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Stimulated light scattering in smectic A liquid crystals

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The non-linear interaction of arbitrary polarized light with smectic layer deformations in smectic A liquids (S_A) is considered. It is shown that the combined effect of anisotropy, fluidity and a characteristic kind of deformation cause a number of specific non-linear optical phenomena. Two-wave mixing in S_A transforms into a partly degenerate four-wave mixing (FWM) when the polarization and the direction of propagation of the coupled electromagnetic (EM) waves are arbitrary. The interference of the EM waves gives rise to a dynamic grating of layer deformations without a change of mass density of S_A. In the resonant case a propagating mode of a second sound (SS) is excited. The non-linear phenomena are analysed by solving the self-consistent system of the Maxwell equations for the non-linear anisotropic inhomogeneous medium and the hydrodynamic equations of S_A in the external EM field. The explicit expressions of the EM and SS waves amplitudes are obtained. It is shown that the coupled fundamental EM waves undergo the parametric amplification and the phase cross-modulation, and their amplitudes as well as the SS wave amplitude are spatially localized. The energy transfer between the coupled EM waves is non-reciprocal. The scattering of the fundamental EM waves by the dynamic grating results in the appearance of additional harmonics with combination frequencies and wavevectors. The light induced dynamic grating also generates a longitudinal electric field due to the flexoelectric effect.

1. Introduction

Stimulated light scattering (SLS) is a result of parametric coupling of light and material excitation [1]. In the past few years a new type of SLS in nematic liquid crystals (NLC) determined by the molecular reorientation in the field of electromagnetic (EM) waves has been investigated [2–7]. It has been shown for the case of constant pumping intensity that the exponential amplification of one EM wave by another one was possible due to the so-called grating orientational non-linearity (ON) [2, 4]. The two-wave mixing on the grating ON in NLCs results in the parametric non-reciprocal energy transfer when one EM wave is amplified by the other one, which is in turn attenuated [6]. Since the change of NLC refractive index is proportional to the pumping intensity, NLC may be characterized as Kerr medium [8]. SLS in NLCs on grating ON was observed experimentally and the gain was measured [5]. It appeared to be of the magnitude of $5\times10^3\,\mathrm{cm\,MW^{-1}}$, that is at least four orders greater than the gain in the case of the ordinary stimulated Brillouin scattering in isotropic liquids [1]. On the other hand, the level of EM wave intensity in SLS on grating ON is limited by the low energy of the orientational deformation. It is clear that for sufficiently strong EM waves the approach based on the purely orientational mechanism is invalid and inapplicable. It should be noted that the orientational modes in NLCs are purely dissipative and overdamped [9, 10], and therefore a resonant excitation of grating ON in NLCs is impossible.

Unlike NLCs, in smectic A liquid crystals (S_A) a resonant propagating mode of the so-called second sound (SS) exists determined by the oscillations of the smectic order parameter [9–15]. This phase is proportional to a smectic layer displacement $u(\mathbf{r},t)$ along the Z axis normal to the layers [12, 14]. SS propagates without the change of S_A mass density and has the dispersion relation of the form [14, 15]

$$\Omega=\surd(B/\rho)\frac{k_{sx}k_{sz}}{k_s},\tag{1}$$

where Ω, $\mathbf{k}_s$ are the SS frequency and wavevector, respectively, B is the elastic constant corresponding to the layer compression, ρ is mass density. It is seen from equation (1) that SS exists only when $\mathbf{k}_s$ is oblique to the layers [9–22]. SS has been experimentally investigated by the spontaneous Brillouin scattering [16], by ultrasonic methods [17–20], and by means of the Rayleigh scattering and the 'interdigital electrode technique' [21, 22]. The elastic constant $B\approx10^8\,\mathrm{erg\,cm^{-3}}$ [12, 21, 22] has the intermediate magnitude between the value $\mathscr{K}k^2$ in NLCs and an elastic constant T_v corresponding to the bulk

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compression in organic liquids [10, 12, 14–22]: $\mathscr{K}k^2 \ll B \ll T_v$ where $\mathscr{K}$ is Frank constant [10].

Therefore the optical non-linearity in S_A caused by SS grating would be stronger than an ordinary Brillouin non-linearity in solids and isotropic liquids. It would also exist in the presence of strong EM waves in contrast to the grating ON in NLCs, although it is expected to be much less than the latter. Some cases of SLS in S_A for the EM waves with particular polarizations and propagation directions were theoretically analysed by one of the authors [6, 23, 24]. It has been shown in [6] that the SLS of the ordinary EM waves propagated in the layer plane was in general analogous to the SLS on grating ON in NLCs [4]. The parametric coupling of the extraordinary EM waves polarized in the incidence plane [25] through the SS grating has been considered, and it has been shown that one wave would be amplified by the other one, which in turn would be depleted [23]. The strong incident extraordinary EM wave polarized in the plane of incidence excited a secondary EM wave and SS wave with the conservation of the frequency and the wavevector [24]. Recently, a special case of the resonant SS excitation by two EM waves polarized either in the incidence plane, or normal to it has been considered [26]. It has been shown that both the intensity of the coupled EM waves and SS amplitude have a spatially localized distribution.

In this paper the general case of SLS in S_A is considered when coupled EM waves have arbitrary polarizations and propagation directions and the combined effect of the anisotropy and layered structure is taken into account. It is shown that in this general case the two-wave mixing in S_A transforms into the four-wave mixing (FWM) [1] because of the essential optical anisotropy of S_A. Each EM wave splits into the extraordinary and ordinary ones with the same frequency and different wavevectors [25]. We define such a process as a partly degenerate FWM, since there are four EM waves with two different frequencies. In the process of SLS the following chain of events takes place.

(i) The interfering EM waves create a dynamic grating of layer deformations which consists of four propagating harmonics with the same frequency and the different wavevectors.
(ii) The parametric amplification of the pair of the fundamental EM waves with the lower frequency by the other pair of EM waves with the higher frequency occurs.
(iii) The fundamental EM waves are scattering on the light-induced grating and create small EM harmonics with combination frequencies and wavevectors. The non-linearity also changes the polarization of the fundamental EM waves. It is also shown that light-induced dynamic grating of layer deformations gives rise to the longitudinal waves [27] with the SS frequency due to the flexoelectric effect [11, 21, 22, 28–30].

It is also shown that the fundamental EM waves amplitudes, SS waves, scattered harmonics and longitudinal electric field are spatially localized states.

The paper is constructed as follows. The partly degenerate FWM is considered in the second part. Using the coupled-wave approach, the slowly varying amplitudes approximation and infinite plane wave approximation [1] and assuming the process to be steady-state, we obtain the reduced equations for the slowly varying amplitudes of the fundamental EM waves and the wave equations for the secondary EM waves. In the third section the parametric amplification of the fundamental EM waves is analysed. The explicit expressions for the amplitudes of the fundamental EM waves and of the SS wave are obtained for the important case when one incident wave is mainly polarized normal to the incidence plane, while the other one mainly belongs to its incidence plane. The stability of the process is studied in the fourth section. It is shown that the fundamental solutions are stable. In the fifth section the Brillouin-like scattering is considered. The sixth section concerns the excitation of the longitudinal waves due to the flexoelectric effect. In the seventh section the results are summarized.

2. The partly degenerate FWM

The analysis of the non-linear optical process is based on the self-consistent solution of the coupled equations of motion of the non-linear medium and of the Maxwell equations for the EM waves propagating in the non-linear medium [1]. The terms responsible for the non-linear coupling are phenomenologically included into the dielectric constant tensor of the medium, while the terms determined by the external EM field are introduced into the equations of motion of the medium. Consider the homeotropically oriented S_A [10] filling the semi-space $z > 0$, while the semi-space $z < 0$ is filled with the homogeneous isotropic dielectric medium with the dielectric constant $\varepsilon_s < \varepsilon_\perp, \varepsilon_\parallel$. Neglecting a bulk compression the dielectric constant tensor ε_{ik} of S_A with the first order in the layer deformations has the form [9, 11]

$$\left.\begin{aligned} &\varepsilon_{xx} = \varepsilon_{yy} = \varepsilon_\perp + a_\perp \frac{\partial u}{\partial z}, \quad \varepsilon_{zz} = \varepsilon_\parallel + a_\parallel \frac{\partial u}{\partial z} \\ &\text{and} \\ &\varepsilon_{xz} = \varepsilon_{zx} = -\varepsilon_a \frac{\partial u}{\partial x}, \qquad \varepsilon_{yz} = \varepsilon_{zy} = -\varepsilon_a \frac{\partial u}{\partial y}, \\ &\varepsilon_a = \varepsilon_\parallel - \varepsilon_\perp. \end{aligned}\right\} \quad (2)$$

The hydrodynamics of S_A with layer displacement

$u(\mathbf{r}, t)$ as the dynamic variable is governed by the following system of equations [9, 10, 12–14]

$$\frac{\partial v_i}{\partial x_i} = 0, \quad (3)$$

$$\rho \frac{\partial v_i}{\partial t} = -\frac{\partial P}{\partial x_i} + \frac{\partial \sigma'_{ik}}{\partial x_k} + g_i, \quad (4)$$

$$\sigma'_{ik} = \alpha_0 \delta_{ik} \mathscr{A}_{ll} + \alpha_1 \delta_{iz} \delta_{zk} \mathscr{A}_{zz} + \alpha_4 \mathscr{A}_{ik} + \alpha_{56}(\delta_{iz} \mathscr{A}_{zk} + \delta_{kz} \mathscr{A}_{zi}) + \alpha_7 \delta_{iz} \delta_{zk} \mathscr{A}_{ll}, \quad (5)$$

$$\mathscr{A}_{ik} = \frac{1}{2}\left(\frac{\partial v_i}{\partial x_k} + \frac{\partial v_k}{\partial x_i}\right), \quad (6)$$

$$v_z = \frac{\partial u}{\partial t}, \quad (7)$$

$$\mathbf{g} = -\frac{\delta F}{\delta \mathbf{u}}, \quad g_{x,y} \equiv 0, \quad (8)$$

and

$$F = \tfrac{1}{2} B \left(\frac{\partial u}{\partial z}\right)^2 - \frac{1}{8\pi} \varepsilon_{ik} E_i E_k, \quad (9)$$

where $\mathbf{v}$ is the hydrodynamic velocity, $\alpha_i \approx 1$ Poise are Leslie viscosity coefficient [9,10], P is the pressure, $\mathbf{g}$ is the density of the generalized force, F is the free energy density, the term

$$\mathscr{K}\left(\frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 u}{\partial y^2}\right)^2 \ll B\left(\frac{\partial u}{\partial z}\right)^2$$

and may be omitted for $k_z \neq 0$ [10, 11]. Equation (7) expresses the continuity of the smectic layers; the slow process of permeation [10, 12] is neglected at high frequencies [21, 22]. In the following the plane of the layer is chosen to be the XY plane, $u(\mathbf{r}, t)$ is normal displacement of the layer, and therefore $\mathbf{g} \equiv g_z \mathbf{z}$.

