

not be as sharp as those we have observed ($\delta g/C \approx 0.1$). However, it would be premature to draw a firm conclusion about this point because we should expect that the "soft optic *R*-corner mode," which becomes statically displaced in the tetragonal phase,^{8,11} may also contribute to the local crystal field.

According to the "zero-barrier-quantum-limit" model as well as the "infinite-barrier" model, Δg should level off below 5°K. To test this the *g* shift at 2.1°K was measured. Interestingly enough, Δg does not level off at 2.1°K (Fig. 3) and the system appears to behave in this region more like the "zero-barrier-classical-limit" model, whereas this is not the case above 5°K. In this connection, a second soft mode in SrTiO₃, the Cochran ferroelectric mode may be of importance. In this temperature region, the latter mode has a very low phonon frequency of $\omega < 10 \text{ cm}^{-1}$,³⁵ which is lower than both

³⁵ P. A. Fleury and J. M. Worlock, Phys. Rev. 174, 613 (1968).

rotational modes. Although linear J-T coupling is not possible for vanishing wave vector, such a coupling to this mode, whose frequency obeys the equation

$$\omega^2(q) = \omega_0^2 + \alpha q^2,$$

becomes allowed for $q \neq 0$. The strength of the $q=0$ in the summation is negligibly small as compared to the $q \neq 0$ values. Moreover, quadratic coupling for $q=0$ is allowed. Thus, it is possible that the ferroelectric mode yields such low rotational levels at this temperature that the system behaves more in the classical manner. To clarify this, more data points between 0.3 and 2°K are needed.

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Resonant Cross Section for 14.4-keV Gamma-Ray Absorption in Fe⁵⁷

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The resonant cross section and internal conversion coefficients of the 14.4-keV level in Fe⁵⁷ are determined from Mössbauer absorption spectra of thin polarized iron foils. We obtain a value for the maximum resonant cross section of $(2.56 \pm 0.05) \times 10^{-18} \text{ cm}^2$, which leads to a total internal conversion coefficient of 8.19 ± 0.18 . This value for the internal conversion coefficient is in excellent agreement with recent results from x-x-ray and x- γ -ray coincident experiments and is nearly 10% lower than the previously accepted value.

I. INTRODUCTION

THERE is some interest in determining the recoilless fraction of resonant absorbers f_a . In order to determine f_a from resonant absorption spectra, it is necessary to secure the maximum resonant cross section σ_0 . In the case of the ^{57}Fe 14.4-keV level in Fe⁵⁷, many attempts have been made to determine σ_0 . After initial very low reported values for σ_0 , the value has risen to a generally accepted value¹ of $(2.36 \pm 0.05) \times 10^{-18} \text{ cm}^2$. This value is based on the average of several Mössbauer determinations²⁻⁵ (2.42 ± 0.19 , 2.39 ± 0.15 , 2.31 ± 0.12 , 2.35 ± 0.09) $\times 10^{-18} \text{ cm}^2$, and three γ - γ -ray coincidence determinations^{1,3,6} (2.36 ± 0.12 , 2.31 ± 0.12 , 2.47 ± 0.23) $\times 10^{-18} \text{ cm}^2$. The latter experiments determine the total internal conversion coefficient α_T , which can in turn be used to give the maximum

resonant cross section through the expression⁷

$$\sigma_0 = 23.6 \times 10^{-18} / (1 + \alpha_T) \text{ cm}^2. \quad (1)$$

Recent x-x- and x- γ -ray coincidence experiments by Rubinson and Gopinathan⁸ have led to a significantly smaller value for α_T than is generally accepted. Through Eq. (1), this leads to a significantly larger value for σ_0 . Depending upon what auxiliary information is used, they arrive at $\alpha_T = 8.04 \pm 0.52$, 8.17 ± 0.25 , and 8.28 ± 0.21 which in turn leads to $\sigma_0 = (2.61 \pm 0.14$, 2.57 ± 0.07 , $2.54 \pm 0.06) \times 10^{-18} \text{ cm}^2$. Their results are further supported by energy extrapolation of the theoretical values of Rose⁹ and by *Z* extrapolation of the theoretical values of Hager and Seltzer¹⁰ which lead to estimates for α_T of 8.17 and 8.12, respectively.⁸

⁶ G. Moreau and G. Ambrosino, Comp. Rend. 261, 4538 (1965).

