Propagation of an electromagnetic wave in an absorbing anisotropic medium and infrared transmission spectroscopy of liquid crystals

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The theory of absorbance is developed for the entire electromagnetic spectrum of radiation in a semi-infinite anisotropic medium with a second rank dielectric tensor, the elements of which are complex and frequency dependent. The theory of the absorbance $\mathcal{A}(\omega, \theta)$ of an optically anisotropic liquid in an infrared (IR) test cell is then outlined and applied to IR transmission experiments. A formula for the dependence of $\mathcal{A}(\omega, \theta)$, on θ (θ being the angle between the electric vector and the principal optical axis) is derived from first principles. The formula, for radiation of angular frequency ω , viz, $\mathcal{A}(\omega, \theta) = -\log_{10}[10^{-\mathcal{A}(\omega, 0)} \cos^2 \theta + 10^{-\mathcal{A}(\omega, \pi/2)} \sin^2 \theta]$ is in agreement with that proposed by Jang, Park, MacLennan, Kim, and Clark [Ferroelectrics **180**, 213 (1996)] and confirms some of the work of Kocot, Wrzalik, and Vij [Liq. Cryst. **21**, 147 (1996)]. The comments on this formula by Jang, Park, Kim, Glaser, and Clark [Phys. Rev. E **62**, 5027 (2000)], and by Kocot *et al.* are discussed. The absorbance $\mathcal{A}(\omega, 0)$ and $\mathcal{A}(\omega, \pi/2)$ have been expressed in terms of the optical properties of the material and the dimensions of the cell. © 2005 American Institute of Physics. [DOI: 10.1063/1.1874833]

I. INTRODUCTION

Infrared absorbance spectroscopy is a powerful technique for studying the molecular structure and this technique has also been applied to the study of liquid crystalline material¹ in the nematic phase since the early 1970's. This technique was first applied in the early 1990's to the study of the ferroelectric liquid crystalline materials and in particular for determining the tilt angle and the orientational order parameter.² Recently, the technique has been used to determine a complete set of second rank orientational order parameters.^{3,4} The orientational distribution function of the directors in smectic layers has been determined for the ferroelectric liquid crystals first by Fukuda *et al.*⁵ In this paper we theoretically investigate the absorbance as a function of the angle of polarization where a ferroelectric liquid crystal is confined to lie between the two windows of a cell. The measurement of the absorption of infrared (IR) radiation in a liquid crystal involves an arrangement shown schematically in Fig. 1. The radiation is divided by a beam splitter *A*; one part constituting a reference beam of intensity I_{ref} , the other enters the test cell *B* with intensity I_{in} .

The *transmittance* of the test cell, $T_{\text{cell}}(\omega)$, for radiation with angular frequency ω , is defined by the relation,

$$T_{\text{cell}}(\omega) \equiv \frac{I_{\text{out}}(\omega)}{I_{\text{in}}(\omega)}, \quad (1.1)$$

in which $I_{\text{out}}(\omega)$ is the intensity of the beam leaving the test cell and $I_{\text{in}}(\omega)$ is the intensity of the beam entering the test cell. We shall assume that

$$I_{\text{in}}(\omega) = I_{\text{ref}}(\omega). \quad (1.2)$$

Suppose that both these beams have the same cross-sectional area σ then we may express $T_{\text{cell}}(\omega)$ in terms of the corresponding power-flux densities, $P_{\text{ref}}(\omega)$ and $P_{\text{out}}(\omega)$, as follows:

$$T_{\text{cell}}(\omega) = \frac{I_{\text{out}}(\omega)}{I_{\text{ref}}(\omega)} = \frac{\sigma P_{\text{out}}(\omega)}{\sigma P_{\text{ref}}(\omega)} = \frac{P_{\text{out}}(\omega)}{P_{\text{ref}}(\omega)}. \quad (1.3)$$

The units of $P_{\text{ref}}(\omega)$ and $P_{\text{out}}(\omega)$ are W m^{-2} .

If the radiation is plane polarized and if the test cell contains an optically anisotropic substance, the power-flux density P_{out} , and hence the transmittance T_{cell} , will depend on θ , the angle between the electric vector of the incident beam and the z (or 3) axis of the test cell. See Fig. 2. Therefore we replace Eq. (1.3) by

$$T_{\text{cell}}(\omega, \theta) = \frac{P_{\text{out}}(\omega, \theta)}{P_{\text{ref}}(\omega, \theta)}. \quad (1.4)$$

In Sec. III we shall show, in Eq. (3.14), that in general,

$$T_{\text{cell}}(\omega, \theta) = \Lambda_{\parallel}(\omega) \cos^2 \theta + \Lambda_{\perp}(\omega) \sin^2 \theta, \quad (1.5)$$

the factors $\Lambda_{\parallel}(\omega)$ and $\Lambda_{\perp}(\omega)$ depend upon the dimensions and optical properties of the test cell and on the optical properties of its contents. Unfortunately there are no simple, general expressions for $\Lambda_{\parallel}(\omega)$ and $\Lambda_{\perp}(\omega)$.

The *absorbance* of the test cell, $\mathcal{A}(\omega, \theta)$, is defined by the equation,

$$\mathcal{A}(\omega, \theta) \equiv -\log_{10} T_{\text{cell}}(\omega, \theta), \quad (1.6)$$

this result with Eq. (1.5) leads to

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FIG. 1. The IR radiation is divided by the beam splitter A. The reflected portion is used as a reference; the transmitted portion enters the test cell B.

$$10^{-A(\omega, \theta)} = T_{\text{cell}}(\omega, \theta) = \Lambda_{\parallel}(\omega) \cos^2 \theta + \Lambda_{\perp}(\omega) \sin^2 \theta \quad (1.7)$$

and hence

$$10^{-A(\omega, 0)} = \Lambda_{\parallel}(\omega), \quad (1.8a)$$

$$10^{-A(\omega, \pi/2)} = \Lambda_{\perp}(\omega). \quad (1.8b)$$

With the following notation:

$$\mathcal{A}(\omega, 0) = \mathcal{A}_{\parallel}(\omega), \quad (1.9a)$$

$$\mathcal{A}(\omega, \pi/2) = \mathcal{A}_{\perp}(\omega), \quad (1.9b)$$

Eq. (1.6) may be written in the form

$$\mathcal{A}(\omega, \theta) = -\log_{10} [10^{-\mathcal{A}_{\parallel}(\omega)} \cos^2 \theta + 10^{-\mathcal{A}_{\perp}(\omega)} \sin^2 \theta]. \quad (1.10)$$

This equation was proposed by Jang *et al.*⁶ who obtained values of $\mathcal{A}_{\parallel}(\omega)$ and $\mathcal{A}_{\perp}(\omega)$ by fitting it to experimental data. Several materials have been examined by means of Eq. (1.10) such as the helical structure of a SmC^* phase as well as the state which is fully unwound by an electric field.⁷⁻¹² The measurements were made on liquid crystalline samples of thicknesses of up to 15 μm . The fully unwound SmC^* state is known to have a finite degree of biaxiality. Jang *et al.* state in one of their recent papers¹⁰ [as a footnote in their reference 13], without providing reasons, that Eq. (1.10) is exact only in the absence of birefringence (no anisotropy of the dielectric permittivity). Because in most experiments the IR radiation is polarized nearly along the principal optical axis of the dielectric, Jang *et al.*¹⁰ assume that the birefringence may be ignored in samples of a few micrometers in thickness. From our derivation of Eq. (1.7) in Sec. III [see Eq. (3.14)] it does not seem that it is subject to such limitations.

To clarify the discussion we outline, in Sec. II A, the theory of the propagation of electromagnetic waves in anisotropic media and in Sec. II B we discuss the passage of an IR beam through a test cell containing an anisotropic medium. Expressions for the Poynting vector are obtained in Sec. III, and those for the density of power absorption in Sec. IV. The molecular aspects of the problem are considered in Sec. V. A general discussion and conclusions will be found in Sec. VI, together with an analysis of the calculations of Kocot *et al.*⁷ It is found that, for a general molecular orientational distri-

FIG. 2. The arrangement for making IR absorption measurements. The radiation of wave vector $\mathbf{k}$ propagates along the y, or 2, axis. The sample lies between two (a,b) CaF_2 windows, each with an indium tin oxide (ITO) electrode (c,d). The smectic layers (f) lie in the x-y plane. The electric vector $\mathbf{E}_0$ of the polarized radiation is perpendicular to $\mathbf{k}$ and makes an angle θ with the z axis which is taken to be the principal optical axis.

bution function, the equation derived by Kocot *et al.*⁷ is obtained in the absence of quadrupolar order [$D=0$ in Eq. (6.9)]; this equation is also valid in the case of thin samples with $C=D=0$.

II. MACROSCOPIC CONSIDERATIONS

A. Homogeneous plane-wave propagation in a semi-infinite anisotropic medium

In this section we derive expressions for the electric field of a polarized, homogeneous plane-wave launched into the infinite plane boundary of a semi-infinite optically anisotropic medium. A homogeneous wave is one in which the planes of constant amplitude and those of constant phase are parallel to one another. The ensuing calculations are based on the treatment by Landau and Lifshitz.¹³ A more general treatment on the propagation of the electromagnetic wave in an infinite medium has been given by Yuan *et al.*^{14,15} in which they also consider spatial dispersion, i.e., permittivity to be spatially dependent on the wave vector, in addition to its frequency dependence. They expand the dielectric tensor as a Taylor series expansion to first order in the wave vector $\mathbf{k}$. However, in the analysis given here, we confine ourselves to investigating the propagation in an absorbing anisotropic medium confined to a cell and ignore spatial dispersion. We can justify this by stating that the wavelengths considered here are too large compared with the intermolecular separations in the liquid crystalline material and furthermore this linear term in $\mathbf{k}$ is likely to introduce only a small correction. By explicitly including just the linear term in $\mathbf{k}$, the calculations become too complicated to be helpful in the problem under discussion. We thus assume that all values of k are allowed for the different frequencies under discussion.

