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Reflection variant of nonlinear polarization spectroscopy

I. I. Gancheryonok, A. V. Lavrinenko

Belarusian State University, Department of Physics, Fr. Skariny av. 4, Minsk 220080, Belarus.

The theoretical analysis of reflection scheme for studying light-induced anisotropy in initially isotropic media is presented. It is based on a very powerful wave operators formalism. Our most significant result is the expression for amplitude reflection operator in the general covariant (coordinate free) form. We also derive the expression for the detected signal in reflection variant of nonlinear polarization spectroscopy (NPS) in the case of linearly polarized interacting pump and probe waves.

1. Wave operators formalism

Let us consider in detail the application of Fresnel's wave operator formalism to the problem of nonlinear polarization spectroscopy. The principles of such a formalism were given in a number of papers (see, for example, [1]–[3] and the literature cited therein). A surface impedance operator $\hat{\gamma}$ (this is a linear operator of matrix form, so it may be considered as a tensor) is one of the fundamental concepts of this theory. The operator $\hat{\gamma}$ generalizes a scalar surface impedance which has been widely known in optics and radiowave theory for many years [4]–[6]. Another important concept is a normal refraction operator $\hat{N}$, which generalizes the refractive index operator [7], [8]. The operator N (or tensor as we showed earlier) describes the space evolution of the field vector amplitudes of an electromagnetic wave under propagation in anisotropic and gyrotropic media. Surface impedance and normal refraction operators are very useful in the theory of electromagnetic or elastic waves propagation in stratified anisotropic media [9], [10]. The boundary problem in such kind of media is strictly formulated by means of operators $\hat{\gamma}$ and $\hat{N}$ which depend on the characteristics of the incident waves and properties of the corresponding media. Thus, the operator method of solving various boundary problems, including reflection and transmission of waves at a single plane interface, may be applied to calculations where high precision is needed.

Let the first medium, from which the plane harmonic wave of frequency ω

$$\boldsymbol{E}(\boldsymbol{r}, t) = \boldsymbol{E}_i \exp[i(k\boldsymbol{m}\boldsymbol{r} - \omega t)] \tag{1}$$

is obliquely incident onto the second one, be isotropic, and the other medium be anisotropic and described by the permittivity tensor $\hat{\varepsilon}$ and permeability tensor $\hat{\mu}$. Here, $\boldsymbol{E}_i$ is the complex vector amplitude of the electric field strength, $k = \omega/c$,

c is the velocity of the light in vacuum, $\boldsymbol{m} = \boldsymbol{b} + m_n \boldsymbol{q}$ is the refraction vector [11] with tangential $\boldsymbol{b}$ and normal $m_n \boldsymbol{q}$ components, $\boldsymbol{q}$ is the unit normal to the interface. For geometry of the problem see Fig. 1. Refraction vector $\boldsymbol{m}$ is connected with wave normal $\boldsymbol{n}$ by means of an index of refraction n

$$\boldsymbol{m} = n\,\boldsymbol{n}. \tag{2}$$

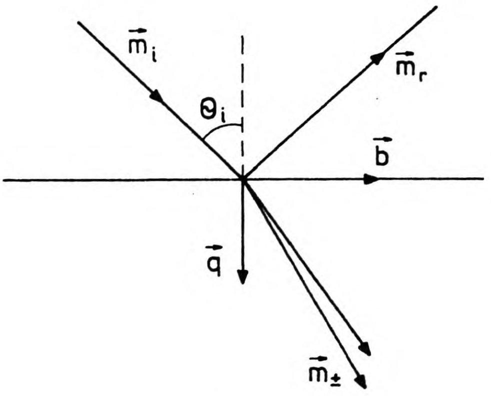

Fig. 1. Geometry of the problem: $\boldsymbol{m}_i$, $\boldsymbol{m}_r$, and $\boldsymbol{m}_\pm$ are the incidence, reflection and refraction vectors, respectively

It is convenient, especially in nonmagnetic media, to use the vector of magnetic field strength $\boldsymbol{H}$ and its tangential component $\boldsymbol{H}_t$ (relatively to the interface), [1]–[3]. It is well known that for the plane waves

$$\boldsymbol{H} = (\boldsymbol{m} \times \boldsymbol{E}) = \boldsymbol{m}^\times \boldsymbol{E} \tag{3}$$

where $(\boldsymbol{m} \times \boldsymbol{E})$ denotes the vector product of $\boldsymbol{m}$ and $\boldsymbol{E}$, $\boldsymbol{m}^\times$ is the antisymmetric second-rank tensor, dual to the vector $\boldsymbol{m}$ [11].

