Coherence and Polarization Effects in Mössbauer Absorption by Single Crystals

R. M. Housley, R. W. Grant, and U. Gonser

Science Center, North American Rockwell Corporation, Thousand Oaks, California 91360

(Received 1 August 1968)

The polarization dependence of the absorption cross section must generally be taken into account in calculating Mössbauer absorption spectra of single crystals which exhibit hyperfine splittings. A method for doing this in an experimentally interesting class of cases is described. In these cases, the incident radiation beam can be divided into two components, each having its own complex index of refraction. A number of conditions under which this division is always possible are described in terms of crystal symmetry considerations. Also, a practical method of computing the polarization of the cross section in terms of the hyperfine interactions for arbitrary nuclear transitions is described. A representation of the polarization in terms of a 2×2 density matrix is introduced which is convenient in cases where two or more nuclear transitions overlap in energy and in cases where resonant nuclei are located at several different or crystallographically equivalent but differently oriented sites in the crystal. A method for applying these results to an analysis of data to obtain electric-field-gradient parameters, mean square displacements, and magnetic structures is outlined.

I. INTRODUCTION

MÖSSBAUER absorption intensity measurements on single-crystal samples can be used to obtain several types of information. Most obviously, they can be used to obtain the parameters which describe the mean square displacement (MSD) of the resonant nuclei as a function of direction.[1] They may also be used to obtain the parameters which describe the electric-field-gradient (EFG) tensor at the sites of the resonant nuclei.[1] Also, particularly in conjunction with polarized sources, they may be used to obtain information on the magnetic structure of magnetically ordered materials.[2]

If a Mössbauer absorption resonance is split by electric-quadrupole or magnetic-dipole interactions, then the absorption cross sections in a single-crystal sample will, except along certain special directions, be polarization-dependent. This clearly will affect the resonant absorption intensities if the source radiation is polarized. It also affects the absorption intensities even for unpolarized source radiation if the absorbing crystal has finite thickness so that saturation effects must be taken into account.

For a polarized absorber, intensities saturate with thickness in a manner quite different than they do for an unpolarized absorber.[3] This may be easily seen by considering the case of an isolated completely polarized absorption line and an unpolarized source.[4] No matter how thick the absorber is made, no more than half the incident radiation can be resonantly absorbed. In addition, the dispersion associated with resonant absorption must also be considered in an absorber of

finite thickness.[5] It produces birefringence which can, for example, cause the plane of polarization of an incident plane polarized beam to rotate and thus cause it to experience a different absorption coefficient with depth in the absorber.[6] These effects alter the relative as well as the absolute intensities of absorption lines in a single crystal of finite thickness.

Our principal objective in this paper is to provide a procedure for analyzing experimental data in order to obtain hyperfine interaction parameters and MSD's. To this end, we first find the measurement conditions under which the analysis will be simplest. We then derive explicit formulas useful in carrying through this analysis, particularly in the case where the absorption lines are well resolved. We also derive a density matrix describing the polarization of the cross section for radiation incident in any direction for an Fe^{57} nucleus in an EFG with arbitrary asymmetry parameter. The use of most of these results is illustrated in the following paper[7] in which the MSD is found and the EFG parameters refined for the Fe positions in sodium nitroprusside.

In Sec. II of this paper, we describe a method for representing the polarization of both the dispersion and the absorption cross section in terms of 2×2 density matrices.

In Sec. III, several cases are explicitly described in which these are both diagonal in the same basis system. In these and only these cases, the incoming γ-ray beam may be decomposed into two components having orthogonal polarizations, which independently propagate through the crystal[8] with their appropriate complex indices of refraction.[9] The real parts of these indices of

---

[1] P. Zory, thesis, Carnegie Institute of Technology, Pittsburgh, Pa., 1964 (unpublished).

[2] U. Gonser, R. W. Grant, H. Wiedersich, and S. Geller, Appl. Phys. Letters **9**, 18 (1966).

[3] We have recently discussed a case where this was very significant. R. M. Housley, U. Gonser, and R. W. Grant, Phys. Rev. Letters **20**, 1279 (1968).

[4] R. H. Nussbaum and R. M. Housley, Nucl. Phys. **68**, 145 (1965).

[5] P. Imbert, Phys. Letters **8**, 95 (1964).

[6] An analysis of the general case is given by M. Blume and O. C. Kistner, Phys. Rev. **171**, 417 (1968).

[7] R. W. Grant, R. M. Housley, and U. Gonser, following paper, Phys. Rev. **178**, 523 (1969).

[8] The necessity of this condition was first pointed out to us by Dr. M. Blume.

[9] R. M. Housley and U. Gonser, Phys. Rev. **171**, 480 (1968).

refraction will, in general, be different and will lead to birefringence effects (Faraday effect, double refraction, etc.).[6,9] The imaginary parts of the indices of refraction determine the absorption line intensities, which is our main interest in this paper. Also, in Sec. III, we discuss possible effects of coherent scattering out of the forward direction and define conditions in which they can be ignored.

In Sec. IV, we present some convenient formulas for the absorption area in well-resolved lines as a function of the fractional polarization of the lines and the thickness of the sample.

In Sec. V, we express the polarization of γ radiation emitted during transitions among general nuclear states in a convenient form. We specialize the general results to the case of a magnetic-dipole transition between $I = \frac{3}{2}$ and $I = \frac{1}{2}$ nuclear levels as is appropriate for Fe^{57}. For a pure quadrupole interaction, we give explicit formulas for the polarizations of the absorption cross section in any direction with respect to the principal axes of the EFG tensor.

II. DESCRIPTION OF POLARIZATION IN TERMS OF DENSITY MATRICES

The polarization of radiation emitted in a definite multipole transition is a function of direction with respect to the quantization axis, but is completely determined in any specified direction. From a classical viewpoint, this polarization can be described in terms of the direction of the electric-field vector. This in turn can be specified in terms of a pair of orthogonal basis vectors both of which are orthogonal to the propagation direction as in Eq. (25). If the generally complex coefficients of the basis vectors are called a_θ and a_φ, then it can be seen that the set of numbers

$$\rho_{11} = a_\theta{}^* a_\theta, \quad \rho_{12} = a_\theta{}^* a_\varphi, \quad \rho_{21} = a_\theta a_\varphi{}^*, \quad \rho_{22} = a_\varphi{}^* a_\varphi, \quad (1)$$

where the * indicates complex conjugation, completely specify the polarization of the wave. From their definition, one can see that

$$\rho_{11}\rho_{22} = \rho_{12}\rho_{21}, \quad (2)$$

and if we, without loss of generality, take a_θ real

$$a_\theta = \sqrt{\rho_{11}} \quad \text{and} \quad a_\varphi = \rho_{12}/\sqrt{\rho_{11}}. \quad (3)$$

Therefore, the description of the polarization by Eq. (25) and by the density matrix

$$\begin{pmatrix} \rho_{11} & \rho_{12} \\ \rho_{21} & \rho_{22} \end{pmatrix} \quad (4)$$

are completely equivalent. Falkoff and MacDonald[10] have pointed out that a quantum-mechanical treatment of the radiation leads to an equivalent density matrix

to describe the polarization and have shown the correspondence between this density matrix and the four parameters originally introduced by Stokes[11] to describe partially polarized light.