For an insulating medium devoid of free electric charges, Maxwell's equations (in SI units) take the form

FIG. 3. The right-handed coordinate system with unit vectors $\mathbf{i}_1$, $\mathbf{i}_2$, and $\mathbf{i}_3$.

$$\begin{aligned}\text{curl } \mathbf{e}(\mathbf{r}, t) &= -\partial_t \mathbf{b}(\mathbf{r}, t), \\ \text{curl } \mathbf{h}(\mathbf{r}, t) &= \partial_t \mathbf{d}(\mathbf{r}, t), \\ \text{div } \mathbf{d}(\mathbf{r}, t) &= \text{div } \mathbf{b}(\mathbf{r}, t) = 0,\end{aligned}\quad (2.1)$$

in which the vectors $\mathbf{e}(\mathbf{r}, t)$, $\mathbf{h}(\mathbf{r}, t)$, $\mathbf{d}(\mathbf{r}, t)$, and $\mathbf{b}(\mathbf{r}, t)$ are, respectively, the electric field, the magnetizing force, the electric flux density, and the magnetic flux density, at position $\mathbf{r}$ at time t . The vector $\mathbf{r}$ has Cartesian components r_1 , r_2 , and r_3 , such that

$$\mathbf{r} = \mathbf{i}_1 r_1 + \mathbf{i}_2 r_2 + \mathbf{i}_3 r_3 = \mathbf{i}_1 x + \mathbf{i}_2 y + \mathbf{i}_3 z, \quad (2.2)$$

in which $\mathbf{i}_1$, $\mathbf{i}_2$, and $\mathbf{i}_3$ (see Fig. 3) are three mutually perpendicular unit vectors of a right-handed coordinate system.

For a plane wave of angular frequency ω and complex, frequency-dependent propagation vector $\mathbf{k}(\omega)$, the vectors $\mathbf{e}$, $\mathbf{h}$, $\mathbf{d}$, and $\mathbf{b}$ may be expressed as follows:

$$\begin{aligned}\mathbf{e}(\mathbf{r}, t) &= \text{Re } \mathbf{E}_0(\omega) \exp[i(\omega t - \mathbf{k} \cdot \mathbf{r})], \\ \mathbf{h}(\mathbf{r}, t) &= \text{Re } \mathbf{H}_0(\omega) \exp[i(\omega t - \mathbf{k} \cdot \mathbf{r})], \\ \mathbf{d}(\mathbf{r}, t) &= \text{Re } \mathbf{D}_0(\omega) \exp[i(\omega t - \mathbf{k} \cdot \mathbf{r})], \\ \mathbf{b}(\mathbf{r}, t) &= \text{Re } \mathbf{B}_0(\omega) \exp[i(\omega t - \mathbf{k} \cdot \mathbf{r})],\end{aligned}\quad (2.3)$$

where $i = \sqrt{-1}$, and Re denotes “real part of”. We shall restrict attention to nonmagnetic media, so that

$$\begin{aligned}\mathbf{b}(\mathbf{r}, t) &= \mu_0 \mathbf{h}(\mathbf{r}, t), \\ \mathbf{B}_0(\omega) &= \mu_0 \mathbf{H}_0(\omega),\end{aligned}\quad (2.4)$$

in which μ_0 is the permeability of free space. The quantities $\mathbf{E}_0(\omega)$, $\mathbf{D}_0(\omega)$, and $\mathbf{H}_0(\omega)$ are the complex vector amplitudes of $\mathbf{e}(\mathbf{r}, t)$, $\mathbf{d}(\mathbf{r}, t)$, and $\mathbf{h}(\mathbf{r}, t)$, at $\mathbf{r} = 0$ and $t = 0$, for a particular propagation vector $\mathbf{k}(\omega)$. This vector may be split into its real and imaginary components, and in keeping with the convention that counter-clockwise rotations are positive, we write

$$\mathbf{k}(\omega) = \mathbf{k}'(\omega) - i \mathbf{k}''(\omega). \quad (2.5)$$

To ensure that the plane wave is homogeneous, the vectors $\mathbf{k}'(\omega)$ and $\mathbf{k}''(\omega)$ must be parallel. The insertion of Eqs. (2.3) and (2.4) into Eqs. (2.1) leads to

$$-i \mathbf{k}(\omega) \times \mathbf{E}_0(\omega) = -i \omega \mu_0 \mathbf{H}_0(\omega), \quad (2.6)$$

$$-i \mathbf{k}(\omega) \times \mathbf{H}_0(\omega) = i \omega \mathbf{D}_0(\omega), \quad (2.7)$$

$$i \mathbf{k}(\omega) \cdot \mathbf{D}_0(\omega) = 0, \quad (2.8)$$

$$i \mathbf{k}(\omega) \cdot \mathbf{B}_0(\omega) = i \mathbf{k}(\omega) \cdot \mathbf{H}_0(\omega) = 0. \quad (2.9)$$

Equations (2.8) and (2.9) indicate that $\mathbf{k}(\omega)$ is perpendicular to $\mathbf{D}_0(\omega)$ and to $\mathbf{H}_0(\omega)$. When we combine Eqs. (2.6) and (2.7) we obtain the relation

$$\mathbf{k}(\omega) \times [\mathbf{k}(\omega) \times \mathbf{E}_0(\omega)] = -\omega^2 \mu_0 \mathbf{D}_0(\omega). \quad (2.10)$$

By means of the standard formula from vector algebra,

$$\mathbf{A} \times (\mathbf{A} \times \mathbf{B}) = \mathbf{A}(\mathbf{A} \cdot \mathbf{B}) - A^2 \mathbf{B}, \quad (2.11)$$

we may rearrange Eq. (2.10) to read

$$\mathbf{k}(\omega) [\mathbf{k}(\omega) \cdot \mathbf{E}_0(\omega)] - [\mathbf{k}(\omega)]^2 \mathbf{E}_0(\omega) = -\omega^2 \mu_0 \mathbf{D}_0(\omega). \quad (2.12)$$

It is clear from this equation that $\mathbf{D}_0(\omega)$ need not be parallel to $\mathbf{E}_0(\omega)$. We cope with this possibility by introducing the complex, frequency-dependent, relative permittivity tensor $\varepsilon_{\alpha\beta}(\omega)$. The suffixes α and β can take the values 1, 2, or 3. We therefore write

$$D_{0\alpha}(\omega) = \varepsilon_0 \sum_{\beta=1}^3 \varepsilon_{\alpha\beta}(\omega) E_{0\beta}(\omega) = \varepsilon_0 \varepsilon_{\alpha\beta}(\omega) E_{0\beta}(\omega), \quad (2.13)$$

in which summation over the repeated suffix β is implied in the last term. Thus Eq. (2.13), written in full, reads as follows:

$$\begin{aligned}D_{01}(\omega) &= \varepsilon_0 [\varepsilon_{1,1}(\omega) E_{01}(\omega) + \varepsilon_{1,2}(\omega) E_{02}(\omega) \\ &\quad + \varepsilon_{1,3}(\omega) E_{03}(\omega)], \\ D_{02}(\omega) &= \varepsilon_0 [\varepsilon_{2,1}(\omega) E_{01}(\omega) + \varepsilon_{2,2}(\omega) E_{02}(\omega) \\ &\quad + \varepsilon_{2,3}(\omega) E_{03}(\omega)], \\ D_{03}(\omega) &= \varepsilon_0 [\varepsilon_{3,1}(\omega) E_{01}(\omega) + \varepsilon_{3,2}(\omega) E_{02}(\omega) \\ &\quad + \varepsilon_{3,3}(\omega) E_{03}(\omega)],\end{aligned}\quad (2.14)$$

where ε_0 is the permittivity of free space. For the materials considered here, the permittivity tensor is symmetrical, that is,

$$\varepsilon_{\alpha\beta}(\omega) = \varepsilon_{\beta\alpha}(\omega). \quad (2.15)$$

Equation (2.12), when expressed in tensor form is

$$\begin{aligned}c_0^2 [k_\alpha(\omega) k_\beta(\omega) E_{0\beta} - k_\beta(\omega) k_\alpha(\omega) E_{0\alpha}(\omega)] \\ = -\varepsilon_{\alpha\beta}(\omega) E_{0\beta}(\omega),\end{aligned}\quad (2.16)$$

in which the summation convention is implied and where $c_0 = \sqrt{(\varepsilon_0 \mu_0)^{-1}}$ is the speed of light in free space.

