The Coherent Forward Scattering Amplitude in Transmission and Grazing Incidence Mössbauer Spectroscopy

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(Dated: Submitted to Physical Review B on July 25, 1995)

The theory of both transmission and grazing incidence Mössbauer spectroscopy is re-analyzed. Starting with the nuclear susceptibility tensor a common concise first order perturbation formulation is given by introducing the forward scattering amplitude into an anisotropic optical scheme. Formulae of Blume and Kistner as well as those of Andreeva are re-derived for the forward scattering and grazing incidence geometries, respectively. Limitations of several previously intuitively introduced approximations are pointed out. The grazing incidence integral propagation matrices are written in a form built up from $2 \times 2$ matrix exponentials which is particularly suitable for numerical calculations and practical fitting of both energy domain (conventional source experiment) and time domain (synchrotron radiation experiment) Mössbauer spectra.

PACS numbers: PACS: 42.25-p,68.35.-p,76.80.+y,78.66.-w

I. INTRODUCTION

A great majority of Mössbauer experiments is performed on polycrystalline samples without applying an external magnetic field. In such cases, the polarization of the $\gamma$-rays plays no role, the Mössbauer spectrum can be described in terms of resonant and non-resonant absorption and the resonant absorption cross-section can be calculated from the parameters of the hyperfine interaction. This naive approach fails if the Mössbauer experiment is performed on a single crystal or a textured sample and/or in an external magnetic field. The resonant cross-section in these latter cases depends on the polarization and the full polarization-dependent scattering problem has to be treated. The numerical difficulties of the scattering approach stem from the great number of randomly distributed scattering centres. These difficulties can be circumvented if, akin to classical optics, a continuum model rather than a microscopic scattering theory can be used. It is by no means trivial, however, that such an optical approach for $\gamma$-rays in condensed matter is feasible since the mean distance of scattering centres is usually greater than the wavelength of the scattered radiation. It has been shown, however, by Lax that, at least for scalar waves, a close to unity index of refraction can be defined and simply related to the coherent forward scattering length $f$, provided that the momentum of the scatterers is small compared to that of the incident wave. Since Lax’s paper the refraction index approach has been used extensively in neutron and x-ray optics. The heuristic generalization of this approach to polarized waves and for an anisotropic medium, although claimed to be trivial by Lax is by no means straightforward and needs further elucidation.

In the forward scattering geometry the polarization dependence of the Mössbauer absorption of $\gamma$-radiation was theoretically studied by Blume and Kistner. Instead of using a $3 \times 3$ index of refraction matrix, they accepted Lax’s intuitive suggestion and used a complex $2 \times 2$ index of refraction matrix $n$, corresponding to the two possible independent states of polarization of the radiation. $n$ was then related to the coherent forward scattering amplitude.

Beside the conventional forward scattering case, grazing incidence Mössbauer spectroscopy (GIMS) has gained considerable recent attention in studying stratified media: surfaces, interfaces and multilayers This method utilizes a geometry such that the $\gamma$-rays are incident on the flat surface of the sample at glancing angles of a few mrad close to the critical angle of the electronic total external reflection. The detected scattered particles are specularly reflected $\gamma$-photons, conversion electrons, conversion x-rays and incoherently scattered $\gamma$-photons. A general treatment of GIMS was published by Andreeva et al. in several papers. Starting from the nucleon current density expression of the susceptibility tensor $\chi$ given by Afanas’ev and Kagan and using a covariant formalism of anisotropic optics first introduced by Fedorov, these authors take into account that both the elements of the susceptibility tensor $\chi$ and the glancing angle $\theta$ are small in GIMS and calculate the $\gamma$-reflectivity. The method of calculation, however, especially for the higher multipolarity nuclear transitions, is rather cumbersome, since the nucleon current densities are directly calculated resulting in quite complex tensor expressions. In view of the extreme requirements to beam divergence, GIMS is certainly more suited for synchrotron radiation than for conventional radioactive source experiments.

Another general description of specular reflection of grazing incidence Mössbauer radiation was given by Hamon et al. Starting from the quantum theory of $\gamma$-radiation, they formulated the dynamic theory of Mössbauer optics. Unfortunately, the dynamic theory provides rather slow algorithms for calculating reflectivity spectra, therefore it is inefficient in spectrum fitting. In the grazing incidence limit, an optical model was derived from the dynamical theory, which has
recently been implemented in numerical calculations.\textsuperscript{15} Without using a covariant formalism, however, this latter approach also results in quite sophisticated algorithms since, in a layered medium, the eigenpolarizations vary from layer to layer.

