to give the *third-moment theorem with internal magnetic field,*

$$(2\pi\hbar)^3F_3=-\frac{3G_1}{5^{1/2}}(-1)^{2I_a}\Big[(f_bd_b{}^2-f_ad_b{}^2W_2-2d_ad_bf_bW_1)(2I_b+1)^{1/2}\begin{Bmatrix}I_b & 1 & I_b\\ 1 & I_b & 2\end{Bmatrix}$$
$$+(f_bd_a{}^2W_2+2d_ad_bf_aW_1-f_ad_a{}^2)(2I_a+1)^{1/2}\begin{Bmatrix}I_a & 1 & I_a\\ 1 & I_a & 2\end{Bmatrix}\Big]-\frac{G_2}{5^{1/2}}(-1)^{2I_a}$$
$$\times\Big[(f_b{}^3-3f_b{}^2f_aW_2)(2I_b+1)^{1/2}\begin{Bmatrix}I_b & 2 & I_b\\ 2 & I_b & 2\end{Bmatrix}+(3f_bf_a{}^2W_2-f_a{}^3)(2I_a+1)^{1/2}\begin{Bmatrix}I_a & 2 & I_a\\ 2 & I_a & 2\end{Bmatrix}\Big], \quad \text{(A3)}$$

where

$$G_\lambda=\sum a(1,\mu_1)a(1,\mu_2)a(\lambda,\mu_3)(-1)^{\mu_3}C(1,1,\lambda;\mu_1,\mu_2,-\mu_3), \quad \text{(A4)}$$

$$G_1=[6(1+\tfrac{1}{3}\eta^2)]^{-1/2}[3\cos^2\theta-1+\eta\sin^2\theta\cos2\phi], \quad \text{(A5)}$$

$$G_2=-(2/7)^{1/2}(1+\tfrac{1}{3}\eta^2)^{-3/2}(1-\eta^2). \quad \text{(A6)}$$

In the Fe^{57} case, the third-moment theorem gives

$$F_3=(e^2/128\pi^3\hbar)qH^2\gamma_a(3\gamma_a-\gamma_b)Q_a(3\cos^2\theta-1+\eta\sin^2\theta\cos2\phi). \quad \text{(A7)}$$

The *third-moment theorem with external magnetic field* is obtained by substituting zero for G_1 in Eq. (A3). In the Fe^{57} example, therefore, $F_3(\Theta)=0$.

There is no difficulty in principle in calculating the fourth and higher moments by repeated application of Eq. (A2) to the trace formulas, but the results become very complicated.

PHYSICAL REVIEW VOLUME 139, NUMBER 3A 2 AUGUST 1965

Mössbauer Cross Section of Fe^{57} in Iron*

S. S. HANNA† AND R. S. PRESTON
Argonne National Laboratory, Argonne, Illinois

(Received 29 September 1964; revised manuscript received 22 March 1965)

The Mössbauer cross section for Fe^{57} nuclei in metallic iron has been measured by determining the Mössbauer absorption as a function of absorber thickness. The method used is independent of the recoilless fraction and the line width of the source. The effects of instrumental resolution and background radiation in the detector are also minimized. The measured cross section is in agreement with the value calculated from measured nuclear quantities and the Debye temperature of iron.

THE resonant cross section for the absorption of gamma rays by a nucleus is given by

$$\sigma_0=2\pi\lambda\!\!\!^{-2}\omega(1/1+\alpha), \quad (1)$$

where $\lambda\!\!\!^{-}$ is the wavelength of the radiation, α is the internal-conversion coefficient, and ω is the statistical factor $(2I_1+1)/(2I_0+1)$. The spins I_1 and I_0 refer to the upper and lower states, respectively. The effective cross section for recoilless absorption is $f\sigma_0$, where f is the fraction of nuclei that interact without recoil.[1] The resonant cross section can be determined from measurements on the total strength of the absorption.[2]

