Resonant Absorption in the Presence of Faraday Rotation*

M. Blume and O. C. Kistner
Physics Department, Brookhaven National Laboratory, Upton, New York
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A general treatment of electro- and magneto-optical phenomena in the presence of resonant absorption is given. The resulting expressions in terms of the real and imaginary parts of a matrix index of refraction are easily utilized in calculations. We consider in detail the effects of these phenomena in Mössbauer experiments with polarized γ rays, presenting expressions for absorption as a function of polarization as well as for the polarization of the transmitted beam, which are valid for arbitrarily thick absorbers. Comparisons with experimental data are presented.

I. INTRODUCTION

THE polarization dependence of the Mössbauer absorption of nuclear γ radiation has been thoroughly studied experimentally and theoretically.[1,2] These treatments consider the cross section for the absorption of polarized radiation by a single nucleus, and they are adequate for most purposes. In considering the absorption of polarized radiation by a relatively thick array of nuclei, however, it is necessary to take into account the reemission of the absorbed radiation and the coherence of this reemitted radiation with the incident wave. In the absence of absorption these multiple scattering effects give rise to such phenomena as birefringence and Faraday rotation.[3]

In this paper we consider the absorption of polarized γ rays by nuclei, taking into account the reemitted radiation. Such a treatment is necessary for the proper interpretation of the delicate experiments recently performed in an attempt to determine time-reversal noninvariance in nuclear γ decays[4] as well as for Mössbauer polarimetry measurements. The reason that the selection rules for absorption of polarized radiation by a single nucleus may not adequately describe the situation in an absorber of finite thickness may be seen by considering a beam of polarized γ rays incident on a resonant absorber. A fraction of the radiation will be absorbed by the first layer of nuclei in the sample, and this fraction will be determined by the selection rules derived in Refs. 1 and 2. Part of this absorbed radiation will be reemitted in the forward direction, and will combine coherently with the incident beam. Since the reemitted radiation will in general have a polarization different from that of the incident beam, the radiation seen by the second layer of nuclei in the sample will be different from that seen by the first layer, and hence a different fraction of the radiation will be absorbed by the second layer. To calculate the total absorption of the sample, then, it is necessary to iterate not only the absorption of the successive layers of nuclei, but also the "rotation" of the polarization induced by the coherent scattering in each successive layer. In effect, we must iterate the *amplitude* of the wave rather than its intensity as it progresses through the absorber, i.e., we must calculate the polarization-dependent index of refraction. This index of refraction will be, in general, a 2×2 matrix, corresponding to the two possible independent states of polarization of the radiation, and will be complex. The real part will be related to the change in polarization of the wave, and the imaginary part to the absorption.

In the following sections we will derive an expression for the matrix index of refraction for an absorber of nuclei in a magnetic field, allowing for a mixture of multipoles in the nuclear transition and for an arbitrary angle between the propagation vector of the γ rays and the direction of the magnetic field. We consider the effects of time-reversal noninvariance in the nuclear transition on the index of refraction. Expressions are given for the transmission as a function of γ-ray energy and initial polarization, and for the polarization of the transmitted beam. Comparison with experimental results[4] will be made.

The principal results derived here are Eqs. (5″) and (6″), which provide a compact and convenient treatment for phenomena such as Faraday rotation and absorption, not only of γ rays, but of electromagnetic radiation in general. It is necessary to use these equations for a correct treatment of Mössbauer absorption if the absorber lines are split by magnetic fields or electric field gradients. The equations are used with expressions for the index of refraction due to resonant nuclear absorption to derive results for the polarization dependence of the absorption of Mössbauer γ rays.

We use the term "Faraday rotation" in a general sense to indicate changes in the polarization of radiation as it propagates through a medium. In the examples considered here the direction of the polarization changes as it propagates, and a circular component appears as well. In our treatment these effects are automatically taken into account.

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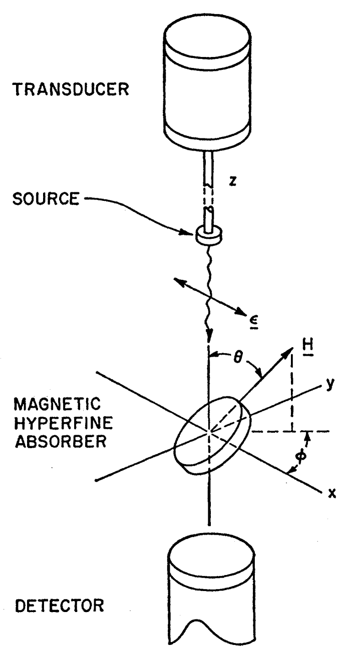

FIG. 1. Schematic of general Mössbauer experiment for observing birefringence and Faraday rotation effects.

II. OUTLINE OF THE THEORY

A schematic illustration of the experimental situation for which the theory is applicable is shown in Fig. 1. Monoenergetic, arbitrarily polarized γ rays are incident on a sample that contains N absorbing nuclei per unit volume. The absorbing nuclei are in a magnetic field $\mathbf{H}$ that makes an angle θ with the propagation vector of the incident radiation. The projection of $\mathbf{H}$ on the azimuthal plane makes an angle ϕ with respect to the component of linear polarization present in the incident radiation. We want to calculate the transmission of the γ radiation as a function of its energy and initial polarization and also the polarization of the transmitted radiation. The γ ray that propagates through the absorbing medium finds itself in a region with an index of refraction n. (We will first consider the development of the theory assuming that the index of refraction is known. In the next section we will turn to its calculation from the properties of the absorbing nuclei.)

The equation for the propagation of a plane wave along the z axis is, then,

$$(\nabla^2+n^2k^2)\psi(z)=0, \tag{1}$$

with the solution

$$\psi(z)=e^{inkz}\psi(0). \tag{2}$$

In an isotropic medium, n is simply a scalar quantity, and we may take $\psi(0)=1$. In the presence of a magnetic field, however, the index of refraction depends on the polarization of the radiation, so that n must be considered as a 2×2 matrix with elements $n_{pp'}$, p, $p'=\pm1$. In this case n_{++} and n_{--} are the indices of refraction for left and right circularly polarized radiation, respectively. The off-diagonal terms represent the possibility of converting left to right circular polarization and vice versa. Diagonalization of n tells us the types of polarization which are transmitted without change. The index of refraction is in general, however, complex (i.e., non-Hermitian): The real parts describe the "rotation" of the polarization and the change in phase velocity, while the imaginary parts describe the polarization dependence of the absorption. This means that n is not necessarily diagonalizable. Any polarization will be altered on passing through a medium with a non-diagonalizable index of refraction.

