Mechanism of the transient stimulated Rayleigh scattering in liquids

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In this paper we present a new theoretical approach to the transient stimulated scattering of light. It was shown that a light-induced grating giving rise to stimulated scattering can be depicted in terms of nonlinear Volterra equations. Taking advantage of this approach we made a detailed study of the mechanism of transient stimulated Rayleigh scattering (SRLS), for which the pulse duration of laser light and the rise time of the isobaric modulations of the dielectric functions are of the same order. Assuming a small depletion of laser-light intensity only, we derive expressions describing the spatially and temporally resolved distributions of isobaric modulations of the dielectric function and of the optical field. From our analysis it follows that SRLS is generated only if a phase mismatch between the optical field and the light-induced isobaric grating occurs. Such a phase mismatch is due to the finite rise time of the grating. The calculations confirm that in absorbing liquids the appearing local heating determines the generation of isobaric gratings. Furthermore they put into evidence that in transparent liquids electrostriction and the optical Kerr effect become significant. The hitherto unexplained shift in the spectrum of SRLS in transparent liquids should now be related to the optical Kerr effect. It was also found that for some experimental conditions a significant part of SRLS reveals a wave front reversed to the incident laser field.

I. INTRODUCTION

Time-resolved stimulated scattering was extensively studied over the last 20 years.[1–4] Several methods were developed which yield an adequate picture of the time evolution of stimulated Brillouin scattering[4–8] (SBS) and stimulated Raman scattering[9] (SRS) for a wide range of experimental conditions. These methods fail, however, for stimulated Rayleigh scattering (SRLS). The question of how to describe, under realistic experimental conditions, the SRLS and the phase gratings involved becomes important again in view of the experimental demonstration of the optical phase conjugation due to Bragg reflection from light-induced phase gratings in linear absorbing liquids.[10,11]

The mechanism of stimulated Rayleigh scattering is now well understood qualitatively. As it was experimentally and theoretically proved, stimulated Rayleigh scattering is due to isobaric modulation of the susceptibility function of the medium caused by density and temperature variations.[2–4,12–15] On the other hand, these modulations are created by the optical field arising from the interference of the laser and scattered waves. In liquids, linear absorption, electrostriction, and the electrocaloric effect are considered to be involved in the interaction of the optical field and the medium.

The role of the linear absorption was first pointed out by Herman and Gray,[16] who found that local heating due to linear absorption created temperature and density variations giving rise to stimulated scattering with the spectrum shifted to the anti-Stokes side. This was later confirmed by several experiments performed in colored liquids.[2,13] The SRLS ruled by linear absorption has been called the stimulated thermal scattering (STS-II).

If the contribution of linear absorption vanishes, as is the case with transparent liquids, the scattered light spectrum is shifted to the Stokes side. The mechanism of this scattering, referred to as STS-I, is not yet clear. The first explanation of STS-I was given by Starunov *et al.*,[3,17] who assigned the modulations of the susceptibility function to the fluctuations of isobaric entropy caused by the electrocaloric effect. This explanation involved, however, the improper form of the source term $(\partial\epsilon/\partial T)_p$. Later, Starunov,[15] Harrison *et al.*,[12] Wang and Herman,[18] and Enns *et al.*[19] proved that in the stationary case the electrocaloric effect has only a small contribution to the susceptibility function.

Searching for other sources of STS-I, Wang and Herman[18] considered the influence of isobaric and adiabatic mode coupling. Taking advantage of the previous calculations of Rother[14] they found that this mechanism contributes significantly to the isobaric modulations of the dielectric function. Their analysis did not include, however, the coupling of modes due to the influence of the scattered radiation.

Several authors[20–22] have independently discussed the influence of the finite rise time to the temperature and density isobaric modulations on SRLS. They have shown that in the cases when the rise time of isobaric fluctuations and the width of the laser pulse are of the same order, the gain factor has a form which cannot be explained in terms of the steady-state theory. To overcome these difficulties several attempts were made to develop a transient theory of SRLS.

For the case of small depletion of the laser-light intensity and small amplification of scattered light, Rother derived expressions[14] which describe the intensity of the scattered electric field as a series expansion of the "z"

coordinate. This approach was extended by Rangnekar and Enns.[22]

A different treatment was based on particular models of the incident optical field. Using these models the time-dependent gain factors and the temporally and spatially resolved distributions of scattered light were calculated analytically[23–25] and numerically.[24–27]

In the present paper a theory of SRLS is developed in which use is made of the integral-equations approach. We show that the isobaric grating giving rise to SRLS can be depicted in terms of the nonlinear integral Volterra equations. Taking advantage of this approach we made a detailed study of the mechanisms of transient SRLS, for which the pulse duration of laser light and the rise time of the isobaric modulations of the dielectric function are of the same order.

In Sec. II, starting from Maxwell's, heat-transport, and the Navier-Stokes equations, we evaluate integral equations describing the distribution of light-induced gratings giving rise to stimulated scattering. We prove that, making use of a simple linearization procedure, these equations can be solved using standard methods. In Sec. III, on the basis of that integral-equations approach, in assuming a small depletion of laser-light intensity only, we derive expressions describing the spatially and temporally resolved distribution of the optical field, as well as the isobaric modulations of the liquid in stimulated Rayleigh scattering. Particular attention was paid to the SRLS in amplifier systems. Section IV contains a detailed discussion of the mechanism of SRLS for various experimental situations and furthermore an analysis of the conditions under which the optical phase conjugation in this kind of light scattering should be observed.

II. GENERAL THEORY

Stimulated Rayleigh scattering may be described by the Maxwell wave equation of the form[2]

$$\Delta \mathbf{E}(\mathbf{r},t)-\frac{\alpha n_0}{c}\frac{\partial \mathbf{E}(\mathbf{r},t)}{\partial t}-\frac{n_0^2}{c^2}\frac{\partial^2 \mathbf{E}(\mathbf{r},t)}{\partial t^2}=\frac{1}{c^2}\frac{\partial^2[\mathbf{E}(\mathbf{r},t)\epsilon_1(\mathbf{r},t)]}{\partial t^2}+\cdots , \quad (1)$$

where $\mathbf{E}(\mathbf{r},t)$ is the optical-field electric vector and $\epsilon_1(\mathbf{r},t)$ describes the modulations of the dielectric function. The ellipsis represents unspecified terms depicting other nonlinear phenomena. The first term on the right-hand side of (1) is related to the perturbations of density and temperature, denoted by $\rho_1(\mathbf{r},t)$ and $T_1(\mathbf{r},t)$, respectively, by

$$\epsilon_1(\mathbf{r},t)=\left[\frac{\partial \epsilon}{\partial T}\right]_\rho T_1(\mathbf{r},t)+\left[\frac{\partial \epsilon}{\partial \rho}\right]_T \rho_1(\mathbf{r},t) . \quad (2)$$

As it has been proved,[2] in liquids these perturbations satisfy the hydrodynamic as well as the heat-conduction equations, the optical field being a superposition of laser and scattered radiation. Since the laser and the backward scattered radiation are the most significant components of the optical field,[1,2,4] we shall consider in our analysis the laser and the scattered light traveling in opposite directions. Taking advantage of this assumption, we take $\mathbf{E}(\mathbf{r},t)$, $\rho_1(\mathbf{r},t)$, and $T_1(\mathbf{r},t)$ in the form

$$\mathbf{E}=\tfrac{1}{2}\{[A_L(z,t)e^{i(\omega_L t-k_L z)-\alpha z/2}+A_s(z,t)e^{i(\omega_L t+k_L z)+\alpha z/2}]+\text{c.c.}\}\mathbf{e} , \quad (3)$$

$$\rho_1=\tfrac{1}{2}[\rho(z,t)e^{-ikz}+\text{c.c.}] , \quad (4)$$

$$T_1=\tfrac{1}{2}[T(z,t)e^{-ikz}+\text{c.c.}] . \quad (5)$$

The complex functions A_L, A_s, ρ, and T denote the slowly varying amplitudes of the laser, Rayleigh, density, and temperature waves, respectively; k_L and $k=2k_L$ are the wave vectors; ω_L is the laser-light frequency; and $\mathbf{e}$ is the polarization vector.