In case two or more of the multipole transitions have the same energy, as will occur, for example, for a pure quadrupole interaction, then the radiation is an incoherent superposition of all the contributions with the same energy. Fano[12] has given arguments to show that the most convenient description of the polarization of the resulting beam is obtained by adding the density matrices for the individual transitions. At energies where two or more resonances overlap, because of their inherent linewidths, the polarization will also be described by a properly weighted sum of the individual density matrices.

For the sum density matrix, Eq. (2) must be replaced by

$$\rho_{11}\rho_{12} \geq \rho_{12}\rho_{21}. \quad (5)$$

The sum density matrix may be broken up into an unpolarized part, which is a multiple of the identity matrix, and a completely polarized part

$$\begin{pmatrix} \rho_{11} & \rho_{12} \\ \rho_{21} & \rho_{22} \end{pmatrix} = \begin{pmatrix} \rho_{00} & 0 \\ 0 & \rho_{00} \end{pmatrix} + \begin{pmatrix} \rho_{11} - \rho_{00} & \rho_{12} \\ \rho_{21} & \rho_{22} - \rho_{00} \end{pmatrix}, \quad (6)$$

where ρ_{00} is chosen to restore the condition

$$(\rho_{11} - \rho_{00})(\rho_{22} - \rho_{00}) = \rho_{12}\rho_{21}. \quad (7)$$

This division is the density matrix equivalent of a statement originally made by Stokes that the most general state of polarization of electromagnetic radiation can be described as a superposition of an unpolarized intensity and an elliptically polarized intensity.[11]

This density matrix notation is also suitable for describing the polarization dependence of scattering amplitudes and total cross sections. In fact, the density matrix describing the polarization of radiation emitted with wave vector **k** due to a particular nuclear transition is proportional to the 2×2 matrix defined in Sec. III to describe the forward coherent scattering due to the transition.

III. OPTIMIZATION OF EXPERIMENTAL CONDITIONS

We first consider coherence of the radiation scattered in other than the forward direction. As is expected in analogy with x ray and neutron diffraction for crystals, this coherence leads to generally destructive interference except along Bragg directions (of the crystallographic or magnetic space group) where strong constructive

[10] D. L. Falkoff and J. E. MacDonald, J. Opt. Soc. Am. **51**, 861 (1951).

[11] For a description of Stokes' work on polarized light, see S. Chandrasekhar, *Radiative Transfer* (Dover Publications, Inc., New York, 1960), pp. 24–35.

[12] U. Fano, Rev. Mod. Phys. **29**, 74 (1957).

interference occurs. If we imagine a slab of single-crystal absorber thin enough that saturation effects are negligible, the total cross section for the slab can differ from the sum of the individual nuclear cross sections because of these coherent effects. Where the total cross section is decreased, there is a corresponding decrease in the resonance linewidth and conversely an increase where the total cross section is increased. These effects have been discussed extensively by Trammell[13,14] whose results we use.

Again in analogy with neutron diffraction, it is possible to define a coherent scattering cross section σ_c for Mössbauer scattering. If we, for simplicity, consider a single resonance line unsplit by hyperfine interactions and a single resonant nucleus per unit cell, then

$$\sigma_c = \frac{1}{2|I_e - I_g| + 1} \left(\frac{2I_e + 1}{2I_g + 1} \right) \frac{f' R \sigma_T}{1 + \alpha}, \qquad (8)$$

where I_e and I_g are, respectively, the excited- and ground-state nuclear spins, α is the internal conversion coefficient, f' is the Debye-Waller factor here assumed isotropic, R is the isotopic abundance of the resonant species, and $\sigma_T = f' R \sigma_0$ is the average total resonant cross section per site, σ_0 being the fixed nucleus cross section. The ratio σ_c / σ_T is a measure of the importance of coherence since the actual cross section of a thin slab is reduced by that fraction for an observation direction not corresponding to any Bragg direction. If σ_c / σ_T is small, then even for an observation direction corresponding to a Bragg direction, the usual beam spread of a few degrees is probably sufficient to insure that any apparent change in cross section due to coherence is small. Hyperfine splittings and the existence of more than one resonant nucleus per unit cell will tend to further reduce the effects of coherence. Therefore, as can be seen by inserting numerical values in Eq. (8), coherence effects may be ignored in a wide variety of experimental situations. However, for highly enriched Fe^{57} or Sn^{119} absorbers, for example, they must be considered.

When coherence effects change the total cross section, it is clear that these changes will complicate attempts to measure f' values. Since σ_c / σ_T will generally be different for each component of a hyperfine split spectrum, they will also complicate attempts to determine magnetic structures and electric-field-gradient parameters.

By controlling the isotopic abundance of the resonance species, coherence can always be made to have a negligible effect on the total cross section. We will therefore assume this to be the case in our following analysis.

Let us now again consider a γ-ray beam of some definite polarization and energy incident on a thin slab of single-crystal absorber. If the energy of the beam

corresponds to a resonant frequency, then resonant modes of the nuclei in the crystal will be excited. These excited nuclei will reradiate with an angular distribution of intensity and a polarization determined by the hyperfine interactions and the multipole character of the transitions. In the forward direction, a large fraction of the scattered radiation is coherent with the incident beam, but, in general, the polarization of the radiation forward scattered by a particular nucleus will be different from that of the incident beam.

For simplicity let us consider a resonance line which is separated by $\gtrsim 100$ natural linewidths Γ from any other resonance lines and a crystal with only one resonant nucleus per unit cell. Then the complex index of refraction for the material is given by[15]

$$n = 1 + (2\pi N / k^2) F(\mathbf{k}, \mathbf{k}'), \qquad (9)$$

where $\mathbf{k}$ and $\mathbf{k}'$ are the wave vectors of the incident and scattered radiation, respectively, N is the number of resonant nuclei per unit volume, and $F(\mathbf{k}, \mathbf{k}')$ is the coherent scattering amplitude for a single nucleus. This expression exactly corresponds to that which can be derived from the classical electron theory for dipole radiation.[9]

The quantities F and hence n are, in general, not sample scalars. In fact, as recently discussed by Blume and Kistner,[6] they may be represented by complex 2×2 matrices. These may be defined in terms of arbitrarily chosen orthogonal basis polarizations. If an incident beam having the first of the basis polarizations is imagined, then F_{11} is the projection of the coherent forward scattering amplitude on that polarization, while F_{12} is its projection on the orthogonal basis polarization. The matrix elements F_{21} and F_{22} are similarly defined with respect to the other basis polarization. From this definition, it can be seen that the elements of the coherent forward scattering amplitude matrix are proportional to the elements of the density matrix describing radiation emitted in the same direction if both are expressed in terms of the same basis system.