Projective operator $\hat{G}$ is applied to cut vector in tangential plane. With the help of dyad $\boldsymbol{q} \otimes \boldsymbol{q}$, where $\otimes$ denotes direct product of two vectors, and unit tensor $\hat{I}$ one can easily find that $\hat{G} = \hat{I} - \boldsymbol{q} \otimes \boldsymbol{q}$. Thus,

$$\boldsymbol{H}_t = \hat{G}\boldsymbol{H}. \tag{4}$$

We follow the intrinsic form of notations [11], meaning in (4) contraction of tensor $\hat{G}$ with vector $\boldsymbol{H}$ in such form: $(H_t)_i = \sum_{j=1}^{3} G_{ij} H_j$. The direct manipulation with tensors and vectors as in (4) simplifies final expressions and provides the results of great generality, eliminating the use of any coordinate system. Moreover, the results obtained are suitable for computer use.

Further, one can find complex vector amplitudes of reflected $\boldsymbol{H}_t^r$ and transmitted waves $\boldsymbol{H}_t^d$ as shown in papers [1]–[3]:

$$\boldsymbol{H}_t^r = \hat{r}\boldsymbol{H}_t, \quad \boldsymbol{H}_t^d = \hat{d}\boldsymbol{H}_t, \quad \hat{r} + \hat{G} = \hat{d} \tag{5}$$

where $\hat{r}$ and $\hat{d}$ are Fresnel's reflection and transmission operators, respectively. These operators are expressed by means of the surface impedance operators of incident $\hat{\gamma}^i$, reflected $\hat{\gamma}^r$ and transmitted $\hat{\gamma}^d$ as follows:

$$\hat{r} = (\hat{\gamma}^d - \hat{\gamma}^r)^- (\hat{\gamma}^i - \hat{\gamma}^d), \quad \hat{d} = (\hat{\gamma}^d - \hat{\gamma}^r)^- (\hat{\gamma}^i - \hat{\gamma}^r) \tag{6}$$

where $(\hat{\gamma}^d - \hat{\gamma}^r)^-$ is a pseudoinverse operator (pseudo — because Fresnel's operators $\hat{r}$ and $\hat{d}$ are the planar tensors, acting in two-dimensional subspace of plane interface, so the common inverse operation for these operators is not determined). Pseudoinverse operator $\hat{\alpha}_t^-$ can be found through the algorithm presented in papers [1]—[3]

$$\hat{\alpha}^- = (\hat{\alpha}_t \hat{I} - \hat{\alpha}) / \bar{\bar{\alpha}}_t \tag{7}$$

where $\bar{\bar{\alpha}}_t$ denotes the trace of adjoint (mutual) tensor [11]. Surface impedance tensor $\hat{\gamma}$ is introduced as a linear operator transforming vector $\boldsymbol{H}_t$ into vector $(\boldsymbol{q} \times \boldsymbol{E})$

$$(\boldsymbol{q} \times \boldsymbol{E}) = \hat{\gamma} \boldsymbol{H}_t. \tag{8}$$

These vectors lie at the plane interface. It is shown [2], [3] that the surface impedance satisfies Riccati tensor equation

$$\hat{\gamma}\hat{B}\hat{\gamma} + \hat{\gamma}\hat{A} - \hat{D}\hat{\gamma} - \hat{C} = 0 \tag{9}$$

where tensor coefficients are:

$$\begin{aligned}
\hat{A} &= \frac{1}{\varepsilon_q} \boldsymbol{q}^\times \hat{\varepsilon} \boldsymbol{q} \otimes \boldsymbol{a} - \frac{1}{\mu_q} \boldsymbol{b} \otimes \boldsymbol{q} \hat{\mu} \hat{G}, \\
\hat{B} &= \frac{1}{\varepsilon_q} \hat{I} \tilde{\hat{\varepsilon}} \hat{I} - \frac{1}{\mu_q} \boldsymbol{b} \otimes \boldsymbol{b}, \\
\hat{C} &= -\frac{1}{\varepsilon_q} \boldsymbol{a} \otimes \boldsymbol{a} - \frac{1}{\mu_q} \boldsymbol{q}^\times \tilde{\hat{\mu}} \boldsymbol{q}^\times, \\
\hat{D} &= -\frac{1}{\varepsilon_q} \boldsymbol{a} \otimes \boldsymbol{q} \hat{\varepsilon} \boldsymbol{q}^\times - \frac{1}{\mu_q} \hat{G} \hat{\mu} \boldsymbol{q} \otimes \boldsymbol{b}, \\
\varepsilon_q &= \boldsymbol{q} \hat{\varepsilon} \boldsymbol{q}, \quad \mu_q = \boldsymbol{q} \hat{\mu} \boldsymbol{q},
\end{aligned} \tag{10}$$