In most cases both ω and $\lambda\!\!\!^{-}$ are known so that a measurement of $f\sigma_0$ determines either f or α, if one or the other of these quantities is known independently. For the two lowest states of Fe^{57} the statistical factor is equal to 2 and the wavelength $\lambda\!\!\!^{-}$ is known accurately from the energy $E_\gamma=14.37$ keV.[3] Measurements on the temperature shift in the Mössbauer spectrum of Fe^{57} have shown[4] that metallic iron is quite well represented by the Debye model with $\theta=400°$K. This Debye temperature gives $f=0.79$. Thus a measurement of the recoilless absorption of Fe^{57} can be used to obtain a value for the internal-conversion coefficient α.

The earliest determination[5] of α, obtained from measurements on the decay of Co^{57}, gave $\alpha=15\pm1$. On the other hand, early Mössbauer measurements[4] indi-

* Work performed under the auspices of the U. S. Atomic Energy Commission.
† Present address: Stanford University, Stanford, California.
[1] R. L. Mössbauer, Z. Physik **151**, 124 (1958).
[2] S. S. Hanna, J. Heberle, C. Littlejohn, G. J. Perlow, R. S. Preston, and D. H. Vincent, Phys. Rev. Letters **4**, 28 (1960).
[3] J. B. Bellicard and A. Moussa, J. Phys. Radium **18**, 115 (1957).
[4] R. S. Preston, S. S. Hanna, and J. Heberle, Phys. Rev. **128**, 2207 (1962).
[5] H. R. Lemmer, O. J. A. Segaert, and M. A. Grace, Proc. Phys. Soc. (London) **A68**, 701 (1955).

cated $\alpha<12$. More recent measurements[6,7] on the Co^{57} decay have given $\alpha=9.9\pm0.6$ and 9.5 ± 0.5, and a current Mössbauer determination[8] gives $\alpha=9.0\pm0.4$.

In this paper we report a measurement of the resonant absorption cross section of Fe^{57} in metallic iron. The method used is independent of the recoilless fraction f_s and line width of the source, and it is practically independent of the instrumental resolution of the equipment and background radiation in the detector.

The intensity distribution of recoilless radiation from a monochromatic Lorentzian source with recoilless fraction f_s and total intensity I_T is

$$I(E,E_s)dE=I_T\frac{(2/\pi)f_s\Gamma_s dE}{4(E-E_s)^2+\Gamma_s^2}, \tag{2}$$

where Γ_s is the full width at half-maximum and E_s the center of the Lorentzian distribution. If radiation from such a source passes through a nonresonant absorber and is detected in a counter whose total counting rate is normalized to unity, then the intensity distribution of the detected recoilless radiation is

$$R(E,E_s)dE=\frac{(2/\pi)f_s\Gamma_s dE}{4(E-E_s)^2+\Gamma_s^2}. \tag{3}$$

$R(E,E_s)$ satisfies the normalization condition that the total intensity of recoilless radiation transmitted by the absorber and detected is

$$T(E_s)=\int_0^\infty R(E,E_s)dE=f_s. \tag{4}$$

However, when resonant absorption also exists, then for an unpolarized absorber with N lines, the cross section for resonant absorption is

$$\sigma(E)=\sigma_0\sum_{i=1}^{N}\frac{q_i\Gamma_a^2}{4(E-E_i)^2+\Gamma_a^2}=\sigma_0\Sigma(E), \quad \sum_{i=1}^{N}q_i=1, \tag{5}$$

where σ_0 is the resonant single-line absorption cross section, E_i is the position and $q_i\sigma_0$ the partial cross section of the ith line. Γ_a is the linewidth of the absorber. The symbol $\Sigma(E)$ is used to denote the sum over the N lines. For Fe^{57}, there are six lines and the q_i are in the ratios[9] $\frac{1}{4}:\frac{1}{6}:\frac{1}{12}:\frac{1}{12}:\frac{1}{6}:\frac{1}{4}$. Therefore, for a given gamma-ray energy (i.e., velocity) of the source E_s, the intensity of recoilless gamma rays transmitted through the absorber and detected is

