	Ge (Li)	180	13.48±0.10	1.08
9	ThO_2 73.9	40[d]	35[d]	NaI (Tl)	550	11.83±0.62	0.93
10	170			NaI (Tl)	450	13.76±0.72	1.04
11	ThO_2 73.9	78±1	78±1	NaI (Tl)	950	12.29±1.12	1.7
12	ThN 73.5	78±1	78±1	NaI (Tl)	410	13.00±1.33	1.0
13	Th 59.2	30.0±1.5	25.5±1.0	NaI (Tl)	900	12.03±0.27	1.1
14[e]	59.2				140	12.29±0.44	1.4
15[f]	59.2			Ge (Li)	100	11.15±0.45	0.92

[a]The count rate depends directly upon the bombarding beam current. The average beam current in these experiments ranged from 0.8 to 1.8 μA. The average variation in beam current for a given experiment was ±0.2 μA. A direct proportion was used to calculate the count rates at 1 μA of He^4 beam from the actual beam currents.

[b]The uncertainty denoted represents the average fluctuation in temperature and not the inaccuracy in the value of the measured temperature.

[c]The velocity in this experiment was reduced about $\frac{1}{4}$ and the data were accumulated in twice as many channels.

[d]These temperatures are estimates, since the exact temperatures are not known due to thermocouple failure.

[e]The bombarding beam was 4.0-MeV He^4 ions.

[f]This experiment used a $\frac{3}{4}$-in. circular collimator in front of the detector.

III. EXPERIMENTAL RESULTS

Thorium spectra were fitted to a single-line Lorentzian with a least-squares fitting program, i.e., no hyperfine splitting was assumed. Fifteen thorium Mössbauer spectra were obtained from which significant conclusions could be drawn. All but four of these experiments involved a thorium metal target and a thorium dioxide absorber. One experiment was done with a thorium metal target and a thorium nitride absorber. Three others were done with a metal target and metal absorber. Six different thicknesses of thorium dioxide absorbers were used. Table I lists the thorium RACE experiments, the absorber thicknesses, and some other distinguishing experimental conditions. A typical RACE run on thorium lasted from 24 to 36 h although a run as long as 114 h was made in the case of the 14-mg/cm^2 absorber.

The value designated for the absorber thickness is liable to two errors. One is the uncertainty due to weight measurement and small losses during the process of making the absorbers. This error is estimated at about ±1.0 mg/cm^2. The second error is the variation in the thickness over the surface of the absorber. This is estimated to be no greater than ±1.5 mg/cm^2.

A. Results of Thorium Observations

1. *Linewidths*

After acquisition of the thorium RACE data, the data were fitted to a single-line Lorentzian. The results of these fits for six different absorber thicknesses are shown in Fig. 2. A 0.002-in. thorium metal target was used throughout, while the ThO_2 absorber thickness varied from 14.3 to 272 mg/cm^2. The line broadening with increasing absorber thickness is clearly manifested.

Figure 3 shows the thorium Mössbauer spectra and the single-line fits for the two experiments (Nos. 9 and 10) carried out at about 40 °K (target temperature), the Th-ThO_2 experiment (No. 12) made at 78 °K, and the Th-ThN experiment (No. 13) at 78 °K. The relatively broad Mössbauer absorption dips at liquid-nitrogen temperature are unmistakable. One can also observe the sharp decrease in percent absorption of the Mössbauer dip at 78 °K.

The HWHM from the single-line fits of the thorium Mössbauer absorption dips are tabulated in Table I. The results from experiment No. 8 are not averaged with Nos. 3 and 4 because the velocity was much smaller and only the lower portion of the Mössbauer-effect dip was displayed.

