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Self-Modulation, Self-Steepening, and Spectral Development of Light in Small-Scale Trapped Filaments

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The spectral broadening associated with light propagating in self-trapped filaments through liquids with large optical Kerr constants is studied. In particular, we treat the influence of a nonzero orientational relaxation time and of linear dispersion upon the phase (and amplitude) development of the light as it interacts with the optically nonlinear medium. Relaxation introduces Stokes–anti-Stokes asymmetry, even in the absence of pulse steepening. The spectrum is compressed, and the degree of interference in various portions of the spectrum is altered. The effect of dispersion is apparently much less important, particularly in the case where propagation distances are short compared with the shock distance. However, dispersion combined with a finite relaxation time does introduce an exponential gain in the forward direction. For a small nonlinearity, the peak gain is equal to the stimulated Rayleigh gain in the backward direction; but it falls off with increasing nonlinearity, because of the Stokes–anti-Stokes interaction.

Spectra computed for a picosecond pulse and for a 100% sinusoidally modulated light beam of infinite extent are compared. Because of its periodicity, the latter possesses a fine structure and is influenced differently by the orientational relaxation.

Comparison of the experimental results with the theoretical calculations for the cases of a zero and a nonzero relaxation time indicates that pulses of the order of 5–10 psec in extent could give rise to the observed spectra. Possible sources of such pulses (or a sequence of such pulses) are discussed.

I. INTRODUCTION

The propagation of optical pulses and other complex wave forms through media with an intensity-dependent index of refraction results in both a self-steepening of the envelope and a phase perturbation driven by the nonlinear dielectric change (which can be called self-phase modulation). These processes, particularly that of self-phase modulation, produce sidebands[1–3] which have been observed[3–7] in the frequency spectra of small-scale trapped filaments of laser light.[8]

The process of self-steepening and the development of a shock in the envelope have been discussed theoretically for the dispersionless case both for zero and finite relaxation times.[9] In particular, it was found that the relaxation inhibits but does not prevent self-steepening. Spectra were also computed for the relaxationless case ($\tau = 0$) assuming propagation distances of the order of the shock distance.

The present paper consists of an extension of this work. An attempt is made to relate the results to those found experimentally by Grieneisen and Sacchi,[4] and Brewer,[5] as well as to those found both experimentally and theoretically by Schimizu,[6] and by Cheung *et al.*[7] Cheung, *et al.* indicated that the down-shifted spectral envelope could be produced by a sinusoidal modulation of laser intensity induced by a process within the liquid. We show that the spectra resulting from single pulses 5–10 psec in extent exhibit a similar envelope without the discrete components present in the sinusoidal case.

Spectra are computed for single pulses in the presence of either a zero or nonzero relaxation time, and for a modulated signal in the presence of finite relaxation. The results indicate that relaxation without dispersion gives rise to a large Stokes–anti-Stokes asymmetry and that dispersion serves to accent this asymmetry.

The characteristics of the light-pulse propagation are based upon a travelling wave equation obtained from a Fourier analysis of the wave equation, using a general approach in which dispersion is easily included. We shall show that neglecting the dispersion, the complex equation reduces to those of Ref. 9.

We also investigate the influence of dispersion upon the propagation of two weak-waves, one up-

shifted and one downshifted from a strong central component at frequency ω_0 and wave-vector $k_0 = \omega_0[\epsilon(\omega_0)]^{1/2}/c$. It is found that relaxation combined with a positive second-order dispersion ($\partial^2 k_0/\partial\omega_0^2 > 0$) can provide exponential gain for weak waves traveling in the same direction as the strong wave. For self-trapped filaments in CS_2, this gain is estimated to peak at about $\Omega = 50/\tau$, τ being the orientational relaxation time. No gain is present for a zero relaxation time.

