Dielectric response and electro-optical effects in suspensions of anisotropic particles

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The dielectric properties of suspensions of microellipsoids may be controlled by reorientation of the particles in an applied electric field. We study this process and the resulting electro-optical effects for fields of arbitrary strength. The magnitude of these effects is determined by the concentration of the particles, their intrinsic dielectric anisotropy and their shape. The approach presented here provides an explicit link between the particle properties, their orientation distribution and the field-induced birefringence and electro-optical phase shift of the suspensions, taking into account the electrostatic interaction among the particles at moderate concentrations. It reproduces well published experimental observations and should therefore be useful for studying electro-optical properties as well as reorientation phenomena in these systems.

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I. INTRODUCTION

Liquid suspensions of shaped microparticles possess relatively large nonlinear susceptibilities arising from the easy reorientation of the particles by an externally applied field. They are therefore interesting candidates for optical applications such as electro-optic phase modulation of the Kerr type. Very dilute suspensions, with particle volume fraction of 1% or less, have been studied as anisotropic artificial Kerr materials (AKM) for various nonlinear optical applications, e.g. optical birefringence and phase conjugation [1-8]. In these applications, which have been studied extensively both experimentally and theoretically, the polarization characteristics of a probe light beam are altered by varying the intensity or the direction of a strong pump beam. Alternatively, electro-optical effects may be induced in these systems by applying a low-frequency electric field, which should lead to stronger effects since the polarizability of dielectric particles is much greater at low frequencies than at optical frequencies. Experimental studies of these effects have shown that AKM may be used as intensity and phase modulators [8-11]. Polarization modulation has also been studied theoretically by solving the wave equation for the two polarization components of a probe beam in an anisotropic medium in the slowly varying envelope approximation [10]. In most of these studies the suspensions are very dilute and may be considered as random collections of independent particles, since the interaction among the particles is negligible. However, ordering phenomena in these suspensions, and the resulting electro-optical effects, are expected to be significantly enhanced with increasing particle concentration.

One attempt to take into account concentration effects in AKM is reported in the literature [8]. A mean field approximation is used to write an effective excluded volume potential for the interaction between the particles. Combined with the electrostatic energy it leads to a spontaneous phase separation, similar to that observed in lyotropic liquid crystals, at high concentrations even at small electric fields. This is not surprising since the original derivation by Onsager of the isotropic-nematic phase separation in liquid crystals considered the liquid crystal as a simple suspension of rigid rods [12] very similar to an artificial Kerr material, which is de-

scribed here as a suspension of ellipsoids. The phases are characterized by the orientational order parameter of the suspension, which is very small in one phase and of order unity in the other. The order parameter is predicted to exhibit hysteresis as a function of applied field at concentrations near the phase separation. However, this type of behavior has not been observed in experiments. This treatment also ignores the electrostatic interaction between the particles, which should be significant even at concentrations much lower than those required for spontaneous phase separation.

In this paper an alternative approach is presented based on calculating the field-dependent dielectric properties of the AKM, from which the electro-optical phase shift and fieldinduced birefringence may be easily derived. This approach takes into account concentration effects and the average electrostatic interaction between the particles. It leads to an expression for the effective dielectric tensor of a suspension, which depends explicitly on the orientation distribution of the microparticles. The field-dependent response of the sample is calculated by relating this distribution to the magnitude of an externally applied low-frequency field. In the dilute limit we obtain analytic expressions for the effective dielectric tensor of the suspension and for the electro-optical effects determined from it. The electrostatic interactions are taken into account using an extension of the well-known Maxwell Garnett approximation to suspensions of anisotropic particles. The electro-optical effects turn out to be quite sensitive to changes in particle shape, concentration, and orientation distribution and may therefore be useful for studying reorientation phenomena, overcoming the problems due to light scattering, in these materials.

The rest of the paper is organized as follows. The Maxwell Garnett approximation for suspensions of anisotropic inclusions is introduced in Sec. II. In Sec. III, we discuss the reorientation of the particles from their initial random orientation distribution to a steady state under thermal fluctuations in the presence of applied field. The electro-optical effects are presented in Sec. IV. Finally, some brief conclusions are included in Sec. V.