Applying rot rot operator to the equation (4) and substituting the relationship (2), (3), (5)–(9) into (4) we obtain the equation of motion of S_A

$$-\rho \nabla^2 \frac{\partial^2 u}{\partial t^2} + \left\{\alpha_1 \left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right)\frac{\partial^2}{\partial z^2} + \frac{1}{2}(\alpha_4 + \alpha_{56})\nabla^2\nabla^2\right\} \times \frac{\partial u}{\partial t} + B\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right)\frac{\partial^2 u}{\partial z^2} = \frac{1}{8\pi}\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right) \times \left\{\frac{\partial}{\partial z}[a_\perp(E_x^2 + E_y^2) + a_\| E_z^2] - 2\varepsilon_a\left[\frac{\partial}{\partial x}(E_x E_z) + \frac{\partial}{\partial y}(E_y E_z)\right]\right\}, \quad (10)$$

where

$$\nabla^2 = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2}.$$

The electric field E_i of the EM waves is determined by the wave equation [1], [25]

$$\text{rot rot}\, \mathbf{E} + \frac{1}{c^2}\frac{\partial^2 \mathbf{D}^{\mathrm{L}}}{\partial t^2} = -\frac{1}{c^2}\frac{\partial^2 \mathbf{D}^{\mathrm{N}}}{\partial t^2}, \quad (11)$$

$$D^{\mathrm{L}}_{x,y} = \varepsilon_\perp E_{x,y}, \quad D^{\mathrm{L}}_x = \varepsilon_\| Ez, \quad (12)$$

and

$$D_i^{\mathrm{N}} = \varepsilon_{ik}^{\mathrm{N}} E_k \quad (13)$$

where $\mathbf{D}^{\mathrm{L}}$, $\mathbf{D}^{\mathrm{N}}$ are the linear and the non-linear parts of the electric induction, respectively; $\varepsilon_{ik}^{\mathrm{N}}$ is the non-linear part of the dielectric constant tensor (2)

$$\varepsilon_{IK}^{\mathrm{N}} = \hat{N}_{ik} u \quad (14)$$

where

$$\hat{N} = \begin{vmatrix} a_\perp \frac{\partial}{\partial z} & 0 & -\varepsilon_a \frac{\partial}{\partial x} \\ 0 & a_\perp \frac{\partial}{\partial z} & -\varepsilon_a \frac{\partial}{\partial y} \\ -\varepsilon_a \frac{\partial}{\partial x} & -\varepsilon_a \frac{\partial}{\partial y} & a_\| \frac{\partial}{\partial z} \end{vmatrix}. \quad (15)$$

The non-linearity is weak for the pumping intensities applicable:

$$|D_i^{\mathrm{N}}| \ll |D_i^{\mathrm{L}}| \quad (16)$$

since the layer deformations are small, as it will be shown below

$$\left|\frac{\partial u}{\partial x_i}\right| \ll 1. \quad (17)$$

SLS on SS in S_A is assumed to be the steady-state process similar to the ordinary Brillouin scattering [1], since the SS velocity is small in comparison with the light velocity c [10, 12, 14],

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

Under such conditions E_i may be represented as the sum of the finite number of the monochromatic harmonics with the time-independent amplitudes $A_i(z)$ slowly varying along the Z axis [1, 31]:

$$\mathbf{E} = \sum_i \mathbf{e}_i A_i(z) \exp i(\mathbf{k}_i \mathbf{r} - \omega_i t) + \sum_i \mathbf{f}_i^{\mathrm{S}} + \text{c.c.}, \quad (18)$$

$$\left|k_i \frac{\partial A_i}{\partial z}\right| \gg \left|\frac{\partial^2 A_i}{\partial z^2}\right|, \quad (19)$$

and

$$|f_i^{\mathrm{S}}| \ll |A_i|, \quad (20)$$

where $\mathbf{e}_i$, $\mathbf{k}_i$, ω_i are the unit polarization vectors, wavevectors and frequencies of the fundamental coupled EM waves, respectively, $\mathbf{f}_i^S$ are the scattered harmonics governed by the non-linear electric induction (13), c.c. means complex conjugate.

Let the two incident EM waves propagating in the semi-space $z<0$ have the form

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

where (21)

$$\mathbf{k}_{1,2}^{\mathrm{I}} = \mathbf{n}_{1,2}^{\mathrm{I}}\omega_{1,2}/c$$

where $e_{1,2}^{\mathrm{I}}$ are supposed to be the three-dimensional vectors. Define the incidence plane of the wave E_1 as the XZ plane. Such a choice is possible since S_A possess the symmetry D_∞ [10].

It is known that in uniaxial medium as S_A is the EM wave in general case splits due to the anisotropy into the two EM waves: extraordinary and ordinary ones with the following dispersion relations [25, 32]:

$$\frac{(k_z^{\mathrm{e}})^2}{\varepsilon_\perp} + \frac{(k_x^{\mathrm{e}})^2 + (k_y^{\mathrm{e}})^2}{\varepsilon_\|} = \left(\frac{\omega}{c}\right)^2, \tag{22}$$

$$(k^{\mathrm{o}})^2 = \left(\frac{\omega}{c}\right)^2 \varepsilon_\perp, \tag{23}$$

where $\mathbf{k}^{\mathrm{e}}$, $\mathbf{k}^{\mathrm{o}}$ are the wavevectors of the extraordinary and of the ordinary waves, respectively.

In our case four fundamental EM waves would propagate in S_A: two ordinary EM waves and two extraordinary ones:

$$\mathbf{E}_1^{\mathrm{o,e}} = \mathbf{e}_1^{\mathrm{o,e}}\{A_1^{\mathrm{o,e}}(z) \exp i(\mathbf{k}_1^{\mathrm{o,e}}\mathbf{r} - \omega_1 t) + \text{c.c.}\} \tag{24}$$

and

$$\mathbf{E}_2^{\mathrm{o,e}} = \mathbf{e}_2^{\mathrm{o,e}}\{A_2^{\mathrm{o,e}}(z) \exp i(\mathbf{k}_2^{\mathrm{o,e}}\mathbf{r} - \omega_2 t) + \text{c.c.}\}. \tag{25}$$

Each pair of EM waves (24) and (25) is frequency degenerate. Hence we may define the mixing of these waves on the non-linearity as the partly degenerate in contrast to the degenerate FWM or FWM with four different frequencies [1].

Using the relationships (22) and (23) and the boundary conditions at $z=0$ [1, 25, 31, 32], we obtain

$$k_{1y}^{\mathrm{I}} = k_{1y}^{\mathrm{o}} = k_{1y}^{\mathrm{e}} = 0, \quad k_{1x}^{\mathrm{I}} = k_{1x}^{\mathrm{o}} = k_{1x}^{\mathrm{e}}, \tag{26}$$

$$k_{1z}^{\mathrm{o}} = \left[\left(\frac{\omega_1}{c}\right)^2 \varepsilon_\perp - (k_{1x}^{\mathrm{I}})^2\right]^{1/2}, \tag{27}$$

$$k_{1z}^{\mathrm{e}} = \left[\left(\frac{\omega_1}{c}\right)^2 - \frac{(k_{1x}^{\mathrm{I}})^2}{\varepsilon_\|}\right]^{1/2} \sqrt{\varepsilon_\perp}, \tag{28}$$

$$k_{2x,y}^{\mathrm{I}} = k_{2x,y}^{\mathrm{o,e}}, \tag{29}$$

$$k_{2z}^{\mathrm{o}} = \left[\left(\frac{\omega_2}{c}\right)^2 \varepsilon_\perp - (k_{2x}^{\mathrm{I}})^2 - (k_{2y}^{\mathrm{I}})^2\right]^{1/2} \tag{30}$$

and

$$k_{2z}^{\mathrm{e}} = \left[\left(\frac{\omega_2}{c}\right)^2 - \frac{(k_{2x}^{\mathrm{I}})^2 + (k_{2y}^{\mathrm{I}})^2}{\varepsilon_\|}\right]^{1/2} \sqrt{\varepsilon_\perp}. \tag{31}$$

Using the relationships (26)–(31) we obtain the possible polarizations of the fundamental modes (24), (25) [25, 32]:

$$\mathbf{e}_1^{\mathrm{o}} = (0, 1, 0), \tag{32}$$

$$\mathbf{e}_1^{\mathrm{e}} = (e_{1x}^{\mathrm{e}}, 0, e_{1z}^{\mathrm{e}}), \tag{33}$$

$$e_{1z}^{\mathrm{e}} = -e_{1x}^{\mathrm{e}}(k_{1x}^{\mathrm{I}}k_{1z}^{\mathrm{e}})\left[\left(\frac{\omega_1}{c}\right)^2 \varepsilon_\perp - (k_1^{\mathrm{e}})^2 - (k_{1x}^{\mathrm{I}})^2\right]^{-1}, \tag{34}$$

$$\mathbf{e}_2^{\mathrm{o}} = (e_{2x}^{\mathrm{o}}, e_{2y}^{\mathrm{o}}, 0), \tag{35}$$

$$e_{2x}^{\mathrm{o}} = -\left(\frac{k_{2y}^{\mathrm{I}}}{k_{2x}^{\mathrm{I}}}\right) e_{2y}^{\mathrm{o}}, \tag{36}$$

$$\mathbf{e}_2^{\mathrm{e}} = (e_{2x}^{\mathrm{e}}, e_{2y}^{\mathrm{e}}, e_{2z}^{\mathrm{e}}), \tag{37}$$

and

$$e_{2z}^{\mathrm{e}} = -k_{2z}^{\mathrm{e}}(e_{2x}^{\mathrm{e}}k_{2x}^{\mathrm{I}} + e_{2y}^{\mathrm{e}}k_{2y}^{\mathrm{I}})\left[\varepsilon_\|\left(\frac{\omega_2}{c}\right)^2 - (k_2^{\mathrm{e}})^2 + (k_{2z}^{\mathrm{e}\,2})\right]^{-1}. \tag{38}$$

It should be noted that both ordinary EM waves are transverse [25]:

$$\operatorname{div} \mathbf{E}_{1,2}^{\mathrm{o}} = 0. \tag{39}$$

Substituting equations (24) and (25) into the equation of motion (10), neglecting the decaying homogeneous solution and retaining in the right-hand side only time-averaged terms with the difference frequency

$$\Delta\omega = \omega_1 - \omega_2 \ll \omega_1 \tag{40}$$

we obtain the layer displacement $u(\mathbf{r}, t)$

$$u(\mathbf{r}, t) = \frac{i}{4\pi\rho}\sum_{j=1}^{4} U_j \exp i(\Delta\mathbf{k}_j\mathbf{r} - \Delta\omega t) + \text{c.c.} \tag{41}$$

where

$$U_j = \frac{\{(\Delta k_{j\perp})^2 h_j M_j\}}{\{(\Delta k_j)^2 G_j(\Delta\omega, \Delta\mathbf{k}_j)\}} \tag{42}$$

and

$$G_j = (\Delta\omega)^2 - \Omega_j^2 + i\Delta\omega\Gamma_j. \tag{43}$$

The explicit form of the magnitudes included into the relationships (42), (43) is sufficiently complicate, and we present these expressions in the Appendix A. For optical frequencies $\omega_{1,2}$ and typical values of the material parameters α_i, ρ, B [9, 10, 21, 22] Γ_j has a magnitude comparable with Ω_j. Substituting equation (41) into (14) we find

$$\varepsilon_{ik}^{\mathrm{N}} = -\frac{1}{4\pi\rho}\sum_{j=1}^{4} L_{ik}^{j} U_j \exp i(\Delta\mathbf{k}_j\mathbf{r} - \Delta\omega t) + \text{c.c.} \tag{44}$$

where

$$L^j = \begin{vmatrix} a_\perp \Delta k_{jz} & 0 & -\varepsilon_a \Delta k_{jx} \\ 0 & a_\perp \Delta k_{jz} & -\varepsilon_a \Delta k_{jy} \\ -\varepsilon_a \Delta k_{jx} & -\varepsilon_a \Delta k_{jy} & -a_\parallel \Delta k_{jz} \end{vmatrix}. \quad (45)$$

It is seen from the relationship (41) that the dynamic grating of the layer displacement consists of the four frequency degenerate harmonics unlike the two-wave mixing [8], or degenerate FWM [1,33]. Substituting equations (24), (25) and (44) into (13) we obtain the non-linear part of the electric induction which represents the superposition of the finite number of harmonics

$$\mathbf{D} = \sum_{l=1}^{32} (\mathbf{D}_l^{\mathrm{N}} \exp i\psi_l + \mathrm{c.c.}). \quad (46)$$

Comparing the expressions (24), (25) and (41)–(46) one may see that the amplitudes $\mathbf{D}_l^{\mathrm{N}}$ are essentially complex. Therefore the amplification of some fundamental waves by the other ones through the non-linear terms (46) phase-matched with the fundamental modes (24), (25) is possible (see [4]). We do not present the explicit form of $\mathbf{D}_l^{\mathrm{N}}$, since they are too complicated, however it is seen from the expressions (13)–(15), (24), (25), (41)–(46) that all $\mathbf{D}_l^{\mathrm{N}}$ are three-dimensional vectors and

$$|\mathbf{D}_l^{\mathrm{N}}| \sim \frac{1}{\rho s^2} |A_m A_n A_k| \quad (47)$$

where $A_{m,n,k}$ are the fundamental mode amplitudes $A_{1,2}^{\mathrm{o,e}}(z)$.

The sum (46) contains the two kinds of terms: (*i*) four harmonics which are phase-matched with the fundamental modes (24), (25), and (*ii*) all other terms with combination frequencies and wavevectors.

Consequently, the non-linear polarization (46) gives rise to the three kinds of the essentially non-linear optical effects. (*i*) The parametric coupling of $\mathbf{E}_{1,2}^{\mathrm{o,e}}$ and amplification are determined by the components of the phase-matched terms of the non-linear polarization (46) which are parallel to the field of the fundamental modes (24), (25). (*ii*) The normal to $\mathbf{e}_{1,2}^{\mathrm{o,e}}$ components of the non-linear terms mentioned above generate the additional components of the fundamental modes. As a result, all fundamental modes become the three-dimensional vectors, while originally $\mathbf{E}_1^{\mathrm{e}}$, $\mathbf{E}_2^{\mathrm{o}}$ were the two-dimensional vectors. (*iii*) The non-linear terms in (46) with combination frequencies and wavevectors create the scattered harmonics (Brillouin-like scattering).

