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FLUCTUATION-INDUCED KERR EFFECT IN CHIRAL SMECTIC-A LIQUID CRYSTAL PHASE

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ABSTRACT

FLUCTUATION-INDUCED KERR EFFECT IN CHIRAL SMECTIC-A LIQUID CRYSTAL PHASE. The second-order electrooptic responses under large orientational fluctuations in a chiral smectic-A liquid crystal phase has been studied theoretically. The formula for analyzing the frequency dispersion of Kerr effect considering the fluctuations has been derived. The possibility of the nonlinear electrooptic response observing the soft mode condensation inducing the smectic-C_α* phase in the smectic-A phase by using the fluctuation-induced Kerr effect has been explored.

Key words : Liquid crystal, smectic, electrooptic, soft mode, fluctuations

ABSTRAK

EFEK KERR YANG DIINDUKSI FLUKTUASI PADA FASA KRISTAL CAIR SMEKTIK-A. Respon elektrooptik orde kedua dalam kondisi fluktuasi orientasi yang besar pada fasa kristal cair smektrik-A telah dipelajari secara teoritis. Telah diturunkan persamaan untuk menganalisis dispersi frekuensi dar efek Kerr dengan mempertimbangkan fluktuasi. Telah dipelajari juga kemungkinan respon elektrooptik nonlinier dapat mengamati *soft mode* yang menginduksi terbentuknya fasa smektik-C_α* di dalam fasa smektik-A.

Kata kunci : Kristal cair, smektik, elektrooptik, *soft mode*, fluktuasi

INTRODUCTION

Chiral smectic liquid crystals have various phases [1]. In the highest symmetry of SmA phase the molecules are perpendicular to smectic layers, while in lower temperature phases the molecules tilted. Although the tilting directions are different in layers to construct different phases. For example, in the ferroelectric smectic-C* (SmC*) phase all the molecules tilt in the same direction and in the antiferroelectric SmC_A* phase the molecules in the neighboring layers tilt in the opposite directions, though these structures are slightly twisted along the layer normal by the chirality. Application of both ferroelectric and antiferroelectric liquid crystal in the display technology needs detailed physical characterization of these materials. The dynamic properties represent one of the most important aspects of the characterization of the ferroelectric and antiferroelectric order and, as a result, they will give the information about the appropriate properties used in the electro-optical systems. Among tilted phase, SmC_α* phase has been attracting much attention, which has an incommensurate, short-period ferroelectric like structure [2]. This phase appears in most of antiferroelectric liquid crystal phase sequence. In the present work the dynamics near the SmA-SmC_α* phase transition theoretically studied.

The phase transitions from the SmA phase to these tilted smectic phases are considered to be brought about by the condensation of overdamped collective orientational soft modes. In general, the observation of the soft mode is quite important to elucidate the mechanism of phase transitions. Actually, for the SmA-SmC* phase transition the soft mode has been observed clearly in both SmA and SmC* phases by means of dielectric spectroscopy [3] and photon correlation spectroscopy [4]. But for the SmA-SmC_α* phase transition, we could not observe it in the SmA by these spectroscopies, because the soft mode is a helically tilted mode with a short pitch, i.e., it exists at a general point of the smectic Brillouin zone in the SmA phase. In the SmC_α* phase the soft mode becomes a zone-center mode by the symmetry breaking. When an electric field is applied parallel to smectic layers two modes can be excited; the ferroelectric mode (the tilting toward the direction perpendicular to the applied field) due to the electroclinic effect and the amplitude mode (the change of tilt angle), which brings about the rotation of indicatrix and the change of birefringence, respectively. Therefore, the amplitude mode can be observed. Actually it has been observed by means of the second-order electrooptic [5], third-order nonlinear dielectric spectroscopy [6]

and the measurement of linear dielectric constant under dc-bias [7].

