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ORDER, DISORDER, AND PHASE TRANSITION IN CONDENSED SYSTEM

Magneto-Optical Parameter *Q* for Structures with Uniaxial Optical Anisotropy

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Abstract—This paper is devoted to the development of reflection magneto-optical ellipsometry. We have solved the inverse problem for structures with uniaxial optical anisotropy: have determined the reflection coefficients for the ambient–sample interface, and have derived an analytic expression for magneto-optical parameter *Q* proportional to the magnetization. This expression makes it possible to determine parameter *Q* exclusively from experimental data obtained using magneto-optical ellipsometry. We present a detailed algorithm for performing experiment on determining the dielectric tensor in the transverse geometry.

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1. INTRODUCTION. FORMULATION OF THE PROBLEM

Magneto-optical ellipsometry (ME) is characterized by a high sensitivity to the magnetic state as well as to optical properties of a reflecting surface. This method is widely used and is very convenient for monitoring optical, structural, magnetic, and magneto-optical (MO) properties of nanostructured materials. Because of the cumbersome mathematical apparatus of ME, most measurements are made for performing simple analysis of magnetic characteristics from the field dependences of ellipsometric angles or for studying simple isotropic and homogeneous media. However, the structure of many materials (especially, in the film and disperse states) depends on direction; therefore, such materials exhibit uniaxial optical anisotropy. The optical properties of such materials in the plane of the film and across it are different [1, 2]. Examples of such structures are photonic crystals [3], polymer films [4], and oriented arrays of carbon nanotubes [5, 6]. In recent years, optical properties of anisotropic 2D systems MXnes [7, 8] and MAX phases [9–11] have been studied actively (ab initio calculations). When magneto-optical properties of such a structure are considered, in addition to existing optical anisotropy, it is necessary to take into account induced anisotropy [12] associated with the application of an external magnetic field, which inevitably complicates the calculation and analysis of the total dielectric tensor. For this reason, such an approach receives little attention in the literature. It has been used predominantly in publications [13–15] based on the formalism of 4 × 4 matrices [16], which has been developed well for ME, but is not applicable in all cases because for determining all matrix elements, measurements must be performed in different geometries; therefore, the sample must be rotated.

We have developed an original method for determining all components of the dielectric tensor for thin magnetic layers from the ME data from optically isotropic bulk and multilayer structures [17–21], which combines classical conventional ellipsometric measurements of ellipsometric angles ψ_0 and Δ_0 without applying an external magnetic field and ellipsometric measurements with magnetization reversal of the sample in an external magnetic field (measurement of $\delta\psi$ and $\delta\Delta$) in the transverse configuration. Such measurements can be made for various angles of light incidence and for various wavelengths of incident radiation. The solution of the inverse problem includes the determination of physical quantities such as the dielectric tensor components from the set of data on ψ_0, Δ_0, $\delta\psi$, and $\delta\Delta$ by analyzing the system of basic equations of ellipsometry:

$$\begin{gathered}\tan(\psi_0)\exp(i\Delta_0) = \frac{R_{p0}}{R_{s0}},\\ \tan(\psi_0 + \delta\psi)\exp(i(\Delta_0 + \delta\Delta)) = \frac{R_{p0} + R_{p1}}{R_{s0} + R_{s1}},\end{gathered} \tag{1}$$

where subscript "0" corresponds to measurements performed in zero external magnetic field and sub-

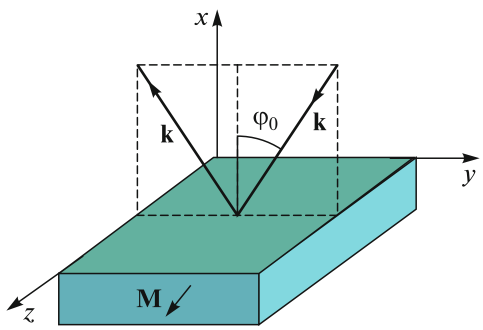

Fig. 1. Geometry of the transverse magneto-optical Kerr effect.

script "1" corresponds to measurements with the applied magnetic field. Subscripts "s" and "p" correspond to the s and p polarizations of light.

In this study, we propose to extend this approach to magnetic optically anisotropic materials; namely, we first solve the inverse ME problem for semi-infinite structures with uniaxial optical anisotropy. Examples of such materials are thick atomic-layer MAX films [22], columnar ferromagnetic films [23–25], and various low-symmetry magneto-optically active bulk crystals [26–28]. The new approach provides information on MO properties of the sample from ME measurements without rotating the sample or the electromagnet producing the external magnetic field.

Let us consider the geometry of ME reflection measurements, which corresponds to the transverse magneto-optical Kerr effect, TMOK in short (Fig. 1). In the case of the transverse configuration, the magnetization lies in the plane of the sample. We choose the direction of the z axis being parallel to the direction of magnetization, as usual [12, 17–22, 29, 30]. Thus, the yz plane is the boundary plane of the reflection surface and xy is the plane of incidence.

In the general case of optically anisotropic media, the dielectric tensor is represented as follows [13, 14]:

$$\varepsilon^{MO} = \begin{bmatrix} \varepsilon_x & -iQ_zM_z & -iQ_yM_y \\ iQ_zM_z & \varepsilon_y & -iQ_xM_x \\ iQ_yM_y & iQ_xM_x & \varepsilon_z \end{bmatrix}, \quad (2)$$

where $Q = (Q_x, Q_y, Q_z)$ is the magneto-optical parameter, which is independent of magnetization, and $M = (M_x, M_y, M_z)$ is the magnetization.

