where σ_0 is the total cross section on resonance ($E = E_\beta - E_\alpha$). Inserting equation (3.29) into equation (3.30) the matrix r_{pq} (α,β is dropped) is the sum ($\delta = M_1/E_2$):

$$\begin{aligned} r_{pq} = (1 + \delta^2)^{-1} \sum_{M,M'} & D^{1+}_{pM} V^*_{1M} V_{1M'} D^1_{M'q} \\ & + p\cdot q\delta^2 D^{2+}_{pM} V^*_{2M} V_{2M'} D^2_{M'q} - \delta(p\cdot D^{2+}_{pM} V^*_{2M} V_{1M'} D^1_{M'q} \\ & + q\cdot D^{1+}_{pM} V^*_{1M} V_{2M'} D^2_{M'q}) \end{aligned} \tag{3.32}$$

The lengthy expressions for the angular dependence of the absorption probability of arbitrary nuclear states are concentrated in equation (3.32). An even more compact equation is obtained introducing the matrices

$$I^L_{MM'} = V^*_{LM} V_{LM'}, \qquad J^{LL'}_{MM'} = V^*_{LM} V_{L'M'} \tag{3.33}$$

The matrix I^L is called the intensity matrix.[11] The rectangular matrix $J^{LL'}$ is needed to describe the interference term of the mixed transition. For the ($M1/E2$) transition in question the superscripts ($L = 1$, $L' = 2$) are dropped in the following: $J_{MM'} = V^*_{1M} V_{2M'}$. The matrices $(I^L)^{\alpha\beta}$ and $J^{\alpha\beta}$ are determined by the expansion coefficients of the nuclear states belonging to the transition ($\alpha \rightarrow \beta$). These states are calculated with respect to the system S^E, the PAS of the EFG. The transformation properties of I^L and J follow from equation (3.32):

$$I^L(S^\gamma) = D^{L+} I^L(S^E) D^L, \qquad J(S^\gamma) = D^{1+} J(S^E) D^2 \tag{3.34}$$

Therewith equation (3.32) is written in the compact form

$$r^{\alpha\beta}_{pq} = \frac{1}{1 + \delta^2} [(I^1)^{\alpha\beta}_{pq} + p\cdot q\delta^2 (I^2)^{\alpha\beta}_{pq} - \delta(p(J^{\alpha\beta}_{pq})^+ + qJ_{pq})] \tag{3.35}$$

The pure magnetic dipole transition ($\delta = 0$) and electric quadrupole transition ($\delta = \infty$) are included in equation (3.35). The pure electric dipole transition ($E1$) is treated similarly. As for the ($E2$) transition the phases of the off-diagonal elements of the absorber matrix are changed by the factor pq: $r_{pq}(E_1) = pq\cdot I^1_{pq}$ so that the matrices of the $E1$ and $M1$ transitions are related by

$$pq\cdot r_{pq}(E1) = r_{pq}(M1) \tag{3.36a}$$

The amplitudes $a^{\alpha\beta}_p$ and the matrices $r^{\alpha\beta}_{pq} \propto (a^{\alpha\beta}_p)^* a^{\alpha\beta}_q$ have a definite meaning. For an electric dipole transition the amplitudes are proportional to the electric field components of the radiation which would be emitted from the excited state $|I_e e_\beta\rangle$ decaying to the ground state $|I_g g_\alpha\rangle$. So the absorber matrix is proportional to the conjugate complex density matrix constructed in Section 3.1.1 giving $\rho^*_{pq}(E_1) \propto pq I^1_{pq}$. The radiation of a magnetic dipole transition ($M1$) is rotated by $\pi/2$ around the γ direction. This behavior is described by equation (3.36a), which also holds for the density matrices

$$\rho_{pq}(M1) = pq \cdot \rho_{pq}(E1) \tag{3.36b}$$

The following generalization of equation (3.31) of the index of refraction is necessary. It has been assumed that all N scatterers have the same orientation, the same f factor, and the same energy level scheme of the nuclear states. Therefore, powder absorbers and also single crystals with several equivalent sites require a different index of refraction. The last assumption of a single level scheme is preserved in the following equation:

$$n = 1_2 - \frac{\sigma_0}{2k} \sum_j f_j(\mathbf{k}) N_j \sum_{\alpha\beta} r_j^{\alpha\beta} \frac{\Gamma/2}{E - (E_\beta - E_\alpha) + i\Gamma/2} \tag{3.37}$$

There are N_j nuclei of type j, which differ in their orientation and contribute to the cross section by different f factors $f_j(\mathbf{k})$. Interchanging the two sums in equation (3.37) the average of the absorber matrix can be defined as follows:

$$\bar{r}^{\alpha\beta} = \sum_j f_j(\mathbf{k}) N_j r_j^{\alpha\beta} / N\bar{f} \tag{3.38a}$$

where

$$\bar{f}(\mathbf{k}) = \sum_j f_j(\mathbf{k}) N_j / N \tag{3.38b}$$

$N = \Sigma_j N_j$ is the total number of Mössbauer atoms per unit volume and $\bar{f}(\mathbf{k})$ an average f factor. If we add the definition of the effective thickness t of the absorber

$$t(\mathbf{k}) = \sigma_0 \bar{f}(\mathbf{k}) N d \tag{3.38c}$$

the index of refraction times kd is written in the final form:

$$nkd = kd 1_2 - \frac{1}{2} t \sum_{\alpha\beta} \bar{r}^{\alpha\beta} \frac{\Gamma/2}{E - (E_\beta - E_\alpha) + i\Gamma/2} \tag{3.39}$$

The sum $\Sigma_{\alpha\beta}$ which runs over all ground states and excited states is in the following reinterpreted as a sum over different energy levels. The summation over degenerate states is thought of as being carried out. Then, to each absorption line an average absorber matrix $\bar{r}^{\alpha\beta}$ can be attributed. Especially for pure multipole transitions it is useful (see Section 5.2) to define the average of the intensity matrix. Inserting equation (3.36) specialized to a magnetic dipole transition ($\delta = 0$) into equation (3.38) we obtain $\bar{r}^{\alpha\beta}_{pq} = \bar{I}^1_{pq}$, where

$$\bar{I}^1 = \sum_j f_j(\mathbf{k}) N_j D^{1+}(\vartheta_j) I^1(S^E) D^1(\vartheta_j)/N\bar{f} \tag{3.40}$$

The average intensity matrix $\bar{I}^1$ again attains the transformation property of equation (3.35) if $f_j(\mathbf{k}) = \bar{f}$ can be taken out of the sum in equation (3.40). This condition always holds for an isotropic f factor but also for an anisotropic f factor of single crystals and special γ-directions $\mathbf{k}$ (see Section 3.3.3). To obtain this transformation property the matrix $\bar{I}^1$ is calculated with respect to the absorber system S^A by an appropriate transformation $D^1(\vartheta_j)$. The transformation of $I^1(S^E)$ to an arbitrary system S^γ is performed by the successive rotation $D^1(\vartheta_j)D^1(\vartheta)$. The second factor of the product can be taken out of the sum, so that

$$\bar{I}^1(S^\gamma) = D^{1+}(\vartheta)\bar{I}^1(S^A)D^1(\vartheta) \tag{3.41}$$

$\bar{I}^1(S^A)$ is then considered as the intensity matrix of the absorber.