In the SmA phase, on the other hand, unusually large pretransitional fluctuations have been observed near the transition point by heat capacity [8], birefringence [9] and layer compression modulus measurements [10]. These large fluctuations enable to observe the soft mode in the SmA phase, as explained below. Quite recently, it has been shown that the pretransitional fluctuations are also observable by means of electrooptic measurements; an anomalous increase in the second-order electrooptic response (Kerr effect) was experimentally found near the transition point from the SmA phase to the SmC_α* phase of 4-(1-methyl-heptyloxycarbonyl) phenyl 4-octylbiphenyl-4-carboxylate(MHPOBC). A simple theory under a dc field has been presented to explain this field-induced Kerr effect [11]. Therefore, the electrooptic measurement has been experimentally and theoretically shown to be useful in studying the pretransitional fluctuations as well as the heat capacity and birefringence measurements. However, it is emphasized that the electrooptic measurements under ac fields have the merit of giving the information on the dynamics of the fluctuations. It has been reported that the electrooptic response shows a characteristic frequency dispersion in the second harmonic [11], though the analysis of it was not adequately made because we had no theoretically derived formula for it.

In this paper, first the formula describing the field-induced Kerr effect under ac fields from the Langevin equation will be derived. Next, it will be shown theoretically that the soft mode at any point of the Brillouin zone can be detected in the SmA phase by using the fluctuation-induced Kerr effect if the low temperature phase is tilted. Finally some conditions for electrooptical measurement to observe the soft mode in SmA phase will be explored.

MODES EXCITED BY AN ELECTRIC FIELD IN THE SMECTIC-A PHASE

The spatially dependent order parameters $(\xi_x(x,y,jd), \xi_y(x,y,jd))$ [12], where j is the layer number and d the layer spacing, by the helicoidal coordinate [1] and the Fourier transformation :

$$\begin{pmatrix}\xi_x(x,y,jd)\\ \xi_y(x,y,jd)\end{pmatrix}=\begin{pmatrix}\xi_{fx}\\ \xi_{fy}\end{pmatrix}+\begin{pmatrix}\cos q_c jd & -\sin q_c jd\\ \sin q_c jd & \cos q_c jd\end{pmatrix}$$

$$\sum_{\mathbf{q}}\begin{pmatrix}\xi_{1\mathbf{q}}\\ \xi_{2\mathbf{q}}\end{pmatrix}\exp\left(i\left(q_x x+q_y y+q_z jd\right)\right) \quad \text{.... (1)}$$

where (ξ_{fx}, ξ_{fy}) and $(\xi_{1\mathbf{q}}, \xi_{2\mathbf{q}})$ are, respectively, the ferroelectric mode and the helically tilted mode in the soft mode branch, and q_c is the wave number related to the SmA-SmC_α* phase transition. It should be noted that the mode with q = 0, (ξ_{10}, ξ_{20}), represents the soft mode (the helically tilted mode with q_c in the laboratory frame) inducing the SmA-SmC_α* phase transition, but the modes with $q \neq 0$ are also necessary for our purpose. In the fluctuation-induced Kerr effect, the nonlinear coupling between the ferroelectric mode (ξ_{fx}, ξ_{fy}) and the modes $(\xi_{1\mathbf{q}}, \xi_{2\mathbf{q}})$ with wave vectors around q = 0 plays an essential role as well as the dielectric anisotropy. Taking these modes into account, we can write the free energy F divided by the sample volume V under an electric field along the x-axis, E_x, in the SmA phase as

$$F/V=\sum_{\mathbf{q}}\left\{\frac{1}{2}a_{\mathbf{q}}\left(\left|\xi_{1\mathbf{q}}\right|^2+\left|\xi_{2\mathbf{q}}\right|^2\right)+ b\left(\left|\xi_{1\mathbf{q}}\right|^2+\left|\xi_{2\mathbf{q}}\right|^2\right)\xi_{fx}^2-\frac{\varepsilon_a'}{4}\left(\left|\xi_{1\mathbf{q}}\right|^2+\left|\xi_{2\mathbf{q}}\right|^2\right)E_x^2\right\}+ c\xi_{fx}E_x+\frac{a_f}{2}\xi_{fx}^2 \quad \text{..... (2)}$$