II. DIELECTRIC RESPONSE OF SUSPENSIONS OF ANISOTROPIC PARTICLES

An exact calculation of the effective properties of an inhomogeneous medium is in general an intractable problem. The literature on this subject therefore includes a wide varietv of approximate schemes, each of which is appropriate for different types of composite microgeometries (see, for example, the review papers [13,14], and references therein). One of those, which is particularly useful for microgeometries in which a host material and isolated inclusions of other materials are clearly identified, is the Maxwell Garnett (MG) approximation. It involves an exact calculation of the field induced in the uniform host by a single spherical or ellipsoidal inclusion and an approximate treatment of its distortion by the electrostatic interaction between the different inclusions. This distortion is caused by the charge dipoles and higher multipoles induced in the other inclusions. The induced dipole moments cause the longest range distortions and their average effect is included in the MG approximation, which results in a uniform field inside all the inclusions. This approach has been extensively used for studying the properties of two-component mixtures in which both the host and the inclusions are isotropic materials with scalar dielectric coefficients. In this paper, we use a variation of the MG approach that is adapted for mixtures where the host is an isotropic material with a scalar dielectric constant ϵ_h and the inclusions are microellipsoids made of an anisotropic component with dielectric tensor

$$\boldsymbol{\epsilon}_{s} = \begin{pmatrix} \boldsymbol{\epsilon}_{\perp} & 0 & 0\\ 0 & \boldsymbol{\epsilon}_{\perp} & 0\\ 0 & 0 & \boldsymbol{\epsilon}_{\parallel} \end{pmatrix}.$$
(1)

We assume that in each inclusion the principal axes of ϵ_s are parallel to the geometric axes of the ellipsoid. The orientation of the dielectric tensor differs from inclusion to inclusion, such that in a common coordinate system ϵ_s is transformed to

$$\widetilde{\boldsymbol{\epsilon}}_{s} = \boldsymbol{R} \boldsymbol{\epsilon}_{s} \boldsymbol{R}^{T}, \qquad (2)$$

where R is the inclusion dependent rotation. The distribution of orientations significantly influences the bulk effective properties of the material.

An AKM thin film is typically sandwiched between two conducting electrodes and is electrically driven to obtain the desired effect. A voltage difference V_0 is applied between the electrodes to modify the orientation distribution of the inclusions. E_0 , the volume averaged field over the entire system, host and inclusions, is determined by this voltage difference and the width of the film d, $E_0 = V_0/d$. Solving the electrostatic problem for an inclusion in this uniform field we find that the uniform field inside the inclusion is $\bar{E}_s = \tilde{\kappa} \bar{E}_L$ and the induced dipole moment is $p_s = (v_s/4\pi)\tilde{\alpha} \bar{E}_L$, where v_s is the volume of the inclusion, $\tilde{\kappa} = R \kappa R^T$, $\tilde{\alpha} = R \alpha R^T$, and \bar{E}_L is the local field in the vicinity of the inclusion. The tensor κ is uniaxial with principal elements

$$\kappa_{\parallel} = \frac{\epsilon_h}{d_{\parallel}\epsilon_{\parallel} + (1 - d_{\parallel})\epsilon_h} \tag{3}$$

$$\kappa_{\perp} = \frac{\epsilon_h}{d_{\perp}\epsilon_{\perp} + (1 - d_{\perp})\epsilon_h},\tag{4}$$

where d_{\parallel} and d_{\perp} are the depolarization coefficients of the ellipsoidal inclusion along its principal axes [15]. The tensor α is the polarizability per unit volume of the inclusion. Its principal elements are

