Self-Induced Stimulated Light Scattering

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The laser-induced collective rotation of molecules in a liquid crystal is treated as a novel stimulated light-scattering process in which the Stokes shift is not constant but is induced by a pump field. The solution following the usual coupled-wave approach appears as a nonlinear eigenvalue problem and exhibits bistable behavior.

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We present in this paper a theoretical discussion on a novel stimulated light-scattering process in which the material excitation has a frequency depending on the pump laser intensity. In a recent publication,¹ it was demonstrated that when a sufficiently intense circularly polarized laser beam is normally incident on a liquidcrystal medium, the output becomes elliptically polarized; the polarization ellipse rotates with a constant angular velocity that depends on the input laser intensity. This phenomenon arises because the laser beam transfers parts of its angular momentum to the medium and causes the molecules in the medium to rotate. In doing so, the beam must also lose energy to the medium. Since the medium is transparent, the energy loss must appear in the form of a red shift on part of the beam. This red shift was also measured.² We realize that the rotation of the polarization ellipse at an angular velocity Ω can be understood as being the result of the presence of both right and left circular polarization components in the medium, one with the incoming laser frequency ω and the other with the shifted frequency $\omega - 2\Omega$. Obviously, the component at $\omega - 2\Omega$ must have been generated by the pump beam via the laser-induced molecular rotation in the medium. Such a process is generally known as stimulated light scattering but, in the present case, since the rotation is induced by the pump field with Ω depending on the laser intensity, we call it self-induced stimulated scattering. In contrast to the usual stimulated scattering, the present process involves a material excitation which is not an intrinsic resonant excitation of the medium but is induced by the pump field, and in the process, only the first Stokes component is generated, the higher-order Stokes and all anti-Stokes components being suppressed. Although self-induced stimulated scattering has only been observed in liquid crystals, we expect that the same process could exist in all transparent birefringent media whose molecules are free to rotate.

We can use the usual coupled-wave approach to describe such a stimulated scattering process. Let us consider a liquid-crystal film in a mesophase with its average local orientation of molecules defined by the unit director $\hat{\mathbf{n}} = (\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta)$, where θ is the angle between $\hat{\mathbf{n}}$ and the surface normal $\hat{\mathbf{z}}$ and ϕ is the azimuthal angle of $\hat{\mathbf{n}}$ with respect to the x axis lying in the film. Initially, $\hat{\mathbf{n}}$ is uniform throughout the film. In the presence of a normally incident laser beam, $\hat{\mathbf{n}}$ can be reoriented as a function of z. We assume here that only the azimuthal angle ϕ can be varied. This corresponds, for example, to the case of the smectic-C film with its layers parallel to the film faces; an extremely strong field would be required to change the angle θ appreciably.

The wave equations for the right and left circularly polarized components, $E_R = (E_x + iE_y)/\sqrt{2}$ and $E_L = (E_x - iE_y)/\sqrt{2}$, of a plane wave propagating in the liquidcrystal film along z can be shown to have the form

$$\frac{\partial^2}{\partial z^2} \begin{pmatrix} E_R \\ E_L \end{pmatrix} = \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \begin{pmatrix} \epsilon & \epsilon_a Q^2 \\ \epsilon_a Q^{*2} & \epsilon \end{pmatrix} \begin{pmatrix} E_R \\ E_L \end{pmatrix}, \quad (1)$$

where

$$\epsilon = n_0^2 (1 - \frac{1}{2} u^2 \sin^2 \theta) / (1 - u^2 \sin^2 \theta),$$

$$\epsilon_a = \frac{1}{2} u^2 n_0^2 / (1 - u^2 \sin^2 \theta),$$

$$u^2 = 1 - n_0^2 / n_e^2,$$
(2)

and

$$Q = \sin\theta e^{i\phi},\tag{3}$$

 n_0 and n_e being the ordinary and extraordinary refractive indices of the liquid-crystal medium, respectively. It

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is seen in Eq. (1) that E_R and E_L are coupled via the Q^2 and Q^{*2} terms, but Q (or ϕ) can actually be varied by the applied optical field. The equation of motion for ψ comes from the balance of optical, elastic, and viscous torques in the medium, namely

$$\alpha \partial \phi / \partial t = \tau_z + k \, \partial^2 \phi / \partial z^2, \tag{4}$$

where α is the effective viscosity coefficient, k the effective elastic constant, and τ_z the optical torque per unit volume on the medium along z.³ Since the time-averaged optical torque on an anisotropic medium is

$$\tau = [(n_e^2 - n_0^2)/8\pi] \operatorname{Re}[(\hat{\mathbf{n}} \cdot \mathbf{E}^*)(\hat{\mathbf{n}} \times \mathbf{E})], \qquad (5)$$

we can show that

$$\tau_z = -\left(\epsilon_a/4\pi\right) \operatorname{Im}(E_R^* E_L Q^2). \tag{6}$$