tilde denotes transposed operator.

For harmonic wave (1) in isotropic media surface impedance is given by formula [3]

$$\hat{\gamma}^i = -\hat{\gamma}^r = \left(\mu_i \hat{G} - \frac{b^2}{\varepsilon_i} \hat{\tau}_a \right) \Big/ m_n$$

where dyad $\hat{\tau}_a = \boldsymbol{a}_{-0} \otimes \boldsymbol{a}_{-0}$ is a projective operator in the direction orthogonal to incident plane, $\boldsymbol{a}_{-0} = (\boldsymbol{b}_{-0} \times \boldsymbol{q})$, $b = |\boldsymbol{b}|$, $\boldsymbol{b}_0 = \boldsymbol{b}/b$. In an anisotropic medium two partial waves are excited [12]. They have different polarizations and refraction vectors $\boldsymbol{m}_{\mp} = \boldsymbol{b} + m_n^{\mp} \boldsymbol{q}$. Therefore, an operator $\hat{\gamma}$ (8) connects vectors $\boldsymbol{H}_t$ and $(\boldsymbol{q} \times \boldsymbol{E})$ which characterize a superposition of harmonic fields of both refracted waves. In particular case, when crystal is nonmagnetic ($\hat{\mu} = \hat{I}$) and is cut in such a way that vector $\boldsymbol{q}$ is an eigenvector of tensor $\hat{\varepsilon}$ ($\boldsymbol{q}\hat{\varepsilon} = \hat{\varepsilon}\boldsymbol{q} = \varepsilon_q \boldsymbol{q}$), tensor $\hat{\gamma}_d$ is expressed in the following form:

$$\hat{\gamma}_d = (m_n^+ + m_n^-)^{-1}\left[\hat{G} - m_n^+ m_n^- \frac{q^\times \hat{\varepsilon}^{-1} q^\times}{1 - a\hat{\varepsilon}^{-1} a} - \left(\frac{1}{\varepsilon_q} + \frac{\varepsilon_q m_n^+ m_n^-}{|\hat{\varepsilon}| - a\bar{\hat{\varepsilon}} a}\right)\hat{\tau}_a\right] \tag{11}$$

where $|\hat{\varepsilon}|$ is the determinant of tensor $\hat{\varepsilon}$. The total field of refracted waves

$$H_t^d(r,t) = H_t^d(0)\exp[i(k\boldsymbol{b}\boldsymbol{r} - \omega t)]\exp(ikz\hat{N}) \tag{12}$$

contains an exponential operator $\exp(ikz\hat{N})^k = \sum_{k=0}^{\infty} \frac{(ikz)^k}{k!}\hat{N}^k$, where $z = \boldsymbol{q}\boldsymbol{r}$,

$$\hat{N} = \hat{A} + \hat{B}\hat{\gamma}. \tag{13}$$

The eigenvalues and eigenvectors of normal refraction operator (13) define normal components $m_n^\pm$ of refraction vector and polarization states of harmonic partial refracted waves. Values $m_n^\pm$ are the solutions of the fourth-power algebraic equation

$$m_n^4 - \hat{P}_t m_n^3 + (\bar{\hat{P}}_t - \hat{Q}_t)m_n^2 + (\hat{P}_t\hat{Q}_t - (\hat{P}\hat{Q})_t)m_n + \bar{\hat{Q}}_t = 0, \tag{14}$$

or one can find them solving the equation of normals [11]

$$(m\overline{\hat{\varepsilon}^{-1}}\, m)(m\overline{\hat{\mu}^{-1}}\, m) + (\hat{\varepsilon}^{-1} m^\times \hat{\mu}^{-1} m^\times)_t + 1 = 0. \tag{15}$$

Tensor coefficients $\hat{P}$ and $\hat{Q}$ in (13) are derived from $\hat{A}, \hat{B}, \hat{C}, \hat{D}$:

$$\hat{P} = \hat{A} + \hat{B}\hat{D}\hat{B}^-, \quad \hat{Q} = \hat{B}(\hat{C} - \hat{D}\hat{B}^-\hat{A}). \tag{16}$$

Some complicated cases with degenerated tensor $\hat{N}$, where refracted waves with linear quadratic and cubic dependence on coordinates appear, are considered in detail in papers [12], [13].