Substituting equations (12), (18), (24), (25), (46) into the wave equation (11), taking into account the dispersion relations (22) and (23), conditions (19), (20) and equating the terms with the same phases we obtain three sets of equations describing the effects mentioned above. (*i*) The reduced equations for the slowly varying amplitudes $A_{1,2}^{\mathrm{o,e}}(z)$. We do not limit the analysis of the parametric coupling to the constant pumping intensity approximation [1], often used [3, 4, 7] and consider the general case taking into account the depletion of pumping waves and the saturation of amplification. (*ii*) The wave equations for the additional components. The extraordinary wave $\mathbf{E}_1^{\mathrm{e}}$ fails to satisfy the condition (39) and propagates in the main cross-section [25] according to equation (26) in such a way that

$$\partial D_y(\mathbf{k}_1^{\mathrm{e}}, \omega_1)/\partial y \equiv 0.$$

The additional components of the ordinary waves $\mathbf{E}_{1,2}^{\mathrm{o}}$ are not longitudinal because of the anisotropy of S_A. Therefore it is more convenient to derive all these additional components directly from the corresponding wave equations instead of using the equation

$$\operatorname{div} \mathbf{D} = 0 \quad (48)$$

along with the relationships (12), (13), (39) according to the known procedure [1]. (*iii*) The wave equations for the Brillouin-like scattered harmonics. The non-linear analysis presented in this paper is based on the assumption that the non-linearity is weak. The criterion of smallness of the non-linearity may be obtained combining the relationships (17), (41)–(43) and (A 1)–(A 6). It has the form

$$\frac{\varepsilon_a (\Delta k_j)^2}{\rho} \frac{|A_i A_k|}{|G_j|} \ll 1. \quad (49)$$

This inequality is valid for any applicable pumping intensity taking into account the magnitude of B [12, 21, 22].

3. The parametric amplification of the coupled fundamental EM waves

Separating the equations for the fundamental modes $\mathbf{E}_{1,2}^{\mathrm{o,e}}$, multiplying each equation by the corresponding polarization vector $\mathbf{e}_1^{\mathrm{o,e}}, 2$ (32), (33), (35), (37), defining

$$A_{1,2}^{\mathrm{o,e}} = |A_{1,2}^{\mathrm{o,e}}| \exp i\gamma_{1,2}^{\mathrm{o,e}} \quad (50)$$

and taking into account the relationship (43) we obtain the reduced equations for the amplitude moduli $|A_{1,2}^{\mathrm{o,e}}|$ and the phases $\gamma_{1,2}^{\mathrm{o,e}}$:

$$\begin{aligned} 2l_1^{\mathrm{o,e}} \frac{\partial |A_1^{\mathrm{o,e}}|}{\partial z} = & -\left(\frac{\omega_1}{c}\right)^2 \frac{\Delta\omega}{4\omega\rho} \left\{ \frac{h_{3,2}^2}{|G_{3,2}|^2} \left(\frac{\Delta k_{3,2\perp}}{\Delta k_{3,2}}\right)^2 \right. \\ & \times |A_2^{\mathrm{o,e}}|^2 \Gamma_{3,2} + \frac{h_{4,1}^2}{|G_{4,1}|^2} \left(\frac{\Delta k_{4,1\perp}}{\Delta k_{4,1}}\right)^2 \\ & \left. \times |A_2^{\mathrm{e,o}}|^2 \Gamma_{4,1} \right\} |A_1^{\mathrm{o,e}}|, \end{aligned} \quad (51)$$

$$2l_2^{o,e}\frac{\partial|A_2^{o,e}|}{\partial z}=\left(\frac{\omega_2}{c}\right)^2\frac{\Delta\omega}{4\omega\rho}\left\{\frac{h_{3,2}^2}{|G_{3,2}|^2}\left(\frac{\Delta k_{3,2\perp}}{\Delta k_{3,2}}\right)^2\right.$$
$$\times|A_1^{o,e}|^2\Gamma_{3,2}+\frac{h_{1,4}^2}{|G_{1,4}|^2}\left(\frac{\Delta k_{1,4\perp}}{\Delta k_{1,4}}\right)^2$$
$$\left.\times|A_1^{e,o}|^2\Gamma_{1,4}\right\}|A_2^{o,e}|, \qquad (52)$$

$$2l_1^{o,e}\frac{\partial\gamma_1^{o,e}}{\partial z}=-\left(\frac{\omega_1}{c}\right)^2\frac{1}{4\omega\rho}\left\{\frac{h_{3,2}^2}{|G_{3,2}|^2}\left(\frac{\Delta k_{3,2\perp}}{\Delta k_{3,2}}\right)^2|A_2^{o,e}|^2\right.$$
$$\times[(\Delta\omega)^2-\Omega_{3,2}^2]+\frac{h_{4,1}^2}{|G_{4,1}|^2}\left(\frac{\Delta k_{4,2\perp}}{\Delta k_{4,1}}\right)^2$$
$$\left.\times|A_2^{e,o}|^2[(\Delta\omega)^2-\Omega_{4,1}^2]\right\}, \qquad (53)$$

$$2l_1^{o,e}\frac{\partial\gamma_1^{o,e}}{\partial z}=-\left(\frac{\omega_2}{c}\right)^2\frac{1}{4\omega\rho}\left\{\frac{h_{3,2}^2}{|G_{3,2}|^2}\left(\frac{\Delta k_{3,2\perp}}{\Delta k_{3,2}}\right)^2|A_2^{o,e}|^2\right.$$
$$\times[(\Delta\omega)^2-\Omega_{3,2}^2]+\frac{h_{1,4}^2}{|G_{1,4}|^2}\left(\frac{\Delta k_{1,4\perp}}{\Delta k_{1,4}}\right)^2|A_1^{e,o}|^2$$
$$\left.\times[(\Delta\omega)^2-\Omega_{1,4}^2]\right\} \qquad (54)$$

where

$$l_{1,2}^{o}=k_{1,2z}^{o},\quad l_{1,2}^{e}=k_{1,2z}^{e}\left[1-\frac{(\mathbf{k}_{1,2}^{e}\mathbf{e}_{1,2}^{e})}{k_{1,2z}^{e}}e_{1,2z}^{e}\right]. \qquad (55)$$

The equations (51)–(54) describe the parametric energy transfer between the pairs of the EM waves with the different frequencies and the phase cross-modulation [4]. The solution of these equations with the appropriate boundary conditions at $z=0$ and $z\to\infty$ provides a full description of the spatial distribution of the amplitude moduli and phases of the four fundamental EM waves in S_A. The nonlinear susceptibility is complex as it is seen from equations (41)–(44), and the energy transfer is determined by the imaginary part of the Fourier transform of SS Green function $G_j(\Delta\omega,\Delta\mathbf{k}_j)$ according to the equations (43), (51) and (52). The energy transfer between the ordinary and extraordinary EM waves with the same frequency is impossible. The light absorption by the dynamic grating of layer deformations may be neglected, since the SS eigenmode relaxation time Γ_j^{-1} determined by the viscosity of S_A is small in comparison with the time required for SS transition through the EM waves interaction region. In such an approximation the coupled system of the EM waves and of the dynamic grating may be characterized as quasi-dissipative. The dynamic grating (41) turns out to be a sort of a channel providing a total energy transfer without losses between the coupled EM modes. This system possesses only one integral of motion which may be easily obtained from the equations (51) and (52). It has the form

$$\left(\frac{\omega_1}{c}\right)^{-2}[l_1^{o}|A_1^{o}|^2+l_1^{e}|A_1^{e}|^2]$$
$$+\left(\frac{\omega_2}{c}\right)^{-2}[l_2^{o}|A_2^{o}|^2+l_2^{e}|A_2^{e}|^2]=I_o=\text{const.} \qquad (56)$$

Introducing the dimensionless variables

$$w_{1,2}^{o,e}=\frac{1}{I_o}\left(\frac{\omega_{1,2}}{c}\right)^{-2}l_{1,2}^{o,e}|A_{1,2}^{o,e}|^2 \qquad (57)$$

and substituting them into equation (56) we obtain

$$w_1^{o}+w_1^{e}+w_2^{o}+w_2^{e}=1. \qquad (58)$$

Substituting equation (57) into the equations (51)–(54) we obtain

$$-\frac{\partial}{\partial z}(\ln w_1^{o,e})=\beta_{3,2}w_2^{o,e}+\beta_{4,1}w_2^{e,o}, \qquad (59)$$

$$\frac{\partial}{\partial z}(\ln w_2^{o,e})=\beta_{3,2}w_1^{o,e}+\beta_{1,4}w_1^{e,o}, \qquad (60)$$

$$\frac{\partial\gamma_1^{o,e}}{\partial z}=-\tfrac{1}{2}(\delta_{3,2}w_2^{o,e}+\delta_{4,1}w_2^{e,o}), \qquad (61)$$

and

$$\frac{\partial\gamma^{o,e}}{\partial z}=-\tfrac{1}{2}(\delta_{3,2}w_1^{o,e}+\delta_{1,4}w_1^{e,o}), \qquad (62)$$

where the coupling constants β_j, δ_j have the form

$$\beta_j=C_j\Delta\omega\Gamma_j,\quad \delta_j=C_j[(\Delta\omega)^2-\Omega_j^2],$$
$$C_j=\left[\frac{\omega_1\omega_2}{c^2}\right]^2\frac{I_0}{4\pi\rho}\frac{h_j^2}{|G_j|^2d_j}\left(\frac{\Delta k_j\perp}{\Delta k_j}\right)^2$$

and

$$d_1=l_1^{e}l_2^{o},\quad d_2=l_1^{e}l_2^{e},\quad d_3=l_1^{o}l_2^{o},\quad d_4=l_1^{o}l_2^{e}. \qquad (63)$$

The calculations of the explicit solutions of the system (59)–(62) in our general case is hardly possible, and we investigate the qualitative behaviour of the intensities $w_{1,2}^{o,e}$ and of the phases $\gamma_{1,2}^{o,e}$. Without the loss of generality we choose $\omega_1>\omega_2$.

Then according to equations (63)

$$\beta_j>0 \qquad (64)$$

and therefore the expressions in the right-hand sides of relations (50) and (60) are positive definite. Hence

$$\frac{\partial w_1^{o,e}}{\partial z}<0,\quad \frac{\partial w_2^{o,e}}{\partial z}>0,\quad z>0, \qquad (65)$$

and it is seen that the intensities of the fundamental EM waves $\mathbf{E}_1^{o,e}$ are monotonically decreasing, while the intensities of the EM waves $\mathbf{E}_2^{o,e}$ with the lower frequency ω_2 are monotonically increasing with z. According to the conservation law (58) all amplitudes $w_{1,2}^{o,e}$ are finite at $z\to\infty$.

The system (59)–(62) has the formal solution in the integral form

$$w_1^{o,e} = w_1^{o,e}(0)\exp\left\{-\int_0^z(\beta_{3,2}w_2^{o,e} + \beta_{4,1}w_2^{e,o})\,dz\right\}, \quad (66)$$

$$w_2^{o,e} = w_2^{o,e}(0)\exp\left\{\int_0^z(\beta_{3,2}w_1^{o,e} + \beta_{1,4}w_1^{e,o})\,dz\right\}, \quad (67)$$

$$\gamma_1^{o,e} - \gamma_1^{o,e}(0) = \int_0^z -\tfrac{1}{2}(\delta_{3,2}w_2^{o,e} + \delta_{4,1}w_2^{e,o})\,dz \quad (68)$$

and

$$\gamma_2^{o,e} - \gamma_2^{o,e}(0) = \int_0^z -\tfrac{1}{2}(\delta_{3,2}w_1^{o,e} + \delta_{1,4}w_1^{e,o})\,dz. \quad (69)$$

Taking into account equations (58) and (65) we obtain from equations (66) and (67)

$$z \to \infty, \quad w_1^{o,e} \to 0, \quad w_2^e + w_2^o \to 1. \quad (70)$$

The relationships (65) and (70) show that the energy transfer from the pair of EM waves with the greater frequency ω_1 to the pair of EM waves with the lower frequency ω_2 occurs. This process is non-reciprocal, since the medium is quasi-dissipative.

In the constant pumping intensity approximation [1], $w_1^{o,e}$ are supposed to be constant, and $w_1^{o,e} \gg w_2^{o,e}$.

In this case the integrands in the expression (67) are constant, and $\mathbf{E}_2^{o,e}$ would increase infinitely with the gain coefficient determined by the imaginary part of the non-linear susceptibility [24]. In other words, in the constant pumping intensity approximation the terms responsible for the amplification are constant and represent the field-induced complex spatial dispersion [25, 34]. In general case when the pumping depletion is taken into account we pass from the linearized theory to the essentially non-linear one, and the mentioned terms become dependent on coordinates. Therefore the terms responsible for the dispersion and for the non-linearity are identical, and the process of the parametric amplification would be stable giving rise to spatially localized states without any threshold [35, 36, 34].

