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Application of generalized ellipsometry to anisotropic crystals*

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DeSmet[1] is to be thanked for directing our attention to the fact that $R_{sp} = -R_{ps}$. It follows that measurement of the normalized reflection matrix $\mathbf{R}(\omega, \phi)$ at one crystal orientation ω and one angle of incidence ϕ enables only four (not all[2]) of the five parameters of a uniaxial crystal (n_o, n_e, k_o, k_e, ω) to be determined, provided the fifth is known.

Practically, it is an easy matter to set the crystal with its optic axis at any desired orientation ω (e.g., $\omega = 45°$ or $135°$, where $|R_{ps}|$ and $|R_{sp}|$ are maximum) so that measurement of $\mathbf{R}$ at one angle of incidence ϕ enables all of the optical parameters n_o, n_e, k_o, and k_e to be determined. With the compensator removed, the polarizer and analyzer can be positioned with their transmission axes, one parallel and the other perpendicular to the plane of incidence. By rotating the uniaxial crystal (whose optic axis is parallel to its surface) around the surface normal until extinction is achieved, the optic axis becomes aligned parallel or perpendicular to the plane of incidence. Rotation of the crystal by a known angle ω from its extinction setting ensures that the azimuth of the optic axis is either ω or $\omega + (\pi/2)$. Inversion of the nonlinear equations of reflection [Eqs. (11)–(13), Ref. 2] to find n_o, n_e, k_o, and k_e by use of the measured reflection matrix $\mathbf{R}$ should be possible only if one of the two values ω or $\omega + (\pi/2)$ is substituted. Thus, the ω or $\omega + (\pi/2)$ initial ambiguity of orientation is resolvable.

Although setting the crystal at a known orientation, as mentioned above, rectifies the difficulty raised by De Smet[1] concerning the number of independent real quantities that are measured and those that are to be determined, we believe there is nothing essential about measurement of the normalized reflection matrix $\mathbf{R}(\omega, \phi)$ once at a single set (ω, ϕ). We now believe that there was no need to emphasize this point in our paper.[2] Fitting data from multiple orientations, such as that in Figs. 2 and 3 of Ref. 2 or Figs. 3 and 4 of Ref. 3 is certainly desirable for better accuracy. Of course, multiple measurements $\mathbf{R}_1(\omega_1, \phi_1)$, $\mathbf{R}_2(\omega_2, \phi_2)$, ... can be automated and the time or labor required for obtaining the additional data becomes no major issue.

Our inversion of Eqs. (11)–(13) by use of one measured reflection matrix, Eqs. (14), to obtain all the unknowns, Eqs. (15), was successful[4] because iteration in the computer program was started with good initial guesses for n_o and n_e (values were taken within ± 0.2 of the published handbook values for these quantities). However, this inversion would not be possible if arbitrary initial guesses are used, unless one of the parameters (e.g., ω) is known, as explained above.

Finally, we believe that the condition

$$|R_{sp}| = |R_{ps}|, \tag{1}$$

embraces a wide range of surface anisotropy and is enforced by a basic physical reciprocity law concerning power exchange in the TM ↔ TE (or $p \leftrightarrow s$) mode conversion that takes place upon reflection (induced by the surface anisotropy). We have found, for example, that Eq. (1) is satisfied (within experimental error) by the off-diagonal elements of the ellipsometrically measured zero-order reflection matrix of a surface whose anisotropy originates from the presence of a line structure (diffraction grating).[5] Other instances can also be found in the literature.[6] At this point, we are unaware of situations where Eq. (1) breaks down. Comments concerning this point will be appreciated.

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[3]D. J. De Smet, J. Opt. Soc. Am. 63, 958 (1973).
[4]All equations cited are those of Ref. 2.
[5]R. M. A. Azzam and N. M. Bashara, J. Opt. Soc. Am. 62, 1521 (1972).
[6]S. Wang, M. L. Shah, and J. D. Crow, IEEE J. Quantum Electron. 8, 212 (1972), Eqs. (4) and (6).