2. Reflection from media with light-induced anisotropy (MLIA)

Let us consider an initially isotropic nonlinear medium in which uniaxial anisotropy is induced by the powerful pumping plane monochromatic wave normally incident from vacuum. If pumping wave is linearly polarized, then dielectric permittivity tensor $\hat{\varepsilon}$ may be written in the form [14]

$$\hat{\varepsilon} = \varepsilon_0[(1 + \chi_0 + \chi_1)\hat{I} + \chi_1 C c \otimes c], \tag{17}$$

where ε_0 is the dielectric constant, χ_0 is the linear scalar susceptibility, $\chi_1 = \chi_{1221}$, $C = (\chi_{1122} + \chi_{1212})/\chi_1$, χ_{ijkl} are the components of the fourth-rank tensor of the third-order nonlinear susceptibility, $\boldsymbol{c}$ is the unit vector of induced optical axis ($\boldsymbol{c}$ is also polarization vector of the pumping field). Just as an incident pump wave normal is perpendicular to the plane interface, so the optical axis of MLIA is at the plane interface. Therefore, one can write

$$\boldsymbol{c} = \cos\varphi\, \boldsymbol{a}_0 + \sin\varphi\, \boldsymbol{b}_0 \tag{18}$$

where φ is the azimuth, counted from direction of vector $\boldsymbol{a}_0$ which is perpendicular to the incidence plane. We point out that the substitution of vector $\boldsymbol{c}$ (18) into (17) yields

$$\hat{\varepsilon}\boldsymbol{q} = \boldsymbol{q}\hat{\varepsilon} = \varepsilon_q \boldsymbol{q}, \tag{19}$$

i.e., vector $\boldsymbol{q}$ is the eigenvector of dielectric permittivity tensor $\hat{\varepsilon}$ (17) with eigenvalue

$$\varepsilon_q = \varepsilon_0(1+\chi_0+\chi_1). \tag{20}$$

Therefore, we may use the exact analytical expression (11) for surface impedance tensor substituting in it the following formula for permittivity tensor $\hat{\varepsilon}$ (17)

$$\hat{\varepsilon} = \varepsilon_q + \varepsilon_1 \hat{\tau}_c \tag{21}$$

where $\varepsilon_1 = \varepsilon_0 \chi_1 C$, $\hat{\tau}_c$ denotes projective dyad operator $\boldsymbol{c}\otimes\boldsymbol{c}$, and we imply that scalar ε_q is multiplied by a unit tensor $\hat{I}$, which from now we shall drop out. The simple but cumbersome calculation gives us expression for impedance tensor of MLIA

$$\hat{\gamma}^d = \frac{\hat{X}}{\varepsilon_q(m_n^+ + m_n^-)} + \frac{m_n^+ m_n^-}{m_n^+ + m_n^-}\,\frac{\hat{X}+\varepsilon_1\hat{\tau}_c}{F} \tag{22}$$

where tensor $\hat{X}$ is

$$\hat{X} = \varepsilon_q \hat{\tau}_b + m_n^2 \hat{\tau}_a, \quad \hat{\tau}_b = \boldsymbol{b}_0 \otimes \boldsymbol{b}_0 \tag{23}$$

and by F we denote

$$F = \varepsilon_q(\varepsilon_q+\varepsilon_1) - b^2(\varepsilon_q + \varepsilon_1 \sin^2\varphi).$$

Eigenvalues $m_n^\pm$ of normal refraction tensor are derived from (13): $m_n^- = \sqrt{\varepsilon_q}\cos\Theta$ for ordinary wave and $m_n^+ = \sqrt{\varepsilon_q - b^2 + \varepsilon_1(1-b^2\sin\varphi/\varepsilon_q)}$ for extraordinary wave. Angle of refraction of ordinary wave Θ_0 is simply derived from Snell's law

$$\frac{\sin\Theta_i}{\sin\Theta_0} = \sqrt{\varepsilon_q}. \tag{24}$$

Further transformations are connected with the small magnitude of light-induced anisotropy. It means that $|\varepsilon_1| \ll |\varepsilon_q|$ and we may consider ε_1 as a small parameter in expansion of m_n^+ in a series.

