of the 2^1S_0 state by a factor of $\sim 10^3$, according to the magnitude of the admixture of 2^3P_1 in 2^1P_1 states [see Ref. 2, pp. 234-235, and R. C. Elton, Astrophys. J. 148, 573 (1967)].

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PHASE MODULATION OF Q-SWITCHED LASER BEAMS IN SMALL-SCALE FILAMENTS*

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It has been observed[1-3] that the spectra of small-scale trapped filaments of laser light in carbon disulfide and other liquids contain discrete bands of frequencies extending to either side of the laser frequency. These bands may be several to several tens in number, with a total frequency spread of a few tens to a few hundreds of wave numbers, as shown in Fig. 1. The regularity of these patterns, and yet the lack of a fixed frequency between bands[3] as expected for various modulation processes, have been puzzling. It is found that the patterns observed correspond to the intensity envelope of an underlying structure of equally spaced sidebands. Their basic features are explained by a more or less periodic variation in intensity of the laser light and the resulting variation of the index of refraction in the filament. The simplest case, which can be best compared with experimental results, involves the admixture with the laser light of a very weak wave of slightly different frequency, giving a sinusoidal phase modulation.

Consider two plane waves of amplitudes E_0 and E_1 and differing in frequency by $\omega_S \ll \omega_0$ traveling through a nonlinear medium where the index of refraction is $n = n_0 + \Delta n \approx n_0 + n_2 E^2$. The electric field is

$$E(z,t) = [E_0 e^{i(\omega_0 t - nk_0 z)} + E_1 e^{i(\omega_1 t - nk_1 z)}], \quad (1)$$

where

$$\omega_i / k_i = c \text{ and } \omega_1 = \omega_0 + \omega_S.$$

The beating between the two frequencies ω_0 and ω_1 will modulate the index of refraction at a frequency ω_S and introduce a time-varying optical path length, and hence a varying phase shift $\Delta\varphi(z,t)$. In the case of a nonzero relaxation time τ of the nonlinear index of refraction,

$$\Delta n(z,t) = \frac{n_2}{\tau}\int_{-\infty}^{t} \langle E^2(t')\rangle e^{-(t-t')/\tau} dt' = n_2\left(\frac{E_0^2 + E_1^2}{2}\right) + \mathrm{Re}\left\{\frac{n_2 E_0 E_1}{[1+(\omega_S\tau)^2]^{1/2}} \exp[i\omega_S t - i\arctan(\omega_S\tau)]\right\}.$$

Hence

$$\Delta\varphi(z,t) = \frac{2kzn_2E_0E_1}{[1+(\omega_S\tau)^2]^{1/2}} \mathrm{Re}\{\exp[i\omega_S t - i\arctan(\omega_S\tau)]\}.$$

The Fourier spectrum, after the filament has traversed a distance L, is given by

$$S(L,\omega) = (cn/8\pi)|E(L,\omega)|^2. \quad (2)$$

The situation described is somewhat similar to a variety of cases of phase modulation. Lallemand[4] treated the case of two strong unperturbed waves separated by a small frequency and modulating a third much weaker wave. Smith[5] has considered a case closer to the self-modulation which will be discussed below, although he included modulation of a single wave only. It has also been shown[6,7]

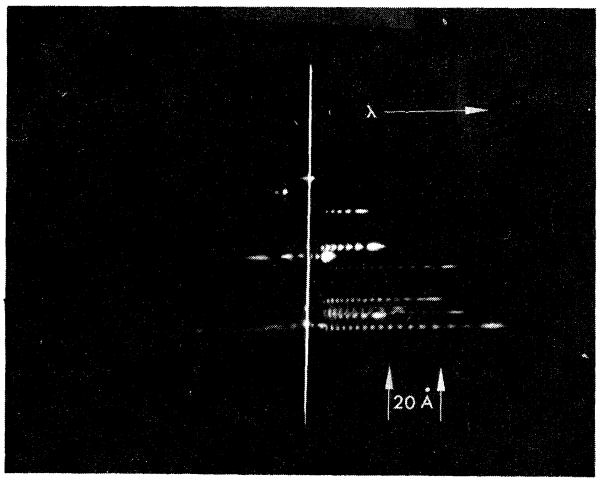

FIG. 1. Spectra of small-scale trapped filaments in liquids. Note: A narrow neutral density filter was placed over the film to reduce intensity at the laser frequency by a factor 5.

that a nonlinear index of refraction gives a frequency shift, on the slope of a light pulse, of

$$\Delta\omega = \omega_0(n_2L/n)(\partial E^2/\partial z),$$

where $\partial E^2/\partial z$ is the spatial rate of change of the intensity envelope. Such a shift is due to the change with time in optical length because of intensity variation, and hence might be called a pseudo-Doppler effect. Shimizu[3] suggested that such effects, also describable as phase modulation, produce the spectra like those of Fig. 1, but associated them with a single pulse of energy and the frequencies separating bands with a difference in the shifts $\Delta\omega$ on either side of the pulse. However, a single pulse produces a continuous spectrum, and it will be seen below that the present model of periodic modulation fits the simpler observed spectra rather well.

