Ring instability of wave front of light beams with regular temporal modulation

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A study has been made of those aspects of a time-varying, small-scale self-focusing of light beams which result from a regular temporal modulation of the beam intensity. In this case, the nonlinear excitation of acoustic waves with frequencies which are multiples of the light modulation frequency is suppressed, while the noisy waves are amplified as a result of the perturbation of the density of the medium by both heating and electrostriction. The result is the appearance of a ring structure in the angular distribution of the scattered light.

One important mechanism in the nonlinear energy dissipation of an intense light wave is the small-scale self-focusing, i.e., the spatial modulation of the field as a result of a nonlinear amplification of noisy waves of the same frequency. In the steady state, this small-scale self-focusing is observed only in media which have a positive cubic susceptibility $\chi^{(3)}$. In this case, it corresponds to an exponential amplification of noisy waves which are propagating at certain small angles with respect to the original plane wave.[1] In this letter we are reporting a study of a time-varying small-scale self-focusing which can occur in media with either $\chi^{(3)}>0$ or $\chi^{(3)}<0$ ("time-varying" here means with respect to the onset of the nonlinear response). For light beams with a regular temporal modulation of the intensity, e.g., for a train of picosecond pulses, it is found that the nonlinear amplification of the noisy waves due to the perturbation of the density of the medium is suppressed for a certain sequence of directions. Spatial struc-

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tures of this type were apparently first observed in Ref. 2.

Let us examine the stability of a plane light wave with respect to small perturbations of the wave front. To describe the variations in the amplitude of the optical field, we use the Helmholtz equation

$$[\Delta + \frac{\omega^2}{c^2} (\epsilon + \delta\epsilon(\mathbf{r}, t))]E(\mathbf{r}, t) = 0. \tag{1}$$

We assume that the perturbations of the dielectric constant, $\delta\epsilon(\mathbf{r},t)$, are caused deviations of the temperature (δT) and the density ($\delta\rho$) from their average values T and ρ as a result of a heating of the medium and the ponderomotive force resulting from the variations in the electric field of the light wave: $\delta\epsilon = (\partial\epsilon/\partial T)_\rho \delta T + (\partial\epsilon/\partial\rho)_T \delta\rho$. To analyze the stability of a plane wave $E_0(t)\exp(ikz - i\omega t)$ which is incident on the medium in the cross section $z = 0$, we write the amplitude of the perturbed wave in the form $E(\mathbf{r},t) = E_0(t)[1 + u_q(z,t)\exp(i\mathbf{qr})]$, $u_q(z=0,t) = u_0$, $|\mathbf{k} + \mathbf{q}| = |\mathbf{k}|$. In the approximation linear in u_q, we then find the following equation from Eq. (1) and from the hydrodynamic equations for $\delta\rho$ under very unsteady conditions, after a linearization with respect to δT and $\delta\rho$ (there is no relaxation of $\delta\rho$ or δT):

$$\frac{\partial}{\partial z} u_q(z, t) = i \frac{\omega}{2c\epsilon^{1/2}} \left(\frac{\partial\epsilon}{\partial\rho}\right)_T \delta\rho(z, t), \tag{2}$$

$$\left(\frac{\partial^2}{\partial t^2} + \Omega_q^2\right)\delta\rho(z, t) = \frac{\alpha\beta c\epsilon^{1/2}}{8\pi C_V} \Omega_q^2 \int_0^t |E_0(t')|^2 u_q(z, t')dt' + \rho\left(\frac{\partial\epsilon}{\partial\rho}\right)_T \frac{q^2}{16\pi} |E_0(t)|^2 u_q(z,t), \tag{3}$$