Neglecting terms higher than linear in ε_1, we obtain

$$m_n^+ \approx m_n^- \left[1 + \frac{\varepsilon_1(1-\sin^2\varphi\sin^2\Theta_0)}{2(m_n^-)^2}\right]. \tag{25}$$

Substitution of (25) into formula (22) gives the tensor of surface impedances in the form

$$\hat{\gamma}^d = \left[\frac{1}{m_n^-} + \Gamma_b\right]\hat{\tau}_b + \left[\frac{m_n^-}{\varepsilon_q} + \Gamma_a\right]\hat{\tau}_a + \lambda(\hat{\tau}_b - \hat{\tau}_a)q^\times. \tag{26}$$

Here we introduce the following notations:

$$\Gamma_a = -\frac{\varepsilon_1 \sin^2\varphi\cos^2\Theta_i}{2\varepsilon_q m_n^-},$$

$$\Gamma_b = \frac{\varepsilon_1}{2\varepsilon_q(m_n^-)^3}((m_n^-)^2\sin^2\varphi + \varepsilon_q b^2 \sin^2\varphi - \varepsilon_q), \tag{27}$$

$$\lambda = \frac{\varepsilon_1 \sin 2\varphi}{4\varepsilon_q m_n^-}.$$

It is clear that $\frac{m_n^-}{\varepsilon_q} \gg \Gamma_a$, $\frac{1}{m_n^-} \gg \Gamma_b, \Gamma_a, \Gamma_b, \lambda$, thus Γ_a, Γ_b, λ are quantities of the first power of small parameter ε_1 and they give small additions to the expression for tensor $\hat{\gamma}$ in the case of isotropic dielectric

$$\hat{\gamma}^d = \frac{1}{m_n^-}\hat{\tau}_b + \frac{m_n^-}{\varepsilon_q}\hat{\tau}_a. \tag{28}$$

This expression definitely follows from (22) for the case of an isotropic medium. In the vacuum or with high accuracy in the air ($\hat{\varepsilon}_i = \hat{I}$, $\hat{\mu}_i = \hat{I}$, $n = 1$) refraction vector $\boldsymbol{m}$ and wave normal $\boldsymbol{n}$ coincide. Let us rewrite relation (26) in the context of such assumption ($m_n = \cos\Theta_i$, $b = \sin\Theta_i$)

$$\hat{\gamma} = -\hat{\gamma}^r = m_n\hat{\tau}_a + \frac{1}{m_n}\hat{\tau}_b. \tag{29}$$

Now, we are ready to derive Fresnel's reflection and transmission operators. Using approximate formulae (26) and (29), it is easy to obtain from (6) tensors of reflection and transmission, generalizing usual Fresnel's coefficients. One has:

$$\hat{d} = \hat{d}_0 + \hat{d}_1, \quad \hat{r} = \hat{r}_0 + \hat{r}_1 \tag{30}$$

where norms of tensors $\hat{d}_0$ and $\hat{r}_0$ are much more than norms of tensors $\hat{d}_1$, $\hat{r}_1$: $\|\hat{d}_1\| \ll \|d_0\|$; $\|\hat{r}_1\| \ll \|\hat{r}_0\|$. The zero-order approximations for $\hat{d}_0$ and $\hat{r}_0$ have been written in the following form:

$$\hat{d}_0 = d_{0a}\hat{\tau}_a + d_{0b}\hat{\tau}_b = \frac{2m_n}{V}\hat{\tau}_a + \frac{2}{m_n\Lambda}\hat{\tau}_b = \frac{2\sqrt{\varepsilon_q}\cos\Theta_i}{\sqrt{\varepsilon_q}\cos\Theta_i + \cos\Theta_0}\hat{\tau}_a$$
$$+ \frac{2\sqrt{\varepsilon_q}\cos\Theta_0}{\cos\Theta_i + \sqrt{\varepsilon_q}\cos\Theta_0}\tau_b, \tag{31}$$