Substituting expressions (3), (4), and (5) into Maxwell's, hydrodynamic, and heat-transport equations and neglecting small terms involving derivatives of slowly varying amplitudes, we obtain in the liquid limit[2] a system of the form

$$\frac{\partial A_s}{\partial z}-\frac{n_0}{c}\frac{\partial A_s}{\partial t}=\frac{ik_L}{4n_0^2}\epsilon^*(z,t)A_L(z,t)e^{-\alpha z}+\frac{6\pi\omega_L i}{n_0 c}\chi^{(3)}(A_L A_L^* e^{-\alpha z}+A_s A_s^* e^{\alpha z})A_s , \quad (6)$$

$$\frac{\partial A_L}{\partial z}+\frac{n_0}{c}\frac{\partial A_L}{\partial t}=-\frac{ik_L}{4n_0^2}\epsilon(z,t)A_s(z,t)e^{\alpha z}-\frac{6\pi\omega_L i}{n_0 c}\chi^{(3)}(A_L A_L^* e^{-\alpha z}+A_s A_s^* e^{\alpha z})A_L , \quad (7)$$

$$\left[-\frac{\partial^2}{\partial t^2}-\left[\frac{v^2k^2}{\gamma}-\frac{\eta k^2}{\rho_0}\frac{\partial}{\partial t}\right]\right]\rho(z,t)-\frac{v^2\beta_T\rho_0 k^2}{\gamma}T(z,t)=-\frac{\rho_0 k^2(\partial\epsilon/\partial\rho)_T}{8\pi}A_L(z,t)A_s^*(z,t) , \quad (8)$$

$$\left[\frac{\partial}{\partial t}-\frac{\lambda_T k^2}{\rho_0 c_v}\right]T(z,t)-\frac{\gamma-1}{\beta_T\rho_0}\frac{\partial\rho(z,t)}{\partial t}=\frac{n_0 c\alpha}{4\pi\rho_0 c_v}A_L(z,t)A_s^*(z,t)-\frac{1}{8\pi}\left[\frac{\partial\epsilon}{\partial T}\right]_\rho\frac{\gamma T_0}{\rho_0 c_v}\frac{\partial A_L A_s^*}{\partial t} , \quad (9)$$

where v is the velocity of sound; $\gamma=c_p/c_v$ is the ratio of the specific heats at constant pressure and volume; λ_T and β_T are the coefficients of heat conduction and thermal expansion, respectively; n_0 is the optical refractive index; α is the linear absorption coefficient; η characterizes the damping of the acoustic waves; T_0 and ρ_0 are the average values of temperature and density; and $\chi^{(3)}$ is the nonlinear susceptibility tensor.

The term on the right-hand side of Eq. (8) describes the contribution due to electrostriction, and the two terms on the right-hand side of (9) represent the influence of linear absorption and electrocaloric effect, respectively. The last terms in (6) and (7) represent the self-action of light connected with the optical Kerr effect. These terms in (6) and (7) were neglected in the previous theoretical considerations.[1–4]

On the basis of Eqs. (6)–(9) we derive our integral-equations approach.

Using the standard methods[2,28] we express the functions $\rho(z,t), T(z,t)$ in terms of optical-field components:

$$\rho(z,t)=\sum_{j=1}^{3}[\rho_{0j}(z,t_0)+c_jF_j(z,t)]e^{\Omega_j t}, \tag{10}$$

$$T(z,t)=\sum_{j=1}^{3}[T_{0j}(z,t_0)+d_jF_j(z,t)]e^{\Omega_j t}+\frac{\gamma T_0}{8c_p\rho_0}\left[\frac{\partial\epsilon}{\partial T}\right]_\rho A_L(z,t)A_s^*(z,t), \tag{11}$$

where $\rho_{0j}(z,t_0), T_{0j}(z,t_0)$ are the values of $\rho(z,t), T(z,t)$ at time t_0 and

$$F_j(z,t)=\int_{t_0}^{t}e^{\Gamma_j t'}A_L(z,t')A_s^*(z,t')dt'. \tag{12}$$

The relations between the particular constants are as follows:

$$\Gamma_R=\frac{2k^2\lambda_T}{\rho_0 c_p},\quad \Gamma_B=\frac{\eta k^2}{\rho_0}+\frac{\gamma-1}{2}\Gamma_R, \qquad \Omega_1=-\frac{\Gamma_R}{2},\quad \Omega_2=\Omega_3^*=i\omega_B-\frac{\Gamma_B}{2}, \tag{13}$$

$$\beta^a=\frac{\beta_T n_0 c}{8\pi c_p},\quad \beta^e=\frac{k^2\rho_0}{32\pi\omega_B}\left[\frac{\partial\epsilon}{\partial\rho}\right]_T,$$

$$\beta^c=\frac{(\gamma-1)\Gamma_R k^2\rho_0}{32\pi\omega_B^2}\left[\frac{\partial\epsilon}{\partial\rho}\right]_T, \tag{14a}$$

$$\beta^d=\frac{T_0\beta_T\Gamma_R}{32c_p}\left[\frac{\partial\epsilon}{\partial T}\right]_\rho,$$

$$c_1=2(\beta^2-\beta^d-\beta^a), \qquad c_3^*=c_2=-\tfrac{1}{2}c_1-2i\left[\beta^e-\frac{\omega_B}{\Gamma_R}\beta^d\right], \tag{14b}$$

$$d_1=-\frac{1}{\beta_T\beta_0}c_1,\quad d_2=d_3^*=\frac{\gamma-1}{\beta_T\rho_0}\left[1+\frac{i\gamma\Gamma_R}{2\omega_B}\right]c_2. \tag{14c}$$

Now the modulations of the dielectric functions are expressed in the form

$$\epsilon(z,t)=\epsilon_R F_1(z,t)e^{-(\Gamma_R/2)t}+(\epsilon_{BR}-i\epsilon_{BJ})F_2(z,t)e^{(i\omega_B-\Gamma_B/2)t}+(\epsilon_{BR}+i\epsilon_{BJ})F_3(z,t)e^{-(i\omega_B+\Gamma_B/2)t}+\sum_{j=1}^{3}\epsilon_j(z,t_0)e^{\Omega_j t}+\epsilon_T A_L A_s^*, \tag{15}$$

where

$$\epsilon_R=-2(\beta^a-\beta^d-\beta^c)\left[\left[\frac{\partial\epsilon}{\partial\rho}\right]_T-\frac{1}{\beta_Q\rho_T}\left[\frac{\partial\epsilon}{\partial T}\right]_\rho\right], \tag{16a}$$

$$\epsilon_{BR}=(\beta^a-\beta^d-\beta^c)\left[\frac{\partial\epsilon}{\partial\rho}\right]_T+\frac{\gamma-1}{\beta_T\rho_0}\left[\frac{\partial\epsilon}{\partial T}\right]_\rho\left[(\beta^a-\beta^d-\beta^c)+\gamma\left[\beta^d+\frac{\Gamma_R}{\omega_B}\beta^e\right]\right], \tag{16b}$$

$$\epsilon_{BJ}=2\left[\left[\frac{\partial\epsilon}{\partial\rho}\right]_T+\frac{\gamma-1}{\beta_T\rho_0}\left[\frac{\partial\epsilon}{\partial T}\right]_\rho\right]\left[\beta^e+\frac{\omega_B}{\Gamma_R}\beta^d\right], \tag{16c}$$

$$\epsilon_T=-\frac{T_0\gamma}{8\pi\rho_0 c_p}\left[\frac{\partial\epsilon}{\partial T}\right]_\rho^2. \tag{16d}$$

The first term in (15) describes the isobaric perturbations; the second and third ones relate to the sound wave traveling parallel and antiparallel to the laser beam, respectively; and the last two terms relate to the initial conditions and to the contributions provided by the electrocaloric effect. The coefficients β^e, β^a, β^c, and β^d are assigned to the electrostriction, linear absorption, mode coupling, and electrocaloric effect, respectively. In expressions (14) and (16) the terms proportional to $(\Gamma_R/\omega_B)^2$ and $(\Gamma_B/\omega_B)^2$ have been neglected.