On the other hand, magnetization is traditionally not separated in the dielectric tensor components; instead, it is assumed that Q is the magneto-optical parameter (also known as the Voigt vector) proportional to the magnetization [12, 29–34]. In this case, for isotropic media, when all diagonal tensor components are identical and equal to ε_0, the off-diagonal dielectric tensor components are equal to the product of the diagonal component and the Voigt vector component:

$$\varepsilon_{ij} = -i\varepsilon_0 Q_k, \quad (3)$$

where i, j, k take value of x, y, z. The geometry of the ME problem being solved here implies that the refractive indices for a uniaxial optical anisotropic semi-infinite ferromagnetic structure in the yz plane are identical:

$$N_x \neq N_y = N_z, \quad (4)$$

which means that the diagonal components of the dielectric tensor in the sample plane are also identical:

$$\varepsilon_{xx} \neq \varepsilon_{yy} = \varepsilon_{zz}. \quad (5)$$

Therefore, taking publications [12, 14] into account, the dielectric tensor in the case of uniaxial anisotropy can be written as

$$\varepsilon = \begin{bmatrix} \varepsilon_{xx} & -i\varepsilon_{zz}Q & 0 \\ i\varepsilon_{zz}Q & \varepsilon_{zz} & 0 \\ 0 & 0 & \varepsilon_{zz} \end{bmatrix}, \quad (6)$$

where $Q = Q_1 - iQ_2$ is the complex magneto-optical parameter, which is proportional to the magnetization lying in the plane of the film, $\varepsilon_{xx} = \varepsilon'_{xx} - i\varepsilon''_{xx}$, and $\varepsilon_{zz} = \varepsilon'_{zz} - i\varepsilon''_{zz}$ (real parts are marked by prime and imaginary parts, by double prime). In this case, the effects quadratic in magnetization are disregarded.

The reflection coefficients for isotropic structures, which take into account the magneto-optical response, are well known [12, 29]:

$$R_s = \frac{N_0\cos(\varphi_0) - N_1\cos(\varphi_1)}{N_0\cos(\varphi_0) + N_1\cos(\varphi_1)}, \quad (7)$$

$$R_p = \frac{N_1\cos(\varphi_0) - N_0\cos(\varphi_1)}{N_1\cos(\varphi_0) + N_0\cos(\varphi_1)} - iQ\frac{N_0^2\sin(2\varphi_0)}{(N_1\cos(\varphi_0) + N_0\cos(\varphi_1))^2}, \quad (8)$$

where N_0 is the complex refractive index of the ambient, N_1 is the complex refractive index of the ferromagnetic material, and φ_0 is the angle of incidence of light on the sample. These expressions are mainly used in the method for determining the dielectric tensor components for isotropic samples, which has been developed in [17–20] and based on the Jones matrices.

It is possible to operate with anisotropic media, still using the Jones matrices and Fresnel coefficients, when the off-diagonal elements of the Jones matrix for the reflection coefficients equal zero [35, 36]:

$$r_{ps} = r_{sp} = 0. \quad (9)$$

In our geometry, this expression is valid because in the case of uniaxial optical anisotropy of a bulk sample, the condition that the optical axis is parallel or perpendicular to the light plane of incidence is sufficient for its fulfillment [35, 36].

The method for obtaining of the reflection coefficients for anisotropic systems in the chosen geometry (see Fig. 1) was described in [36]. These coefficients can be presented in the following way:

$$r_{ss} = \frac{N_0\cos(\varphi_0) - N_z\cos(\varphi_{ts})}{N_0\cos(\varphi_0) + N_z\cos(\varphi_{ts})}, \tag{10}$$

$$r_{pp} = \frac{N_y\cos(\varphi_0) - N_0\cos(\varphi_{tp})}{N_y\cos(\varphi_0) + N_0\cos(\varphi_{tp})}, \tag{11}$$

where angles φ_{ts} and φ_{tp} are the angles of refraction, which are defined via

$$\cos(\varphi_{ts}) = \frac{\sqrt{N_z^2 - N_0^2\sin^2(\varphi_0)}}{N_z}, \tag{12}$$

$$\cos(\varphi_{tp}) = \frac{\sqrt{N_x^2 - N_0^2\sin^2(\varphi_0)}}{N_x}. \tag{13}$$

In [36], the minus sign in relation $N = n - ik$ has been taken into account from the very outset, which is traditional for the representation of the refraction index in ellipsometry and ME. In isotropic limit $N_x = N_y = N_1$, Eqs. (10) and (11) yield the standard Fresnel coefficients [35].

However, reflection coefficients (10) and (11) in [36] do not take into account the effect of the external magnetic field and are insufficient for analyzing the MO properties of the sample. We must supplement the familiar expressions for the reflection coefficients of anisotropic media with expressions taking into account the magneto-optical response, connect them with parameters measured using ME technique, and find the expression for obtaining information on MO parameter $Q = Q_1 - iQ_2$ and on total dielectric tensor (6).

2. TAKING INTO ACCOUNT FOR THE MAGNETO-OPTICAL RESPONSE WHILE CALCULATING THE REFLECTION COEFFIIENTS FOR THE INTERFACE BETWEEN THE AMBIENT AND AN OPTICALLY ANISOTROPIC UNIAXIAL BULK SAMPLE WITH

To add the magneto-optical response to expressions (10) and (11), let us follow the approach used by Sokolov and Krinchik [12, 29], who considered the situation at the interface between two isotropic media. To take into account the TMOKE while analyzing the reflection at the interface between an ambient and an optically anisotropic bulk crystal, the same steps are to be taken as in the case when it is considered at the interface with an isotropic sample. All expressions presented below are valid for visible spectral range and have been obtained assuming that $\mu \approx \mu_0 \approx 1$, and MO parameter $Q \ll 1$ [12, 30, 32, 37].

We use tensor (6) and solve the Maxwell equations with the following relations between the electric field intensity and electric displacement:

$$\begin{aligned} D_x &= \varepsilon_{xx}E_x - i\varepsilon_{zz}QE_y, \\ D_y &= i\varepsilon_{zz}QE_x + \varepsilon_{zz}E_y, \\ D_z &= \varepsilon_{zz}E_z. \end{aligned} \tag{14}$$

