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Two-wave mixing on the cubic non-linearity in the smectic A liquid crystals

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We present a theoretical study of two-wave mixing in smectic A liquid crystals (S_ALC) due to the resonant interaction of the electromagnetic (EM) waves through the second sound (SS), which is a new type of the cubic non-linearity. It is shown that a parametric amplification of one EM wave by the other occurs. Explicit expressions for the EM and SS wave amplitudes are obtained, which appear to be of the kink and transverse spatial soliton form, respectively. It is also shown that the SS wave creates flexoelectric polarization and space charge. The estimated gain is one or two orders of magnitude greater than the gain for the stimulated Brillouin scattering in isotropic organic liquids. The theoretical results obtained may be used as a basis for an experimental investigation of SS high frequency behaviour.

1. Introduction

The non-linear optics of smectic A liquid crystals (S_ALC) have not been sufficiently studied so far, especially in comparison with the non-linear optics of nematic liquid crystals (NLC) [1–3]. The unique properties of S_ALCs, which are viscous fluids with a layered structure [4–6], give rise to a specific kind of optical non-linearity connected with the so-called second sound (SS). SS is the propagating mode due to the smectic layer deformations [4–9, 16–19]. SS is the macroscopic manifestation of the oscillations of the phase of the smectic order parameter. This phase is proportional to the layer displacement $u(\mathbf{r}, t)$ along the normal to the layers, which is conventionally chosen to be the z axis [6–9]. Unlike normal sound, SS propagates without a change of mass density [4–9, 10–13].

The dispersion relation of SS has the form [4]

$$\Omega = \left(\frac{B}{\rho}\right)^{1/2} \frac{k_{sx}k_{sz}}{k_s} \tag{1}$$

where Ω, $\mathbf{k}_s$, k_{sx}, k_{sz} are the SS frequency, wave vector and its components along and normal to the layers, respectively; B is the elastic constant corresponding to the layer compression, ρ is the mass density of the S_ALC. It is known [4, 6, 9, 14, 15] that $B \sim 10^8\,\mathrm{erg\,cm^{-3}}$ and $\rho \sim 1\,\mathrm{g\,cm^{-3}}$. The SS velocity

$$s = \left(\frac{B}{\rho}\right)^{1/2} \sim 10^4\,\mathrm{cm\,s^{-1}}$$

is much lower than the ordinary sound velocity [6, 14]. It is seen from (1) that a SS propagating mode may exist only if $\mathbf{k}_s$ is oblique to the layers. Otherwise the SS mode degenerates into a purely dissipative over damped mode [4]. Experimentally, SS was

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first observed in elastic Brillouin scattering from monodomain samples of S_ALCs [16]. It was then investigated by ultrasound methods [17–19] and by means of Rayleigh scattering and the 'interdigital electrodes technique' [14, 15].

It is well-known that every kind of light scattering can become stimulated, if the pump intensity is sufficiently high [20]. As a consequence of the fact that B is much smaller than the elastic constant corresponding to bulk compression [4, 6], stimulated Brillouin light scattering on SS in S_ALCs is expected to occur at a lower level of pumping. Attempts have been made to consider theoretically the possible non-linear optical effects in S_ALCs [21–24, 25, 26]. The studies [23, 25] have been concerned with the case characterized by

$$k_{sz}=0, \quad \frac{\partial u}{\partial z}=0 \tag{2}$$

in which there has been no SS.

The situation was also analysed when the non-linearity of S_ALCs was determined by the thermal and mass density oscillations, while layer deformations were ignored [26]. The opposite case, in which SS existed, while thermal oscillations and ordinary sound were ignored, was considered in [21, 22, 24]. Parametric amplification in the non-resonant case had been predicted [21]. The threshold of the stimulated scattering and the gain have been calculated for the case of non-vanishing light absorption in S_ALCs with only one strong incident EM wave [22], using the constant pump approximation [20]. In the time-dependent case, the EM wave undergoes self-modulation and a SS soliton occurs, provided that the pumping is sufficiently strong [24]. It has been shown [21], that the SS and EM waves are strongly coupled when the EM waves propagate obliquely to the layers and their frequency shift

$$\Delta\omega \equiv \omega_1 - \omega_2$$

is of the order of $(s/c)\omega_1$, where c is the vacuum light velocity, ω_1 is the EM wave frequency and $\Delta\omega$ is much smaller than either ω_1 or the other EM wave frequency, ω_2.

In this paper we consider two-wave mixing due to SS in S_ALCs in the resonant case, for which the difference between the frequencies of the two EM waves is equal to the SS frequency

$$\Delta\omega = \Omega. \tag{3}$$

It is shown that parametric amplification of one EM wave by the other is possible. The kink states [28–30] of the coupled EM wave, SS wave and the small amplitude scattered harmonics may be excited. The gain at the resonant two-wave mixing due to SS in S_ALCs is one or two orders of magnitude larger than the one at the stimulated Brillouin scattering in isotropic organic liquids. The pump intensity applied may be much larger in S_ALCs than at the stimulated scattering on the orientational non-linearity in NLCs. The phenomenon considered in this paper is a new kind of two-wave mixing. S_ALCs may be characterized in the framework of the general approach [41] as a Kerr medium with a specific mechanism of the Kerr effect determined by the deformation of the smectic layers, which results in a Brillouin-like stimulated light scattering. The stimulated SS gives rise to flexoelectric polarization and space charge.