A last generalization of n becomes necessary mainly for electric dipole transitions. The interference between nuclear absorption followed by internal conversion and photoelectric absorption can be taken into account by a modification[31,32] of the resonance term $(\Gamma/2)/\{[E - (E_\beta - E_\alpha)] + i\Gamma/2\}$ of equation (3.31). This term is multiplied by $(1-2i\xi)$, where ξ is a real number characteristic for the atom. The interference is large for $E1$ transitions because the photoeffect has predominantly dipole character. The cross section of the photoelectric absorption is constant in the energy range of the Mössbauer γ ray and contributes to the index of refraction by a small complex number. This absorption has no effect on the background corrected and normalized Mössbauer spectrum and therefore need not be considered.

Now we return to the commutation relation of n and n^+ which lead to the simplified equation for the transmitted intensity of equation (3.23). Inserting equation (3.37) the difference $nn^+ - n^+n = [nn^+]_-$ gives

$$[n,n^+]_- = -\frac{\sigma_0}{2k} N\bar{f} \sum_{\alpha\alpha'\beta\beta'} [\bar{r}^{\alpha\beta},\bar{r}^{\alpha'\beta'}] \times \frac{\Gamma^2/4}{[E-(E_\beta - E_\alpha)+i\Gamma/2][E-(E_{\beta'}-E_\alpha)-i\Gamma/2]} \tag{3.42}$$

There are two ways to fulfil the condition $[nn^+]_- = 0$ at least approximately:

a. The average absorber matrices $\bar{r}^{\alpha\beta}$ commute with each other. In Section 3.2 it will be shown that for a 3/2 → 1/2 transition with two absorption lines (π, σ) the two absorber matrices $\bar{r}^{\pi}$ and $\bar{r}^{\sigma}$ always commute.

b. The energy separation $|(E_\beta - E_\alpha) - (E_{\beta'} - E_{\alpha'})|$ is large compared with $\Gamma/2$. Then $[nn^+]_- \approx 0$ is approximately fulfilled at all energies E.

3.1.3. The Mössbauer Absorption Spectrum

The theoretical Mössbauer spectrum is obtained by the convolution integral of the intensity function $I(d,E)$ of equation (3.8) and the source function $S(E,v)$ of the moving source. We consider the general case of a split source, for example ^{57}Co in iron. The separated Lorentzian lines ($i = 1,2,\ldots$)

$$L_{si}(E,v) = \frac{\Gamma^2/4}{[E - E_{si}(1 + v/c)]^2 + \Gamma^2/4} \tag{3.43}$$

are centered at the energies E_{si}. If their intensities are denoted by I_{si}, which conveniently are normalized to $\Sigma_i I_{si} = 1$, and the density matrix of the radiation by ρ_{si} the source function is given by

$$S(E,v) = \sum_i I_{si}\rho_{si}L_{si}(E,v) \tag{3.44}$$

The source function $I_0\rho_0$ of equation (3.8) now has to be replaced by $S(E,v)$:

$$I(d,E,v) = \mathrm{Tr}[\exp(inkd)S(E,v)\exp(-in^+kd)] \tag{3.45}$$

The background corrected counting rates are then written as

$$N(d,v) = N(d,\infty)\left[1 - f_s\left(1 - \frac{2}{\Gamma\pi}\int_{-\infty}^{\infty} I(d,E,v)\, dE\right)\right] \tag{3.46}$$

f_s is the Debye–Waller factor of the source. The prefactor of the integral equals $\int_{-\infty}^{+\infty} \mathrm{Tr}[S(E,v)]\,dE$. The normalized spectrum $\mathrm{SP}(v) = [N(d,\infty) - N(d,v)]/N(d,v)$ will be considered for two cases which are frequently realized:

i. The single line source is unpolarized; equation (3.46) reduces to

$$\mathrm{SP}(v) = f_s \left\{ 1 - \frac{2}{\Gamma\pi} \int_{-\infty}^{\infty} L_s(E,v) \times \frac{1}{2} \mathrm{Tr}[\exp(inkd)\exp(-in^{+}kd)]\,dE \right\} \tag{3.47}$$

ii. For a thin absorber the integral in equation (3.44) can be evaluated. The spectrum $\mathrm{SP}(v)$ is a superposition of the subspectra $\mathrm{SP}(v)_i^{\alpha\beta}$:

$$\mathrm{SP}(v)_i^{\alpha\beta} = f_s t I_{si} \mathrm{Tr}(\bar{r}^{\alpha\beta}\rho_{si}) \frac{\Gamma^2}{[E_{si}(v/c) - (E_\beta - E_\alpha)]^2 + \Gamma^2} \tag{3.48a}$$

$$\mathrm{SP}(v) = \sum_{\alpha,\beta,i} \mathrm{SP}(v)_i^{\alpha\beta} \tag{3.48b}$$

The area $A_i^{\alpha\beta}$ of the subspectra of the transition (α, β) and source line i is given by

$$A_i^{\alpha\beta} = \int_{-\infty}^{+\infty} \mathrm{SP}_i^{\alpha\beta}(v)\,dv = f_s t \frac{\Gamma\pi}{2} \mathrm{Tr}(\bar{r}^{\alpha\beta}\rho_{si}) \tag{3.49a}$$

In the thin absorber approximation the areas are proportional to the trace of the product of the absorber matrix and the density matrix of the radiation. For an unpolarized source (single line) the area is proportional to the trace of the absorber matrix itself:

$$A^{\alpha\beta} = f_s t \frac{\Gamma\pi}{2} \cdot \frac{1}{2} \mathrm{Tr}(\bar{r}^{\alpha\beta}) \tag{3.49b}$$

The sum over all absorption lines is obtained by equation (3.52) of the next section. The sum $\Sigma_{\alpha\beta}\mathrm{Tr}(\bar{r}^{\alpha\beta}) = 2$ gives

$$\sum_{\alpha\beta} A^{\alpha\beta} = f_s t \cdot \frac{\Gamma\pi}{2} \tag{3.49c}$$

In the thin absorber approximation the total area of the spectrum is proportional to the effective thickness $t = \sigma_0 \bar{f}(\mathbf{k}) N d$.

3.2. *The Intensity Matrix of a Single Transition*

Before the intensity matrices are calculated for several transitions the general properties of the intensity matrices will be summarized. These properties are easily deduced from their definition.