After substituting (15), Eqs. (6) and (7) may be transformed to take the form

$$\frac{\partial^2F_j(z,t)}{\partial z\,\partial t}+\frac{2n_0}{cA_L}\frac{\partial A_L}{\partial t}\frac{\partial F_j}{\partial t}-\frac{n_0}{c}\frac{\partial^2F_j}{\partial t^2}-\frac{n_0\Omega_j}{c}\frac{\partial F_j}{\partial t}+\frac{ik_L}{4n_0^2}\left[e^{-\alpha z}I_L+e^{\alpha z}I_s\right]\left[2\frac{\partial F_j(z,t)}{\partial t}\epsilon_k+\sum_{l=1}^{3}[\epsilon_l(z,t_0)+\epsilon_lF_l(z,t)]e^{(\Omega_l-\Omega_j)t}\right]=0, \tag{17}$$

$$\frac{\partial I_s}{\partial z}-\frac{n_0}{c}\frac{\partial I_s}{\partial t}=\frac{ik_Le^{-\alpha z}}{4n_0^2}\left[\left[\sum_{l=1}^{3}\epsilon_l^*F_l^*+\epsilon_l^*(z,t_0)e^{(\Omega_l^*-\Omega_j^*)t}\right]\frac{\partial F_j}{\partial t}e^{-\Gamma_j t}-\text{c.c.}\right], \tag{18}$$

$$\frac{\partial I_L}{\partial z}+\frac{n_0}{c}\frac{\partial I_L}{\partial t}=\frac{ik_L e^{\alpha z}}{4n_0^2}\left[\left[\sum_{l=1}^{3}\epsilon_l^* F_l^* +\epsilon_l^*(z,t_0)e^{(\Omega_l^*-\Omega_j^*)t}\right]\frac{\partial F_j}{\partial t}e^{-\Gamma_j t}-\text{c.c.}\right], \tag{19}$$

where $I_s=A_sA_s^*$ and $\epsilon_k=24\pi\chi^{(3)}+\epsilon_T$.

From (18) and (19) we have immediately that

$$I_s(z,t+zn_0/c)=\int_L^z dz'' e^{+2\alpha z''}\frac{\partial}{\partial z''}I_L(z'',t-z''n_0/c)+I_s(L,t+Ln_0/c)\ . \tag{20}$$

Let us introduce new variables $t'=t+zn_0/c$, $z'=L-z$ and denote

$$F_j(z,t)=e^{\Omega_j(L-z')n_0/c}\bar{F}_j(z',t')\ . \tag{21}$$

Substituting (20) and (21) into (17) we finally get

$$-\frac{\partial^2\bar{F}_j(z',t')}{\partial z'\partial t'}+\frac{ik_L e^{-\alpha L}}{4n_0^2}\left[e^{\alpha z'}I_L(z',t'-2z'n_0/c)+\left[e^{-\alpha z'}\int_0^{z'}dz''e^{2\alpha z''}\frac{\partial}{\partial z''}I_L(z'',t'-2z''n_0/c)\right]+e^{-\alpha z'}I_s(0,t')\right]$$
$$\times\left[2\frac{\partial\bar{F}_j(z',t')}{\partial t'}\epsilon_k+\sum_{l=1}^{3}[\epsilon_l(z',t_0)+\epsilon_l\bar{F}_l(z',t')]e^{(\Omega_l-\Omega_j)t'}+\frac{2n_0}{cA_L}\frac{\partial A_L}{\partial t'}\frac{\partial\bar{F}_j(z',t')}{\partial t'}\right]=0\ . \tag{22}$$

Since Γ_L is of the order of Γ_R, we have the following relations:

$$\Gamma_R\approx\Gamma_L\ll\Gamma_B\ll\omega_B\ . \tag{23}$$

For real experimental conditions the case considered involves also[2]

$$\frac{2n_0L}{cA_L}\frac{\partial A_L}{\partial t'}\ll 1\ . \tag{24}$$

Integrating the system (22) and eliminating terms which are small in the meaning of (23) and (24) we get a set of nonlinear Volterra equations of the form (Appendix A)

$$\bar{F}_j(z',t')=f_j(z',t')+\lambda\int_{t_0}^{t'}dt''\int_0^{z'}dz''K_j(z',t';z'',t'')\bar{F}_j(z'',t'')\ , \tag{25}$$

where

$$f_j(z',t')=\bar{F}_j(0,t')+\frac{ik_L e^{-\alpha L}}{4n_0^2}\int_{t_0}^{t'}\int_0^{z'}J(z'',t'')\left[\sum_{l=1}^{3}\bar{\epsilon}_l(z'',t_0)e^{(\Omega_l-\Omega_j)t''}\right]dz''dt''\ , \tag{26}$$

$$\lambda=i\lambda_0=\frac{ik_L e^{-\alpha L}}{4n_0^2}\ , \tag{27}$$

$$K_j(z',t';z'',t'')=J(z'',t'')[2\delta(t'-t'')\epsilon_k+\epsilon_j]+\frac{\partial J(z'',t'')}{\partial t''}\left[\sum_{l\,(\neq j)}\frac{\epsilon_l}{\Omega_l-\Omega_j}-\epsilon_k\right]$$
$$+\sum_{l\,(\neq j)}\epsilon_l\left[J(z'',t')-\frac{\partial J(z'',t')}{\partial t'(\Omega_l-\Omega_j)}\right]\exp[(\Omega_l-\Omega_j)(t'-t'')]\ , \tag{28}$$

$$J(z',t')=e^{\alpha z'}I_L(z',t')+e^{-\alpha z'}\int_0^{z'}dz''e^{2\alpha z''}\frac{\partial}{\partial z''}I_L(z'',t'')+e^{-\alpha z'}I_s(0,t'')\ . \tag{29}$$

Since for each finite pulse of laser light the integral operator in (25) satisfies the Lipschitz condition,[29] the system of equations (19) and (25) has an iterative solution. However, in cases when a strong amplification of the modulations of the dielectric functions occurs, the iterated series obtained is slowly convergent.