The analysis of the non-linear optical effects mentioned above is carried out using the Maxwell equations in the non-linear anisotropic and inhomogeneous medium and the equation of motion of the medium itself in the presence of the external EM field [31, 20].

The non-linearity is taken into account by including the terms proportional to the normal and tangential layer deformations $(\partial u/\partial x_i) \ll 1$ in the dielectric constant tensor ε_{ik}. These deformations turn out to be the small parameters of the problem. It should be noted that we do not use the constant pump approximation and linearized theory. Consequently, we obtained the distribution of both EM wave amplitudes in the general case. In the final part of the paper the results obtained are summarized.

2. Resonant coupling of EM waves through the second sound in S_ALCs

Consider the bulk effect, assuming the sample to be infinite. Since the liquid crystals of interest are transparent [1], there is no heating due to light absorption and the temperature can be taken as constant. The dielectric constant tensor ε_{ik} with the non-linear terms has the form [5]

$$\left.\begin{aligned} &\varepsilon_{xx}=\varepsilon_{\perp}+a_{\perp}\frac{\partial u}{\partial z}; \quad \varepsilon_{zz}=\varepsilon_{\parallel}+a_{\parallel}\frac{\partial u}{\partial z}, \\ &\text{and} \\ &\varepsilon_{xz}=\varepsilon_{zx}=-\varepsilon_a\frac{\partial u}{\partial x} \end{aligned}\right\} \tag{4}$$

where

$$\varepsilon_a=\varepsilon_{\parallel}-\varepsilon_{\perp}.$$

We take for ε_{ik} its value at the optical frequency. According to the relationship (3), the frequency dispersion of the ε_{ik} may be neglected. To analyse the non-linear optical process in a continuous inhomogeneous medium, we must find a self-consistent solution of the equations of motion of the non-linear medium and the Maxwell equations, with the non-linear polarization determined by the coupling between this medium and the external field [31, 20]. In our case, the equations of motion are the hydrodynamic equations of S_ALCs with the EM field-dependent terms, while the non-linear polarization is determined by the inhomogeneous part of ε_{ik}.

The free energy density of S_ALCs in the presence of the EM field E_i has the form [4, 6, 32]

$$F=\tfrac{1}{2}B\left(\frac{\partial u}{\partial z}\right)^2-\frac{1}{8\pi}\varepsilon_{ik}E_iE_k, \tag{5}$$

where the term $K(\partial^2u/\partial x^2)^2$ is neglected. The Frank constant $K\sim(10^{-7}$–$10^{-6})$ dyn and therefore [4, 6]

$$Kk_s^2 \ll \mathrm{B}.$$

The field-dependent terms in (5) are assumed to be averaged over the optical frequencies and only a slowly varying part remains [32, 20, 33]. The equations of the incompressible S_ALCs hydrodynamics have the form [4–6, 9]

$$\frac{\partial v_i}{\partial x_i}=0, \tag{6}$$

$$\frac{\partial u}{\partial t}=v_z, \tag{7}$$

$$\rho\frac{\partial v_i}{\partial t}=-\frac{\partial P}{\partial x_i}+g_i+\frac{\partial \sigma'_{ik}}{\partial x_k}, \tag{8}$$

$$\sigma'_{ik}=\alpha_0\delta_{ik}\mathscr{A}_{ll}+\alpha_1\delta_{iz}\delta_{kz}\mathscr{A}_{zz}+\alpha_4\mathscr{A}_{ik}+\alpha_{56}(\delta_{iz}\mathscr{A}_{zk}+\delta_{kz}\mathscr{A}_{zi})+\alpha_7\delta_{iz}\delta_{kz}\mathscr{A}_{ll}, \tag{9}$$

$$\mathscr{A}_{ik}=\frac{1}{2}\left(\frac{\partial v_i}{\partial x_k}+\frac{\partial v_k}{\partial x_i}\right), \tag{10}$$

$$\mathbf{g}=-\frac{\delta F}{\delta \mathbf{u}}, \tag{11}$$

where v_i is the hydrodynamic velocity, P is the pressure, σ'_{ik} is the viscosity stress tensor, α_i are the Leslie coefficients, $\mathbf{g}$ is the generalized force density, $\delta_{ik}=0$, $i\neq k$, $\delta_{ik}=1$, $i=k$.

Since $u(\mathbf{r},t)$ is determined as the layer displacement along the z axis [4–6, 9],

$$g_{x,y}\equiv 0,\quad g_z=\frac{d}{dx}\left[\frac{\partial F}{\partial(\partial u/\partial x)}\right]+\frac{d}{dz}\left[\frac{\partial F}{\partial(\partial u/\partial z)}\right]. \tag{12}$$