The influence of the phase cross-modulation on the parametric process is determined by the ratio of the coupling constants β_j and δ_j. According to the condition (64) for $\omega_1 > \omega_2$ β_j are positive definite while δ_j may be positive as well as negative. Four essentially different cases of the interplay of the phase cross-modulation and the parametric amplification may occur.

(*i*) $\Delta\omega \gg \Omega_j$

Then

$$\delta_j > 0, \quad \beta_j/\delta_j \approx \Gamma_j/\Delta\omega \ll 1$$

and the phase cross-modulation process dominates. The phase-shifts are negative which corresponds to the defocusing condition [1]. The amplification at the same distance is weak.

$$(ii) \qquad \Delta\omega \gg \Omega_j \quad \delta_j < 0, \quad |\beta_j/\delta_j| \sim \frac{\Delta\omega\Gamma_j}{\Omega_j^2} \ll 1.$$

In this case the phase cross-modulation prevails, too, but the phase-shifts (68) and (69) are positive which corresponds to the focusing condition.

$$(iii) \qquad \Delta\Omega \sim (s/c)\omega_1 \sim \Omega_j, \quad j = (1\text{–}4),$$

$$[(\Delta\omega^2) - \Omega_j^2]/(\Delta\omega)^2 \ll 1.$$

In this case $\beta_j \gg |\delta_j|$, $j = (1\text{–}4)$. The process of the parametric amplification is predominant while the phase-shifts would be comparatively small.

(*iv*) The exact resonance of the one of the SS harmonics u_j: $(\Delta\omega)^2 = \Omega_j^2$.

Let, for example, the resonant coupling occurs between the wave $\mathbf{E}_1^e$ and the wave $\mathbf{E}_2^o$. The resonant condition in this case has the form

$$(\Delta\omega)^2 = \Omega_1^2 = s^2\frac{(\Delta k_{1\perp})^2(\Delta k_{1z})^2}{(\Delta k_1)^2}. \quad (71)$$

Substituting the dispersion relations (22) and (23) into the equation (71) and using the relationships (26) and (28)–(30) we obtain

$$\left(\frac{\Delta\omega}{\omega_1}\right)^2 = \left(\frac{s}{c}\right)^2 \varepsilon_\perp\{(n_{1x}^I - n_{2x}^I)^2 + (n_{2y}^I)^2\}\left\{\left[1 - \frac{(n_{1x}^I)^2}{\varepsilon_\parallel}\right]^{1/2} - \left[1 - \frac{(n_{1\perp}^I)^2}{\varepsilon_\perp}\right]^{1/2}\right\}^2\left\{(n_{1x}^I - n_{2x}^I)^2 + (n_{2y}^I)^2 + \varepsilon_\perp\left[\left(1 - \frac{(n_{1x}^I)^2}{\varepsilon_\parallel}\right)^{1/2} - \left(1 - \frac{(n_{2\perp}^I)^2}{\varepsilon_\perp}\right)^{1/2}\right]^2\right\}^{-1} \quad (72)$$

where the small terms of higher orders $O(\Delta\omega/\omega_1)$ are neglected. It is clear that the equation (71) may be satisfied only for $\Delta\omega/\omega_1 \sim s/c$. The relationship (72) represents in the implicit form the limitations on the propagation directions of the coupled EM waves which is specific for the SS excitation. Comparing the expressions (43) and (63) it is seen that the resonant coupling constant $\beta_1^r \gg \beta_j$ $(j \neq 1)$ while $\delta_1^r = 0$.

The phase shifts for the resonantly coupled waves are much less than for other pairs.

The interaction of the non-resonant pairs of waves may be considered as perturbation. It is clear that the third and the fourth cases are the most favourable for the strong coupling since the energy transfer occurs at the almost constant phases.

The behaviour of the grating harmonics amplitudes (42) is determined by the field-dependent factors M_j. Substituting equations (57), (66) and (67) into (A 6) we obtain

$$|M_j| = I_0 \frac{\omega_1\omega_2}{c^2} \cdot (d_j)^{-1/2}(m_j)^{1/2} \exp\left(\frac{1}{2}\int_0^z I_j dz\right) \quad (73)$$

where

$$m_1 = w_1^e(0)w_2^o(0), \quad m_2 = w_1^e(0)w_2^e(0), \quad m_3 = w_2^o(0)w_2^o(0),$$
$$m_4 = w_1^o(0)w_2^e(0), \quad (74)$$

$$\left.\begin{aligned} I_1 &= \beta_3 w_1^o + \beta_1 w_1^e - \beta_2 w_2^e - \beta_1 w_2^o \\ I_2 &= \beta_2 w_1^e + \beta_4 w_1^o - \beta_2 w_2^e - \beta_1 w_2^o \\ I_3 &= \beta_3 w_1^o + \beta_1 w_1^e - \beta_3 w_2^o - \beta_4 w_2^e \\ I_4 &= \beta_2 w_1^e + \beta_4 w_1^o - \beta_3 w_2^o - \beta_4 w_2^e \end{aligned}\right\} \quad (75)$$

Comparing the relationships (57), (58), (70) and (73)–(75) we see that at $z = 0$ the dynamic grating is determined by the input intensity of the coupled fundamental EM waves while at $z \to \infty$ all harmonics (41) vanish since $z \to \infty$, $|M_j| \to 0$. The condition of maximum for each grating harmonic amplitude has the form

$$\left.\begin{aligned} &\frac{\partial |M_j|}{\partial z} = 0, \quad z = z_{oj} > 0 \\ &\frac{\partial |M_j|}{\partial z} > 0, \quad z_{oj} > z > 0 \\ \text{and} \quad &\frac{\partial |M_j|}{\partial z} < 0, \quad z > z_{oj} \end{aligned}\right\} \quad (76)$$

The solution of relations (76) exists, if at least $I_j(0) > 0$. The latter condition may be satisfied for all harmonics, if

$$w_1^{o,e}(0) > w_2^{o,e}(0). \quad (77)$$

Therefore the amplitudes of the layer displacement grating are spatially localized, and four harmonics (41) are travelling along the directions $\Delta \mathbf{k}_j$ obliquely to the layers which is characteristic for SS in S_A.

Consider the practically important case when each of the incident EM waves has mainly one kind of polarization and only their small components have the other polarization. Let, for example, the pumping wave $\mathbf{E}_1^I$ is polarized mainly in the incidence plane while the signal wave $\mathbf{E}_2^I$ is mainly polarized normal to its incidence plane:

$$w_1^e \gg w_1^o, \; w_2^o \gg w_2^e. \quad (78)$$

In this case the system (59)–(60) splits into the following equations:

$$\frac{\partial w_1^e}{\partial z} = -\beta_1 w_1^e w_2^o, \quad \frac{\partial w_2^o}{\partial z} = \beta_1 w_1^e w_2^o, \quad (79a)$$

and

$$-\frac{\partial w_1^o}{\partial z} = \beta_3 w_2^o w_1^o, \quad \frac{\partial w_2^e}{\partial z} = \beta_2 w_1^e w_2^e. \quad (79b)$$

The first two equations (79 a) describe the two-wave mixing [8] and may be solved separately. These two equations have an integral of motion

$$J_1 = w_1^e(0) + w_2^o(0) \quad (80)$$

and the solution

$$w_1^e = \tfrac{1}{2}J_1\{1 - \tanh(\eta - \eta_0)\} \quad (81a)$$

and

$$w_2^o = \tfrac{1}{2}J_1\{1 + \tanh(\eta - \eta_0)\} \quad (81b)$$

where

$$\eta = \tfrac{1}{2}\beta_1 J_1 z, \quad z_0 = \frac{1}{\beta_1 J_1} \ln\{w_1^e(0)/w_2^o(0)\}.$$

The intensities w_1^e and w_2^o have a form of a spatial kink and an anti-kink [35, 36], respectively. Their crossing-point $z_0 > 0$ exists, if

$$w_1^e(0)/w_2^o(0) > 1 \quad (82)$$

The solutions (81) for the different pumping-signal ratios (82) are represented in figure 1.

Substituting (81) into the pair of equations (79 b) we obtain the expressions for the small components

$$w_1^o = w_1^o(0)\{\cosh(\eta_0)\exp(-\eta)/\cosh(\eta - \eta_0)\}^{\beta_3/\beta_1} \quad (83)$$

and

$$w_2^e = w_2^e(0)\{\cosh(\eta_0)\exp(\eta)/\cosh(\eta - \eta_0)\}^{\beta_2/\beta_1} \quad (84)$$

It is seen from the relationships (83) and (84) that the ordinary component of the pumping wave vanishes as η tends to infinity, while the extraordinary component of the signal wave, on the contrary, increases and reaches the saturation at sufficiently large $\eta \gg \eta_0$:

$$\eta \to \infty, \; w_1^o \to 0, \quad (w_1^o)_{max} = w_1^o(0) \quad (85)$$

and

$$\eta \to \infty, \quad w_2^e \to w_2^e(0)\{1 + w_1^e(0)/w_2^o(0)\}^{\beta_2/\beta_1} > w_2^e(\eta). \quad (86)$$

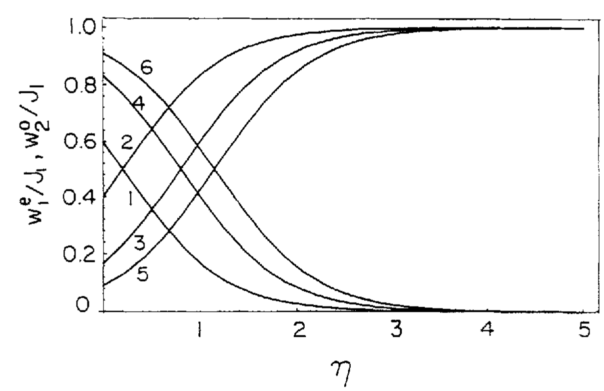
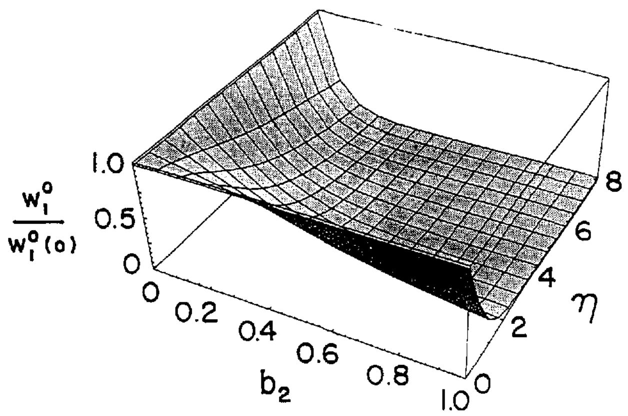
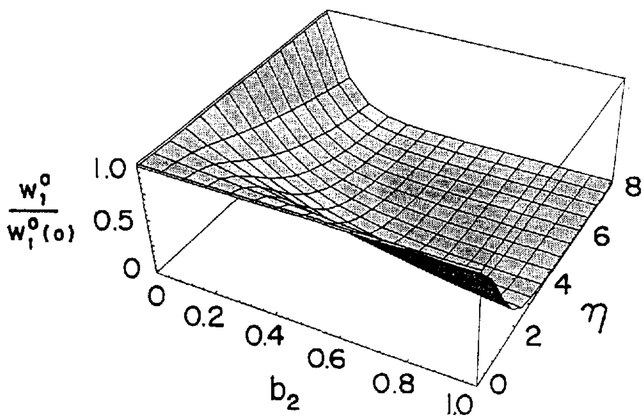


Figure 1. The dependence of the reduced pumping intensity (w_1^e/J_1) and the reduced signal intensity (w_2^o/J_1) on the dimensionless coordinate η for the pumping-signal ratio $(w_1^e(0)/w_2^o(0)) = 1{\cdot}5$ (curves 1, 2), $(w_1^e(0)/w_2^o(0)) = 5$ (curves 3, 4), $(w_1^e(0)/w_2^o(0)) = 10$ (curves 5, 6).



(*a*)



(*b*)

Figure 2. The dependence of the reduced intensity $(w_1^o/w_1^o(0))$ of the ordinary pumping wave on the dimensionless coordinate η and on $0 \leqslant b_2 \leqslant 1$: (*a*) $\eta_0 = 0{\cdot}2$ $((w_1^e(0)/w_2^o(0)) = 1{\cdot}5)$, (*b*) $\eta_0 = 1$ $((w_1^e(0)/w_2^o(0)) = e^2)$.