 $\alpha_{\parallel} = \kappa_{\parallel} (\epsilon_{\parallel} - \epsilon_{h})$

and

$$\alpha_{\perp} = \kappa_{\perp} (\epsilon_{\perp} - \epsilon_h). \tag{5}$$

A simple method to calculate \overline{E}_L , the average field acting on each inclusion, usually referred to as the excluded volume approach [13], was proposed by Bragg and Pippard [16]. In a mixture that is not too dense, \overline{E}_L is the average field in the host medium. The averaged field over the entire system, inside and outside the inclusions, must still be \overline{E}_0 . The difference between \overline{E}_L and \overline{E}_0 is due to the correlations between positions of different spheres that arise from the prohibition of overlap between them [16]. This leads to a simple relation between the average fields in the host and in the inclusions

$$f\langle \bar{E}_s \rangle + (1-f)\bar{E}_L = \bar{E}_0, \qquad (6)$$

where the angular brackets denote a volume average over the inclusions and f is their volume fraction. Substituting \overline{E}_s , we solve for \overline{E}_L and find

$$\bar{E}_L = \frac{\bar{E}_0}{(1-f) + f\langle \tilde{\kappa} \rangle}.$$
(7)

The denominators here and in the following equations should be interpreted as inverse matrices. The induced dipole moment of a single inclusion is

$$p_{s} = \frac{v_{s}}{4\pi} \frac{\tilde{\alpha} \bar{E}_{0}}{(1-f) + f\langle \tilde{\kappa} \rangle}, \tag{8}$$

which depends not only on the dielectric tensor of the inclusion but on an average of the polarization field of all the other inclusions in the system. The bulk effective dielectric tensor can be defined by the ratio between the volume averaged displacement field $D_0 = \langle D \rangle = \epsilon_h \bar{E}_0 + 4 \pi \langle P \rangle$ and the volume averaged electric field $\bar{E}_0 = \langle E \rangle$. This leads to the bulk effective dielectric tensor

$$\boldsymbol{\epsilon}_{eff} = \boldsymbol{\epsilon}_h \boldsymbol{I} + \frac{f\langle \tilde{\boldsymbol{\alpha}} \rangle}{1 - f + f\langle \tilde{\boldsymbol{\kappa}} \rangle}.$$
(9)

This is the Maxwell Garnett result for mixtures of anisotropic inclusions. It depends on the shape anisotropy of the inclusions, their dielectric tensor ϵ_s , and the orientation distribution of the rotation matrices *R*. It is valid in the static case and in the quasistatic regime, where the wavelength of the applied field is much larger than the particles. Experi-

and

ments have been reported for suspensions of particles with semimajor axis of 40 μ m or less and applied field wavelength at the microwave range or larger [4,8–11], which are well within the quasistatic regime.

For a mixture of many such inclusions, embedded in an isotropic host, it is convenient to define the coordinate system such that the external field \overline{E}_0 is applied in the positive *z* direction. In the absence of applied field the inclusion axes are uniformly distributed in all possible orientations $\theta \in [0, \pi/2]$ and $\varphi \in [0, 2\pi]$, where θ and φ are the polar and azimuthal orientation angles, respectively. The application of an external field in the *z* direction does not change the distribution of the azimuthal angle φ . The angle θ , on the other hand, decreases as the field is increased. The average tensors $\langle \tilde{\kappa} \rangle$ and $\langle \tilde{\alpha} \rangle$ are therefore uniaxial with elements

$$\langle \tilde{\kappa} \rangle_{xx} = \langle \tilde{\kappa} \rangle_{yy} = \frac{1}{2} [\kappa_{\parallel} + \kappa_{\perp} - (\kappa_{\parallel} - \kappa_{\perp}) \langle \cos^2 \theta \rangle]$$

and

$$\langle \tilde{\kappa} \rangle_{zz} = \kappa_{\perp} + (\kappa_{\parallel} - \kappa_{\perp}) \langle \cos^2 \theta \rangle \tag{10}$$

and

$$\langle \tilde{\alpha} \rangle_{xx} = \langle \tilde{\alpha} \rangle_{yy} = \frac{1}{2} [\alpha_{\parallel} + \alpha_{\perp} - (\alpha_{\parallel} - \alpha_{\perp}) \langle \cos^2 \theta \rangle]$$

and

$$\langle \tilde{\alpha} \rangle_{zz} = \alpha_{\perp} + (\alpha_{\parallel} - \alpha_{\perp}) \langle \cos^2 \theta \rangle, \qquad (11)$$

respectively. The effective dielectric tensor also remains uniaxial throughout the switching process.