S_ALCs possesses D_∞ symmetry, being rotationally symmetrical relative to the optical axis z, which is normal to the layers [4]. We may therefore choose the coordinate system such that the xz plane coincides with the plane of incidence of the EM wave. Nevertheless, for arbitrary polarization of the incident wave, the transverse component E_y remains. In this paper, we consider the case in which all wave vectors belong to the xz plane. Substituting the relationships (4), (5), (9)–(12) into the system (6)–(8), we obtain the equation of motion of S_ALCs in the external electric field [21, 34]

$$-\rho\nabla^2\frac{\partial^2 u}{\partial t^2}+\left[\alpha_1\frac{\partial^4}{\partial x^2\partial z^2}+\tfrac{1}{2}(\alpha_4+\alpha_{56})\nabla^2\nabla^2\right]\frac{\partial u}{\partial t}$$
$$+B\frac{\partial^4 u}{\partial x^2\partial z^2}=\frac{1}{8\pi}\frac{\partial^2}{\partial x^2}\left\{\frac{\partial}{\partial z}(a_\perp E_\perp^2+a_\parallel E_z^2)-2\varepsilon_a\frac{\partial}{\partial x}(E_xE_z)\right\}, \tag{13}$$

where

$$E_\perp^2=E_x^2+E_y^2.$$

It is seen from (13) that, unlike the ordinary sound determined by the bulk compression [35], SS exists without mass density change. If the external field is absent and the viscosity term is omitted, we obtain the SS equation in the de Gennes approximation with the dispersion relation (1) [4]. The equation (13) contains third and fourth order spatial derivatives and fourth and fifth order mixed derivatives, while the equation describing Brillouin scattering on ordinary sound includes second order spatial and time derivatives and a third order mixed derivative [31, 20]. The reason is that unlike solids or isotropic liquids, the system (6)–(8) contains the specifically smectic equation (7). This equation expresses the continuity of the smectic layers, which means that permeation is neglected [14, 15]. It is also seen from equation (13), that the EM waves must propagate obliquely to the layers, displaying a dependence on both x and z. Otherwise, the right hand side of (13) vanishes. This requirement is in accord with SS dispersion (1). The Maxwell equations for the non-linear inhomogeneous anisotropic medium yield the wave equation [31, 20]

$$\operatorname{rot}\operatorname{rot}\mathbf{E}^{\mathrm{tot}}+\frac{1}{c^2}\frac{\partial^2\mathscr{D}^{\mathrm{L}}}{\partial t^2}=-\frac{1}{c^2}\frac{\partial^2\mathscr{D}^{\mathrm{N}}}{\partial t^2},\tag{14}$$

where

$$\mathscr{D}_i^{\mathrm{L}}+\mathscr{D}_i^{\mathrm{N}}=\varepsilon_{ik}E_k^{\mathrm{tot}},$$

$$|\mathscr{D}_i^{\mathrm{N}}|\ll|\mathscr{D}_i^{\mathrm{L}}|,$$

$\mathscr{D}_i^{\mathrm{L}}$, $\mathscr{D}_i^{\mathrm{N}}$ are the linear and non-linear parts of the electric induction.

In the two-wave mixing case the total electric field in $\mathrm{S_A}$LCs $\mathbf{E}^{\mathrm{tot}}$, consists of a finite number of Fourier components [31, 20]

$$\mathbf{E}^{\mathrm{tot}}=\mathbf{E}_1+\mathbf{E}_2+\mathbf{f}_1+\mathbf{f}_2,\quad |\mathbf{f}_{1,2}|\ll|\mathbf{E}_{1,2}|.\tag{15}$$

We take the incident waves $\mathbf{E}_{1,2}$ in the form

$$\mathbf{E}_{1,2}=\mathbf{e}_{1,2}\{A_{1,2}(\mathbf{r})\exp i(\mathbf{k}_{1,2}\mathbf{r}-\omega_{1,2}t)+\text{c.c.}\},\tag{16}$$

where $\mathbf{e}_{1,2}$, $\mathbf{k}_{1,2}$ are the unit polarization vectors and wave vectors, respectively, c.c. means complex conjugation, and it is assumed that $A_{1,2}(\mathbf{r})$ are slowly varying [31, 20, 33]

$$|\nabla^2A_i|\ll|k_i\nabla A_i|.\tag{17}$$

The time dependence of A_i may be neglected, since $s\ll c$, and therefore $A_{1,2}$ vary insignificantly during the time needed for light to traverse the interaction region [20]. We use the infinite plane-wave approximation [20], neglecting the transverse part of (∇A_i). Taking into account (4), (15), (17), we divide the equation (14) in the standard way [31, 20, 33] into linear and non-linear parts. In the linear approximation, we have for the incident wave $\mathbf{E}_{1,2}$ the homogeneous system of linear algebraic equations [32]

$$k_{1,2}^2e_{1,2i}-k_{1,2i}(k_{1,2k}e_{1,2k})-\varepsilon_{ik}^{\mathrm{o}}e_{1,2k}\left(\frac{\omega_{1,2}}{c}\right)^2=0,\tag{18}$$

where

$$\varepsilon_{zz}^{\mathrm{o}}=\varepsilon_{\|},\quad \varepsilon_{xx}^{\mathrm{o}}=\varepsilon_{\perp}.$$

For uniaxial media such as $\mathrm{S_A}$LCs, the system (18) has two kinds of solution: extraordinary waves and ordinary waves [32]. The extraordinary waves have the dispersion relation [32]

$$\frac{(k_{1,2z}^{\mathrm{e}})^2}{\varepsilon_{\perp}}+\frac{(k_{1,2x}^{\mathrm{e}})^2}{\varepsilon_{\|}}=\left(\frac{\omega_{1,2}}{c}\right)^2.\tag{19}$$