It is natural to try to choose a basis system in which the complex index of refraction is diagonal. For the simple case under consideration (single isolated resonance line, only one resonant site per unit cell), this is always possible since both the real and imaginary parts of F have the same polarization.

Generalization of the above to complicated crystals having several resonant nuclear sites per unit cell, each with its own hyperfine interactions, is trivial. We merely replace Eq. (9) by

$$n = 1 + \frac{2\pi}{k^2} \sum_j N_j F_j(\mathbf{k}, \mathbf{k}) \equiv 1 + \frac{2\pi}{k^2} F_r(\mathbf{k}, \mathbf{k}), \qquad (10)$$

where the sum runs over all the sites. In saying this, we have assumed that the electromagnetic interaction is so

[13] G. T. Trammell, in *Chemical Effects of Nuclear Transformations* (International Atomic Energy Agency, Vienna, 1961), Vol. I, p. 75.
[14] G. T. Trammell, Phys. Rev. **126**, 1045 (1962).

[15] M. Lax, Rev. Mod. Phys. **23**, 287 (1951).

weak that the index of refraction has a negligible influence on an incident beam in a distance of order of the unit cell dimensions. In practice, this appears to always be the case, even for resonant scattering. Similarly, neglect of polarization effects in a random powder is strictly correct only if each grain absorbs a negligible fraction of the beam.

Expressed in terms of the density matrix notation introduced in Sec. II, Eq. (10) becomes

$$n = 1 + i\frac{\sigma_0}{2k}\sum_{ij}\begin{pmatrix}\rho_{11}^{ij} & \rho_{12}^{ij}\\ \rho_{21}^{ij} & \rho_{22}^{ij}\end{pmatrix}\frac{N_j f_j'}{(x-x_{ij})^2+1}$$

$$-\frac{\sigma_0}{2k}\sum_{ij}\begin{pmatrix}\rho_{11}^{ij} & \rho_{12}^{ij}\\ \rho_{21}^{ij} & \rho_{22}^{ij}\end{pmatrix}\frac{N_j f_j'(x-x_{ij})}{(x-x_{ij})^2+1}, \quad (11)$$

where sites are labeled by j and hyperfine components of a resonance by i. The quantity x is related to the energy E by $x=2E/\Gamma$ and the x_{ij}'s correspond to the resonant energies at the different sites. The assumption that all lines have Lorentz shapes of natural width Γ is inessential and has only been introduced to keep the formulas simple. The Debye-Waller factors are no longer assumed isotropic but are the ones appropriate for the direction $\mathbf{k}$. The normalization for the density matrices may be found by imagining the hyperfine splittings of the different sites to collapse to zero. Then the cross section must be unpolarized and of magnitude $f_j'\sigma_0$. This gives

$$\sum_i \rho_{12}^{ij}=\sum_i \rho_{21}^{ij}=0 \quad (12)$$

and

$$\sum_i \rho_{11}^{ij}=\sum_i \rho_{22}^{ij}=1. \quad (13)$$

It is easy to see from Eq. (11) that, in general, the real and imaginary parts of n cannot be diagonalized at the the same time, even if there is only one resonant site per unit cell, since the differently polarized contributions depend on energy in different ways.

Blume and Kistner[6] have derived a general expression for the intensity of a beam transmitted through a slab of material of any thickness having an index of refraction described by a complex matrix. In addition, they have shown how this index of refraction matrix can be calculated if the nuclear wave functions, the multipolarity of the transition, and the Debye-Waller factor are known. While this procedure is straightforward, it appears that inverting it in the general case to obtain f' values and nuclear wave functions (and hence EFG parameters or magnetic structures) would be difficult.

In the special cases where n is diagonal, an incident radiation beam may be decomposed into two components with polarizations corresponding to the basis polarizations in which n is diagonal. Each of these beams may be regarded as separately propagating through the crystal with its own index of refraction.

An effective thickness matrix σ may be defined from Eq. (11) and will be diagonal when n is diagonal

$$\sigma \equiv \begin{pmatrix}\sigma_{11} & \sigma_{12}\\ \sigma_{21} & \sigma_{22}\end{pmatrix} \equiv \sigma_0 \sum_{ij}\begin{pmatrix}\rho_{11}^{ij} & \rho_{12}^{ij}\\ \rho_{21}^{ij} & \rho_{22}^{ij}\end{pmatrix}\frac{n_j f_j'}{(x-x_{ij})^2+1}, \quad (14)$$

where n_j are the number of resonant nuclei of type j per unit area. When n is diagonal, the matrix elements of σ can be extracted from experimental line intensities in a straightforward manner if the spectra are well resolved. Procedures for doing this are discussed in Sec. IV. These matrix elements can then be related to the f' values and nuclear wave functions. Assuming enough independent measurements are available, these equations can be solved for the parameters of interest.

Fortunately, several restrictions can be placed on the index of refraction matrix from crystal symmetry considerations alone. These allow an experimentally interesting class of cases to be found in which it can be concluded that n is diagonal in a basis system which is defined in terms of symmetry elements of the crystal. Since magnetic hyperfine splittings are important in magnetically ordered crystals, the symmetry operations we must consider are those of the magnetic space group of the crystal.[16,17] These involve combinations of time reversal, which inverts $\mathbf{k}$, with the more familiar elements of rotation, reflection, etc.

To derive these restrictions, we imagine an infinite crystal with a plane wave or Bloch wave of propagation vector $\mathbf{k}$ and polarization $\mathbf{P}$ traveling through it. Any symmetry operation of the crystal which also leaves $\mathbf{k}$ and $\mathbf{P}$ unchanged must leave the resultant coherent forward scattering amplitude F_r of Eq. (11) unchanged. By applying this test for various $\mathbf{k}$ and $\mathbf{P}$, we arrive at the following general conclusions.