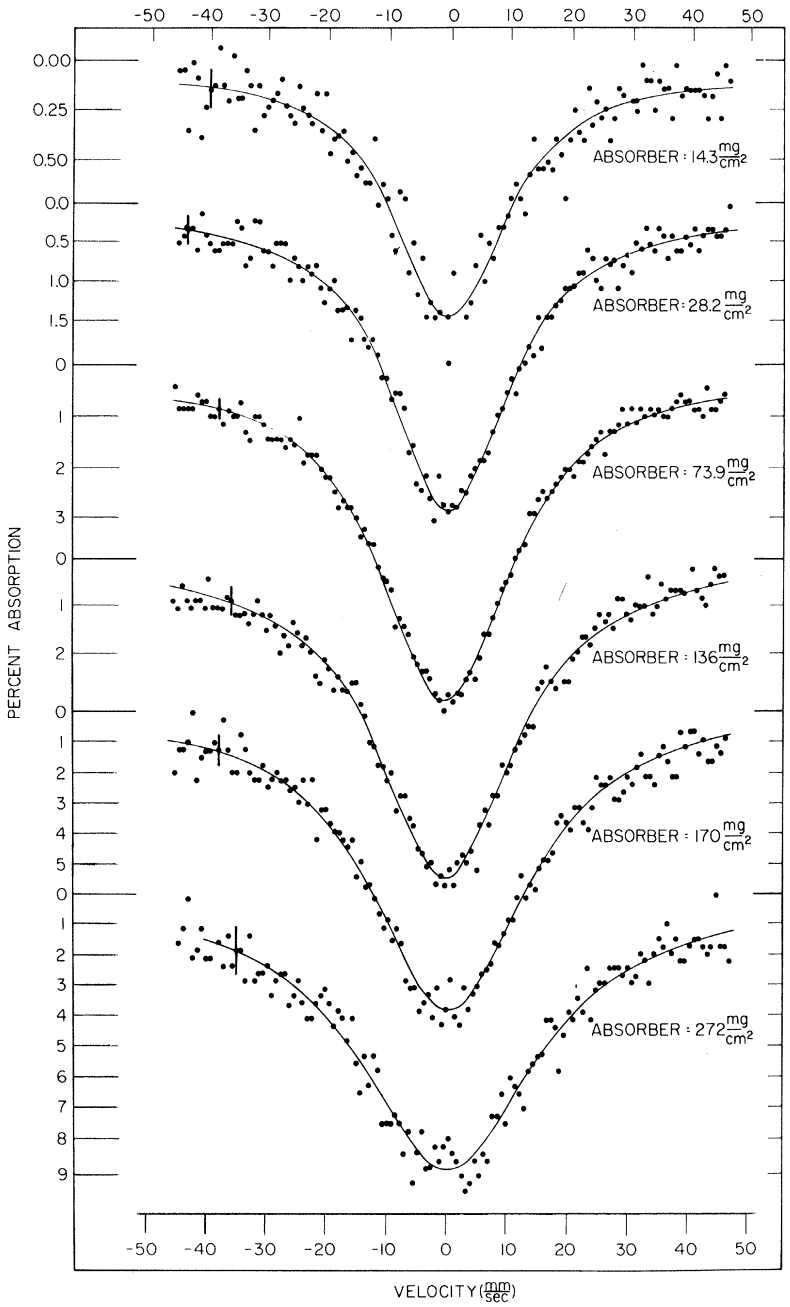

FIG. 2. Thorium Mössbauer-effect spectra for six different absorber thicknesses.

The HWHM of the Mössbauer spectra for these experiments at the target temperature of 30 °K are given as a function of absorber thickness in Fig. 4 along with the three observations (experiment Nos. 13–15) made with metal absorbers at the same temperature. The HWHM of the Th-ThO_2 spectra at 30 °K (target temperature) were fitted to a linear function of the form[13]

$$\text{FWHM (observed)} = 2\Gamma_a(1 + 0.135t), \quad 0 < t < 5$$

or

$$\text{HWHM (observed)} = \Gamma_a(1 + 6.32 f_a x),$$

where t refers to the effective absorber thickness, x is the thickness in g/cm^2, the constant is for the internal-conversion coefficient equal to 255 ± 16,[9] f_a is the absorber recoilless fraction, and Γ_a represents the natural linewidth of the absorber or the Mössbauer HWHM at zero-thickness absorber. The results of the above fits determined the zero-thickness intercept, and the slope of the curve determines f_a for this temperature (25 °K). These results were

$$\Gamma_a = 11.35 \pm 0.26 \text{ mm/sec}$$

and

$$f_a = 0.35 \pm 0.04 .$$

The final fit by the least-squares routine had a variance ratio of 0.477.