In view of the symmetry condition in Eq. (2.15), it is always possible to choose a set of coordinate axes so that the permittivity tensor becomes diagonal. Therefore, in matrix form, we have

$$[\varepsilon_{\alpha\beta}(\omega)] = \begin{bmatrix} \varepsilon_1(\omega) & 0 & 0 \\ 0 & \varepsilon_2(\omega) & 0 \\ 0 & 0 & \varepsilon_3(\omega) \end{bmatrix}, \quad (2.17)$$

where we have used the abbreviations: $\varepsilon_{1,1}(\omega) = \varepsilon_1(\omega)$, $\varepsilon_{2,2}(\omega) = \varepsilon_2(\omega)$, and $\varepsilon_{3,3}(\omega) = \varepsilon_3(\omega)$. With this result we may express Eq. (2.16) in matrix form as follows:

$$c_0^2 \begin{bmatrix} k_0^2 \varepsilon_1 - (k_2^2 + k_3^2) & k_1 k_2 & k_1 k_3 \\ k_1 k_2 & k_0^2 \varepsilon_2 - (k_1^2 + k_3^2) & k_2 k_3 \\ k_1 k_3 & k_2 k_3 & k_0^2 \varepsilon_3 - (k_1^2 + k_2^2) \end{bmatrix} \times \begin{bmatrix} E_{01}(\omega) \\ E_{02}(\omega) \\ E_{03}(\omega) \end{bmatrix} = 0, \quad (2.18)$$

where we have introduced k_0 , the free-space wave number of the radiation, defined by the relation

$$k_0 \equiv \frac{\omega}{c_0} = \frac{2\pi}{\lambda_0}, \quad (2.19)$$

λ_0 being the free-space wavelength. To save space in Eq. (2.18) we have dropped the dependence on ω of k_1 , k_2 , and k_3 . The condition that the matrix Eq. (2.18) has a solution is that the determinant of the left-hand matrix should vanish, that is,

$$\begin{vmatrix} k_0^2 \varepsilon_1 - k_2^2 - k_3^2 & k_1 k_2 & k_1 k_3 \\ k_1 k_2 & k_0^2 \varepsilon_2 - k_1^2 - k_3^2 & k_2 k_3 \\ k_1 k_3 & k_2 k_3 & k_0^2 \varepsilon_3 - k_1^2 - k_2^2 \end{vmatrix} = 0. \quad (2.20)$$

After considerable algebra this equation reduces to

$$\begin{aligned} \varepsilon_1 \varepsilon_2 \varepsilon_3 - \varepsilon_1 (\varepsilon_2 + \varepsilon_3) k_1^2 k_0^{-2} - \varepsilon_2 (\varepsilon_1 + \varepsilon_3) k_2^2 k_0^{-2} \\ - \varepsilon_3 (\varepsilon_1 + \varepsilon_2) k_3^2 k_0^{-2} + \varepsilon_1 k_1^2 k_2^2 k_0^{-4} + \varepsilon_2 k_2^2 k_3^2 k_0^{-4} \\ + \varepsilon_3 k_3^2 k_1^2 k_0^{-4} = 0, \end{aligned} \quad (2.21)$$

where we have used the facts that

$$\mathbf{k}(\omega) = \mathbf{i}_1 k_1(\omega) + \mathbf{i}_2 k_2(\omega) + \mathbf{i}_3 k_3(\omega) \quad (2.22)$$

and

$$\begin{aligned} k^2 &\equiv [k(\omega)]^2 = |\mathbf{k}(\omega)|^2 \\ &= \mathbf{k}(\omega) \cdot \mathbf{k}(\omega) = [k_1(\omega)]^2 + [k_2(\omega)]^2 + [k_3(\omega)]^2 \\ &= k_1^2 + k_2^2 + k_3^2. \end{aligned} \quad (2.23)$$

Now we restrict our attention to a plane-wave traveling along the 2 (or y) axis, so that $k_1(\omega) = k_3(\omega) = 0$. In this case Eq. (2.21) reduces to the following quadratic in $k_2^2 k_0^{-2}$:

$$\begin{aligned} \varepsilon_1(\omega) \varepsilon_2(\omega) \varepsilon_3(\omega) - \varepsilon_2(\omega) [\varepsilon_1(\omega) + \varepsilon_3(\omega)] k_2^2 k_0^{-2} \\ + \varepsilon_2(\omega) k_2^4 k_0^{-4} = 0. \end{aligned} \quad (2.24)$$

On dividing throughout by $\varepsilon_2(\omega)$, we find the solutions of this equation to be

FIG. 4. To illustrate the two possible transverse waves propagating along the 2 axis with propagation vectors $\mathbf{k}_{\parallel}$ and $\mathbf{k}_{\perp}$.

$$\begin{aligned} k_2^2 k_0^{-2} &= \frac{[\varepsilon_1(\omega) + \varepsilon_3(\omega)] \pm \sqrt{[\varepsilon_1(\omega) + \varepsilon_3(\omega)]^2 - 4\varepsilon_1(\omega)\varepsilon_3(\omega)}}{2} \\ &= \frac{[\varepsilon_1(\omega) + \varepsilon_3(\omega)] \pm \sqrt{[\varepsilon_1(\omega) - \varepsilon_3(\omega)]^2}}{2} \\ &= \begin{cases} \varepsilon_1(\omega) \\ \varepsilon_3(\omega) \end{cases}. \end{aligned} \quad (2.25)$$

These two solutions correspond, respectively, to plane waves with their electric vectors parallel either to the 1 (or x) axis [$\mathbf{E}_{\perp}(\omega) = \mathbf{i}_1 E_{\perp}(\omega)$] or to the 3 (or z) axis [$\mathbf{E}_{\parallel}(\omega) = \mathbf{i}_3 E_{\parallel}(\omega)$], see Fig. 4. If the medium is dissipative, the quantities $\varepsilon_1(\omega)$ and $\varepsilon_3(\omega)$ will be complex and, hence, so also will be the values of $k_2(\omega)$ obtained from Eq. (2.25). We shall use the following notation:

$$\mathbf{k}_{\perp} = \mathbf{k}_{\perp}(\omega) = \mathbf{i}_2 k_{\perp}(\omega) = \mathbf{i}_2 [k'_{\perp}(\omega) - \iota k''_{\perp}(\omega)], \quad (2.26)$$

$$\mathbf{k}_{\parallel} = \mathbf{k}_{\parallel}(\omega) = \mathbf{i}_2 k_{\parallel}(\omega) = \mathbf{i}_2 [k'_{\parallel}(\omega) - \iota k''_{\parallel}(\omega)],$$

so that

$$\begin{aligned} k_{\perp}^2 &= [k_{\perp}(\omega)]^2 = [k'_{\perp}(\omega) - \iota k''_{\perp}(\omega)]^2 \\ &= k_0^2 \varepsilon_1(\omega) = \frac{\omega^2}{c_0^2} [\varepsilon'_1(\omega) - \iota \varepsilon''_1(\omega)], \end{aligned} \quad (2.27a)$$

$$\begin{aligned} k_{\parallel}^2 &= [k_{\parallel}(\omega)]^2 = [k'_{\parallel}(\omega) - \iota k''_{\parallel}(\omega)]^2 \\ &= k_0^2 \varepsilon_3(\omega) = \frac{\omega^2}{c_0^2} [\varepsilon'_3(\omega) - \iota \varepsilon''_3(\omega)]. \end{aligned} \quad (2.27b)$$

After separating the real and imaginary parts of these expressions, we find that

$$[k'_{\perp}(\omega)]^2 - [k''_{\perp}(\omega)]^2 = \frac{\omega^2}{c_0^2} \varepsilon'_1(\omega), \quad (2.28a)$$

$$2k'_{\perp}(\omega)k''_{\perp}(\omega) = \frac{\omega^2}{c_0^2} \varepsilon''_1(\omega), \quad (2.28b)$$

$$[k'_{\parallel}(\omega)]^2 - [k''_{\parallel}(\omega)]^2 = \frac{\omega^2}{c_0^2} \varepsilon'_3(\omega), \quad (2.28c)$$

$$2k'_{\parallel}(\omega)k''_{\parallel}(\omega) = \frac{\omega^2}{c_0^2} \varepsilon''_3(\omega). \quad (2.28d)$$

Thus, in view of these results and Eqs. (2.1) and (2.3), we have

$$\begin{aligned}\mathbf{k}_\perp(\omega) \times \mathbf{E}_\perp(\omega) &= \mathbf{i}_2 \times \mathbf{i}_1 k_\perp(\omega) E_\perp(\omega) \\ &= -\mathbf{i}_3 k_\perp(\omega) E_\perp(\omega) \\ &= -\mathbf{i}_3 \omega \mu_0 H_\perp(\omega) = \omega \mu_0 \mathbf{H}_\perp(\omega),\end{aligned}\quad (2.29a)$$

$$\begin{aligned}\mathbf{k}_\parallel(\omega) \times \mathbf{E}_\parallel(\omega) &= \mathbf{i}_2 \times \mathbf{i}_3 k_\parallel(\omega) E_\parallel(\omega) \\ &= \mathbf{i}_1 k_\parallel(\omega) E_\parallel(\omega) \\ &= \mathbf{i}_1 \omega \mu_0 H_\parallel(\omega) = \omega \mu_0 \mathbf{H}_\parallel(\omega).\end{aligned}\quad (2.29b)$$

Since $\text{div } \mathbf{b}(\mathbf{r}, t) = \text{div } \mathbf{h}(\mathbf{r}, t) = 0$ everywhere, the vectors $\mathbf{k}$ and $\mathbf{H}$ must be perpendicular to one another. Consequently the set of vectors $\mathbf{k}_\perp$, $\mathbf{E}_\perp$, and $\mathbf{H}_\perp$ and the set $\mathbf{k}_\parallel$, $\mathbf{E}_\parallel$, and $\mathbf{H}_\parallel$ are each mutually orthogonal. Therefore the vector pairs $(\mathbf{E}_\perp, \mathbf{H}_\perp)$ and $(\mathbf{E}_\parallel, \mathbf{H}_\parallel)$ both lie in the 1-3 plane perpendicular to the direction of propagation as in Fig. 4.