To solve the coupled equations (1) and (4) for a steady-state solution, we first reduce them to first-order differential equations. Let

$$E_{\beta} = (\omega_{\beta}/\sqrt{\epsilon})^{1/2}G_{\beta}(z)\exp(ik_{\beta}z - i\omega_{\beta}t),$$

with $k_{\beta} = \omega_{\beta} \sqrt{\epsilon}/c$ ($\beta = R, L$) and $Q = q(z) \exp(-i\Omega t)$ with $q(z) = \sin\theta \exp[i\psi(z)]$. Then the slowly-varyingamplitude approximation in Eq. (1) gives $\omega_R - \omega_L = 2\Omega$ and

$$dG_R/dz = \frac{1}{2} igq^2 G_L \exp(ik_L z - ik_R z),$$

$$dG_L/dz = \frac{1}{2} igq^{*2} G_R \exp(ik_R z - ik_L z),$$
(7)

where $g = (\omega_R \omega_L / \epsilon)^{1/2} (\epsilon_a / c)$. With the help of the above equations, the expression for τ_z in Eq. (6) can be written as

$$\tau_z = dL_z/dz, \tag{8}$$

with $L_z = (c/8\pi)(|G_R|^2 - |G_L|^2)$. We recognize that $L_z(z)$ is actually the angular momentum flux carried by the beam at position z, and therefore Eq. (8) simply describes the usual relation between torque and angular momentum. Equation (4), with $\phi = \psi(z) - \Omega t$ and τ_z given by Eq. (8), can then be integrated to yield

$$kd\psi/dz + (c/8\pi)(|G_R|^2 - |G_L|^2 + C) + \alpha \Omega z = 0,$$
(9)

with C an integration constant. Since $d\psi/dz = (dq/dz)/ig$, we have

$$dq/dz + (ic/8\pi k)(|G_R|^2 - |G_L|^2 + C)q + (ia \Omega/k)zq = 0.$$
(10)

Our stimulated light-scattering process is now fully described by the nonlinearly coupled set of equations (7) and (10). Note that the anti-Stokes and higher-order Stokes fields never appear in the equations.

For a right circularly polarized input beam with intensity I and frequency ω the boundary conditions are $|G_R(0)| = (8\pi I/\omega c)^{1/2}$ and $G_L(0) = 0$ at z=0. The conservation of the total number of photons in such a scattering process requires $|G_R(z)|^2 + |G_L(z)|^2 = |G_R(0)|^2$, which can be easily obtained from Eq. (7). For the molecular reorientation, we notice that the term $k d\psi/dz$ at the boundaries z=0 and z=d actually describes the mechanical torques exerted by the liquid-crystal molecules on the sample walls. If the molecules are free to rotate at the walls, then we should have $d\psi/dz=0$ at z=0 and z=d. The former condition leads to the determination of the integration constant $C=-|G_R(0)|^2$ in Eq. (9) or (10). The latter condition yields

$$\alpha \Omega d = L_z(0) - L_z(d) = (c/4\pi) |G_L(d)|^2, \quad (11)$$

which describes the conservation of the angular momentum flux. The elastic torques are absent in the above relation because they come from internal forces. With the ellipticity of the beam polarization given by the Stokes parameter

$$s_3 = (|G_R|^2 - |G_L|^2) / (|G_R|^2 + |G_L|^2), \quad (12)$$

Eq. (11) can be written as

$$\Omega = (I/\omega a d) \Delta s_3, \tag{13}$$

where $\Delta s_3 = 1 - s_3(d)$ is the change of ellipticity suffered by the beam in traversing the sample. The above equation shows that Ω is directly proportional to Δs_3 , both being easily measurable quantities.

Equations (7) and (10) can now be solved with the initial conditions $G_R(0) = (8\pi I/\omega c)^{1/2}$, $G_L(0) = 0$, $q(0) = \sin\theta$ [with $\psi(0)$ chosen as 0], and a prescribed value of Ω . In particular, the ellipticity change Δs_3 across a sample thickness *d* can be calculated. In view of the constraint of Eq. (13), this means that Ω cannot have any arbitrary value. In other words, the solution of Eqs. (7) and (10) must be consistent with Eq. (13). Therefore, what we have here is a sort of nonlinear eigenvalue problem with Ω being the eigenvalue which depends on *I* and the process itself.

Analytical solution of the problem is difficult, but numerical solution is straightforward. A standard Runge-Kutta subroutine can be used to integrate Eqs. (7) and (10) for given I and d. The function $s_3(\Omega)$ and hence $\Delta s_3(\Omega)$ can be calculated for various Ω . The result, coupled with Eq. (13), then allows us to find the values of Δs_3 and Ω , either numerically or graphically.