II. LIGHT-PULSE EQUATION

We concern ourselves with materials which have a dielectric coefficient dependent upon the intensity of the electric field (e.g., Kerr liquids). We assume that the displacement D can be written in the form

$$D(t) = \int_0^t f(t-t')E(t')\,dt' + \delta\epsilon(t)E(t), \tag{1}$$

where $\delta\epsilon(t)$ is the intensity-dependent part of the dielectric coefficient and the Fourier transform of f gives the frequency-dependent dielectric function $\epsilon(\omega)$. The superposition integral implies a medium with dispersion of the linear dielectric constant. In writing the second term as a product, the dispersive effects associated with the nonlinearity are neglected. If $\delta\epsilon(t)$ arises from the alignment of anisotropic molecules along the electric field, the Boltzmann equation can be employed to show that to first order in the field intensity

$$\tau\,\partial\delta\epsilon/\partial t = -(\delta\epsilon - \epsilon_2 E^2). \tag{2}$$

Since $1/\tau$ is much less than optical frequencies, molecular orientation cannot respond to optical variations in the torque resulting from the applied electric field and the induced molecular dipole moment. Consequently $\delta\epsilon$ effectively responds only to the part of E^2 which does not vary at optical frequencies. For CS_2 the value of ϵ_2 is about 4.5×10^{-11} esu.

For a linearly polarized plane wave, the propagation of the optical field is governed by the wave equation

$$\frac{\partial^2 E}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2 D}{\partial t^2} = 0, \tag{3}$$

where z is the direction of propagation. The effects resulting from the above nonlinearity can be treated conveniently by introducing Fourier transforms of the complex amplitudes which describe the spread about the optical frequency. Thus if the electric field is specified by

$$E(z,t) = \tfrac{1}{2}[\mathcal{E}(z,t)e^{i(k_0 z - \omega_0 t)} + c.c.],$$

k_0 and ω_0 being the carrier propagation vector and frequency, the envelope can be written as

$$\mathcal{E}(z,t) = \frac{1}{2\pi}\int_{-\infty}^{\infty} dQ \int_{-\infty}^{\infty} d\Omega\, \mathcal{E}(Q,\Omega)e^{+i(Qz - \Omega t)}, \tag{4a}$$

where $\omega = \omega_0 + \Omega$, $k = k_0 + Q$ and the components $\mathcal{E}(Q,\Omega)$ are given by

$$\mathcal{E}(Q,\Omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} dz \int_{-\infty}^{\infty} dt\, \mathcal{E}(z,t)e^{-i(Qz - \Omega t)}. \tag{4b}$$

The displacement field can be handled similarly. The Fourier transform of its amplitude is composed of two portions: the linear portion $\epsilon(\omega)\mathcal{E}(Q,\Omega)$, and the transform of $\delta\epsilon\mathcal{E}$ the nonlinear portion, which will be denoted as $[\delta\epsilon\mathcal{E}]_T$.

The Fourier transform of the wave equation is,

$$\left[-k^2 + \left(\frac{\omega}{c}\right)^2 \epsilon(\omega)\right]\mathcal{E}(Q,\Omega) = -\left(\frac{\omega}{c}\right)^2 [\delta\epsilon\mathcal{E}]_T. \tag{5}$$

The evolution of modulated signals possessing either a large number of discrete Fourier components or a continuum of such components, all coupled nonlinearly, cannot be treated easily in the transform space. However, (5) provides a way of deducing a travelling wave equation containing higher-order time derivatives of the field. These occur as a result of expanding (5) in powers of Ω to treat the effects of dispersion. The final equation has also been deduced directly from the wave equation; however, the procedure is much more tedious and the general form is not as transparent.