We have the following relations between the magnitudes $E_\perp(\omega)$, and $H_\perp(\omega)$, and between the magnitudes $E_\parallel(\omega)$ and $H_\parallel(\omega)$:

$$H_\perp(\omega) = |\mathbf{H}_\perp(\omega)| = \frac{k_\perp(\omega)}{\omega \mu_0} |\mathbf{E}_\perp(\omega)| = \frac{k_\perp(\omega)}{\omega \mu_0} E_\perp(\omega), \quad (2.30a)$$

$$H_\parallel(\omega) = |\mathbf{H}_\parallel(\omega)| = \frac{k_\parallel(\omega)}{\omega \mu_0} |\mathbf{E}_\parallel(\omega)| = \frac{k_\parallel(\omega)}{\omega \mu_0} E_\parallel(\omega). \quad (2.30b)$$

For the two modes of propagation we obtain from Eqs. (2.3) and (2.26) the following expressions for the electric field vectors:

$$\mathbf{e}_\perp(y, t) = \mathbf{i}_1 \exp[-k''_\perp(\omega)y] \text{Re } E_\perp(\omega) \exp\{i[\omega t - k'_\perp(\omega)y]\}, \quad (2.31a)$$

$$\mathbf{e}_\parallel(y, t) = \mathbf{i}_3 \exp[-k''_\parallel(\omega)y] \text{Re } E_\parallel(\omega) \exp\{i[\omega t - k'_\parallel(\omega)y]\} \quad (2.31b)$$

and for the magnetizing force vectors:

$$\begin{aligned}\mathbf{h}_\perp(y, t) &= -\mathbf{i}_3 \exp[-k''_\perp(\omega)y] \text{Re } \frac{k_\perp(\omega)}{\omega \mu_0} E_\perp(\omega) \\ &\quad \times \exp\{i[\omega t - k'_\perp(\omega)y]\},\end{aligned}\quad (2.31c)$$

$$\begin{aligned}\mathbf{h}_\parallel(y, t) &= \mathbf{i}_1 \exp[-k''_\parallel(\omega)y] \text{Re } \frac{k_\parallel(\omega)}{\omega \mu_0} E_\parallel(\omega) \\ &\quad \times \exp\{i[\omega t - k'_\parallel(\omega)y]\}.\end{aligned}\quad (2.31d)$$

If $E_\perp(\omega)$ and $E_\parallel(\omega)$ are real quantities, these equations take the simpler forms

$$\mathbf{e}_\perp(y, t) = \mathbf{i}_1 \exp[-k''_\perp(\omega)y] \cos[\omega t - k'_\perp(\omega)y] E_\perp(\omega), \quad (2.32a)$$

$$\mathbf{e}_\parallel(y, t) = \mathbf{i}_3 \exp[-k''_\parallel(\omega)y] \cos[\omega t - k'_\parallel(\omega)y] E_\parallel(\omega), \quad (2.32b)$$

$$\begin{aligned}\mathbf{h}_\perp(y, t) &= -\mathbf{i}_3 \exp[-k''_\perp(\omega)y] \left\{ \frac{k'_\perp(\omega)}{\omega \mu_0} \cos[\omega t - k'_\perp(\omega)y] \right. \\ &\quad \left. + \frac{k''_\perp(\omega)}{\omega \mu_0} \sin[\omega t - k'_\perp(\omega)y] \right\} E_\perp(\omega),\end{aligned}\quad (2.32c)$$

$$\begin{aligned}\mathbf{h}_\parallel(y, t) &= \mathbf{i}_1 \exp[-k''_\parallel(\omega)y] \left\{ \frac{k'_\parallel(\omega)}{\omega \mu_0} \cos[\omega t - k'_\parallel(\omega)y] \right. \\ &\quad \left. + \frac{k''_\parallel(\omega)}{\omega \mu_0} \sin[\omega t - k'_\parallel(\omega)y] \right\} E_\parallel(\omega).\end{aligned}\quad (2.32d)$$

These are equations of damped positive traveling waves, i.e., waves along the positive z (or y) axis, with phase velocities,

$$\nu_{\text{ph}\perp}(\omega) = \frac{\omega}{k'_\perp(\omega)} = \frac{c_0}{\text{Re } \sqrt{\epsilon_1(\omega)}} = \frac{c_0}{n'_1(\omega)}, \quad (2.33a)$$

$$\nu_{\text{ph}\parallel}(\omega) = \frac{\omega}{k'_\parallel(\omega)} = \frac{c_0}{\text{Re } \sqrt{\epsilon_3(\omega)}} = \frac{c_0}{n'_3(\omega)}. \quad (2.33b)$$

These relations involve the real parts of the appropriate complex refractive indices,

$$n_1(\omega) \equiv n'_1(\omega) - i n''_1(\omega) = \sqrt{\epsilon_1(\omega)}, \quad (2.34a)$$

$$n_3(\omega) \equiv n'_3(\omega) - i n''_3(\omega) = \sqrt{\epsilon_3(\omega)}, \quad (2.34b)$$

in which the positive root is to be taken for positive traveling waves. If we express the complex, relative permittivity in the form

$$\epsilon(\omega) = |\epsilon(\omega)| \exp(-i\delta) \quad (2.35)$$

with

$$|\epsilon(\omega)| = \sqrt{[\epsilon'(\omega)]^2 + [\epsilon''(\omega)]^2} \quad (2.36)$$

and

$$\delta = \arctan[\epsilon''(\omega)/\epsilon'(\omega)] \quad (2.37)$$

then

$$\begin{aligned}n(\omega) &= \sqrt{|\epsilon(\omega)|} \exp(-i\delta/2) \\ &= |n(\omega)| [\cos(\delta/2) - i \sin(\delta/2)].\end{aligned}\quad (2.38)$$

Because, for a nonmagnetic medium, δ must lie somewhere in the range from 0 to π it follows that $n''(\omega)$ can never be negative. Corresponding to Eqs. (2.28) we have

$$[n'(\omega)]^2 - [n''(\omega)]^2 = \epsilon'(\omega) \quad (2.39a)$$

and

$$2n'(\omega)n''(\omega) = \epsilon''(\omega). \quad (2.39b)$$

B. Propagation within the test cell

The analysis of the passage of radiation through the test cell is made difficult by the presence of the windows (see Fig. 2) which give rise to multiple reflections within the windows themselves and within the sample. An exact analysis would require the calculation of the reflection and transmission coefficients at each interface in the cell. Such calculations would require detailed knowledge of the optical properties of the windows of the cell and its contents. Unfortunately this information is lacking. However, as we shall show, it is possible to evade this difficulty when the sample is strongly absorbing.

The electric field within the sample is the sum of the fields of a succession of damped forward and backward traveling waves.

We shall take the origin of the coordinates to be just inside the sample as indicated in Fig. 2. The sample lies between the two 1-3 (x - z) planes passing through the points $y=0$, and $y=L$, L being the width of the cell. We shall treat the interfaces as infinite planes. It is permissible to consider each mode of propagation separately. We shall, first, concentrate on the mode in which the electric field is perpendicular to the 3 (or z) axis. If the amplitude of the electric field entering, and just inside, the sample is denoted by

$$\mathbf{E}_\perp(\omega, 0) = \mathbf{E}_\perp(\omega) = \mathbf{i}_1 E_\perp(\omega), \quad (2.40)$$

then in view of the results obtained in Sec II A, the electric field on some plane y within the sample may be expressed in the following manner:

$$\mathbf{e}_\perp(y, t) = \mathbf{i}_1 \text{Re } E_\perp(\omega, y) e^{i\omega t}, \quad (2.41)$$

in which

$$\begin{aligned} E_\perp(\omega, y) = & E_\perp(\omega) \{ \varepsilon^{-\iota k_\perp y} + R_\perp(\omega) \varepsilon^{-\iota k_\perp (2L-y)} \\ & + [R_\perp(\omega)]^2 \varepsilon^{-\iota k_\perp (2L+y)} + [R_\perp(\omega)]^3 \varepsilon^{-\iota k_\perp (3L-y)} \\ & + [R_\perp(\omega)]^4 \varepsilon^{-\iota k_\perp (3L+y)} + \dots \} \end{aligned} \quad (2.42)$$

and $R_\perp(\omega)$ takes account of the reflection at the sample-window interface, including the multiple reflections within the window. In a similar way we have for the magnetizing force vector

$$\mathbf{h}_\perp(y, t) = -\mathbf{i}_3 \text{Re } H_\perp(\omega, y) e^{i\omega t}, \quad (2.43)$$

where, in view of Eq. (2.30a),

$$\begin{aligned} H_\perp(\omega, y) = & \frac{k_\perp(\omega)}{\omega \mu_0} E_\perp(\omega) \{ \varepsilon^{-\iota k_\perp y} - R_\perp(\omega) \varepsilon^{-\iota k_\perp (2L-y)} \\ & + [R_\perp(\omega)]^2 \varepsilon^{-\iota k_\perp (2L+y)} - [R_\perp(\omega)]^3 \varepsilon^{-\iota k_\perp (3L-y)} \\ & + [R_\perp(\omega)]^4 \varepsilon^{-\iota k_\perp (3L+y)} + \dots \}. \end{aligned} \quad (2.44)$$

Note that the odd powers of $R_\perp(\omega)$ now have a negative sign because these terms arise from backward waves for which the magnetizing force vector is the reverse of that in a forward wave.