It is clear that if $E_0 = 0$ the orientation distribution is uniform over $\theta \in [0, \pi]$ and $\varphi \in [0, 2\pi]$ and the material should be isotropic with a scalar dielectric coefficient. Indeed, in this case $\langle \cos^2 \theta \rangle = 1/3$, $\langle \tilde{\kappa} \rangle$ and $\langle \tilde{\alpha} \rangle$ are isotropic and

$$\boldsymbol{\epsilon}_{eff} = \boldsymbol{\epsilon}_h + f \frac{2 \,\boldsymbol{\alpha}_\perp + \boldsymbol{\alpha}_\parallel}{3(1-f) + f(2 \,\boldsymbol{\kappa}_\perp + \boldsymbol{\kappa}_\parallel)}. \tag{12}$$

For spherical particles with a scalar dielectric coefficient ϵ_s this is reduced to

$$\boldsymbol{\epsilon}_{eff} = \boldsymbol{\epsilon}_h + \frac{3f\boldsymbol{\epsilon}_h(\boldsymbol{\epsilon}_s - \boldsymbol{\epsilon}_h)}{(1 - f)(\boldsymbol{\epsilon}_s - \boldsymbol{\epsilon}_h) + 3\boldsymbol{\epsilon}_h},\tag{13}$$

which is the well-known Maxwell Garnett result for mixtures of isotropic components [13].

III. THE ORIENTATIONAL ORDER PARAMETER AND DIELECTRIC RESPONSE

In the preceding section a simple method was presented for calculating the dielectric behavior of AKM films, based on the Maxwell Garnett approximation. The results obtained depend explicitly on the orientation distribution of the microellipsoids. Averages of this distribution have to be evaluated for determining the orientational order parameter of the AKM

$$S = \frac{1}{2} (3\langle \cos^2 \theta \rangle - 1) \tag{14}$$

and the different elements of the bulk effective dielectric tensor. The order parameter S varies from 0 at $E_0=0$, where the orientation distribution is random, to 1 at large fields where all the ellipsoids are aligned with their principal axis parallel to the field. At intermediate situations S is determined by a thermal average of the electrostatic energy that seeks to orient the particles in the direction of the applied field. The electrostatic energy of a single particle is

$$\mathcal{E} = -\frac{1}{2} p_s \bar{E}_L = -\frac{v_s \tilde{\alpha}_{zz} E_0^2}{8 \pi (1 - f + f\langle \tilde{\kappa} \rangle_{zz})^2}, \qquad (15)$$

where p_s is the dipole moment of the inclusion. Similarly to p_s this energy depends not only on the dielectric tensor of the inclusion itself but on an average of the polarization field of all the other inclusions in the system.

Let us define the dimensionless electrostatic energy $U = [v_s E_0^2(\alpha_{\parallel} - \alpha_{\perp})]/8\pi kT$. We then obtain

$$\langle \cos^2 \theta \rangle = \frac{1}{Q} \int e^{-\mathcal{E}/kT} \cos^2 \theta d\Omega = -\frac{1}{2\tilde{U}} + \frac{e^U}{\sqrt{\pi \tilde{U}} \operatorname{Erfi}(\sqrt{\tilde{U}})},$$
(16)

where

$$Q = \int e^{-\mathcal{E}/kT} d\Omega = 2 \pi e^{\widetilde{U} \frac{\alpha_{\perp}}{\delta \alpha}} \sqrt{(\pi/\widetilde{U})} \operatorname{Erfi}(\sqrt{\widetilde{U}}), \quad (17)$$

$$\tilde{U} = \frac{U}{\left(1 - f + f\langle \tilde{\kappa} \rangle_{zz}\right)^2},\tag{18}$$