These waves are polarized in the incidence plane

$$\mathbf{e}_{1,2}^{\mathrm{e}}=(e_{1,2x},\,0,\,e_{1,2z}).\tag{20}$$

The ordinary waves have the dispersion relation [32]

$$(k_{1,2}^{\mathrm{o}})^2=\varepsilon_{\perp}\left(\frac{\omega_{1,2}}{c}\right)^2,\tag{21}$$

and their polarization is normal to the the incidence plane

$$e^{o}_{1,2}=e^{o}_{1,2y}=1. \tag{22}$$

It is seen from equations (19)–(21) that for each frequency, four modes may be realized

$$\mathbf{E}\sim \exp i[\pm k_x x \pm k_z z-\omega t]. \tag{23}$$

We define as positive the directions k_{1x}, k_{1z} and consider the two cases

$$\left.\begin{aligned}&(1)\quad k_{2x},\,k_{2z}>0\\ \text{and}\qquad &(2)\quad k_{2x},\,k_{2z}<0.\end{aligned}\right\} \tag{24}$$

Consider first of all the interaction of the extraordinary waves with the wave vectors

$$k^{e}_{1,2x}>0,\quad k^{e}_{1,2z}>0. \tag{25}$$

Substituting (16) into (13) we obtain for the layer displacement $u(\mathbf{r},t)$

$$u(\mathbf{r},t)=U\exp i\{(\mathbf{k}^{e}_1-\mathbf{k}^{e}_2)\mathbf{r}-(\omega_1-\omega_2)t\}+\text{c.c.}, \tag{26}$$

$$U=\frac{i(k^{e}_{1x}-k^{e}_{2x})^2\,hA_1A_2^*}{4\pi\rho(\mathbf{k}^{e}_1-\mathbf{k}^{e}_2)^2\,G(\mathbf{k}_{1,2},\omega_{1,2})}, \tag{27}$$

where the asterisk denotes complex conjugation,

$$h=(k^{e}_{1z}-k^{e}_{2z})(a_\perp e_{1x}e_{2x}+a_\parallel e_{1z}e_{2z})-\varepsilon_a(k^{e}_{1x}-k^{e}_{2x})(e_{1x}e_{2z}+e_{1z}e_{2x}),$$

$$G(\mathbf{k}_{1,2},\omega_{1,2})=(\omega_1-\omega_2)^2-\Omega^2+i(\omega_1-\omega_2)\Gamma$$

and

$$\Gamma=\frac{1}{\rho}\left[\alpha_1\frac{(k^{e}_{1x}-k^{e}_{2x})^2\,(k^{e}_{1z}-k^{e}_{2z})^2}{(\mathbf{k}^{e}_1-\mathbf{k}^{e}_2)^2}+\tfrac{1}{2}(\alpha_4+\alpha_{56})\times(\mathbf{k}^{e}_1-\mathbf{k}^{e}_2)^2\right].$$

Combining the resonance condition (3) and the SS dispersion relation (1) we obtain

$$(\Delta\omega)^2=s^2\left(\frac{k_{sx}k_{sz}}{k_s}\right)^2 \tag{28}$$

where

$$\mathbf{k}_s=\mathbf{k}^{e}_1-\mathbf{k}^{e}_2.$$

Unlike ordinary Brillouin scattering, the relationships (3) and (28) give not only the frequency and wave vector conservation conditions, but also the selection rule for the directions of propagation of EM waves. The substitution of (28) into (27) yields

$$U=\left(\frac{k_{sx}}{k_s}\right)^2\frac{hA_1A_2^*}{4\pi\rho\Delta\omega\Gamma}. \tag{29}$$

The magnitude of $\mathbf{k}_s$ may be determined using the cosine theorem

$$k_s^2=k_1^2+k_2^2-2k_1k_2\cos\theta, \tag{30}$$

where θ is the angle between $\mathbf{k}_1$ and $\mathbf{k}_2$

$$\theta=\varphi_1-\varphi_2,$$

where $\varphi_{1,2}$ are the angles between $\mathbf{k}_{1,2}$ and the x axis, respectively.

Substituting relationships (4), (16), (19), (26) and (29) into (14), separating the non-linear part, and neglecting $\nabla^2 A_{1,2}$ according to the condition (17), we obtain the reduced equations for the slowly varying amplitudes

$$A_{1,2} = |A_{1,2}| \exp i\gamma_{1,2} \tag{31}$$

as well as wave equations for the scattered harmonics

$$\mathbf{f}_{1,2} = \mathbf{f}^{\circ}_{1,2} \exp i[(\mathbf{k}_{1,2} \pm \mathbf{k}_s)\mathbf{r} - (\omega_{1,2} \pm \Omega)t] + \text{c.c.} \tag{32}$$