The iterating procedure may be avoided by taking the laser-light intensity as a sum:

$$I_L(z',t')=I_{L0}(z',t')+\Delta I_L(z',t')\ , \tag{30}$$

where $I_{L0}(z',t')$ is an arbitrarily chosen function. If we find such a $I_{L0}(z',t')$ for which we have

$$I_{L0}(z',t')\gg\Delta I_L(z',t')\ , \tag{31}$$

then the equations (25) assume the form

$$\bar{F}_j(z',t')=f_j(z',t')+\int_{t_0}^{t'}dt''\int_0^{z'}dz''K_{j0}(z',t';z'',t'')\bar{F}_j(z'',t''+\int_{t_0}^{t'}dt''\int_0^{z'}dz''K_{jNL}(z',t';z'',t'')\bar{F}_j(z'',t'')\ . \tag{32}$$

The kernel of the first integral operator is expressed in terms of I_{L0}, while the kernel of the second one includes ΔI_L only. From the general theory of the nonlinear Volterra equation[29] it follows that due to the assumptions (24), (30), and (31) the second order causes only small changes of the shape of $\bar{F}_j(z',t')$. Hence the solution of (32) may be written in the form

$$\bar{F}_j(z',t')=f_j(z',t') + \int_{t_0}^{t'} dt'' \int_0^{z'} dz'' R_j(z',t';z'',t'') f_j(z'',t'') , \quad (33)$$

where the $R_j(z',t';z'',t'')$ resolvent is given by

$$R_j(z',t';z'',t'') = K_j(z',t';z'',t'') + \sum_{n=1}^{\infty} \lambda^n J_{jn}(z',t';z'',t'') , \quad (33a)$$

$$J_{j1}(z',t';z'',t'') = \int_{t''}^{t'} dt''' \int_{z''}^{z'} dz''' K_j(z',t';z''',t''') \times K_j(z''',t''';z'',t'') , \quad (33b)$$

$$J_{jn}(z',t';z'',t'') = \int_{t''}^{t'} dt''' \int_{z''}^{z'} dz''' K_j(z',t';z''',t''') \times J_{j(n-1)}(z''',t''';z'',t'') . \quad (33c)$$

Since $\epsilon(z',t')$ is now expressed in terms of the initial conditions and $I_{L0}(z',t')$ only, A_s and A_L may be determined using standard methods. Let

$$A_L(z',t')=A_{L0}(z',t')\exp[+i\lambda_0\zeta(z',t')] , \quad (34a)$$

$$A_s(z',t')=A_{s0}(z',t')\exp[-i\lambda_0\zeta(z',t')] , \quad (34b)$$

$$\epsilon_j(z',t_0)=\epsilon_{j0}(z',t')\exp[2i\lambda_0\zeta(z',t')] , \quad (34c)$$

$$\bar{F}_j(z',t')=\bar{F}_{j0}(z',t')\exp[+2i\lambda_0\zeta(z',t')] , \quad (34d)$$

where the function $\zeta(z',t')$ is given according to (20) and (31) by

$$\zeta(z',t')=\epsilon_k \int_0^{z'} \left[e^{\alpha z''} I_{L0}(z'',t'') + \left[e^{-\alpha z''} \int_0^{z''} dz''' e^{2\alpha z'''} \frac{\partial I_{L0}(z''',t'')}{\partial z'''} \right] + e^{-\alpha z''} I_s(0,t') \right] dz'' ; \quad (35)$$

then system (6) takes the form

$$\frac{\partial A_{s0}(z',t')}{\partial z'} = -i\lambda_0 \left[\sum_{j=1}^{3} [\epsilon_j^* F_j^*(z',t') + \epsilon_{j0}^*(z',t')] e^{\Omega_j^* t'} \right] \times A_{L0}(z't') e^{\alpha z''} , \quad (36)$$

$$\frac{\partial A_{L0}}{\partial z'} = i\lambda_0 A_{s0}(z't') \left[\sum_{j=1}^{3} [\epsilon_j \bar{F}_{j0}(z',t') + \epsilon_{j0}(z',t')] e^{\Omega_j t'} \right] e^{-\alpha z'} . \quad (37)$$

By integrating this system and making use of Fubini's theorem[28] we get immediately

$$A_{s0}(z',t') = \sum_{j=1}^{3} A_{sj} e^{(\Omega_j + \Gamma_j/2)t'} , \quad (38)$$

where

$$A_{sj}(z',t') = A_{sj}(0,t') - i\lambda_0 A_{L0}(0,t') \int_0^{z'} dz'' [\epsilon_j^* \bar{F}_{j0}^*(z'',t') + \epsilon_{j0}^*(z'',t')] e^{-\Gamma_j t'/2} e^{\alpha z''} + \lambda_0^2 \int_0^{z'} dz'' A_{sj}(z'',t') \left[\sum_{l=1}^{3} [\epsilon_l \bar{F}_{l0}(z'',t') + \epsilon_{l0}(z'',t')] \int_{z''}^{z'} dz''' [\epsilon_l^* \bar{F}_{j0}^*(z''',t') + \epsilon_{l0}^*(z''',t')] e^{-\Gamma_l t'/2} e^{\alpha(z'''-z'')} \right] . \quad (38a)$$

The second right-hand-side term in (38a) describes the interaction of the unperturbated laser wave with the isobaric modulations, and the last one is the correction associated with the change of laser field due to stimulated scattering.

Since (38a) is a linear equation with respect to $A_{sj}(z',t')$, its solution can be expressed using formulas analogous to (33) and (33a)–(33c) in which the definitions of kernel and resolvent are only changed. It follows from (33)–(38) that all information about the mechanisms of scattering processes including the coupling of the adiabatic and isobaric modes are involved in the resolvents which can be determined regardless of the initial conditions.

The approach as given in Sec. III provides a qualitatively new insight into the SRLS, being only an extension of the previous treatments[7,8] for SBS.

III. INTEGRAL-EQUATIONS APPROACH IN STIMULATED RAYLEIGH SCATTERING

A. The resolvent for the SRLS

Our linearization procedure of the equations describing SRLS is based on the assumption

$$I_s(z',t') \ll I_L(z',t') . \quad (39)$$

Hence, according to (24), we can write

$$I_L(z',t')=I_L(t')+\frac{\partial I_L(t')}{\partial t'}\frac{2z'n_0}{c}+\Delta I_L(z',t') , \quad (40)$$

where the last two terms, concerning the time retardation and the depletion of the laser pulse, respectively, are small with respect to the initial laser-light intensity given by $I_L(t')$. In this case

$$K_1(z',t';z'',t'')=e^{\alpha z''}\left[I_L(t'')[2\delta(t'-t'')\epsilon_k+\epsilon_R]+\frac{\partial I_L(t'')}{\partial t''}\left[\frac{\epsilon_{BJ}}{2\omega_B}-2\epsilon_k\right]\right] + \left[\epsilon_2\left[I_L(t'')-\frac{\partial I_L(t'')}{\partial t''(\Omega_2+\Gamma_R/2)}\right]\exp[(\Omega_2+\Gamma_R/2)(t'-t'')]+\text{c.c.}\right] , \quad (41)$$

while $K_{1NL}(z',t';z'',t'')$ is expressed in terms of I_s, ΔI_L, and $\partial I_L(t')/\partial t'$ only.

According to relations (23) the term

$$\frac{\partial I_L(t'')}{\partial t''(\Omega_2+\Gamma_R/2)}$$

can be neglected. Furthermore, since $\bar{F}_1(z',t')$ is definite by the convolution (12), it varies slowly with time with respect to $\exp[\Omega_2(t'-t'')]$. Thus

$$\int_{t_0}^{t'}\epsilon_2\left[\left[I_L(t''')-\frac{\partial I_L(t''')}{\partial t'''(\Omega_2+\Gamma_R/2)}\right]\exp[(\Omega_2+\Gamma_R/2)(t'-t'')]\right]\bar{F}_1(z'',t'')dt''\approx-\frac{\epsilon_2 I_L(t'')\bar{F}_1(z'',t')}{\Omega_2} . \quad (42)$$