The value for f_a yielded values for the effective absorber thickness that were consistent with the assumptions of the fitting equation. The above linear equation is valid for $t < 5$; it also assumes that the source and the absorber have natural linewidths. If $f_a = 0.35$, then $t = 1$ corresponds to an absorber thickness of 57.7 mg/cm^2. This give a t value for the thickest absorber (272 mg/cm^2) of 4.71, which is consistent with the initial assumption.

Experiment Nos. 13 and 14, consisting of a Th metal target and metal absorber, were done to check the assumption that the linewidths for ThO_2 equaled the linewidth for Th metal. Experiment No. 14 was made with a 4.0-MeV α beam to see if any energy dependence from the bombarding beam existed. The resulting linewidths with Th metal

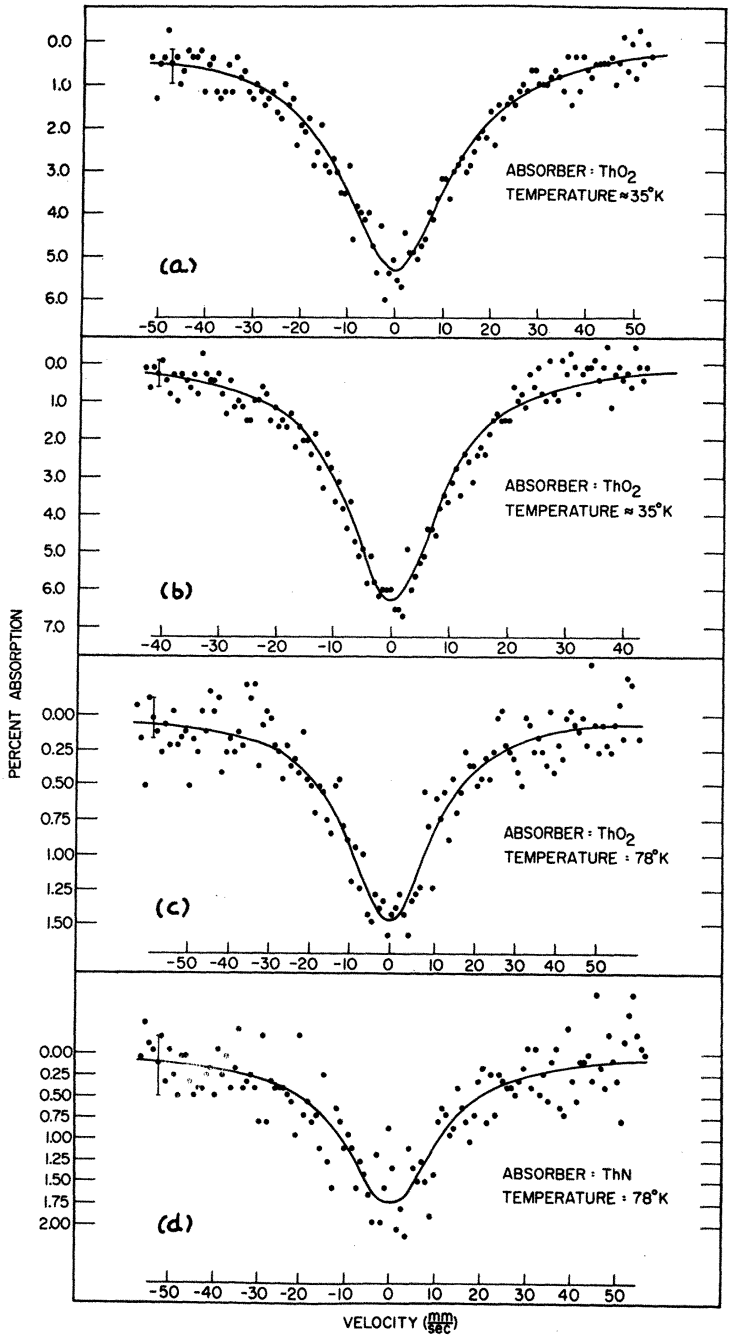

FIG. 3. (a) Th-ThO_2 (target-absorber) Mössbauer-effect experiment with a 73.9-mg/cm^2 absorber at about 35°K; (b) Th-ThO_2 experiment with 170-mg/cm^2 absorber at 35°K; (c) Th-ThO_2 experiment with a 73.9-mg/cm^2 absorber at 78°K; (d) Th-ThN experiment with a 73.5-mg/cm^2 absorber at 78°K.