We begin by writing $(\omega/c)[\epsilon(\omega)]^{1/2}$ in terms of the frequency deviation Ω, obtaining

$$(\omega/c)[\epsilon(\omega)/\epsilon_0]^{1/2} = (\omega_0 + \Omega + \mathcal{L}\Omega)/c, \tag{6}$$

in which $\epsilon_0 = \epsilon(\omega_0)$ and $\mathcal{L}$ is an infinite series in Ω. The first few terms are

$$\mathcal{L} = \tfrac{1}{2}a + (A/\omega_0)\Omega/2 + (B/\omega_0^2)\Omega^2/6 + \cdots. \tag{7}$$

The coefficients a, A, and B are in turn determined by the frequency derivatives of $\epsilon(\omega)$ at ω_0. They are

$$a = \frac{\omega_0}{\epsilon_0}\frac{\partial\epsilon}{\partial\omega}\bigg|_{\omega_0},\quad A = a + \tfrac{1}{2}b - \tfrac{1}{4}a^2, \tag{8a}$$

$$B = \tfrac{3}{2}b + \tfrac{1}{2}c - \tfrac{3}{4}ab - \tfrac{3}{4}a^2 + \tfrac{3}{8}a^3, \tag{8b}$$

with

$$b = \frac{\omega_0^2}{\epsilon_0}\frac{\partial^2\epsilon}{\partial\omega^2}\bigg|_{\omega_0},\quad \text{and } c = \frac{\omega_0^3}{\epsilon_0}\frac{\partial^3\epsilon}{\partial\omega^3}\bigg|_{\omega_0}.$$

By factoring the left-hand side of Eq. (5) and dividing through by $k + (\omega/c)[\epsilon(\omega)]^{1/2}$, and noting that $k_0 = \omega_0\sqrt{\epsilon_0}/c$, we obtain

$$[-Q + \Omega\,\epsilon_0^{1/2}/c]\mathcal{E}(Q,\Omega) + (\epsilon_0^{1/2}/c)\mathcal{L}\Omega\mathcal{E}(Q,\Omega)$$

$$= -\frac{(\omega_0^2 + 2\omega_0\Omega + \Omega^2)}{c^2[2k_0 + Q + (\sqrt{\epsilon_0}/c)(\Omega + \mathcal{L}\Omega)]}[\delta\epsilon\mathcal{E}]_T. \tag{9}$$

For the initial pulse, the frequency components of the electric field amplitude, $\mathcal{E}(Q,\Omega)$, are assumed clustered about $\Omega = 0$. Moreover, if dispersion is able to overcome the steepening effect before the envelope attains variations less than or equal to a wavelength, then Ω will always be less than the optical frequency, ω_0. One can expand the denominator occurring in the nonlinear term of the above equation. Taking the inverse

transform of the resultant equation, one obtains the following complex propagation equation for the electric field envelope

$$O^+\mathcal{E}(z,t)=\frac{i}{2\omega_0}\left[\omega_0^2+2\omega_0 i\frac{\partial}{\partial t}-\left(\frac{\partial}{\partial t}\right)^2\right]\times\left[1+\sum_{n=1}^{\infty}\left(\frac{i}{2\omega_0}\right)^n\left(O^- - \mathfrak{L}\frac{\partial}{\partial t}\right)^n\right]\times\left[\frac{\delta\epsilon}{\epsilon_0}\mathcal{E}(z,t)\right]-\mathfrak{L}\frac{\partial}{\partial t}\mathcal{E}(z,t). \quad (10)$$

$\mathfrak{L}$ here is an operator obtained by setting $\Omega = i\partial/\partial t$ in Eq. (7). Two directional derivatives have also been defined; $O^+ = (c/\sqrt{\epsilon_0})\,\partial/\partial z + \partial/\partial t$, a derivative in the direction of propagation in the $z-t$ plane for the linear and dispersionless case, and $O^- = (c/\sqrt{\epsilon_0})\,\partial/\partial z - \partial/\partial t$, a derivative in a direction perpendicular to this direction. The latter, if it became of the order of the optical variation, would lead to a significant reflection of the pulse from the nonlinear variations in the medium. Although the change in the refractive index is found experimentally to be of the order of 0.01 or larger in CS_2, because of the finiteness of the pulse and the relaxation of the medium, this change is not abrupt and thus such reflections are negligible. In this case, only the lowest-order term in the operator series is significant, so that the pulse equation simplifies considerably becoming

