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D. N. Ghosh Roy
University of Massachusetts Boston

D.V.G.L.N. Rao
University of Massachusetts Boston, raod@umb.edu

H. Bronk
University of Massachusetts Boston

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Optical pulse compression in a cholesteric liquid crystal

D. N. Ghosh Roy,[a] D. V. G. L. Narasimha Rao, and H. Bronk
Physics Department, University of Massachusetts at Boston, Boston, Massachusetts 02125

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A 20-ns laser pulse is compressed to nearly 2.5 ns in a 10-cm-long sample of liquid-crystal cholesteryl oleate in the isotropic phase. Pulse compression in a length as short as only 5 cm has been observed. A semiquantitative explanation is given in terms of stimulated Brillouin scattering.

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In recent years compression of optical pulses has received considerable attention, especially for applications in laser fusion.[1,2] Pulse compression by backward-stimulated Raman (SRS),[3,4] and Rayleigh wing scattering (SRWS)[5] is considerably more established than that by backward-stimulated Brillouin scattering (SBS). Controlled pulse compression by SBS has been very recently demonstrated[6] in methane (CH_4) gas at 130 atm in a 1.3-m-long tapered glass tube. In this letter we report backward SBS compression of a 20-ns ruby laser pulse to about 2.5 ns in the isotropic phase of the liquid-crystal cholesteryl oleate using a length of 10 cm only.

A schematic of the experimental arrangement is shown in Fig. 1. A water-cooled, passively Q-switched, giant pulse ruby laser operating in a single mode with peak power in the range of 1-20 MW and pulse width of 20 ns was used as the optical pump. The cholesteryl oleate obtained from Aldrich Chemical Company was 97% pure and had a cholesteric to isotropic transition at 49 °C. The cell was kept sufficiently distant from the ruby laser to avoid any regenerative Brillouin feedback. Signals were monitored by a fast pulse photodetector with an S-1 photocathode and 0.5-ns rise time and displayed on a Tektronix 519 oscilloscope. Energy in the laser pulse was measured by a ballistic thermopile connected to a Keithely microvoltammeter.

Figures 2(a) and 2(b) show the incident laser and a typical backscattered pulse for a 10-cm cell focussed by a 10-cm lens, respectively. Figure 2(c) shows a Fabry-Perot interferogram for a sample temperture of 51 °C. The backscattered pulses were highly compressed with a compression ratio of about 10. For reasons discussed below, the observed pulse compression was believed to be due to backward SBS.

No evidence of filament formation and self-focusing of the incident beam in the sample was found, and a thorough investigation revealed no Raman shift. Backward SRS usually occurs under self-focusing conditions[3] at the power levels used in this experiment. In generator-type experiments, as in the present case, stimulated thermal Brillouin scattering (STBS) is usually not observed.[7] The occurrence of SRWS was considered unlikely as it is usually accompanied by a band emission in the backscattering. Of the remaining two competing scattering processes, namely SBS and stimulated thermal Rayleigh scattering (STRS), the latter was improbable for the following reasons.

First, an analysis of the Fabry-Perot interferograms of the backscattered radiation showed a Stokes shift in frequency, the magnitude of the shift decreasing with increasing temperature (0.252 cm^{-1} at 51 °C and 0.191 cm^{-1} at 99 °C). This is typical of SBS, but not of STRS or of SRWS processes. Moreover, the shifts observed were orders of magnitude larger than those of the absorption-assisted STRS, a typical value for which is 10^{-3} cm^{-1} (Ref. 7). Second, STRS in generator-type experiments is not observed if the linear optical absorption coefficient α of the scattering medium is less than the critical absorption coefficient α_{cr} defined as[7]

$$\alpha_{cr} = (\gamma_e C_p \omega_B / 2\beta n c v_p^2)\,(\Gamma_R + \Gamma_L)/(\Gamma'_B + \Gamma_L). \qquad (1)$$

In Eq. (1), γ_e is the electrostrictive constant, C_p the specific heat at constant pressure, ω_B the Brillouin shift, β the coefficient of volume expansion, n the refractive index, Γ_R and Γ_L the Rayleigh and the laser linewidth, respectively. An approximate estimate of the critical absorption coefficient is obtained using Eq. (1). γ_e may be approximated by[8] $(n^2-1)(n^2+2)/3$ using the Lorentz-Lorenz relation. The hypersound speed for backward scattering is given by $v_p = c\omega_B/2n\omega_L$, c/n being the speed of light in the medium and ω_L the frequency of the incident laser beam. The linewidth $\Gamma'_B = \Gamma_B + (\gamma-1)/2\Gamma_R$, where Γ_B is the Brillouin linewidth and γ is the ratio of specific heats. We may assume $\Gamma'_B \approx \Gamma_B$, since $\Gamma_R \ll \Gamma_B$. Using the observed values of Brillouin shift from SBS and Brillouin linewidth from spontaneous Brillouin scattering,[9] the value of α_{cr} comes out as 0.23 cm^{-1} at 51 °C which is larger than our observed value for the linear absorption coefficient $\alpha = 0.15$ cm^{-1} at the ruby laser wavelength. Furthermore, the estimated gain for this experiment using Eq. (3) is nearly an order of magnitude smaller than the gain typical of an STRS process. SBS appears to be the only nonlinear process going in the backward direction and causing the observed pulse compression.