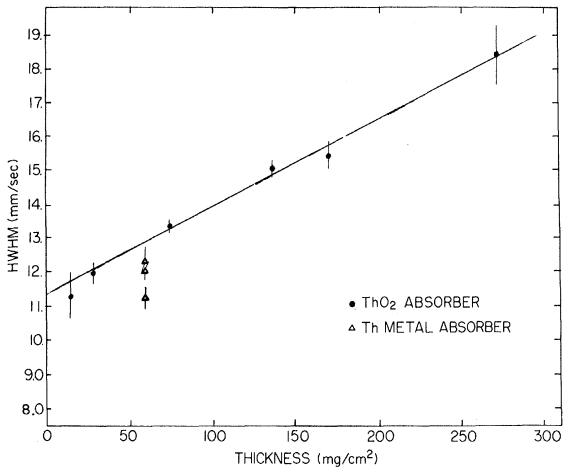

FIG. 4. HWHM of the Th^{232} Mössbauer spectra as a function of absorber thickness at a target temperature of 30 °K.

absorbers were only slightly less than the linewidths with ThO_2 absorbers, thus vindicating the assumption. No essential difference was found in the linewidths when using 6.0- and 4.0-MeV α beams.

2. *Source Recoilless Fraction*

Once the recoilless fraction of the absorber, f_a, is known and the percent effects at resonant energy are corrected for noise, the recoilless fraction of the source can be calculated. Using[13]

$$f_s = (\text{corrected percent effect})/[1 - e^{-t/2} J_0(it/2)] ,$$

where t refers to the effective absorber thickness and J_0 is the zeroth-order Bessel function, we calculated the recoilless fraction of the source, f_s, for each experimental run and tabulated them in Table II. Experimental run No. 1(14.3-mg/cm^2 absorber) has not been included because the percent absorption corrected for noise was not accurately determined. The corrected percent absorptions for experiments Nos. 3 and 4, both of which used a 73.9-mg/cm^2 absorber, were averaged in order to find a single value for f_s at that absorber thickness. With these corrections the weighted average recoilless fraction of the source was found to be $f_s = 0.31 \pm 0.06$.

A second approach in finding the recoilless fraction of Th^{232} metal was taken in which the data from the experiments consisting of a metal absorber as well as target (Nos. 13 and 14) were used. The above equation for the fractional absorption at resonance can again be used and the recoilless fraction solved for directly since the f_a in the effective absorber thickness can be assumed to be equal to f_s within the statistical uncertainties. The result after averaging the two experiments was $f = 0.36 \pm 0.04$.

3. *Debye Temperature*

For low temperatures ($T < 0.35\,\Theta_D$), f can be approximated by[13]

$$f = \exp\left\{ -\frac{3E_0^2}{Mc^2 K_B \Theta_D} \left[\frac{1}{4} + \frac{\pi^2}{6} \left(\frac{T}{\Theta_D} \right)^2 \right] \right\} ,$$

where E_0 signifies the nuclear transition energy, M refers to the mass of the nucleus, K_B is Boltzmann's constant, and Θ_D is the Debye temperature. When $T = 25$ °K and $f_a = 0.35 \pm 0.04$, it is found that $\Theta_D = (121 \pm 13)$ °K. The result is consistent with the approximation assumption, viz., $T < 0.25\Theta_D$. When $T = 30$ °K and $f_s = 0.31 \pm 0.06$ (the value for the Th metal target), then $\Theta_D = (121 \pm 20)$ °K also. This lends further support to the conclusion that the recoilless fractions of Th metal and ThO_2 are the same.

4. *Isomer Shift and Hyperfine Structure*

In order to determine whether thorium (metal vs ThO_2) exhibited an isomer shift and any hyperfine structure, an experiment (No. 8) was undertaken at a small velocity compared to the usual thorium velocities in order to focus on the bottom portion of the absorption dip. In addition, the data were accumulated in 512 channels of the multichannel analyzer, twice as many as usually are used. Thus, the resolution was improved about 9 times.