$$O^+\mathcal{E}(z,t)=\frac{i}{2\omega_0}\left[\omega_0^2+2\omega_0 i\frac{\partial}{\partial t}-\frac{\omega_0}{2i}\left(O^+-2\frac{\partial}{\partial t}-\mathfrak{L}\frac{\partial}{\partial t}\right)\right]\times\left(\frac{\delta\epsilon}{\epsilon_0}\mathcal{E}(z,t)\right)-\mathfrak{L}\frac{\partial}{\partial t}[\mathcal{E}(z,t)]. \quad (10a)$$

From Maxwell's equations we also find the magnetic field in terms of the electric field as follows

$$\mathcal{H}(z,t)=\left\{1+\frac{1}{i\omega_0}O^+\left[\sum_{j=0}^{\infty}\left(\frac{1}{i\omega_0}\frac{\partial}{\partial t}\right)^j\right]\right\}\times\epsilon_0^{1/2}\mathcal{E}(z,t). \quad (11)$$

The relaxation equation for the nonlinearity written in terms of the amplitude, $\mathcal{E}$, is

$$\tau\frac{\partial\delta\epsilon}{\partial t}=-(\delta\epsilon-\tfrac{1}{2}\epsilon_2\mathcal{E}\mathcal{E}^*). \quad (12)$$

Equations (10)–(12), then, provide the basic expressions upon which a discussion of the evolution of a quasi-monochromatic pulse will be based. Neglecting dispersion in the lowest order of nonlinearity, these yield the pulse equations of Ref. 9 (see Appendix A).

III. GAIN ARISING FROM THE EFFECTS OF DISPERSION

Without dispersion, Stokes–anti-Stokes linear phase-matching occurs in the forward direction and the gain is zero. Dispersion, however, introduces a mismatch and hence a gain which we examine briefly by linearizing in weak-fields shifted from the strong central optical frequency. We thus write the field, $\mathcal{E}$, in the form

$$\mathcal{E}(z,t)=\mathcal{E}_0+\mathcal{E}_1 e^{i[(k_1-k_0)z-\Omega t]}+\mathcal{E}_2 e^{i[(k_2-k_0)z+\Omega t]} \quad (13a)$$

where $\mathcal{E}_1=\mathcal{E}_{01}e^{i(\Delta k/2+\gamma)z}$ (13b)

and $\mathcal{E}_2^*=\mathcal{E}_{02}^* e^{-i(\Delta k/2-\gamma)z}$. (13c)

Here, $k_i=(\omega_i/c)\sqrt{\epsilon_i}$ with $\epsilon_i=\epsilon(\omega_i)$, for $i=0,1,2$. Also Δk is the linear mismatch, $2k_0-k_1-k_2$. The constant amplitudes $\mathcal{E}_{01}$, $\mathcal{E}_{02}$ are assumed to be much less than the laser field amplitude $\mathcal{E}_0$ which is itself assumed not to change. The Stokes gain is then $|2\,\mathrm{Im}\gamma|$, where γ is found by substituting Eq. (13) into (10a). If

$$(\Delta k/k_0)(\omega_0/\Omega)^2 \gg \epsilon_2\mathcal{E}_0\mathcal{E}_0^*/\epsilon_0,$$

we obtain

$$2\gamma\simeq(\Delta k)^{1/2}\left[\Delta k-\epsilon_0\left(\frac{k_2}{\epsilon(\omega_2)}+\frac{k_1}{\epsilon(\omega_1)}\right)\frac{\delta}{D(\Omega)}\right]^{1/2} \quad (14)$$

in which $D(\Omega)=1+i\Omega\tau$, τ being the orientational relaxation time defined previously, and δ is equal to $\epsilon_2|\mathcal{E}_0|^2/2\epsilon_0$.