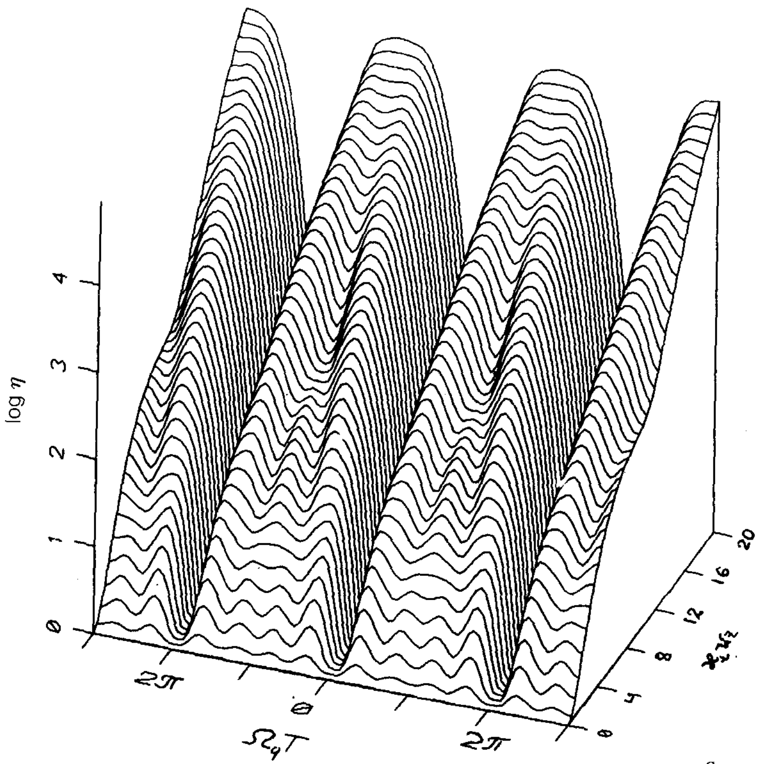
where $\Omega_q = qv$, v is the sound velocity, ρ and C_V are the density and heat capacity of the medium, α is the optical absorption coefficient, and β is the bulk expansion coefficient. Following Ref. 3, we have omitted from system (2), (3) small terms with $(\partial\epsilon/\partial T)_\rho$, which correspond to an electrocaloric effect.

Let us assume that a train of short pulses with a repetition period T (from a mode-locked laser) is incident on the medium. Assuming that the length of an individual pulse in the train satisfies $t_p \ll T, \Omega_q^{-1}$, we approximate the time evolution of the light intensity by a sequence of δ-pulses: $|E_0(t)|^2 = (8\pi w/Nc\epsilon^{1/2})\Sigma_{j=1}^N \delta(t - jT)$, where w is the energy flux density in the light wave. Integrating Eq. (3), and substituting the expression found for $\delta\rho$ into Eq. (2), we then find

$$\frac{\partial}{\partial z} u_q(z, nT) = \frac{iw}{N} \sum_{j=1}^{n-1} u_q(z, jT)\{\kappa_t[1 - \cos(\Omega_q T(n-j))] + \kappa_e \sin(\Omega_q T(n-j))\}, \tag{4}$$

where $\kappa_t = \alpha\beta\omega(\partial\epsilon/\partial\rho)_T/2c\epsilon^{1/2}C_V$ and $\kappa_e = \omega\Omega_q\rho(\partial\epsilon/\partial\rho)_T^2/4v^2c^2\epsilon$. Since the energy flux density of the noisy waves is determined by the perturbation amplitudes $u_q(z,t)$ at the times $t = nT - u_q(z,nT)$ $(n = 1,...,N)$, we have restricted Eq. (4) to those times. For perturbations with $\Omega_q T = 2\pi m$ (where m is an integer), the right side of Eq. (4) vanishes; i.e., these perturbations are not amplified. This result in turn means that density perturbations corresponding to sound waves with frequencies which are multiples of the pulse repetition frequency in the train do not grow in the medium. This result can be explained at a qualitative level as follows: A short light pulse excites density perturbations with a wide frequency spectrum and a common initial phase in the medium. This initial phase is determined by the time of arrival of the exciting light. Those frequency components of the perturbations for which the wave amplitude of the density oscillation crosses zero at the time at which each successive light pulse arrives at the medium do not generate a corresponding angular component in the scattered light.[4] Since there are no density perturbations in the medium at the time of arrival of the first exciting light pulse, the frequencies of such components are multiples of the pulse repetition frequency.