3. Parametric amplification and Brillouin-like scatering

The reduced equations for the amplitudes (31) and wave equations for the harmonics (32) have the form

$$\frac{\partial \gamma_{1,2}}{\partial R} = 0 \tag{33}$$

$$-l_{1,2}\left(\frac{\omega_{1,2}}{c}\right)^{-2} \frac{\partial |A_{1,2}|^2}{\partial R} = \pm\left(\frac{k_{sx}}{k_s}\right)^2 \frac{h^2 |A_1 A_2|^2}{4\pi\rho\Delta\omega\Gamma} \tag{34}$$

and

$$\operatorname{rot}\operatorname{rot}\mathbf{f}_{1,2} + \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\mathscr{D}^{\mathrm{L}}(\omega_{1,2} \pm \Omega)$$

$$= -\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\mathscr{D}^{\mathrm{N}}(\omega_{1,2} \pm \Omega), \tag{35}$$

where

$$R = x\cos\alpha + z\sin\alpha,$$

$$l_{1,2} = k^{\mathrm{e}}_{1,2z}\sin\alpha + k^{\mathrm{e}}_{1,2x}\cos\alpha - (\mathbf{k}^{\mathrm{e}}_{1,2}\mathbf{e}_{1,2}) \times (e_{1,2z}\sin\alpha + e_{1,2x}\cos\alpha),$$

$$\mathscr{D}^{\mathrm{L}}_x(\omega_{1,2} \pm \Omega) = \varepsilon_\perp f_{1,2x}, \quad \mathscr{D}^{\mathrm{L}}_z(\omega_{1,2} \pm \Omega) = \varepsilon_\parallel f_{1,2z},$$

$$\mathscr{D}^{\mathrm{N}}(\omega_1 + \Omega) = i\left(\frac{k_{sx}}{k_s}\right)^2 \frac{hA_1^2 A_2^*}{4\pi\rho\Delta\omega\Gamma}\mathbf{d}_1 \exp i[(\mathbf{k}_1 + \mathbf{k}_s)\mathbf{r} - (\omega_1 + \Omega)t] + \text{c.c.},$$

$$\mathscr{D}^{\mathrm{N}}(\omega_2 - \Omega) = -i\left(\frac{k_{sx}}{k_s}\right)^2 \frac{hA_2^2 A_1^*}{4\pi\rho\Delta\omega\Gamma}\mathbf{d}_2 \exp i[(\mathbf{k}_2 - \mathbf{k}_s)\mathbf{r} - (\omega_2 - \Omega)t] + \text{c.c.},$$

$$d_{1,2x} = a_\perp k^{\mathrm{e}}_{sz} e_{1,2x} - \varepsilon_a k^{\mathrm{e}}_{sx} e_{1,2z}$$

and

$$d_{1,2z} = a_\parallel k^{\mathrm{e}}_{sz} e_{1,2z} - \varepsilon_a k^{\mathrm{e}}_{sx} e_{1,2x}.$$

The angle α determines the direction along which the amplitudes are varying. For $\theta \ll 1$, this angle determines the beam vector direction, which does not coincide with the wave vector in anisotropic media [32]:

$$\alpha = \varphi_1 + \Delta\theta' \tag{36}$$

$$\operatorname{tg}\Delta\theta' \cong \frac{(\varepsilon_\perp - \varepsilon_\parallel)}{\varepsilon_\parallel} \frac{\operatorname{ctg}\varphi_1}{1 + \dfrac{\varepsilon_\perp}{\varepsilon_\parallel}\operatorname{ctg}^2\varphi_1} \tag{37}$$

The angle $\Delta\theta'$ is sufficiently small since the optical anisotropy of S_ALCs satisfies the condition [1]

$$\frac{1}{\varepsilon_{\parallel}}(\varepsilon_{\parallel}-\varepsilon_{\perp})\ll 1.$$

We shall consider the case in which the two incident waves $\mathbf{E}_1$ and $\mathbf{E}_2$ propagate in the direction close to parallel, i.e.

$$\Delta\theta'\approx\theta\ll 1.$$

In this case the resonance condition (28) has the form

$$\Delta\omega=\frac{s}{c}\omega_1|\theta|\sin\varphi_1\sin 2\varphi_1\frac{\varepsilon_{\perp}}{\varepsilon_{\parallel}}\left(\varepsilon_{\perp}-\frac{\varepsilon_{\perp}}{\varepsilon_{\parallel}}\cos^2\varphi_1\right)^{-1/2}$$
$$\times\left[\sin^2\varphi_1+\left(\frac{\varepsilon_{\perp}}{2\varepsilon_{\parallel}}\right)^2\frac{\sin^2 2\varphi_1}{\varepsilon_{\perp}-(\varepsilon_{\perp}/\varepsilon_{\parallel}\cos^2\varphi_1)}\right]^{-1/2} \tag{38}$$

The system (33)–(34) has the following solution

$$\gamma_{1,2}=\text{constant}, \tag{39}$$

$$c^2\left\{\frac{l_1}{\omega_1^2}|A_1|^2+\frac{l_2}{\omega_2^2}|A_2|^2\right\}=I_0=\text{constant}, \tag{40}$$

where I_0 is the integral of motion.

$$|A_{1,2}|^2=\frac{1}{2}I_0\left(\frac{\omega_{1,2}}{c}\right)^2\frac{1}{l_{1,2}}\left\{1\mp\tanh\frac{\beta}{2}(R-R_0)\right\}. \tag{41}$$

$$\beta=\left(\frac{\omega_1\omega_2}{c^2}\right)^2 I_0\left(\frac{k_{sx}}{k_s}\right)^2\frac{h^2}{4\pi\rho l_1 l_2\Delta\omega\Gamma}. \tag{42}$$

R_0 is the crossing-point, where

$$c^2\frac{l_1}{\omega_1^2}|A_1|^2=c^2\frac{l_2}{\omega_2^2}|A_2|^2=\frac{I_0}{2}. \tag{43}$$

If $\Delta\omega>0$, $\beta>0$ and we have

$$|A_1|^2\to 0,\quad |A_2|^2\to I_0\left(\frac{\omega_2}{c}\right)^2\frac{1}{l_2},$$

for $R\to\infty$ and

$$|A_1|^2\to I_0\left(\frac{\omega_1}{c}\right)^2\frac{1}{l_1},\quad |A_2|^2\to 0, \tag{44}$$

for $R\to-\infty$.