We seek the solution in the form of a plane inhomogeneous wave propagating in a magnetic medium:

$$E = E_0\exp\left(i\omega\left(t - \frac{\alpha^*x + \beta^*y + \gamma^*z}{v^*}\right)\right), \tag{15}$$

where α^*, β^*, and γ^* are direction cosines and v^* is the velocity of the wave propagating in the medium. This gives the following system of equations:

$$\begin{aligned} &\varepsilon_{xx}E_x - i\varepsilon_{zz}QE_y \\ &= N_x^2[E_x - \alpha^*(\alpha^*E_x + \beta^*E_y + \gamma^*E_z)], \\ &i\varepsilon_{zz}QE_x + \varepsilon_{zz}E_y \\ &= N_y^2[E_y - \beta^*(\alpha^*E_x + \beta^*E_y + \gamma^*E_z)], \\ &\varepsilon_{zz}E_z \\ &= N_z^2[E_z - \gamma^*(\alpha^*E_x + \beta^*E_y + \gamma^*E_z)]. \end{aligned} \tag{16}$$

After a number of transformations (see Appendix A) with account for relations (4) and (5), we obtain

$$\begin{aligned} &(\varepsilon_{xx}\varepsilon_{zz}\cos(\varphi_{tp}) + i\varepsilon_{xx}\varepsilon_{zz}Q\sin(\varphi_{tp}))E_x \\ &+ (\varepsilon_{xx}\varepsilon_{zz}\sin(\varphi_{tp}) - i\varepsilon_{zz}^2Q\cos(\varphi_{tp}))E_y = 0. \end{aligned} \tag{17}$$

With account for the coordinate system introduced here, the boundary conditions for the s polarization are

$$E_{0is} + E_{0rs} = E_{d3} = E_{ts}, \tag{18}$$

$$N_0\cos(\varphi_0)(E_{0is} - E_{0rs}) = N_z\cos(\varphi_{ts})E_{ts}, \tag{19}$$

while for the p polarization, they are

$$\cos(\varphi_0)(E_{0ip} - E_{0rp}) = E_{d2}, \tag{20}$$

$$\begin{aligned} &N_0(E_{0ip} + E_{0rp}) \\ &= N_y(\cos(\varphi_{tp})E_{d2} - \sin(\varphi_{tp})E_{d1}), \end{aligned} \tag{21}$$

where subscripts i, r, and d on E correspond to the incident wave, reflected wave, and the wave transmitted through the medium, respectively; E_{d1}, E_{d2}, and E_{d3} are the x, y, and z components of the electric field intensity amplitude of the transmitted wave, respectively [12, 29].

The resulting reflection coefficient $r_{ssTMOKE}$ coincides with reflection coefficient (10) for anisotropic media in zero magnetic field,

$$r_{sTMOKE} = \frac{N_0\cos(\varphi_0) - N_z\cos(\varphi_{ts})}{N_0\cos(\varphi_0) + N_z\cos(\varphi_{ts})}, \tag{22}$$

accordingly, for the s polarization, the effect of the magnetic field on the reflection coefficient for the TMOKE is absent not only for isotropic, but also for anisotropic systems:

$$R_{s1} = 0. \tag{23}$$

We obtain the reflection coefficient for the p polarization from expressions (20) and (21), which are supplemented with the expression connecting E_{d1} and E_{d2} (see Appendix A):

$$r_{ppTMOKE} = \frac{E_{0rp}}{E_{0ip}} = \frac{\cos(\varphi_0) - FN_0}{\cos(\varphi_0) + FN_0}, \tag{24}$$

where

$$F = \frac{\cos(\varphi_{tp}) + iQ\sin(\varphi_{tp})}{N_z(1 + i(1 - \varepsilon_{zz}/\varepsilon_{xx})Q\sin(\varphi_{tp})\cos(\varphi_{tp}))}. \tag{25}$$

Since for taking into account the MO contribution, the changes in ellipsometric parameters are measured in the experiment (i.e., there is the change in the reflection coefficient as compared to expression (11) characterizing the behavior of light in zero magnetic field), it would be interesting to obtain an explicit expression describing the variation of reflection coefficient R_{p1}. For this purpose, we write expression (24) for $r_{ppTMOKE}$ as the sum of r_{pp} disregarding the external magnetic field (i.e., expression (11)) and the term responsible for the contribution of the TMOK (see Appendix A). Therefore, we obtain the reflection coefficient for the p polarization, where the MO response in the geometry of the TMOK is considered:

$$\begin{aligned} r_{ppTMOKE} &= \frac{N_z\cos(\varphi_0) - N_0\cos(\varphi_{tp})}{N_z\cos(\varphi_0) + N_0\cos(\varphi_{tp})} \\ &- iQ\frac{2N_0N_z\sin(\varphi_{tp})\cos(\varphi_0)(1 - (1 - N_z^2/N_x^2)\cos^2(\varphi_{tp}))}{(N_z\cos(\varphi_0) + N_0\cos(\varphi_{tp}))^2}. \end{aligned} \tag{26}$$

Using this expression, we can derive the expression for the reflection coefficient for the interface with the isotropic crystal, assuming that $N_x = N_z = N_1$ and $\varphi_{tp} = \varphi_1$:

$$\begin{aligned} r_p &= \frac{N_1\cos(\varphi_0) - N_0\cos(\varphi_1)}{N_1\cos(\varphi_0) + N_0\cos(\varphi_1)} \\ &- iQ\frac{2N_0N_1\sin(\varphi_1)\cos(\varphi_0)}{(N_1\cos(\varphi_0) + N_0\cos(\varphi_1))^2}. \end{aligned} \tag{27}$$

Using the Snell law

$$N_1\sin(\varphi_1) = N_0\sin(\varphi_0) \tag{28}$$

we obtain familiar expression (8) for the reflection coefficient in the p-polarization at the interface between two isotropic media [12, 38].