Our aim is to rigorously derive general formulae for the transmissivity and the reflectivity of $\gamma$-radiation in the forward scattering and the grazing incidence case, respectively. Moreover, we shall try to obtain these formulae in such form that is suitable for fast numerical calculations in order to fit the experimental data. Like Andreeva et al.\textsuperscript{6-8} we start from the Afanas'ev–Kagan nucleon current density expression of the dielectric tensor\textsuperscript{10} and use a covariant anisotropic optical formalism.\textsuperscript{11,12} Instead of calculating the susceptibility tensor from the current densities of the nucleons, however, we reduce the problem to the calculation of the transmittance (forward scattering case) and the reflectivity (grazing incidence case) from the coherent forward scattering amplitude. We show that, in the case of forward scattering, this approach is equivalent to the theory of Blume and Kistner.\textsuperscript{2} The present treatment is based on no intuitive assumption and represents, thereby, a firm basis of the Blume–Kistner theory\textsuperscript{2} and of the Andreeva approximation.\textsuperscript{6-9}

\section{The Nuclear Susceptibility}

Let us consider the collective system of the nuclei and the electromagnetic field. The effect of the electromagnetic field will be treated as a perturbation on the randomly distributed nuclei. The interaction Hamiltonian $H$ between the nucleus and the electromagnetic field may be written as

\[ H = -\frac{1}{c} \sum_{i} j(r_i) \cdot A(r_i), \]  

where $j(r_i)$ is the current density of the $i$-th nucleon and $A(r_i)$ is the vector potential of the electromagnetic field at the point $r_i$:

\[ A(r_i) = \sum_{k,p} \left( \frac{2\pi \hbar c}{V_k} \right)^{1/2} \{ c_{k,p} \hat{u}_{k,p} \exp (i k \cdot r_i) + \text{H.C.} \}. \]  

In this formula, $c_{k,p}$ denotes a photon annihilation operator and $\hat{u}_{k,p}$ a unit polarization vector.

The matrix elements of the interaction Hamiltonian $H$ are scalar products of the current density matrix elements $J_{m_g m_e}$ and the polarization vectors $\hat{u}_{k,p}$:

\[ \langle \hat{u}_{k,p} | c_{k,p} \hat{u}_{k,p} \exp (i k \cdot r_i) | I_e m_e \rangle \]  

\[ J_{m_g m_e}(k) = \left\langle I_g m_g \left| \sum_{i} j(r_i) \exp (-i k \cdot r_i) \right| I_e m_e \right\rangle \]  

where $I_g$ and $I_e$ are the nuclear spin quantum numbers in the excited and ground state with the corresponding magnetic quantum numbers $m_g$ and $m_e$, respectively. $J(k)$ is the $k$-representation of the current density produced by a single nucleus. (Throughout the calculations we shall use the same letters for physical quantities in $r$- and $k$-representation letting the argument make evident which representation is meant.)

In first order perturbation of the electromagnetic field the average nucleon current density reads:\textsuperscript{19}

\[ \left\langle J^{(1)}(k) \right\rangle = \sigma(k) E(k) = \frac{i \omega}{c} \sigma(k) A(k), \]  

with $\sigma(k)$ being the conductivity tensor, which in turn defines the susceptibility tensor of the medium by

\[ \chi(k) = \frac{4\pi \sigma}{\omega} \sigma(k). \]  

Afanas'ev and Kagan\textsuperscript{10} calculated the susceptibility tensor in first order of the vector potential for randomly distributed nuclei in terms of the change of the average nucleon current density:

\[ \chi(k) = \frac{4\pi}{c^2 \omega^2} \frac{N}{2} \sum_{m_g m_e} \frac{J_{m_e m_g}(k) \circ J^{(1)m_e m_g}(k)}{E_k - E_{m_e m_g} + \frac{i\Gamma}{2}}. \]  

where $N$ is the number of resonant nuclei per unit volume, $E_k$ is the energy of the $\gamma$-photon, $E_{m_e m_g} = E_{m_e} - E_{m_g}$ is the energy difference between the nuclear excited and ground states, $\Gamma$ is the natural width of the excited state and $\circ$ is the dyadic vector product sign. The susceptibility tensor $\chi(k)$ depends on the propagation vector $k$ of the unperturbed wave. Instead of Eq. \textsuperscript{19} a $J^{(1)}(k)$ expression is obtained for non-random distribution of the scatterers with $(2\pi)^{-1} k$ being a reciprocal lattice vector. Only the random scatterer case will be further considered here.