It is fortunately not necessary to diagonalize n to solve the wave equation (1). The solution, which we denote by $|\psi(z)\rangle$, will be a column vector with two components,

$$|\psi(z)\rangle=\begin{pmatrix}\psi_+(z)\\ \psi_-(z)\end{pmatrix},$$

where $\psi_+(z)$ is the amplitude for left circular polarization and $\psi_-(z)$ is the amplitude for right circular polarization. The solution (2) is still formally correct provided we interpret e^{inkz} as a matrix that acts on $|\psi(0)\rangle$. The vector $|\psi(0)\rangle$ describes the polarization of the radiation incident on the absorber:

$$|\psi(0)\rangle=\begin{pmatrix}1\\0\end{pmatrix}$$

represents left circular polarization;

$$|\psi(0)\rangle=\begin{pmatrix}0\\1\end{pmatrix},$$

right circular polarization;

$$|\psi(0)\rangle=\frac{1}{\sqrt{2}}\begin{pmatrix}1\\1\end{pmatrix},$$

linear polarization along the x axis; etc. The intensity of radiation at z is given by

$$I(z)=\langle\psi(z)\mid\psi(z)\rangle=\langle\psi(0)\mid\exp(-in^\dagger kz)\times\exp(inkz)\mid\psi(0)\rangle, \tag{3}$$

where $\langle a\mid b\rangle$ represents the vector product of the two-dimensional column vector $|b\rangle$ with the row vector $\langle a|$. Note that if n is real (i.e., no absorption), $n^\dagger=n$ and $I(z)=\langle\psi(0)\mid\psi(0)\rangle=I(0)$. Also, if n and $n^\dagger$ commute, we may write

$$\exp(-in^\dagger kz)\exp(inkz)=\exp[i(n-n^\dagger)kz]=\exp(-2\,\mathrm{Im}nkz)$$

and

$$I(z)=\langle\psi(0)\mid\exp(-2\,\mathrm{Im}nkz)\mid\psi(0)\rangle.$$

In this case, the real part of the index of refraction, and hence the rotation of the polarization, plays no role in the determination of the intensity of the transmitted radiation. The most interesting situation that we will consider occurs when the real and imaginary parts of the index of refraction do not commute with one another, so that Eq. (3) must be used. This non-commutativity of the two parts of the index of refraction implies that rotation of polarization followed by absorption yields an intensity different from that obtained by absorption followed by rotation.

The expression (3) for the transmitted intensity is correct for arbitrary but complete initial polarization. To generalize this to the case of partially polarized radiation it is convenient to introduce the Poincaré-Stokes representation for the polarization, and the density matrix for the incident beam. To do this we rewrite (3) as

$$I(z)=\sum_{m=\pm 1}\langle\psi(0)\mid\exp(-in^{\dagger}kz)\mid m\rangle \times\langle m\mid\exp(inkz)\mid\psi(0)\rangle$$

by inserting a complete set of polarization states. Rearranging this gives

$$I(z)=\sum_{m}\langle m\mid\exp(inkz)\mid\psi(0)\rangle \times\langle\psi(0)\mid\exp(-in^{\dagger}kz)\mid m\rangle.$$

Averaging over the initial polarization $|\psi(0)\rangle$ then yields an expression for the transmitted intensity of a partially polarized beam. We have

$$I(z)=\sum_{m}\langle m\mid\exp(inkz)\rho\exp(-in^{\dagger}kz)\mid m\rangle,$$

where

$$\rho=\overline{|\psi(0)\rangle\langle\psi(0)|}$$

is the density matrix for the incident beam. Since it is the averaged outer product of a two-dimensional vector, ρ is a 2×2 matrix, and hence may be written in terms of the unit matrix and the three Pauli matrices:

$$\rho=\tfrac{1}{2}(1+\mathbf{P}\cdot\boldsymbol{\sigma}). \qquad (4)$$

Here the σ's are the 2×2 Pauli matrices, and the three parameters P_ξ, P_η, P_ζ give the Poincaré representation of the polarization. In the basis that we use here, $P_\zeta=\pm1$ represent left and right circular polarization, $P_\xi=\pm1$ represent linear polarization along the x and y axes, and $P_\eta=\pm1$ represent linear polarization at $+$ and $-45°$ to the x axis. If $|\mathbf{P}|=0$, the beam is unpolarized, and $|\mathbf{P}|=1$ represents a completely polarized beam. An elliptically polarized beam has P_ξ, P_η, and $P_\zeta\neq0$. It should be noted that $\mathbf{P}$ is a vector in an abstract space. This has been emphasized by denoting the axes by ξ, η, ζ, and the corresponding Pauli matrices by σ_ξ, σ_η, σ_ζ. Coordinates in real space are given by x, y, z. For a detailed discussion of the Poincaré representation, see the papers of Fano.[5]

[5] U. Fano, J. Opt. Soc. Am. **39**, 859 (1949); Phys. Rev. **93**, 121 (1954).

In terms of the density matrix, then, Eq. (3) can be written

$$I(z)=\mathrm{Tr}[\exp(inkz)\rho\exp(-in^{\dagger}kz)], \qquad (5)$$

where the trace is over the two-dimensional polarization variables. We may also write down the expression for the polarization vector in Poincaré space, $\mathbf{P}'$, of the transmitted beam:

$$\mathbf{P}'I(z)=\mathrm{Tr}[\boldsymbol{\sigma}\exp(inkz)\rho\exp(-in^{\dagger}kz)]. \qquad (6)$$

Equations (5) and (6) contain expressions for phenomena such as Faraday rotation, birefringence, the Cotton-Mouton effect, etc. All of these are represented by rotations of the vector $\mathbf{P}$ in the abstract Poincaré space of the polarization. We may further simplify these expressions by using again the fact that any 2×2 matrix can be written as a linear combination of the unit matrix and the Pauli matrices. In particular,

$$nkz=a+\mathbf{b}\cdot\boldsymbol{\sigma}=a+b_\xi\sigma_\xi+b_\eta\sigma_\eta+b_\zeta\sigma_\zeta. \qquad (7)$$

From the standard forms for the Pauli matrices,

$$\sigma_\xi=\begin{pmatrix}0&1\\1&0\end{pmatrix},\quad \sigma_\eta=\begin{pmatrix}0&-i\\i&0\end{pmatrix},\quad \sigma_\zeta=\begin{pmatrix}1&0\\0&-1\end{pmatrix},$$

we find

$$\begin{aligned}a&=\tfrac{1}{2}kz(n_{++}+n_{--}),\\ b_\xi&=\tfrac{1}{2}kz(n_{+-}+n_{-+}),\\ b_\eta&=\tfrac{1}{2}ikz(n_{+-}-n_{-+}),\\ b_\zeta&=\tfrac{1}{2}kz(n_{++}-n_{--}).\end{aligned} \qquad (8)$$