In the limit of small intensity *I*, Eqs. (7) and (10) can be solved analytically, yielding $s_3(d) \approx \cos(gd)$ and $\Omega \approx \{[1 - \cos(gd)]/\omega ad\}I$. We see that Ω starts to increase linearly with *I* for small *I*, unless $\cos(gd) = 1$, which corresponds to a sample of optical thickness $n\lambda$, with integer *n*. In this case, for an undistorted sample, the polarization of the beam remains unchanged in traversing the medium and no angular momentum is transferred. Therefore, for an optical thickness of $n\lambda$, Ω



FIG. 1. (a) Angular velocity Ω as a function of the light intensity I for a sample having an optical thickness of λ ($d=116 \mu$ m). (b) Angular velocity Ω as a function of the light intensity I for a sample having an optical thickness of $\lambda/2$ ($d=58 \mu$ m).

starts with a term quadratic in the bean intensity I.

As an example, we consider the Ar⁺-laser optical wavelength (515 nm) and a liquid-crystal medium with typical material constants $\theta = 10^{\circ}$, $k_{22} \approx k_{33} = 10^{-7}$ dyne, and $\alpha = 1$ P. The calculated results of Ω vs *I* for a sample having an optical thickness of λ ($d = 116 \mu$ m) and $\lambda/2$ ($d = 58 \mu$ m) are shown in Figs. 1(a) and 1(b). The corresponding curves of $s_3(d)$ vs *I* are given in Figs. 2(a) and 2(b).

For the λ sample, it is seen that, starting with $s_3 = 1$ and $\Omega = 0$, both $|\Delta s_3|$ and Ω first increase with increasing *I*. As previously anticipated, Ω grows from 0 with a term quadratic in the intensity *I*. Then, as s_3 approaches its lowest possible value -1, Ω also approaches its maximum. This is expected since maximum Ω should correspond to the maximum net deposition of angular momentum of light into the medium. The maximum of Ω , how-



FIG. 2. (a) Output polarization ellipticity s_3 as a function of the light intensity *I* for a sample having an optical thickness of λ . The ellipticity of the input beam is $s_3(0) = +1$. (b) Output polarization ellipticity s_3 as a function of the light intensity *I* for a sample having an optical thickness of $\lambda/2$. The ellipticity of the input beam is $s_3(0) = +1$.

ever, is reached at an intensity higher than the maximum of Δs_3 because Ω is proportional to $I\Delta s_3$. With further increase of *I*, the ellipticity s_3 changes back to +1 and Ω correspondingly decreases to 0, and then the cycle starts over again. When the laser intensity is sufficiently high, s_3 and Ω become multivalued functions of *I*. As seen in Figs. 1(a) and 1(b), this clearly happens already in the second cycle. Intrinsic optical bistable operations can result in such multivalued portions of the curves. It should be stressed, however, that the transitions shown in the figures connect time-dependent limit-cycle states of the system and not fixed states, as in usual optical bistability. The intensity required to observe the bistable behavior is in the range of a few kilowatts per square centimeter, which can be easily obtained from a focused cw

Ar⁺ laser beam.

A cyclic behavior is also seen in the function $\Omega(I)$ for the $\lambda/2$ sample [Fig. 1(b)], but now the multivalued behavior appears at lower intensity. The branch starts linearly from $\Omega = 0$. This is expected, since in this case we have a large angular momentum transfer at low intensity, because of the linear birefringence of the sample. As seen from Figs. 1(b) and 2(b), switching from the first branch to the second branch occurs at ≈ 7.15 kW/cm² as I increases, and back to the first branch at ≈ 0.5 kW/cm² as I decreases.

In summary, we have described a self-induced stimulated light-scattering process in which the Stokes frequency shift is not a characteristic constant of the medium, but depends on the pump intensity. The solution of the coupled-wave equation appears as a nonlinear eigenvalue problem and can exhibit bistable behavior at sufficiently high pump intensity. The description should apply to the process of laser-induced collective rotation of molecules in a liquid-crystalline medium. Such an effect has been experimentally observed in a nematic film. This latter case is, however, more complicated because both the polar angle θ and the azimuthal angle ϕ of the molecules can be reoriented by the pump field. When the variation of θ is small, one could indeed observe an azimuthal rotation of molecules with a constant velocity $\Omega(I)$ in effective agreement with the theory presented here.

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¹E. Santamato, B. Daino, M. Romagnoli, M. Settembre, and Y. R. Shen, Phys. Rev. Lett. **57**, 2423 (1986).

²E. Santamato, B. Daino, M. Romagnoli, M. Settembre, and Y. R. Shen, Mol. Cryst. Liq. Cryst. **143**, 89 (1986).

³The effective constants α and k are related to Leslie's viscosity coefficient γ_1 and Franck's elastic constants k_{22} and k_{33} by $\alpha = \gamma_1 \sin^2 \theta$ and $k = (k_{22} \sin^2 \theta + k_{33} \cos^2 \theta) \sin^2 \theta$ [see, for example, P. G. de Gennes, *The Physics of Liquid Crystals* (Oxford Univ. Press, Oxford, 1974)].