Under these assumptions, one obtains

$$\bar{F}_1(z',t')=\bar{f}_1(z',t')+\lambda I_L(t')\epsilon_s\int_0^{z'}dz''e^{\alpha z''}\bar{F}_1(z'',t')+\int_{t_0}^{t'}dt''\int_0^{z'}dz''\bar{F}_1(z'',t'')e^{\alpha z''}\left[\epsilon_R I_L(t'')-\epsilon_s\frac{\partial I_L(t'')}{\partial t''}\right] , \quad (43)$$

where

$$\epsilon_s=2\epsilon_k-\frac{\epsilon_{BJ}}{2\omega_B} . \quad (43a)$$

Using infinite iteration, again taking advantage of the Fubini theorem and summing up the series obtained, we finally get the integral equation (Appendix B)

$$\bar{F}_1(z',t')=g_1(z',t')+\lambda\int_{t_0}^{t'}dz''\int_0^{z'}dz''\{\exp[\lambda\epsilon_s I_L(t'')(q'-q'')+\alpha z'']\}\left[\epsilon_R I_L(t'')-\epsilon_s\frac{\partial I_L(t'')}{\partial t''}\right]\bar{F}_1(z'',t'') , \quad (44)$$

where

$$g_1(z',t')=f_1(z',t')+\lambda\epsilon_s I_L(t')\int_0^{z'}dz''\{\exp[\alpha z''+\lambda\epsilon_s I_L(t')(q'-q'')]\}f_1(z'',t') , \quad (44a)$$

$$q'=\frac{e^{\alpha z'}-1}{\alpha} . \quad (44b)$$

Now, taking advantage of expressions (34a)–(34c), we get the resolvent of (44) in the form

$$R_1(z',t';z'',t'')=\{\exp[\alpha z''+\lambda\epsilon_s I_L(t'')(q'-q'')]\}\left[\epsilon_R I_L(t'')-\epsilon_s\frac{\partial I_L(t'')}{\partial t''}\right]B(z',t';z'',t'') . \quad (45)$$

The function $B(z',t';z'',t'')$ is given by

$$B(z',t';z'',t'')=\sum_{n=0}^{\infty}B_n(z',t';z'',t'') , \quad (46)$$

where

$$B_0(z',t';z'',t'')=1 , \quad (46a)$$

$$B_n(z',t';z'',t'')=\int_{t''}^{t'}dt'''\int_{z''}^{z'}dz'''\left[\epsilon_R I(t''')-\epsilon_s\frac{\partial I_L(t''')}{\partial t'''}\right]$$
$$\times(\exp\{\alpha z'''+\lambda\epsilon_s[I_L(t''')-I_L(t'')](q'-q''')\})B_{n-1}(z''',t''';z'',t'') . \tag{46b}$$

In the approximation considered we get the distribution of the scattered field after the one-step iteration of (38a). Then, neglecting small terms, we may write

$$A_{s1}(z',t')=A_s(0,t')+A_L(0,t')\eta(z',t') , \tag{47}$$

where

$$\eta(z',t')=\lambda\epsilon_R\int_0^{z'}dz''\{\exp[\alpha z''-(\Gamma_R/2)t']\}$$
$$\times\left[g_{10}(z'',t')+\int_0^{z''}\int_{t_0}^{t'}\left[\exp\frac{\epsilon_{BJ}}{2\omega_B}[I_L(t'')q'''-I_L(t')q'']\right]g_{10}(z''',t'')e^{\alpha z'''}\left[\epsilon_R I_L(t'')-\epsilon_s\frac{\partial I_L(t'')}{\partial t''}\right]\right.$$
$$\left.\times(\exp\{\lambda\epsilon_s[I_L(t'')-I_L(t')]q'\})B(z'',t';z''',t'')\right] \tag{47a}$$

and

$$g_{10}(z',t')=g_1(z',t')e^{-2\epsilon_k\lambda I_L(t')q'} . \tag{47b}$$

One should note that according to (15) we can limit our considerations to time periods $|(t''-t')|<2/\Gamma_R$. If the function $\exp\{\lambda\epsilon_s[I_L(t''')-I_L(t'')](q'-q'')\}$ is slowly varying within this period, i.e.,

$$|\lambda\epsilon_s[I_L(t''')-I_L(t'')](q'-q''')|\ll|\tfrac{1}{2}\Gamma_R(t''-t')| , \tag{48}$$

then $B(z',t';z'',t'')$ becomes (Appendix C)

$$B(z',t';z'',t'')=I_0\left\{\left[2\lambda(q'-q'')\int_{t''}^{t'}dt'''\left[\epsilon_R I_L(t''')-\epsilon_s\frac{\partial I_L(t''')}{\partial t'''}\right]\right]^{1/2}\right\} , \tag{49}$$

where $I_0\{\ \}$ is here the modified Bessel function of zeroth order.

B. Special solution for amplifier systems

From the experimental point of view two cases are of particular interest: the amplifier system and the generator system. In the amplifier system a weak optical wave enters the medium at point $z'=0$ and gives rise to the initial modulation of the medium,[1,2] while in the generator system the initial signal giving rise to SRLS is produced by a random fluctuation of entropy.[1] So far, however, we have no theoretical or experimental hints as to the shape of these random fluctuations involved, so one cannot determine the forcing function $g_1(z',t')$ for this case. These difficulties do not occur, however, in the case of the amplifier system, where as it follows from (26) and (44) the forcing functions take the form

$$f_1(z',t')=\bar{F}_1(0,t')=\int_{t_0}^{t'}dt''A_L(0,t'')A_s^*(0,t'')e^{(\Gamma_R/2)t''} , \tag{50}$$

$$g_1(z',t')=\bar{F}_1(0,t')\exp[i\lambda_0\epsilon_s I_L(t')q'] , \tag{51}$$

regardless of the envelope of the initial light pulses. If we substitute (50) into (33) and make use of (38) and (45), then

$$F_1(z',t')=\exp[i\lambda_0\epsilon_s I(t')q']$$
$$\times\int_{t_0}^{t'}e^{\Gamma_R(t''/2}A_L(0,t'')A_s^*(0,t'')\left[1+i\lambda_0\int_{t''}^{t'}\int_0^{z'}B(z',t';z'',t''')\left[\epsilon_R I_L(t''')-\epsilon_s\frac{\partial I_L(t''')}{\partial t'''}\right]\right.$$
$$\left.\times\exp\{\alpha z''+i\lambda_0\epsilon_s[I_L(t''')-I_L(t')]q''\}dz''dt'''\right]dt'' , \tag{52}$$

$$A_{s1}(z',t')=A_s(0,t')-i\lambda_0\epsilon_R A_L(0,t')\int_{t_0}^{t'}dt''\int_0^{z'}dz''\left[A_L^*(0,t'')A_s(0,t'')\exp\left[\alpha z''-i\lambda_0\frac{\epsilon_{BJ}}{2\omega_B}I_L(t')q''+\tfrac{1}{2}\Gamma_R(t''-t')\right]\right]$$
$$\times\left[1+i\lambda_0\int_{t''}^{t'}dt'''\int_0^{z'}dz'''B^*(z'',t'';z''',t''')\right.$$
$$\times\left[\epsilon_R I_L(t''')-\epsilon_s\frac{\partial I(t''')}{\partial t'''}\right]$$
$$\left.\times\exp\{-i\lambda_0\epsilon_s[I_L(t')-I_L(t''')]q'''\}\right] . \tag{53}$$

If condition (48) is satisfied, then

$$\bar{F}_1(z',t')=\int_{t_0}^{t'}dt''A_L(0,t'')A_s^*(0,t'')e^{(\Gamma_R/2)t''}I_0\left\{\left[2i\lambda_0 q'\epsilon_R\int_{t''}^{t'}dt'''I_L(t''')\right]^{1/2}\right\}\exp[i\lambda_0\epsilon_s I_L(t')q'] . \tag{54}$$

$$A_{s1}(z',t')=A_s(0,t')+(-i\epsilon_R\lambda_0)^{1/2}A_L(0,t')$$
$$\times\exp\left[-i\lambda_0\frac{\epsilon_{BJ}}{2\omega_B}I_L(t')q'\right]\int_{t_0}^{t'}dt''\frac{A_L^*(0,t')A_s(0,t'')e^{(\Gamma_R/2(t''-t')}}{\left[\int_{t''}^{t'}dt'''I_L(t''')\right]^{1/2}}$$
$$\times I_1\left\{\left[-2i\lambda_0 q'\epsilon_R\int_{t''}^{t'}dt'''I_L(t''')\right]^{1/2}\right\} . \tag{55}$$