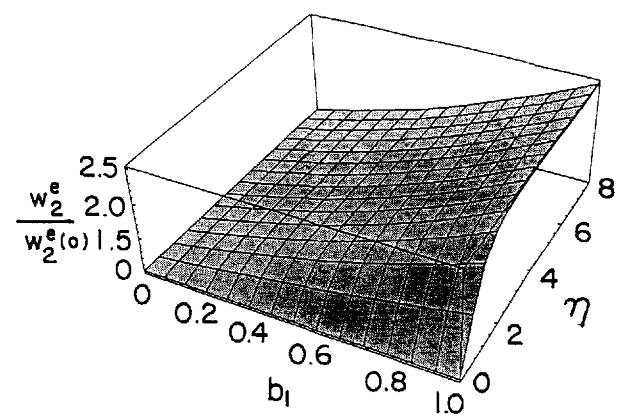
The numerical estimations show that both saturation and pumping depletion occur at $\eta \sim (4\text{–}5)$. Comparing the expressions (63), (83) and (84), we may see that far from the resonance when all β_j are approximately equal both components (83), (84) are changing with an approximately same rate. In the opposite case, near the resonance $\beta_{2,3} \ll \beta_1$, and the small components are kept almost constant since they are weakly coupled with the main waves. The function (83) and (84) with the different values of the exponents $b_{1,2} = \beta_{2,3}/\beta_1$ are presented in figures 2 and 3, respectively.

Consider finally the behaviour of the phases. In general case the phase evolution of each wave is rather complicate. Each phase evolves independently from other ones because the cubic susceptibility is complex. We investigate two limit cases: first, when the coupling is far from the resonance, and secondly, the resonant coupling. Substituting the relationships (81), (83) and (84) into the equations (68) and (69) and assuming that far from the resonance $b_{1,2} = \beta_{2,3}/\beta_1 \approx 1$, we obtain that the phases $\gamma_1^{o,e}$ of the pumping waves $\mathbf{E}_1^{o,e}$ rapidly increase with η tending to $\pm\infty$ for $(\Delta\omega)^2 < \Omega_{1,3}^2$ or $(\Delta\omega)^2 < \Omega_{1,3}^2$, respectively:

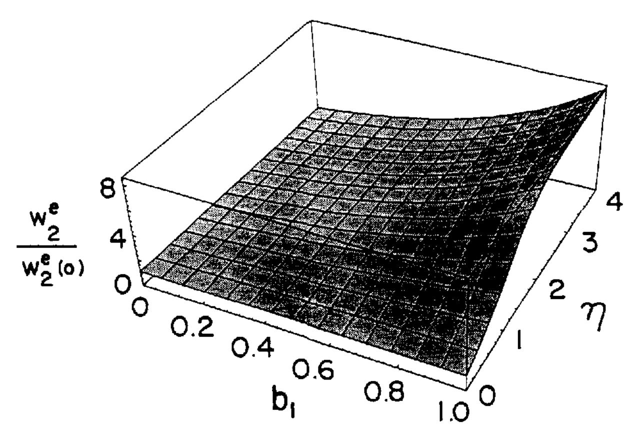
$$\gamma_1^{o,e} - \gamma_1^{o,e}(0) \sim \frac{\delta_{1,3}}{2\beta_1} \ln\{\cosh(\eta_0) \times \exp(-\eta)/\cosh(\eta - \eta_0)\}. \quad (87)$$

That means that the depletion of the pumping waves is accompanied by the oscillations of the corresponding amplitudes (50). The phases $\gamma_2^{o,e}$ of the signal waves $\mathbf{E}_2^{o,e}$ reach the constant values at sufficiently large $\eta \gg \eta_0$:

$$\eta \to \infty, \quad \{\gamma_2^{o,e} - \gamma_2^{o,e}(0)\} \to -\frac{\delta_{1,2}}{2\beta_1} \ln\{1 + w_1^e(0)/w_2^o(0)\}. \quad (88)$$



(*a*)



(*b*)

Figure 3. The dependence of the reduced intensity $(w_2^e/w_2^e(0))$ of the extraordinary signal wave on the dimensionless coordinate η and on $0 \leqslant b_1 \leqslant 1$: (*a*) $\eta_0 = 0{\cdot}2$ $((w_1^e(0)/w_2^o(0)) = 1{\cdot}5)$, (*b*) $\eta_0 = 1$ $((w_1^e(0)/w_2^o(0)) = e^2)$.

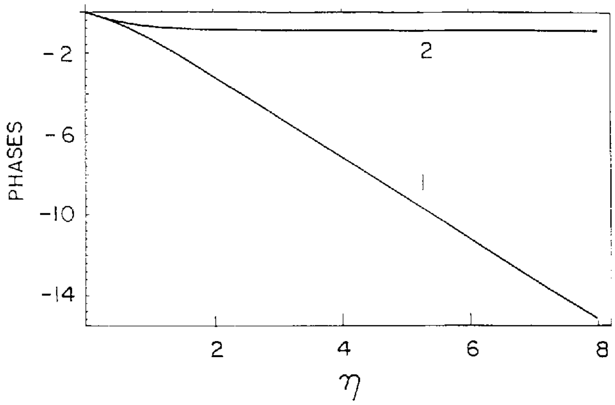


Figure 4. The distribution of the reduced phases: curve $1-(2\beta_1/\delta_{1,3})[\gamma_1^{o,e}-\gamma_1^{o,e}(0)]$, curve $2-(2\beta_1/\delta_{1,2})[\gamma_2^{o,e}-\gamma_2^{o,e}(0)]$.

The evolution of the phases (87) and (88) is shown in the figure 4.

The medium behaves as focusing or as defocusing one in respect to the amplified waves depending on the sign of $\delta_{1,2}$. The case when

$$(\Delta\omega)^2<\Omega_{1,2}^2$$

and both phase shifts (88) are positive is the most favourable since the signal waves are at the same time parametrically amplified and focused.

In the resonant case the main terms in the phases of the fundamental coupled waves $\mathbf{E}_1^e$ and $\mathbf{E}_2^o$ vanish, since $\delta_1^r=0$. Near the resonance we may also suppose the integrands in (68) and (69) to be constant: $w_1^o\approx w_1^o(0)$, $w_2^e\approx w_2^e(0)$. The phases of the waves $\mathbf{E}_1^e$ and $\mathbf{E}_2^e$ have, consequently, the form which is analogous to a phase shift of a plane wave:

$$\gamma_1^e-\gamma^e(0)\approx-\frac{\delta_2}{2}w_2^e(0)z,\quad \gamma_2^o-\gamma_2^o(0)\approx-\frac{\delta_3}{2}w_1^o(0)z. \quad (89)$$

The phase shifts on the interaction interval $2z_0$ are small:

$$\left.\begin{aligned}\Delta\gamma_1^e&\sim\left[\frac{w_2^e(0)|\delta_2|}{J_1\beta_1}\right]\ln\{w_1^e(0)/w_2^o(0)\}\ll 1\\ \Delta\gamma_2^o&\sim\left[\frac{w_1^o(0)|\delta_3|}{J_1\beta_1}\right]\ln\{w_1^e(0)/w_2^o(0)\}\ll 1.\end{aligned}\right\}\quad (90)$$

The resonant energy transfer occurs at the almost constant phases.

The results obtained permit to calculate explicitly the layer displacement $u(\mathbf{r},t)$. Combining the relationship (A 6), (57), (81 a,b) and (84) we find the amplitude factors of the dynamic grating

$$|M_1|=M_0\frac{I}{2\sqrt{d_1}}J_1\operatorname{sech}(\eta-\eta_0),$$

$$|M_2|=M_0\frac{I}{\sqrt{(2d_2)}}\{J_1w_2^e(0)\}^{1/2}\exp(\tfrac{1}{2}\eta_0)\times\{\cosh(\eta_0)\}^{1/2b_1}\exp\left\{-\frac{\eta}{2}(1-b_1)\right\}\times\{\operatorname{sech}\eta-\eta_0)\}^{1/2(1+b_1)},$$

$$|M_3|=M_0\frac{I}{\sqrt{(2d_3)}}\{J_1w_1^o(0)\}^{1/2}\exp(-\tfrac{1}{2}\eta_0)\times\{\cosh(\eta_0)\}^{1/2b_2}\exp\left\{\frac{\eta}{2}(1-b_2)\right\}\times\{\operatorname{sech}(\eta-\eta_0)\}^{1/2(1+b_2)},$$

and

$$|M_4|=M_0\frac{I}{\sqrt{d_4}}\{w_2^e(0)w_2^e(0)\}^{1/2}\{\cosh(\eta_0)\}^{1/2(b_1+b_2)}\times\exp\{\tfrac{1}{2}\eta(b_2-b_1)\}\{\operatorname{sech}(\eta-\eta_0)\}^{1/2(b_1+b_2)},$$

where $M_0=I_0\omega_1\omega_2/c^2$ and $\eta\to\infty$, $|M_j|\to 0$.

It is seen from the latter expressions that the profile $|M_1|$ of the harmonic excited by the EM waves with the strongest coupling has a spatially localized distribution with the centre at $\eta=\eta_0$. All other profiles have displaced distribution in respect to η_0. The smallest amplitude $|M_4|$ may decay rapidly or slowly which depends on the sign of the difference $(\beta_3-\beta_2)$. The first case corresponds to $\beta_3<\beta_2$, while the second case occurs when $\beta_3>\beta_2$. The profiles $|M_j|$ are shown in figures 5 and 6.

Let us evaluate numerically the coupling constant β_1^r in the resonant case which plays the role of the gain coefficient for the EM wave $\mathbf{E}_2^o$ according to (81 b). Taking into account the relations (26)–(31), (43), (55), (56), (63), (A 1), (A 2), (A 7) and (A 8) we obtain the gain per unit intensity

$$\frac{\beta_1^r}{\mathscr{P}}\sim\frac{\varepsilon_a^2}{c\sqrt{(\varepsilon_\perp)}\alpha_i s\cos\phi_2^o}\left(\frac{\Delta k_{1\perp}}{\Delta k_1}\right)^2$$

where ϕ_2^o is the angle between $\mathbf{k}_2^o$ and the Z axis, and the pumping intensity $\mathscr{P}$ has the form [25] $\mathscr{P}=|A_1^e|^2c/4\pi$.

It is seen that in the resonant case the gain coefficient does not depend on the EM wave frequency and strongly depends on the propagation directions of the amplified EM wave $\mathbf{E}_2^o$ and SS wave. The strongest amplification occurs when both $\mathbf{E}_2^o$ and the SS wave propagate at small angles with the plane of smectic layer. Using typical values of the material parameters $\alpha_i=1$ Poise [10, 21, 22], $s\sim 10^4\,\mathrm{cm\,s^{-1}}$ [12, 21, 22], $\varepsilon_a=0{\cdot}6$, $\varepsilon_\perp\sim 2$ [7], we obtain that for different ϕ_2^o and the SS wave propagation directions

$$\frac{\beta_1^r}{\mathscr{P}}\sim(0{\cdot}01\text{–}10)\,\mathrm{cm\,MW^{-1}}.$$

This value of $\beta_1^r/\mathscr{P}$ is up to three orders of magnitude greater than a gain coefficient for the ordinary stimulated

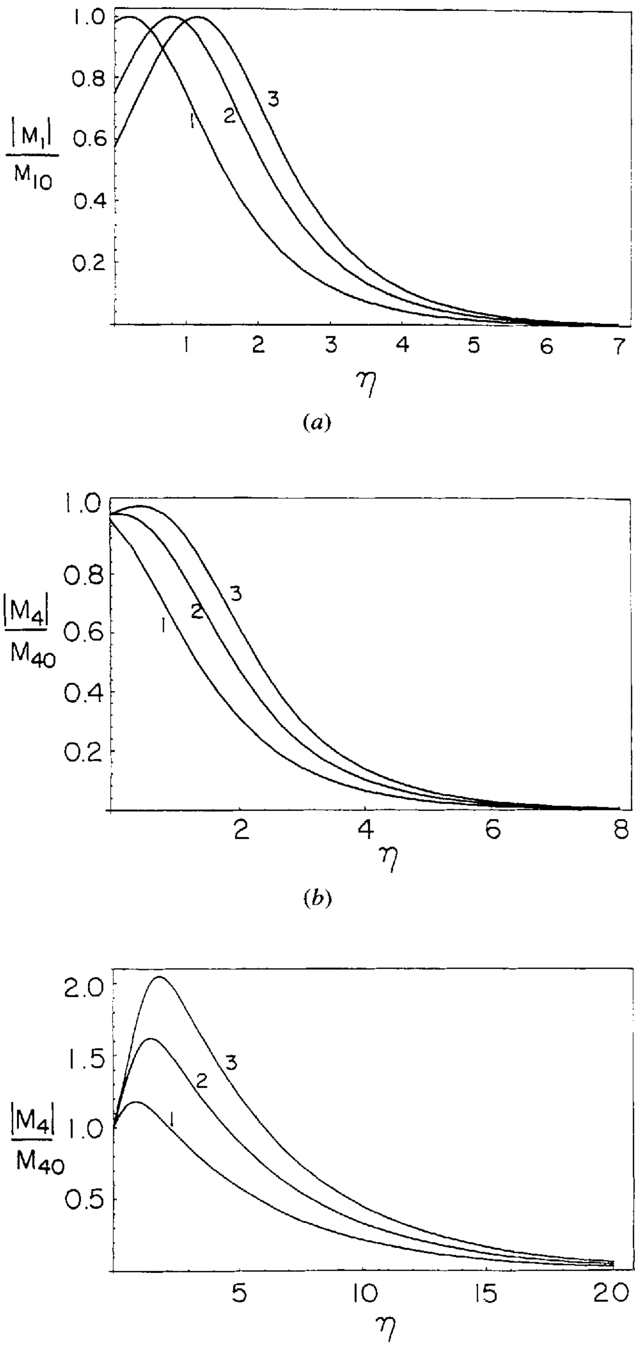


Figure 5. The dependence of the reduced moduli ($|M_1|/M_{10}$) and ($|M_4|/M_{40}$) of the dynamic grating amplitudes on the dimensionless coordinate η for the pumping-signal ratio: $(w_1^e(0)/w_2^o(0)) = 1{\cdot}5$ (curve 1), $(w_1^e(0)/w_2^o(0)) = 5$ (curve 2), $(w_1^e(0)/w_2^o(0)) = 10$ (curve 3): (*a*) ($|M_1|/M_{10}$), (*b*) ($|M_4|/M_{40}$) with rapid decay, (*c*) ($|M_4|/M_{40}$) with slow decay.