To lowest order in the dispersion, Δk is equal to $-A(\Omega/\omega_0)^2 k_0$, where A is given by (8a). For most liquids at the ruby frequency, A is greater than zero. There is then a single maximum in the gain curve given by

$$(\Omega\tau)_{\mathrm{opt}}\simeq\left\{\tfrac{1}{2}+\left[\tfrac{1}{4}+\delta(\omega_0\tau)^2/|A|\right]^{1/2}\right\}^{1/2}. \quad (15)$$

In contrast to the dispersionless case, where the phase-matching angle was zero, it is now finite. The phase-matching dip increases in width with increasing laser power, and eventually spreads into the forward direction. The increase in frequency shift with power, as given by (15), occurs because the greater the phase-matching angle (due to a large frequency shift), the less the dip can spread into the forward direction.

Thus, as illustrated in Table I and Fig. 1, for a very small nonlinearity, the gain is equal to the stimulated-Rayleigh gain in the backward direction. With increasing nonlinearity, the Stokes–anti-Stokes interaction suppresses the gain, so that for usual small-scale filaments it is well below the stimulated Rayleigh gain. It should be pointed out that because of the finite angular spread in k vectors in a trapped filament, the average stimulated Rayleigh gain may be greater than that found above for collinear propagation.

We will in Sec. V investigate whether the enhancement of the Stokes components, due to the exponential gain as given by (14), is as important as the phase development effects due only to relaxation of the nonlinearity.

Finally, we should point out that Ostrovskii[12] has shown that for the case of negative second-

TABLE I. Gain per unit length versus laser intensity I_l. Here $\delta\epsilon$ is the nonlinear change in the dielectric constant, $\max g_f$ the maximum value of the forward gain shown in the corresponding curves of Fig. 1, and g_b the maximum backward or large scattering-angle gain.

Curve in Fig. 1	$I_l(MW/cm^2)$	$\frac{1}{2}\delta\epsilon$	$\max g_f$	g_b
(a)	18×10^{-4}	10^{-10}	5.6×10^{-6}	5.6×10^{-6}
(b)	18×10^{-2}	10^{-8}	5.0×10^{-4}	5.6×10^{-4}
(c)	18	10^{-6}	1.7×10^{-2}	5.6×10^{-2}
(d)	18×10^{2}	10^{-4}	0.4	5.6
(e)	18×10^{4}	10^{-2}	9.4	5.6×10^{2}
not shown in Fig. 1	18×10^{5}	10^{-1}	4.4×10	5.6×10^{3}

order dispersion, an instability is present in the forward direction even when relaxation is absent. This instability provides gain for finite frequency shifts. In this case the phase-matching angle is not real, giving rise to a phase mismatch in the forward direction which is of such a sign as to allow four-photon light-by-light scattering to occur. This case is unimportant for all liquids studied at ruby frequencies.

IV. PULSE PROPAGATION NEGLECTING RELAXATION AND DISPERSION

The propagation of a pulse through the medium is determined by Eqs. (11) and (12); and the power spectrum, by

$$S(Q,\Omega)=(c/8\pi)\,\mathrm{Re}[\mathcal{E}(Q,\Omega)\mathcal{K}^*(Q,\Omega)]$$
$$\simeq(c/8\pi)\sqrt{\epsilon_0}\,|\mathcal{E}(Q,\Omega)|^2. \qquad (16)$$

There are basically three effects whose influence upon the pulse propagation we wish to consider; the nonlinearity, relaxation of the nonlinearity, and the linear dispersion of the medium.

The primary effect is that of the nonlinearity. It generates both self-phase modulation and self-steepening (envelope distortion). Experimental observations of these effects have so far been restricted to the frequency spectrum. It will be shown in Sec. V that relaxation effects can have a marked influence particularly in the qualitative behavior of the upshifted spectrum; thus only the downshifted (Stokes) spectrum is considered in the present section.