Eq. \textsuperscript{20} is the starting equation of Andreeva in calculating grazing incidence Mössbauer spectra.\textsuperscript{8} In order to calculate $\chi(k)$ for an arbitrary orientation of the hy-
perline fields with respect to $\mathbf{k}$, the currents $J_{m_1m_2}$ are expanded in terms of irreducible tensors. For cases, like transitions of higher multipolarity, mixed multipole transitions, variation of hyperfine fields within the medium, texture, etc. the formalism therefore becomes cumbersome and numerically intractable. Having calculated the dielectric tensor of the Mössbauer medium, Andreeva et al. apply a very elegant covariant formalism and solve the problem of grazing incidence nuclear scattering by stratified media.

The numerical difficulties of the higher multipolar terms, perline field distributions, texture, etc. have been overcome years ago by Spiering in treating the thick absorber case in the Blume–Kistner formalism. The Hamiltonian, the scalar product of the current density $J_{m_1m_2}$ and the polarization vector $\hat{u}_{k\nu}$, has simpler transformation properties than $J_{m_1m_2}$, therefore, unlike Andreeva et al., the forward scattering amplitude

$$ f^{k,p\rightarrow k',p'} = -\frac{kV}{2\pi \hbar c 2I_g + 1} \sum_{m_1m_2m_3} \frac{H_{m_1m_2}^{k,p'} H_{m_2m_3}^{k,p'}}{E_k - E_{m_1m_2} + \frac{i\pi}{2}} $$

rather than $J_{m_1m_2}$ is calculated for an arbitrary $k$-direction.

In what follows we shall show that for $\gamma$-quanta in the physically relevant representation the $3 \times 3$ properties of the dielectric tensor are not fully used by the optical theory. Since the $k$-directions involved in the scattering problem are either equivalent (forward scattering) or extremely close to each other (grazing incidence case), the relevant block of the dielectric tensor is fully described by the four components of the forward scattering amplitude. This latter ensures that the present theory remains valid for nuclear transitions of any multipolarity. Indeed, expressing the susceptibility tensor in the polarization vector system $P = (\hat{e}_{\sigma}, \hat{e}_{\pi}, \epsilon_3 = \mathbf{k}/|\mathbf{k}|)$ of the unperturbed incident radiation the significant matrix elements are:

$$ \chi_{p'p}(\mathbf{k}) = \frac{4\pi N}{k^2} f^{k,p\rightarrow k',p'} \quad p, p' = \sigma, \pi; $$

$\sigma$ and $\pi$ being arbitrary polarizations. Once the susceptibility (the refractive index or the dielectric) tensor of the medium is defined the problem of calculating the propagation of electromagnetic field in the medium becomes an optical problem. Since the nuclear dielectrics is anisotropic, a polarization-dependent optical formalism will be used.

### III. COVARIANT ANISOTROPIC OPTICS OF A NUCLEAR DIELECTRICS

The covariant optical formalism of stratified anisotropic media developed by Borzdov, Barskovskii and Lavrukovich and applied by Andreeva et al. will be introduced here for three reasons:

1. Approximations made by Andreeva et al. are based on the assumption that the square of the scattering angle is of the order of the susceptibility tensor elements. The borderline of the Andreeva approximation will be specified here.

2. The Blume–Kistner theory will be derived from the covariant optical formalism.

3. In a practical application of the Blume–Kistner theory one calculates the exponentials of $2 \times 2$ complex matrices. The covariant optics uses $4 \times 4$ matrices in the exponentials leading to rather time consuming calculations. It will be shown that in a suitably chosen basis, the $4 \times 4$ matrices reduce to $2 \times 2$ ones both in forward scattering and in grazing incidence geometry.