$$T(E_s)=\int_0^\infty R(E,E_s)\exp[-f_a n\sigma_0\Sigma(E)]dE, \tag{6}$$

where n is the thickness of the absorber in nuclei per cm^2 and f_a is the resonant fraction of absorbing nuclei. Then if $x=f_a n\sigma_0$, the total area in the absorption dips is

$$\begin{aligned}\Phi(x)&=\int_0^\infty[f_s-T(E_s)]dE_s\\&=f_s\int_0^\infty[1-e^{-x\Sigma(E)}]dE=f_sA(x).\end{aligned} \tag{7}$$

Thus, the area depends on f_s, $x=f_a n\sigma_0$, q_i, and E_i/Γ_a, but not on the linewidth Γ_s of the source.

In practice the radiation detector always records an extraneous background which has the effect of reducing the measured area in exactly the same way as does the presence of nonresonant gamma rays of energy E_γ. Thus, one can write

$$[\Phi(x)]_{\text{meas}}=f_b\Phi(x)=f_bf_sA(x), \tag{8}$$

where f_b is the ratio of the number of recorded gamma rays of energy E_γ to the total number of gamma rays recorded, as measured at a source velocity for which the recoilless absorption is negligible.

If the apparatus used to measure the absorption spectrum introduces an instrumental line broadening which is not velocity (energy)-dependent, this broadening enters into the calculation in the same way as does the line width Γ_s of the source. Thus, the area is independent of a constant instrumental broadening. If the instrumental resolution varies with velocity in a known manner, the resolution function which then appears in Eq. (7) can be taken into account in computing the areas. In the ratio method described below, the effect of unknown small variations in the resolution function will be greatly reduced.

For an absorber polarized by a transverse magnetic field[10] it is necessary to modify[11] the above equations, since lines with different polarizations will absorb independently. Recoilless radiation from the unpolarized source may be expressed as the sum of two equal components with mutually perpendicular linear polarizations:

$$I(E,E_s)=I_{\parallel}(E,E_s)+I_{\parallel}(E,E_s), \quad I_{\parallel}=I_{\perp}. \tag{9}$$

---

[6] H. C. Thomas, C. F. Griffin, W. E. Phillips, and E. C. Davis, Jr., Nucl. Phys. **44**, 268 (1963).

[7] A. H. Muir, Jr., E. Kankeleit, and F. Boehm, Phys. Letters **5**, 161 (1963).

[8] R. H. Nussbaum and R. M. Housley, Nucl. Phys. (to be published).

Note added in proof. O. C. Kistner and A. W. Sunyar [Phys. Rev. (to be published)] report values of $\alpha=9.0\pm0.5$ from a gamma-gamma coincidence measurement, and $\alpha=8.9\pm0.6$ from a Mössbauer measurement.

[9] S. S. Hanna, J. Heberle, C. Littlejohn, G. J. Perlow, R. S. Preston, and D. H. Vincent, Phys. Rev. Letters **4**, 177 (1960).

[10] G. J. Perlow, S. S. Hanna, M. Hamermesh, C. Littlejohn, D. H. Vincent, R. S. Preston, and J. Heberle, Phys. Rev. Letters **4**, 74 (1960).

[11] In our earlier work (Ref. 4) which gave $\alpha<12$, unpolarized absorbers were used and the analysis was based on Eq. (7). In the present work polarized absorbers were used to obtain fixed and known line intensities. In a preliminary report [Rev. Mod. Phys. **36**, 469 (1963)] of this work the analysis was not modified to take account of the polarization and a value of $\alpha<8$ was reported. We are indebted to R. H. Nussbaum, R. M. Housley, and E. Kankeleit for calling our attention to this error.