 Ω is a solid angle, k is the Boltzmann constant, and T is the temperature. Erfi(x) = $(2/\sqrt{\pi})\int_0^x e^{t^2} dt = -i \cdot \operatorname{erf}(ix)$ is the modified error function. In the dilute limit $f \rightarrow 0$, \tilde{U} is reduced to U and the calculation of the order parameter is straightforward. However, taking into account the electrostatic interaction between particles we have to consider finite f and the explicit dependence of \tilde{U} on the orientation distribution of all particles in the system. The solution in this case is obtained by a simultaneous numerical solution of Eqs. (16) and (18). The numerical calculation is easy in principle. The only difficulty arises in the computation of the modified error function Erfi(x), for which we have used an algorithm for rapid computation of complex error functions developed by Hui *et al.* [17].

The result (16) applies for prolate and oblate microparticles. It is assumed here, for simplification, that the suspension is monodispersed, i.e., v_s is the same for all particles, however, similar results are obtained for any distribution of v_s . As expected, when $E_0=0$, θ is distributed uniformly and $\langle \cos^2 \theta \rangle = 1/3$. It varies sharply at small fields and reaches saturation ($\langle \cos^2 \theta \rangle = 0,1$, for $\delta \alpha = \alpha_{\parallel} - \alpha_{\perp} < 0,>0$, respectively) at large fields where all the ellipsoids are aligned parallel to the field. For small *U* we find $\langle \cos^2 \theta \rangle = 1/3 + 4\tilde{U}/45 + O(\tilde{U}^2)$.

The electric fields applied to induce reorientation are low frequency (typically 100 kHz or less) [10,11]. The dielectric coefficients ϵ_{\parallel} , ϵ_{\perp} , and ϵ_{h} used in Eq. (16) should therefore be the dielectric coefficients of the inclusions and liquid host at these low frequencies. The field strength required for saturation may be easily estimated from the definition of U. For a typical sample with $\delta \alpha \sim 1$ and $v_s \approx 10^{-15}$ cm³ at T= 300 K, $U \sim 10$ leads to $E_0 \approx 1 \text{ V/}\mu\text{m}$, i.e., voltages of the order of 20 V are needed to bring a 20- μ m-thick film to saturation, in excellent agreement with the experimental results of Ref. [11].

Typical results demonstrating the dependence of the order parameter *S* on the volume fraction of the particles and their aspect ratio are shown in Fig. 1. *S* is plotted as a function of the dimensionless energy *U* and the corresponding voltage V_0 applied on a 20- μ m-thick sample. In Fig. 1(a) results are shown for suspensions of particles with axes aspect ratio of 1.7:1, corresponding to a principal depolarization factor d_{\parallel} =0.2, and four different concentrations f=0.01, 0.1, 0.3, and 0.4. The rise of *S* with increasing field is more rapid at higher concentrations. It is evident that although the applied field is strong enough to saturate the rotational degree of freedom of the particles the concentrational effects are relatively modest. This is in very good agreement with the experimental results of Kralik *et al.* [8].

In Fig. 1(b) results are shown for suspensions with a fixed concentration f=0.3 and particles with different axes aspect ratios: prolate spheroids with aspect ratios of 3.2:1 and 1.7:1, perfect spheres, and oblate spheroids with aspect ratio of 1.8:1, corresponding to the principal depolarization factors $d_{\parallel}=0.1, 0.2, 1/3, \text{ and } 0.5$, respectively. The effect of these shape variations on *S* as a function of *U* is again quite modest. However, since shape variations also affect *U*, the effect on *S* as a function of applied voltage V_0 is significant. This effect has not been examined experimentally.

Once *S* is calculated, it is easy to evaluate the principal elements of the effective dielectric tensor (9). Typical results for ϵ_{eff} are shown in Figs. 2 and 3. The elements of the effective dielectric tensor are shown as a function of applied voltage for AKM films of microellipsoids with different aspect ratios and different volume fractions. It is clearly seen that smaller depolarization coefficients, larger volume fractions, and larger intrinsic anisotropies of the particles, lead to stronger dependence of the dielectric properties on the applied field. In these examples, the effect also appears for spherical inclusions, $d_{\parallel} = 1/3$, because of the intrinsic anisotropy of the inclusions $\epsilon_{\parallel} \neq \epsilon_{\perp}$.