⁷ This is an evaluation of the expression $\sigma_0 = 2\pi\lambda^2\omega[1/(1+\alpha_T)]$ with $\lambda^2 = 1.88 \times 10^{-18} \text{ cm}^2$, and $\omega = 2$. See, for example, H. Frauenfelder, *The Mössbauer Effect* (Benjamin, New York, 1962).

⁸ W. Rubinson and R. P. Gopinathan, Phys. Rev. 170, 969 (1968).

⁹ M. E. Rose, *Internal Conversion Coefficient* (Interscience, New York, 1958).

¹⁰ R. S. Hager and E. C. Seltzer, California Institute of Technology Report No. CALT-63-60, 1967 (unpublished).

¹ A. H. Muir, K. J. Ando, and H. M. Coogan, *Mössbauer Effect Data Index, 1958-1965* (Interscience, New York, 1966).

² S. S. Hanna and R. S. Preston, Phys. Rev. 139, A722 (1965); 142, 286(E) (1966).

³ O. C. Kistner and A. W. Sunyar, Phys. Rev. 139, B295 (1965).

⁴ G. R. Isaak and U. Isaak, Phys. Letters 17, 51 (1965).

⁵ R. H. Nussbaum and R. M. Housley, Nucl. Phys. 68, 145 (1965).

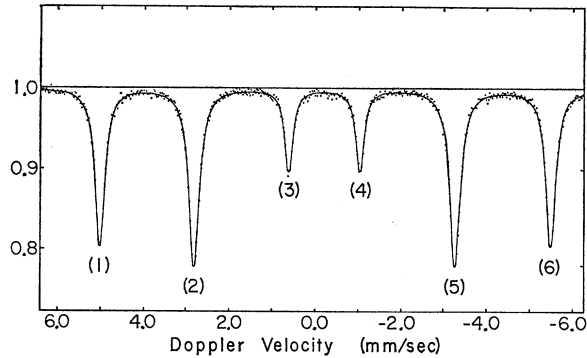


FIG. 1. Mössbauer absorption spectrum of the $\text{Cu}(\text{Co}^{57})$ source versus the 14.5-mg/cm^2 unenriched Fe foil fully polarized in the plane perpendicular to the transmitted beam of radiation. The solid line is the least-square fit to the spectrum. The circles are the normalized experimental data. The rms deviation of the experimental points from the fitted curve is $\pm 0.2\%$.

In view of this 9% discrepancy between the results of Rubinson and Gopinathan and the accepted value, we have performed a careful Mössbauer determination of σ_0 and α_T . Our results are in excellent agreement with that of Rubinson and Gopinathan.

II. EXPERIMENTAL METHOD

The method used for determining σ_0 and α_T is similar to that outlined in Ref. 5. The recoilless resonant cross section $f_a\sigma_0$, of two thin, polarized, unenriched, 99.996% iron foils are determined from the integrated intensity of Mössbauer absorption spectra and independent determinations of the recoilless fraction associated with the source f_s .

The integrated intensity A_i of each of the six hyperfine absorption lines is determined from a nonlinear least-square fit of the absorption data to a sum of six Lorentzian equations (see Fig. 1). The results of this investigation are based on six such spectra. As a check on the value of the base line determined by the computer fit, the ratio of the count rate at zero velocity to that at high velocity (20 mm/sec) was measured and compared to the ratio of the zero velocity count to the base line count given by the computer fit. Excellent agreement between these two ratios is found, from which it is inferred that the value of the base line is accurate to better than 0.05%. An error of 0.1% in the base line will produce an error of approximately 2% in the apparent integrated intensity.