It is seen from relationships (41) and (44) that a parametric amplification of the signal EM wave $\mathbf{E}_2$ with the lower frequency ω_2 and the depletion of the pumping EM wave $\mathbf{E}_1$ with the higher frequency occur, while the SS wave plays a role of an idler wave [20]. The amplification and depletion of the respective EM waves take place mainly at the interval $|R-R_0|\leqslant 2/\beta$. Since we have taken into account the depletion of the pumping, the process of amplification reaches saturation at $|R-R_0|\gg 2/\beta$. The stable states described by the relationships (41) are usually referred to as kinks [27, 28, 36].

Substituting (32) and (41) into (35), we calculate the harmonics $\mathbf{f}_{1,2}$, taking into account only the particular solutions governed by the expressions on the right hand side of (35), since the frequencies $(\omega_{1,2} \pm \Omega)$ and wave vectors $(\mathbf{k}_{1,2} \pm \mathbf{k}_s)$ cannot satisfy the homogeneous part of the equations (35), and therefore homogeneous solutions in that case do not exist [3, 10]. We have the following explicit expressions for the amplitudes $\mathbf{f}^{\circ}_{1,2}$:

$$\mathbf{f}^{\circ}_{1,2} = \pm i \frac{\beta}{h} (I_0 l_{2,1})^{1/2} \frac{(\omega_{1,2} \pm \Omega)^2}{2^{3/2} \omega_{2,1} c d_0} \left[\cosh \frac{\beta}{2} (R - R_0) \right]^{-3/2}$$

$$\times \exp\left\{ \mp \frac{\beta}{4} (R - R_0) \right\} [p_{1,2} \mathbf{x} + q_{1,2} \mathbf{z}] \tag{45}$$

where

$$d_0 = 4\varepsilon_{\|}\varepsilon_{\perp} \left(\frac{k^{\mathrm{e}}_{1z} k^{\mathrm{e}}_{2z}}{\varepsilon_{\perp}} + \frac{k^{\mathrm{e}}_{1x} k^{\mathrm{e}}_{2x}}{\varepsilon_{\|}} - \frac{\omega_1 \omega_2}{c^2} \right),$$

$$p_{1,2} = d_{1,2x} \left[(k^{\mathrm{e}}_{1,2x} \pm k_{sx})^2 - \varepsilon_{\|} \frac{(\omega_{1,2} \pm \Omega)^2}{c^2} \right] + d_{1,2z} (k^{\mathrm{e}}_{1,2z} \pm k_{sz}) (k^{\mathrm{e}}_{1x} \pm k_{sx}),$$

$$q_{1,2} = d_{1,2z} \left[(k^{\mathrm{e}}_{1,2z} \pm k_{sz})^2 - \varepsilon_{\perp} \frac{(\omega_{1,2} \pm \Omega)^2}{c^2} \right] + d_{1,2x} (k^{\mathrm{e}}_{1,2z} \pm k_{sz}) (k^{\mathrm{e}}_{1,2x} \pm k_{sx}).$$

The amplitudes $|f^{\circ}_{1,2}|$ have the form of spatial solitons [27, 28, 29] with symmetric maxima at the points $R_{1,2m}$

$$R_{1,2m} = R_0 \mp \frac{1}{\beta} \ln 2, \tag{46}$$

while

$$\frac{|f^{\circ}_{1,2}|_{\max}}{|f^{\circ}_{1,2}(R_0)|} = 2 \left(\frac{2}{3} \right)^{3/2}, \tag{47}$$

$$|f^{\circ}_{1,2}| \to 0 \quad \text{at} \quad R \to \pm\infty. \tag{48}$$

Inserting (41) into (29) we obtain the SS wave amplitude

$$|U| = I_0 \left(\frac{k_{sx}}{k_s} \right)^2 \frac{h \omega_1 \omega_2}{4\pi\rho (l_1 l_2)^{1/2} c^2 \Delta\omega \Gamma} \left[\cosh \frac{\beta}{2} (R - R_0) \right]^{-1}. \tag{49}$$

The SS wave represents the transverse spatial soliton travelling in the direction of $\mathbf{k}_s$, which is approximately normal to the wave fronts of the coupled EM waved $\mathbf{E}_{1,2}$. The width of the soliton (49) is

$$W = \frac{2}{\beta},$$

and therefore the region in which the EM waves couple effectively is confined mainly to the interval near the crossing-point R_0

$$|R - R_0| < \frac{2}{\beta}.$$

A comparison of the expressions (45), (49) indicates that the scattered harmonics $\mathbf{f}_{1,2}$ are localized in the same interval. The expressions (41), (45), (49) show that all the excitations are of the convective type, since they exist only within the non-linear medium [36]. It may be demonstrated that the coupling of ordinary waves would yield the same results for $A_{1,2}$, U if we replaced E_x, $E_\perp$ by E_y, $\mathbf{k}^e$ by $\mathbf{k}^o$ and h by the expression

$$h_0 = a_\perp k_{sz} \tag{50}$$

in further relationships.