The sum over all ground states or all excited states yields simple relations for the trace of the matrices:

$$\sum_{\alpha} \mathrm{Tr}(I^{L})^{\alpha\beta} = (2L + 1)/(2I_{e} + 1) \tag{3.50a}$$

$$\sum_{\beta} \mathrm{Tr}(I^{L})^{\alpha\beta} = (2L + 1)/(2I_{g} + 1) \tag{3.50b}$$

The total sum over all states gives

$$\sum_{\alpha\beta} (I^{L})^{\alpha\beta}_{MM'} = \delta_{MM'} \tag{3.51a}$$

$$\sum_{\alpha\beta} J^{\alpha\beta}_{MM'} = 0 \tag{3.51b}$$

Equation (3.51a) implies the same relation for the absorber matrices $r^{\alpha\beta}$:

$$\sum_{\alpha\beta} r^{\alpha\beta}_{pq} = \delta_{pq} \tag{3.52}$$

This relation is of course also preserved for the average of the absorber matrix. The symmetry elements of the EFG, the three orthogonal twofold axes, and the time inversion symmetry, are necessarily preserved for the intensity matrices of the transition between the degenerate nuclear levels. The following constraints are inferred by the symmetry relations:

$$\begin{aligned}
&\text{(a) } I^{L} \text{ and } J \text{ are real}\\
&\text{(b) } I_{MM'} = J_{MM'} = 0 \qquad \text{if } M + M' \text{ is odd}\\
&\text{(c) } I_{MM'} = I_{M'M'},\ I_{MM'} = I_{-M-M'}\\
&\quad\ J_{MM'} = -J_{-M-M'}
\end{aligned} \tag{3.53}$$

Exchanging the nuclear spins of the excited and the ground state (I_1, I_2) the intensity matrices are generally related by

$$\begin{aligned}
I_{MM'}(I_1 \rightarrow I_2) &= (-1)^{M+M'}\, I_{-M'-M}(I_2 \rightarrow I_1)\\
J_{MM'}(I_1 \rightarrow I_2) &= (-1)^{M+M'}\, J^{+}_{-M'-M}(I_2 \rightarrow I_1)
\end{aligned} \tag{3.54}$$

With the restrictions equation (3.53) for pure quadrupole interaction the I matrices are unchanged and the interference term changes the sign.

(i) The 3/2 → 1/2 Transition ($M1$). To obtain the intensity matrices first the tensors $V_{LM}^{\alpha\beta}$ for each transition have to be calculated with the eigenstates explicitly given in Section 2.1. The Clebsch–Gordan coefficients in equation (3.30) are replaced by the more common $3j$ symbols of Wigner:

$$V_{LM}^{\alpha\beta} = (2L + 1)^{1/2} \sum_{m_e m_g} e_{\beta m_e}^{*} g_{\alpha m_g} (-1)^{I_g - L + m_e} \begin{pmatrix} I_g & L & I_e \\ m_g & M & -m_e \end{pmatrix} \tag{3.55}$$

The quantum numbers are $I_g = 1/2$, $L = 1$, $I_e = 3/2$. The coefficients $g_{\alpha m_g}$ of the Kramers doublet of the ground states $|I_g, \pm\frac{1}{2}\rangle$ are $g_{\alpha m_g} = \delta_{\alpha m_g}$. The tensor components $V_{1M}^{\pm\frac{1}{2}\beta}$ are written as a row vector:

$$(V_{1M}^{\frac{1}{2}\beta}) = \left(\frac{\sqrt{3}}{2}\, e_{\beta\frac{3}{2}}^{*}, \frac{1}{\sqrt{2}}\, e_{\beta\frac{1}{2}}^{*}, \frac{1}{2}\, e_{\beta-\frac{1}{2}}^{*}\right) \tag{3.56a}$$

$$(V_{1M}^{-\frac{1}{2}\beta}) = \left(\frac{1}{2}\, e_{\beta\frac{1}{2}}^{*}, \frac{1}{\sqrt{2}}\, e_{\beta-\frac{1}{2}}^{*}, \frac{\sqrt{3}}{2}\, e_{\beta-\frac{3}{2}}^{*}\right) \tag{3.56b}$$

The coefficients $e_{\beta m_e}$ are obtained from equation (2.4). The eight vectors ($\alpha = \pm 1/2$, $\beta = 1,2,3,4$) are given by

$$\begin{aligned}
(V_{1M}^{\frac{1}{2}1}) &= \left(\frac{\sqrt{3}}{2}\, e_{+}, 0, \frac{1}{2}\, e_{-}\right), &\quad (V_{1M}^{-\frac{1}{2}1}) &= \left(0, \frac{1}{\sqrt{2}}\, e_{-}, 0\right) \\
(V_{1M}^{\frac{1}{2}2}) &= \left(0, \frac{1}{\sqrt{2}}\, e_{-}, 0\right), &\quad (V_{1M}^{-\frac{1}{2}2}) &= \left(\frac{1}{2}\, e_{-}, 0, \frac{\sqrt{3}}{2}\, e_{+}\right) \\
(V_{1M}^{\frac{1}{2}3}) &= \left(\frac{\sqrt{3}}{2}\, e_{-}, 0, -\frac{1}{2}\, e_{+}\right), &\quad (V_{1M}^{-\frac{1}{2}3}) &= \left(0, -\frac{1}{\sqrt{2}}\, e_{+}, 0\right) \\
(V_{1M}^{\frac{1}{2}4}) &= \left(0, -\frac{1}{\sqrt{2}}\, e_{+}, 0\right), &\quad (V_{1M}^{-\frac{1}{2}4}) &= \left(-\frac{1}{2}\, e_{+}, 0, \frac{\sqrt{3}}{2}\, e_{-}\right)
\end{aligned} \tag{3.57}$$

In each case four intensity matrices $I_{MM'}^{1,\alpha\beta}$ belonging to the transitions at the same energy have to be summed up to one matrix I^{π} and I^{σ} (the superscript $L = 1$ is dropped):

$$I^{\pi} = I^{\frac{1}{2}1} + I^{\frac{1}{2}2} + I^{-\frac{1}{2}1} + I^{-\frac{1}{2}2} \tag{3.58}$$

I^σ is immediately obtained by equation (3.51a):

$$I^\sigma = 1_3 - I^\pi \tag{3.59}$$

Inserting the tensor components $V^{\alpha\beta}_{1M}$ of equation (3.57) the matrix I^π is given by

$$I^\pi = \begin{pmatrix} \frac{3}{4}e_+^2 + \frac{1}{4}e_-^2 & 0 & \frac{\sqrt{3}}{2}e_+e_- \\ 0 & e_-^2 & 0 \\ \frac{\sqrt{3}}{2}e_+e_- & 0 & \frac{3}{4}e_+^2 + \frac{1}{4}e_-^2 \end{pmatrix} \tag{3.60}$$

The symmetry relations [equation (3.53)] are easily verified. The trace of the matrix is independent of η: Tr (I^π) = Tr (I^σ) = 3/2. A sufficient condition that the index of refraction n commutes with its adjoint matrix n^+ was the commuting property of the absorber matrices $\bar{r}^{\alpha\beta}$. The matrices I^π and $I^\sigma = 1_3 - I^\pi$ obviously commute with each other and so do $\bar{r}^\pi$ and $\bar{r}^\sigma = 1_2 - \bar{r}^\pi$.