IV. FIELD-INDUCED BIREFRINGENCE AND OPTICAL PHASE SHIFT

Once the order parameter *S* is known, Eq. (9) can be used to calculate the ordinary and extraordinary refractive indices of the AKM. In this calculation, the dielectric coefficients $\epsilon_{\parallel} = n_{\parallel}^2$, $\epsilon_{\perp} = n_{\perp}^2$, and $\epsilon_h = n_h^2$ substituted in Eqs. (10) and



FIG. 1. The order parameter *S* as a function of the dimensionless energy *U* and the applied voltage V_0 (inset) for suspensions of ellipsoids with $\epsilon_{\parallel}=3$, $\epsilon_{\perp}=2$, $\epsilon_{h}=1$, $d=20 \ \mu$ m. In (a) the principal depolarization coefficient is $d_{\parallel}=0.2$ and the volume fraction varies: f=0.4 (solid lines), f=0.3 (dotted lines), f=0.1(dashed lines), and f=0.01 (dash-dot lines). In (b) f=0.3 and $d_{\parallel}=0.1$ (solid lines), $d_{\parallel}=0.2$ (dotted lines), $d_{\parallel}=1/3$ (dashed lines), and $d_{\parallel}=0.5$ (dash-dot lines).

(11), are those of the inclusions and host at the frequency of light incident on the sample. n_{\parallel} , n_{\perp} , and n_h are the corresponding refractive indices. The effective dielectric tensor is uniaxial and its principal axis is perpendicular to the film plane. The ordinary refractive index of the AKM is therefore

$$n_o = \sqrt{\epsilon_h + \frac{f\langle \tilde{\alpha} \rangle_{xx}}{1 - f + f\langle \tilde{\kappa} \rangle_{xx}}}$$
(19)

and the extraordinary index is

$$n_e = \sqrt{\epsilon_h + \frac{f\langle \tilde{\alpha} \rangle_{zz}}{1 - f + f\langle \tilde{\kappa} \rangle_{zz}}}.$$
 (20)



FIG. 2. The diagonal elements of the effective dielectric tensor (9) as a function of applied voltage for the same samples as in Fig. 1(b). The upper (lower) curve in each pair is $\epsilon_{eff,zz}$ ($\epsilon_{eff,xx}$).

The optical phase shift experienced by light of wavelength λ incident obliquely at angle ϑ on the plane surface of the AKM film is

$$\phi = \frac{2\pi d}{\lambda \cos\vartheta} \,\delta n,\tag{21}$$

where

$$\delta n = \frac{n_o n_e}{\sqrt{n_e^2 \cos^2 \vartheta + n_o^2 \sin^2 \vartheta}} - n_o \,. \tag{22}$$



FIG. 3. The diagonal elements of the effective dielectric tensor (9) as a function of applied voltage for suspensions of ellipsoids with the same principal depolarization coefficients $d_{\parallel}=0.2$ and $\epsilon_{\perp}=2$, $\epsilon_{h}=1$ but different principal dielectric coefficients and volume fractions: $\epsilon_{\parallel}=3$, f=0.1 (dash-dot lines), and f=0.3 (dashed lines). $\epsilon_{\parallel}=5$, f=0.1 (dotted lines), and f=0.3 (solid lines). The upper (lower) curve in each pair is $\epsilon_{eff,zz}$ ($\epsilon_{eff,xx}$).



FIG. 4. The electro-optical phase shift ϕ (radians) as a function of applied voltage V_0 (in volts) for the same samples as in Fig. 3, with $n_{\parallel} = 1.4$, $n_{\perp} = 1.35$, $n_h = 1$, and incidence angle $\vartheta = 10^{\circ}$.