The scattered harmonics would be polarized along the y axis, but they would keep the form of spatial solitons. The resonance condition (38) must be replaced by the relation

$$|\Delta\omega| = \frac{s}{c}\omega_1\sqrt{(\varepsilon_\perp)}\frac{|\theta|}{2}\sin 2\varphi_1, \tag{51}$$

since the dispersion relation (21) replaces relationship (19).

Consider now the case in which

$$k_{1x}, k_{1z} > 0, \quad k_{2x}, k_{2z} < 0.$$

In this case the condition $\theta \ll 1$ must be replaced by the condition

$$\pi - \theta \ll 1 \tag{52}$$

Then the wave $\mathbf{E}_2$ and the SS wave u have the form

$$\mathbf{E}_2 = \mathbf{e}_2\{A_2(\mathbf{r})\exp i(\mathbf{k}_2\mathbf{r} + \omega_2 t) + \text{c.c.}\}, \tag{53}$$

$$u = U\exp i[(\mathbf{k}_1 + \mathbf{k}_2)\mathbf{r} - (\omega_1 - \omega_2)t] + \text{c.c.} \tag{54}$$

All further results would remain the same, if we replaced A_2^* by A_2 in the expression (27), transformed the second equation in the system (34) into the complex conjugate one for A_2^* and introduced $(-|l_2|)$ instead of l_2, since now $l_2 < 0$. In the resonance condition (28) we must use

$$\mathbf{k}_s = \mathbf{k}_1 + \mathbf{k}_2. \tag{55}$$

It should be noted that for parallel or antiparallel propagation ($\theta = 0, \pi$), the resonance condition (28) cannot be fulfilled. Indeed in such a case

$$k_s = k_1 - |k_2| \sim \frac{\omega_1 - \omega_2}{c},$$

so that the left-hand side of (28) is proportional to $(\omega_1 - \omega_2)$, while the right-hand side is proportional to

$$\frac{s}{c}(\omega_1 - \omega_2) \ll (\omega_1 - \omega_2).$$

It is clear from the relationship (42) that, in the resonant case, the gain β is determined by the frequency difference $\Delta\omega$ and SS time decay Γ, just as in the case of stimulated scattering in NLCs [3, 23].

As shown in [4, 5], the decay constant of the purely dissipative overdamped orientational mode in NLCs is

$$\Gamma_{\text{N}} \sim \frac{\mathcal{K}k^2}{\alpha_i},$$

which is much shorter than the SS decay constant Γ. It is known that the low energy of the molecular reorientation in NLCs $\mathscr{K}k^2$, determines the so-called giant optical non-linearity (GON) [3]; on the other hand, the small magnitude of $\mathscr{K}k^2$ and Γ_N limits the pumping level. In the case of SS-determined non-linearity in S_ALCs, there is no GON, but the pumping level may be much greater, since it is limited by the elastic constant B. We may conclude that the magnitude of the non-linearity in S_ALCs is intermediate between the GON in NLCs and the non-linearity determined by the bulk compression in solids or isotropic liquids.

Introducing the intensity $\mathscr{P}$ according to [31, 32, 20]

$$\mathscr{P}=\frac{c}{4\pi}|A|^2, \tag{56}$$

we may evaluate numerically the relative gain per unit intensity $\beta/\mathscr{P}$, measured in cm (MW)$^{-1}$

$$\frac{\beta}{\mathscr{P}}\sim\frac{\omega_1\varepsilon_a^2\Delta\omega}{\rho s^2c^2\Gamma}. \tag{57}$$

Using typical values for the material parameters $\alpha_i\sim(0{\cdot}1\text{–}1)$ Poise [4, 14, 15], $\varepsilon_a=0{\cdot}6$ [1] and choosing $\theta\sim0{\cdot}1$, $\omega_1\sim10^{15}$ Hz, we obtain

$$\frac{\beta}{\mathscr{P}}\sim(0{\cdot}1\text{–}1)\,\mathrm{cm\,(MW)^{-1}}.$$

This value of $\beta/\mathscr{P}$ is one or two orders of magnitude greater than the gain for the ordinary stimulated Brillouin scattering in isotropic organic liquids [20]. For the input intensity $\mathscr{P}\sim100$ MW cm^{-2}, we obtain the gain $\beta\sim(10\text{–}100)$ cm^{-1} and the gain length L would be

$$L\sim\frac{2}{\beta}\sim(0{\cdot}02\text{–}0{\cdot}2)\,\mathrm{cm}.$$

This estimation is in a good agreement with the experimental data obtained by I.C. Khoo *et al.* [37].

4. The light induced polarization, space charge and electric field

In liquid crystals, a specific kind of polarization may exist which is connected with the orientational deformations—the so-called flexoelectric polarization [6, 38, 39]. In S_ALCs, the flexoelectric effect is determined by the layer deformation [7, 38, 39]. It has been shown theoretically and experimentally that SS may be excited in S_ALCs due to flexoelectric couplings, if an inhomogeneous high-frequency electric field is applied [14, 15]. We show that the inverse effect may occur, when the SS wave (26), (27) governed by the interfering waves generates the flexoelectric polarization $\mathbf{P}_{fe}$, space charge Q_{fe} and electrostatic longitudinal wave $\mathbf{E}_{fe}$.