1. If the crystal contains an anti-inversion center, then for any original linear polarization $\mathbf{P}$ there can be no elliptical component in the scattering amplitude. Although any linear polarization is unchanged by the operation, an elliptical component would be reversed in sense. Therefore, a crystal containing an anti-inversion center cannot be optically active. In a nonmagnetically ordered crystal, an inversion center and an anti-inversion center are equivalent.

2. If $\mathbf{k}$ is in a mirror or glide plane, then polarizations normal and parallel to this plane are unchanged. This applies, in particular, to both the real and imaginary parts of the forward scattering amplitude. Therefore, the index of refraction is diagonal in a system having basis polarizations normal and parallel to the plane.

3. If $\mathbf{k}$ is along a three-fold or higher simple rotation or screw axis, then a $\mathbf{P}$ corresponding to right or left circular polarization is unchanged by the rotations. Then both real and imaginary parts of $F_r(\mathbf{k},\mathbf{k})$ must

[16] M. Atoji, Am. J. Phys. **33**, 212 (1965).
[17] G. Donnay, L. M. Corliss, J. D. H. Donnay, N. Elliott, and J. M. Hastings, Phys. Rev. **112**, 1917 (1958).

have circular polarization and n must be diagonal in a system with right and left circular basis polarizations.

4. If $\mathbf{k}$ is normal to a 2-, 4-, or 6-fold axis of rotary inversion or normal to a mirror plane which contains a 2-, 4-, or 6-fold rotation or screw axis, we can conclude that $\mathbf{P}$'s parallel or normal to the axis will be unchanged by the symmetry operation of rotation by 180° followed by inversion or reflection. Therefore, n will be diagonal in a system with basis vectors parallel and normal to the symmetry axis. Also, if $\mathbf{k}$ is normal to a 2-, 4-, or 6-fold axis in a nonmagnetically ordered crystal so that time reversal is a symmetry element, then n will be diagonal in the same system.

We have thus found three classes of $\mathbf{k}$ directions in which n can always be diagonalized, namely $\mathbf{k}$ in a mirror or glide plane, $\mathbf{k}$ along a 3-fold or higher rotation or screw axis, and $\mathbf{k}$ normal to a suitable 2-, 4-, or 6-fold rotation axis.

Since the above considerations are independent of wavelength, these conclusions must hold at all wavelengths, including the visible. These directions are experimentally generally convenient and frequently measurements along them will be sufficient to determine the desired parameters to the extent that this is possible from absorption measurements.

If the multipolarity of the transition and the type of hyperfine interaction are specified, additional symmetry restrictions may be found. For example, if there are only two lines in the spectrum as occurs for an $I = \frac{3}{2} \to \frac{1}{2}$ or $\frac{1}{2} \to \frac{3}{2}$ nuclear transition in a pure quadrupole field gradient and only one resonant site per unit cell, then the index of refraction may be diagonalized for any $\mathbf{k}$ direction.

It should be mentioned that there is an intrinsic difficulty in any attempt to determine hyperfine-field parameters, for crystals containing more than one crystallographically equivalent resonant site per unit cell, from transmission measurements alone. Although a set of equations may be found which, when applied to the experimental data yield a number of sets of internal field parameters equal to the number of resonant sites, there is nothing to identify which internal field corresponds to which site.

This identification could, in principle, be made from resonant scattering measurements. In practice, it can sometimes be made with some confidence from the local symmetry and known chemical properties of the resonant atom.

It seems plausible that if the initial γ-ray beam is unpolarized, changes in polarization due to the real part of the index of refraction will not become important until a substantial polarization has built up due to the polarization dependence of the absorption. This can be verified by expanding Eq. (5) of Blume and Kistner.[6] The attenuation in this case is given correctly to second order in the thickness by keeping only diagonal elements in the index of refraction matrix. Therefore, if the

incident radiation is unpolarized and the absorber is thin enough, for example, to be adequately described by the first two terms of Eqs. (20) and (21) of Sec. IV, it becomes unimportant to restrict measurements to directions in which n can be diagonalized. In other cases, it may be very important.

IV. ABSORPTION BY POLARIZED OR PARTIALLY POLARIZED ABSORBER

In this section, we assume that we have a γ-ray beam that can be described as a plane wave incident on a slab of homogeneous absorbing material of uniform thickness.

We also assume one of the conditions of the previous sections holds so that we may describe absorption without considering dispersion. It is natural to work in the basis system where σ of Eq. (14) is diagonal. We can describe the polarization and intensity of the unshifted component of the source radiation at any energy by a density matrix S in the same basis system

$$S \equiv \frac{I}{\pi} \begin{pmatrix} S_{11} & S_{12} \\ S_{21} & S_{22} \end{pmatrix}$$

$$= \frac{I}{2\pi} \sum_{k,l} \begin{pmatrix} \rho_{11}{}^{kl} & \rho_{12}{}^{kl} \\ \rho_{21}{}^{kl} & \rho_{22}{}^{kl} \end{pmatrix} \frac{r_l f_l}{(x - x_{kl} - v)^2 + 1}, \quad (15)$$

where I is the total intensity due to the transition of interest and r_l is the fraction of the emitting nuclei in site l. The source resonance energies x_{kl} and site positions l will not generally correspond to those in the absorber. The quantity v describes the Doppler shift in the resonance energies due to relative motion of the source and absorber. The normalization of the density matrices is the same as given by Eqs. (12) and (13), which accounts for the 2 in the denominator on the right. Generally, sources are used in which all sites are equivalent and the different lines are well resolved.

The general expression for the transmitted intensity at a particular energy x when radiation from a source described by Eq. (15) is incident on an absorber described by Eq. (14) can easily be found since S_{11} gives the projection of the unshifted source intensity on one of the basis polarizations and S_{22} that on the other. We get

$$I(x,v) = \frac{I}{\pi} [S_{11} e^{-\sigma_{11}} + S_{22} e^{-\sigma_{22}}], \quad (16)$$

which is similar to the corresponding expression for unpolarized radiation except for containing two terms instead of one on the right. It may be subtracted from the incident intensity $S_{11} + S_{22}$ and the result integrated over x to obtain the fractional absorption dip at any Doppler velocity v. This may then be integrated over v to obtain the total dimensionless area in the absorption spectrum.