3. ANALYTICAL CALCULATION OF MAGNETO-OPTICAL PARAMETER Q

As shown in our previous works on investigation of MO properties of isotropic structures [20, 39], in the case when the magnetic field contribution to the reflection coefficients is small, one can use in calculations small parameters such as ratios of the magnetic to nonmagnetic parts of the reflection coefficient for the p-polarization:

$$\alpha = R'_{p1}/R'_{p0}, \tag{29}$$

$$\beta = R''_{p1}/R''_{p0}, \tag{30}$$

where subscripts "0" and "1" correspond to the measurements taken in zero magnetic field and in the presence of an external magnetic field (TMOKE), respectively. These small parameters are expressed in terms of experimentally measured ellipsometric (ψ_0, Δ_0) and magneto-ellipsometric ($\delta\psi$, $\delta\Delta$) parameters in the following way [20]:

$$\alpha \approx \frac{\delta\psi(1 + \tan^2(\psi_0))}{\tan(\psi_0)} + \frac{R''_{p0}}{R'_{p0}}\delta\Delta, \tag{31}$$

$$\beta \approx \frac{\delta\psi(1 + \tan^2(\psi_0))}{\tan(\psi_0)} - \frac{R'_{p0}}{R''_{p0}}\delta\Delta, \tag{32}$$

i.e., the reflection coefficient for the p-polarization can be written as follows:

$$R_p = R_{p0} + R_{p1} = R'_{p0} - iR''_{p0} + R'_{p1} - iR''_{p1}, \tag{33}$$

$$R_{p0} = R'_{p0} - iR''_{p0}, \tag{34}$$

$$R_{p1} = R'_{p1} - iR''_{p1} = \alpha R'_{p0} - i\beta R''_{p0}. \tag{35}$$

Let us compare expressions (34) and (35) with expression (26) derived above:

$$R'_{p0} - iR''_{p0} = \frac{N_z\cos(\varphi_0) - N_0\cos(\varphi_{tp})}{N_z\cos(\varphi_0) + N_0\cos(\varphi_{tp})}, \tag{36}$$

$$\alpha R'_{p0} - i\beta R''_{p0} = -iQ\frac{2N_0N_z\sin(\varphi_{tp})\cos(\varphi_0)(1 - (1 - N_z^2/N_x^2)\cos^2(\varphi_{tp}))}{(N_z\cos(\varphi_0) + N_0\cos(\varphi_{tp}))^2}. \tag{37}$$

Then we take into account Eqs. (31) and (32) and express Q (see Appendix B):

$$Q = \frac{N_0^2\cos^2(\varphi_{tp}) - N_z^2\cos^2(\varphi_0)}{2N_0N_z\sin(\varphi_{tp})\cos(\varphi_0)(1 - (1 - N_z^2/N_x^2)\cos^2(\varphi_{tp}))}\left(\delta\Delta - i\frac{\delta\psi(1 + \tan^2(\psi_0))}{\tan(\psi_0)}\right). \tag{38}$$

Thus, we can calculate analytically the magneto-optical parameter for a uniaxial anisotropic bulk crystal from the results of ME measurements performed in the TMOK configuration.

If we set $N_x = N_z = N_1$ and $\varphi_{tp} = \varphi_1$ in expression (37), the resulting expression is transformed to

$$Q = \frac{N_0^2\cos^2(\varphi_1) - N_1^2\cos^2(\varphi_0)}{2N_0N_1\sin(\varphi_1)\cos(\varphi_0)} \times\left(\delta\Delta - i\frac{\delta\psi(1+\tan^2(\psi_0))}{\tan(\psi_0)}\right). \quad (39)$$

Using the Snell law (28), we obtain the following expression coinciding with the one for the MO parameter for an isotropic crystal [20]:

$$Q = \frac{N_0^2\cos^2(\varphi_1) - N_1^2\cos^2(\varphi_0)}{N_0^2\sin(2\varphi_0)} \times\left(\delta\Delta - i\frac{\delta\psi(1+\tan^2(\psi_0))}{\tan(\psi_0)}\right). \quad (40)$$

4. DISCUSSION OF RESULTS

As follows from expression (37) derived above, for calculating all components of dielectric tensor (6), it is sufficient to have information on

(i) the angle of incidence φ_0,

(ii) the refractive index N_0 of the ambient,

(iii) the refractive indices of the anisotropic structure in the plane of the sample ($N_y = N_z$) and perpendicular to it (N_x),

(iv) the ellipsometric parameter ψ_0 measured in zero magnetic field, and

(v) the magneto-ellipsometric parameters

$$\delta\psi = \psi(+H) - \psi(-H), \quad \delta\Delta = \Delta(+H) - \Delta(-H),$$

measured in the transverse configuration of the magneto-optical Kerr effect, where $\pm H$ is the external magnetic field on the sample.

Experimental and mathematical approaches to measuring the refractive indices of an anisotropic (including opaque) film of an arbitrary thickness by means of conventional ellipsometry have been developed long ago and require no additional explanations [40–42]. The realization of a specific approach can depend on the experimental conditions and on the available instrumental base of the experimenter.

It should be borne in mind that the proposed method for finding out the MO parameter Q is limited not only by the features of the model of a homogeneous semi-infinite medium and the geometry of the TMOKE, but also by the condition of smallness of the MO contribution to reflection coefficient R_p, ensuring the smallness of parameters α and β [20]. The latter condition can be violated in the case of closeness to the Brewster angle (especially for weakly absorbing samples). Therefore, in our opinion, the approach involving multiangle ellipsometric measurements appears to be most reliable in view of the simplicity of the optical scheme and the possibility to avoid the closeness to the Brewster angle. The sample must have a smooth reflecting surface, which is opaque in the spectral range used in experiments, and as thin as possible nonferromagnetic oxide layer on the surface. In this case, the algorithm of a ME experiment can be described as follows.

(1) We measure the spectral dependences of ψ_0 and Δ_0 in zero magnetic field for arbitrary angle φ_0. Then we calculate the spectral dependence of the real component of the Brewster angle for the sample in the isotropic semi-infinite medium approximation. We choose at least two angles of incidence φ_0, which are accessible for setting on the magnetoellipsometer and do not coincide with the values of the Brewster angle in the required spectral range.

(2) For one of the chosen angles φ_0, we measure field dependences of ψ_0 and Δ_0 at a fixed wavelength corresponding to the maximal value of the signal-to-noise ratio for the magnetoellipsometer.

(3) From the field dependences of ψ_0 and Δ_0, we choose the best values of magnetic field $\pm H$ for further spectral ME measurements (i.e., if the sample is ferromagnetic, it is expedient to choose the value of H from the condition of ferromagnetic saturation of the sample).

(4) For the first chosen angle φ_0, we measure the spectral dependences of ψ_0 and Δ_0 of the demagnetized sample and the changes in $\delta\psi$ and $\delta\Delta$ during the magnetization reversal of the sample in fields $\pm H$.