where ε_a' is the dielectric anisotropy at low frequencies, and we assume that the dispersion is parabolic in the soft-mode branch, $a_{\mathbf{q}}=a_0+\kappa_\perp\left(q_x^2+q_y^2\right)+\kappa_{//}q_z^2$, and a_0 is linearly dependent on the temperature and become zero at the SmA-SmC_α* transition point, and the above summation is made over q around q_c since only large fluctuations around q_c contribute significantly to the fluctuation-induced Kerr effect. With the free energy and the dissipation function D,

$$D/V=\frac{\gamma_s}{2}\sum_{\mathbf{q}}\left(\left|\dot{\xi}_{1\mathbf{q}}\right|^2+\left|\dot{\xi}_{2\mathbf{q}}\right|^2\right)+\frac{\gamma_f}{2}\dot{\xi}_{fx}^2 \quad \text{..... (3)}$$

the dynamics can be expressed as

$$\frac{\partial D}{\partial\dot{\xi}_{fx}}=-\frac{\partial F}{\partial\xi_{fx}},\frac{\partial D}{\partial\dot{\xi}_{n\mathbf{q}}^*}=-\frac{\partial F}{\partial\xi_{n\mathbf{q}}}+R_{n\mathbf{q}}(t)\ \ (n=1,2) \quad \text{..... (4)}$$

where $R_{nq}(t)$ is the random force satisfying the following relation:

$$\left\langle R_{n\mathbf{q}}(t)^* R_{n'\mathbf{q}'}(t')\right\rangle=2V\gamma_s k_B T\delta_{nn'}\delta_{\mathbf{q}\mathbf{q}'}\delta(t-t')$$

Solving the above set of equations under an ac electric field, $E_x=E_0\cos\omega t$, by the perturbation method with respect to E_0, yields

$$\xi_{fx}=\mathrm{Re}\left\{\chi_f(\omega)\exp(i\omega t)\right\}E_0,$$

$$\left\langle\left|\xi_{1\mathbf{q}}\right|^2\right\rangle=\left\langle\left|\xi_{2\mathbf{q}}\right|^2\right\rangle=\frac{k_B T}{a_{\mathbf{q}}}-$$

$$\frac{k_B T\gamma}{a_{\mathbf{q}}^2}\left[c_0(\omega)+\mathrm{Re}\left\{\frac{c_2(\omega)}{1+i\omega\gamma_s/a_{\mathbf{q}}}\right\}\exp(i\omega t)\right]E_0^2 \quad \text{..... (5)}$$

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with

$$\chi_f(\omega)=\left(a_f + \sum_{\mathbf{q}} \left\langle \left|\xi_{1\mathbf{q}}\right|^2 + \left|\xi_{2\mathbf{q}}\right|^2 \right\rangle_{E_x=0} + i\omega\gamma_f \right)^{-1} \quad \text{..... (6)}$$

$$c_0(\omega)=bc^2\left|\chi_f(\omega)\right|^2 - \varepsilon_a'/4 \quad \text{..... (7)}$$

$$c_2(\omega)=bc^2\chi_f(\omega)^2 - \varepsilon_a'/4 \quad \text{..... (8)}$$

where $\chi_f(\omega)$ is the susceptibilities of the ferroelectric mode, which are of the Debye-type relaxation. Note that in eq.(8), $c_0(\omega)$ and $c_2(\omega)$ contain $c_f(\omega)$ and ε_a'; the former means that the modes around q_c couple with the field through the ferroelectric mode and the latter through the dielectric anisotropy. $\langle \xi_y^2 \rangle$ is given as $\sum_{\mathbf{q}} \langle |\xi_{1\mathbf{q}}|^2 \rangle$ and $\langle \xi_x^2 \rangle = \langle \xi_y^2 \rangle + \xi_{fx}^2$. For convenience $\langle \xi_y^2 \rangle$ is divided into the field-independent part $\langle \xi_y^2 \rangle_0$ and the field-dependent part $\langle \xi_y^2 \rangle_E$ as $\langle \xi_y^2 \rangle = \langle \xi_y^2 \rangle_0 + \langle \xi_y^2 \rangle_E$. They are