If the index of refraction has no time lag, the phase of the term in E_1 lags that of E_0 by $\frac{1}{2}\pi$, so that there are no cross terms of the type E_0E_1, and the spectral intensity of light emerging from a filament can be obtained from (1) and (2) as

$$S(L, \omega_0 - m\omega_s) = (cn/8\pi)[E_0^2 J_m^2(\alpha) + E_1^2 J_{m+1}^2(\alpha)], \quad (3)$$

where

$$\alpha \equiv 2k_0 L n_2 E_0 E_1. \quad (4)$$

For small intensity modulation, $E_1 \ll E_0$, a symmetric series of M sidebands ($M \simeq \alpha$) separated by ω_S is generated on both sides of ω_0 with the intensity of the mth sideband proportional to $J_m^2(\alpha)$. A resulting set of sidebands for $\alpha = 50$ is shown in Fig. 2(a), where it may be seen that the intensity variations are semiperiodic, giving the appearance of sidebands with slowly varying spacing. The spectra of

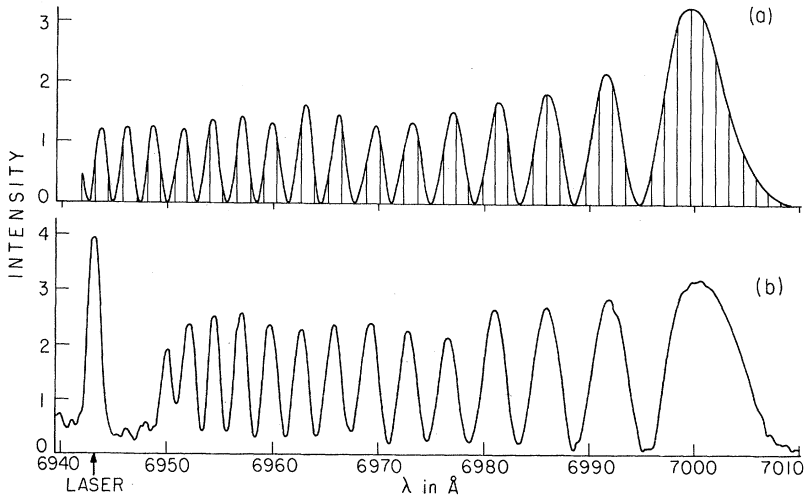

FIG. 2. (a) Calculation of the Stokes sidebands for $\alpha = 50$, $\omega = 2.50$ cm^{-1}, from Eq. (2). Individual sidebands and the intensity envelope are both shown. (b) Microphotometer trace showing the Stokes sidebands of a typical filament. [Best fit with calculated spectrum has been obtained by shifting the entire spectrum 2.5 cm^{-1}; this shift is presumably due to the laser pulse shape.]

typical filaments show reasonably good quantitative agreement with expression (3) for the Stokes-shifted sidebands as shown in Fig. 2(b). However, the anti-Stokes sidebands of the filaments are observed to be much weaker than the Stokes. Also they generally correspond to a different modulation frequency and total extent. Such asymmetries indicate that the modulation of the laser light is not completely sinusoidal.

For E_1 and E_0 of comparable size, both terms of the series in (3) are important, and the intensity pattern due to variation of $J_m^2(\alpha)$ is somewhat smoothed out. However, the last peaks are not much changed, since $J_m^2(\alpha)$ changes slowly with m for m comparable with α. The total extent of a filament's sidebands can be obtained from expression (3), and is close to $\alpha\omega_S$ in frequency units. Hence, the maximum frequency shifts in the spectra of filaments produced by laser pulses of constant amplitude should be proportional to the length of the liquid cell. Spectra taken in carbon disulfide with cell lengths from 5 to 20 cm agree well with this conclusion. The laser power used (a few megawatts) gave a trapping distance in carbon disulfide of about 5 cm. A 7.5-cm cell produced spectra up to 100 wave numbers in length. With increasing cell length, the length of the sidebands increased to 100 or 200 cm^{-1} beyond the first Raman line, or close to 1000 cm^{-1}.

While Eq. (3) clearly does not contain all details observed in the spectra of small-scale filaments, it does represent a very good first approximation to the Stokes spectra and therefore allows a quantitative analysis of the sidebands in terms of α and ω_S. It can be seen that the period of the spectrum close to the laser frequency is $2\omega_S$ and from a more detailed analysis it can be demonstrated that $\alpha = 3.2$ times the number of intensity maxima from the laser frequency to the last sideband. From expressions (3) and (4) for α, E_1/E_0 may be estimated. For typical filaments in a 10-cm. cell of CS_2, $\alpha = 30$ to 100. The size of the filaments gives a nonlinear index change of $\Delta n \approx 10^{-2}$ due to E_0, and hence from the value of α, $E_1/E_0 \sim 10^{-2}$.