#### A. The Borzdov–Barskovskii–Lavrukovich formalism

We may write the basic equation for the tangential components of the electric and magnetic fields $\mathbf{q} \times \mathbf{E}(\mathbf{q} \cdot \mathbf{r})$ and $\mathbf{H}_t(\mathbf{q} \cdot \mathbf{r}) = -\mathbf{q} \times [\mathbf{q} \times \mathbf{H}(\mathbf{q} \cdot \mathbf{r})]$ at the point $\mathbf{r}$ as follows:

$$ (\mathbf{q} \cdot \nabla) \left( \frac{\mathbf{H}_t(\mathbf{q} \cdot \mathbf{r})}{\mathbf{q} \times \mathbf{E}(\mathbf{q} \cdot \mathbf{r})} \right) = i k M(\mathbf{q} \cdot \mathbf{r}) \left( \frac{\mathbf{H}_t(\mathbf{q} \cdot \mathbf{r})}{\mathbf{q} \times \mathbf{E}(\mathbf{q} \cdot \mathbf{r})} \right), $$

were $\mathbf{q}$ represents the unit normal vector of the surface. The material parameters are allowed to vary only in the $\mathbf{q}$-direction (stratified medium) and the fields depend only on the $\mathbf{q} \cdot \mathbf{r}$ scalar product. $M$ is the differential propagation matrix defined by

$$ M = \begin{pmatrix} A & B \\ C & D \end{pmatrix}, $$

with

$$ A = (\mathbf{q} \cdot \varepsilon \mathbf{q})^{-1} \mathbf{q} \times \varepsilon \mathbf{a} - (\mathbf{q} \cdot \mu \mathbf{q})^{-1} \mathbf{b} \circ \mathbf{q} \mu I, $$

$$ B = (\mathbf{q} \cdot \varepsilon \mathbf{q})^{-1} \mathbf{q} \times \varepsilon \mathbf{b} - (\mathbf{q} \cdot \mu \mathbf{q})^{-1} \mathbf{a} \circ \mathbf{q} \mu I, $$

$$ C = - (\mathbf{q} \cdot \varepsilon \mathbf{q})^{-1} \mathbf{a} \circ \mathbf{a} - (\mathbf{q} \cdot \mu \mathbf{q})^{-1} \mathbf{q} \times \mu \mathbf{q}, $$

$$ D = (\mathbf{q} \cdot \varepsilon \mathbf{q})^{-1} \mathbf{a} \circ \mathbf{q} \varepsilon \mathbf{q} - (\mathbf{q} \cdot \mu \mathbf{q})^{-1} I \mu \mathbf{q} \circ \mathbf{b}. $$

Here $\varepsilon = 1 + \chi$ is the dielectric tensor, $\mu$ denotes the dual tensor of an arbitrary vector $\mathbf{v}$ and the tilde sign stands for the transpose of a tensor. The $I - (\mathbf{q} \times \mathbf{q})^2$ operator projects a vector into the plane of the sample surface. The tangential component of the incident wave vector is $\mathbf{b} = \mathbf{I} k / |k|$, and $\mathbf{a} := \mathbf{b} \times \mathbf{q}$ is a vector perpendicular to the reflection plane, $\varepsilon = \text{det}(\varepsilon) \varepsilon^{-1}$, $\mu = \text{det}(\mu) \mu^{-1}$. Strictly speaking, $A, B, C$ and $D$ are 3-dimensional tensors acting only, as it can be seen, in the $\mathbf{a}, \mathbf{b}$ plane. Consequently, $M$ can be properly represented by $4 \times 4$ matrices. The permeability tensor $\mu$ will play no further role. The solution of Eq. (10) relates $\mathbf{H}_t$ and $\mathbf{q} \times \mathbf{E}$ to each other at the lower and upper surfaces of the
layered medium. In a homogeneous film of thickness $d$, the solution is given by the so-called integral propagation matrix $L = \exp (ikdM)$, by the matrix exponential of the differential propagation matrix. For an $n$-layer system, the total integral propagation matrix is the product of the individual integral propagation matrices $L_{(l)}$ of layer $l$, thus

$$L = L_{(n)}...L_{(2)}L_{(1)}.$$ (12)

The expression of the planar reflectivity, $r$ defined by $H^0_r = rH^0_t$, where $H^0_r$ and $H^0_t$ are the tangential amplitudes of the reflected and transmitted waves, respectively, writes as

$$r = \left( (\gamma^t, -I_2) L \left( I_2 \gamma^r \right) \right)^{-1} \times \left( (-\gamma^t, I_2) L \left( I_2 \gamma^r \right) \right).$$ (13)

Here $I_2$ is the $2 \times 2$ unity matrix and the $\gamma^0$, $\gamma^r$ and $\gamma^t$ tensors are the impedance tensors for the incident, specularly reflected and transmitted waves, respectively, defined by the

$$\gamma^{0,r,t} = \hat{q} \times E^{0,r,t}$$ (14)
equation. Since the $\gamma$ tensors act in the plane perpendicular to $\hat{q}$, they can be represented by $2 \times 2$ matrices.