The results of this experiment along with the simultaneously acquired Fe^{57} Mössbauer spectrum are shown in Fig. 5. The measured isomer shift was $+0.08 \pm 0.09$ mm/sec. The isomer shift was determined from the Fe-Pd isomer shift of the simultaneously acquired Fe spectrum.[14]

5. *Mössbauer Experiments with* ThC, ThC_2, *and* ThN

The results of a thorium RACE experiment with a metal target and a ThN absorber have been shown in Fig. 3 and listed in Table I. The experiment was done at 78 °K. The observed linewidth (13.0 ± 1.3 mm/sec) was broader than the zero-

TABLE II. Recoilless fractions of Th^{232} at 30 °K as determined from the effective absorber thicknesses and the fractional absorptions at resonance.

Experiment No.	Absorber thickness (mg/cm^2)	Effective absorber thickness	Corrected percent effects (%)	f_s
2	28.2	0.488	8.85 ± 1.00	0.43 ± 0.16
3,4	73.9	1.28	16.68 ± 1.28	0.40 ± 0.14
5	136	2.36	16.55 ± 1.10	0.29 ± 0.12
6	170	2.94	18.28 ± 1.32	0.30 ± 0.13
7	272	4.72	14.97 ± 2.25	0.22 ± 0.12

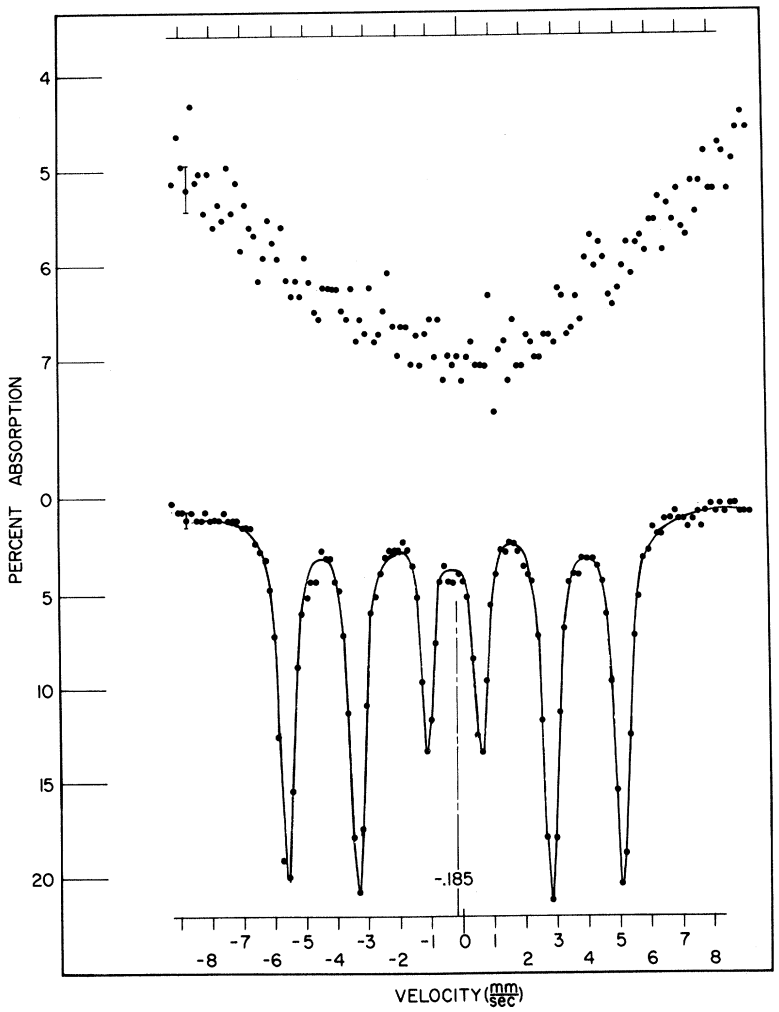

FIG. 5. Bottom portion of a Th-ThO_2 (73.9-mg/cm^2) Mössbauer absorption dip. The simultaneous Fe^{57} calibration spectrum is also shown.

thickness ThO_2 linewidth (11.35 ± 0.26 mm/sec).

Another Mössbauer experiment was carried out using a ThC target and a Th metal absorber. The observed Mössbauer effect at 78 °K was barely discernable after several hours of running and no conclusions could be made.

A third experiment with a thorium metal target and a ThC_2 absorber was also conducted at 78 °K. A very small percent effect (0.7%) and a line about as broad as the ThN experiment was observed.