As we shall later require the derivatives of $E_\perp(\omega, y)$ and of $H_\perp(\omega, y)$ with respect to y , it is convenient to give their values here

$$\begin{aligned} \frac{\partial E_\perp(\omega, y)}{\partial y} = & [-\iota k_\perp(\omega)] E_\perp(\omega) \{ \varepsilon^{-\iota k_\perp y} \\ & - R_\perp(\omega) \varepsilon^{-\iota k_\perp (2L-y)} + [R_\perp(\omega)]^2 \varepsilon^{-\iota k_\perp (2L+y)} \\ & - [R_\perp(\omega)]^3 \varepsilon^{-\iota k_\perp (3L-y)} \\ & + [R_\perp(\omega)]^4 \varepsilon^{-\iota k_\perp (3L+y)} + \dots \} \end{aligned} \quad (2.45)$$

and using Eq. (2.30a) we obtain

$$\frac{\partial E_\perp(\omega, y)}{\partial y} = -\iota \omega \mu_0 H_\perp(\omega, y). \quad (2.46)$$

In the same way we find from Eqs. (2.42) and (2.44) that

$$\frac{\partial H_\perp(\omega, y)}{\partial y} = -\iota k_\perp^2(\omega) E_\perp(\omega, y). \quad (2.47)$$

In the following section, where we treat the Poynting vector, we shall require the value of

$$\begin{aligned} \frac{\partial}{\partial y} E_\perp(\omega, y) H_\perp^*(\omega, y) = & H_\perp^*(\omega, y) \frac{\partial}{\partial y} E_\perp(\omega, y) \\ & + E_\perp(\omega, y) \frac{\partial}{\partial y} H_\perp^*(\omega, y) \end{aligned} \quad (2.48)$$

where $*$ indicates the complex conjugate. By means of Eqs. (2.46) and (2.47) we get

$$\begin{aligned} \frac{\partial}{\partial y} E_\perp(\omega, y) H_\perp^*(\omega, y) = & -\iota \omega \mu_0 |H_\perp(\omega, y)|^2 + \iota \frac{k_\perp^{*2}(\omega)}{\omega \mu_0} |E_\perp(\omega, y)|^2 \\ = & -\frac{2k'_\perp(\omega)k''_\perp(\omega)}{\omega \mu_0} |E_\perp(\omega, y)|^2 - \iota \frac{1}{\omega \mu_0} [(\omega \mu_0)^2 |H_\perp(\omega, y)|^2 \\ & - \{[k'_\perp(\omega)]^2 - [k''_\perp(\omega)]^2\} |E_\perp(\omega, y)|^2]. \end{aligned} \quad (2.49)$$

Note that the first term in the second part of this equation is entirely real, and the second term is entirely imaginary.

The series which appear in Eqs. (2.42) and (2.44) can be summed, and we find it possible to write these equations in the following way:

$$E_\perp(\omega, y) = E_\perp(\omega) \varepsilon^{-\iota k_\perp y} C_{\perp, E}(\omega, y) \quad (2.50a)$$

and

$$\begin{aligned} H_\perp(\omega, y) = & H_\perp(\omega) \varepsilon^{-\iota k_\perp y} C_{\perp, H}(\omega, y) \\ = & \frac{k_\perp(\omega)}{\omega \mu_0} E_\perp(\omega) \varepsilon^{-\iota k_\perp y} C_{\perp, H}(\omega, y). \end{aligned} \quad (2.50b)$$

The coefficients $C_{\perp, E}(\omega, y)$, and $C_{\perp, H}(\omega, y)$ have the values,

$$C_{\perp, E}(\omega, y) = \left[1 + R_\perp(\omega) \frac{[R_\perp(\omega) + \varepsilon^{i2k_\perp y}] \varepsilon^{-i2k_\perp L}}{\{1 - [R_\perp(\omega)]^2 \varepsilon^{-i2k_\perp L}\}} \right] \quad (2.51a)$$

and

$$C_{\perp, H}(\omega, y) = \left[1 + R_\perp(\omega) \frac{[R_\perp(\omega) - \varepsilon^{i2k_\perp y}] \varepsilon^{-i2k_\perp L}}{\{1 - [R_\perp(\omega)]^2 \varepsilon^{-i2k_\perp L}\}} \right]. \quad (2.51b)$$

It is clear from these last equations that the sum

$$C_{\perp, E}(\omega, y) + C_{\perp, H}(\omega, y) = 2 \left[1 + \frac{[R_\perp(\omega)]^2 \varepsilon^{-i2k_\perp L}}{\{1 - [R_\perp(\omega)]^2 \varepsilon^{-i2k_\perp L}\}} \right] \quad (2.52)$$

and is independent of y .

Finally we note that the relations for the mode in which the electric vector is parallel to the 3 (or z) axis may be obtained from the foregoing equations by replacing the subscript $\perp$ by the subscript $\parallel$ throughout, and by changing the vector $-\mathbf{i}_3$ in Eq. (2.43) to the vector $\mathbf{i}_1$.

FIG. 5. The total electric vector E_0 lies in the 1-3 plane and makes an angle θ with the 3 axis.

III. THE POYNTING VECTOR

The time-dependent Poynting vector $\mathbf{s}(\mathbf{r}, t)$ is defined by the equation

$$\mathbf{s}(\mathbf{r}, t) \equiv \mathbf{e}(\mathbf{r}, t) \times \mathbf{h}(\mathbf{r}, t) \quad (3.1)$$

and is a measure of the power-flux density in an electromagnetic field. Equation (3.1) implies that for a plane electromagnetic wave, the Poynting vector $\mathbf{s}$ is parallel to the propagation vector $\mathbf{k}$.

To calculate the Poynting vector in the test cell we must take into consideration the angle θ between the electric vector of the incoming radiation and the 3 (or z) axis of the test cell. Thus the electric field, $\mathbf{E}_{\text{in}}(\omega, \theta) = \mathbf{E}_0(\omega)$, in the wave approaching the test cell has two components (see Fig. 5),

$$\mathbf{E}_{\text{in}}(\omega, \theta) = (\mathbf{i}_1 \sin \theta + \mathbf{i}_3 \cos \theta) E_0(\omega). \quad (3.2)$$

Each component of this approaching wave creates its own plane wave with propagation vectors: $\mathbf{k}_{\perp}(\omega) = \mathbf{i}_2 k_{\perp}(\omega)$ and $\mathbf{k}_{\parallel}(\omega) = \mathbf{i}_3 k_{\parallel}(\omega)$. The magnitudes of the electric vectors of the waves just entering the test cell will be directly proportional to the components of $\mathbf{E}_0(\omega)$. Therefore we write

$$\mathbf{E}_{\perp}(\omega, 0) = \mathbf{E}_{\perp}(\omega) = \mathbf{i}_1 F_{\perp, E}(\omega) E_0(\omega) \sin \theta, \quad (3.3a)$$

$$\mathbf{E}_{\parallel}(\omega, 0) = \mathbf{E}_{\parallel}(\omega) = \mathbf{i}_3 F_{\parallel, E}(\omega) E_0(\omega) \cos \theta, \quad (3.3b)$$

in which the coefficients $F_{\perp, E}(\omega)$ and $F_{\parallel, E}(\omega)$ take account of the properties of the windows and of the window-sample interface. The magnitudes of the magnetic vectors, $\mathbf{H}_{\perp}(\omega)$ and $\mathbf{H}_{\parallel}(\omega)$, may be calculated from the corresponding electric vectors by means of Eqs. (2.30), and their orientations are indicated in Fig. 4.

Using the relations obtained in Sec. II B, we find that the total electric field vector within the sample is

$$\begin{aligned} \mathbf{e}(\mathbf{r}, t) = & \text{Re} \{ \mathbf{i}_1 E_{\perp}(\omega) C_{\perp, E}(\omega, y) \varepsilon^{i(\omega t - k_{\perp} y)} \\ & + \mathbf{i}_3 E_{\parallel}(\omega) C_{\parallel, E}(\omega, y) \varepsilon^{i(\omega t - k_{\parallel} y)} \} \\ = & \text{Re} \varepsilon^{i\omega t} E_{\text{in}}(\omega) \{ \mathbf{i}_1 \sin \theta F_{\perp, E}(\omega) C_{\perp, E}(\omega, y) \varepsilon^{-ik_{\perp} y} \\ & + \mathbf{i}_3 \cos \theta F_{\parallel, E}(\omega) C_{\parallel, E}(\omega, y) \varepsilon^{-ik_{\parallel} y} \} \end{aligned} \quad (3.4a)$$

and the magnetizing force vector is

$$\begin{aligned} \mathbf{h}(\mathbf{r}, t) = & \text{Re} \{ -\mathbf{i}_3 H_{\perp}(\omega) C_{\perp, H}(\omega, y) \varepsilon^{i(\omega t - k_{\perp} y)} \\ & + \mathbf{i}_1 H_{\parallel}(\omega) C_{\parallel, H}(\omega, y) \varepsilon^{i(\omega t - k_{\parallel} y)} \} \\ = & (\omega \mu_0)^{-1} \text{Re} \varepsilon^{i\omega t} \{ -\mathbf{i}_3 k_{\perp}(\omega) E_{\perp}(\omega) \end{aligned}$$

$$\begin{aligned} & \times C_{\perp, H}(\omega, y) \varepsilon^{-ik_{\perp} y} \\ & + \mathbf{i}_1 k_{\parallel}(\omega) E_{\parallel}(\omega) C_{\parallel, H}(\omega, y) \varepsilon^{-ik_{\parallel} y} \} \\ = & (\omega \mu_0)^{-1} \text{Re} \varepsilon^{i\omega t} E_{\text{in}}(\omega) \\ & \times \{ -\mathbf{i}_3 \sin \theta k_{\perp}(\omega) F_{\perp, E}(\omega) C_{\perp, H}(\omega, y) \varepsilon^{-ik_{\perp} y} \\ & + \mathbf{i}_1 \cos \theta k_{\parallel}(\omega) F_{\parallel, E}(\omega) C_{\parallel, H}(\omega, y) \varepsilon^{-ik_{\parallel} y} \}. \end{aligned} \quad (3.4b)$$

The time average over one cycle, of duration $T = 2\pi/\omega$, of the product of the two sinusoidal functions of time,

$$\begin{aligned} a(t) &= \text{Re} A(\omega) \varepsilon^{i\omega t} \\ &= |A(\omega)| \text{Re} \varepsilon^{i\alpha(\omega)} \varepsilon^{i\omega t} \\ &= |A(\omega)| \cos[\omega t + \alpha(\omega)], \\ b(t) &= \text{Re} B(\omega) \varepsilon^{i\omega t} \\ &= |B(\omega)| \text{Re} \varepsilon^{i\beta(\omega)} \varepsilon^{i\omega t} \\ &= |B(\omega)| \cos[\omega t + \beta(\omega)], \end{aligned} \quad (3.5)$$

is given by

$$\begin{aligned} \overline{a(t)b(t)}^T &= \frac{1}{T} \int_0^T dt a(t)b(t) \\ &= \frac{1}{2} \text{Re} A(\omega) B^*(\omega) \\ &= \frac{1}{2} \text{Re} A^*(\omega) B(\omega) \\ &= \frac{1}{2} |A(\omega)| |B(\omega)| \cos[\alpha(\omega) - \beta(\omega)]. \end{aligned} \quad (3.6)$$