$$\left\langle \xi_y^2 \right\rangle_0 = A\left(1 - B\sqrt{a_0}\right),$$
$$\left\langle \xi_y^2 \right\rangle_E = -\frac{k_B T}{8\pi\kappa_\perp \sqrt{\kappa_{//}}\sqrt{a_0}}\left[c_0(\omega) + \mathrm{Re}\{c_2(\omega) g(i\omega\tau_s)\exp(i2\omega t)\}\right]E_0^2 \quad (9)$$

with $g(x) \equiv 2\left(\sqrt{1+x}-1\right)/x$ where $\tau_s (= \gamma_s / a_0)$ is the relaxation time of the soft mode at q = 0 and the thermal average $\sum_{\mathbf{q}} \langle |\xi_{1\mathbf{q}}|^2 + |\xi_{2\mathbf{q}}|^2 \rangle_{E_x=0}$ in the absence of field is calculated as :

$$2k_B T/V \sum_{\mathbf{q}} a_{\mathbf{q}}^{-1} \approx k_B T A\left(1 - B a_0^{1/2}\right) \quad \text{..... (10)}$$

(*A* and *B* are constants depending on the higher cut-off wave number). Note that $g(i\omega\tau_s)$ is a broad frequency dispersion coming from the modes in the soft mode branch.

ELECTROOPTIC RESPONSE IN THE SMECTIC-A PHASE

As mentioned above, the ferroelectric and the soft modes modulate the refractive indices and so bring about the electrooptical effect. In investigating the change of refractive indices under a field, it is assumed that the molecules are optically uniaxial and the optical properties are fully determined by the directors.

In general, it is difficult to calculate the transmitted intensity through a cell between polarizers. For simplicity, it is considered such a case that the wavelength of the helix is smaller than that of the light. Under this condition it is regarded the cell is spatially homogeneous. In electrooptic measurements of a homogeneous cell between the crossed polarizers, where the *z*-axis is set to be along the layer normal and the *x*-axis along the electric field direction, the transmitted light intensity *I* is written as

$$I / I_0 = \sin^2(2\alpha)\sin^2(\beta n_a) \quad \text{..... (11)}$$

with $\beta = \pi d/\lambda$ where I_0 is the incident light intensity, *d* the cell gap, λ the light wavelength, α and n_a are the angle between one of the principal axes of the indicatrix nearly parallel to the *z*-axis and the direction of one polarizer and the anisotropy of refractive indeces nearly along *z*- and *y*- axes, respectively. The modes induced by applied field modify both α and n_a. We put

$$\alpha = \alpha_0 + \Delta\alpha$$
$$n_a = n_{a0} + \Delta n_a \quad \text{..... (12)}$$

where α_0 and n_{a0} are equilibrium values in the absence of the field, and $\Delta\alpha$ and Δn_a are electrically-induced parts.

Substitution of eq. (12) into eq. (11) yields the change of the intensity ΔI, due to the field application to the cell between crossed polarizers, is expanded up to the second order with respect to the field strength [13]:

$$\begin{aligned} \Delta I / I_0 &= 2\sin 4\alpha_0 \sin^2(\beta n_{a0})\Delta\alpha \\ &+ 4\cos 4\alpha_0 \sin^2(\beta n_{a0})\Delta\alpha^2 \\ &+ \beta \sin^2 2\alpha_0 \sin(2\beta n_{a0})\Delta n_a \end{aligned} \quad \text{..... (13)}$$

Δa, Δn_a and n_{a0} are calculated from the dielectric tensor, the components of which are expressed as [13]

$$\begin{aligned} \varepsilon_{yy} &= \varepsilon_\perp + \varepsilon_a \langle \xi_x^2 \rangle, \\ \varepsilon_{yz} &= \varepsilon_a \langle \xi_x \rangle, \\ \varepsilon_{zz} &= \varepsilon_{//} - \varepsilon_a\left(\langle \xi_x^2 \rangle + \langle \xi_y^2 \rangle\right) \end{aligned} \quad \text{..... (14)}$$

where $\varepsilon_\perp$ and $\varepsilon_{//}$ are the dielectric constants perpendicular and parallel to the director, respectively, and $\varepsilon_a = \varepsilon_{//} - \varepsilon_\perp$, ξ_x and ξ_y are order parameters and $\langle \cdots \rangle$ means the statistical average. The above averages are calculated on the basis of the Langevin equation as mentioned above.