$\mathbf{P}_{fe}$ and Q_{fe} have the form [7]

$$\mathbf{P}_{fe}=-e_3^f\frac{\partial^2u}{\partial z\partial x}\mathbf{x}-\left(e_1^f\frac{\partial^2u}{\partial x^2}+e_2^f\frac{\partial^2u}{\partial z^2}\right)\mathbf{z} \tag{58}$$

and

$$Q_{fe} = -\operatorname{div} \mathbf{P}_{fe} = (e_1^f + e_3^f)\frac{\partial^3 u}{\partial x^2 \partial z} + e_2^f \frac{\partial^3 u}{\partial z^3}, \tag{59}$$

where $e_{1,2,3}^f$ are the flexoelectric coupling constants [7].

Substituting (26) and (49) into (58), (59) we obtain

$$\mathbf{P}_{fe} = I_0\left(\frac{k_{sx}}{k_s}\right)^2 \frac{h\omega_1\omega_2}{4\pi\rho(l_1 l_2)^{1/2}c^2\Delta\omega\Gamma}\left[\cosh\frac{\beta}{2}(R-R_0)\right]^{-1}$$
$$\times [e_3^f k_{sz}k_{sx}\mathbf{x} + (e_1^f k_{sx}^2 + e_2^f k_{sz}^2)\mathbf{z}] \exp i(\mathbf{k}_s\mathbf{r} - \Omega t) + \text{c.c.}, \tag{60}$$

$$Q_{fe} = iI_0\left(\frac{k_{sx}}{k_s}\right)^2 \frac{h\omega_1\omega_2}{4\pi\rho(l_1 l_2)^{1/2}c^2\Delta\omega\Gamma}\left[\cosh\frac{\beta}{2}(R-R_0)\right]^{-1}$$
$$\times [(e_1^f + e_3^f)k_{sz}k_{sx}^2 + e_2^f k_{sz}^3] \exp i(\mathbf{k}_s\mathbf{r} - \Omega t) + \text{c.c.} \tag{61}$$

It is seen from the relationship (60), that that the magnetic field $\mathbf{H}_{fe}$ connected with the flexoelectric polarization $\mathbf{P}_{fe}$ is small

$$|\mathbf{H}_{fe}| \sim \frac{s}{c}|\mathbf{P}_{fe}|,$$

and may therefore be neglected. Consequently, the polarization $\mathbf{P}_{fe}$ creates the electrostatic longitudinal wave $\mathbf{E}_{fe}$ [40]

$$\operatorname{rot} \mathbf{E}_{fe} = 0, \quad \operatorname{div} \mathscr{D}_{fe} = 0 \tag{62}$$

and

$$\mathscr{D}_{fe} = \varepsilon_\perp E_{fex}\mathbf{x} + \varepsilon_\parallel E_{fez}\mathbf{z} + 4\pi\mathbf{P}_{fe}. \tag{63}$$

Combining equations (60), (62) and (63), we find that

$$E_{fe} = -\mathbf{k}_s I_0\left(\frac{k_{sx}}{k_s}\right)^2 \frac{h\omega_1\omega_2}{\rho(l_1 l_2)^{1/2}c^2\Delta\omega\Gamma}$$
$$\times\left[\cosh\frac{\beta}{2}(R-R_0)\right]^{-1}\frac{[(e_1^f - e_3^f)k_{sz}k_{sx}^2 + e_2^f k_{sz}^3]}{(\varepsilon_\perp k_{sx}^2 + \varepsilon_\parallel k_{sz}^2)} \exp i(\mathbf{k}_s\mathbf{r} - \Omega t) + \text{c.c.} \tag{64}$$

It is seen from (60), (61) and (64), that the flexoelectric polarization, space charge and longitudinal electrostatic field have a form identical to that of the SS wave and are travelling in the direction determined by $\mathbf{k}_s$. The amplitudes of the waves $\mathbf{P}_{fe}$, Q_{fe}, $\mathbf{E}_{fe}$ are proportional to the intensity I_0 of the EM waves $\mathbf{E}_{1,2}$.

5. Conclusions

We now summarize the main results obtained. It is demonstrated that coupling through the SS may give rise to a strong parametric amplification of one EM wave at the expense of another. The amplitudes of the incident and scattered EM waves and the amplitude of the SS are calculated explicitly. The signal and pumping waves undergo amplification with saturation and depletion, respectively. The light stimulated SS wave creates the waves of the flexoelectric polarization and the space charge. Numerical estimates show that the gain per unit intensity at the stimulated scattering on SS in S_ALCs is one or two orders of magnitude greater than that of stimulated Brillouin scattering on ordinary sound in isotropic organic liquids. The scattering on SS occurs without a change of the S_ALCs mass density. In terms of [41], such a scattering may be described as a new kind of two-wave mixing in a Kerr medium, with the specific mechanism of a Kerr effect due to the smectic layer deformations. The gain depends on the material parameters of S_ALCs such as the Leslie viscosity coefficients α_i, the elastic constant B, and the dielectric constant anisotropy ε_a, as well as on the intensity, polarization and propagation direction of the coupled EM waves. Therefore, the stimulated scattering on SS may be used for the experimental investigation of the high-frequency hydrodynamics of S_ALCs in the presence of a strong external field.

The theoretical results obtained in the present article may be applied as a basis for such an investigation.

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