$$\hat{r}_0 = r_{0a}\hat{\tau}_a + r_{0b}\hat{\tau}_b = \left(\frac{2m_n}{V} - 1\right)\hat{\tau}_a + \left(\frac{2}{m_n\Lambda} - 1\right)\hat{\tau}_b$$
$$= \frac{\sqrt{\varepsilon_q}\cos\Theta_i - \cos\Theta_0}{\sqrt{\varepsilon_q}\cos\Theta_i + \cos\Theta_0}\hat{\tau}_a + \frac{\sqrt{\varepsilon_q}\cos\Theta_0 - \cos\Theta_i}{\sqrt{\varepsilon_q}\cos\Theta_0 + \cos\Theta_i}\hat{\tau}_b, \tag{32}$$

$$\Lambda = \frac{1}{m_n} + \frac{1}{m_n^-}, \quad V = m_n + \frac{m_n^-}{\varepsilon_q}. \tag{33}$$

Direct comparison shows that tensors $\hat{r}_0$ and $\hat{d}_0$ have two eigenvectors $\boldsymbol{a}_0$, $\boldsymbol{b}_0$ and two non-zero eigenvalues—coefficients before the corresponding dyad projectors. These eigenvalues exactly coincide with Fresnel's reflection and transmission coefficients (see, for example, [15]). Thus, operators $\hat{d}_0$ and $\hat{r}_0$ (31), (32) give us appropriate coefficients for reflection and transmission of TE- or TM-plane monochromatic wave at the isotropic media interface. One can obtain by direct calculation the equality

$$\hat{r}_0 + \hat{G} = \hat{d}_0, \tag{34}$$

which means that tangential components of strength vector of magnetic field are continuous on the boundaries in the main approximation. Taking into account the second equality (5), we can write

$$\hat{r}_1 = \hat{d}_1, \tag{35}$$

so it is sufficient to find only the first-order approximation for reflection operator. The further operation produces the formula

$$\hat{r}_1 = \frac{2}{\Lambda V}\left[-\frac{m_n\Lambda}{V}\Gamma_a\hat{\tau}_a - \frac{V}{m_n\Lambda}\Gamma_b\hat{\tau}_b + \lambda\left(\frac{\hat{\tau}_a}{m_n} - m_n\hat{\tau}_b\right)q^{\times}\right]. \tag{36}$$

With the help of (27) and (33) this expression is reduced to the following form:

$$\hat{r}_1 = \frac{\varepsilon_1 m_n \sin^2\varphi\cos^2\Theta_i}{m_n^-(\varepsilon_q m_n + m_n^-)^2}\hat{\tau}_a - \frac{\varepsilon_1 m_n(\sin^2\varphi(\cos^2\Theta_0 + \sin^2\Theta_i) - 1}{m_n^-(m_n^- + m_n)^2}\hat{\tau}_b$$
$$+ \frac{\varepsilon_1 m_n \sin 2\varphi q^{\times}\left(\frac{1}{m_n}\hat{\tau}_b - m_n\hat{\tau}_a\right)}{2(m_n^- + m_n)(\varepsilon_q m_n + m_n^-)} \tag{37}$$

or with proper designations:

$$\hat{r}_1 = r_{1a}\hat{\tau}_a + r_{1b}\hat{\tau}_b + \hat{r}_{1ab} \tag{38}$$

where r_{1a}, r_{1b} are scalar coefficients and $\hat{r}_{1ab}$ is the second-rank tensor. The initial tensors of reflection and transmissions $\hat{r}$ and $\hat{d}$ can be obtained by combining equations (30)—(33), (35), (38):

$$\hat{r} = r_a\hat{\tau}_a + r_b\hat{\tau}_b + \hat{r}_1, \quad \hat{d} = d_a\hat{\tau}_a + d_b\hat{\tau}_b + \hat{r}_1,$$
$$r_a = r_{0a} + r_{1a}, \quad r_b = r_{0b} + r_{1b}, \quad d_a = d_{0a} + r_{1a}, \quad d_b = d_{0b} + r_{1b}. \tag{39}$$

Our subsequent scheme includes the following procedures:

1. To deduce vector $\boldsymbol{H}_i$ (3) from the given vector amplitude of incident wave $\boldsymbol{E}$ and refraction vectors $\boldsymbol{m}_i$.

2. To cut vector amplitude $\boldsymbol{H}_i$ at the plane interface of two media (4), defined by the vector $\boldsymbol{q}$.