B. Discussion

One result of the foregoing thorium RACE experiments was the determination of the zero-thickness absorber linewidth. In order to accurately state the experimentally determined zero-thickness linewidth of the first excited state of Th^{232}, one needs to correct the linewidth at zero-thickness absorber for self-absorption in the target. Since the range of 6-MeV α particles bombarding U^{238} is 24 mg/cm^2, the range of 6-MeV α in Th^{232} should be approximately the same. From other Coulomb excitation experiments[10] it is believed that most of the Coulomb excitation takes place within about one-half of the range, i.e., 12 mg/cm^2. Thus, the correction for self-absorption in the target can be estimated to be 12 mg/cm^2. Extrapolating the line fitting the Mössbauer HWHM as a function of absorber thickness (Fig. 4) to a point 12 mg/cm^2 to the left of zero-thickness absorber, one finds the value of 11.0 ± 0.3 mm/sec. The extrapolation is justified by the fact that the recoilless fraction of the absorber essentially equals that of the source.

Mössbauer-effect measurements of natural linewidths are often suspected of being too large, i.e., broader linewidths than actually exist, because any uncontrolled experimental factor will tend to broaden the observed Mössbauer linewidths. If 11.4 mm/sec is taken as the value of the natural linewidth for the first excited state of Th^{232}, it is about 40% broader than Bell's value from electronic measurements of the half-life. Radiation damage caused by Th nuclei recoiling out of their lattice

TABLE III. Summary of results.

Recoilless fraction of the absorber (f_a)	0.35 ± 0.04
Recoilless fraction of source (f_s)	0.31 ± 0.06
Absorber thickness at $t = 1$	57.7 mg/cm
Debye temperature (Θ_D) at $T = 25$ °K	(121 ± 13) °K
Isomer shift (Th metal target with ThO_2 absorber)	$+0.08 \pm 0.09$ mm/sec
Corrected Mössbauer HWHM at zero-thickness absorber	9.6 ± 0.4 mm/sec

sites and producing local vacancies is a possibility, but is usually not observed to any significant degree in other kinds of metal targets. However, Mekshes and Hershkowitz[15] have reported a 27% broadening in RACE spectra of W^{182} which can be attributed to radiation damage. On the other hand, electronic half-life measurements of less than 1 nsec are reaching the limits of the electronic timing capabilities. Nevertheless, the large discrepancy between previous electronic measurements of the half-life and our Mössbauer measurements leads one to suscept some sort of systematic error. Consider the effect due to collimation geometry. Numerous researchers have pointed to the necessity to control, or at least, to be cognizant of the effect of γ-ray collimation geometry on Mössbauer-effect measurements. For the special case of a point source and a thin flat absorber, and when emission and absorption lines both have Lorentzian line shapes, Hershkowitz[16] has shown that the resonant absorption energy scale is shifted by a factor $2/(1+\cos\theta)$ where θ is half-angle subtended by the absorber. Aramu and Maxia[17] have developed analytical expressions for the energy shift in the case of cylindrical geometry with a source and absorber of the same radius R separated by a distance d and where $R/d \ll 1$. The factor in this case is $1 + (2\pi/9\sqrt{3})(R/d)^2$. Neither of these two models applies directly to our experimental situation because (1) our target is smaller than the absorber yet obviously not a point source (target diameter $\cong 0.38$ in.; effective absorber diameter for NaI detector $\cong 0.94$ in.); (2) the shape of the source is nearly square and the Ge(Li) detector is rectangular in shape with a length almost twice the width; and (3) the radius of the absorber R_{ab} divided by the target-absorber distance d is not much less than one (for NaI detector, $R_{ab}/d = 0.50$). Nevertheless, one would expect a correction for geometrical effects somewhere between the two above extreme cases—a point source and a source which is as large as the absorber. Efforts to find analytically a correction for our particular geometry, viz., a small extended source with R/d as large as 0.5, have so far been unsuccessful. If we use the corrections of Hershkowitz for the geometrical configuration presented by our NaI(Tl), we find the resonant absorption energy shifted by a factor of 1.057. If we use the corrections of Aramu and Maxia we get 1.101.