(ii) The $1/2^+ \rightarrow 3/2^+$ mixed ($M1/E2$) transition (^{197}Au). The mixed transition requires the calculation of three matrices I^1, I^2, and J. The I^1 matrix is identical with that obtained for the 3/2 → 1/2 transition. The 5×5 matrix I^2 and the 3×5 matrix J are as follows:

$$(I^2)^\pi = \begin{pmatrix} e_+^2 & 0 & \frac{1}{\sqrt{2}}e_+e_- & 0 & 0 \\ 0 & \frac{1}{4}e_+^2 + \frac{3}{4}e_-^2 & 0 & \frac{\sqrt{3}}{2}e_+e_- & 0 \\ \frac{1}{\sqrt{2}}e_+e_- & 0 & e_-^2 & 0 & \frac{1}{\sqrt{2}}e_+e_- \\ 0 & \frac{\sqrt{3}}{2}e_+e_- & 0 & \frac{1}{4}e_+^2 + \frac{3}{4}e_-^2 & 0 \\ 0 & 0 & \frac{1}{\sqrt{2}}e_+e_- & 0 & e_+^2 \end{pmatrix} \tag{3.61a}$$

$$J^{\pi} = \begin{pmatrix} 0 & +\frac{\sqrt{3}}{4}(e_+^2 - e_-^2) & 0 & +\frac{1}{2}e_+e_- & 0 \\ -\frac{1}{\sqrt{2}}e_+e_- & 0 & 0 & 0 & +\frac{1}{\sqrt{2}}e_+e_- \\ 0 & -\frac{1}{2}e_+e_- & 0 & -\frac{\sqrt{3}}{4}(e_+^2 - e_-^2) & 0 \end{pmatrix} \tag{3.61b}$$

The corresponding matrices for the σ transition are again obtained by equation (2.52)

$$(I^2)^{\sigma} = 1_5 - (I^2)^{\pi}, \qquad J^{\sigma} = -J^{\pi} \tag{3.62}$$

The trace of I^2 is 5/2.

(iii) The $2^+ \to 0^+$ transition ($E2$). The $V_{2M}^{\alpha\beta}$ tensors components are just the expansion coefficients of the excited states:

$$V_{2M}^{0\beta} = e_{\beta M}^{*} \tag{3.63}$$

The intensity matrices of the six transitions of the eigenstates of equation (2.7) and (2.10) are given by (the superscripts $L = 2$ and $\alpha = 0$ are dropped)

$$I^{1,5} = \begin{pmatrix} \frac{1}{2}e_{\mp}^2 & 0 & \mp\frac{1}{\sqrt{2}}e_+e_- & 0 & \frac{1}{2}e_{\mp}^2 \\ 0 & 0 & 0 & 0 & 0 \\ \mp\frac{1}{\sqrt{2}}e_+e_- & 0 & e_{\pm}^2 & 0 & \mp\frac{1}{\sqrt{2}}e_+e_- \\ 0 & 0 & 0 & 0 & 0 \\ \frac{1}{2}e_{\mp}^2 & 0 & \mp\frac{1}{\sqrt{2}}e_+e_- & 0 & \frac{1}{2}e_{\mp}^2 \end{pmatrix} \tag{3.64a}$$

$$I^{2,3} = \begin{pmatrix} 0 & 0 & 0 & 0 & 0 \\ 0 & 1/2 & 0 & \mp 1/2 & 0 \\ 0 & 0 & 0 & 0 & 0 \\ 0 & \mp 1/2 & 0 & 1/2 & 0 \\ 0 & 0 & 0 & 0 & 0 \end{pmatrix} \tag{3.64b}$$

$$I^{4} = \begin{pmatrix} 1/2 & 0 & 0 & 0 & -1/2 \\ 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 \\ -1/2 & 0 & 0 & 0 & 1/2 \end{pmatrix} \tag{3.64c}$$

The trace of all matrices is $\mathrm{Tr}(I^k) = 1$, $k = 1, \ldots, 6$. The absorption probabilities of the transitions 2, 3, and 4 are independent of the asymmetry parameter η.

(iv) The $5/2^+ \rightarrow 7/2^+$ transition ($M1$). The eigenstates and eigenvalues result from a 4×4 Hermitian matrix, which has not been solved for the general case in Section 2.1. Therefore, only the simple case of an axial field gradient is considered where the eigenstates are $|Im\rangle$. The 12 transitions from the four ground states to the three excited states are partially forbidden. The eight nonvanishing matrices are all diagonal. Each of them is again a sum of four matrices, for example

$$I(7,5) = I^{\frac{7}{2}\frac{5}{2}} + I^{\frac{7}{2}-\frac{5}{2}} + I^{-\frac{7}{2}\frac{5}{2}} + I^{-\frac{7}{2}-\frac{5}{2}} \tag{3.65}$$

The diagonal elements of the diagonal matrices are written as row vectors; then we have

$$\begin{aligned} &I(7,5) = (\tfrac{3}{8}, 0, \tfrac{3}{8}) \\ &I(5,5) = (0, \tfrac{3}{14}, 0), \quad I(5,3) = (\tfrac{15}{56}, 0, \tfrac{15}{56}) \\ &I(3,5) = (\tfrac{1}{56}, 0, \tfrac{1}{56}), \quad I(3,3) = (0, \tfrac{5}{14}, 0), \quad I(3,1) = (\tfrac{5}{28}, 0, \tfrac{5}{28}) \\ &I(1,3) = (\tfrac{3}{56}, 0, \tfrac{3}{56}), \quad I(1,1) = (\tfrac{3}{28}, \tfrac{3}{7}, \tfrac{3}{28}) \end{aligned} \tag{3.66}$$

The pure $\Delta m = 0$ transitions $I(3,3)$ and $I(5,5)$ consist of one nonzero element I_{00}, so that they are proportional to each other. Since the intensity matrix determines the intensities of the absorption lines, the ratio of the intensities of these two lines is independent of the γ-direction. The same holds for the five $\Delta m = \pm 1$ transitions. Only the $I(1,1)$ transition is of mixed type and not proportional to any other transition.

It is easy to construct sum rules for the intensities. For example the sum $2I(3,1) + I(3,3)$ is proportional to the unit matrix and therefore the intensity is independent of the γ-direction.

3.3. Texture

The calculation of the absorber matrices $\bar{r}_{pq}$ in the case of an isotropic Debye–Waller factor $f(\mathbf{k}) = \bar{f}$, but for a textured absorber, is the topic of this section. The single crystal is considered as a powder absorber with a δ-like distribution function $T(\alpha, \beta, \gamma)$. The concept of the texture opens the possibility of avoiding the transformation of the intensity matrices to the new γ-direction and instead of that transforming the distribution function $T(\alpha, \beta, \gamma)$. For this purpose the transformation properties of the texture components are required. Examples of texture components of single crystals will be given. The last point of this section describes the construction of a texture-free absorption spectrum (random powder absorber) by the superposition of spectra of a textured absorber measured at definite directions. This possibility is applicable for thin absorbers.

3.3.1. The Absorber Matrices

The absorber matrix r_{pq} will be averaged by use of the texture function. The probability of finding a system S^E at the orientation $\boldsymbol{\beta}$ is given by equation (3.1):

$$T(\boldsymbol{\beta}) = \sum_{Lm'm} t^L_{m'm} D^L_{m'm}(\boldsymbol{\beta})$$

The angles α, γ of $\boldsymbol{\beta} = (\alpha, \beta, \gamma)$ are the rotations around the z axes of the systems S^γ and S^E, respectively (see Figure 3). An axial symmetric texture does not depend on α so that m', which is multiplied to α in $\exp(im'\alpha)$, must vanish. If the hyperfine Hamiltonian is axial the angle γ does not appear, so that $m = 0$. The normalization condition ($\Omega = 8\pi^2$)

$$\frac{1}{\Omega} \int T(\boldsymbol{\beta})\, d\boldsymbol{\beta} = 1, \qquad d\boldsymbol{\beta} = \sin\beta\, d\beta\, d\alpha\, d\gamma \tag{3.67}$$

requires that $t^0_{00} = 1$. The random powder is defined by $t^L_{mm'} = 0$, $L \geq 1$.