3. To calculate tangential component of reflected wave (or transmitted wave as it is needed) $\boldsymbol{H}_{rt}$ with the help of expressions (5), (39).

4. To restore vector strength of electric field of reflected wave $\boldsymbol{E}_r$ from the vector $\boldsymbol{H}_{rt}$ using the restoring operator $\hat{v}$ [3]

$$\boldsymbol{E}=\hat{v}\boldsymbol{H}_t,\quad \hat{v}=-\boldsymbol{q}^{\times}\hat{\gamma}+\frac{1}{\varepsilon_q}\boldsymbol{q}\otimes(\boldsymbol{a}+\boldsymbol{q}\hat{\varepsilon}\boldsymbol{q}^{\times}\hat{\gamma}). \tag{40}$$

Taking into accout that the first medium is isotropic and $\hat{\gamma}_r=-\hat{\gamma}$ (29), we may simplify (40):

$$\boldsymbol{E}_r=\hat{v}_r\boldsymbol{H}_{rt},\quad \hat{v}_r=m_n\boldsymbol{q}^{\times}\hat{\tau}_a+\frac{1}{m_n}\boldsymbol{q}^{\times}\hat{\tau}_b+\boldsymbol{q}\otimes\boldsymbol{a}. \tag{41}$$

Finally, we obtain the desired expression connecting the electric field of incident and reflected waves by uniting equations (3)–(5), (41)

$$\boldsymbol{E}_r=\hat{R}_E\boldsymbol{E}_i \tag{42}$$

where amplitude reflection operator for electric field is defined as

$$\hat{R}_E=\hat{v}_r\hat{r}\boldsymbol{m}_i^{\times}. \tag{43}$$

Substituting relations for $\hat{v}_r$ (41), $\hat{r}$ (39) and for tensor $\boldsymbol{m}_i^{\times}$ dual to the refraction vector of incident wave $\boldsymbol{m}_i^{\times}=\boldsymbol{b}^{\times}+m_n\boldsymbol{q}^{\times}$ into (43) gives us the final formula

$$\hat{R}_E=m_nr_ab(-\boldsymbol{a}_0^{\times})(\hat{\tau}_q+\hat{\tau}_b)-m_n^2r_a\hat{\tau}_b+r_ab^2\hat{\tau}_q-r_b\hat{\tau}_a-\sigma m_n\boldsymbol{q}^{\times}(\hat{\tau}_a-\hat{\tau}_b)$$
$$+\lambda\boldsymbol{b}^{\times}(\hat{\tau}_a+\hat{\tau}_q) \tag{44}$$

where

$$\sigma=\frac{\varepsilon_1m_n\sin2\varphi}{2(m_n+m_n^-)(\varepsilon_qm_n+m_n^-)}. \tag{45}$$

This expression is written in a very useful form for applied calculations because all terms are explicitly connected with the natural basis of boundary problem $(\boldsymbol{a}_0,\boldsymbol{b}_0,\boldsymbol{q})$.

3. Reflection configuration of nonlinear polarization spectroscopy detection scheme

Now, we try to apply the obtained results to reflection-transmission problem in the scheme of nonlinear polarization spectroscopy. A powerful normally incident pumping wave induces the anisotropy structure in nonlinear medium. This anisotropy is taken properly into account through the complex dielectric permittivity tensor $\hat{\varepsilon}$ (43). Thus, we use covariant expression (44) to derive the vector amplitude of reflected probe wave. Let obliquely incident plane wave $\boldsymbol{E}_0$ pass through a linear polarizer described by unit vector $\boldsymbol{U}_p$. So, we have the incident (at MLIA) wave in the form $\boldsymbol{E}_i=\hat{P}\boldsymbol{E}_0$, where dyad $\hat{P}=\boldsymbol{U}_p\otimes\boldsymbol{U}_p$ is related with the polarizer action. If the reflected beam

$$\boldsymbol{E}_r = \hat{R}_E \hat{P} \boldsymbol{E}_0 \tag{46}$$

is blocked by the crossed analyzer, described by dyad operator $A = \boldsymbol{U}_A \otimes \boldsymbol{U}_A$, where $\boldsymbol{U}_A$ is parallel to analyzer axis, we may write the detected field as

$$\boldsymbol{E} = \hat{A}\boldsymbol{E}_r = \hat{A}\hat{R}_E\hat{P}\boldsymbol{E}_0. \tag{47}$$