The unknown parameters of the experiment are contained in the σ's and hopefully can be extracted from measurements made in different directions and/or with different thickness absorbers. This extraction is greatly simplified if the absorption lines are well resolved. In that case we can define individual areas for each one, and assuming both source and absorber resonance lines have natural widths, these are given simply by a generalization of Eq. (11) of Bykov and Hein.[18] For the overlap of the kth source line from site l with the ith absorber line from site j, this gives

$$A_{ij}{}^{kl} = S_{11}(x_{kl})\sigma_{11}(x_{ij})\ \exp[\sigma_{11}(x_{ij})/2]\{I_0[\sigma_{11}(x_{ij})/2]$$
$$+I_1[\sigma_{11}(x_{ij})/2]\}+S_{22}(x_{kl})\sigma_{22}(x_{ij})\ \exp[\sigma_{22}(x_{ij})/2]$$
$$\times\{I_0[\sigma_{22}(x_{ij})/2]+I_1[\sigma_{22}(x_{ij})/2]\}, \quad (17)$$

where I_0 and I_1 are the zeroth- and first-order Bessel functions of imaginary argument. When different absorber lines have the same energy, their contributions are added in the arguments of the functions in Eq. (17). The dimensionless area $A_{ij}{}^{kl}$ is related to the background corrected experimental area B by

$$A_{ij}{}^{kl} = 2B/\pi\Gamma. \quad (18)$$

A simpler approximate formula which underestimates $A_{ij}{}^{kl}$ by less than 1% for σ_{11}, $\sigma_{22}\leq 2$ is

$$A_{ij}{}^{kl}\simeq S_{11}(x_{kl})\frac{\sigma_{11}(x_{ij})}{1+\sigma_{11}(x_{ij})/4}+S_{22}(x_{kl})\frac{\sigma_{22}(x_{ij})}{1+\sigma_{22}(x_{ij})/4}. \quad (19)$$

For relatively thin absorbers, power-series expansions of Eq. (17) are useful. If we restrict ourselves for simplicity to a single line unpolarized source $S_{11}=S_{22}=f/2$ and if we define the average cross section on resonance $p=[\sigma_{11}(x_{ij})+\sigma_{22}(x_{ij})]/2$ and fractional polarization $a=(\sigma_{11}-\sigma_{22})/(\sigma_{11}+\sigma_{22})$ of the absorption, we may write

$$A=p-\tfrac{1}{4}(1+a^2)p^2+\tfrac{1}{16}(1+3a^2)p^3$$
$$-(5/384)(1+6a^2+a^4)p^4+\cdots \quad (20)$$

or inverting Eq. (20)

$$p=A+\tfrac{1}{4}(1+a^2)A^2+\tfrac{1}{16}(1+a^2+2a^4)A^3$$
$$+(5/384)(1+a^4+6a^6)A^4+\cdots, \quad (21)$$

where $A=A_{ij}/f$. We notice that the lowest power of the fractional polarization which enters is the square. Therefore, for a *relatively thin* absorber, the effects of polarization may be quite small unless the fractional polarization becomes large.

Equations for area ratios which have previously been used in analyzing experimental data[19–24] can be ob-

tained by dropping all but the first terms in Eqs. (20) or (21).

V. POLARIZATION OF MÖSSBAUER LINES

A. General

The polarization of the hyperfine components of Mössbauer γ rays has previously been discussed by Frauenfelder et al.[25] and by Dehn, Marzolf, and Salman.[26] They primarily considered the case of magnetized simple ferromagnetic materials without quadrupole splitting. Therefore, they have (1) only considered a pure magnetic hyperfine interaction such that the nuclear states could be described by the quantum numbers I and m_I in a suitable coordinate system. A quadrupole interaction with an EFG tensor not having axial symmetry or a noncolinear mixed magnetic and quadrupole interaction do not satisfy this requirement. (2) They have not considered the case where different nuclear transitions have the same energy which always occurs for a pure quadrupole interaction. (3) They have only considered the case where all nuclei are in a field having the same direction as well as magnitude. This will not generally be the case if there is more than one resonant nucleus per unit cell. When conditions (2) or (3) are violated, the cross section will be only partially polarized and something equivalent to our density matrix method of description is required.

In order to derive the results we need in as simple a manner as possible, we make use of the fact that the polarization of the scattering amplitude due to a given transition is the same as the polarization for emission with the same **k**. We start out following the methods of Blatt and Weisskopf.[27] We choose a system of coordinates which is convenient for the description of the nuclear states at one site and make a multipole expansion of the amplitude of the electric-field vector of the radiation field.

$$\mathcal{E}(\mathbf{r})=\sum_{l=1}^{\infty}\sum_{m=-l}^{l}a_E(l,m)\,\mathcal{E}_E(l,m,\mathbf{r})$$
$$+a_M(l,m)\,\mathcal{E}_M(l,m,\mathbf{r}), \quad (22)$$

where $\mathbf{r}$ is the position vector, $\mathcal{E}_E(l,m,\mathbf{r})$ and $\mathcal{E}_M(l,m,\mathbf{r})$ are, respectively, the electric and magnetic multipole fields of order l, m, and $a_E(l,m)$, and $a_M(l,m)$ are the source terms for the multipole fields and depend on the

[18] G. A. Bykov and Pham Zuy Hien, Zh. Eksperim. i Teor. Fiz. **43**, 909 (1962) [English transl.: Soviet Phys.—JETP **16**, 646 (1963)].

[19] P. Zory, Phys. Rev. **140**, A1401 (1965).

[20] J. Danon and L. Iannarella, J. Chem. Phys. **47**, 382 (1967).

[21] R. Ingalls, K. Ono, and L. Chandler, Phys. Rev. **172**, 295 (1968).

[22] Pham Zuy Jien, Zh. Eksperim. i Teor. Fiz. **49**, 1576 (1965) [English transl.: Soviet Phys.—JETP **22**, 1080 (1966)].

[23] S. V. Karyagin, Fiz. Tverd. Tela **8**, 1739 (1966) [English transl.: Soviet Phys.—Solid State **8**, 1387 (1966)].

[24] S. V. Karyagin, Fiz. Tverd. Tela **9**, 2514 (1967) [English, transl.: Soviet Phys.—Solid State **9**, 1979 (1968)].

[25] H. Frauenfelder, D. E. Nagle, R. D. Taylor, D. R. F. Cochran, and W. M. Visscher, Phys. Rev. **126**, 1065 (1962).

[26] J. T. Dehn, J. G. Marzolf, and J. F. Salman, Phys. Rev. **135**, B1307 (1964).

[27] J. M. Blatt and W. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley & Sons, Inc., New York, 1952), Chap. XII, pp. 583–600 and Appendices A and B.

time variation of the charge and magnetization distribution in the nucleus.