Substituting (7) in (5) and (6),

$$I(z)=\exp[i(a-a^*)] \times\mathrm{Tr}\{[\exp(i\mathbf{b}\cdot\boldsymbol{\sigma})]\rho[\exp(-i\mathbf{b}^*\cdot\boldsymbol{\sigma})]\}, \qquad (5')$$

$$\mathbf{P}'I(z)=\exp[i(a-a^*)] \times\mathrm{Tr}\{\boldsymbol{\sigma}[\exp(i\mathbf{b}\cdot\boldsymbol{\sigma})]\rho[\exp(-i\mathbf{b}^*\cdot\boldsymbol{\sigma})]\}. \qquad (6')$$

We then make use of the identity

$$\exp(i\mathbf{b}\cdot\boldsymbol{\sigma})=\cos b+i(\hat{b}\cdot\boldsymbol{\sigma})\sin b,$$

where $b=(b_\xi^2+b_\eta^2+b_\zeta^2)^{1/2}$, $\hat{b}=\mathbf{b}/b$, and the expressions

$$\begin{aligned}&\mathrm{Tr}1=2,\\ &\mathrm{Tr}\sigma_\mu=0,\\ &\mathrm{Tr}\sigma_\mu\sigma_\nu=2\delta_{\mu\nu},\\ &\mathrm{Tr}\sigma_\mu\sigma_\nu\sigma_\beta=2i\epsilon_{\mu\nu\beta},\\ &\mathrm{Tr}\sigma_\mu\sigma_\nu\sigma_\beta\sigma_\gamma=2(\delta_{\mu\nu}\delta_{\beta\gamma}+\delta_{\mu\gamma}\delta_{\nu\beta}-\delta_{\mu\beta}\delta_{\nu\gamma})\end{aligned}$$

(where μ, ν, β, γ run over ξ, η, ζ, and $\epsilon_{\mu\nu\beta}$ is the unit antisymmetric tensor of third rank). Substituting these and (4) in (5′) and (6′) gives expressions for the intensity and polarization of the transmitted beam in terms of the initial polarization $\mathbf{P}$ and the parameters a and $\mathbf{b}$, which are related through (8) to the components of the index of refraction. The results of the algebra are

$$I(z)=\exp[i(a-a^*)]\{\cos b^*\cos b+(\hat{b}^*\cdot\hat{b})\sin b^*\sin b -i(\hat{b}^*\cdot\mathbf{P})\sin b^*\cos b+i(\hat{b}\cdot\mathbf{P})\sin b\cos b^* +i\mathbf{P}\cdot(\hat{b}^*\times\hat{b})\sin b^*\sin b\} \tag{5''}$$

and

$$\mathbf{P}'I(z)=\exp[i(a-a^*)]\{i\hat{b}\sin b\cos b^*-i\hat{b}^*\sin b^*\cos b -i(\hat{b}^*\times\hat{b})\sin b^*\sin b+\mathbf{P}\cos b^*\cos b +(\mathbf{P}\times\hat{b})\sin b\cos b^*+(\mathbf{P}\times\hat{b}^*)\sin b^*\cos b +[\hat{b}(\mathbf{P}\cdot\hat{b}^*)+\hat{b}^*(\mathbf{P}\cdot\hat{b})-\mathbf{P}(\hat{b}\cdot\hat{b}^*)]\sin b^*\sin b\}. \tag{6''}$$

These relations are very easy to use once the index of refraction has been calculated, and in the following section we consider this problem.

III. CALCULATION OF THE INDEX OF REFRACTION

We will assume that the scattering and absorption of the γ rays is done entirely by the nuclei of the absorber, so that electronic effects are neglected. In this case, the index of refraction is given by the well-known expression[6]

$$n=1+(2\pi/k^2)N\bar{f}, \tag{9}$$

where $\bar{f}$ is the coherent forward scattering amplitude for γ rays from a single nucleus. This amplitude will, in general, be a 2×2 matrix, since a nucleus may absorb a γ ray of one polarization and emit one of another polarization coherently in the forward direction. Also, the forward scattering amplitude for right circular polarization will differ from that for left circular polarization. The problem then reduces to the calculation of $\bar{f}$.

Let the spins and magnetic quantum number for the ground and excited state of the nucleus be given by I_0, m_0, and I_1, m_1, respectively. The resonant scattering amplitude for the scattering of a photon $\mathbf{k}_0$ with polarization p to $\mathbf{k}p'$ is then given by[7]

$$f=\left(-\frac{k_0V}{2\pi\hbar c}\right)R \times\sum_{m_1}\frac{\langle\mathbf{k}p'I_0m_0'\,|\,\mathcal{H}'\,|\,I_1m_1\rangle\langle I_1m_1\,|\,\mathcal{H}'\,|\,\mathbf{k}_0pI_0m_0\rangle}{E_{k_0}+E_{m_0}-E_{m_1}+\frac{1}{2}i\Gamma}. \tag{10}$$

Here $\mathcal{H}'$ is the interaction between the nucleus and the electromagnetic field, R is the recoil-free fraction, and Γ is the total width of the excited state. The electromagnetic field is assumed normalized in a volume V, which accounts for the presence of this factor. In writing Eq. (10) it has been assumed that the eigenstates of the system can be labeled by the magnetic quantum numbers m_1 and m_0 of the excited and ground states. If this is not the case (as, for example, in the presence of a nonaxial electric field gradient or when magnetic field and electric field gradient lie along different axes), then we simply introduce transformation coefficients $\langle I_0m_0\,|\,I_0\alpha_0\rangle$ between the correct eigenstates $|\,I_0\alpha_0\rangle$ and the $|\,I_0m_0\rangle$. These complicate the appearance of the expressions, but the procedure is straightforward in principle. Also, in including the recoil-free fraction, we have assumed that the incident energy of the photon is close to resonance, i.e., that we have a "slow" collision. This point is discussed in detail by Trammell.[7] Otherwise, Eq. (10) is general and allows for a change of the sublevel of the ground state from m_0 to m_0' in the course of the scattering. The coherent forward amplitude, which we need for (9), is obtained from (10) by taking the wave vector $\mathbf{k}$ of the scattered photon equal to that of the incident photon $\mathbf{k}_0$. In addition, the final state m_0' must be taken equal to the initial m_0, and an average over m_0 is performed. This gives

$$\bar{f}=\left(-\frac{k_0VR}{2\pi\hbar c}\right)\frac{1}{2I_0+1} \times\sum_{m_0m_1}\frac{\langle\mathbf{k}_0p'I_0m_0\,|\,\mathcal{H}'\,|\,I_1m_1\rangle\langle I_1m_1\,|\,\mathcal{H}'\,|\,\mathbf{k}_0pI_0m_0\rangle}{E_{k_0}+E_{m_0}-E_{m_1}+\frac{1}{2}i\Gamma}. \tag{11}$$

The two possible values of p and $p'(=\pm1)$ denote left and right circular polarization. This expression for $\bar{f}$ is then a 2×2 matrix. The presence of magnetic fields or electric field gradients at the nucleus are accounted for by the dependence of E_{m_0} and E_{m_1} in the denominator on the magnetic quantum number.

