OBSERVATION OF SELF-PHASE MODULATION AND SMALL-SCALE FILAMENTS IN CRYSTALS AND GLASSES

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Frequency broadening and small-scale filaments were observed in calcite, quartz, sodium chloride, and several glasses under picosecond pulse excitation. The physical mechanism responsible for these processes is the change in refractive index resulting from electronic distortion.

This Letter reports the first direct measurement of frequency broadening in crystals and glasses. We have observed self-modulation of the broadened spectra and have identified the broadening mechanism as an electronic self-phase modulation. We have also observed small-scale filament formation in solids due to electronic self-trapping.

In the experiment high-power picosecond pulses at a wavelength of 5300 Å were either reduced in area by an inverted Galilean telescope or focused into solids. Under high powers the output spectra of the solids are shifted thousands of wave numbers to the red and blue and show modulation. A similar effect of self-phase modulation from filaments was first observed by Shimizu[1] in liquid CS_2 due to the orientational Kerr effect. In a Nd-glass laser, Treacy[2] interpreted the picosecond laser pulses as being linearly frequency swept since he was able to compress the pulses by passing them through two gratings. Fisher and Fleck[2] have shown that more complex sweeping can lead to similar two-photon fluorescence results. The nonlinear susceptibility coefficients in solids are ~10 to 10^3 times less than in liquids, but with high powers available from mode-locked picosecond lasers combined with electronic self-trapping the change in the refractive index of solids is large enough to cause an observable sweep.

An optical pulse traveling through a medium with an intensity-dependent refractive index becomes distorted in phase (self-phase modulation) and in envelope shape (self-steepening). The effect of envelope-shape distortion is negligible[1,3,4] while self-phase modulation has been shown to produce an optical-frequency broadening in liquids over several hundred wave numbers in extent. Mechanisms in liquids which give rise to an intensity-dependent refractive index are the orientational Kerr effect, electrostriction, molecular redistribution, librations, and electronic distortion. In suitably chosen liquids[5] these frequency-broadening mechanisms may be distinguished from the electronic mechanism through their different time responses. Typical calculated relaxation-time responses for diffusional motions are ~10^{-12} sec while the electronic distortion response time is ~10^{-16} sec.[5] Thus with picosecond light pulses Brewer and Lee have shown that the dominant mechanism for filament formation should be electronic in very viscous liquids. Recently, however, molecular librations have been suggested as causing broadening and trapping in CS_2.[6] In solids, mechanisms giving rise to an intensity-dependent refractive index for picosecond pulse excitation are electronic distortion, electronic lattice distortion, and electronic molecular libration. The electrostriction mechanism[5,7] is rejected because it exhibits a negligible effect for picosecond pulses.

The Nd-glass mode-locked laser beam, Q switched with type 9860 Kodak dye, was passed through a 2-cm potassium-dihydrogen-phosphate (KDP) second-harmonic generator (SHG) crystal. The laser consisted of a Brewster-Brewster cut $7\frac{1}{2}$-in.$\times\frac{1}{2}$-in. Owen Illinois glass rod (ED-2), a Korad (K-1) laser head, a 10-m 100% reflectivity rear dielectric mirror, a 50% dielectric wedge output mirror (~10′), and a Kodak Q-switch dye cell. The laser output at 1.06 μ was typically ~5×10^9 W which was converted to ~2×10^8 W at the harmonic. The laser output consisted typically of 14 intense pulses separated from each other by the cavity round-trip time of 5.5 nsec. Pulse widths of 4 psec were measured by the two-photon fluorescence technique.

The SHG beam from KDP was reduced in size to a collimated 1.2-mm-diam beam across the sample by an inverted telescope. The intensity distribution of the light at the exit face of the sample was magnified 10 times and imaged on the 2-mm slit of a Jarrell Ash grating $\frac{3}{4}$-m spectrograph. Hence the spectrum of each individual filament was displayed. Usually there were five to ten filaments. A thin quartz-wedge beam split-

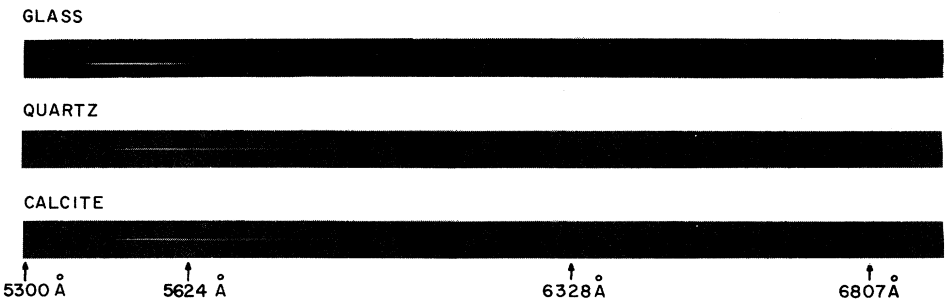

FIG. 1. Stokes frequency sweeps in flint glass, quartz, and calcite with a focused laser beam. Solid exit face was not focused on the slit.

ter was used in order to photograph filament formation at the exit face simultaneously. In observations of the Stokes (anti-Stokes) side of the spectra, three type 3-68 and three type 3-67 (two type 5-60) Corning filters were used to prevent the 5300-Å direct laser light from entering the spectrograph. Spectra were taken on Polaroid type 57 film or Kodak type 1N plates.