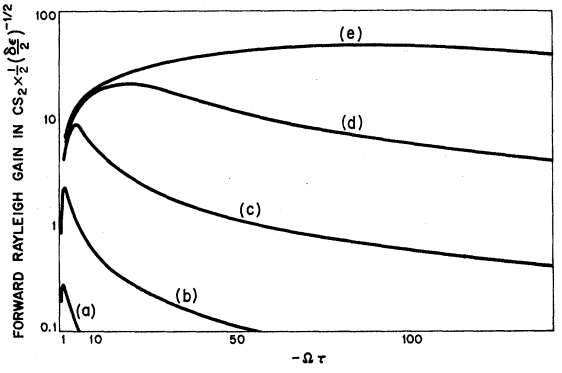

FIG. 1. Forward Rayleigh gain per unit length in CS_2 versus frequency shift for values of the nonlinearity given in Table I. The curve labels (a)–(e) correspond to those of the Table. The second-order dispersion coefficient A is equal to 0.056.

The complex amplitude $\mathcal{E}(z,t)$ can be written as $\mathfrak{A}(z,t)e^{+i\delta\phi(z,t)}$, where $\mathfrak{A}(z,t)$ and $\delta\phi(z,t)$ are real. For distances which are very short compared to the shock distance,[9] the self-steepening of the envelope $\mathfrak{A}^2(z,t)$ is not appreciable. In this case, the spectral development is almost entirely determined by $\delta\phi(z,t)$, which is described in the dispersionless case by

$$O^+\delta\phi(z,t)=(\omega_0/2\epsilon_0)\delta\epsilon(z,t), \qquad (17)$$

where $\delta\epsilon$ is found from (2). If we assume that $\mathfrak{A}^2$ has a fixed shape for all z, and that $\delta\phi(z=o,t)=0$, then $\delta\phi$ becomes

$$\delta\phi(z,t)=\alpha\int_{-\infty}^{t}\frac{e^{-(t-t')/\tau}}{\tau}\left(\frac{\mathfrak{A}(z,t')}{\mathfrak{A}_0}\right)^2dt'. \qquad (18)$$

We define the phase parameter α to be $k_0z\epsilon_2\mathfrak{A}_0^2/4\epsilon_0$, in which $\mathfrak{A}_0^2$ normalizes the initial intensity variation to unity.[13]

The methods of stationary phase applied to Eq. (4b) show that the frequency shift is approximately given by (minus) the time derivative of $\delta\phi$. Thus the slope of the phase curve determines, to an important degree, the Fourier component associated with that part of the pulse. There are generally two points in the phase curve with the same slope and, as Shimizu[6] has pointed out, the corresponding contributions can interfere constructively or destructively depending on whether they differ in phase by an even or odd multiple of π. It can be shown that the maximum number of interference peaks is $\alpha/2\pi$. While the phase development is very significant, we also must consider the shape of the envelope, since the degree of interference will be determined in part by the amplitude ratio. Furthermore, the rate of change of slope of the phase will also influence the size of the contributions of each region.

The downshifted part of a filament spectrum is shown in Fig. 2. Curve (a) of this figure shows the theoretical self-modulation obtained by Cheung *et al.*[7] from Eq. (18), when $\tau=0$. The optical wave envelope $\mathfrak{A}^2(z,t)$ is assumed to possess a