With this result we find that the time average of the Poynting vector with $\mathbf{e}$ and $\mathbf{h}$ given by Eqs. (3.4a) and (3.4b) is

$$\begin{aligned} \mathbf{S}(y) = & \mathbf{i}_2 S(y) = \overline{\mathbf{e}(r, t) \times \mathbf{h}(r, t)}^T \\ = & \mathbf{i}_2 \frac{1}{2} (\omega \mu_0)^{-1} \text{Re} |\mathbf{E}_0(\omega)|^2 \{ \varepsilon^{-2k_{\perp}'' y} \sin^2 \theta \\ & \times [F_{\perp, E}(\omega)]^2 k_{\perp}^*(\omega) C_{\perp, E}(\omega, y) C_{\perp, H}^*(\omega, y)] \\ & + \varepsilon^{-2k_{\parallel}'' y} \cos^2 \theta [F_{\parallel, E}(\omega)]^2 k_{\parallel}^*(\omega) C_{\parallel, E}(\omega, y) C_{\parallel, H}^*(\omega, y) \}. \end{aligned} \quad (3.7)$$

We may assume that $E_0(\omega)$ is a real quantity so that

$$S(y) = \frac{1}{2} \sqrt{\frac{\varepsilon_0}{\mu_0}} |\mathbf{E}_0(\omega)|^2 [\cos^2 \theta G_{\parallel}(\omega, y) + \sin^2 \theta G_{\perp}(\omega, y)] \quad (3.8)$$

with

$$\begin{aligned} G_{\parallel}(\omega, y) &= (c_0/\omega) \exp[-2k_{\parallel}''(\omega)y] \\ &\times [F_{\parallel, E}(\omega)]^2 \text{Re} [k_{\parallel}^*(\omega) C_{\parallel, E}(\omega, y) C_{\parallel, H}^*(\omega, y)] \\ &= \exp[-2k_{\parallel}''(\omega)y] \\ &\times [F_{\parallel, E}(\omega)]^2 \text{Re} k_0^{-1} [k_{\parallel}^*(\omega) C_{\parallel, E}(\omega, y) C_{\parallel, H}^*(\omega, y)] \end{aligned} \quad (3.9a)$$

and

$$\begin{aligned}
 G_{\perp}(\omega, y) &= (c_0/\omega) \exp[-2k''_{\perp}(\omega)y] \\
 &\times |F_{\perp,E}(\omega)|^2 \operatorname{Re}[k_{\perp}^*(\omega) C_{\perp,E}(\omega, y) C_{\perp,H}^*(\omega, y)] \\
 &= \exp[-2k''_{\perp}(\omega)y] \\
 &\times |F_{\perp,E}(\omega)|^2 \operatorname{Re} k_0^{-1} [k_{\perp}^*(\omega) C_{\perp,E}(\omega, y) C_{\perp,H}^*(\omega, y)].
 \end{aligned} \quad (3.9b)$$

The mean-power density in the beam approaching the test cell is

$$P_{\text{in}}(\omega, \theta) = P_{\text{ref}}(\omega, \theta) = \frac{1}{2} \sqrt{\frac{\epsilon_0}{\mu_0}} |\mathbf{E}_0(\omega)|^2, \quad (3.10)$$

so that the mean-power density entering the sample is

$$S(0) = P_{\text{in}}(\omega, \theta) [\cos^2 \theta G_{\parallel}(\omega, 0) + \sin^2 \theta G_{\perp}(\omega, 0)] \quad (3.11)$$

and the mean-power density leaving the sample is

$$S(L) = P_{\text{in}}(\omega, \theta) [\cos^2 \theta G_{\parallel}(\omega, L) + \sin^2 \theta G_{\perp}(\omega, L)]. \quad (3.12)$$

The mean-power density emerging from the cell, $P_{\text{out}}(\omega, \theta)$, will be directly proportional to $S(L)$ but slightly reduced in intensity by the effect of the window of the test cell. Hence we write

$$\begin{aligned}
 P_{\text{out}}(\omega, \theta) &= P_{\text{in}}(\omega, \theta) [\cos^2 \theta w_{\parallel}(\omega) G_{\parallel}(\omega, L) \\
 &+ \sin^2 \theta w_{\perp}(\omega) G_{\perp}(\omega, L)].
 \end{aligned} \quad (3.13)$$

The factors $w_{\parallel}(\omega)$ and $w_{\perp}(\omega)$ take account of the small absorption of energy as the radiation passes through the exit window of the test cell.

Recalling Eq. (1.4), we see that Eq. (3.13) leads to the following expression for the transmittance of the test cell:

$$T_{\text{cell}}(\omega, \theta) = \frac{P_{\text{out}}(\omega, \theta)}{P_{\text{in}}(\omega, \theta)} = [\Lambda_{\parallel}(\omega) \cos^2 \theta + \Lambda_{\perp}(\omega) \sin^2 \theta], \quad (3.14)$$

where

$$\begin{aligned}
 \Lambda_{\parallel}(\omega) &= \exp[-2k''_{\parallel}(\omega)L] w_{\parallel}(\omega) \\
 &\times |F_{\parallel,E}(\omega)|^2 \operatorname{Re} k_0^{-1} [k_{\parallel}^*(\omega) C_{\parallel,E}(\omega, L) C_{\parallel,H}^*(\omega, L)]
 \end{aligned} \quad (3.15a)$$

and

$$\begin{aligned}
 \Lambda_{\perp}(\omega) &= \exp[-2k''_{\perp}(\omega)L] w_{\perp}(\omega) \\
 &\times |F_{\perp,E}(\omega)|^2 \operatorname{Re} k_0^{-1} [k_{\perp}^*(\omega) C_{\perp,E}(\omega, L) C_{\perp,H}^*(\omega, L)].
 \end{aligned} \quad (3.15b)$$

The form of Eq. (3.14) is that first proposed by Jang *et al.*⁶ Spatial dispersion is ignored in this analysis.¹⁴

IV. THE DENSITY OF POWER ABSORPTION

Consider two parallel planes perpendicular to the 2 (or y) axis and separated by a small distance δy , as shown in Fig. 6. The time-averaged Poynting vector S was defined in Eq.

FIG. 6. The Poynting vector $\mathbf{s}$ parallel to the 2 axis, decreases in magnitude as the wave progresses from the 1-3 plane at y to the 1-3 plane at $y + \delta y$.

(3.7). The magnitude of S at the plane $(y + \delta y)$ may be expressed in terms of its magnitude at the plane y by means of the Taylor expansion,

$$S(y + \delta y) = S(y) + \delta y \frac{\partial S(y)}{\partial y} + (\dots). \quad (4.1)$$

The mean power absorbed in the slab of thickness δy and of unit (1 m^2) cross-sectional area is

$$\begin{aligned}
 W(y) \delta y &= S(y) - S(y + \delta y) \\
 &\cong S(y) - S(y) - \delta y \frac{\partial S(y)}{\partial y} \cong -\delta y \frac{\partial S(y)}{\partial y}.
 \end{aligned} \quad (4.2)$$

In the limit when δy is vanishingly small we obtain

$$W(y) = -\frac{\partial S(y)}{\partial y}. \quad (4.3)$$

Here $W(y)$ is the mean power absorbed per unit volume on the plane y . It is convenient here to split the time-averaged Poynting vector into two parts, $S_{\parallel}(y)$ and $S_{\perp}(y)$, for the two modes of propagation, so that in the notation of Sec. II B, we have

$$S_{\perp}(y) = e_{\perp}(y, t) h_{\perp}(y, t)^T = \frac{1}{2} \operatorname{Re} E_{\perp}(\omega, y) H_{\perp}^*(\omega, y) \quad (4.4)$$

with a similar expression for $S_{\parallel}(y)$. From Eq. (2.49) and (4.4) we get

$$\begin{aligned}
 W_{\perp}(y) &= -\frac{\partial S_{\perp}(y)}{\partial y} \\
 &= -\frac{1}{2} \operatorname{Re} \frac{\partial}{\partial y} [E_{\perp}(\omega, y) H_{\perp}^*(\omega, y)] \\
 &= \frac{1}{2} \frac{2k'_{\perp}(\omega) k''_{\perp}(\omega)}{\omega \mu_0} |E_{\perp}(\omega, y)|^2.
 \end{aligned} \quad (4.5)$$

By means of Eq. (2.28b) we see that

$$W_{\perp}(y) = \frac{1}{2} \omega \epsilon_0 \epsilon''_1(\omega) |E_{\perp}(\omega, y)|^2 \quad (4.6)$$

and hence

$$\begin{aligned}
 W(y) &= W_{\parallel}(y) + W_{\perp}(y) \\
 &= \frac{1}{2} \omega \epsilon_0 [\epsilon''_3(\omega) |E_{\parallel}(\omega, y)|^2 + \epsilon''_1(\omega) |E_{\perp}(\omega, y)|^2]
 \end{aligned} \quad (4.7)$$

in agreement with standard dielectric theory.¹⁶

FIG. 7. The angular coordinates β and γ specify the orientation of a molecule with respect to the axes 1, 2, and 3.