The interaction $\mathcal{H}'$ between the nucleus and the electromagnetic field is given by

$$\mathcal{H}'=-(1/c)\sum_i\mathbf{j}(\mathbf{r}_i)\cdot\mathbf{A}(\mathbf{r}_i), \tag{12}$$

where $\mathbf{j}(\mathbf{r}_i)$ is the current density of the ith nucleon and $\mathbf{A}(\mathbf{r}_i)$ is the vector potential of the electromagnetic field:

$$\mathbf{A}(\mathbf{r}_i)=\sum_{\mathbf{k}p}(2\pi\hbar c/Vk)^{1/2} \times\{a(\mathbf{k}p)\hat{u}(\mathbf{k}p)\exp(i\mathbf{k}\cdot\mathbf{r}_i)+\text{c.c.}\}. \tag{13}$$

Here $a(\mathbf{k}p)$ is a phonon annihilation operator and $\hat{u}(\mathbf{k}p)$ is a unit polarization vector. In order to calculate the matrix elements necessary for the evaluation of (11) we make use of the multipole expansion of Rose.[8] In all of the following expressions we will use the definitions and phase conventions of Rose for rotations, Clebsch-Gordan coefficients, etc. Equation (13) can

[6] M. Lax, Rev. Mod. Phys. **23**, 287 (1951).
[7] G. T. Trammell, Phys. Rev. **126**, 1045 (1962).

[8] M. E. Rose, *Elementary Theory of Angular Momentum* (John Wiley & Sons, Inc., New York, 1957).

be expanded to give [Ref. 8, Eq. (7.38)]

$$\mathbf{A}(\mathbf{r}_i)=\sum_{\mathbf{k}p}(2\pi\hbar c/Vk)^{1/2}\times\{a(\mathbf{k}p)(2\pi)^{1/2}\sum_{LM}i^L(2L+1)^{1/2}D_{Mp}^{(L)}(\phi\theta 0)\times(\mathbf{A}_{LM}(m)+ip\mathbf{A}_{LM}(e))+\text{c.c.}\}. \quad (14)$$

The angles θ and ϕ in the rotation $D_{Mp}^{(L)}$ are, respectively, the polar and azimuthal angles of the direction $\mathbf{k}$ of propagation of the γ ray. The vectors $\mathbf{A}_{LM}(m)$ and $\mathbf{A}_{LM}(e)$ represent the components of the magnetic and electric 2^L-pole moment operators. Expressions for them in terms of vector spherical harmonics are given on pp. 133 ff. of Ref. 8. The only property of these quantities that we shall use is that (Ref. 8, p. 138) $\mathbf{v}\cdot\mathbf{A}_{LM}$ is an irreducible tensor operator of rank L, where $\mathbf{v}$ is any vector. This fact, together with the expansion (14), enables us to calculate the matrix elements of the interaction $\mathcal{H}'$, which are necessary for the evaluation of (11). We need

$$\begin{aligned}\langle I_1m_1|(1/c)\sum_i\mathbf{j}(\mathbf{r}_i)\cdot\mathbf{A}(\mathbf{r}_i)|I_0m_0\mathbf{k}p\rangle &=2\pi\sum_{LM}(\hbar c/Vk)^{1/2}i^L(2L+1)^{1/2}D_{Mp}^{(L)}(\phi\theta 0)\\ &\times\langle I_1m_1|(1/c)\sum_i\mathbf{j}(\mathbf{r}_i)\cdot\mathbf{A}_{LM}(m)+ip(1/c)\\ &\times\sum_i\mathbf{j}(\mathbf{r}_i)\cdot\mathbf{A}_{LM}(e)|I_0m_0\rangle. \quad (15)\end{aligned}$$

Since $\mathbf{j}\cdot\mathbf{A}_{LM}$ is a tensor of rank L, however, we can write

$$(1/c)\langle I_1m_1|\sum_i\mathbf{j}(\mathbf{r}_i)\cdot\mathbf{A}_{LM}|I_0m_0\rangle=\langle I_1||(1/c)\sum_i\mathbf{j}(\mathbf{r}_i)\cdot\mathbf{A}_L||I_0\rangle C(I_0LI_1;m_0Mm_1), \quad (16)$$

where C is the Clebsch-Gordan coefficient, and the reduced matrix element is independent of the magnetic quantum number. This expression holds separately for $\mathbf{A}_{LM}(m)$ and $\mathbf{A}_{LM}(e)$. For convenience we introduce the notation

$$M_L=\langle I_1||(1/c)\sum_i\mathbf{j}(\mathbf{r}_i)\cdot\mathbf{A}_L(m)||I_0\rangle,$$

$$E_L=\langle I_1||(1/c)\sum_i\mathbf{j}(\mathbf{r}_i)\cdot\mathbf{A}_L(e)||I_0\rangle.$$

Substituting in (15) gives

$$\langle I_1m_1|\mathcal{H}'|I_0m_0\mathbf{k}p\rangle=2\pi(\hbar c/Vk)^{1/2}\sum_{LM}i^L(2L+1)^{1/2}D_{Mp}^{(L)}(\phi\theta 0)\times\{M_L+ipE_L\}C(I_0LI_1;m_0Mm_1). \quad (17)$$