For an iron absorber fully polarized by an external magnetic field in a direction perpendicular to the transmitted beam of radiation, the six integrated intensities give six independent determinations of $f_a\sigma_0$ through the expression¹¹

$$A_i = f_s \Gamma_0 (\pi/2) Z_i e^{-Z_i} [I_0(Z_i) + I_1(Z_i)] (1-R), \quad (2)$$

where I_n is a hyperbolic Bessel function of order n ,

¹¹ G. A. Bykov and P. Z. Hein, Zh. Eksperim. i Teor. Fiz. **43**, 909 (1962) [Soviet Phys. JETP **16**, 646 (1963)].

$Z_i = q_i f_a \sigma_0 n_a t / 2$, n_a is the number density of Fe^{57} nuclei in the absorber, t is the thickness of the absorber in cm, $\Gamma_0 = 0.097$ mm/sec is the full width at half-maximum,¹ R is the background fraction, and

$$q_1 = q_6 = \frac{3}{8}, \quad q_2 = q_5 = \frac{1}{2}, \quad q_3 = q_4 = \frac{1}{8}. \quad (3)$$

In order to obtain the maximum resonant cross section σ_0 from the recoilless resonant cross section of iron $f_a\sigma_0$, it is necessary to obtain a value for the recoilless fraction of iron f_a . The recoilless fraction of iron is estimated from the iron phonon frequency distribution $g(\omega)$ of Minkiewicz, Shirane, and Nathans.¹² This phonon distribution is based on a Born-von-Kármán fifth-neighbor general-force-constant model in which the force constants are determined from a fit to the dispersion relation for the phonons as determined by coherent scattering of thermal neutrons. The Debye portion of the phonon distribution corresponds to a $\Theta_D = 463^\circ\text{K}$. Three frequency moments are

$$\begin{aligned} \omega(-2) &= \left[\int_0^\infty g(\omega) \omega^{-2} d\omega \right]^{-1/2} = 5.29 \times 10^{12} \text{ cps}, \\ \Theta_D(-2) &= 439^\circ\text{K}, \\ \omega(1) &= \int_0^\infty g(\omega) \omega d\omega = 6.45 \times 10^{12} \text{ cps}, \\ \Theta_D(1) &= 413^\circ\text{K}, \\ \omega(2) &= \left[\int_0^\infty g(\omega) \omega^2 d\omega \right]^{-1/2} = 6.73 \times 10^{12} \text{ cps}, \\ \Theta_D(2) &= 416^\circ\text{K}, \end{aligned} \quad (4)$$

where $\Theta_D(p)$ is the effective Debye temperature associated with the p th moment.¹³ Essentially the same values are obtained from the frequency distribution of Brockhouse *et al.*¹⁴

The last two moments give the limiting mean-square velocity of the iron nuclei¹⁵ and are in good agreement with the effective Debye temperature of $400 \pm 30^\circ\text{K}$ obtained from the second-order Doppler shift of iron.¹⁶

The mean-square displacement of the Fe^{57} nucleus at 297°K is given by¹⁷

$$\langle x^2 \rangle = \frac{kT}{ma[2\pi\omega(-2)]^2} \left[1 + \frac{a}{12} \left(\frac{h\omega(-2)}{kT} \right)^2 - \frac{a}{720} \left(\frac{h\omega(2)}{kT} \right)^2 \left(\frac{h\omega(-2)}{kT} \right)^2 + \dots \right], \quad (5)$$

¹² V. J. Minkiewicz, G. Shirane, and R. Nathans, Phys. Rev. **162**, 528 (1967).

¹³ R. M. Housley and F. Hess, Phys. Rev. **146**, 517 (1966).

¹⁴ B. N. Brockhouse, H. E. Abou-Helal, and E. D. Hallman, Solid State Commun. **5**, 211 (1967).

¹⁵ D. P. Johnson and A. J. Kassman, Phys. Rev. **188**, 1385 (1969).

¹⁶ R. S. Preston, S. S. Hanna, and J. Heberle, Phys. Rev. **128**, 2207 (1962).