Brillouin scattering in isotropic organic liquids [1]. For the high input intensity $\mathscr{P} \sim 100\,\mathrm{MW\,cm^{-2}}$ we obtain the gain coefficient $\beta_1^r \sim (1\text{–}10^3)\,\mathrm{cm^{-1}}$, and the length of the interaction interval L would be $L \sim 2/\beta_1^r \sim (2 \times 10^{-3}\text{–}2)\,\mathrm{cm}$.

This estimation is in agreement with the experimental results [37].

4. Analysis of the stability

For the analysis of the stability of the solutions (81 *a, b*) we expand the intensities (57) into a series:

$$w_1^e = w_{10}^e + w_{11}^e + \dots, \; |w_{11}^e| \ll w_{10}^e \qquad (91\,a)$$

and

$$w_2^o = w_{20}^o + w_{21}^o + \dots, \; |w_{21}^o| \ll w_{20}^o \qquad (91\,b)$$

Substituting the expansions (91 *a, b*) into the system of equations (59) and (60) and taking into account the relationships (78) and (79 *a, b*) we obtain

$$\frac{\partial w_{11}^e}{\partial z} = \beta_1(w_{20}^o w_{11}^e + w_{10}^e w_{21}^o) + \beta_2 w_2^e w_{10}^e, \qquad (92)$$

$$\frac{\partial w_{11}^o}{\partial z} = \beta_1(w_{20}^o w_{11}^e + w_{10}^e w_{21}^o) + \beta_3 w_1^o w_{20}^o, \qquad (93)$$

and

$$w_{11}^e + w_{21}^o + w_1^o + w_2^e = J_2 = \text{const}. \qquad (94)$$

where J_2 is an integral of motion of the system of equations (79 *b*), (92) and (93), and the expressions (81 *a, b*) are used as the first approximation solutions w_{10}^e, w_{20}^o. Using the conservation law (94) and the expression (81 *a, b*) we obtain the general solution of the equations (92) and (93). It has the form [38]

$$w_{11}^e = C_1\{\cosh(\eta_0)\,\mathrm{sech}(\eta - \eta_0)\}^2 + \{\mathrm{sech}(\eta - \eta_0)\}^2 \int_0^\eta d\eta' \{\cosh(\eta' - \eta_0)\}^2 \times \{1 - \tanh(\eta' - \eta_0)\}\{w_1^o + w_2^e(1 - b_1) - J_2\} \quad (95)$$

and

$$w_{21}^o = C_2\{\cosh(\eta_0)\,\mathrm{sech}(\eta - \eta_0)\}^2 + \{\mathrm{sech}(\eta - \eta_0)\}^2 \int_0^\eta d\eta' \{\cosh(\eta' - \eta_0)\}^2 \times \{1 + \tanh(\eta' - \eta_0)\}\{J_2 - w_2^e - w_1^o(1 - b_2)\}. \quad (96)$$

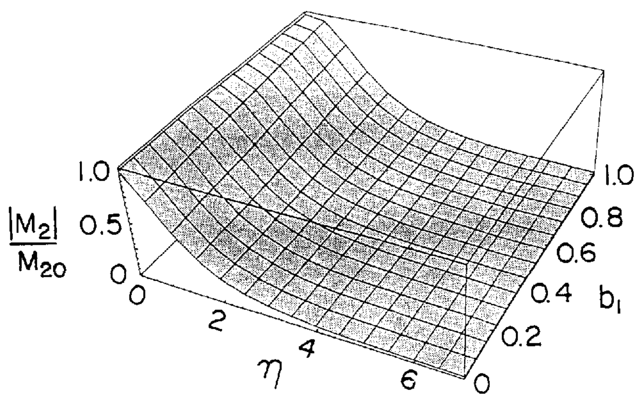
The equations (79 *b*), (92) and (93) describe the influence of the small components w_1^o and w_2^e on the main components w_1^e and w_2^o, and it is clear that at the boundary of the non-linear medium $z = 0$ the small corrections w_{11}^e and w_{21}^o do not exist:

$$w_{11}^e(0) = w_{21}^o(0) = 0. \qquad (97)$$

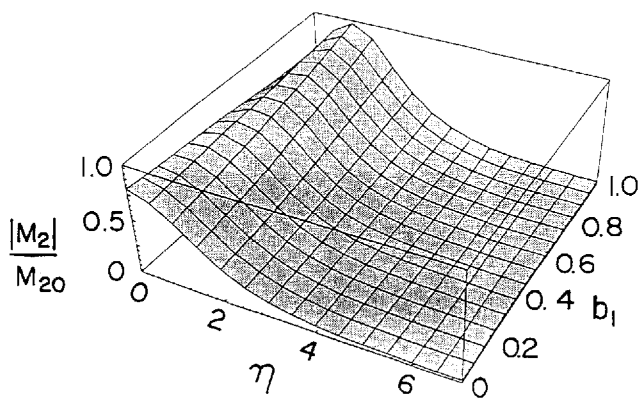
As a result we find that

$$C_1 = C_2 = 0, \tag{98}$$

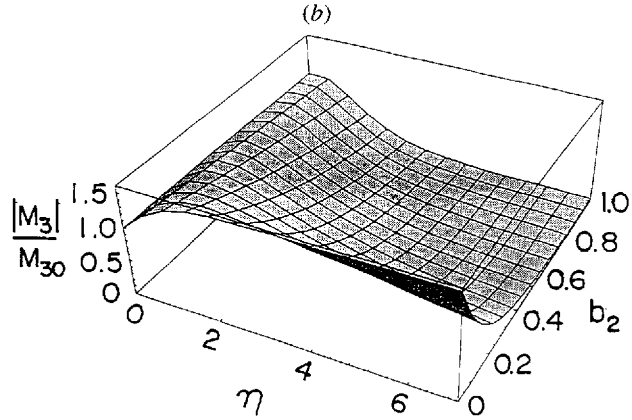
$$w_{11}^e = \{\operatorname{sech}(\eta - \eta_0)\}^2 \int_0^\eta d\eta' \{\cosh(\eta' - \eta_0)\}^2 \times \{1 - \tanh(\eta' - \eta_0)\} \times \{w_1^o + w_2^e(1 - b_1) - J_2\}, \tag{99a}$$



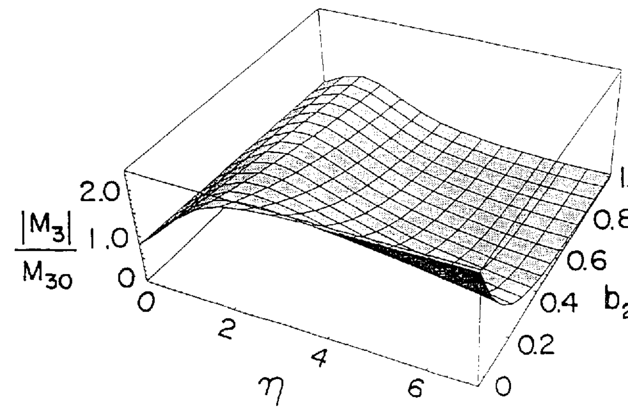
(*a*)



(*b*)



(*c*)



(*d*)

Figure 6. The dependence of the reduced moduli ($|M_2|/M_{20}$) (6*a,b*) and ($|M_3|/M_{30}$) (6*c,d*) of the dynamic grating amplitudes on the dimensionless coordinate η and exponents $0 \leqslant b_1 \leqslant 1$, $0 \leqslant b_2 \leqslant 1$, respectively, for the pumping -signal ratio $(w_1^e(0)/w_2^o(0)) = 1{\cdot}5$ (6*a,c*), $(w_1^e(0)/w_2^o(0)) = e^2$(6*b,d*).

$$w_{21}^o = \{\operatorname{sech}(\eta - \eta_0)\}^2 \int_0^\eta d\eta' \{\cosh(\eta' - \eta_0)\}^2 \times \{1 - \tanh(\eta' - \eta_0)\} \times \{J_2 - w_2^e - w_1^o(1 - b_2)\} \tag{99b}$$

and

$$J_2 = w_1^o(0) + w_2^e(0). \tag{100}$$

The solutions (99*a,b*) satisfy the conservation law automatically, which may be proved immediately. Combining the relationships (79*b*), (81*a,b*) and (99*a,b*) we obtain

$$w_1^o + w_2^e + w_{11}^e + w_{21}^o = w_1^o + w_2^e + \{\operatorname{sech}(\eta - \eta_0)\}^2 \times \int_0^\eta d\eta' \frac{\partial}{\partial \eta'} \{[\cosh(\eta' - \eta_0)]^2 \times (J_2 - w_1^o - w_2^e)\} \equiv J_2. \tag{101}$$

The explicit calculation of the integrals (99*a,b*) is hardly possible, however we may evaluate the lower and upper limits of these expressions using the relationships (81*ab*), (85) and (86), and assuming without the loss of generality that

$$b_{1,2} = \frac{\beta_{2,3}}{\beta_1} < 1.$$

Consider firstly w_{11}^e. Substituting the relationships (85) and (86) into (99*a*) and taking into account that

$$\exp(2\eta_0) = w_1^e(0)/w_2^o(0),$$

we obtain

$$W_{11}^- < w_{11}^e < W_{11}^+,$$

$$W_{11}^{-} = -(\tfrac{1}{2})\{\operatorname{sech}(\eta - \eta_0)\}^2\{\eta + \{1 - \exp(-2\eta)] \times w_1^e(0)/2w_2^o(0)\}\{w_1^o(0) + b_1 w_2^e(0)\},$$

and (102)

$$W_{11}^{+} = (\tfrac{1}{2})\{\operatorname{sech}(\eta - \eta_0)\}^2\{\eta + [1 - \exp(-2\eta)] \times w_1^e(0)/2w_2^o(0)\}w_2^e(0)\{[1 + w_1^e(0)/w_2^o(0)]^{b_2} \times (1 - b_2) - 1\}.$$

The inequality (102) shows that w_{11}^e is finite for all η and $\eta \to \infty$ and $w_{11}^e \to 0$ since both the upper limit W_{11}^{+} and the lower limit W_{11}^{-} of w_{11}^e tend to zero as $\eta \to \infty$.