Equation (4) has been solved numerically for $\kappa_e = 0$ and arbitrary values of Ω_q. Figure 1 shows the effective perturbation growth rate η, defined by $\eta = \Sigma_{j=1}^{N} |u_q(z,jT)|^2 / N|u_0|^2$, as a function of the parameters $\Omega_q T$ and $\kappa_t wz$ for (a) $N = 5$ and (b) $N = 21$. These results, as well as results calculated for other values of



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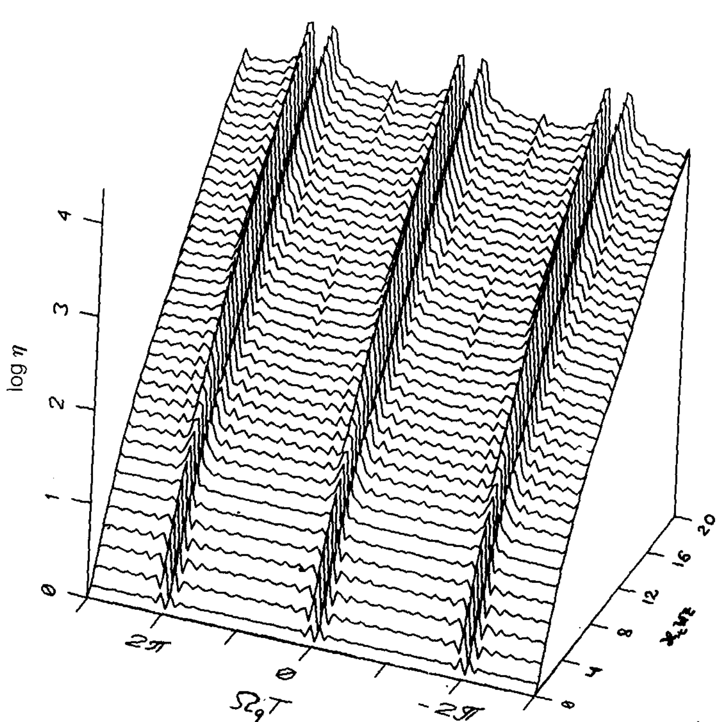

b

FIG. 1. Effective growth rate of the noisy waves, η, versus the parameters $\kappa_t wz$ and $\Omega_q T$ for (a) $N=5$ and (b) $N=21$.

N, show that the maximum value of η is determined by the energy of the train, w, and is essentially independent of the number of pulses in the train (N). In contrast, the angular dependence of η, which is determined by the parameter $\Omega_q T(\Omega_q T = 4\pi\epsilon^{1/2} vT \sin(\vartheta/2)/\lambda$, where λ is the wavelength of the light, and ϑ is the propagation direction of a noisy wave, depends on the particular number (N) of pulses at which a given perturbation growth rate, say $\eta \approx 10^3$, is reached.

Working from system (2), (3), we can also study the effective perturbation growth rate in the case in which the intensity of the light incident on the medium is not modulated. Omitting the time derivative from Eq. (3) for $t_p \gg \Omega_q^{-1}$, and setting $\kappa_e = 0$, we find $u_q(z,t) = u_0 J_0(2(-i\kappa_t w(t)z)^{1/2})$, where $w(t)$ is the energy density of the light which has passed through the medium by the time t. Using an asymptotic representation of the Bessel function $J_0(\xi)$ for $\kappa_t w(t)z \gg 1$, we find $\eta \approx \exp[2(2\kappa_t w(t)z)^{1/2}]/4\pi(\kappa_t^{1/2} w(t)z)^{1/2}$. Estimates of η from this expression yield values close to those found in the case of a train of pulses.

For an experimental study of this time-varying, small-scale self-focusing we used the second harmonic of the beam from a $YAlO_3$:Nd^{2+} laser ($\lambda = 0.54\ \mu m$) with three types of output: a single pulse with a length $t_p \approx 80$ ns (one longitudinal mode); a train of pulses each ≈ 20 ps long with a repetition period $T = 7.4$ ns; or two neighboring longitudinal modes. In the latter case the light was sinusoidally modulated with a period equal to the axial period of the resonator ($T \approx 7.4$ ns). The collimated laser beam, with a diameter $d \approx 0.5$ mm (and a divergence $\approx 10^{-3}$ rad), entered a cell $L = 1$ cm thick filled with one of the following liquids: nitrobenzene, phenyl salicylate, aniline, solutions of iodine in acetone and ethyl alcohol, and a solution of rhodamine 6G in ethyl alcohol. The optical absorption coefficient in the pure liquids was $\alpha \approx 10^{-2}$ cm^{-1}, and the transmission of the solutions was $\approx 50\%$.