The multipole fields are defined in terms of products of spherical Bessel functions and vector spherical harmonics which are in turn defined in terms of the scalar spherical harmonics and the complex unit vectors

$$\hat{e}_{-1} = (1/\sqrt{2})(\hat{e}_x - i\hat{e}_y), \quad \hat{e}_0 = \hat{e}_z, \quad \hat{e}_1 = -(1/\sqrt{2})(\hat{e}_x + i\hat{e}_y). \quad (23)$$

The expression Eq. (22) for the polarization of the radiation can be simplified by transforming to a new system of coordinates in which the z' axis is parallel to the direction of propagation $\mathbf{k}$ of the radiation. Since electromagnetic radiation is transverse, we expect the coefficient of $\hat{e}_z$, to be zero.[28] A simple way to specify such a new coordinate system is to perform a rotation of φ about the z axis followed by a rotation of θ about the resulting y axis, where θ and φ are the usual polar angles describing the direction $\mathbf{k}$. The unit vectors in this new system are the usual spherical unit vectors $\hat{r}$, $\hat{\theta}$, and $\hat{\phi}$. For further simplicity we will write our result directly in terms of these unit vectors rather than in terms of the transformed complex unit vectors. The expressions for our original complex unit vectors in terms of these new unit vectors are easily obtained by performing this transformation and the two rotations in succession.

$$\begin{bmatrix}\hat{e}_{-1}\\\hat{e}_1\\\hat{e}_0\end{bmatrix} = 1/\sqrt{2}\begin{bmatrix}1 & -i & 0\\-1 & -i & 0\\0 & 0 & \sqrt{2}\end{bmatrix}\begin{bmatrix}\cos\varphi & -\sin\varphi & 0\\\sin\varphi & \cos\varphi & 0\\0 & 0 & 1\end{bmatrix}$$

$$\times\begin{bmatrix}\cos\theta & 0 & \sin\theta\\0 & 1 & 0\\-\sin\theta & 0 & \cos\theta\end{bmatrix}\begin{bmatrix}\hat{\theta}\\\hat{\phi}\\\hat{r}\end{bmatrix} \quad (24)$$

$$= 1/\sqrt{2}\begin{bmatrix}e^{-i\phi}\cos\theta & -ie^{-i\varphi} & e^{-i\varphi}\sin\theta\\-e^{i\varphi}\cos\theta & -ie^{i\varphi} & -e^{i\varphi}\sin\theta\\-\sqrt{2}\sin\theta & 0 & \sqrt{2}\cos\theta\end{bmatrix}\begin{bmatrix}\hat{\theta}\\\hat{\phi}\\\hat{r}\end{bmatrix}.$$

Substituting Eq. (24) in Eq. (22) and dropping terms which fall off faster than r^{-1} and do not contribute to the radiation,[28] we get the simple form

$$\boldsymbol{\varepsilon}(\mathbf{r}) = R(r)[a_\theta(\theta,\phi)\hat{\theta} + a_\phi(\theta,\phi)\hat{\phi}]. \quad (25)$$

We must now see how to determine the coefficients $a_\theta(\theta,\phi)$ for an arbitrary nuclear transition. The coefficients $a_E(l,m)$ and $a_M(l,m)$ in Eq. (22) are defined in terms of reduced nuclear matrix elements which cannot be explicitly evaluated at the present stage of nuclear theory. In the case that the z projections m_I of the nuclear spin I are good quantum numbers for the angular part of the nuclear wave functions, we can apply the vector addition law of angular momenta to the radiation of any multipole order and obtain relative

[28] This is strictly true for the magnetic-multipole fields and is true for the radiation component of the electric-multipole fields. However, in the transition region the electric-multipole fields have a radial component which falls off as $1/r^2$. See, for example, W. K. H. Panofsky and M. Phillips, *Classical Electricity and Magnetism* (Addison-Wesley Publishing Co., Inc., Reading, Mass., 1955), Sec. 13–6.

values for the $a(l,m)$ involved in a particular transition $m_e \to m_g$ in terms of Clebsch-Gordan coefficients

$$a(l,m) \sim C_{lI_g}(I_e, m_e, m, m_g), \quad (26)$$

where the subscript e stands for the nuclear excited state and g for the ground state. If more than one multipole order is involved, the $a(l,m)$ for each should be weighted in proportion to the experimentally determined amplitude mixing ratio X, $= E2/M1$, for example.

For the more general case where m_e is not a good quantum number to use in describing the nuclear states, we will write the angular part of the ith excited and the jth ground-state nuclear wave functions in terms of expansions in m as

$$|e\rangle_i = \sum_{m_e=-I_e}^{I_e} \alpha_i(m_e)|m_e\rangle \quad (27)$$

and

$$|g\rangle_j = \sum_{m_g=-I_g}^{I_g} \alpha_j(m_g)|m_g\rangle. \quad (28)$$

From this we see that in the nuclear transition $|e\rangle_i \to |g\rangle_j$ the relative values of the different $a(l,m)$ are

$$a_{Eij}(l,m) \sim X \sum_{m_e-m_g=m} \alpha_i(m_e)\alpha_j^*(m_g)$$
$$\times C_{lI_g}(I_e, m_e, m, m_g) \quad (29)$$

and

$$a_{Mij}(l,m) \sim \sum_{m_e-m_g=m} \alpha_i(m_e)\alpha_j^*(m_g)C_{lI_g}(I_e, m_e, m, m_g). \quad (30)$$

By substituting Eqs. (29) and (30) in Eq. (22), we can determine the coefficients in Eq. (25) except for a constant factor which to a very good approximation we can assume is the same for all hyperfine components of a given nuclear transition.

B. Fe[57] Quadrupole Interaction

The 14.4-keV Mössbauer level of Fe[57] has very nearly pure magnetic dipole character. For pure $M1$ radiation Eq. (22) becomes

$$\boldsymbol{\varepsilon}(\mathbf{r}) = R(r)[a_M(1,1)e^{i\varphi}(\hat{\theta}+i\cos\theta\hat{\phi}) + a_M(1,0)i\sqrt{2}\sin\theta\hat{\phi}$$
$$+ a_M(1,-1)e^{-i\varphi}(\hat{\theta}-i\cos\theta\hat{\phi})], \quad (31)$$

where numerical factors have been absorbed in $R(r)$. Rewriting this in the form of Eq. (25), we have

$$\boldsymbol{\varepsilon}(\mathbf{r}) = R(r)\{[a_M(1,1)e^{i\varphi} + a_M(1,-1)e^{-i\varphi}\hat{\theta}]$$
$$+ i[a_M(1,1)e^{i\varphi}\cos\theta + a_M(1,0)\sqrt{2}\sin\theta$$
$$- a_M(1,-1)e^{-i\varphi}\cos\theta]\hat{\phi}\}. \quad (32)$$

We now consider the case where the nucleus is subjected to a pure quadrupole interaction. We specify our coordinate system to be the principal axis system in which $|V_{zz}| \geq |V_{yy}| \geq |V_{xx}|$ and define $\eta = (V_{xx} - V_{yy})/V_{zz}$. It is clear that $0 \leq \eta \leq 1$. In this system one can show[19] that the angular parts of the wave functions for the