Expression (55) has a form similar to that describing the distribution of the scattered field in SBS and SRS under small signal approximation.[7,9]

IV. DISCUSSION

It follows from (47a)–(47c) that in cases when the intensity of the light beam is time independent, SRLS does not occur. This fact becomes evident if we examine Eq. (18). Since ϵ_R is real,

$$\frac{\partial I_s}{\partial z'}\propto-\epsilon_R\frac{\partial\phi(z',t')}{\partial t'} , \tag{56}$$

where $\bar{F}_1(z',t')=|\bar{F}_1(z',t')|\exp[-i\phi(z',t')]$. As it is proved by (56), SRLS arises when $\epsilon_R(\partial\phi/\partial t')<0$. A detailed analysis of the optical-field distributions and the isobaric perturbations involving expressions (44) and (47) requires the knowledge of the initial conditions. This difficulty may be avoided, if one takes advantage of the fact that $3\Gamma_L<\Gamma_R$ in most experiments where SRLS was investigated.[2] In such a case we get from (44) and (43a)

$$\frac{\partial I_s}{\partial z'}\propto-\epsilon_R\left[\frac{2\epsilon_R}{\Gamma_R}+\epsilon_s\right]\frac{\partial I_L(t')}{\partial t'}q'+\cdots . \tag{57}$$

The physical explanation of (56) is fairly simple. In the stationary case, two plane waves of the same frequency produce a phase grating in perfect phase matching with each of the waves so the intensity of both waves does not change. Transfer of energy between these two beams occurs only if the phase-matching condition is not fulfilled. In view of the finite rise time of isobaric perturbations, it will take place, however, if the phases of the optical waves are time dependent. Expression (57) shows that the generation of scattered radiation takes place either on the rising or on the falling slope of the laser pulse, depending on the sign of the material parameters ϵ_R and ϵ_s. In the model presented, these parameters are a linear combination of contributions provided by electrostriction, linear absorption, the electrocaloric effect, and molecular reorientation (Kerr effect). The values of these contributions calculated for several liquids are summarized in Table I [Eqs. (14a)–(14c), (16a)–(16d), and (43)].

As it follows from (16) and Table I, the values of ϵ_R in absorbing as well as in transparent liquids are determined by the term attributed to linear absorption. The compensating influence of the coupling of modes may be significant if $\alpha<0.001\ \mathrm{cm}^{-1}$. The electrocaloric effect is of no importance here.

The dependence of ϵ_s on various mechanisms of interactions between light and the liquid is more complicated. The value of ϵ_k is related to the anisotropy of the molecules and lies in the limits from 10^{-13} esu in liquids where this anisotropy is small (i.e., CCl_4) to 10^{-10} esu in liquids where a large optical Kerr effect is observed (CS_2, nitrobenzene). The other term in (43a) gives the contributions of the coupling of modes and is of the order of 10^{-12} esu. So, in liquids with anisotropic molecules, ϵ_s of the order of 10^{-10} esu and is positive. On the other hand, in liquids consisting of isotropic molecules, ϵ_s is of the order of 10^{-12} esu and is negative.

Numerical estimations prove that the terms involving ϵ_s may be significant only in nonabsorbing liquids. If $\alpha>0.1\ \mathrm{cm}^{-1}$, the terms due to linear absorption dominate regardless of the shape of the laser pulse and the anisotropy of the molecules of the medium. Therefore, as it follows from (57), the pulse of the scattered radiation is delayed in absorbing liquids by a value of $2/\Gamma_L$ with respect to the laser pulse. This conclusion is in excellent agreement with the experimental results of Rother *et al.*[13] On the other hand, the sign of $\partial I_s/\partial z'$ is determined in transparent liquids by the anisotropy of the molecules. If $[\epsilon_s+(2\epsilon_R/\Gamma_R)]>0$, then the spectrum of scattered light is shifted to the Stokes side. It becomes evident, therefore, that STS-I is caused by the self-action of the optical wave and not by the electrocaloric effect, as was suggested by Starunov and Fabielinski.[3]

A. Phase conjugation in SRLS

From (47) it follows directly that in cases when small depletion of the laser-beam intensity occurs, the scattered radiation leaving the liquid is directed by

$$\begin{aligned}A_{s1}(L,t')=&A_s(0,t')-i\lambda_0\epsilon_R A_L(0,t')\\&\times\int_{t_0}^{t'}dt''\int_0^L dz''A_L(L,t'')A_s(0,t'')\\&\times\exp\left[\alpha z''+\tfrac{1}{2}\Gamma_R(t''-t')+i\lambda_0\frac{\epsilon_{BJ}}{2\omega_B}I_L(t')q''+i\lambda_0\epsilon_k I_L(t')q(L)\right]\\&\times\left[1-i\lambda_0\int_{t''}^{t'}dt'''\int_0^{z''}dz'''\exp\{\alpha z'''-i\lambda_0\epsilon_s q'''[I_L(t''')-I_L(t')]\}\right.\\&\left.\times\left[\epsilon_R I_L(t''')-\epsilon_s\frac{\partial I_L(t''')}{\partial t'''}\right]B(z'',t'';z''',t''')\right]+\cdots\,,\end{aligned}\tag{58}$$

where the centered dots denote terms depending on $\epsilon_j(z',t_0)$. It follows from numerous theoretical and experimental investigations of optical phase conjugation[30] that the dependence in form (58) is sufficient to make a part of the spectrum of the scattered light obey the phase front reversed with respect to the incident laser light. Usually the efficiency of the process of optical phase conjugation is characterized by the ratio of the intensities of the outcoming scattered and the incident laser waves:

$$R=\frac{I_s(L,t')}{I_L(t')}\,.\tag{59}$$

The detailed study of parameter R requires, in view of the nonstationary nature of SRLS, extensive numerical calculations. However, if we restrict our considerations to colored liquids of isotropic molecules, for nanosecond-width laser pulses we get, after dropping small terms, the approximate expressions

$$\begin{aligned}R=\Bigg|&\frac{A_s(0,t')}{A_L(0,t')}+\frac{A_s(0,t_0)}{A_L(0,t_0)}\exp[-\tfrac{1}{2}\Gamma_R(t'-t_0)]C(t',t')\\&-\int_0^{t-t_0}\left[\frac{d}{dt''}\exp(-\tfrac{1}{2}\Gamma_R t'')\frac{A_s(0,t'-t'')}{A_L(0,t'-t'')}\right]C(t',t'')dt''\Bigg|^2\,,\end{aligned}\tag{60}$$

where the function $C(t',t'')$ is defined as

$$C(t',t'')=I_0\left\{\left[-2i\epsilon_R\lambda_0 q(L)\int_0^{t''}dt'''I_L(t'-t''')\right]^{1/2}\right\}-1\,.\tag{61}$$

TABLE I. Values of the contributions provided by electrostriction, linear absorption, the electrocaloric effect, and molecular reorientation, calculated for several liquids. All values in esu. The data have been taken from Refs. 1, 2, and 4 and from references therein.