Since an analytic expression for our geometrical configuration could not be derived and since an uncertainty in the widths of less than 2.5% could be achieved, comparison of two experiments, identical in all aspects except for the detectors, was performed. The results are listed below and indicate that there is no significant difference in the broadening due to collimation geometry for the geometrical configurations presented by the Ge(Li) and NaI(Tl) detectors:

	Ge(Li)	NaI(Tl)
HWHM	13.42 mm/sec	13.26 mm/sec
	±2.46%	±1.83%
Variance ratio	1.28	1.08

A final experiment, No. 15, was carried out to examine more carefully the effects of collimation geometry. A $\frac{3}{4}$-in.-diam circular collimator was placed in front of the Ge(Li) detector and an experiment was done with a Th metal target and a 0.002-in. Th metal absorber. This made R_{ab}/d approximately equal to 0.152. The observed linewidth was about 7.3% narrower than the linewidth without collimation (experiment No. 13).

Since f_a in the linear function fitting the HWHM vs thickness is not affected by geometrical corrections, a line with identical slope to the one drawn in Fig. 4 but now placed through the experimental point representing experiment No. 15 will estimate the zero-thickness absorber linewidth corrected for geometrical broadening. After correcting for self-absorption in the source by extrapolating the line fit 12 mg/cm^2 beyond the zero-thickness point, one finds an estimate for the natural linewidth equal to 9.6 ± 0.4 mm/sec. This procedure is justified by the fact that the recoilless fractions of ThO_2 and Th metal have been shown to be approximately equal. The above estimate for the natural linewidth agrees with the earlier determination by Hershkowitz *et al.*[7] but is slightly broader than would be expected from electronic measurements.

IV. CONCLUSION

The Mössbauer effect was observed following Coulomb excitation of Th^{232} in a Th metal target whose recoilless emission was resonantly absorbed by a ThO_2 or Th metal absorber. Six different ThO_2 absorber thicknesses were employed in order to determine the HWHM of the Mössbauer absorption dips as a function of absorber thickness. The fit of a linear function to the HWHM observations yielded a value for the recoilless fraction of the absorber equal to 0.35 ± 0.04. Experiments performed with Th metal absorbers confirmed the as-

sumption that the recoilless fractions of ThO_2 and Th metal were approximately equal. A summary of the results can be found in Table III.

The natural linewidth determined for the first excited state, after corrections were made for broadening due to the geometry of the experimental configuration and for self-absorption in the source, agreed with previous Mössbauer measurements but was slightly broader than would be expected from previous electronic half-life measurements. Experiments made at about 40 and 78 °K were consistent with the Mössbauer results at 30 °K.

No significant isomer shift was measured; no hyperfine structure was observed. RACE observations carried out on a ThN absorber at 78 °K indicated a linewidth at least as broad as with a ThO_2 absorber.

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Nuclear-Resonance Studies of Critical Fluctuations in FeF_2 above T_N*

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A detailed experimental study is made of the behavior of the F^{19} nuclear-resonance linewidth in the paramagnetic state of the uniaxial anisotropic antiferromagnet FeF_2. It is concluded that the observed linewidths are due to nuclear-electronic hyperfine interaction modulated by electron-spin motion. The portion $\delta\nu_{\parallel}$ of the linewidth due to the longitudinal component of the local field fluctuation is determined. The critical behavior of $\delta\nu_{\parallel}$ is found to be described by a power law $\delta\nu_{\parallel} \propto (T - T_N)^{0.67\pm0.02}$ for $0.04 < T - T_N < 1.5$ °K. This is in good agreement with predictions based on the extension by Riedel and Wegner of the dynamical-scaling theory to anisotropic systems.

I. INTRODUCTION

We have made detailed measurements of the F^{19} nuclear magnetic resonance (NMR) linewidth in the paramagnetic state of the anisotropic antiferromagnet FeF_2. A pronounced anomaly is observed near the critical temperature $T_N = 78.366$ °K. Our data provide information on the statics and dynamics of the critical fluctuation behavior.

Over the past few years, there has been considerable interest in the broad range of phenomena taking place near critical points.[1-3] Let us now briefly summarize these facts in a qualitative way, with reference to an antiferromagnet such as FeF_2. The order parameter, i.e., the sublattice magnetization,[4] approaches zero as the critical point $T_c = T_N$