Evaluating w_{21}^o by the same method we obtain

$$W_{21}^{-} < w_{21}^o < W_{21}^{+},$$

$$W_{21}^{-} = (1/2)\{\operatorname{sech}(\eta - \eta_0)\}^2\{\eta + [\exp(2\eta) - 1] \times w_2^o(0)/2w_1^e(0)\}\{w_2^e(0)[1 - (1 + w_1^e(0)/w_2^o(0))^{b_1}] + b_2 w_1^o(0)\}$$

and (103)

$$W_{21}^{+} = (1/2)\{\operatorname{sech}(\eta - \eta_0)\}^2 w_1^o(0)\{\eta + [\exp(2\eta) - 1] \times w_2^o(0)/2w_1^e(0)\}.$$

The inequality (103) shows that w_{21}^o is finite for all η since

$$\lim_{\eta \to \infty} W_{21}^{+} = w_1^o(0)$$

and (104)

$$\lim_{\eta \to \infty} W_{21}^{-} = w_2^e(0)\{1 - [1 + w_1^e(0)/w_2^o(0)]^{b_1}\} + b_2 w_1^o(0).$$

For the analysis of the behaviour of w_{21}^o at $\eta \to \infty$ we divide the interval of integration in (99 *b*) into two parts $(0, \eta_1)$ and (η_1, ∞) where $\eta_1 = \text{const}$ is a sufficiently large number such that for $\eta > \eta_1$, w_1^o and w_2^e are assumed to be constant according to the conditions (85) and (86). It is seen from the condition (103) that the integral over the first part of the interval is a finite number for any fixed η_1 however large it would be, and therefore

$$\lim_{\eta \to \infty} \left\{[\operatorname{sech}(\eta - \eta_0)]^2 \int_0^{\eta_1} [\cosh(\eta' - \eta_0)]^2 \, d\eta' \times [1 + \tanh(\eta' - \eta_0)][J_2 - w_2^e - w_{o1}(1 - b_2)]\right\} = 0.$$

The integral over the second part of the interval increases as $\exp[2(\eta - \eta_0)]$. Then substituting the values of w_2^e and w_1^o at $\eta \to \infty$ into (99 *b*) we obtain

$$\lim_{\eta \to \infty} w_{21}^o = \lim_{\eta \to \infty} \left\{[\operatorname{sech}(\eta - \eta_0)]^2 \int_{\eta_1}^{\eta} d\eta' [\cosh(\eta' - \eta_0)]^2 \times [1 + \tanh(\eta' - \eta_0)][J_2 - w_2^e(0) \times (1 + w_1^e(0)/w_2^o(0))^{b_1}]\right\} = w_1^o(0) + w_2^e(0) \times \{1 - (1 + w_1^e(0)/w_2^o(0))^{b_1}\} < w_1^o(0). \quad (105)$$

It is seen from the relationships (85), (86), (94) and (102)–(105) that the corrections w_{11}^e and w_{21}^o are finite, small and they satisfy the conservation law (94) for all η. The system is stable in respect to the thickness of the non-linear medium and to the pumping intensity. The numerical estimations show that the values of $\eta \sim (4\text{–}8)$ may be considered as the sufficiently large number η_1 for different $b_{1,2}$ as it is seen from the figures 2 and 3.

5. The Brillouin-like scattering on SS grating and the generation of the additional components

The Brillouin-like scattered harmonics $\mathbf{f}_i^S$ with the combination frequencies and wavevectors result from the coupling of the fundamental EM waves with the dynamic grating induced by them. They are governed by the terms in the non-linear polarization (46) which are not phase-matched to the fundamental modes (24) and (25). These harmonics evolve according to the wave equation

$$\operatorname{rot}\operatorname{rot}\mathbf{f}_i^S + (1/c^2)\frac{\partial^2}{\partial t^2}[\varepsilon_\perp(f_{ix}^S\mathbf{x} + f_{iy}^S\mathbf{y}) + \varepsilon_\parallel f_{iz}^S\mathbf{z}] = (-1/c^2) \times \frac{\partial^2}{\partial t^2}(\mathbf{D}_i^N \exp i\psi_i). \quad (106)$$

The combination frequencies and wavevectors do not satisfy the dispersion relations (22) and (23) and therefore we would take into account only the inhomogeneous particular solution determined by the right-hand side of (106) and having the form

$$\mathbf{f}_i^S = \mathbf{F}_i^S \exp i\psi_i + \text{c.c.} \quad (107)$$

These stimulated harmonics are essentially weak in comparison with the fundamental modes [15]. The analyses of the relationships (13), (24), (25) and (41) shows that there are 20 different phases of the harmonics f_i^S including the series of Stokes and anti-Stokes terms with the shifted frequencies

$$\omega_S = 2\omega_1 - \omega_2 = \omega_1 + \Omega$$

and (108)

$$\omega_A = 2\omega_2 - \omega_1 = \omega_2 - \Omega$$

and the terms with the fundamental frequencies $\omega_{1,2}$ and combination wavevectors, unlike the ordinary Brillouin scattering [1]. All phases are presented in appendix *B*. The explicit form of the amplitude F_i^S is too involved, and we do not present them here. However, we may conclude using the relationships (47), (70) and (74) that all harmonics are spatially localized and their amplitudes vanish at infinity:

$$z \to \infty; \quad |\mathbf{F}_i^S| \to 0.$$

The wave equations for the additional components f_{1x}, f_{1z}, f_{2y}, f_{3z} have the form

$$\operatorname{grad}\operatorname{div}\mathbf{f}_1-\nabla^2\mathbf{f}_1=\left(\frac{\omega_1}{c}\right)^2\{\varepsilon_\perp f_{1x}\mathbf{x}+\varepsilon_\parallel f_{1z}\mathbf{z}+(D^N_{1x}\mathbf{x}+D^N_{1z}\mathbf{z})\times\exp i(\mathbf{k}^o_1\mathbf{r}-\omega_1 t)\},\qquad(109)$$

$$\nabla^2 f_{2y}=\left(\frac{\omega_1}{c}\right)^2\left\{\varepsilon_\perp f_{2y}-\frac{1A^e_1}{4\pi\rho}\left[\frac{a_\perp\Delta k_{1z}e^o_{2y}}{G_1}\left(\frac{\Delta k_{1\perp}}{\Delta k_1}\right)^2\times h_1|A^o_2|^2+(a_\perp\Delta k_{2z}e^e_{2y}-\varepsilon_a\Delta k_{2y}e^e_{2z})\times G_2^{-1}\times h_2|A^e_2|^2\right]\exp i(\mathbf{k}^e_1\mathbf{r}-\omega_1 t)\right\}\qquad(110)$$

$$-\left(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y^2}\right)f_{3z}=\left(\frac{\omega_2}{c}\right)^2\left\{\varepsilon_\parallel f_{3z}-\frac{1}{4\pi\rho}\left[\frac{h_1}{G^*_1}\left(\frac{\Delta k_{1\perp}}{\Delta k_1}\right)^2\times(a_\parallel\Delta k_{1z}e^e_{1z}+\varepsilon_a\Delta k_{1x}e^e_{1x})|A^e_1|^2-\varepsilon_a\Delta k_{3y}\frac{h_3}{G^*_3}|A^o_1|^2\left(\frac{\Delta k_{3\perp}}{\Delta k_3}\right)^2\right]\times A^o_2\exp i(k^o_2\mathbf{r}-\omega_2 t)\right\},\qquad(111)$$

where

$$D^N_{1x}=-\frac{1}{4\pi\rho}\left[a_\perp\frac{h_3}{G_3}\left(\frac{\Delta k_{3\perp}}{\Delta k_3}\right)^2e^o_{2x}|A^o_2|^2+(a_\perp\Delta k_{4z}e^e_{2x}-\varepsilon_a\Delta k_{4x}e^e_{2z})\frac{h_4}{G_4}\left(\frac{\Delta k_{4\perp}}{\Delta k_4}\right)^2|A^e_2|^2\right]A^o_1,$$

$$D^N_{1z}=\frac{1}{4\pi\rho}\left\{[-a_\parallel\Delta k_{4z}e^e_{2z}+\varepsilon_a(\Delta k_{4x}e^e_{2x}+\Delta k_{4y}e^e_{2y})]\frac{h_4}{G_4}\left(\frac{\Delta k_{4\perp}}{\Delta k_4}\right)^2|A^e_2|^2+\varepsilon_a(\Delta k_{3z}e^o_{2x}+\Delta k_{3y}e^e_{2y})\frac{h_3}{G_3}\left(\frac{\Delta k_{3\perp}}{\Delta k_3}\right)^2|A^o_2|^2\right\}A^o_1.$$

It is seen from the equations (109)–(111) that the phases in the right-hand sides do not satisfy the corresponding homogeneous equations, and therefore the homogeneous solutions of (109)–(111) should be omitted. The inhomogeneous harmonics driven by the non-linear polarization have the form

$$f_{1x}=\frac{1}{(\varepsilon_\parallel-\varepsilon_\perp)(k^o_{1x})^2}\left\{-D^N_{1x}\left[\frac{\omega_1^2}{c^2}(\varepsilon_\parallel-\varepsilon_\perp)+(k^o_{1z})^2\right]+D^N_{1z}k^o_{1x}k^o_{1z}\right\}\exp i(\mathbf{k}^o_1\mathbf{r}-\omega_1 t),\qquad(112)$$

and

$$f_{1z}=\frac{1}{(\varepsilon_\parallel-\varepsilon_\perp)(k^o_{1x})^2}\{D^N_{1x}k^o_{1x}k^o_{1z}-D^N_{1z}(k^o_{1x})^2\}\times\exp i(\mathbf{k}^o_1\mathbf{r}-\omega_1 t),\qquad(113)$$

$$f_{2y}=\left(\frac{\omega_1}{c}\right)^2\frac{\varepsilon_\parallel A^e_1}{4\pi\rho(\varepsilon_\parallel-\varepsilon_\perp)(k^o_{1x})^2}\left\{\frac{a_\perp\Delta k_{1z}e^o_{2h}h_1}{G_1}\left(\frac{\Delta k_{1\perp}}{\Delta k_1}\right)^2\times|A^o_2|^2+(a_\perp\Delta k_{2z}e^e_{2y}-\varepsilon_a\Delta k_{2y}e^e_{2z})\frac{h_2}{G_2}\left(\frac{\Delta k_{2\perp}}{\Delta k_2}\right)^2\times|A^e_2|^2\right\}\exp i(\mathbf{k}^e_1\mathbf{r}-\omega_1 t),\qquad(114)$$

$$f_{3z}=\left(\frac{\omega_2}{c}\right)^2\frac{A^o_2}{4\pi\rho[(\omega_2/c)^2(\varepsilon_\parallel-\varepsilon_\perp)+(k^o_{2z})^2]}\times\left\{(a_\parallel\Delta k_{1z}e^e_{1z}+\varepsilon_a\Delta k_{1x}e^e_{1x})\left(\frac{\Delta k_{1\perp}}{\Delta k_1}\right)^2\frac{h_1}{G^*_1}|A^e_1|^2-\varepsilon_a\Delta k_{3y}\left(\frac{\Delta k_{3\perp}}{\Delta k_3}\right)^2\frac{h_3}{G^*_3}|A^o_1|^2\right\}\times\exp i(\mathbf{k}^o_2\mathbf{r}-\omega_2 t).\qquad(115)$$

It is seen from the relationships (112)–(115) that both anisotropy of the linear part of ε_{ik} and the non-linearity are essential for the existence of the components f_{1x}, f_{1z} and f_{2y}. The reason is that the wavevectors $\mathbf{k}^{o,e}_1$ and the optical axis OZ of S_A determine the XZ plane coinciding with the main cross-section [25], while the unit vector $\mathbf{e}^o_1$ (32) determines the OY axis thus breaking the rotational symmetry of S_A with respect to the ordinary wave $\mathbf{E}^o_1$ and the extraordinary wave $\mathbf{E}^e_1$. On the contrary, the ordinary wave $\mathbf{E}^o_2$ is polarized in the plane of the smectic layer, and for such a wave the OX and OY axes are degenerate. In this case the longitudinal component f_{3z} appears due to the non-linearity even in the case when a medium is isotropic in the linear approximation [1]. The amplitudes of the additional components are proportional to $|A^{o,e}_1|$ and therefore they are spatially localized

$$z\to\infty,\ |f_{1x}|,\ |f_{1z}|,\ |f_{2y}|,\ |f_{3z}|\to 0.\qquad(116)$$

The expressions (112)–(115) along with the fundamental modes (24) and (25) satisfy the condition (48) $\operatorname{div}\mathbf{D}=0$ as it may be shown directly.