The coefficients M_L and E_L represent the strengths of the magnetic and electric 2^L poles, respectively, involved in the absorption of the photon. Conservation of parity requires that either all odd-L M_L and even-L E_L or all even-L M_L and odd-L E_L vanish. In the majority of cases it is sufficient to consider a single non-negligible M_L, together with a non-negligible E_{L+1}, and we will treat this situation. (For $L=1$ we have a mixed magnetic dipole–electric quadrupole transition.) Equation (17) becomes

$$\begin{aligned}\langle I_1m_1|\mathcal{H}'|I_0m_0\mathbf{k}p\rangle &=2\pi(\hbar c/Vk)^{1/2}\sum_M i^L\{(2L+1)^{1/2}M_LD_{Mp}^{(L)}(\phi\theta 0)\\ &\times C(I_0LI_1;m_0Mm_1)-p(2L+3)^{1/2}E_{L+1}D_{Mp}^{(L+1)}(\phi\theta 0)\\ &\times C(I_0L+1I_1;m_0Mm_1)\}. \quad (18)\end{aligned}$$

Since

$$\langle I_0m_0\mathbf{k}p'|\mathcal{H}'|I_1m_1\rangle=\langle I_1m_1|\mathcal{H}'|I_0m_0\mathbf{k}p'\rangle^*,$$

and also

$$D_{Mp}^{(L)}(\phi\theta 0)=e^{-iM\phi}d_{Mp}^{(L)}(\theta),$$

where $d_{Mp}^{(L)}(\theta)$ is real, we have for (11)

$$\begin{aligned}\bar{f}_{p'p}=&-[2\pi R/(2I_0+1)]\\ &\times\sum_{m_0m_1M}[(2L+1)^{1/2}M_L^*d_{Mp'}^{(L)}(\theta)C(I_0LI_1;m_0Mm_1)\\ &-p'(2L+3)^{1/2}E_{L+1}^*d_{Mp'}^{(L+1)}(\theta)\\ &\times C(I_0L+1I_1;m_0Mm_1)][(2L+1)^{1/2}M_Ld_{Mp}^{(L)}(\theta)\\ &\times C(I_0LI_1;m_0Mm_1)-p(2L+3)^{1/2}E_{L+1}d_{Mp}^{(L+1)}(\theta)\\ &\times C(I_0L+1I_1;m_0Mm_1)](E_{k_0}+E_{m_0}-E_{m_1}+\tfrac{1}{2}i\Gamma)^{-1}. \quad (19)\end{aligned}$$

This expression is independent of the azimuthal angle ϕ, as expected for a cylindrically symmetric system.

The two quantities M_L and E_{L+1} can be expressed in terms of their ratio and in terms of the partial width for γ emission of the excited nuclear state. The latter quantity can be determined experimentally, and the relation is derived in the Appendix. From Eq. (A2) we have

$$\Gamma_\gamma=8\pi k_0(|M_L|^2+|E_{L+1}|^2).$$

Setting

$$\mu_L=M_L/(|M_L|^2+|E_{L+1}|^2)^{1/2},$$

$$\epsilon_{L+1}=E_{L+1}/(|M_L|^2+|E_{L+1}|^2)^{1/2},$$

so that

$$|\mu_L|^2+|\epsilon_{L+1}|^2=1,$$

we obtain for (19)

$$\begin{aligned}\bar{f}_{p'p}=&-(R/2I_0+1)(\Gamma_\gamma/4k_0)\sum_{m_0m_1M}[(2L+1)^{1/2}\mu_L^*\\ &\times d_{Mp'}^{(L)}(\theta)C(I_0LI_1;m_0Mm_1)-p'(2L+3)^{1/2}\epsilon_{L+1}^*\\ &\times d_{Mp'}^{(L+1)}(\theta)C(I_0L+1I_1;m_0Mm_1)][(2L+1)^{1/2}\mu_L\\ &\times d_{Mp}^{(L)}(\theta)C(I_0LI_1;m_0Mm_1)-p(2L+3)^{1/2}\epsilon_{L+1}\\ &\times d_{Mp}^{(L+1)}(\theta)C(I_0L+1I_1;m_0Mm_1)]\\ &\times(E_{k_0}+E_{m_0}-E_{m_1}+\tfrac{1}{2}i\Gamma)^{-1}.\end{aligned}$$

Finally we define

$$\epsilon_{L+1}/\mu_L = \delta e^{i\alpha}, \tag{20}$$

where δ is real, so that

$$|\mu_L|^2 = 1/(1+\delta^2).$$

Also, if the nucleus is in a magnetic field H along the z axis, $E_{m_0} = g_0\mu_N m_0 H$, and $E_{m_1} = g_1\mu_N m_1 H + E_1$, where g_0 and g_1 are the g factors for the ground and excited states, E_1 is the energy of the excited state relative to the ground state in the absence of a magnetic field, and μ_N is the nuclear magneton, we find for $\bar{f}_{p'p}$:

$$\begin{aligned}\bar{f}_{p'p} = &-(2I_0+1)^{-1}(\Gamma_\gamma/4k_0)[R/(1+\delta^2)]\\ &\times \sum_{m_0 m_1 M} [(2L+1)^{1/2} d_{Mp'}{}^{(L)}(\theta) C(I_0 L I_1; m_0 M m_1)\\ &-p'(2L+3)^{1/2}\delta e^{-i\alpha} d_{Mp'}{}^{(L+1)}(\theta)\\ &\times C(I_0 L+1 I_1; m_0 M m_1)][(2L+1)^{1/2} d_{Mp}{}^{(L)}(\theta)\\ &\times C(I_0 L I_1; m_0 M m_1) - p(2L+3)^{1/2}\delta e^{i\alpha} d_{Mp}{}^{(L+1)}(\theta)\\ &\times C(I_0 L+1 I_1; m_0 M m_1)]\\ &\times (E_{k_0} - E_1 + (g_0 m_0 - g_1 m_1)\mu_N H + \tfrac{1}{2} i\Gamma)^{-1}.\end{aligned} \tag{21}$$

This is the expression that we will use for the calculation of the index of refraction.

At this point we may recall that the assumption of time-reversal invariance requires that the phase factor α in Eq. (20) should be equal either to 0 or to π,[9] i.e., that ϵ_{L+1} and μ_L be relatively real. Examination of (21) shows that this implies that $\bar{f}_{p'p} = \bar{f}_{pp'}$, and hence that, for the case under consideration, the index of refraction is a symmetric matrix. From Eq. (8), the symmetry of n implies that $b_\eta = 0$. An experiment which aims to detect time-reversal noninvariance should thus be designed to detect a nonvanishing b_η. In general, the antisymmetric part of $\bar{f}$ is proportional to $\sin\alpha$, so that b_η is linear in α for small α. As we will see in the following section, the proper design of an experiment to detect a nonzero b_η requires some care, since, in a thick absorber, other terms in the index of refraction may combine to imitate the effect of an antisymmetric term in n.