(5) We set the second chosen angle of incidence φ_0 and measure the spectral dependences of ψ_0 and Δ_0 for the demagnetized sample.

(6) Using numerical methods, we obtain complex values of N_x and $N_z = N_y$ for the demagnetized sample, from which Q is calculated using expression (37).

Having determined the values of the MO parameter Q and refractive indices N_x, N_y, and N_z, we can calculate all components of dielectric tensor (6).

5. CONCLUSIONS

Thus, we have derived expressions for the reflection coefficients in the p- and s-polarizations for the interface between the ambient and the sample with uniaxial optical anisotropy, which take into account the magneto-optical response in the geometry of the transverse magneto-optical Kerr effect. Analytical calculation of the magneto-optical parameter Q for the chosen experimental geometry has been proposed. The scheme of magneto-ellipsometric measurements for obtaining all components of the dielectric tensor has been demonstrated for materials with uniaxial anisotropy, e.g., MAX phases.

APPENDIX A

Below, we describe below the derivation of the reflection coefficients at the interface between the ambient and a bulk medium with uniaxial optical anisotropy and the dielectric tensor

$$\varepsilon = \begin{bmatrix} \varepsilon_{xx} & -i\varepsilon_{zz}Q & 0 \\ i\varepsilon_{zz}Q & \varepsilon_{zz} & 0 \\ 0 & 0 & \varepsilon_{zz} \end{bmatrix}. \quad \text{(A.1)}$$

All expressions given below are valid for the visible spectral range and have been obtained under the assumption that $\mu \approx \mu_0 \approx 1$ and the magneto-optical parameter $Q \ll 1$ [12].

We seek the solutions to the Maxwell equations

$$\begin{aligned} D_x &= \varepsilon_{xx}E_x - i\varepsilon_{zz}QE_y, \\ D_y &= i\varepsilon_{zz}QE_x + \varepsilon_{zz}E_y, \\ D_z &= \varepsilon_{zz}E_z \end{aligned} \quad \text{(A.2)}$$

in the form of a plane inhomogeneous wave propagating in a magnetic medium:

$$E = E_0 \exp\left(i\omega\left(t - \frac{\alpha^*x + \beta^*y + \gamma^*z}{v^*}\right)\right), \quad \text{(A.3)}$$

where α^*, β^*, and γ^* are the direction cosines and v^* is the velocity of wave propagation in the medium. This gives the following system of equations:

$$\begin{aligned} &\varepsilon_{xx}E_x - i\varepsilon_{zz}QE_y \\ &= N_x^2[E_x - \alpha^*(\alpha^*E_x + \beta^*E_y + \gamma^*E_z)], \\ &i\varepsilon_{zz}QE_x + \varepsilon_{zz}E_y \\ &= N_y^2[E_y - \beta^*(\alpha^*E_x + \beta^*E_y + \gamma^*E_z)], \\ &\varepsilon_{zz}E_z = N_z^2[E_z - \gamma^*(\alpha^*E_x + \beta^*E_y + \gamma^*E_z)]. \end{aligned} \quad \text{(A.4)}$$

We then multiply the lines of this system by $N_y^2N_z^2\alpha^*$, $N_z^2N_x^2\beta^*$, and $N_x^2N_y^2\gamma^*$, respectively, sum them, take into account the fact that $N_y = N_z$, $\varepsilon = N^2$, $\alpha^* = \cos\varphi_{tp}$, $\beta^* = \sin\alpha_{tp}$, and $\gamma^* = 0$, and finally, we obtain

$$\begin{aligned} &(\varepsilon_{xx}E_x - i\varepsilon_{zz}QE_y)\cos(\varphi_{tp})\varepsilon_{zz} \\ &+ (i\varepsilon_{zz}QE_x + \varepsilon_{zz}E_y)\sin(\varphi_{tp})\varepsilon_{xx} = 0, \end{aligned} \quad \text{(A.5)}$$

$$\begin{aligned} &(\varepsilon_{xx}\varepsilon_{zz}\cos(\varphi_{tp}) + i\varepsilon_{xx}\varepsilon_{zz}Q\sin(\varphi_{tp}))E_x \\ &+ (\varepsilon_{xx}\varepsilon_{zz}\sin(\varphi_{tp}) - i\varepsilon_{zz}^2Q\cos(\varphi_{tp}))E_y = 0. \end{aligned} \quad \text{(A.6)}$$

Given the chosen system of coordinates (see Fig. 1), we obtain the boundary conditions for the s-polarization:

$$E_{0is} + E_{0rs} = E_{d3} = E_{ts}, \quad \text{(A.7)}$$

$$N_0\cos(\varphi_0)(E_{0is} - E_{0rs}) = N_z\cos(\varphi_{ts})E_{ts}, \quad \text{(A.8)}$$

while for the p-polarization, we get

$$\cos(\varphi_0)(E_{0ip} - E_{0rp}) = E_{d2}, \quad \text{(A.9)}$$

$$\begin{aligned} &N_0(E_{0ip} + E_{0rp}) \\ &= N_y(\cos(\varphi_{tp})E_{d2} - \sin(\varphi_{tp})E_{d1}), \end{aligned} \quad \text{(A.10)}$$

where subscripts i, r, and d on E correspond to the incident, reflected, and transmitted wave, respectively, and E_{d1}, E_{d2}, and E_{d3} are the x, y, and z components of the electric field intensity amplitude of the transmitted wave, respectively [12, 29].