The rotation $\boldsymbol{\beta}$ is opposite to the rotation $\boldsymbol{\vartheta}$ in equation (3.39), so that the intensity matrices are transformed to the system S^γ by the inverse rotation matrix $D^{L+}(\boldsymbol{\beta})$:

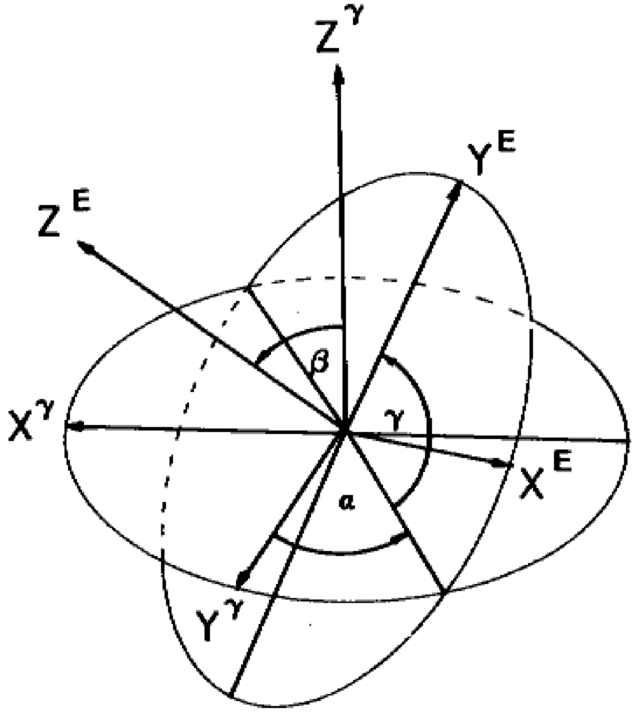

FIGURE 3. The coordinate system S^γ is fixed at the laboratory. The Euler angles $\boldsymbol{\beta} = (\alpha, \beta, \gamma)$ rotate S^γ to the principal axis system S^E of the EFG.

$$I^L(S^\gamma) = D^L(\boldsymbol{\beta}) I^L(S^E) D^{L+}(\boldsymbol{\beta}) \tag{3.68a}$$

$$J(S^\gamma) = D^1(\boldsymbol{\beta}) J(S^E) D^{2+}(\boldsymbol{\beta}) \tag{3.68b}$$

The average of r_{pq}

$$\bar{r}_{pq} = \frac{1}{\Omega} \int T(\boldsymbol{\beta}) r_{pq}(\boldsymbol{\beta})\, d\boldsymbol{\beta} \tag{3.69}$$

leads to two terms, the average of the Hermitian matrix I^L of a pure L-pole transition and of the rectangular matrix J describing the interference term of the $(M1/E2)$ transition:

$$\bar{I}^L_{pq} = \frac{1}{\Omega} \int T(\boldsymbol{\beta}) [D^L(\boldsymbol{\beta}) I^L D^{L+}(\boldsymbol{\beta})]_{pq}\, d\boldsymbol{\beta} \tag{3.70}$$

$$\Delta_{pq} = \frac{1}{\Omega} \int T(\boldsymbol{\beta}) [p(D^1(\boldsymbol{\beta}) J D^{2+}(\boldsymbol{\beta}))^+_{pq} + q(D^1(\boldsymbol{\beta}) J D^{2+}(\boldsymbol{\beta}))_{pq}]\, d\boldsymbol{\beta} \tag{3.71}$$

The integrals are easily evaluated by the formula for the integral of the product of three rotation matrices[28]:

$$\frac{1}{\Omega}\int D^{L_1}_{m'_1m_1}(\boldsymbol{\beta})\, D^{L_2}_{m'_2m_2}(\boldsymbol{\beta})\, D^{L_3}_{m'_3m_3}(\boldsymbol{\beta})\, d\boldsymbol{\beta} = \begin{pmatrix} L_1 & L_2 & L_3 \\ m'_1 & m'_2 & m'_3 \end{pmatrix} \begin{pmatrix} L_1 & L_2 & L_3 \\ m_1 & m_2 & m_3 \end{pmatrix} \tag{3.72}$$

The average of the intensity matrix I^L is given by

$$\bar{I}^L_{pq} = \sum_{L',m',m,j,k} (-)^{q+k} \begin{pmatrix} L' & L & L \\ m' & p & -q \end{pmatrix} \begin{pmatrix} L' & L & L \\ m & j & -k \end{pmatrix} I^L_{jk} t^{L'}_{m'm} \tag{3.73}$$

It should be noted that the average matrix for a random powder absorber ($t^L_{mm'} = 0, L \geq 1$) is proportional to the unit matrix:

$$\bar{I}^L_{pq}(r \text{ powder}) = \frac{1}{2L+1}\operatorname{Tr}(I^L)\delta_{pq} \tag{3.74}$$

The components of the Δ matrix are written separately:

$$\begin{aligned}
\Delta_{\pm1\pm1} &= \pm \sum_{L',m,M_1,M_2} \begin{pmatrix} L' & 2 & 1 \\ 0 & \mp1 & \pm1 \end{pmatrix} \begin{pmatrix} L' & 2 & 1 \\ m & -M_2 & M_1 \end{pmatrix} \\
&\quad \times [(-)^{M_2} J_{M_1M_2} t^{L'}_{0m} + (-)^{M_1} J^{+}_{M_1M_2} t^{L'}_{0-m}] \\
\Delta_{1-1} &= \sum_{L',m,M_1,M_2} \begin{pmatrix} L' & 2 & 1 \\ -2 & 1 & 1 \end{pmatrix} \begin{pmatrix} L' & 2 & 1 \\ m & -M_2 & M_1 \end{pmatrix} \\
&\quad \times [(-)^{M_2} J_{M_1M_2} t^{L'}_{-2m} + (-)^{M_1} J^{+}_{M_2M_1} t^{L'}_{-2-m}] \\
\Delta_{-11} &= \Delta^{*}_{1-1}
\end{aligned} \tag{3.75}$$