$I = \frac{3}{2}$ nuclear excited state are

$$|\tfrac{3}{2}\rangle \rightleftharpoons \gamma | \quad \tfrac{3}{2}\rangle + \delta | -\tfrac{1}{2}\rangle,$$

$$|-\tfrac{3}{2}\rangle \rightleftharpoons \gamma | -\tfrac{3}{2}\rangle + \delta | \quad \tfrac{1}{2}\rangle, \qquad (33)$$

$$|-\tfrac{1}{2}\rangle \rightleftharpoons \delta | \quad \tfrac{3}{2}\rangle - \gamma | -\tfrac{1}{2}\rangle,$$

$$|\tfrac{1}{2}\rangle \rightleftharpoons \delta | -\tfrac{3}{2}\rangle - \gamma | \quad \tfrac{1}{2}\rangle,$$

where

$$\gamma = \frac{1}{\sqrt{2}}\left[1 + \left(\frac{3}{3+\eta^2}\right)^{1/2}\right]^{1/2},$$

$$\delta = \frac{1}{\sqrt{2}}\left[1 - \left(\frac{3}{3+\eta^2}\right)^{1/2}\right]^{1/2}, \qquad (34)$$

and the states have been labeled by the m_I values which become good quantum numbers when $\eta = 0$. The first two states have the energy $E_3 = (e^2 qQ/4)(1+\eta^2/3)^{1/2}$ while the second two have the energy $E_1 = -(e^2 qQ/4) \times (1+\eta^2/3)^{1/2}$, where Q is the nuclear quadrupole moment and q is V_{zz}/e. Transitions between these excited states and the doubly degenerate ground state have equal probability. If we, without loss of generality,

TABLE I. Multipole source terms.

A. Transitions with energy E_3				
	$|\tfrac{3}{2}\rangle \rightarrow |\tfrac{1}{2}\rangle$	$|\tfrac{3}{2}\rangle \rightarrow |-\tfrac{1}{2}\rangle$	$|-\tfrac{3}{2}\rangle \rightarrow |\tfrac{1}{2}\rangle$	$|-\tfrac{3}{2}\rangle \rightarrow |-\tfrac{1}{2}\rangle$
$a_M(1,1)$	γ	0	0	$\delta/\sqrt{3}$
$a_M(1,0)$	0	$\sqrt{2}\delta/\sqrt{3}$	$\sqrt{2}\delta/\sqrt{3}$	0
$a_M(1,-1)$	$\delta/\sqrt{3}$	0	0	γ

B. Transitions with energy E_1				
	$|-\tfrac{1}{2}\rangle \rightarrow |\tfrac{1}{2}\rangle$	$|-\tfrac{1}{2}\rangle \rightarrow |-\tfrac{1}{2}\rangle$	$|\tfrac{1}{2}\rangle \rightarrow |\tfrac{1}{2}\rangle$	$|\tfrac{1}{2}\rangle \rightarrow |-\tfrac{1}{2}\rangle$
$a_M(1,1)$	δ	0	0	$-\gamma/\sqrt{3}$
$a_M(1,0)$	0	$-\sqrt{2}\gamma/\sqrt{3}$	$-\sqrt{2}\gamma/\sqrt{3}$	0
$a_M(1,-1)$	$-\gamma/\sqrt{3}$	0	0	δ

label the ground-state wave functions with their m values $|\tfrac{1}{2}\rangle$ and $|-\tfrac{1}{2}\rangle$, we can use Eqs. (28) and (30) to evaluate the $a_M(l,m)$ appearing in Eq. (32). We take $X = 0$.

For the groups of states having energy E_3 and E_1, we find that the $a_M(l,m)$ values are, respectively, proportional to the quantities tabulated in part A and part B of Table I. From the definition of the elements of the density matrix, Eqs. (25) and (1), we have in the $\hat{\theta}, \hat{\varphi}$

$$\rho_{11}{}^j = c \sum \left[|a_M(1,1)|^2 + |a_M(1,-1)|^2 + a_M{}^*(1,1)a_M(1,-1)e^{-2i\varphi} + a_M(1,1)a_M{}^*(1,-1)e^{2i\varphi} \right],$$

$$\rho_{22}{}^j = c \sum \left[|a_M(1,1)|^2 \cos^2\theta + 2|a_M(1,0)|^2 \sin^2\theta + |a_M(1,-1)|^2 \cos^2\theta + a_M{}^*(1,1)a_M(1,0)\sqrt{2}e^{-i\varphi}\sin\theta\cos\theta \right.$$
$$- a_M{}^*(1,1)a_M(1,-1)e^{-2i\varphi}\cos^2\theta + a_M{}^*(1,0)a_M(1,1)\sqrt{2}e^{i\varphi}\sin\theta\cos\theta - a_M{}^*(1,0)a_M(1,-1)\sqrt{2}e^{-i\varphi}\cos\theta\sin\theta$$
$$\left. - a_M{}^*(1,-1)a_M(1,1)e^{2i\varphi}\cos^2\theta - a_M{}^*(1,-1)a_M(1,0)\sqrt{2}e^{i\varphi}\sin\theta\cos\theta \right], \qquad (35)$$

$$\rho_{12}{}^j = \rho_{21}{}^{*j} = ic \sum \left[|a_M(1,1)|^2 \cos\theta + a_M{}^*(1,1)a_M(1,0)\sqrt{2}e^{-i\varphi}\sin\theta - a_M{}^*(1,1)a_M(1,-1)e^{-2i\varphi}\cos\theta \right.$$
$$\left. + a_M{}^*(1,-1)a_M(1,1)e^{2i\varphi}\cos\theta + a_M{}^*(1,-1)a_M(1,0)\sqrt{2}e^{i\varphi}\sin\theta - |a_M(1,-1)|^2\cos\theta \right],$$

where the sums extend over the group of states with the same energy. The various sums over products of $a_M(l,m)$ appearing above are tabulated in Table II where the definitions in Eqs. (34) have been used to eliminate γ and δ. Using these results and the normalization condition Eq. (13), the above equations simplify to

$$\rho_{11}{}^j = \tfrac{1}{2} \pm \tfrac{1}{4}[3/(3+\eta^2)]^{1/2}(1 + \eta \cos2\varphi),$$

$$\rho_{22}{}^j = \tfrac{1}{2} \pm \tfrac{1}{4}[3/(3+\eta^2)]^{1/2}(1 - 3 \sin^2\theta - \eta \cos^2\theta \cos2\varphi),$$

$$\rho_{12}{}^j = \rho_{21}{}^j = \mp (\eta/4)[3/(3+\eta^2)]^{1/2} \cos\theta \sin2\varphi, \qquad (36)$$

where the upper signs correspond to the E_3 transitions and the lower ones to the E_1 transitions.