Consequently, for the s-polarization, the reflection coefficient for the case of TMOKE is

$$r_{ssTMOKE} = \frac{N_0\cos(\varphi_0) - N_z\cos(\varphi_{ts})}{N_0\cos(\varphi_0) + N_z\cos(\varphi_{ts})}. \quad \text{(A.11)}$$

We obtain the reflection coefficient for the p-polarization for TMOKE from expressions (A.9) and (A.10), which must be supplemented with the relation connecting E_{d1} and E_{d2}. The required relation can be obtained from (A.6):

$$E_{d2} = \frac{(\varepsilon_{xx}\cos(\varphi_{tp}) + i\varepsilon_{xx}Q\sin(\varphi_{tp}))E_{d1}}{i\varepsilon_{zz}Q\cos(\varphi_{tp}) - \varepsilon_{xx}\sin(\varphi_{tp})}. \quad \text{(A.12)}$$

Accordingly, we solve the following system of three equations:

$$\begin{aligned} &E_{d2} = \frac{(\varepsilon_{xx}\cos(\varphi_{tp}) + i\varepsilon_{xx}Q\sin(\varphi_{tp}))E_{d1}}{i\varepsilon_{zz}Q\cos(\varphi_{tp}) - \varepsilon_{xx}\sin(\varphi_{tp})}, \\ &\cos(\varphi_0)(E_{0ip} - E_{0rp}) = E_{d2}, \\ &N_0(E_{0ip} + E_{0rp}) = N_y(\cos(\varphi_{tp})E_{d2} - \sin(\varphi_{tp})E_{d1}). \end{aligned} \quad \text{(A.13)}$$

Expressing E_{d1} from the first equation of this system and substituting it into the third equation, we obtain

$$N_0E_{0ip} + N_0E_{0rp} = N_yE_{d2}\left(\cos(\varphi_{tp}) - \sin(\varphi_{tp})\frac{i\varepsilon_{zz}Q\cos(\varphi_{tp}) - \varepsilon_{xx}\sin(\varphi_{tp})}{\varepsilon_{xx}\cos(\varphi_{tp}) + i\varepsilon_{xx}Q\sin(\varphi_{tp})}\right). \quad \text{(A.14)}$$

This gives the following expression for E_{d2}:

$$E_{d2} = \frac{(N_0E_{0ip} + N_0E_{0rp})(\varepsilon_{xx}\cos(\varphi_{tp}) + i\varepsilon_{xx}Q\sin(\varphi_{tp}))}{N_y\varepsilon_{xx} + iN_yQ\sin(\varphi_{tp})\cos(\varphi_{tp})(\varepsilon_{xx} - \varepsilon_{zz})}. \quad \text{(A.15)}$$

We now equate resulting expression (A.15) to the left-hand side of the second equation in system (A.13):

$$\cos(\varphi_0)(E_{0ip} - E_{0rp}) = \frac{(N_0E_{0ip} + N_0E_{0rp})(\varepsilon_{xx}\cos(\varphi_{tp}) + i\varepsilon_{xx}Q\sin(\varphi_{tp}))}{N_y\varepsilon_{xx} + iN_yQ\sin(\varphi_{tp})\cos(\varphi_{tp})(\varepsilon_{xx} - \varepsilon_{zz})}. \quad \text{(A.16)}$$

Since we seek the expression for reflection coefficient $r_p = E_{0rp}/E_{0ip}$, it is convenient to introduce notation

$$F = \frac{\varepsilon_{xx}\cos(\varphi_{tp}) + i\varepsilon_{xx}Q\sin(\varphi_{tp})}{N_y\varepsilon_{xx} + iN_yQ\sin(\varphi_{tp})\cos(\varphi_{tp})(\varepsilon_{xx} - \varepsilon_{zz})}. \quad (A.17)$$

Considering that $N_y = N_z$, we divide the numerator and denominator of F by ϵ_{xx}:

$$F = \frac{\cos(\varphi_{tp}) + iQ\sin(\varphi_{tp})}{N_z(1 + i(1 - \varepsilon_{zz}/\varepsilon_{xx})Q\sin(\varphi_{tp})\cos(\varphi_{tp}))}. \quad (A.18)$$

Expression (A.16) takes the form

$$\begin{aligned} E_{0ip}\cos(\varphi_0) - E_{0rp}\cos(\varphi_0) \\ = FN_0E_{0ip} + FN_0E_{0rp}, \end{aligned} \quad (A.19)$$

$$E_{0ip}(\cos(\varphi_0) - FN_0) = E_{0rp}(\cos(\varphi_0) + FN_0). \quad (A.20)$$

Thus, we can see that the reflection in the p-polarization is given by

$$r_{ppTMOKE} = \frac{E_{0rp}}{E_{0ip}} = \frac{\cos(\varphi_0) - FN_0}{\cos(\varphi_0) + FN_0}. \quad (A.21)$$

In this expression, we single out two terms, R_{p0} and R_{p1}, where nonmagnetic term R_{p0} is defined by expression (11), while R_{p1} is responsible for the TMOKE contribution:

$$\begin{aligned} r_{ppTMOKE} &= \left(\cos(\varphi_0) - \frac{N_0}{N_z}\frac{\cos(\varphi_{tp}) + iQ\sin(\varphi_{tp})}{1 + i(1 - \varepsilon_{zz}/\varepsilon_{xx})Q\sin(\varphi_{tp})\cos(\varphi_{tp})}\right) \\ &\quad / \left(\cos(\varphi_0) + \frac{N_0}{N_z}\frac{\cos(\varphi_{tp}) + iQ\sin(\varphi_{tp})}{1 + i(1 - \varepsilon_{zz}/\varepsilon_{xx})Q\sin(\varphi_{tp})\cos(\varphi_{tp})}\right), \end{aligned} \quad (A.22)$$

$$\begin{aligned} r_{ppTMOKE} &= \left[N_z\cos(\varphi_0)\left(1 + i\left(i - \frac{\varepsilon_{zz}}{\varepsilon_{xx}}\right)Q\sin(\varphi_{tp})\cos(\varphi_{tp})\right) - N_0\cos(\varphi_{tp}) - iN_0Q\sin(\varphi_{tp})\right] \\ &\quad / \left[N_z\cos(\varphi_0)\left(1 + i\left(i - \frac{\varepsilon_{zz}}{\varepsilon_{xx}}\right)Q\sin(\varphi_{tp})\cos(\varphi_{tp})\right) + N_0\cos(\varphi_{tp}) + iN_0Q\sin(\varphi_{tp})\right], \end{aligned} \quad (A.23)$$