The absorption cross section is also split into two parts corresponding to the two polarizations:

$$\sigma_{||}(E)=\sigma_0\sum_{i=1}^{N}\frac{q_i^{||}\Gamma_a^2}{4(E-E_i)^2+\Gamma_a^2}=\sigma_0\Sigma_{||}(E),\quad\sum_{i=1}^{N}q_{i||}=1,$$
$$\sigma_{\perp}(E)=\sigma_0\sum_{i=1}^{N}\frac{q_i^{\perp}\Gamma_a^2}{4(E-E_i)^2+\Gamma_a^2}=\sigma_0\Sigma_{\perp}(E),\quad\sum_{i=1}^{N}q_i^{\perp}=1. \tag{10}$$

For Fe^{57} the $q_i^{||}$ are in the ratio[9] $\frac{3}{8}:0:\frac{1}{8}:\frac{1}{8}:0:\frac{3}{8}$ and the $q_i^{\perp}$ are in the ratio $0:\frac{1}{2}:0:0:\frac{1}{2}:0$. For an energy E_s of the source, the intensity of recoilless radiation transmitted by the absorber and detected is

$$T(E_s)=\int_0^{\infty}R_{||}(E,E_s)\exp[-f_a n\sigma_0\Sigma_{||}(E)]dE+\int_0^{\infty}R_{\perp}(E,E_s)\exp[-f_a n\sigma_0\Sigma_{\perp}(E)]dE. \tag{11}$$

The total area in the absorption dips is then

$$\Phi(x)=f_s\int_0^{\infty}\{1-\tfrac{1}{2}\exp[-x\Sigma_{||}(E)]-\tfrac{1}{2}\exp[-x\Sigma_{\perp}(E)]\}dE=f_sA(x). \tag{12}$$

In this case, as for an unpolarized absorber,

$$[\Phi(x)]_{\text{meas}}=f_bf_sA(x). \tag{13}$$

Both f_s and f_b can be effectively eliminated in the determination of $f_a\sigma_0$ by using the ratio method.[12,13] This method consists in measuring the ratio $A(x_2)/A(x_1)$ for a thick absorber (thickness x_2) and a thin absorber (thickness x_1). Because of the nonlinear nature of the absorption, the ratios $A(x_2)/A(x_1)$ and x_2/x_1 uniquely determine x_1. The use of a very thick and a thin absorber increases the sensitivity of the method. The ratio $x_2/x_1=n_2/n_1$ can be determined very accurately from the weights, areas, and compositions of the absorber foils. The value of f_s is independent of the absorber. If both the atomic absorption and the setting of the pulse-height window at the output of the detector are kept constant during the measurements on both foils, then f_b will also be constant. From Eq. (8) or Eq. (13) it will then follow that

$$A(x_2)/A(x_1)=[\Phi(x_2)]_{\text{meas}}/[\Phi(x_1)]_{\text{meas}}.$$

The atomic absorption can be kept constant by using absorbing foils of equal total iron content or by interposing suitable foils with nearly the same Z.

In principle $A(x)$ is determined by evaluating the integral in Eq. (7) or Eq. (12), whichever is appropriate. For the specified thickness ratio $r=x_2/x_1$, the area ratio $A(rx_1)/A(x_1)$ is computed as a function of x_1. The measured area ratio then determines $x_1=n_1f_a\sigma_0$ from which $f_a\sigma_0$ is obtained.

Because of the practical difficulty of determining areas from the measured curves, the procedure is modified as follows: Theoretical Mössbauer spectra $T(E_s)$ are computed from Eq. (6) or Eq. (11) for representative values of x over the complete range of x. Next the calculated points for each theoretical spectrum are fitted with six Lorentzian lines by the method of least squares. The areas obtained from these Lorentzian fits (Fig. 1) are then used to obtain the ratios $A(x_2)/A(x_1)$. In like manner, the experimentally measured spectra are also fitted with Lorentzian lines to obtain the measured area ratios which are then compared with the computed ones to determine $x_1=n_1f_a\sigma_0$. All the computations were performed with a CDC-3600 computer.