	I	II	III	IV	V	VI	VII	VIII	IX	X	XI	XII
	β^e	β^a	β^d	β^c	ϵ_T	$\chi^{(3)}$	Γ_B	Γ_R	ϵ_R[a]	ϵ_{BR}[a]	ϵ_{BJ}	$\Gamma_B\epsilon_K/2$
	(10^{-2})		(10^{-6})	(10^{-6})	(10^{-14})	(10^{-14})	(10^{-9})	(10^{-8})	(10^{-4})	(10^{-4})	(10^{-2})	(10^{-2})
Nitrobenzene	3.07	0.111α	−5.68	107.1	−2.37	90	4.30	2.35	−3.60	1.5	7.6	29.2
CS_2	4.93	0.246α	−10.32	181.0	−2.34	114	3.66	2.26	−10.54	5.1	15.3	31.4
Acetone	2.07	0.105α	+0.110	38.7	−0.016	1	1.38	1.32	−4.50	2.1	5.24	0.096
C_6H_6	2.70	0.126α	−0.515	44.3	−0.26	21	2.19	1.51	−7.40	3.7	9.23	3.46
CCl_4	3.61	0.302α	+0.041	67.6	−0.012	1.85	3.96	1.07	−9.09	4.5	5.24	0.550

[a]$\alpha=0.002\ \text{cm}^{-1}$.

If we consider, by way of example, CCl_4 colored with J_2 of concentration giving $\alpha=0.2$ cm^{-1}, then $\epsilon_R=9\times10^{-2}$, $\lambda_0=4.2\times10^3$, and $\Gamma_R=1.07\times10^8$ s^{-1}. For a 1-cm-length sample, illuminated by two laser pulses of the maximum power densities 40 and 0.01 MW/cm^2, respectively, and the width of 10 ns (full width at half maximum), owing to the same initial optical-field distribution, i.e.,

$$\frac{A_s(0,t'-t'')}{A_L(0,t'-t'')}=\frac{A_s(0,t_0)}{A_L(0,t_0)} , \tag{62}$$

we get $R\lesssim0.04$. The value of R increases, if

$$\frac{A_s(0,t'-t'')}{A_L(0,t'-t'')}=\frac{A_s(0,t_0)}{A_L(0,t_0)}\exp[i\omega(t'-t'')] ,$$
$$\omega<\tfrac{1}{2}\Gamma_R \tag{63}$$

which means that the frequency of the initial scattered field is shifted to the anti-Stokes side. For $\omega=\frac{1}{2}\Gamma_R$, which corresponds to the maximum of the steady-state gain for SRLS, we obtain $R\lesssim0.07$. These values of R are much smaller than the values of R obtained for SBS (Ref. 31) and are of the same order as the values of R obtained in experiments in which the optical phase conjugation via four-wave mixing in linear absorbing liquids was investigated.[10,11,31]

V. CONCLUSIONS

From our considerations it follows that the proposed integral-equations approach provides a new insight into the mechanism of transient SRLS. All information about the processes contributing to SRLS including the coupling of adiabatic and isobaric modes are involved in the resolvents of the linearized Volterra equations. These resolvents can be determined regardless of the initial conditions. The expressions derived on the basis of this approach show that SRLS is generated only if a phase mismatch between the laser field and the isobaric grating occurs, which happens when the duration of the laser pulse and the rise time of the grating are of the same order.

The significance of various mechanisms contributing to the generation of light-induced phase gratings depends on the value of the linear absorption coefficient. In colored liquids the local heating dominates, while the increase of transparency, electrostriction, and the optical Kerr effect become important. The hitherto unexplained shift in the spectrum of STS-I should be related to the optical Kerr effect. We also found that for certain experimental conditions a significant part of STS-II reveals a wave front reversed to the incident laser field.

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APPENDIX A: EVALUATION OF INTEGRAL EQUATION FOR E_j

Let us consider the equation

$$-\frac{\partial^2\bar{F}_j(z',t')}{\partial z'\partial t'}+\frac{ik_Le^{-\alpha L}}{4n_0^2}\left[e^{\alpha z'}I_L(z',t'-2z'n_0/c)+\left[e^{-\alpha z'}\int_0^{z'}dz''e^{2\alpha z''}\frac{\partial}{\partial z''}I_L(z'',t'-2z''n_0/c)\right]+e^{-\alpha z'}I_s(0,t')\right]$$
$$\times\left[2\frac{\partial\bar{F}_j(z',t')}{\partial t'}\epsilon_k+\sum_{l=1}^{3}[\epsilon_l(z',t_0)+\epsilon_l\bar{F}_l(z',t)]e^{(\Omega_l-\epsilon_j)t'}+\frac{2n_0}{cA_L(z',t')}\frac{\partial A_L(z',t')}{\partial t'}\frac{\partial\bar{F}_j(z',t')}{\partial t'}\right]=0 . \tag{A1}$$

If we expand

$$I_L(z',t'-2z'n_0/c)=I_L(z',t')-\frac{\partial I_L(z',t')}{\partial t'}\frac{2z'n_0}{c}+\cdots , \tag{A2}$$

eliminate the terms which are small in the meaning of (24), and take advantage of the fact that the functions $\bar{F}_j$ satisfy the formal relation following directly from (12),

$$\bar{F}_j(z',t')=e^{(\Omega_j-\Omega_l)t'}\bar{F}_1(z',t')+(\Omega_l-\Omega_j)\int_0^{t'}dt''e^{(\Omega_j-\Omega_l)t''}\bar{F}_l(z',t'') , \tag{A3}$$

then we get

$$
-\frac{\partial^2\bar{F}_j(z',t')}{\partial z'\partial t'}+\frac{ik_L e^{-\alpha L}}{4n_0^2}\left[e^{\alpha z'}I_L(z',t')+\left[e^{-\alpha z}\int_0^{z'}dz''e^{2\alpha z''}\frac{\partial}{\partial z''}I_L(z'',t')\right]+e^{-\alpha z'}I_s(0,t)\right]
$$
$$
\times\left[2\frac{\partial\bar{F}_j(z',t')}{\partial t'}\epsilon_k+\sum_{l=1}^{3}\bar{\epsilon}_l(z',t_0)e^{(\Omega_l-\Omega_j)t'}+(\epsilon_R+2\epsilon_{BR})\bar{F}_j(z',t')\right.
$$
$$
\left.+\sum_{l=1}^{3}\epsilon_l(\Omega_l-\Omega_j)\int_{t_0}^{t'}e^{(\Omega_l-\Omega_j)(t'-t'')}\bar{F}_j(z',t'')dt''\right]=0\,. \qquad \text{(A4)}
$$

After integrating (A4) and making use of Fubini's theorem we get a nonlinear Volterra equation of the form

$$
\bar{F}_j(z',t')=f_j(z',t')+\lambda\int_{t_0}^{t'}dt''\int_0^{z'}dz''K_j(z',t';z'',t'')\bar{F}_j(z'',t'')\,, \qquad \text{(A5)}
$$

where

$$
f_j(z',t')=\bar{F}_j(0,t')+\frac{ik_L e^{-\alpha L}}{4n_0^2}\int_{t_0}^{t'}\int_0^{z'}\left[e^{\alpha z''}I_L(z'',t'')+\left[e^{-\alpha z''}\int_0^{z''}dz'''e^{2\alpha z'''}\frac{\partial}{\partial z'''}I_L(z''',t'')\right]+e^{-\alpha z''}I_s(0,t'')\right]
$$
$$
\times\left[\sum_{l=1}^{3}\bar{\epsilon}_l(z'',t_0)e^{(\Omega_l-\Omega_j)t''}\right]dz''dt''\,, \qquad \text{(A6)}
$$

$$
\lambda=i\lambda_0=\frac{ik_L e^{-\alpha L}}{4n_0^2}\,, \qquad \text{(A7)}
$$

$$
K_j(z',t';z'',t'')=2\epsilon_k\delta(t'-t'')J(z'',t'')-\frac{\partial J(z'',t'')}{\partial t''}\epsilon_k+\epsilon_j J(z'',t'')
$$
$$
+\sum_{l\,(\neq j)}\epsilon_j J(z'',t')e^{(\Omega_l-\Omega_j)(t'-t'')}-\int_{t''}^{t'}\frac{\partial J(z'',t''')}{\partial t'''}e^{(\Omega_l-\Omega_j)(t''-t''')}dt'''\,, \qquad \text{(A8)}
$$

$$
J(z',t')=e^{\alpha z'}I_L(z',t')+e^{-\alpha z'}\int_0^{z'}dz''e^{2\alpha z''}\frac{\partial}{\partial z''}I_L(z'',t'')+e^{-\alpha z'}I_s(0,t'')\,. \qquad \text{(A9)}
$$