The elements of the reflectivity matrix $R$ are geometrically related to the elements of the planar reflectivity, $r$ i.e.

$$R_{\sigma\sigma} = -r_{22}, \quad R_{\sigma\pi} = -r_{21}\sin^{-1}\theta, \quad R_{\pi\sigma} = r_{12}\sin\theta, \quad R_{\pi\pi} = r_{11},$$ (15)

where $\sigma$ and $\pi$ are polarizations corresponding to $E$ perpendicular and parallel to the plane of incidence, respectively. For numerical calculations we shall choose different appropriate coordinate systems. The laboratory system $S$ will be defined so that the $x$, $y$ and $z$-axes are parallel to $a$, $b$ and $q$, respectively. The field components in Eq. (10) define a natural permutation $K = (1, 2, 3, 4)$ basis of the 4-component field vectors $[H_x, H_y, (\hat{q} \times E)_x, (\hat{q} \times E)_y]$ with respect to the $S$-system. A convenient permutation of $K = (1, 2, 3, 4)$ viz. $K' = (2, 3, 4, 1)$ shall also be used. The differential and integral propagation matrices $M$ and $L$ will be denoted by $M'$ and $L'$, respectively in the $K'$-system.

B. The forward scattering case: The Blume–Kistner equation

The (11) differential propagation matrix for the case of normal incidence in the $K$-system has the following form of

$$M = \begin{pmatrix} 0 & 0 & \varepsilon_{22} & -\varepsilon_{21} \\ 0 & 0 & -\varepsilon_{12} & \varepsilon_{11} \\ 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \end{pmatrix}.$$ (16)

The transmissivity may be expressed in terms of the integral propagation matrix, $L = \exp (ikdM)$ as

$$t = 2 \left( (I_2, I_2) L^{-1} \left( I_2' I_2 \right) \right)^{-1}$$ (17)
deefined by $H^t_{I} = tH^0_{I}$ can be explicitly elaborated to obtain the Blume–Kistner formulae. Indeed, using the identity (which can easily be proved by expanding the exponentials):
Kistner²

$$n_{pp'} = \delta_{pp'} + \frac{2\pi N}{k^2} f^{k,p \rightarrow k',p'},$$

(22)

where \(\delta\) is the Kronecker symbol and \(p, p' = \sigma, \pi\).

C. The grazing incidence case: The Andreeva approximation

1. The differential propagation matrix

In order to see which elements in Eq. (15) are of the same order of magnitude, we eliminate the explicit \(\theta\)-dependence of \(R\) by applying a linear transformation \(T\) (in the \(\mathcal{K}'\)-system) of the form:

$$T = \begin{pmatrix}
\sin^{-1}\theta & 0 & 0 & 0 \\
0 & \sin^{-1}\theta & 0 & 0 \\
0 & 0 & 1 & 0 \\
0 & 0 & 0 & 1
\end{pmatrix}.$$  

(23)

It can be easily seen that only the integral propagation matrix

$$L''_l = TL'_l T^{-1}$$

(24)

depends on \(\theta\), and the reflectivity matrix depends on the elements of \(L''_l\) only. The transform of the differential propagation matrix \(M''_l = TM'_l T^{-1}\) of layer \(l\) is obtained with the same similarity transformation:

$$M''_l = \sin \theta \begin{pmatrix}
0 & 0 & 1 & 0 \\
0 & 0 & 1 & 0 \\
1 & 0 & 0 & 0 \\
0 & 1 & 0 & 0
\end{pmatrix} + \frac{1}{\sin \theta} \begin{pmatrix}
\chi_{l}^{(1)11} & \chi_{l}^{(1)13} & 0 & 0 \\
\chi_{l}^{(1)31} & \chi_{l}^{(1)33} & 0 & 0 \\
0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0
\end{pmatrix} + \begin{pmatrix}
0 & -\chi_{l}^{(1)12} & 0 & 0 \\
0 & -\chi_{l}^{(1)32} & 0 & 0 \\
0 & 0 & 0 & 0 \\
0 & \chi_{l}^{(2)22} \sin \theta & -\chi_{l}^{(2)21} & -\chi_{l}^{(2)23}
\end{pmatrix},$$

(25)

\(\chi_{l}^{(i)j} (i, j = 1, 2, 3)\) being the matrix elements of the susceptibility tensor of layer \(l\).