$$\begin{aligned} r_{ppTMOKE} &= \left[N_z\cos(\varphi_0) - N_0\cos(\varphi_{tp}) + i\left(1 - \frac{\varepsilon_{zz}}{\varepsilon_{xx}}\right)Q\sin(\varphi_{tp})\cos(\varphi_{tp})N_z\cos(\varphi_0) - iN_0Q\sin(\varphi_{tp})\right] \\ &\quad / \left[N_z\cos(\varphi_0) + N_0\cos(\varphi_{tp}) + i(1 - \varepsilon_{zz}/\varepsilon_{xx})Q\sin(\varphi_{tp})\cos(\varphi_{tp})N_z\cos(\varphi_0) + iN_0Q\sin(\varphi_{tp})\right], \end{aligned} \quad (A.24)$$

$$\begin{aligned} r_{ppTMOKE} &= \left[(N_z\cos(\varphi_0) - N_0\cos(\varphi_{tp})) - iQ\sin(\varphi_{tp})\left(N_0 - \left(1 - \frac{\varepsilon_{zz}}{\varepsilon_{xx}}\right)\cos(\varphi_{tp})N_z\cos(\varphi_0)\right)\right] \\ &\quad / \left[(N_z\cos(\varphi_0) + N_0\cos(\varphi_{tp}))\left(1 + iQ\sin(\varphi_{tp})\frac{(1 - \varepsilon_{zz}/\varepsilon_{xx})\cos(\varphi_{tp})N_z\cos(\varphi_0) + N_0}{N_z\cos(\varphi_0) + N_0\cos(\varphi_{tp})}\right)\right]. \end{aligned} \quad (A.25)$$

Let us multiply the numerator and denominator of expression (A.25) by the complex conjugate of the second factor in the denominator, namely, by

$$\left(1 - iQ\sin(\varphi_{tp})\frac{(1 - \varepsilon_{zz}/\varepsilon_{xx})\cos(\varphi_{tp})N_z\cos(\varphi_0) + N_0}{N_z\cos(\varphi_0) + N_0\cos(\varphi_{tp})}\right).$$

Now, since the magneto optical parameter $Q \ll 1$, we disregard the terms proportional to Q^2 and higher powers of Q in view of their smallness:

$$\begin{aligned} r_{ppTMOKE} &= \left[\frac{(N_z\cos(\varphi_0) - N_0\cos(\varphi_{tp}))}{N_z\cos(\varphi_0) + N_0\cos(\varphi_{tp})} - \frac{iQ\sin(\varphi_{tp})(N_0 - (1 - \varepsilon_{zz}/\varepsilon_{xx})\cos(\varphi_{tp})N_z\cos(\varphi_0))}{N_z\cos(\varphi_0) + N_0\cos(\varphi_{tp})}\right] \\ &\quad \times \left(1 - iQ\sin(\varphi_{tp})\frac{(1 - \varepsilon_{zz}/\varepsilon_{xx})\cos(\varphi_{tp})N_z\cos(\varphi_0) + N_0}{N_z\cos(\varphi_0) + N_0\cos(\varphi_{tp})}\right), \end{aligned} \quad (A.26)$$

$$\begin{aligned} r_{ppTMOKE} &= \frac{N_z\cos(\varphi_0) - N_0\cos(\varphi_{tp})}{N_z\cos(\varphi_0) + N_0\cos(\varphi_{tp})} - \frac{iQ\sin(\varphi_{tp})(N_0 - (1 - \varepsilon_{zz}/\varepsilon_{xx})\cos(\varphi_{tp})N_z\cos(\varphi_0))}{N_z\cos(\varphi_0) + N_0\cos(\varphi_{tp})} \\ &\quad - \frac{iQ\sin(\varphi_{tp})(N_0 + (1 - \varepsilon_{zz}/\varepsilon_{xx})\cos(\varphi_{tp})N_z\cos(\varphi_0))}{(N_z\cos(\varphi_0) + N_0\cos(\varphi_{tp}))^2}(N_z\cos(\varphi_0) - N_0\cos(\varphi_{tp})). \end{aligned} \quad (A.27)$$

Thus, we have singled out the first term, which is given by expression (11) as expected. Let us transform the second and third terms:

$$r_{pTMOKE} = \frac{N_z\cos(\varphi_0) - N_0\cos(\varphi_{tp})}{N_z\cos(\varphi_0) + N_0\cos(\varphi_{tp})} - \frac{iQ\sin(\varphi_{tp})}{(N_z\cos(\varphi_0) + N_0\cos(\varphi_{tp}))^2}$$
$$\times\left(\left(N_0 - \left(1 - \frac{\varepsilon_{zz}}{\varepsilon_{xx}}\right)\cos(\varphi_{tp})N_z\cos(\varphi_0)\right)(N_z\cos(\varphi_0) + N_0\cos(\varphi_{tp})) + (N_z\cos(\varphi_0) - N_0\cos(\varphi_{tp}))\right. \tag{A.28}$$
$$\left.\times\left(N_0 + \left(1 - \frac{\varepsilon_{zz}}{\varepsilon_{xx}}\right)\cos(\varphi_{tp})N_z\cos(\varphi_0)\right)\right).$$

Let us consider separately the expression in the parentheses of the second term, denoting it by A:

$$\begin{aligned} A = (N_0 - (1 - \varepsilon_{zz}/\varepsilon_{xx})\cos(\varphi_{tp})N_z\cos(\varphi_0))(N_z\cos(\varphi_0) + N_0\cos(\varphi_{tp}))\\ +(N_z\cos(\varphi_0) - N_0\cos(\varphi_{tp}))(N_0 + (1 - \varepsilon_{zz}/\varepsilon_{xx})\cos(\varphi_{tp})N_z\cos(\varphi_0)). \end{aligned} \tag{A.29}$$

Removing the parentheses, we obtain

$$\begin{aligned} A = N_0N_z\cos(\varphi_0) - N_z^2(1 - \varepsilon_{zz}/\varepsilon_{xx})\cos(\varphi_{tp})\cos^2(\varphi_0) + N_0^2\cos(\varphi_{tp}) - N_0N_z(1 - \varepsilon_{zz}/\varepsilon_{xx})\cos(\varphi_0)\cos^2(\varphi_{tp})\\ +N_0N_z\cos(\varphi_0) + N_z^2(1 - \varepsilon_{zz}/\varepsilon_{xx})\cos(\varphi_{tp})\cos^2(\varphi_0) - N_0^2\cos(\varphi_{tp}) - N_0N_z(1 - \varepsilon_{zz}/\varepsilon_{xx})\cos(\varphi_0)\cos^2(\varphi_{tp}), \end{aligned} \tag{A.30}$$

$$A = 2N_0N_z\cos(\varphi_0)(1 - (1 - \varepsilon_{zz}/\varepsilon_{xx})\cos^2(\varphi_{tp})). \tag{A.31}$$