An estimation of phonon lifetime was made by using the following relation proposed by Tang[10]:

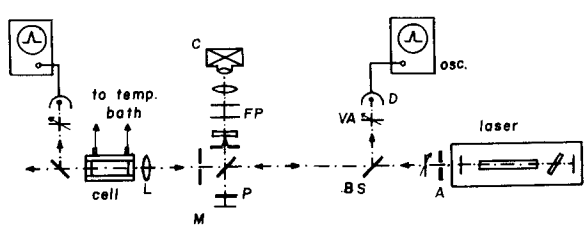

FIG. 1. Schematic of the experimental arrangement. M: mirror, L: lens, BS: beam splitter, VA: variable attenuator, FP: Fabry-Perot etalon, C: camera; A: aperture, P: λ/4-plate.

[a]Present address: Consiglio Nazionale Delle Ricerche, 73 Strada delle Cacce, Torino 10135, Italy.

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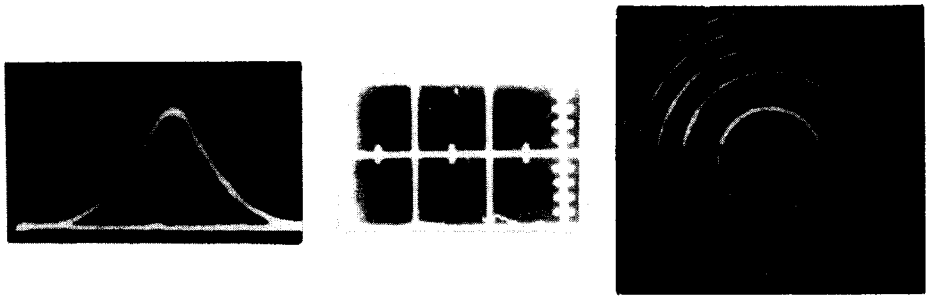

(a) (b) (c)

FIG. 2. (a) Incident laser pulse, 20-ns/cm scale. Power = 10 MW. (b) Typical backscattered pulse. 10-ns/cm scale. Power = 2 MW. (c) Fabry-Perot interferogram for 51 °C. The upper half shows ruby and backscattered radiation and the lower half ruby alone.

$$N_S(0) = (\Delta\Omega\omega_s{}^3 kT/8\pi^3 c^2 h\omega_s\omega_B)\alpha_p v_p \times [1n\, 2/N_L(0)gL]^{1/2} \exp\{N_L(0)gL\} \quad (2)$$

In Eq. (2), $N_{S,L}(0)$ are the photon flux densities of the Stokes or laser light at the entrance window, related to the amplitudes $E_{S,L}$ of the Stokes or laser wave by $N_{S,L} = (cn/8\pi h\omega_{S,L})||E_{S,L}||^2$; $\Delta\Omega$ the solid angle subtended by the primary beam at the sample, ω_s the frequency of the backscattered radiation, k the Boltzmann constant, T the temperature, g the gain coefficient, and α_p the phonon damping constant. α_p was calculated from Eq. (2) for 10-cm sample length using observed intensities of incident and backscattered beams. The phonon lifetime is estimated as 0.41 ns from the relation $\tau_p = 2/\alpha_p v_p$. A direct experimental measurement[9] of thermal Brillouin scattering at 90° angle using the 5145-Å green line of an argon-ion laser with a multipass interferometer yielded a value 760 MHz for the Brillouin linewidth. Considering the dependence on phonon wave vector and incident laser wavelength, it translates to 417 MHz for this experiment leading to a phonon lifetime 0.38 ns in close agreement with the estimated value.

The gain coefficient g is given by

$$g = \mu\omega_s\gamma_e^2 k_p/2\varepsilon k\alpha_p, \quad (3)$$

where μ, ε are the permeability and the dielectric constant of the medium respectively and k_p the phonon wave vector. The elastic constant of the medium is given by $k = 1/\rho_0 v_p^2$, ρ_0 being the density. Equation (3) gives $g = 0.01$ cm/MW using α_p obtained from Eq. (2).

A semiquantitative explanation of the observed pulse compression can be given in terms of the mechanism first proposed by Maier *et al.*,[3] and Culver *et al.*,[9] for SRS and subsequently used for SRWS.[5] After the incident radiation reaches a threshold value inside the Brillouin active medium, SBS process is initiated. As the SBS pulse sweeps backwards, its leading edge "sees" the incoming light beam, interacts with it, and grows as its expense. The trailing edge, on the other hand, is in the "shadow" and consequently grows considerably less rapidly. This leads to a dramatic sharpening of the pulse to a width of $\delta T \simeq ln/c$, if the full interaction length l is accessible.

Under the condition of Bragg reflection, an acoustic wave has a reflectivity r given by[11] $r = \tan h^2 [\pi l\sqrt{MI_0}/\sqrt{2}\ \lambda]$, where I_0 is the acoustic power density and $M = n^6 p_{12}^2/\rho_0 v_p{}^3$ the acoustic-optic figure of merit, the pockel constant p_{12} being γ_e/n^4. From this relation for r and for phonon lifetime longer than the width of the laser pulse, a semiclassical argument shows[6] that the interaction length $l = (\lambda c/2n^4 p_{12})(\rho_0 v_p/P)^{1/2}$, where $P = r'P_L$, r' being the SBS efficiency and P_L the power density of the incident laser beam. For phonon lifetime shorter than the pulse width, as in the present study, a qualitative estimate of l can be made by replacing P with P' where $P' = P\exp(-T/\tau_p)$, where $T = nl/c$. For our experiment with the 10-cm-long cell, the interaction length was estimated to be nearly 40 cm, which should give a limiting pulse width of 2 ns. Thus, in our case, the SBS process did not obtain the full interaction length resulting in backscattered pulses slightly wider than expected.

It is interesting that we have observed pulse compression in cholesteryl oleate in a sample lengths as short as only 5 cm. More detailed studies are in progress and the results will be published elsewhere.

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