In terms of calculation results, Dn_a in eq. (13) is given as

$$\Delta n_a \approx -\frac{\varepsilon_a'}{2}\left(\frac{2}{\sqrt{\varepsilon_{//}}} + \frac{1}{\sqrt{\varepsilon_\perp}}\right)\langle \xi_y^2 \rangle_E - \cdot$$
$$\frac{3\varepsilon_a'}{2}\left(\frac{1}{\sqrt{\varepsilon_{//}}} - \frac{1}{\sqrt{\varepsilon_\perp}}\right)\langle \xi_y^2 \rangle_0 \xi_{fx}^2 \quad \text{..... (15)}$$

In addition, Δa in eq. (13) is

$$\Delta\alpha = \varepsilon_a' \xi_{fx} \quad \text{..... (16)}$$

By using the above equations we can fully analyze the results obtained in the electrooptic measurements.

Note that α_0 in eq. (13) is determined by the experimental geometry; the angle between the *z*-axis and

the direction of one polarizer. Thus, the physical meaning of each term in eq. (13) is clarified. The first term comes from the ferroelectric mode and is proportional to the field (see eqs. (13), (6), (8), (16)), and the origin is the tilt of the indicatrix induced by the electric field, i.e., the Pockels effect, which is called the electroclinic effect in the SmA phase. The second term is also related to the ferroelectric mode, but it is proportional to the square of the field. The third term is also proportional to the square of the field and the origin is both the soft mode and the ferroelectric mode, which all contribute to the Kerr effect (see eqs. (13), (15), (9)). Since Δa and Δn_a are proportional to the field and the square of the field (see eqs. (6), (9), (13), (15), (16)), respectively, the first term in eq. (13) can be detected in the first-order electrooptic response and the two others in the second-order electrooptic response.

It should be emphasized here that without considering the fluctuation, in the SmA phase the application of a field brings about the spatially uniform tilt of molecules due to the electroclinic effect, resulting only in the rotation of indicatrix, i.e., the Pockels effect [13]. However, in the vicinity of the transition point even in the SmA phase the Kerr effect can be observed since small clusters of the tilted phases are developed, in which the field can change the tilt and the azimuthal angles of the director, resulting in the Kerr effect. It is seen from eq. (9) that in the presence of thermal fluctuations, the order parameter can be excited by the applied field. Thus we have the electrooptic effect induced by fluctuations, as seen in eq. (15).

RESULTS AND DISCUSSION

Most of the electrooptical measurements were conducted only at α_0 =22.5º. For α_0 =22.5º, the first-order response originates in the indicatrix rotation Δα and the second-order response only in the birefringence change Dn_a, as seen in eq. (13). From eq. (13), (6), (8), (16) the frequency dispersion of the first-order electro optic response can be expressed by the Debye-type formula,

$$\Delta I_{1f} = \frac{A}{1+i\omega\tau_f} \quad \text{..... (17)}$$

where $\tau_f = 1/2\pi f_f$, f_f is relaxation frequency of ferroelectric mode, with the typical calculated results is shown Figure 1. From the equation, we know that only the ferroelectric mode is involved in the linear electrooptical response with the peak frequency of the imaginary part indicated the relaxation frequency of ferroelectric mode.