The last equation is given by the Hermiticity of the absorber matrix. The above equations are quite general; they can also be used for a magnetic term in the hyperfine interaction. Using the symmetry properties of the intensity matrices equation (3.53) inferred by the pure quadrupole interaction, the equations simplify considerably. The explicit formulas of the matrix elements are given below. As a result of the time inversion symmetry the diagonal elements are identical ($\bar{r}_{11} = \bar{r}_{-1-1}$). The superscripts α, β denoting the transitions are dropped in the following:

$$\bar{r}_{11} = A_{00} + A_{20} + A_{40} \tag{3.76a}$$

$$\bar{r}_{\pm1\mp1} = B_{2\mp2} + B_{4\mp2} \tag{3.76b}$$

The $A_{Lm'}$, $B_{Lm'}$ are linear combinations of the texture components $t^L_{m'm}$:

$$A_{Lm'} = \sum_m a^L_m t^L_{m'm} (2L + 1)^{-1} \tag{3.77a}$$

$$B_{Lm'} = \sum_m b^L_m t^L_{m'm} (2L + 1)^{-1} \tag{3.77b}$$

The coefficients a^L_m, b^L_m vanish for odd values of m and L. They are given by

$$\begin{aligned}
a^0_0 &= \frac{1}{1 + \delta^2}\left[\frac{1}{3}\mathrm{Tr}(I^1) + \frac{1}{5}\,\delta^2\mathrm{Tr}(I^2)\right] \\
a^2_0 &= \frac{1}{1 + \delta^2} \cdot \frac{1}{21}\,[7(I^1_{11} - I^1_{00}) + 42\delta J_{11} \\
&\quad -3\delta^2(2I^2_{22} - I^2_{11} - I^2_{00})] \\
a^2_{\pm 2} &= \frac{1}{1 + \delta^2}\,\frac{1}{21\sqrt{6}}\,[21I^1_{1-1} - 42\delta(\sqrt{2}\,J_{02} - J_{1-1}) \\
&\quad -3\delta^2(2\sqrt{6}\,I^2_{20} + 3I^2_{1-1})] \\
a^4_0 &= -\frac{\delta^2}{1 + \delta^2}\,\frac{2}{7}\,\frac{2}{5}\,(I^2_{22} - 4I^2_{11} + 3I^2_{00}) \\
a^4_{\pm 2} &= -\frac{2}{1 + \delta^2}\,\frac{2}{7}\,\sqrt{\tfrac{2}{5}}\,(\sqrt{6}\,I^2_{20} - 4I^2_{1-1}) \\
a^4_{\pm 4} &= -\frac{2}{1 + \delta^2}\,\frac{2}{7}\,\sqrt{\tfrac{2}{5}}\,\sqrt{7} \cdot I^2_{2-2} \\
b^2_0 &= \frac{1}{1 + \delta^2}\,\frac{\sqrt{6}}{21}\,[7(I^1_{11} - I^1_{00}) + 14\delta J_{11} \\
&\quad + 3\delta^2(2I^2_{22} - I^2_{11} - I^2_{00})] \\
b^2_{\pm 2} &= \frac{1}{1 + \delta^2}\,\frac{1}{21}\,[21I^1_{1-1} - 14\delta(\sqrt{2}\,J_{02} - J_{1-1}) \\
&\quad + 3\delta^2(2\sqrt{6}\,I^2_{20} + 3I^2_{1-1})] \\
b^4_m &= \frac{\sqrt{5}}{4}\,a^4_m
\end{aligned} \tag{3.78}$$

At first sight these formulas by no means give an impression of simplicity and clearness. But the reader is reminded of the complicated problem which has been solved. Equation (3.76) describes the intensity of a mixed transition of an arbitrarily textured absorber which is readily avoided in the standard Mössbauer experiment. Some special cases will be of interest; these are the axial texture which is easily produced during the preparation of powder absorbers[33] and the single crystal with several equivalent sites per unit cell.

It will be shown that the quantities A_{Lm}, B_{Lm} transform under rotations as irreducible tensors of rank L

$$\begin{aligned} A_{Lm}(\vartheta) &= \sum_n A_{Ln}\, D^L_{nm}(\vartheta) \\ B_{Lm}(\vartheta) &= \sum_n B_{Ln}\, D^L_{nm}(\vartheta) \end{aligned} \tag{3.79}$$

so that the absorber matrix elements are expanded in a series of rotation matrices:

$$\begin{aligned} \bar{r}_{pq}(\vartheta) &= \sum_{L,n} A_{Ln}\, D^L_{n,q-p}(\vartheta), \qquad p-q = 0 \\ \bar{r}_{pq}(\vartheta) &= \sum_{L,n} B_{Ln}\, D^L_{n,q-p}(\vartheta), \qquad p-q = \pm 2 \end{aligned} \tag{3.80}$$

This expansion series has been announced in the introduction of this chapter.

The angle ϑ has the same meaning as in equation (3.41). It rotates S^A to S^γ. The tensor components can be completely determined from the Mössbauer spectra measured at different directions and therefore they define the cut between the experimental results and their interpretation.

For the three transitions represented by ^{57}Fe, ^{197}Au, and ^{156}Gd where the intensity matrices have been calculated in Section 3.2, the tensors A_{Lm} will be explicitly evaluated.

(i) The $3/2^- \rightarrow 1/2^-$ transition ($M1$). The average of the absorber matrix is, according to equation (3.76),

$$\bar{r}^{\alpha\beta}_{11} = A^{\alpha\beta}_{00} + A^{\alpha\beta}_{20}, \qquad \bar{r}^{\alpha\beta}_{1-1} = B^{\alpha\beta}_{2-2}, \qquad (\alpha\beta) = \pi \text{ and } \sigma \tag{3.81}$$

With the intensity matrix of equation (3.59) the A_{Lm}, B_{Lm} are calculated to be

$$\begin{aligned} A^{\pi,\sigma}_{00} &= 1/2 \\ A^{\pi,\sigma}_{2m} &= \pm \frac{1}{20\sqrt{6}} [\sqrt{6}\, t^2_{m0} + \eta(t^2_{m2} + t^2_{m-2})](1 + \eta^2/3)^{-\frac{1}{2}} \\ \mathrm{B}^{\pi,\sigma}_{2m} &= \sqrt{6}\, A^{\pi,\sigma}_{2m} \end{aligned} \tag{3.82}$$

The measurement at five independent directions yield the A_{2m} components uniquely. The off-diagonal matrix elements $\bar{r}^{\pi,\sigma}_{1-1}$ contain the same information as the diagonal elements. The measurement of $\bar{r}^{\pi,\sigma}_{1-1}$ with polarized radiation can only be used to reduce the number of measurements at different directions (see Section 5).

(ii) The 1/2$^+$ → 3/2$^+$ mixed ($M1/E2$) transition. The tensors $A^{\pi,\sigma}_{2m}$, $B^{\pi,\sigma}_{2m}$ are proportional to $A^{\pi,\sigma}_{2m}$ of equation (3.82):

$$A^{\varkappa}_{20}(\delta) = (1 - \delta^2 + 2\sqrt{3}\,\delta)/(1 + \delta^2)\cdot A^{\varkappa}_{20}(\delta = 0) \tag{3.83a}$$

$$A^{\varkappa}_{2\pm 2}(\delta) = [1 + \delta^2 - (2/\sqrt{3})\,\delta]/(1 + \delta^2)\cdot\sqrt{6}\,A^{\varkappa}_{2\pm 2}(\delta = 0) \tag{3.83b}$$

$$\varkappa = \pi,\ \sigma$$

so that we obtain for the matrix elements of the absorber matrix

$$\bar{r}^{\varkappa}_{11} = \frac{1}{2} + \left(1 + 2\sqrt{3}\,\frac{\delta}{1 - \delta^2}\right)\frac{1 - \delta^2}{1 + \delta^2}\,A^{\varkappa}_{20}(\delta = 0) \tag{3.84a}$$