Spectra taken from all samples showed modulation in filaments. The modulation ranged from as small as a few wave numbers to hundreds of wave numbers. The filament size was approximately 20 μ. Typical Stokes sweeps from these filaments were 4400 cm^{-1} in a calcite crystal of length 4 cm, 3900 cm^{-1} in a quartz crystal of length 4.5 cm, 1100 cm^{-1} in extra-dense flint glass of length 7.55 cm, 3900 cm^{-1} in NaCl of length 4.7 cm, and 4200 cm^{-1} in both borosilicate crown (BK-7) and light barium crown (LBC-1) glass of length 8.9 cm. Sweeps on the anti-Stokes side were typically to 6100 cm^{-1} in calcite, 5500 cm^{-1} in quartz, 7300 cm^{-1} in NaCl and 7400 cm^{-1} in BK and LBC glasses. The anti-Stokes sweeps from the flint glass were not observed probably due to the two 5-60 filters used.

We have also observed a much weaker Stokes sweep of ~1000 cm^{-1} from the KDP SHG crystal itself, and on one intense laser pulse a modulated anti-Stokes sweep of thousands of cm^{-1} was detected. To insure that the sweeps in the other materials were not complicated by the KDP sweep, a 5300-Å narrow-band-pass filter ($\Delta\lambda$ = 100 Å) was placed after the KDP crystal. Sweeps in the other materials remained unchanged. Figure 1 shows the sweeps from calcite, quartz, and flint glass taken with a 25-cm lens focusing the 5300-Å light into the samples to a spot size of ~50 μ. The length of sweep for the focused beam was similar to that of the collimated beam except in flint glass where it was necessary to focus the beam to see the sweep.

Spectra taken with the collimated beam and with the filaments imaged on the slit were often complicated. The filaments that were in focus always showed modulation. Figure 2 shows a modulated spectrum from calcite. Some spectra show parabolic structure with apexes near the center of the spectra with the parabolas going toward the red for Stokes and toward the blue for anti-Stokes. Typical filament spectra show an aperiodic structure similar to that observed by Shimizu[1] although periodic structures similar to those observed by Lifsitz and Grieneisen[3] were sometimes also observed.

In a calcite crystal the threshold for the sweep is lower than the threshold for stimulated Raman scattering (SRS) when the laser travels as an extraordinary (E) wave, but the sweep threshold is higher than the SRS threshold when the laser travels as an ordinary (O) wave. The SRS threshold is lower for the laser traveling as an O wave than for an E wave by ~3. The thresholds for the sweep for E and O waves are ~3 and 6 GW/cm^2, respectively. The length of the frequency sweep is not changed when the collection angle was varied from 0.5 to 10 deg indicating that no parametric fluorescence effect is involved. The sweep is also polarized in the direction of the incident laser polarization. This is also true for unstrained NaCl and glasses. Damage was occasionally observed in calcite but since the emission is modulated, collimated, and polar-

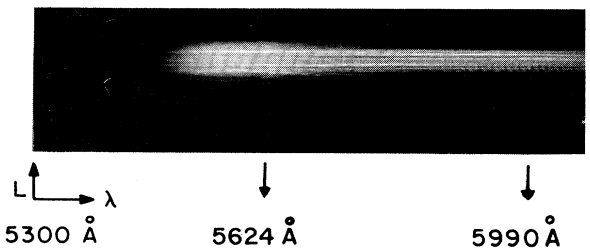

FIG. 2. Spectrum showing self-phase modulation from calcite for filaments imaged on the slit.

ized it is not produced from dielectric breakdown.

For relaxation time much less than the laser pulse width, the approximate frequency sweep $\delta\omega$ can be calculated for an isotropic material from[8,9]

$$\delta\omega \sim 2\Delta\omega n_2 k E_0^2 l, \quad (1)$$

where $\Delta\omega$ is the spectral width of the pulse, n_2 is the third-order nonlinear refractive index, E_0 is the field amplitude, k is the propagation constant, and l is the distance the light propagates through the sample. However, for an anisotropic crystal the sweep equation is more complicated than (1) because of the tensor form of the susceptibility χ_3 and the polarization of the optical field. Since the third-order susceptibility coefficients are not well known in crystals, only a rough estimate of the sweep span can be made using Eq. (1). Using n_2 for calcite[10] of $\sim 10^{-14}$ esu, a field strength $\sim 2.7\times 10^5$ esu, and beam waist length of 0.15 cm, the calculated sweep for calcite is ~ 4500 cm^{-1}. A change in refractive index corresponding to a 20-μ-diam filament[11] is $\gtrsim (1/8n_0)(1.22\lambda/d)^2 = 7\times 10^{-5}$.

Electronic librations are not possible in NaCl, and electronic lattice distortions are ruled out in glasses. However, all electronic mechanisms can contribute in calcite and quartz. Kaminow and Johnston[12] have discussed relative contributions of direct electronic and electronic lattice distortions in crystals.

With a large power density, $\sim 10^4$ GW/cm^2, in the filaments, multiphoton effects such as four-photon processes may be taking place.

The large frequency sweeps obtained in crystals indicate that these materials might be used as ultrafast light gates with response times of $\sim 10^{-15}$ sec, an improvement in response of 10^3 over the molecular-orientation light gate introduced by Duguay and Hansen.[13] Self-phase-modulation processes are probably present inside mode-locked lasers.[14]

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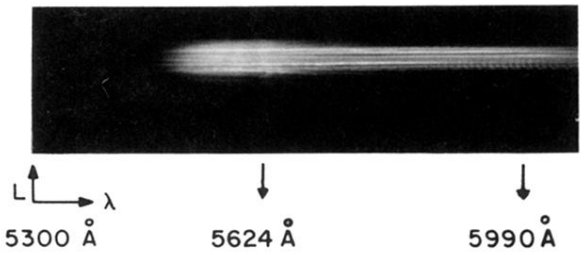

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