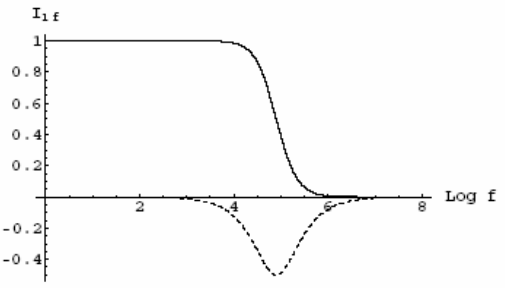

Figure 1. The frequency dispersion of the first-order electrooptical response in SmA phase calculated from eq. (17) with A=1, ff=80kHz. Solid and dashed lines show the real part and the imaginary part, respectively.

On the other hand, the dispersion curve of the second-order response for α_0=22.5º can be expressed by the formula,

$$\left\{A_1 + \frac{A_2}{(1+i\omega\tau_f)^2}\right\} g(i\omega\tau_s) + \frac{A_3}{(1+i\omega\tau_f)^2}, \quad \text{..... (18)}$$

where $\tau_s = 1/2\pi f_s$ is the relaxation time of the soft mode inducing the SmA-SmC$_\alpha$* phase transition, and A_1, A_2 and A_3 are adjustable parameters for the least-square fitting. We also obtained the similar equation for the frequency dispersion in SmC$_\alpha$* phase, though we have to change $g(i\omega\tau_s)$ to susceptibility of amplitude mode $\chi_s(2\omega)$ [5]. Figure 2 shows the frequency dispersion of both modes. $\chi_s(2\omega)$ shows Debye-type relaxation , while $g(i\omega\tau_s)$ has a wide distribution of relaxation time which comes from not only the mode located at q_c but also the modes around q_c in the soft mode branch. The peak frequency becomes higher than Debye-type.

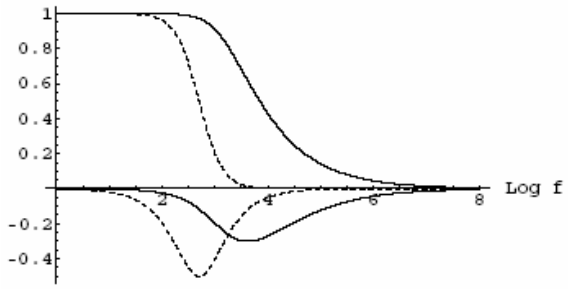

Figure 2. Comparison between Debye-type relaxation $\chi_s(2\omega) = 1/(1+i2\omega\tau_s)$ (dashed line) in the SmC$_\alpha$* phase and the function $g(i\omega\tau_s) = 2\left(\sqrt{1+i\omega\tau_s}-1\right)/i\omega\tau_s$ in the SmA phase, f_s=1kHz.

Figure 3 shows the typical frequency dispersion obtained from the eq. (18). In the imaginary part, there are two modes as predicted in eq. (15); one peak at a high frequency corresponds to the ferroelectric mode, and another one at a low frequency corresponds to the soft mode. It should be emphasized here that when the frequency dispersion measurement is performed at α_0=22.5º, the ferroelectric mode in the second-order response is originated only from the Δn_a term, and not from the $\Delta\alpha^2$. term.

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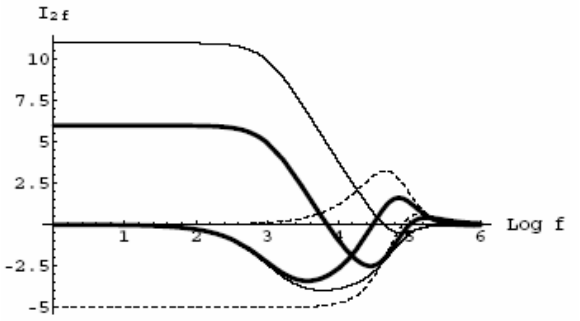

Figure 3. Typical calculated frequency dispersion of the second-order electrooptical response in SmA phase, with A_1=1, A_2=10, A_3=-5, f_s=1kHz, f_f=80kHz (Bold line). The thin line shows the contribution of first term in eq. (18), while dashed line the second term.