The three matrices in Eq. (25) are of the order of magnitude of \(\chi / \theta\) and \(\chi / \theta^2\), respectively. Without a rigorous explanation, Andreeva et al.²² intuitively drop the third term containing only those elements of the \(\chi\) tensor which are not related to the forward scattering amplitude. This approximation is obviously valid if \(\chi\) is small compared to \(\chi / \theta\) and \(\chi / \theta^2\). Since typically \(\chi \approx 10^{-5}\), the interval for \(\theta\) in order the third term to remain below 1% of the first two is: \(10^{-4} < \theta \ll 10^{-2}\), which is, indeed, the typical region of a grazing incidence experiment. From the (25) form of \(M\) it is clearly seen what conditions have to be fulfilled for the Andreeva approximation to be valid. Note that there is not only an upper but also a lower bound for \(\theta\).

Returning to the covariant notation, the differential propagation matrix \(M''_l\) of layer \(l\) with a 1% accuracy in the \(\mathcal{K}\)-system is of the form

$$M''_l = \begin{pmatrix}
(a - b) \chi_{l}^{(i)q} \circ a & I - b \circ a [1 - (q : \chi_{l}^{(i)} \circ a)] \\
I - a \circ a [1 - (q : \chi_{l}^{(i)} \circ a)] & (q : \chi_{l}^{(i)} \circ a) a \circ b
\end{pmatrix},$$

(26)

which is identical to the form suggested by Andreeva et al.²². In the grazing incidence case the \(a\) and \(b\) vectors are approximate unit vectors \(|a| = |b| = \cos \theta \approx 1\). This approximation is equivalent to neglecting terms of the order of \(\sin^2 \theta\) as compared to 1. In this limit \(k||b\). We can choose the two polarization vectors so that \(\hat{u}_1 = a\) and \(\hat{u}_2 = q\).

Transforming the \(\chi\) matrix given in the polarization vector system \(P\) by Eq. (9) into the \(S\) system and substituting into Eq. (26) the differential propagation matrix can be expressed in terms of the forward scattering amplitude:

$$M''_l \approx \begin{pmatrix}
\frac{4\pi N_{l}(l)}{k^2} f^{k,\sigma \rightarrow k,\pi} & 0 & 0 & 0 \\
\frac{4\pi N_{l}(l)}{k^2} f^{k,\pi \rightarrow k,\sigma} & 0 & 0 & 0 \\
0 & \frac{4\pi N_{l}(l)}{k^2} f^{k,\sigma \rightarrow k,\pi} + \sin^2 \theta & 0 & 0 \\
1 & 0 & 0 & 0
\end{pmatrix}.$$  

(27)

which, as we shall see, is a particularly suitable form for numerical calculations (\(N_{l}(l)\) is the number of resonant nuclei per unit volume and \(f^{k,p \rightarrow k',p'}\) is the coherent forward scattering amplitude in layer \(l\)). Starting with Eq. (27) a time-effective numerical algorithm is derived in the following sub-section.

2. Numerical calculations

The matrix (27) contains small quantities of the order of \(\sin^2 \theta < 10^{-4}\) and the much larger number unity. The calculation of the exponential of \(M''_l\) to a sufficient accuracy is rather time consuming even if the approximation \(\exp y \approx (1 + \frac{y}{2})^2^n\) is applied. For each energy channel
the exponential of $M$ should be calculated thus typically $2^{10}$ times per M"ossbauer spectrum.

The corresponding transformed integral propagation matrix in the $K'$-system [cf. Eq. (25)] can be written as

$$L''(t) = \exp \left( \frac{i kd(t) TM'(t) T^{-1}}{x(t) I_2 + \frac{1}{2} x(t) \phi(t)} \right),$$

where $x(t) = ikd(t) \sin \theta$, with $d(t)$ being the thickness of layer $l$. $\phi(t) = -4\pi N(t)d(t)f(t)$ is proportional to the forward scattering amplitude $f(t)$.