For definition let us take one of the characteristic polarizations of incident wave, for example, TE-wave of unit intensity. The polarizer transmits this wave without losses, which means that

$$\boldsymbol{E}_0 = \boldsymbol{a}_0, \quad \boldsymbol{U}_p = \boldsymbol{a}_0, \quad \boldsymbol{E}_i = \hat{P}\boldsymbol{E}_0 = (\boldsymbol{U}_p \boldsymbol{a}_0)\boldsymbol{U}_p = \boldsymbol{a}_0.$$

Then, from (42)–(44), it follows that

$$\begin{aligned}\boldsymbol{E}_r = \hat{R}_E \boldsymbol{a}_0 &= (-r_b \hat{\tau}_a - \sigma m_n \boldsymbol{q}^{\times} \hat{\tau}_a + \sigma \boldsymbol{b}^{\times} \hat{\tau}_a)\boldsymbol{a}_0 = -r_b \boldsymbol{a}_0 - \sigma m_n (\boldsymbol{q} \times \boldsymbol{a}_0) + \sigma(\boldsymbol{b} \times \boldsymbol{a}_0) \\ &= -r_b \boldsymbol{a}_0 - \sigma(m_n \boldsymbol{b}_0 + b\boldsymbol{q}).\end{aligned} \tag{48}$$

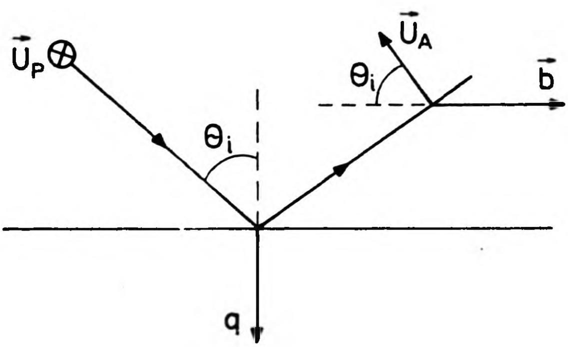

Fig. 2. Illustration to the projective dyad of the crossed analyser

Here, we took into consideration that $\hat{\tau}_b \boldsymbol{a}_0 = \hat{\tau_q} \boldsymbol{a}_0 = 0$, $\hat{\tau}_a \boldsymbol{a}_0 = \boldsymbol{a}_0$, $(\boldsymbol{q} \times \boldsymbol{a}_0) = \boldsymbol{b}_0$, $(\boldsymbol{b}_0 \times \boldsymbol{a}_0) = -\boldsymbol{q}$. Projective dyad of crossed analyzer has an axis

$$\boldsymbol{U}_A = -\cos\Theta_i \boldsymbol{b} - \sin\Theta_i \boldsymbol{q} = -m_n \boldsymbol{b}_0 - b\boldsymbol{q} \tag{49}$$

as one can see in Fig. 2. Therefore, the resulting electric field behind the analyzer is reduced from (47), (49)

$$\boldsymbol{E}_{ex} = \hat{A}\boldsymbol{E}_r = (\boldsymbol{U}_A \boldsymbol{E}_r)\boldsymbol{U}_A = (\sigma m_n^2 + \sigma b^2)\boldsymbol{U}_A = \sigma \boldsymbol{U}_A. \tag{50}$$

Finally, for the detected signal in our approximations, we have

$$I_{\mathrm{NPS}} = |\sigma|^2 = \left|\frac{\varepsilon_1}{2(m_n^- + m_n)(\varepsilon_q m_n + m_n^-)}\right|^2 m_n^2 \sin^2 2\varphi. \tag{51}$$

In (51), we took into account that ε_1 and m_n^- may be complex quantities due to absorbing properties of nonlinear medium.

Thus, we theoretically verified the principal possibility of using the reflection scheme in nonlinear polarization spectroscopy as it was noted for the first time in paper [16].

4. Concluding remarks

A very general method based on a wave operator formalism has been proposed to describe reflection at the interface between isotropic medium and MLIA. In the configuration in which both pump and probe beam copropagate onto the surface of an investigated sample, the polarization of reflected wave is related with nonlinear additions to the dielectric permittivity tensor. As a result, one can extract the spectroscopic data from the detected signal. On the other hand, the general theoretical expressions obtained seem to be very useful in the light of recent experimental research on nonlinear selective reflection [17], [18].

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