It is interesting to notice that $\rho_{12}{}^j$ and $\rho_{21}{}^j$ are always real for this case which indicates that there is no el-

lipticity in the polarization in any direction, the lines being only partially linearly polarized. The fractional polarizations along several interesting directions as a function of η are

$$\mathbf{x}: \quad a = \frac{3+\eta}{4[\tfrac{1}{3}(3+\eta^2)]^{1/2} \mp (1-\eta)},$$

$$\mathbf{y}: \quad a = \frac{3-\eta}{4[\tfrac{1}{3}(3+\eta^2)]^{1/2} \mp (1+\eta)}, \qquad (37)$$

$$\mathbf{z}: \quad a = \frac{2\eta}{4[\tfrac{1}{3}(3+\eta^2)]^{1/2} \pm 2},$$

$$\mathbf{x+y+z}: \quad a = \tfrac{1}{2},$$

TABLE II. Sums over products of source terms for pure quadrupole interaction.

	E_3	E_1
$\sum |a_M(1,1)|^2$	$\gamma^2 + \tfrac{1}{3}\delta^2 = \tfrac{2}{3} + \tfrac{1}{3}[3/(3+\eta^2)]^{1/2}$	$\delta^2 + \tfrac{1}{3}\gamma^2 = \tfrac{2}{3} - \tfrac{1}{3}[3/(3+\eta^2)]^{1/2}$
$\sum |a_M(1,-1)|^2$	$\gamma^2 + \tfrac{1}{3}\delta^2 = \tfrac{2}{3} + \tfrac{1}{3}[3/(3+\eta^2)]^{1/2}$	$\delta^2 + \tfrac{1}{3}\gamma^2 = \tfrac{2}{3} - \tfrac{1}{3}[3/(3+\eta^2)]^{1/2}$
$\sum |a_M(1,0)|^2$	$\tfrac{4}{3}\delta^2 = \tfrac{2}{3} - \tfrac{2}{3}[3/(3+\eta^2)]^{1/2}$	$\tfrac{4}{3}\gamma^2 = \tfrac{2}{3} + \tfrac{2}{3}[3/(3+\eta^2)]^{1/2}$
$\sum a_M{}^*(1,1)a_M(1,-1)$	$(2\delta\gamma/\sqrt{3}) = \tfrac{1}{3}\eta[3/(3+\eta^2)]^{1/2}$	$-(2\delta\gamma/\sqrt{3}) = -\tfrac{1}{3}\eta[3/(3+\eta^2)]^{1/2}$
$\sum a_M{}^*(1,1)a_M(1,0)$	0	0
$\sum a_M{}^*(1,0)a_M(1,-1)$	0	0

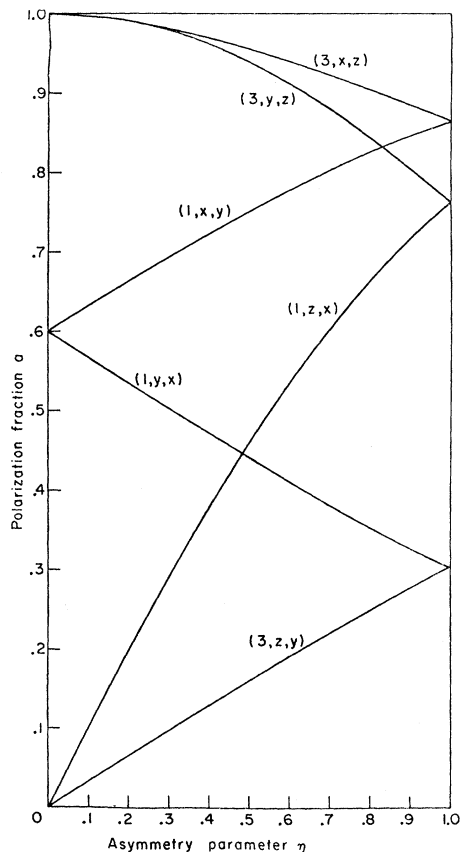FIG. 1. Fractional polarization of radiation emitted by Fe^{57} in a pure quadrupole field gradient as a function of asymmetry parameter η for three high-symmetry directions. Radiation from the excited states with $m=\pm\frac{3}{2}$ when $\eta=0$ is indicated by 3 and similarly that with $m=\pm\frac{1}{2}$ by 1. The other quantities in parentheses indicate, respectively, the observation direction and the polarization direction.

where again the upper signs correspond to the E_3 and the lower to the E_1 transitions. These results together with the directions of the polarization are shown in Fig. 1.

When the crystal of interest contains more than one set of nuclei whose electric and/or magnetic fields have different directions, we must add the density matrices from all these sites. In general, for a given observation direction, the $\hat{\theta}$ and $\hat{\varphi}$ directions for each site will be different. It is therefore necessary to transform all the density matrices to a common basis system. Calculating

the appropriate rotation angles α_i in terms of the parameters describing the fields at the nuclei is a straightforward geometrical problem. The new density matrices can then be found by applying the standard similarity transformation used to express a matrix in a rotated coordinate system.

VI. SUMMARY

Our analysis has been motivated by the desire to obtain information on hyperfine fields and lattice dynamics from the analysis of Mössbauer absorption measurements on single crystals. Therefore, we have attempted to find conditions in which the relations between these quantities and the experimental intensities are as simple as possible, taking into consideration coherence, polarization, and saturation effects. We found three general types of situations in which the relation can take a simple form. (1) The absorber is so thin that effects higher than second order in the thickness can be neglected. (The second-order saturation correction must still be made.) (2) There are only two lines in the spectrum and one site per unit cell. (3) The wave vector **k** is along a suitable symmetry direction in the crystal.

This last condition appears to be the most important since it applies even if there are several resonant sites in the unit cell, each with a complicated spectrum.

As an outline of how our results can be used, we suggest the following. (1) Make measurements along symmetry directions in the crystal where the index of refraction matrix is known to be diagonal if possible. Otherwise, use very thin samples. In the special case in which the spectrum contains only two lines and there is only one resonant site per unit cell, these restrictions are not necessary. (2) If the spectra are well resolved, obtain the average cross section p and fractional polarization a values from the equations of Sec. IV. Using the techniques described in Sec. V, relate these to the unknown parameters whose values are to be determined. This procedure is used in the following paper on Fe in sodium nitroprusside.[7]

ACKNOWLEDGMENT