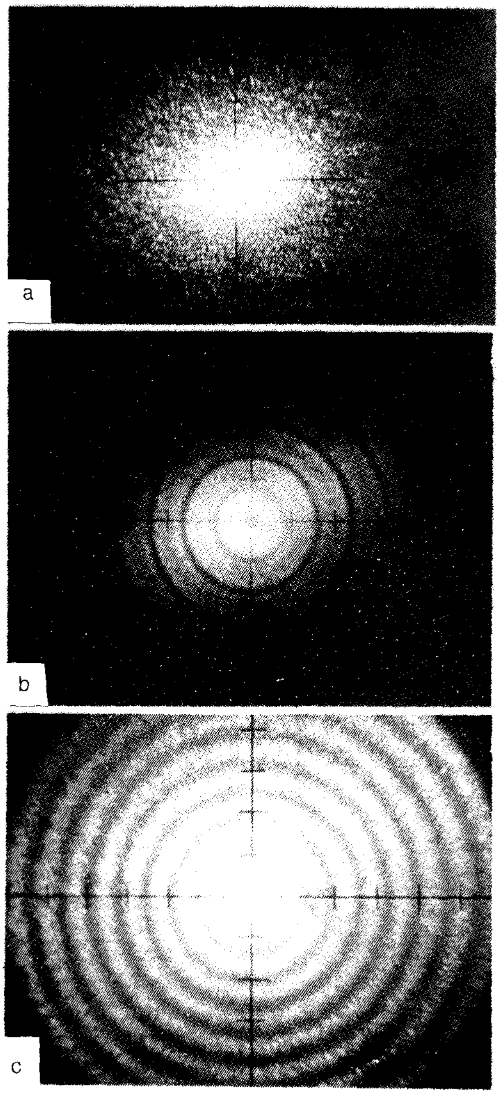

FIG. 2. Transverse distribution of the scattered light which appears during the passage of (a) a single pulse or (b,c) a train of picosecond pulses through a cell filled with (a,b) a solution of iodine in ethyl alcohol of (c) nitrobenzene. The scale division is 0.1 rad.

As the single pulse passed through the liquid-filled cell, a pronounced diffuse scattering into a solid angle ≈ 1 rad, with a smooth angular distribution, began at an energy ≈ 1 mJ [Fig. 2(a)]. For the pulse train, however, we observed a clearly defined ring structure with narrow minima in directions specified by $\vartheta'_m = m\lambda/vT$, $m = 1,2,...$ in the spatial distribution of the scattered light [Figs. 2(b) and 2(c)], although the scattering efficiency and the solid angle into which the light was scattered remained the same. As the energy of the laser beam was increased, the ring structure formed after the first three or four pulses of the train, according to a time sweep of the scattered light. The angular width of the minima increased substantially [Fig. 2(c)]. We observed a similar picture—similar in terms of both the spatial distribution of the scattered light and the efficiency of the excitation of this scattering—when the laser beam intensity was sinusoidally modulated with a depth close to 100%.

Let us estimate the value of the parameter $\kappa_t wL$, which determines the effective growth rate of the noisy waves, and the relative component of the perturbation of the density of the medium due to electrostriction, κ_e/κ_t. For the ethyl alcohol [$\rho = 0.78$ g/cm^3, $\rho(\partial\epsilon/\partial\rho)_T = 0.99$, $C_V = 2.4$ J/(g·K), $\epsilon = 1.8$, and $\beta = 1.1\times10^{-3}\cdot$K^{-1}], for example, we have $\kappa_t = 26\alpha$ cm^{2}J^{-1} and $\kappa_e/\kappa_t \approx 10^{-2}/\alpha$ ($[\alpha] = $ cm^{-1}). At $\alpha \approx 1$ cm^{-1}, we can thus ignore the electrostriction $\kappa_e/\kappa_t \approx 10^{-2}$, and at a train energy ≈ 1 mJ we have $\kappa_t wL \approx 13$. This figure corresponds to an effective growth rate $\eta = 5\times10^2$–10^3; i.e., the instability of the beam should be manifested even at an initial noise level $\approx 10^{-4}$.

As we have already mentioned, a ring structure of this sort arising in the scattered light during the passage of a train of picosecond pulses through a weakly absorbing medium has been observed previously.[2] However, both the onset of the scattering and the appearance of structures of this type were attributed to the excitation of sound waves with a period equal to the product of the sound velocity and the pulse repetition period, vT, in the medium. It follows from our study that the scattering is not itself associated with the periodic modulation of the laser beam. On the contrary, the temporal modulation suppresses the scattering at a certain discrete set of angles, since sound waves with lengths vT/m ($m = 1,2,...$) are not excited.

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