¹⁷ This is a slight modification of the expression for the mean square displacement to include the mass dependence of the various moments as described in Ref. 13.

where $T=297^\circ\text{K}$, k is the Boltzmann constant, h is Planck's constant, m is the mass of an Fe^{57} nucleus, and $a=0.981$ is the ratio of the atomic weight of iron to the atomic weight of Fe^{57} . The third term in the expansion contributes approximately 0.1% to the mean-square displacement of iron at 297°K . Using the values from Eq. (4) for the frequency moments leads to

$$\langle x^2 \rangle = 0.00423 \text{ \AA}^2. \quad (6)$$

If the frequency distribution of Brockhouse *et al.*¹⁴ is used, the value for the mean-square displacement is within 2% of the value given in Eq. (6). Although the uncertainty in the mean-square displacement is difficult to evaluate, it is probably less than $\pm 2\%$.

The recoilless fraction is given by¹⁸

$$f_a = \exp(-k^2 \langle x^2 \rangle) = 0.799 \pm 0.004, \quad (7)$$

where k is the wave number for the 14.4-keV γ radiation, and the uncertainty reflects the 4% uncertainty in the mean-square displacement. This value for the recoilless fraction compares favorably with other estimates used in determining σ_0 , which range from 0.77 to 0.80.²⁻⁵

The background fraction R was determined in a manner similar to that described in Ref. 19. The measured values are 0.134 ± 0.001 and 0.127 ± 0.001 for the 14.5-mg/cm² and the 13.6-mg/cm² iron absorbers, respectively.

The average thickness of the two absorbers was determined by weighing the thin iron disks and dividing by the area. The two absorbers have an average thickness of 14.48 and 13.56 mg/cm². The natural relative abundance of Fe^{57} was used to determine the thickness of Fe^{57} in the unenriched absorbers.²⁰

The possible effects on the measured value of the resonant cross section resulting from nonuniformities in the thickness of the absorbers have not previously been discussed. In view of the fact that the thickness of the iron foils are of the order of 10 μ , variations of tenths of microns can produce significant errors in the measured value of the resonant cross section.

There are two different ways in which nonuniformity in the thickness can effect the accuracy of the measurement. A significant error can result if the region of the absorber through which the γ radiation is transmitted has a different thickness than the average thickness. One way to reduce the possibility of this occurrence is to arrange the geometry such that the beam passes through a major fraction of the foil. With the geometry used in this investigation, the beam had a diameter of 1.3 cm compared with a total iron disk diameter of 1.6 cm. However, even if the beam diameter is the same as the disk diameter, significant variations in the thickness

will produce an underestimation of the resonant cross section because the smaller electronic absorption associated with the thinner region give a larger weight to the thinner regions and more importantly because of the nonlinear relationship between the integrated intensity and the resonant cross section.²¹

For the above reasons, it is important to establish the thickness uniformity of the iron foils. To this end we have examined the count rate of 14.4-keV γ radiation through several regions of the iron foil, each region having an area of approximately 0.07 cm². The largest variation in thickness observed in this manner was approximately 0.6%. No significant error was introduced by the beam size being smaller than the iron absorbers. The absorbers were also examined under a microscope in order to estimate the microscopic variation in thickness. The root-mean-square variation in thickness was estimated to be approximately $\pm 0.5 \mu$ which will lead to an underestimation of the resonant cross section of less than 0.1%.

The primary source used in these measurements was a 5.0-mCi $\text{Cu}(\text{Co}^{57})$ source purchased from New England Nuclear. Its recoilless fraction f_s , as measured by the black absorber technique,²² is 0.690 ± 0.005 , which is 3% lower than the intrinsic value for f_s of Fe^{57} in Cu .²³ The uncertainty in f_s reflects the sum of statistical uncertainty (2 standard deviations) and possible systematic errors due to the uncertainty in the blackness of the black absorber and other small corrections.

The linewidths associated with the spectra have an average thickness-corrected value²⁴ of 0.203 ± 0.002 mm/sec, which is narrower than previously reported linewidths associated with spectra used in determining the resonant cross section, but is 4.5% broader than the value¹ of 0.194 mm/sec expected from an ideal spectrometer and a natural width source and absorber. The relationship between the integrated intensity and the recoilless resonant cross section is independent of the source line shape or velocity-independent instrumental broadening, but it is dependent on the absorber linewidth.¹⁹ For example, if the observed broadening is entirely attributable to a broadening in the absorber, than the resonant cross section from the inner lines would be approximately 2% lower than reported here and from the outer lines would be approximately 4% lower.