$$\bar{r}^{\varkappa}_{1-1} = \left(1 - \frac{2}{\sqrt{3}}\,\frac{\delta}{1 + \delta^2}\right)\sqrt{6}\,A^{\varkappa}_{2-2}(\delta = 0) \tag{3.84b}$$

$$\varkappa = \pi,\ \sigma$$

The absorption of unpolarized radiation becomes isotropic if the coefficient of $A^{\varkappa}_{20}$ vanishes. The condition is[34] $\delta = \sqrt{3} \pm 2$. The value[34] $\delta = -0.354$ of ^{197}Au is very near $\delta = \sqrt{3} - 2$. Therefore, the angular part of $\bar{r}^{\varkappa}_{11}$ and the off-diagonal element $r^{\varkappa}_{1-1}$ are appreciably decreased:

$$\bar{r}^{\varkappa}_{11}(^{197}\mathrm{Au}) = \tfrac{1}{2} - 0.305\,A^{\varkappa}_{20}(\delta = 0) \tag{3.85a}$$

$$\bar{r}^{\varkappa}_{1-1}(^{197}\mathrm{Au}) = 0.717\,B^{\varkappa}_{2-2}(\delta = 0), \qquad \varkappa = \pi,\ \sigma \tag{3.85b}$$

(iii) The 2$^+$ → 0$^+$ transition ($E2$). There are five absorber matrices ($\alpha = 1$, $\beta = 1, 2, \ldots, 5$)

$$\bar{r}^{\varkappa}_{11} = A^{\varkappa}_{00} + A^{\varkappa}_{20} + A^{\varkappa}_{40} \tag{3.86a}$$

$$\bar{r}^{\varkappa}_{1-1} = B^{\varkappa}_{2-2} + B^{\varkappa}_{4-2}, \qquad \varkappa = (\alpha,\ \beta) \tag{3.86b}$$

The tensors $A^{\varkappa}_{L,m'}$, $B^{\varkappa}_{Lm}$ are constructed using the intensity matrices of equation (3.64):

$$A^{\varkappa}_{00} = 1/5, \qquad \varkappa = (\alpha,\ \beta) = 1, 2, 3, 4, 5 \tag{3.87a}$$

$$A^{1,5}_{2m} = -\frac{1}{35}\,(e^2_{\mp} - e^2_{\pm})t^2_{m0} \pm \frac{\sqrt{2}}{35}\,e_{+}e_{-}(t^2_{m2} + t^2_{m-2})$$

$$A^{2,3}_{2m} = \frac{1}{70}\, t^2_{m0} \pm \frac{\sqrt{6}}{4\cdot 35}\,(t^2_{m2} + t^2_{m-2}) \tag{3.87b}$$

$$A^{4}_{2m} = -\frac{1}{35}\, t^2_{m0}$$

$$B^{\varkappa}_{2m} = -\sqrt{6}\, A^{\varkappa}_{2m}$$

$$A^{1,5}_{4m} = -\frac{2}{7\cdot 9}\left[\frac{1}{5}\,(e^2_{\mp} + 6e^2_{\pm})t^4_{m0} \mp \frac{1}{\sqrt{10}}\, 2\cdot\sqrt{3}\; e_{+}e_{-}(t^4_{m2} + t^4_{m-2}) \right.$$
$$\left. + \sqrt{\tfrac{7}{10}}\; e^2_{\mp}(t^4_{m4} + t^4_{m-4})\right]$$

$$A^{2,3}_{4m} = -\frac{2}{7\cdot 9}\left[-\frac{4}{5}\, t^4_{m0} \pm \sqrt{\tfrac{2}{5}}\,(t^4_{m2} + t^4_{m-2})\right] \tag{3.87c}$$

$$A^{4}_{4m} = -\frac{2}{7\cdot 9}\left[\frac{1}{5}\, t^4_{m0} - \sqrt{\tfrac{7}{10}}\,(t^4_{m4} + t^4_{m-4})\right]$$

$$B^{\varkappa}_{4m} = \frac{\sqrt{5}}{4}\, A^{\varkappa}_{4m}$$

The sum $\Sigma_{\varkappa} A^{\varkappa}_{Lm}$ vanishes for $L > 0$ as a result of the sum rule of the $\bar{r}$ matrices [according to equation (3.52)]. The $A^{\varkappa}_{00}$ values represent the intensities of the absorption lines of a random powder sample.

3.3.2. *Transformation Properties of Texture Components*

In Figure 4 two situations are shown. First, the absorber has a position in space where the system S^{γ} coincides with the system S^{A} of the absorber (a). If the absorber is rotated by the angle $-\boldsymbol{\vartheta}$ we have the second situation (b). This definition of $\boldsymbol{\vartheta}$ is in accordance with equations (3.29) and (3.68). Per definition the texture functions in S^{A} and S^{γ} of Figure 4 are given by

$$T(\boldsymbol{\beta}) = \sum_{L,m',m} t^L_{m'm}(A)\; D^L_A(\boldsymbol{\beta})_{m'm} \tag{3.88a}$$

$$T(\boldsymbol{\beta}') = \sum_{L,m',m} t^L_{m'm}(\gamma)\; D^L_{\gamma}(\boldsymbol{\beta}')_{m'm} \tag{3.88b}$$

The labels A and γ at the rotation matrix $D^L_{A,\gamma}$ and at the texture components $t^L_{m'm}(A,\gamma)$ denote the systems S^A and S^{γ}, respectively, which the quantities belong to. From Figure 4 the relationship between the rotation matrices can be derived:

$$D^L_A(\boldsymbol{\beta}')D^L_A(\boldsymbol{\vartheta}) = D^L_A(\boldsymbol{\beta}) \tag{3.89a}$$

or by changing the coordinate system (the rotation axes)

$$D^L_A(\boldsymbol{\vartheta})\cdot D^L_{\gamma}(\boldsymbol{\beta}') = D^L_A(\boldsymbol{\beta}) \tag{3.89b}$$

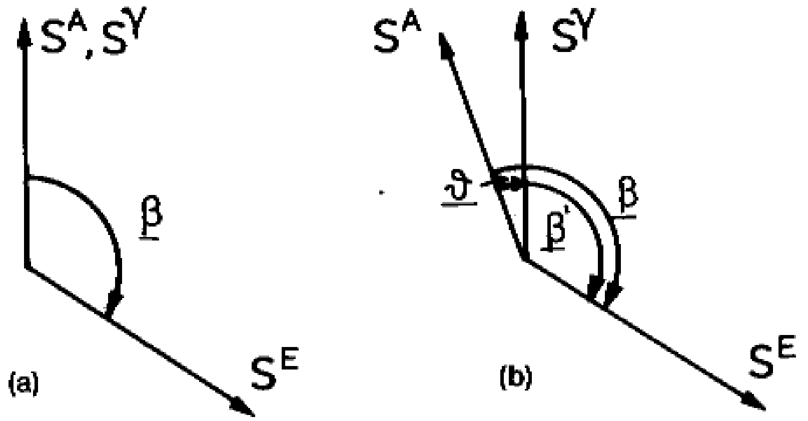

FIGURE 4. In (a) the system of the absorber S^A and the system S^γ coincide. The z axis of S^γ is the γ direction. S^E is one representative PAS of the EFG in the absorber. In (b) the absorber is rotated by the angle $-\vartheta$. The texture components of the absorber are transformed to the system S^γ by the rotation matrices $D^L(\vartheta)$ (see text).