The modulation frequency ω_S shows considerable variability even in filaments in CS_2, $1.75 < \omega_S < 4$ cm^{-1}. The individual sideband components separated by ω_S are not resolvable in the simple type of spectra depicted in Figs. 1 and 2. In these cases ω_S is not a sharp frequency and therefore must correspond to a spread of frequencies with a central maximum component at ω_S. For a particular filament the values of ω_S determined from a measurement of α and $2\omega_S$ determined from the distance between intensity maxima near the laser line agree to within 20%. In some filaments with spectra showing more complicated intensity envelopes and a total extent corresponding to a large phase modulation (α = several hundred) the individual sideband components separated by ω_S are resolvable. The values of ω_S determined from these filaments agree very well with those observed in simpler but unresolved spectra.

A nonzero relaxation time τ introduces a lag between susceptibility and the electric field. Therefore the trailing edge of the response will be distorted and the intensity envelope is no longer symmetrical. Calculations show that as τ is increased, the anti-Stokes spectrum becomes longer and less intense. The correlation between these calculations and the observed anti-Stokes spectra is only qualitative since their periodicities do not agree. For CS_2, $\omega_S\tau$ is estimated[8] to be ~1, which does not modify the Stokes spectrum greatly.

Self-steepening of the modulation envelope occurs at the same time as phase modulation, and is part of the same phenomenon. It will distort the pulse such that the trailing edge will steepen while the leading edge will become shallower. Thus one expects an increase in intensity of the Stokes-shifted light and the extent of the anti-Stokes sidebands. In particular, the centroid of the spectrum is shifted to the Stokes side. The fractional amount of steepening approximately equals the fractional frequency modulation, and hence in some cases it would not have a large effect.

A combination of a finite relaxation time and pulse steepening could account for the asymmetry in many of the filaments. However, in some others, the anti-Stokes sidebands are observed to be shorter than the Stokes sidebands which seems to indicate that other mechanisms are involved in the distortion of the modulation envelope.

Calculations show that the asymmetry in intensity is much more sensitively dependent on finite relaxation times and deviations from sinusoidal modulations than is the pattern of the Stokes sidebands.

We have as yet been unable to determine the

process which generates the basic frequency ω_S. The most obvious possible process is the beating of laser modes. However, the observed frequencies do not correlate with our mode structure and different frequencies occur even with the same laser pulse. Typically we have several laser modes spaced by from 0.2 to 0.4 cm^{-1}. The effect does seem to depend on the modes to the extent that it is not produced with our laser operating in a single mode. Lifsitz,[9] however, has been able to produce this type of frequency broadening with an apparently single-mode laser operating at a higher power than that which we can obtain. It seems most probable that the frequency is generated by the molecules of the liquid. The Brillouin frequency is much too small to explain ω_S. Possible mechanisms which could give frequencies as large as a few reciprocal centimeters are the stimulated Rayleigh scattering due to damped rotation, and a molecular pendular motion in which the polarization of the molecules causes them to librate in the strong electric field of the filament. Such a motion is anharmonic, but its frequency for molecules approximately aligned with the field is

$$\omega_L = |E|(\alpha/2I)^{1/2},$$

where α = anisotropic polarizability, E = electric field, I = moment of inertia. The libration would generate a light wave differing in frequency from the laser by $2\omega_L$. This model involves the same molecular motion as Rayleigh scattering, but is a resonant oscillation with damping rather than a viscously damped rotation without potential energy. In small-scale filaments where the electric fields are as large as 10^5 esu, these librational frequencies can be as high as a few wave numbers, and hence larger than the Rayleigh frequency. The latter is τ^{-1}, where τ is the damping time. ω_S has been measured in CS_2 over a temperature range of -10 to 40°C. Over this range the Rayleigh frequency should change by 75% in normal liquid outside of the filamentary region where high fields and compression exist. There was no significant change in the average value of ω_S for filaments with this temperature change, although there was, as expected, a decrease of α with increasing viscosity. Experiments have also been performed in which carbon disulfide was mixed with nitrobenzene, benzene, toluene, and carbon tetrachloride. In all cases ω_S was found to vary about a mean of 3 cm^{-1}. We were unable with power levels available to produce long sidebands in other pure liquids. However, measurements on some short sidebands in toluene and benzene ($\alpha = 10$ to 15) yielded an ω_S which was not much different from that in CS_2.

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†Alfred P. Sloan Research Fellow.

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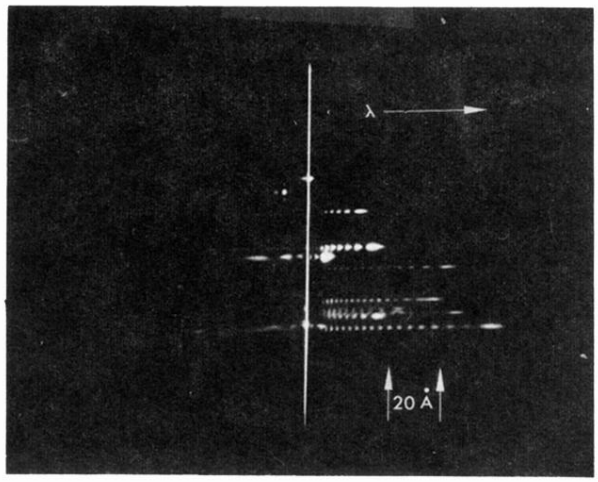

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