Since according to (23) the function $\partial J(z'',t'')/\partial t''$ varies slowly with respect to $\exp[(\Omega_l-\Omega_j)(t''-t''')]$, (A8) can be replaced by

$$
K_j(z',t';z'',t'')=J(z'',t'')[2\delta(t'-t'')\epsilon_k+\epsilon_j]+\frac{\partial J(z'',t'')}{\partial t''}\left[\sum_{l\,(\neq j)}\frac{\epsilon_l}{\Omega_l-\Omega_j}-\epsilon_k\right]
$$
$$
+\sum_{l\,(\neq j)}\epsilon_l\left[J(z'',t')-\frac{\partial J(z'',t')}{\partial t'(\Omega_l-\Omega_j)}\right]\exp[(\Omega_l-\Omega_j)(t'-t'')]\,. \qquad \text{(A10)}
$$

APPENDIX B: TRANSFORMATION OF AN INTEGRAL EQUATION BY INFINITE ITERATION

Let us consider the equation

$$
F(z,t)=f(z,t)+a(t)\int_0^z d\tilde{z}\,e^{\alpha\tilde{z}}F(\tilde{z},t)+\int_{t_0}^{t}\int_0^z b(\tilde{t})e^{\alpha\tilde{z}}F(\tilde{z},\tilde{t})d\tilde{z}\,d\tilde{t}\,. \qquad \text{(B1)}
$$

If the forcing function is written in the form

$$
g(z,t)=f(z,t)+\int_{t_0}^{t}d\tilde{t}\int_0^z d\tilde{z}b(\tilde{t})e^{\alpha\tilde{z}}F(\tilde{z},\tilde{t})\,, \qquad \text{(B2)}
$$

Eq. (B1) can be written as

$$
F(z,t)=g(z,t)+a(t)\int_0^z d\tilde{z}\,e^{\alpha\tilde{z}}F(\tilde{z},t)\,. \qquad \text{(B3)}
$$

After infinite iteration of Eq. (B3) we get a series

$$
F(z,t)=g(z,t)+\sum_{n=0}^{\infty}G_n(z,t)\,, \qquad \text{(B4)}
$$

where

$$
G_0(z,t)=a(t)\int_0^z d\tilde{z}\,e^{\alpha\tilde{z}}g(\tilde{z},t)\,, \qquad \text{(B5)}
$$
$$
G_n(z,t)=a(t)\int_0^z d\tilde{z}\,e^{\alpha\tilde{z}}G_{n-1}(\tilde{z},t)\,. \qquad \text{(B6)}
$$

From (B5) and (B6) it follows that $G_n(z,t)$ has the form

$$
G_n(z,t)=\int_0^z d\tilde{z}\,e^{\alpha\tilde{z}}\frac{(q-\tilde{q})^n}{n!}a(t)^n\,, \qquad \text{(B7)}
$$

where $q=\int_0^z d\tilde{z}\,e^{\alpha\tilde{z}}$. For $n=1$,

$$
G_1(z,t)=\int_0^z d\tilde{z}\,e^{\alpha\tilde{z}}\int_0^{\tilde{z}}d\bar{z}\,e^{\alpha\bar{z}}g(\bar{z},t)=\int_0^z d\bar{z}\,g(\bar{z},t)e^{\alpha\bar{z}}(q-\bar{q})\,. \qquad \text{(B8)}
$$

The latter transformation in (B8) is the consequence of Fubini's theorem. Let us suppose that (B7) is true for n; for $n+1$, then, (B6) gives

$$G_{n+1}(z,t)=a(t)\int_0^z d\tilde{z}\,e^{\alpha\tilde{z}}\int_0^{\tilde{z}} d\bar{z}\,a^n(t)\times\frac{(q-\bar{q})^n}{n!}e^{\alpha\bar{z}}g(\bar{z},t)$$

$$=\int_0^z d\tilde{z}\,g(\tilde{z},t)a^{n+1}(t)\int_{\tilde{z}}^z d\bar{z}\,e^{\alpha\bar{z}}\frac{(q-\bar{q})^n}{n!}$$

$$=\int_0^z d\tilde{z}\,g(\tilde{z},t)\frac{(q-\tilde{q})^{n+1}}{(n+1)!}a^{n+1}(t)\,. \qquad \text{(B9)}$$

Relations (B4), (B8), and (B9) imply that

$$\sum_{n=0}^{\infty}G_n(z,t)=a(t)\int_0^z e^{\alpha\tilde{z}}e^{a(t)(q-\tilde{q})}g(\tilde{z},t)d\tilde{z}\,. \qquad \text{(B10)}$$

By substituting (B2) and (B10) into (B4) we finally get

$$F(z,t)=f(z,t)+a(t)\int_0^z d\tilde{z}\,e^{\alpha\tilde{z}}e^{a(t)(q-\tilde{q})}f(\tilde{z},t)+\int_{t_0}^t\int_0^z b(\tilde{t})e^{a(\tilde{t})(q-\tilde{q})}F(\tilde{z},\tilde{t})d\tilde{z}\,d\tilde{t}\,. \qquad \text{(B11)}$$

APPENDIX C: RESOLVENT FOR LIQUIDS REVEALING SMALL ac KERR EFFECT

Let us consider the function $B(z',t';z'',t'')$ if the condition

$$|\lambda\epsilon_s[I_L(t''')-I_L(t'')](q'-q''')|\ll 1 \qquad \text{(C1)}$$

is fulfilled. This assumption means that in all expressions the phase factor $\exp\{\lambda\epsilon_s[I_L(t''')-I_L(t'')](q'-q''')\}$ provides only a small contribution of the value of B_n. Therefore, we can approximately write

$$B_n=\int_{t''}^{t'}dt'''\int_{z''}^{z'}dz'''\left[\epsilon_R I(t''')-\epsilon_s\frac{\partial I(t''')}{\partial t'''}\right]\times S_{n-1}(z''',t''';z'',t'')e^{\alpha z'''}\,. \qquad \text{(C2)}$$

Since

$$B_0=1\,,$$

from (B2) it follows immediately that

$$B_1=(q'-q'')\int_{t''}^{t'}dt'''\left[\epsilon_R I(t''')-\epsilon_s\frac{\partial I_L(t''')}{\partial t'''}\right],$$

$$B_n=\frac{(q'-q'')^n\left[\int_{t''}^{t'}dt'''\left[\epsilon_R I(t''')-\epsilon_s\frac{\partial I_L(t''')}{\partial t'''}\right]\right]^n}{(n!)^2}\,.$$

Making use of (46) we obtain finally

$$B(z',t';z'',t'')=\sum_{n=0}^{\infty}\frac{\lambda^n(q'-q'')^n\left[\int_{t''}^{t'}dt'''\left[\epsilon_R I(t''')-\epsilon_s\frac{\partial I_L(t''')}{\partial t'''}\right]\right]^n}{(n!)^2}$$

$$=I_0\left\{\left[2\lambda\int_{t''}^{t'}dt'''\left[\epsilon_R I_L(t''')-\epsilon_s\frac{\partial I_L(t''')}{\partial t'''}\right](q'-q'')\right]^{1/2}\right\}, \qquad \text{(C3)}$$

where $I_0\{\ \}$ denotes the modified Bessel function of the zeroth order.

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