Insertion of $D^L_A(\boldsymbol{\beta})$ in equation (3.88a) and the comparison of the coefficients with equation (3.88b) yield the transformation properties of the texture components

$$t^L_{m'm}(\gamma) = \sum_n D^L_A(\boldsymbol{\vartheta})_{nm'} t^L_{nm}(A) \tag{3.90a}$$

The label A is dropped and instead of that the $\boldsymbol{\vartheta}$ dependence of $t^L_{m'm}$ is emphasized:

$$t^L_{m'm}(\boldsymbol{\vartheta}) = \sum_n D^L(\boldsymbol{\vartheta})_{nm'} t^L_{nm} \tag{3.90b}$$

The tensor property of the quantities A_{Lm} is easily proved by inserting this transformation equation of the texture components into equation (3.77).

3.3.3. *The Texture of a Single Crystal*

In the beginning we consider a single crystal which has only one lattice site of the Mössbauer atom per unit cell. The coordinate system S^E fixed to the nucleus will be parallel to the system S^C of the single crystal. Then the probability of finding a system S^E at $\boldsymbol{\beta} = 0$ is one and otherwise zero:

$$T(\boldsymbol{\beta})\, d\boldsymbol{\beta} = \delta_{0\boldsymbol{\beta}} \tag{3.91}$$

The integral

$$\frac{1}{\Omega}\int T(\boldsymbol{\beta})D^L(\boldsymbol{\beta})^+_{nn'}\,d\boldsymbol{\beta} = \frac{1}{\Omega}\int \sum_{L',m',m} t^{L'}_{m'm}D^{L'}_{m'm}(\boldsymbol{\beta})D^L(\boldsymbol{\beta})^+_{nn'}\,d\boldsymbol{\beta} \quad (3.92)$$

gives, inserting equation (3.91),

$$D^L_{nn'}(0)^+ = \frac{1}{2L+1}\,t^L_{n'n} \quad (3.93a)$$

so that the texture component is proportional to the Kronecker symbol:

$$t^L_{m'm} = (2L+1)\delta_{m'm} \quad (3.93b)$$

The texture components of a single crystal with n equivalent sites are constructed as follows. Let the orientation of the kth site be given by the Euler angles $\boldsymbol{\theta}_k$, which rotate the system S^C to the system S^E of site k. The texture components defined in the system S^E [equation (3.93)] are transformed to S^C by the equation (3.90):

$$t^L_{m'm}(S^C,k) = \sum_n D^L(\boldsymbol{\theta}_k)^+_{nm'}(2L+1)\delta_{nm} \quad (3.94)$$

The rotation matrix is replaced by its inverse, because the rotation angles $\boldsymbol{\theta}_k$ rotate the old system to the new one contrary to the definition of Figure 4. The texture components of the N sites have to be averaged to obtain the components of the crystal:

$$t^L_{m'm}(S^C) = \frac{1}{N}\sum_{k=1}^{N} D^L(\boldsymbol{\theta}_k)^+_{mm'}(2L+1) \quad (3.95)$$

Three examples will be given.

(i) Monoclinic crystal with two equivalent sites. The twofold axis is chosen to be the z axis of the crystal system S^C. The two equivalent sites have the general orientations $\boldsymbol{\theta}_1 = (\phi, \theta, \Psi)$ and $\boldsymbol{\theta}_2 = (\phi + \pi, \theta, \Psi)$, respectively. The texture components are then given by

$$t^L_{m'm} = \tfrac{1}{2}(2L+1)D^L(\boldsymbol{\theta}_1)^+_{mm'}\,[1 + \exp(-im'\pi)] \quad (3.96)$$

so that

$$t^L_{m'm} = 0 \qquad \text{if } m' \text{ is odd}$$

and

$$t^L_{m'm} = (2L+1)D^L(\boldsymbol{\theta}_1)^+_{mm'} \qquad \text{if } m' \text{ is even}$$

This property is of course transferred to the tensor components $A_{Lm'}$ by equation (3.77).

(ii) Axial crystal (C_3 axis). The threefold axis is the z axis. The orientations of the three equivalent sites are $\boldsymbol{\theta}_k = (\phi + (\pi/3)(k - 1), \theta, \Psi)$, $k = 1,2,3$, so that

$$t^L_{m'm}(S^C) = (2L + 1)\,\tfrac{1}{3}\,(1 + 2\cos(2\pi/3m'))D^L(\boldsymbol{\theta}_1)^+_{mm'} \qquad (3.97)$$

(iii) Orthorhombic crystal (D_2 symmetry). The rotation matrices $D^L(\boldsymbol{\theta}_k)$ are written as a succession of two rotations $D^L(C_{2i})D^L(\boldsymbol{\theta}_1)$. They are the three twofold rotations of the D_2 point group. With the relations

$$\begin{aligned} D^L_{mm'}(C_{2x}) &= (-)^L\,\delta_{m-m'} \\ D^L_{mm'}(C_{2y}) &= (-)^{L-m}\,\delta_{m-m'} \\ D^L_{mm'}(C_{2z}) &= (-)^m\,\delta_{mm'} \end{aligned} \qquad (3.98)$$

the texture components are easily obtained to be

$$t^L_{m'm} = \tfrac{1}{4}(2L + 1)[1 + (-)^{m'}][D^L(\boldsymbol{\theta}_1)^+_{mm'} + D^L(\boldsymbol{\theta}_1)^+_{m-m'}] \qquad (3.99)$$

The $A_{Lm'}$ tensor components vanish for odd m' and $A_{Lm'} = A_{L-m'}$. The expressions are conveniently formulated in terms of the invariants of finite point groups, the identical representations A_{1g}. There are three irreducible A_{1g} representations of the second-rank tensor A_{2m} in C_2 symmetry. These are the A_{20}, A_{22}, A_{2-2} components. In C_3 symmetry there is only one component A_{20} in accordance with equation (3.97). In cubic symmetry there is no invariant for $L \leq 4$, so that in the case of a dipole transition (A_{2m}) the intensities are the same as for a random powder.

The pure quadrupole interaction imposes further restrictions on the texture components. The system S^E cannot be uniquely fixed to the EFG tensor. There are four equivalent PAS related by the three twofold axes parallel to the principal axes. The texture components have to be invariant with respect to these transformations. These transformations are already applied in equation (3.99) so that we obtain the texture components of a single site setting $\boldsymbol{\theta}_1 = 0$:

$$t^L_{m'm} = \tfrac{1}{4}(2L + 1)[1 + (-)^{m'}][\delta_{mm'} + (-)^L\delta_{m-m'}] \qquad (3.100)$$

The equation (3.93) has to be replaced by this equation and in turn the rotation matrix $D^{L+}_{mm'}$ by the linear combination $\tfrac{1}{2}[D^{L+}_{mm'} + (-)^L D^{L+}_{-mm'}]$, the index m being even. The texture components of a distribution of EFG tensors therefore have the symmetry property[16]