V. MICROSCOPIC CONSIDERATIONS

As a model we assume that the medium consists of long, rodlike molecules with *only* an axial electrical polarizability,

$$\alpha(\omega) = \alpha'(\omega) - i\alpha''(\omega). \quad (5.1)$$

The problem of the local field arises;¹⁶ given the postulated shape of the molecules, it is not unreasonable to assume that the local field is equal to the macroscopic electric field. (The depolarizing factor for an exceedingly prolate spheroid is negligibly small.¹⁶)

If the angular coordinates of a molecule are β and γ as in Fig. 7, the orientational distribution function $\rho(\beta, \gamma)$ will be taken to be⁷

$$4\pi\rho(\beta, \gamma) = 1 + 3A \sin \beta \sin \gamma + 5BP_2(\cos \beta) + (15/4)[C \sin 2\beta \cos \gamma + D \sin^2 \beta \cos 2\gamma + E \sin 2\beta \sin \gamma + F \sin^2 \beta \sin 2\gamma], \quad (5.2)$$

Here A, B, C, D, E , and F are the expansion coefficients and serve as the order parameters with the maximum value of 1 in completely ordered systems. A measures the polar order and for the ferroelectric phase $\neq 0$ in the unwound state. B measures the order relative to the axis 3. C, D, E , and F are the four order parameters measuring the quadrupolar order. $P_2(\cos \beta) = (1/2)(3 \cos^2 \beta - 1)$ is a Legendre polynomial.

For a static electric field $\mathbf{e} = \mathbf{i}_1 e_1$, the polarization components are

$$p_{1,1} = \chi_{1,1} \epsilon_0 e_1 = \frac{N}{V} \langle \alpha_s e_1 \cos^2 \gamma \sin^2 \beta \rangle = (\epsilon_{1,1} - 1) \epsilon_0 e_1, \quad (5.3a)$$

$$p_{2,1} = \chi_{2,1} \epsilon_0 e_1 = \frac{N}{V} \langle \alpha_s e_1 \cos \gamma \sin \gamma \sin^2 \beta \rangle = (\epsilon_{2,1} - 1) \epsilon_0 e_1, \quad (5.3b)$$

$$p_{3,1} = \chi_{3,1} \epsilon_0 e_1 = \frac{N}{V} \langle \alpha_s e_1 \cos \gamma \sin \gamma \sin \beta \rangle = (\epsilon_{3,1} - 1) \epsilon_0 e_1. \quad (5.3c)$$

In these equations N denotes the number of molecules in volume V ,

$$\alpha_s = \alpha'(0) \quad (5.4)$$

and the angular brackets indicate an average given by

$$\langle (\dots) \rangle = \int_0^{2\pi} d\gamma \int_0^\pi d\beta \sin \beta \rho(\beta, \gamma) (\dots). \quad (5.5)$$

Similarly, for $\mathbf{e} = \mathbf{i}_2 e_2$ we have

$$p_{1,2} = \chi_{1,2} \epsilon_0 e_2 = \frac{N}{V} \langle \alpha_s e_2 \sin \gamma \cos \gamma \sin^2 \beta \rangle = (\epsilon_{1,2} - 1) \epsilon_0 e_2, \quad (5.6a)$$

$$p_{2,2} = \chi_{2,2} \epsilon_0 e_2 = \frac{N}{V} \langle \alpha_s e_2 \sin^2 \gamma \sin^2 \beta \rangle = (\epsilon_{2,2} - 1) \epsilon_0 e_2, \quad (5.6b)$$

$$p_{3,2} = \chi_{3,2} \epsilon_0 e_2 = \frac{N}{V} \langle \alpha_s e_2 \sin \gamma \sin \beta \cos \beta \rangle = (\epsilon_{3,2} - 1) \epsilon_0 e_2, \quad (5.6c)$$

and for $\mathbf{e} = \mathbf{i}_3 e_3$,

$$p_{1,3} = \chi_{1,3} \epsilon_0 e_3 = \frac{N}{V} \langle \alpha_s e_3 \cos \gamma \sin \beta \cos \beta \rangle = (\epsilon_{1,3} - 1) \epsilon_0 e_3, \quad (5.7a)$$

$$p_{2,3} = \chi_{2,3} \epsilon_0 e_3 = \frac{N}{V} \langle \alpha_s e_3 \sin \gamma \sin \beta \cos \beta \rangle = (\epsilon_{2,3} - 1) \epsilon_0 e_3, \quad (5.7b)$$

$$p_{3,3} = \chi_{3,3} \epsilon_0 e_3 = \frac{N}{V} \langle \alpha_s e_3 \cos^2 \beta \rangle = (\epsilon_{3,3} - 1) \epsilon_0 e_3. \quad (5.7c)$$

Clearly

$$\chi_{\alpha,\beta} = \chi_{\beta,\alpha}, \quad \epsilon_{\alpha,\beta} = \epsilon_{\beta,\alpha}. \quad (5.8)$$

For an electric field

$$\mathbf{e} = \mathbf{i}_1 e_1 + \mathbf{i}_2 e_2 + \mathbf{i}_3 e_3, \quad (5.9)$$

the induced-polarization vector

$$\mathbf{p} = \mathbf{i}_1 p_1 + \mathbf{i}_2 p_2 + \mathbf{i}_3 p_3, \quad (5.10)$$

where the components of $\mathbf{p}$ are given by the matrix equation:

$$\begin{bmatrix} p_1 \\ p_2 \\ p_3 \end{bmatrix} = \begin{bmatrix} p_{1,1} + p_{1,2} + p_{1,3} \\ p_{2,1} + p_{2,2} + p_{2,3} \\ p_{3,1} + p_{3,2} + p_{3,3} \end{bmatrix} = \epsilon_0 \begin{bmatrix} \chi_{1,1} & \chi_{1,2} & \chi_{1,3} \\ \chi_{2,1} & \chi_{2,2} & \chi_{2,3} \\ \chi_{3,1} & \chi_{3,2} & \chi_{3,3} \end{bmatrix} \begin{bmatrix} e_1 \\ e_2 \\ e_3 \end{bmatrix}. \quad (5.11)$$

On evaluating the averages in Eqs. (5.3), (5.6), and (5.7) by means of Eq. (5.5) (see the Appendix) we find that

$$\begin{bmatrix} \chi_{1,1} & \chi_{1,2} & \chi_{1,3} \\ \chi_{2,1} & \chi_{2,2} & \chi_{2,3} \\ \chi_{3,1} & \chi_{3,2} & \chi_{3,3} \end{bmatrix} = \frac{N\alpha_s}{\epsilon_0 V} \begin{bmatrix} \left[\frac{1-B}{3} + \frac{D}{2}\right] & \frac{F}{2} & \frac{C}{2} \\ \frac{F}{2} & \left[\frac{1-B}{3} - \frac{D}{2}\right] & \frac{E}{2} \\ \frac{C}{2} & \frac{E}{2} & \left[\frac{1+2B}{3}\right] \end{bmatrix}. \quad (5.12)$$

The condition that the susceptibility matrix, $[\chi_{\alpha,\beta}]$, be diagonal is that

$$C = E = F = 0 \quad (5.13)$$

and under this condition we get

$$\epsilon_1(\omega) - 1 = \frac{N\alpha(\omega)}{\epsilon_0 V} \left[\frac{1-B}{3} + \frac{D}{2} \right], \quad (5.14a)$$

$$\epsilon_2(\omega) - 1 = \frac{N\alpha(\omega)}{\epsilon_0 V} \left[\frac{1-B}{3} - \frac{D}{2} \right], \quad (5.14b)$$

$$\epsilon_3(\omega) - 1 = \frac{N\alpha(\omega)}{\epsilon_0 V} \left[\frac{1+2B}{3} \right]. \quad (5.14c)$$

Hence the susceptibility of the sample in its isotropic state is given by

$$\begin{aligned} \chi_{\text{iso}}(\omega) &\equiv \epsilon_{\text{iso}}(\omega) - 1 \\ &= \frac{1}{3}[\epsilon_1(\omega) + \epsilon_2(\omega) + \epsilon_3(\omega)] \\ &= \frac{N\alpha(\omega)}{3\epsilon_0 V} \left[\frac{1-B}{3} + \frac{D}{2} + \frac{1-B}{3} - \frac{D}{2} + \frac{1+2B}{3} \right] \\ &= \frac{N\alpha(\omega)}{3\epsilon_0 V} \end{aligned} \quad (5.15)$$

as expected.

Explicit expressions for $k_{\perp}(\omega)$ and $k_{\parallel}(\omega)$ in terms of $\epsilon_{\text{iso}}(\omega)$, B , and D may be obtained by combining Eq. (2.17) with the results of this section.