6. The light-induced longitudinal waves

It is known in S_A the layer deformations produce the so-called flexoelectric polarization $\mathbf{P}_f$ [11, 29], which has the form [11]

$$P_{fx}=-e^f_3\frac{\partial^2 u}{\partial z\partial x},\quad P_{fy}=-e^f_3\frac{\partial^2 u}{\partial z\partial y},$$
$$P_{fz}=-e^f_1\left(\frac{\partial^2 u}{\partial x^2}+\frac{\partial^2 u}{\partial y^2}\right)-\frac{\partial^2 u}{\partial z^2}e^f_2\qquad(117)$$

where $e^f_{1,2,3}$ are the flexoelectric coupling constants. Comparing the relationship (41) and (117) one may see that the light-induced SS grating (41) gives rise to the high frequency travelling waves of polarization (117) which in turn would cause the high frequency electric field $\mathbf{E}_f$ according to the Maxwell equation [25]

$$\operatorname{div}\{\varepsilon_\perp \mathbf{E}_{f\perp} + \varepsilon_\| E_{fz}\mathbf{z} + 4\pi \mathbf{P}_f\} = 0. \tag{118}$$

The magnetic field $\mathbf{H}_f$ connected to $\mathbf{P}_f$ would be small and may be neglected:

$$H_f \sim (1/c)\frac{\Delta\omega}{\Delta k}P_f \sim (s/c)P_f \ll E_f.$$

Therefore we have

$$\operatorname{rot}\mathbf{E}_f = 0 \tag{119}$$

$\mathbf{E}_f$ is a kind of longitudinal wave [27]. Substituting (41) into (117) and solving together (118) and (119) we obtain taking into account (42)

$$\mathbf{E}_f = \left\{ -\sum_{j=1}^{4} i[(e_1^f + e_3^f)(\Delta k_{j\perp})^2 + e_2^f(\Delta k_{jz})^2] \times \frac{\Delta \mathbf{k}_j}{[\varepsilon_\perp(\Delta k_{j\perp})^2 + \varepsilon_\|(\Delta k_{jz})^2]}\frac{(\Delta k_{jz})(\Delta k_{j\perp})^2}{\rho(\Delta k_j)^2} \times \frac{h_j M_j}{G_j(\Delta\omega, \Delta\mathbf{k}_j)}\exp i\Delta\mathbf{k}_j\mathbf{r}\right\}\exp(-i\Delta\omega t) + \text{c.c.} \tag{120}$$

The flexoelectric polarization $\mathbf{P}_f$ also gives rise to the space charge Q_f [11], [25]:

$$Q_f = -\operatorname{div}\mathbf{P}_f = \left\{\sum_{j=1}^{4}\frac{1}{4\pi\rho}\frac{(\Delta k_{jz})(\Delta k_{j\perp})^2 h_j M_j}{(\Delta k_j)^2 G_j(\Delta\omega, \Delta\mathbf{k}_j)} \times [(e_1^f + e_3^f)(\Delta k_{j\perp})^2 + e_2^f(\Delta k_{jz})^2]\exp i\Delta\mathbf{k}_j\mathbf{r}\right\} \times \exp(-i\Delta\omega t) + \text{c.c.} \tag{120}$$

Comparing the expressions (71), (72) and (121) one may see that the flexoelectric polarization $\mathbf{P}_f$, the longitudinal field $\mathbf{E}_f$ and the space charge Q_f are spatially localized just like the SS harmonics. They are finite at $z = 0$, they have their extreme at $z = z_{oj}$ and they vanish at infinity:

$$z \to \infty,\ |\mathbf{E}_f|,\ |\mathbf{P}_f|,\ |Q_f| \sim |M_j| \to 0. \tag{122}$$

The longitudinal field $\mathbf{E}_f$ cannot penetrate into the linear medium $z < 0$ and represents the superposition of the high frequency surface waves at the interface $z = 0$ periodically distributed along $\Delta k_{j\perp}$ directions. The wavevectors $\mathbf{k}_j^s$ of the harmonics E_{fj} in the linear medium $z < 0$ must meet the boundary conditions [1], [25]:

$$k_{jx,y}^s = \Delta k_{jx,y}. \tag{123}$$

On the other hand

$$(k_{jz}^s)^2 = \left(\frac{\Delta\omega}{c}\right)^2 \varepsilon_s - (\Delta k_{j\perp})^2 \tag{124}$$

For $\Delta\omega \sim \Omega_j \sim (s/c)\omega_1$,

$$(k_{jz}^s)^2 \cong \left(\frac{s}{c}\omega_1\right)^2\left[\frac{\varepsilon_s}{c^2} - \frac{1}{s^2}\left(\frac{\Delta k_j}{\Delta k_{jz}}\right)^2\right] < 0, \tag{125}$$

$$\Delta k_{jz} \neq 0$$

and k_{jz}^s is an imaginary magnitude. The high frequency electric field with the artificially created periodicity on the interface $z = 0$ excites SS when applied to S_A [21,22]. The equations (120) and (123)–(125) show that the inverse effect is possible when the light-induced SS wave excites the periodical high frequency electric field at the interface between S_A and a linear medium.

7. Conclusions

We consider a stimulated scattering of an arbitrary polarized light on a new kind of Kerr non-linearity determined by the smectic layer deformations. Unlike the EM waves polarized either in the incidence plane or normal to it, the EM waves with the three-dimensional wavevectors split into the extraordinary and ordinary waves due to the birefringence of S_A [10, 25]. Consequently, the SLS in general case appeared to be FWM instead of the two-wave mixing discussed earlier [26]. The interaction of four EM waves on the Kerr non-linearity mentioned above results in the following chain of events. Four coupled fundamental waves are interfering and create the dynamic grating of the displacement $u(\mathbf{r}, t)$. The nonlinear part ε_{ik}^N of the dielectric constant tensor of S_A determined by this grating is complex due to the viscosity of S_A. As a result the coupled fundamental EM waves undergo simultaneously the parametric nonreciprocal energy transfer and the phase cross-modulation. It is shown that the pair of EM waves with the lower frequency is amplified while the pair of EM waves with the greater frequency is attenuated and finally depleted. In the resonant case when the frequency difference of the coupled EM waves is equal to the SS frequency and the SS dispersion relation is met the parametric amplification is essential and the phase cross-modulation is negligible. Far from the resonance S_A may behave as a focusing or defocusing medium depending on the ratio of the frequency difference of the EM waves and the resonant SS frequency. The analysis shows that the solutions obtained are stable at $z \to \infty$. The scattering of the fundamental EM waves on the field-induced grating results in the excitation of the number of secondary EM waves. Unlike ordinary Brillouin scattering, in the spectrum not only Stokes and anti-Stokes components exist, but there are the harmonics with the fundamental frequencies and combination wavevectors, too. The anisotropy and the non-linearity of S_A give rise to the additional components of the fundamental EM waves. It is shown that all excitations are spatially localized and stable since the terms describing the spatial dispersion and the non-linear terms are identical. The explicit expressions of the slowly varying amplitudes of the fundamental EM waves and of the SS waves are obtained for the important particular case when the coupling EM waves are mainly polarized in the incidence plane or normal to it. The light-induced dynamic grating of the layer deformations generates the high frequency polarization due to the flexoelectric effect. This polarization creates the longitudinal electric field and the space charge waves. All these excitations are spatially localized. The longitudinal electric field has the frequency and the wavevectors of the dynamic grating which fail to satisfy the EM dispersion relation and cannot penetrate into the linear medium. At the interface of S_A and the linear medium longitudinal waves behave like surface waves. The numerical estimations show that in the resonant case the coupling constant per unit intensity

$$\frac{\beta_1''}{\mathscr{P}} \sim (0{\cdot}01\text{–}10)\,\mathrm{cm\,MW^{-1}}$$

which is one–three orders of magnitude greater than the gain coefficient at the ordinary stimulated Brillouin scattering in isotropic organic liquids [1]. For the high pumping intensity $\mathscr{P} \sim 100\,\mathrm{MW\,cm^{-2}}$ the length of the interaction interval belongs to the range $(2\times10^{-3}\text{–}2)$ cm, which is in agreement with the experimental results [37].

Appendix A

The amplitudes U_j (42) are determined by the wavevectors, polarizations, frequencies and amplitudes of the interacting EM waves and by the material parameters of S_A. The wavevectors of the dynamic grating have the form

$$\Delta\mathbf{k}_1 = \mathbf{k}_1^e - \mathbf{k}_2^o,\ \Delta\mathbf{k}_2 = \mathbf{k}_1^e - \mathbf{k}_2^e, \quad \Delta\mathbf{k}_3 = \mathbf{k}_1^o - \mathbf{k}_2^o,\ \Delta\mathbf{k}_4 = \mathbf{k}_1^o - \mathbf{k}_2^e. \tag{A 1}$$

The constants h_j containing the polarization dependence have the form

$$h_1 = a_\perp \Delta k_{1z} e_{1x}^e e_{2x}^o - \varepsilon_a[\Delta k_{1x} e_{1z}^e e_{2x}^o + \Delta k_{1y} e_{1z}^e e_{2y}^o], \tag{A 2}$$

$$h_2 = a_\perp \Delta k_{2z} e_{1x}^e e_{2x}^e + a_\| \Delta k_{2z} e_{1z}^e e_{2z}^e - \varepsilon_a[\Delta k_{2x}\times(e_{1x}^e e_{2z}^e + e_{1z}^e e_{2x}^e) + \Delta k_{2y} e_{1z}^e e_{2y}^e], \tag{A 3}$$

$$h_3 = a_\perp \Delta k_{3k} e_{2y}^o, \tag{A 4}$$

and

$$h_4 = a_\perp \Delta k_4 z e_{2y}^e - \varepsilon_a \Delta k_{4y} e_{2z}^e. \tag{A 5}$$

The amplitude factors M_j are

$$M_1 = A_1^e(A_2^o)^*,\quad M_2 = A_1^e(A_2^e)^*,\quad M_3 = A_1^o(A_2^o)^*,\quad M_4 = A_1^o(A_2^e)^*, \tag{A 6}$$

Ω_j and Γ_j have the meaning of the frequency and the time decay of the SS eigenmode.

$$\Omega_j^2 = s^2\frac{(\Delta k_{j\perp})^2(\Delta k_{kj})^2}{(\Delta k_j)^2}, \tag{A 7}$$

$$\Gamma_j = \frac{1}{\rho}\left\{\alpha_1\left(\frac{\Delta k_{j\perp}\Delta k_{jz}}{\Delta k_j}\right)^2 + \tfrac{1}{2}(\alpha_4+\alpha_{56})(\Delta k_j)^2\right\} \tag{A 8}$$

where

$$(\Delta k_{j\perp})^2 = (\Delta k_{jx})^2 + (\Delta k_{jy})^2.$$

An asterisk means the complex conjugation operation.

Appendix B

The multiplication of the expressions (24) and (25) by the dielectric constant (44) yields 12 terms with the following combination frequencies and wavevectors:

$$\begin{aligned}
&[(2\mathbf{k}_1^e - \mathbf{k}_2^o)\mathbf{r} - (\omega_1+\Delta\omega)t],\\
&[(2\mathbf{k}_1^o - \mathbf{k}_2^o)\mathbf{r} - (\omega_1+\Delta\omega)t],\\
&[(2\mathbf{k}_1^e - \mathbf{k}_2^e)\mathbf{r} - (\omega_1+\Delta\omega)t],\\
&[(2\mathbf{k}_1^o - \mathbf{k}_2^e)\mathbf{r} - (\omega_1+\Delta\omega)t],\\
&[(\mathbf{k}_1^e + \mathbf{k}_1^o - \mathbf{k}_2^o)\mathbf{r} - (\omega_1+\Delta\omega)t],\\
&[(\mathbf{k}_1^e + \mathbf{k}_1^o - \mathbf{k}_2^e)\mathbf{r} - (\omega_1+\Delta\omega)t],\\
&[(2\mathbf{k}_2^e - \mathbf{k}_1^o)\mathbf{r} - (\omega_2-\Delta\omega)t],\\
&[(2\mathbf{k}_2^e - \mathbf{k}_1^e)\mathbf{r} - (\omega_2-\Delta\omega)t],\\
&[(2\mathbf{k}_2^o - \mathbf{k}_1^o)\mathbf{r} - (\omega_2-\Delta\omega)t],\\
&[(2\mathbf{k}_2^o - \mathbf{k}_1^e)\mathbf{r} - (\omega_2-\Delta\omega)t],\\
&[(\mathbf{k}_2^o + \mathbf{k}_2^e - \mathbf{k}_1^o)\mathbf{r} - (\omega_2-\Delta\omega)t],\\
&[(\mathbf{k}_2^o + \mathbf{k}_2^e - \mathbf{k}_1^e)\mathbf{r} - (\omega_2-\Delta\omega)t].
\end{aligned} \tag{B 1}$$

Four of these terms are doubly degenerate. Besides that there are 8 terms with the fundamental frequencies $\omega_{1,2}$ and with the combination wavevectors:

$$\begin{aligned}
&[(\mathbf{k}_2^e - \mathbf{k}_2^o + \mathbf{k}_1^{o,e})\mathbf{r} - \omega_1 t],\\
&[(\mathbf{k}_2^o - \mathbf{k}_2^e + \mathbf{k}_1^{o,e})\mathbf{r} - \omega_1 t],\\
&[(\mathbf{k}_1^e - \mathbf{k}_1^o + \mathbf{k}_2^{o,e})\mathbf{r} - \omega_2 t],\\
&[(\mathbf{k}_1^o - \mathbf{k}_1^e + \mathbf{k}_2^{o,e})\mathbf{r} - \omega_2 t],
\end{aligned} \tag{B 2}$$

It is easy to see that such terms are specific for the partly degenerate FWM. The four terms with the fundamental phases are also doubly degenerate since only two different frequencies exist: ω_1 and ω_2. The total number of the terms is consequently equal to 32, including 24 terms with essentially different phase factors: 4 phase matched terms and 20 scattered harmonics, as mentioned above.

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