It is possible to distinguish between Faraday and time-reversal contributions to b_η because the Faraday contribution is unchanged on going from M to $-M$, while the time-reversal contribution changes sign. Assuming the absence of quadrupole interactions, the two effects may be separated by making measurements at a symmetric pair of $M = \pm 1$ lines and taking the sum and difference of the observed effects.

IV. COMPARISON WITH EXPERIMENT

We will now apply the theory of the previous sections to the results of a Mössbauer experiment[4] performed to test time-reversal invariance in the 90-keV $M1$-$E2$ mixed transition in ruthenium-99. In this experiment a measurement is made of the resonant absorption of polarized 90-keV γ rays by the $m_0 = \frac{5}{2} \to m_1 = \frac{3}{2}$ and $m_0 = -\frac{5}{2} \to m_1 = -\frac{3}{2}$ magnetic hyperfine components in a magnetized absorber of ruthenium-99 in metallic iron. The experimental arrangement, which is reproduced in Fig. 2, is that of a conventional Mössbauer transmission experiment with the exception that *two*, individually magnetized ruthenium-iron absorbers are used. The single line source is Doppler shifted to coincide in energy with either of the selected pair of symmetric $\Delta m = \pm 1$ hyperfine absorption resonances. Absorber No. 1 which is magnetized at 90° to the propagation vector serves to polarize linearly (by selective resonant absorption) the radiation passing through it. The transmitted radiation is in fact polarized transverse to the direction $\mathbf{H}'$. The field in the second absorber is oriented so as to maximize the value of the b_η (i.e., $\sin\alpha$) term in the absorption cross section for linearly polarized radiation. This occurs at $\theta = 54\frac{3}{4}°$ and $\phi = 45°$. The test for a nonvanishing $\sin\alpha$ is made by looking for a change in the intensities of the two resonances with a reversal of the magnetic field in the second absorber. Field reversal measurements were made for various reflections of the direction of $\mathbf{H}$ into adjacent quadrants. The effect observed is obtained from

$$E = \frac{I(\theta) - I(\pi-\theta)}{I(\theta) + I(\pi-\theta)}, \tag{22}$$

where $I(\theta)$ is the intensity of transmitted radiation linearly polarized at 45° to the x axis, i.e.,

$$I(\theta) = \mathrm{Tr}\{\exp[in(\theta)kz]\tfrac{1}{2}(1+\sigma_\eta)\exp[-in^\dagger(\theta)kz]\}.$$

From the relation

$$d_{Mp}{}^{(j)}(\pi-\theta) = (-)^{j-p} d_{-Mp}{}^{(j)}(\theta) = (-)^{j-M-2p} d_{M-p}{}^{(j)}(\theta),$$

it follows that $n_{p'p}(\pi-\theta) = n_{-p'-p}(\theta)$, or, in matrix notation $n(\pi-\theta) = \sigma_\xi n(\theta)\sigma_\xi$. Hence

$$\begin{aligned} I(\pi-\theta) &= \mathrm{Tr}\{\exp[in(\pi-\theta)kz]\tfrac{1}{2}(1+\sigma_\eta)\\ &\quad\times\exp[in^\dagger(\pi-\theta)kz]\}\\ &= \mathrm{Tr}\{\sigma_\xi \exp[in(\theta)kz]\sigma_\xi \tfrac{1}{2}(1+\sigma_\eta)\sigma_\xi\\ &\quad\times \exp[-in^\dagger(\theta)kz]\sigma_\xi\}\\ &= \mathrm{Tr}\{\exp[in(\theta)kz]\tfrac{1}{2}(1-\sigma_\eta)\\ &\quad\times\exp[-in^\dagger(\theta)kz]\},\end{aligned}$$

and the effect E is given by

$$E = \frac{\mathrm{Tr}\{\exp[in(\theta)kz]\sigma_\eta \exp[-in^\dagger(\theta)kz]\}}{\mathrm{Tr}\{\exp[in(\theta)kz]\exp[-in(\theta)kz]\}}.$$

From Eq. (5″) we find

$$E = \frac{i\hat{b}_\eta \sin b \cos b^* - i\hat{b}_\eta{}^* \sin b^* \cos b + i(\hat{b}^* \times \hat{b})_\eta \sin b^* \sin b}{\cos b^* \cos b + (\hat{b}^* \cdot \hat{b}) \sin b^* \sin b}, \tag{23}$$

[9] S. P. Lloyd, Phys. Rev. **81**, 161 (1951).

so that, even if $\hat{b}_\eta = 0$, the effect does not vanish, since the term $(\hat{b}^* \times \hat{b})_\eta \neq 0$. The latter term corresponds to the combined effect of Faraday rotation and absorption.[10] To simplify the physical interpretation of (23) we consider a thin absorber. From (8) we see that $b \propto kz$, so we may expand the terms in (23), keeping only those quadratic or linear in b. We find

$$
\begin{aligned}
E &\simeq i(b_\eta - b_\eta^*) + i(\mathbf{b}^* \times \mathbf{b})_\eta \\
&= -kz(n_{+-}{}^r - n_{-+}{}^r) - \tfrac{1}{2}(kz)^2\{(n_{++}{}^r - n_{--}{}^r) \\
&\quad \times (n_{+-}{}^i + n_{-+}{}^i) - (n_{++}{}^i - n_{--}{}^i)(n_{+-}{}^r + n_{-+}{}^r)\},
\end{aligned} \tag{24}
$$

where n^r and n^i are the real and imaginary parts of the index of refraction: $n = n^r + in^i$. In addition to the opposite symmetry with respect to $\pm M$, there are two other basic characteristics that differentiate between a time-reversal effect and a Faraday effect. First, the time reversal noninvariance part is linear in the thickness z, since it is a single-nucleus effect. The Faraday rotation term, on the other hand, depends quadratically on the thickness because it is a product of the rotation and absorption, each of which is linear in z. Second, the variation with respect to the polar angle θ subtended by H in absorber No. 2 is markedly different for the two effects.

The effect for linearly polarized γ rays of energy E_0 may be calculated using Eqs. (8), (9), (21), and (23).

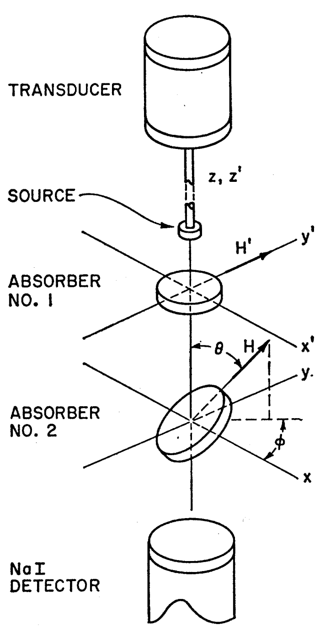

FIG. 2. Schematic of experimental arrangement used in Ref. 4 for testing time-reversal invariance with a single line source and two stationary Ru-Fe hyperfine absorbers. The absorbers are magnetized along the directions indicated by $\mathbf{H}$ and $\mathbf{H}'$.