Ultimately, expression (A.28) takes the form

$$\begin{aligned} r_{ppTMOKE} = \frac{N_z\cos(\varphi_0) - N_0\cos(\varphi_{tp})}{N_z\cos(\varphi_0) + N_0\cos(\varphi_{tp})}\\ -iQ\frac{2N_0N_z\sin(\varphi_{tp})\cos(\varphi_0)(1 - (1 - \varepsilon_{zz}/\varepsilon_{xx})\cos^2(\varphi_{tp}))}{(N_z\cos(\varphi_0) + N_0\cos(\varphi_{tp}))^2}, \end{aligned} \tag{A.32}$$

where the first term is R_{p0} and the second term is R_{p1}.

Thus, we have obtained the reflection coefficient for the p-polarization, which takes into account for the MO response in the TMOKE geometry:

$$r_{ppTMOKE} = \frac{N_z\cos(\varphi_0) - N_0\cos(\varphi_{tp})}{N_z\cos(\varphi_0) + N_0\cos(\varphi_{tp})} - iQ\frac{2N_0N_z\sin(\varphi_{tp})\cos(\varphi_0)(1 - (1 - N_z^2/N_x^2)\cos^2(\varphi_{tp}))}{(N_z\cos(\varphi_0) + N_0\cos(\varphi_{tp}))^2}. \tag{A.33}$$

APPENDIX B

Below, we describe the derivation of the expression for the MO parameter Q from system of equations (36). From the second equation, we get

$$Q = \frac{(i\alpha R'_{p0} + \beta R''_{p0})(N_z\cos(\varphi_0) + N_0\cos(\varphi_{tp}))^2}{2N_0N_z\sin(\varphi_{tp})\cos(\varphi_0)(1 - (1 - N_z^2/N_x^2)\cos^2(\varphi_{tp}))}. \tag{B.1}$$

Given expressions (31) and (32) for small parameters α and β. let us write the first factor in the numerator of expression (B.1):

$$\beta R''_{p0} + i\alpha R'_{p0} = \left(\frac{\delta\psi(1 + \tan^2(\psi_0))}{\tan(\psi_0)} - \frac{R'_{p0}}{R''_{p0}}\delta\Delta\right)R''_{p0} + i\left(\frac{\delta\psi(1 + \tan^2(\psi_0))}{\tan(\psi_0)} + \frac{R''_{p0}}{R'_{p0}}\delta\Delta\right)R'_{p0}. \tag{B.2}$$

Removing the parentheses and collecting like terms, we obtain

$$\begin{aligned} \beta R''_{p0} + i\alpha R'_{p0} = \frac{\delta\psi(1 + \tan^2(\psi_0))}{\tan(\psi_0)}R''_{p0} - R'_{p0}\delta\Delta + i\frac{\delta\psi(1 + \tan^2(\psi_0))}{\tan(\psi_0)}R'_{p0} + iR''_{p0}\delta\Delta\\ = \frac{\delta\psi(1 + \tan^2(\psi_0))}{\tan(\psi_0)}(R''_{p0} + iR'_{p0}) - \delta\Delta(R'_{p0} + iR''_{p0}) = R_{p0}\left(i\frac{\delta\psi(1 + \tan^2(\psi_0))}{\tan(\psi_0)} - \delta\Delta\right). \end{aligned} \tag{B.3}$$

Then the magneto-optical parameter takes form

$$Q = R_{p0}\left(i\frac{\delta\psi(1+\tan^2(\psi_0))}{\tan(\psi_0)} - \delta\Delta\right)(N_z\cos(\varphi_0) + N_0\cos(\varphi_{tp}))^2 \Bigg/ \left(2N_0N_z\sin(\varphi_{tp})\cos(\varphi_0)\left(1-\left(1-\frac{N_z^2}{N_x^2}\right)\cos^2(\varphi_{tp})\right)\right). \quad \text{(B.4)}$$

Substituting the value of R_{p0} from expression (36), we obtain

$$\begin{aligned} Q &= \frac{N_z\cos(\varphi_0) - N_0\cos(\varphi_{tp})}{N_z\cos(\varphi_0) + N_0\cos(\varphi_{tp})}\left(i\frac{\delta\psi(1+\tan^2(\psi_0))}{\tan(\psi_0)} - \delta\Delta\right)(N_z\cos(\varphi_0) + N_0\cos(\varphi_{tp}))^2 \\ &\quad \Bigg/ \left(2N_0N_z\sin(\varphi_{tp})\cos(\varphi_0)\left(1-\left(1-\frac{N_z^2}{N_x^2}\right)\cos^2(\varphi_{tp})\right)\right) \\ &= \frac{(N_z\cos(\varphi_0))^2 - (N_0\cos(\varphi_{tp}))^2}{2N_0N_z\sin(\varphi_{tp})\cos(\varphi_0)(1-(1-N_z^2/N_x^2)\cos^2(\varphi_{tp}))}\left(i\frac{\delta\psi(1+\tan^2(\psi_0))}{\tan(\psi_0)} - \delta\Delta\right). \end{aligned} \quad \text{(B.5)}$$

This leads to the following expression for calculating the magneto-optical parameter for auniaxial anisotropic bulk crystal:

$$Q = \frac{N_0^2\cos^2(\varphi_{tp}) - N_z^2\cos^2(\varphi_0)}{2N_0N_z\sin(\varphi_{tp})\cos(\varphi_0)(1-(1-N_z^2/N_x^2)\cos^2(\varphi_{tp}))}\left(\delta\Delta - i\frac{\delta\psi(1+\tan^2(\psi_0))}{\tan(\psi_0)}\right). \quad \text{(B.6)}$$

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