From eq. (13), it is understood that the transmitted intensity should depend on α_0, n_{a0}, λ and d, and that the expression for the second-order response is more complicated than that for the linear response because it includes the contributions of both the fluctuation-induced soft mode and the ferroelectric mode. Most of the electrooptical measurements were conducted only at α_0=22.5º. Therefore, the above complexity have not been fully understood.

Now, let us investigate eqs. (13)-(16) in more detail. From eq. (13), $\Delta\alpha$ is proportional to ξ_{fx}, and thus the second term in eq. (13) is proportional to ξ_{fx}^2. On the other hand, Δn_a also includes ξ_{fx}^2 as is seen in eq. (15). When eq. (13) is regarded as a function of ξ_{fx}^2 and $\left\langle \xi_y^2 \right\rangle$, their coefficients may depend on α_0, n_{a0}, λ and d in different manners. Therefore, the intensity ratio of the soft mode to the ferroelectric mode depends on the above parameters. In particular, we may select such a set of parameters to suppress the contribution of the ferroelectric mode to the second-order response, i.e., the sum of the coefficients of ξ_{fx}^2 in the second and third terms in eq. (13) vanishes. In this case, the soft mode can be observed more clearly.

Figure 4 shows the calculated frequency dispersion of second-order electrooptic response for various weight contribution of ferroelectric mode (A_3) in SmA phase. For A_3=90, 20, -90 where ferroelectric mode is dominant, it is difficult to observe the contribution of soft mode appeared as the peak in lower frequency in imaginary part of frequency dispersion. Previous experimental reports showed that the electrooptic measurements at α_0=22.5º gives difficulty to obtain the contribution of soft mode to second-order electrooptic response in SmA phase since the contribution of ferroelectric mode was dominant [5]. When the high-frequency peak related to the ferroelectric mode is suppressed (A_3 small), then the low-frequency peak related to the soft mode can be seen. This result is due to the cancellation of two terms related to ξ_{fx}^2 in eq. (13). In the real experiment, this ferroelectric mode contribution can be suppressed by changing the measurement geometry, such as setting α_0 at certain angles other than 22.5º, changing n_{a0} by introducing a compensator between sample and objective lense or change the cell gap or select appropriate liquid crystal samples.

CONCLUSION

The formula for analyzing the frequency dispersion of second-order electrooptic response in SmA phase has been derived. The possibility of electrooptic measurement to observe soft mode in the SmA phase by using fluctuation-induced Kerr effect has been explored. By suppressing the ferroelectric mode weight contribution to the second-order response the peak in the imaginary part of frequency dispersion corresponds to soft mode should be obtained. In this paper we emphasized that the second-order electrooptic measurement is a powerful method for observing orientational fluctuations with non-zero wave numbers.

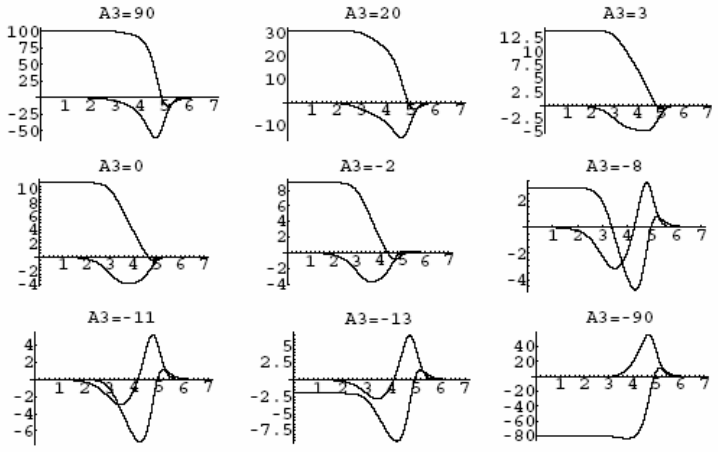

Figure 4. Typical calculated frequency dispersion of the second-order electrooptical response in SmA phase with variation of A_3, with A_1=1, A_2=10, f_s=1kHz, f_f=80kHz

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