The positions of the lines E_i/Γ_a used in calculating the theoretical spectra were taken from earlier measurements.[4] The natural line width was used for Γ_a. All the absorbing foils were prepared by a standard procedure of purification at 900°C in wet hydrogen gas and subsequent annealing in vacuum. It was demonstrated that this procedure produced practically the natural line width in thin foils.

A series of measurements was made on a sequence of foils having thicknesses of 1.56×10^{18}, 7.71×10^{18}, 29.0×10^{18}, and 58.2×10^{18} Fe^{57} nuclei per cm^2. A velocity drive of the loudspeaker type was employed. The Mössbauer measurements were performed in as identical a fashion as possible. Measurements of the pulse-height spectrum and the window setting of the detector were made before and after each run, and were used to calculate corrections for variations in f_b. These were found to be small. The foils were magnetized in a transverse magnetic field in order to obtain the same known distribution of fractional cross sections in each

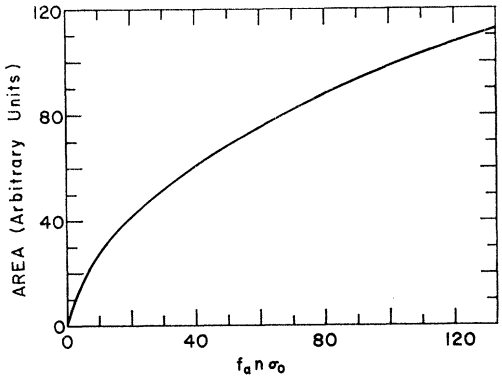

FIG. 1. Area of a polarized six-line absorption spectrum of Fe^{57} in iron at room temperature. This is the Lorentzian area computed in the manner outlined in the text, and is plotted as a function of absorber thickness expressed as $f_an\sigma_0$, where f_a is the recoilless fraction, n the number of Fe^{57} nuclei per cm^2, and σ_0 the resonant cross section.

[12] W. W. Havens, Jr., and L. J. Rainwater, Phys. Rev. **83**, 1123 (1951); E. Melkonian, W. W. Havens, Jr., and L. J. Rainwater, *ibid.* **92**, 702 (1953).

[13] S. S. Hanna and L. Meyer-Schützmeister, Phys. Rev. **115**, 986 (1959).

foil. The curves obtained from the least-squares fit of Lorentzian lines to the data are shown in Fig. 2. Since the absorbing foils were polarized, the analysis given in Eqs. (9)–(13) was used. The curve of $A(x)$ versus x for this case is shown in Fig. 1. From this curve appropriate curves of $A(x_2)/A(x_1)$ versus x_1 were obtained (Fig. 3). The measured area ratios, obtained from the least-square curves of Fig. 2, were then used to obtain values of $x_1=f_a n_1\sigma_0$ as illustrated in Fig. 3. The results are tabulated in Table I.