To evaluate the integral propagation matrix one may notice that the differential propagation matrix is block-anti-diagonal. We show that the problem, like in the Blume–Kistner case in Sec. III B reduces to the calculation of a single $2 \times 2$ matrix exponential of a small quantity. Indeed, using again the identity with $B_{(t)} = x(t)I_2 + \frac{1}{2} x(t) \phi(t)$ and $C_{(t)} = x(t)I_2$ the integral propagation matrix of Eq. (28) with $F_{(t)} = (x(t)B_{(t)})^{1/2}$ is given by:

$$L''(t) = \begin{pmatrix} \cosh F_{(t)} & \frac{1}{x(t)} F_{(t)} \sinh F_{(t)} \\ x(t) F_{(t)}^{-1} \sinh F_{(t)} & \cosh F_{(t)} \end{pmatrix}. \tag{29}$$

Eq. (29) is well suited for numerical calculations since it contains only the $2 \times 2$ matrix exponential $\exp F_{(t)}$.

By the present method the large matrix elements are separated from the small ones. If the argument of the exponential is of the order of $10^{-4}$ the $\exp y \approx (1 + \frac{y}{2})^2$ approximation gives a sufficient accuracy with $n$ as small as $2$.

Using Eqs. (13), (15), (12), and (29) the reflectivity in the $\sigma, \pi$ basis is given by

$$R = \left( L''_{[11]} - L''_{[12]} - L''_{[21]} + L''_{[22]} \right)^{-1} \left( L''_{[11]} + L''_{[12]} - L''_{[21]} - L''_{[22]} \right), \tag{30}$$

in time domain.

ACKNOWLEDGMENT

Fruitful discussions with Dr. M. A. Andreeva are gratefully acknowledged. This work was partly supported by the PHARE ACCORD Program under Contract No. H–9112–0522 and by the Hungarian Scientific Research Fund (OTKA) under Contract Nos. 1809 and T016667. The authors also thank for the partial support by the Deutsche Forschungsgemeinschaft and the Hungarian Academy of Sciences in frames of a bilateral project.

DERIVATION OF THE REFLECTIVITY FORMULA

The integral propagation matrix $L$ of Eq. (13) is expressed by $L''$ of Eq. (24):

$$r = \left( \begin{pmatrix} \gamma^t & -I_2 \end{pmatrix} V^{-1} T^{-1} L'' T V \begin{pmatrix} I_2 \gamma^r \end{pmatrix} \right)^{-1} \times \begin{pmatrix} \gamma^t, I_2 \end{pmatrix} V^{-1} T^{-1} L'' T V \begin{pmatrix} I_2 \gamma^0 \end{pmatrix}, \tag{31}$$

where $T$ is given in Eq. (23), $\gamma^{t,r,0}$ are the impedance tensors for the transmitted, reflected and incident radiation as defined in Eq. (13). Assuming vacuum on both sides of the stratified sample (which — by allowing for a thick enough substrate — imposes no further restriction)
the $\gamma$'s are of the form\cite{9}:

$$\gamma^0 = \gamma^t = -\gamma^r = \begin{pmatrix} \sin \theta & 0 \\ 0 & \sin^{-1} \theta \end{pmatrix}$$

(32)

$V$ is the matrix of the $(1, 2, 3, 4)$ \textit{viz.} $(2, 3, 4, 1)$ ($K \rightarrow K'$) transformation of the form:

$$V = \begin{pmatrix} 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \\ 1 & 0 & 0 & 0 \end{pmatrix}$$

(33)

Performing the calculations in Eq. (31) with the above matrices the planar reflectivity:

$$r = - \left( \begin{pmatrix} 0 & \sin^{-1} \theta \\ -1 & 0 \end{pmatrix} \right)^{-1} \left( L''_{[1]} - L''_{[12]} - L''_{[2]} + L''_{[22]} \right)^{-1}$$

$$\times \left( L''_{[11]} + L''_{[12]} - L''_{[21]} - L''_{[22]} \right) \begin{pmatrix} 0 & \sin^{-1} \theta \\ 1 & 0 \end{pmatrix}$$

(34)

From Eqs. (34) and (15) we obtain the reflectivity formula.

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