[10] We again note, as mentioned in the Introduction, that we use the term "Faraday rotation" in a general sense. The polarization emerging from the sample is, in the case considered here, elliptical rather than linear.

TABLE I. Parameters used in comparing Eq. (28) with experiment.

N	$=3.96\times10^{+21}$ cm^{-3}
k_0	$=4.55\times10^{9}$ cm^{-1}
z_0	$=0.241$ cm
Γ_γ/Γ	$=0.385$
δ	$=-1.64$
R	$=0.08$
g_1	$=-0.189$
g_0	$=-0.249$
H	$=500$ kOe

The thickness z through which the radiation travels is given by $z = z_0/\sin\theta$, where z_0 is the perpendicular thickness of the sample. In order to compare our calculations with experiment it is necessary to make a number of corrections. The absorption and rotation must be integrated over the line shape of the incident resonant γ rays. Broadening in the resonance lines of the absorber, the efficiency of the polarizing filter, the recoil-free fraction of the source, and the background counts under the 90-keV photopeaks in the detector window must be considered. These corrections can be accounted for empirically, to a good approximation by normalization to the variation of the resonant absorption with azimuthal angle ϕ in Fig. 2. This azimuthal variation, which was measured experimentally,[4] can be calculated from Eq. (5″). For linearly polarized radiation making an angle ϕ with the projection of the magnetic field (Fig. 1), we have in Poincaré space

$$
\begin{aligned}
P_\xi &= P\cos 2\phi, \\
P_\eta &= P\sin 2\phi, \\
P_\zeta &= 0,
\end{aligned} \tag{25}
$$

where $0 < P < 1$ is the degree of polarization. This representation follows from the discussion of the Poincaré parameters following Eq. (4). Substituting in Eq. (5″), we find

$$
\begin{aligned}
I(z) = \exp[i(a-a^*)][&|\cos b|^2 + \hat{b}^* \cdot \hat{b}\,|\sin b|^2 \\
&+ P\sin 2\phi\{i(\hat{b}^* \times \hat{b})_\eta\,|\sin b|^2\} \\
&- P\cos 2\phi\{2\,\mathrm{Im}\hat{b}_\xi \sin b \cos b^*\} \\
&- P\sin 2\phi\{2\,\mathrm{Im}\hat{b}_\eta \sin b \cos b^*\}].
\end{aligned} \tag{26}
$$

Equation (26) gives the absorption as a function of the angle ϕ of the linear polarization. The coefficient of the first $\sin 2\phi$ term vanishes in the absence of Faraday rotation, while the coefficient of the second vanishes for time-reversal invariance, i.e., for $b_\eta = 0$. In the absence of these two effects, then, the $\cos 2\phi$ term is the only polarization-dependent term present and $I(z)$ is a maximum or minimum for $\phi = 0°$ or $\phi = 90°$, i.e., for polarization parallel to or perpendicular to the projection of the magnetic field. It should be noted that the $\cos 2\phi$ dependence on ϕ is exact even for a thick absorber.

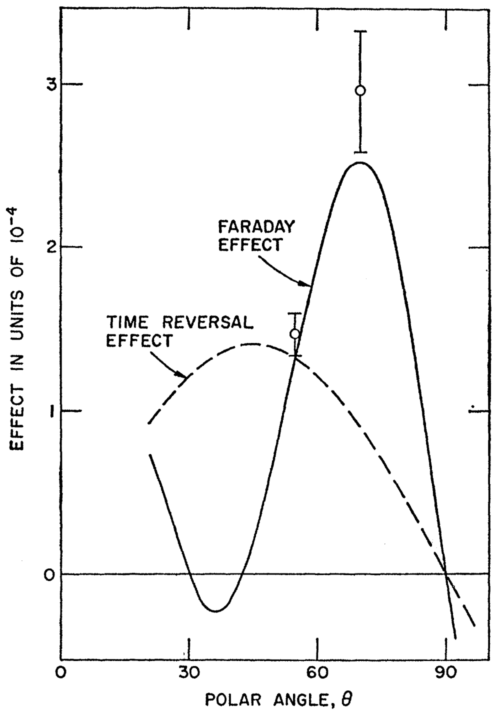

FIG. 3. Comparison of experimental data with the computed Faraday rotation effect as a function of the angle θ subtended by the magnetic field in absorber No. 2 with $\phi=45°$. The computed θ dependence for a time-reversal violation effect is shown for comparison. The experimental points are taken directly from run 2 of Table I in Ref. 4. A small correction has been applied to the point at 70° to account for the change in background at 70° relative to that at 55°.

Faraday rotation or time-reversal noninvariance cause a simple shift in the azimuthal curve. In this case, the angles at which the extrema occur are given by $dI/d\phi=0$ with

$$\begin{aligned} dI/d\phi=P\exp[i(a-a^*)][2\cos 2\phi\{i(\hat{b}^*\times\hat{b})\ |\sin b|^2 \\ -2\,\mathrm{Im}\hat{b}_\eta \sin b\cos b^*\}+2\sin 2\phi \\ \times\{2\,\mathrm{Im}\hat{b}_\xi \sin b\cos b^*\}], \end{aligned} \tag{27}$$

so that

$$\tan 2\phi=-\frac{i(\hat{b}^*\times\hat{b})_\eta\,|\sin b|^2-2\,Im\{\hat{b}_\eta\sin b\cos b^*\}}{2\,\mathrm{Im}\{b_\xi\sin b\cos b^*\}}. \tag{28}$$

This quantity gives us a significant comparison with experiment, since it is independent of the efficiency of the polarizer, the recoil-free fraction of the source, and the background in the counter window. It is also relatively insensitive to moderate line broadening in the absorber. Using the parameters given in Table I, we find, for γ rays in resonance with the $m_0=-\frac{5}{2}\rightarrow m_1=-\frac{3}{2}$ transition at θ of 55°, a shift in the azimuthal rotation curve of $\Delta\phi=0.65°$. This is to be compared with the experimental value[4] $\Delta\phi_{\mathrm{expt}}=(0.71\pm0.05)°$. This shift in the azimuthal absorption extrema, $\Delta\phi$, is in effect an average Faraday rotation experienced by the γ rays absorbed by the second absorber.