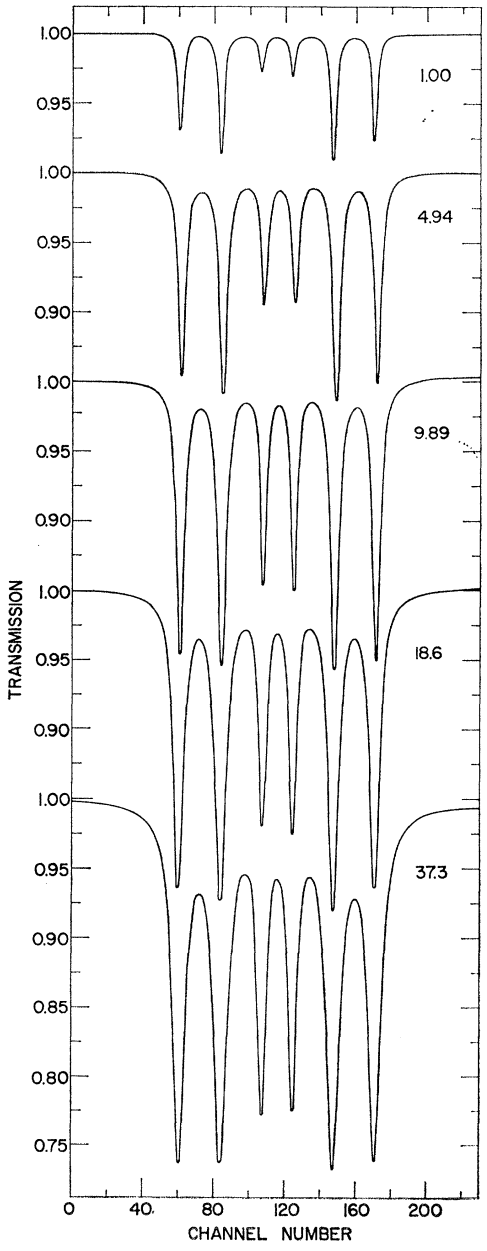

FIG. 2. Transmission curves for the five foils measured in the experiment. The curves are least-squares fits to the experimental points (not shown). The numbers on the right give the foil thickness (Fe^{57} nuclei/cm²) relative to the thinnest sample.

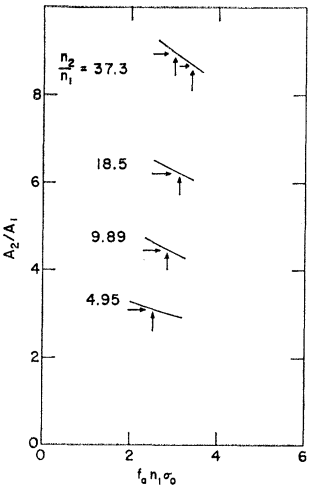

FIG. 3. Determination of $f_a n_1\sigma_0$ for the five measurements in Table I. The curves of A_2/A_1 versus $f_a n_1\sigma_0$ have been constructed from the curve of Fig. 1.

These values of $f_a\sigma_0$ are in reasonable agreement with each other and give an average $f_a\sigma_0=(1.91\pm0.14)\times10^{-18}$ cm². With $f_a=0.79$, one obtains $\sigma_0=(3.20\pm0.25)\times10^{-18}$ cm². With $E_\gamma=14.37$ keV and $\omega=2$ in Eq. (1), one finds $(1+\alpha)\sigma_0=23.89\times10^{-18}$ cm². Combining this with the measured value of σ_0 gives $\alpha=8.9\pm0.7$. This result agrees with the recent measurements listed above.

TABLE I. Calculation of Mössbauer cross sections from area measurements on five foils. The subscript "1" refers to the thinnest foil relative to which ratios for the other foils are determined.

n_2/n_1	A_2/A_1	$f_a n_1\sigma_0$	$f_a\sigma_0\times10^{18}$ (cm²)
37.3	8.67	3.40	2.18
18.5	6.20	3.12	2.00
9.89	4.45	2.87	1.84
4.95	3.09	2.53	1.62
37.3	8.94	3.01	1.93
		Average:	1.91±0.14

The value obtained for the resonant Mössbauer absorption cross section at room temperature, $f_a\sigma_0=(1.91\pm0.14)\times10^{-18}$ cm², is in agreement with the earlier measurement[4] of $(2.3\pm0.4)\times10^{-18}$ cm². Because the earlier measurements were obtained with unpolarized absorbers, the above value at room temperature was subject to some uncertainty because of the known difficulty in achieving complete depolarization in the foils. The value $f_a\sigma_0=(0.60\pm0.06)\times10^{-18}$ cm² at $T=1050$°K (above the Curie point) was not subject to this uncertainty. If we use a mean value of $\alpha=9.3$ computed from the recent measurements, we obtain $\sigma_0=2.32\times10^{-18}$ cm² and $f_a=0.26\pm0.03$ at $T=1050$°K. This value of f_a corresponds to a Debye temperature of (310 ± 30)°K at $T=1050$°K. This confirms the trend observed earlier[4] that the Debye temperature of iron decreases with increasing temperature.

We would like to thank W. S. Denno and D. J. Bailey for valuable assistance in accumulating and analyzing the data.