The most sensitive feature which could contribute to the broadening of an iron absorber is a slight variation in the magnetic field at the iron nuclei. This type of broadening would produce five times as large an effect on the outer lines as on the inner lines. The spectra

²¹ J. D. Bowman, E. Kankeleit, E. N. Kaufmann, and B. Persson, Nucl. Instr. Methods 50, 13 (1967).

²² R. M. Housley, N. E. Erickson, and J. G. Dash, Nucl. Instr. Methods 27, 29 (1964).

²³ See for example, R. H. Nussbaum, D. G. Howard, W. L. Nees, and C. F. Steen, Phys. Rev. 173, 653 (1968).

²⁴ H. Frauenfelder, D. E. Nagle, R. D. Taylor, D. R. Cochran, and W. M. Visscher, Phys. Rev. 126, 1065 (1962).

¹⁸ H. J. Lipkin, Ann. Phys. (N. Y.) 26, 115 (1964).

¹⁹ D. P. Johnson and J. G. Dash, Phys. Rev. 172, 983 (1968).

²⁰ The average thickness of Fe^{57} in each of the two absorbers is 0.323 mg/cm² and 0.297 mg/cm² assuming the relative abundance of Fe^{57} is 0.0219 [A. H. Wapstra, J. Inorg. Nucl. Chem. 3, 329 (1957)].

TABLE I. Resonant cross section σ_0 .

Absorption line	With 14.5-mg/cm ² absorber (10 ⁻¹⁸ cm ²)	With 13.6-mg/cm ² absorber (10 ⁻¹⁸ cm ²)
3, 4	2.58	2.57
2, 5	2.55	2.56
1, 6	2.53	2.59
Average	2.55	2.57

reported here have an equal broadening of all lines, indicating the broadening is velocity independent. If the broadening of the spectra was due to a broadening of the absorber lines, then under the assumption used in this work, one would find that the value of the resonant cross section determined from the outer lines would be 2% higher than the value determined from the inner lines. No such systematic variation is observed (see Table I).

From the above evidence it was concluded that the extra broadening is not due to the absorber but is due to either velocity-independent instrumental broadening or broadening of the source. Hence, no corrections to the relationship between the integrated intensity and the recoilless resonant cross section given in Eq. (2) are required.

There is additional evidence that suggests the primary source may be the major contributor to the broadening. The fact that the recoilless fraction of the source was 3% lower than the intrinsic value for Fe⁵⁷ in Cu could be due to either self-absorption in the source, or some of the Fe⁵⁷ being in nonequivalent sites. Both of these effects would produce a broadening of the emitted radiation. To further explore this possibility, a second 0.5-mCi Cu(Co⁵⁷) source was prepared in a manner similar to that described in Ref. 25. The thickness-

²⁵ R. M. Housley, J. G. Dash, and R. H. Nussbaum, Phys. Rev. 136, A464 (1964).

corrected line width of this secondary source versus the 14.5-mg/cm² absorber is 0.197 ± 0.006 mm/sec, which is consistent with the extra broadening of the principal spectra being primarily due to the source line shape.

III. RESULTS AND CONCLUSION

The results of this investigation are summarized in Table I. The experimental values for the maximum resonant cross section and the total internal conversion coefficient obtained from this investigation are

$$\begin{aligned}\sigma_0 &= (2.56 \pm 0.05) \times 10^{-18} \text{ cm}^2, \\ \alpha_T &= 8.19 \pm 0.18.\end{aligned}\quad (8)$$

The quoted experimental uncertainty is the sum of a systematic uncertainty of approximately $\pm 1.7\%$ resulting from the uncertainties in f_s , f_a , and the baseline, and an uncertainty of $\pm 0.4\%$, resulting from counting statistics, uncertainties in thickness and other small effects.

In conclusion, these results are in excellent agreement with those of Rubinson and Gopinathan⁸ and strongly support a value of σ_0 approximately 9% higher than previously accepted.

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