The electric field induced birefringence is

$$\Delta n = n_e - n_o = \frac{f \,\delta\alpha}{4n_h} (3\langle \cos^2\theta \rangle - 1) = \frac{N_0 (\delta\beta)^2 E_0^2}{120n_h \pi kT} \quad (23)$$

to first order in U and f, where $N_0 = f/v_s$ is the number of particles per unit volume and $\delta\beta = v_s \delta\alpha$ is their polarizability. For a suspension where $f = 10^{-3}$, $v_s = 10^{-9}$ cm³, and, if the orienting field is at microwave frequencies leading to $\delta\alpha \sim 0.5$ (typical for particles with an aspect ratio of $\sim 5:1$), we find $n_2 = 8 \pi \Delta n/(cE_0^2) \approx 2 \times 10^{-4}$ cm²/W, where *c* is the speed of light. This is in very good agreement with the experimental result of Ref. [4] for the Kerr coefficient of such an AKM.

Typical results for the electro-optical phase shift ϕ as a function of applied voltage for AKM films of microellipsoids with different aspect ratios and volume fractions are shown in Figs. 4 and 5. Again, larger anisotropy, either intrinsic or shape anisotropy, of the particles and higher concentrations lead to a stronger effect. It is interesting to note that for oblate particles with $d_{\parallel} = 1/2$, ϕ is negative (Fig. 5). This happens because in this example $\delta \alpha$ is positive at the low frequency of the applied field but is negative at the optical frequency of the incident light. The same behavior is obtained for the electric field induced birefringence Δn . These results agree with the observation that the electro-optic activity of AKM is strongly influenced by the intrinsic anisotropy of the particles, in addition to shape anisotropy, and the phase shift obtained at low concentrations is smaller than 0.1 rad [9].

V. CONCLUSIONS

In this paper, the reorientation of microellipsoids in liquid suspensions and the consequent changes in their electro-optic properties are studied using a simple model for the bulk effective dielectric response. The model is an extension of the well-known Maxwell Garnett approximation for the dielec-



FIG. 5. The electro-optical phase shift ϕ (radians) as a function of applied voltage V_0 (in volts) for 20- μ m-thick samples with f=0.2, ϵ_{\parallel} =3, ϵ_{\perp} =2, ϵ_{h} =1, n_{\parallel} =1.4, n_{\perp} =1.35, n_{h} =1, ϑ =10°, and different principal depolarization coefficients: d_{\parallel} =0.2 (solid line), d_{\parallel} =0.25 (dash-dotted line), d_{\parallel} =1/3 (dotted line), and d_{\parallel} =0.5 (dashed line).

tric properties of mixtures of isotropic particles and, similarly, it is exact to second order in the volume fraction of the inclusions. As in previous studies of this problem, the essential physics involves a balance between the electrostatic energy, which favors alignment with the applied field, and thermal fluctuations. However, the approach presented here gives

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explicit results for the field-dependent orientation distribution of the ellipsoids and provides a convenient framework for the evaluation of the electrostatic effects. It also allows explicit consideration of the electrostatic interaction between the particles at moderate concentrations. Analytical results are obtained for dilute systems and a very simple numerical scheme is given for systems with higher concentrations of microellipsoids. The results of these calculations reproduce published experimental observations. In particular, the calculated order parameter at moderate concentrations (f=0.1-0.4) agrees well with the experiments of Kralick *et al.* [8] which are, to the best of our knowledge, the only results published for this regime.

It is demonstrated that the optical phase shift and birefringence in AKM films should be very sensitive to changes in the orientation distribution, which in turn is sensitive to properties of the microparticles. Measurement of these effects may therefore offer a good approach for studying the reorientation process and the physical parameters (e.g., particle shape, intrinsic anisotropy, and concentration) that influence it. In addition to these fundamental questions, electro-optical effects in AKM are also interesting from the applied point of view. Like any birefringent material of the Kerr type, they may be used in electro-optical shutters and other devices. Their potential advantage over other materials is the variety of electro-optical behaviors under various conditions, demonstrated above. Using AKM we may select the operation voltage and the magnitude of the effects by varying particle concentration and shape as well as the intrinsic dielectric coefficients of the components.

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