The normalizing factor may be determined as follows. The experimental measurement of the rotation curve gives

$$N_{\mathrm{expt}}=2P\frac{I'(\phi=0)-I'(\phi=90)}{I'(\phi=0)+I'(\phi=90)},$$

where $I'=I_0I+B$, I_0 is the intensity of incident resonant radiation, I is given by (26), and B is the nonresonant background.

Hence,

$$N_{\mathrm{expt}}=2P\frac{I(0)-I(90)}{I(0)+I(90)+2B/I_0}$$

$$\approx (I(0)-I(90))I_0P/B,$$

where we assume the background to be large. The experimental effect corresponding to (22) is

$$\begin{aligned} E'&=P(I_+'-I_-')/(I_+'+I_-') \\ &=P(I_+-I_-)/[I_++I_-+(2B/I_0)] \\ &\approx(I_0P/2B)(I_+-I_-). \end{aligned}$$

Calculation gives $I(0)-I(90)=0.528$. From the average experimental value of $N_{\mathrm{expt}}=1.16\times10^{-2}$ (run 2 of Ref. 4), we find $I_0P/2B=0.011$. Combining this with the calculated value $I_+-I_-=0.012$, we have, at $\theta=55°$, $E'=1.32\times10^{-4}$, compared with the average experimental value $E'=1.47\times10^{-4}$. This point has been used to scale the results for the angular dependence of the effect shown in Fig. 3. The agreement with experiment is excellent.

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APPENDIX: PARTIAL WIDTH FOR γ EMISSION

To derive the relation between M_L, E_{L+1}, and the partial width Γ_γ, we use the fact that Γ_γ is given by the total probability of a transition out of the excited state:

$$\Gamma_\gamma=2\pi\sum_{m_0p}\int d\hat{k}_0\,|\langle I_0m_0\mathbf{k}_0p\,|\,\mathcal{H}'\,|\,I_1m_1\rangle|^2\rho(E_F), \tag{A1}$$

where $\rho(E_F)$ is the density of final states of one polarization, given by $\rho(E_F)=[V/(2\pi)^3][k_0^2/\hbar c]$. Substituting

Eq. (18) in (A1) gives

$$\Gamma_\gamma=(2\pi)^3\sum_{m_0p}\int d\hat{k}_0(\hbar c/Vk_0)\{(-i)^L M_L^*(2L+1)^{1/2}$$
$$\times\sum_M D_{Mp}^{(L)*}(\phi\theta 0)C(I_0LI_1;m_0Mm_1)+(-i)^{L+2}$$
$$\times pE_{L+1}^*(2L+3)^{1/2}\sum_M D_{Mp}^{(L+1)*}(\phi\theta 0)C(I_0L+1I_1;$$
$$m_0Mm_1)\}\{i^L M_L(2L+1)^{1/2}\sum_M D_{Mp}^{(L)}(\phi\theta 0)$$
$$\times C(I_0LI_1;m_0Mm_1)+i^{L+2}pE_{L+1}(2L+3)^{1/2}$$
$$\times\sum_M D_{Mp}^{(L+1)}(\phi\theta 0)C(I_0L+1I_1;m_0Mm_1)\}\rho(E_F).$$

Using the orthogonality relation for the D coefficients, given in (Ref. 8, p. 74),

$$\int d\hat{k}_0 D_{M'p}^{(L')*}(\phi\theta 0)D_{Mp}^{(L)}(\phi\theta 0)=[4\pi/(2L+1)]\delta_{MM'}\delta_{LL'},$$

we find

$$\Gamma_\gamma=8\pi k_0\{|M_L|^2\sum_{m_0M}C^2(I_0LI_1;m_0Mm_1)+|E_{L+1}|^2\sum_{m_0M}C^2(I_0L+1I_1;m_0Mm_1)\},$$
$$=8\pi k_0\{|M_L^2+|E_{L+1}|^2\},\tag{A2}$$

the required relation.

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Time Dependence of Mössbauer Scattered Radiation*

P. Thieberger, J. A. Moragues,† and A. W. Sunyar
Brookhaven National Laboratory, Upton, New York

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Time distributions of resonantly scattered 14.4-keV Fe^{57} γ rays were measured by using as time-zero signals the preceding 122-keV γ rays. A Co^{57} source coplated with Fe was used with two different metallic scatterers (2.54×10^{-2}-mm-thick 92.8% Fe^{57} and 4.53×10^{-3}-mm-thick 2.19% Fe^{57}) in a cylindrical geometry. Measurements were performed at different relative source velocities. From the measured time spectra, contributions due to random coincidences and to Rayleigh scattering were subtracted. A theory based on a classical model in which the scattering nuclei are represented by randomly situated harmonic oscillators was developed. The experimental results are compared with computed theoretical curves. Good agreement was found in most of the cases. Discrepancies observed in two instances are discussed.

I. INTRODUCTION

The purpose of the present work is to measure the time spectrum of resonantly scattered γ rays and to develop a theory for this process based on a classical model. The first experimental results were reported previously.[1] It has been shown by Lynch *et al.*[2] that experimental results on the time distribution of Fe^{57} γ rays transmitted through a resonant absorber are well described by a theory in which each Fourier component of the incident radiation is changed in amplitude and phase according to a complex index of refraction. This complex index of refraction is given by the frequency response of the resonant oscillators in the absorber and by their number per unit volume.

It can be written as[3]

$$n=[1+r(\omega_0'^2-\omega^2+i\omega\lambda)^{-1}]^{1/2}$$
$$\simeq 1+\tfrac{1}{2}[r/(\omega_0'^2-\omega^2+i\omega\lambda)].\tag{1}$$

Here, ω_0' and λ are the frequency and the decay constant of the oscillators in the absorber and r is a constant which contains the oscillator density.

It may perhaps seem surprising that it should be possible to describe the absorber by a macroscopic quantity in this case, where the wavelength of the 14.4-keV radiation of Fe^{57} is about three times smaller than the minimum distance between neighboring oscillators in a highly enriched Fe^{57} scatterer and about ten times smaller than the average distance in the case of a natural-iron scatterer. This can, however, be explained by the fact[4] that the average contribution modifying the incident wave at a point inside the absorber is due to the mutually in-phase, coherently forward scattered waves from the oscillators.

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† Fellow of The Consejo Nacional de Investigaciones Científicas y Técnicas, Argentina. Permanent address: Comisión Nacional de Energía Atómica, Buenos Aires, Argentina.

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