VI. DISCUSSION

In order to simplify the discussion we shall assume that the windows of the test cell are only weakly reflecting and hence that their transmittances are close to unity. Under these assumptions Eqs. (3.15a) and (3.15b) simplify considerably and take the approximate forms,

$$\begin{aligned} \Lambda_{\parallel}(\omega) &\equiv k_0^{-1} k'_{\parallel}(\omega) \exp[-2k''_{\parallel}(\omega)L] \\ &\equiv n'_3(\omega) \exp[-2(2\pi/\lambda_0)n''_3(\omega)L], \end{aligned} \quad (6.1a)$$

$$\begin{aligned} \Lambda_{\perp}(\omega) &\equiv k_0^{-1} k'_{\perp}(\omega) \exp[-2k''_{\perp}(\omega)L] \\ &\equiv n'_1(\omega) \exp[-2(2\pi/\lambda_0)n''_1(\omega)L]. \end{aligned} \quad (6.1b)$$

The absorbance $\mathcal{A}(\omega, \theta)$, defined by Eq. (1.5), has the following particular values:

$$\mathcal{A}(\omega, 0) = \mathcal{A}_{\parallel}(\omega) = -\log_{10} n'_3(\omega) + 2(2\pi L/\lambda_0) n''_3(\omega) \log_{10} \epsilon, \quad (6.2a)$$

$$\begin{aligned} \mathcal{A}(\omega, \pi/2) &= \mathcal{A}_{\perp}(\omega) \\ &= -\log_{10} n'_1(\omega) + 2(2\pi L/\lambda_0) n''_1(\omega) \log_{10} \epsilon. \end{aligned} \quad (6.2b)$$

Kocot *et al.*⁷ assume that the modulus squared of the electric field within the sample varies with distance according to the equations:

$$|E_{\parallel}(\omega, y)|^2 = |E_{\parallel}(\omega)|^2 10^{-\mathcal{A}_{\parallel}(\omega)(y/L)}, \quad (6.3)$$

$$|E_{\perp}(\omega, y)|^2 = |E_{\perp}(\omega)|^2 10^{-\mathcal{A}_{\perp}(\omega)(y/L)}.$$

With the approximations introduced in this section, our versions of these equations, based on Eq. (2.50a), read

$$|E_{\parallel}(\omega, y)|^2 = |E_{\parallel}(\omega)|^2 \exp[-2k''_{\parallel}(\omega)y] \quad (6.4)$$

$$|E_{\perp}(\omega, y)|^2 = |E_{\perp}(\omega)|^2 \exp[-2k''_{\perp}(\omega)y].$$

For Eqs. (6.3) and (6.4) to be compatible it is necessary that

$$\mathcal{A}_{\parallel}(\omega) = 2k''_{\parallel}(\omega)L \log_{10} \epsilon, \quad \mathcal{A}_{\perp}(\omega) = 2k''_{\perp}(\omega)L \log_{10} \epsilon. \quad (6.5)$$

This condition could be satisfied if the absorption was so strong as to make the $\log_{10} n'(\omega)$ term, in Eq. (6.2), small in comparison with the term in $n''(\omega)$. Kocot *et al.*⁷ also derived an expression for $P_{\text{abs}}(\omega, \theta)$, the mean-power density absorbed by the cell, which seems to be in error.

From the discussion in Sec. I and the definition in Eq. (1.4) it is clear that with

$$\frac{P_{\text{abs}}(\omega, \theta)}{P_{\text{in}}(\omega, \theta)} = \frac{P_{\text{in}}(\omega, \theta) - P_{\text{out}}(\omega, \theta)}{P_{\text{in}}(\omega, \theta)} = 1 - T_{\text{cell}}(\omega, \theta), \quad (6.6)$$

then

$$\begin{aligned} 1 - T_{\text{cell}}(\omega, \theta) &= 1 - [\Lambda_{\parallel}(\omega) \cos^2 \theta + \Lambda_{\perp}(\omega) \sin^2 \theta] \\ &= [1 - \Lambda_{\parallel}(\omega)] \cos^2 \theta + [1 - \Lambda_{\perp}(\omega)] \sin^2 \theta, \end{aligned} \quad (6.7)$$

which differs from the result obtained by Kocot *et al.* [Ref. 7, Eq. (8)]. For the case of *weak* absorption, we have the approximation,

$$\begin{aligned} P_{\text{abs}}(\omega, \theta) &\equiv P_{\text{in}}(\omega, \theta) \frac{2\pi L}{\lambda_0} \{ [2n'_3(\omega)n''_3(\omega)] \cos^2 \theta \\ &\quad + [2n'_1(\omega)n''_1(\omega)] \sin^2 \theta \} \\ &= P_{\text{in}}(\omega, \theta) \frac{2\pi L}{\lambda_0} [\epsilon''_3(\omega) \cos^2 \theta + \epsilon''_1(\omega) \sin^2 \theta]. \end{aligned} \quad (6.8)$$

When the sample is isotropic this equation becomes

$$P_{\text{abs}}^{(\text{iso})}(\omega, \theta) = P_{\text{in}}(\omega, \theta) \frac{2\pi L}{\lambda_0} \epsilon''_{\text{iso}}(\omega). \quad (6.9)$$

On dividing Eq. (6.8) by Eq. (6.9), we find, by using Eqs. (5.14) and (5.15), that

$$\frac{P_{\text{abs}}(\omega, \theta)}{P_{\text{abs}}^{(\text{iso})}(\omega, \theta)} = \left\{ [1 + 2B] \cos^2 \theta + \left[1 - B + \frac{3D}{2} \right] \sin^2 \theta \right\} \\ = 1 + B(2 - 3 \sin^2 \theta) + \frac{3D}{2} \sin^2 \theta. \quad (6.10)$$

This equation agrees with that obtained by Kocot *et al.* for $D=0$ [Ref. 7, Eq. (12)].

VII. CONCLUSIONS

We have developed the theory of an electromagnetic homogeneous plane-wave in a semi-infinite, optically anisotropic and absorbing medium. Based on this, and from first principles, we have derived the empirical equation first proposed by Jang, Park, Kim, and Clark⁶ for the IR absorbance as a function of the angle of polarization for normal incidence of an IR beam in a ferroelectric liquid crystalline sample aligned in the homogeneous configuration and the sample confined to lie between the windows of a cell. We find that this equation is not subject to the limitation that the IR birefringence of the sample should be zero. In the derivation, we considered the elements of the dielectric tensor to be complex and frequency dependent but neglected the explicit dependence on the wave vector. We thus assume that all values of the wavevector are allowed for the various frequencies. This is justified by the fact the smectic layer thickness (~ 3 nm) is much smaller than the shortest wavelength in the IR beam ($\sim 3 \mu\text{m}$). The absorbances along the direction of polarization of the IR beam, and normal to it, are expressed in terms of the properties of the materials and the dimensions of the cell. We remark that an earlier simplified attempt by Kocot, Wrzalik, and Vij⁷ to derive this formula was based on certain assumptions and in some cases the equations were in error. It has been shown that the order parameters in certain cases can also be calculated from the three different orthogonal components of the relative permittivity.

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APPENDIX: RESULTS OF MATHEMATICAL AVERAGES OF TRIGONO-METRIC FUNCTIONS OF THE ANGULAR COORDINATES OF A MOLECLUE

We list here the values of the averages used in the calculation of $\chi_{\alpha\beta}$ in Sec. V:

$$\langle \sin^2 \beta \cos^2 \gamma \rangle = \frac{1}{3} \left[1 - B + \frac{3D}{2} \right], \quad (A1)$$

$$\langle \sin^2 \beta \sin^2 \gamma \rangle = \frac{1}{3} \left[1 - B - \frac{3D}{2} \right], \quad (A2)$$

$$\langle \cos^2 \beta \rangle = \frac{1}{3} (1 + 2B), \quad (A3)$$

$$\langle \sin^2 \beta \sin \gamma \cos \gamma \rangle = \frac{F}{2}, \quad (A4)$$

$$\langle \sin \beta \cos \beta \sin \gamma \rangle = \frac{E}{2}, \quad (A5)$$

$$\langle \sin \beta \cos \beta \cos \gamma \rangle = \frac{C}{2}. \quad (A6)$$

¹B. J. Bulkin, *Liquid Crystals, the Fourth State of Matter*, edited by F. D. Saeva (Dekker, New York, 1975).

²A. Kocot, G. Kruk, R. Wrzalik, and J. K. Vij, *Liq. Cryst.*, **12**, 1005 (1992).

³K. Merkel, A. Kocot, J. K. Vij, G. H. Mehl, and T. Meyer, *J. Chem. Phys.* **121**, 5012 (2004).

⁴E. Hild, A. Kocot, J. K. Vij, and R. Zentel, *Liq. Cryst.* **16**, 783 (1994).

⁵K. H. Kim, K. Ishikawa, H. Takezoe, and A. Fukuda, *Phys. Rev. E* **51**, 2166 (1995).

⁶W. G. Jang, C. S. Park, J. E. MacLennan, K. H. Kim, and N. A. Clark, *Ferroelectrics* **180**, 213 (1996).

⁷A. Kocot, R. Wrzalik, and J. K. Vij, *Liq. Cryst.* **21**, 147 (1996).

⁸A. Kocot, R. Wrzalik, B. Orgasinska, T. S. Perova, J. K. Vij, and H. T. Nguyen, *Phys. Rev. E* **59**, 551 (1999).

⁹A. A. Sigarev, J. K. Vij, Yu. P. Panarin, and J. W. Goodby, *Phys. Rev. E* **62**, 2269 (2000); A. A. Sigarev, J. K. Vij, R. A. Lewis, M. Hird, and J. W. Goodby, *ibid.* **68**, 031707 (2003).

¹⁰W. G. Jang, C. S. Park, K. H. Kim, M. A. Glaser, and N. A. Clark, *Phys. Rev. E* **62**, 5027 (2000).

¹¹W. G. Jang, C. S. Park, and N. A. Clark, *Phys. Rev. E* **62**, 5154 (2000).

¹²A. Kocot, J. K. Vij, and T. S. Perova, *Adv. Chem. Phys.* **113**, 203 (2000); T. S. Perova, J. K. Vij, and A. Kocot *Adv. Liq. Cryst.* in a special issue of *Advances in Chemical Physics*, edited by J. K. Vij (series editors I. Prigogine and S. Rice) (John Wiley & Sons, New York) **113**, 341 (2000).

¹³L. D. Landau, E. M. Lifshitz, and L. P. Pitaevskii, *Electrodynamics of Continuous Media*, 2nd ed. (Pergamon, Oxford 1984), Chap. XI.

¹⁴H. Yuan, W. E. Palffy-Muhoray, and P. Palffy-Muhoray, *Mol. Cryst. Liq. Cryst. Sci. Technol., Sect. A* **358**, 311 (2001).

¹⁵H. Yuan, W. E. Palffy-Muhoray, and P. Palffy-Muhoray, *Phys. Rev. E* **61**, 3264 (2000).

¹⁶B. K. P. Scaife, *Principles of Dielectrics*, Revised ed. (Clarendon, Oxford, 1998).