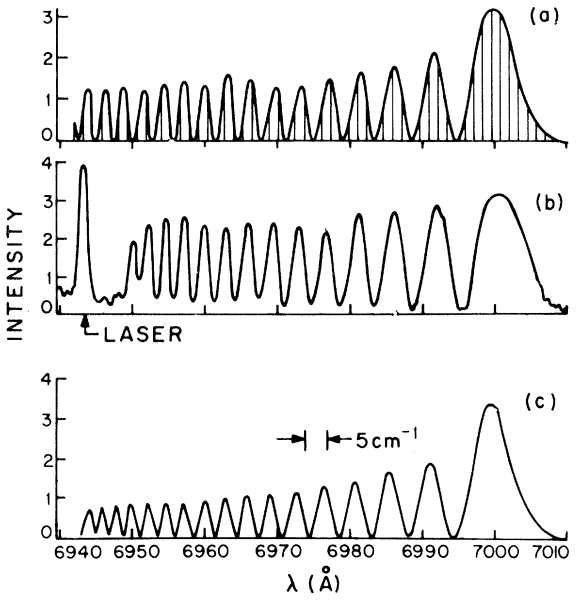

FIG. 2. Spectral broadening in CS_2. (a) Theoretical calculation of Cheung *et al.*[7] (b) Experimental spectrum of Cheung *et al.* (c) Theoretical calculation assuming an initially Gaussian intensity profile. The full $(1/e)$ width equals 7.4 psec and $\alpha = 107$.

weak modulation at a frequency $\omega_S = 2.5\ \mathrm{cm}^{-1}$. This envelope is independent of z, since self-steepening was ignored. In comparison, curve (c) indicates the self-modulation obtained when $\tau = 0$, assuming a Gaussian envelope $\mathcal{E}^2(z,t)$ of full width 7.4 psec. Pulse distortion is taken into account but has little effect on the spectrum in this case, since the distance is about 1/100 of the shock distance. The curves of both Figs. 2(a) and 2(c) fit the experimental spectrum [curve (b)] of Cheung *et al.*[7] for CS_2.

It is evident that there is little difference between the overall spectral development for a single pulse and that for a sinusoidal modulation whose period is of the order of twice the width of the pulse. The spectrum for the sinusoid, however, does possess a discrete underlying structure whose components are separated by ω_S, in contrast to the continuous spectrum for the pulse. A libration effect arising from an oscillation of the molecules about the electric field has been suggested[7] as a model giving rise to a sinusoidal modulation. Experimentally there are a few cases which show fine structure, possibly due to sinusoidal modulation.[14] There is apparently no fine structure associated with the largest frequency shifts contrary to what is expected for pure sinusoidal modulation. Variation in modulation frequency might blur out the discrete spectra in the region of large frequency shifts. The finite filament lifetime, if it is only a few modulation cycles, can also lead to a blurring of the fine structure.

If the spectra arise from pulses[15] of the order of 7 psec, their origin is still uncertain. Spectra with the above characteristics have been observed in experiments using laser pulses at least as long as 100 psec. We therefore infer that pulses of a few psec, if present, are formed in the liquid. Nevertheless, the initial time structure of the laser is important, since it plays a role in the nonlinear processes, including self-focusing.

The self-focusing process and stimulated scattering might give rise to trapped filaments whose temporal extent is of the order of 7 psec. Marburger and Wagner[16] have pointed out that a pulse will become shortened in time as it travels from the cell entrance to the shortest self-focusing point in the beam. This process of chopping the wings of the pulse should continue until its width is the order of the relaxation time for orientation. This process can also turn a smoothly modulated signal into a series of short pulses, which would have a spectrum exhibiting fine structure at the modulation frequency.

Stimulated scattering processes, for example Brillouin scattering[17] in the backward (opposite to the laser beam) direction, could lead to a shortening of the self-trapped light pulse in a filament. This shortening would continue until the gain dropped to a low enough value to cut off the loss due to stimulated scattering. The Brillouin gain in this region will take on transient character.[18,19] The transient gain exponent can be given by

$$G = (2g_0 v_0/\tau')^{1/2}\tau_p .$$

Here g_0 is the steady-state gain per unit length, τ' is the phonon relaxation time, v_0 is the velocity of light, and τ_p the pulse length in time. Note that $g_0 \propto \tau'$; therefore G does not depend on τ'. In small scale filaments $g_0 \approx 10^4\ \mathrm{cm}^{-1}$, $\tau' \approx 10^{-9}$ sec, and assuming $G < 20$ for the instability to be ineffective, we find $\tau_p < 25$ psec.

V. INFLUENCE OF RELAXATION AND DISPERSION

In the present section, we wish to consider the influence of relaxation upon the spectra and to discuss the role which dispersion should play. We will also include pulse distortion; however, the effect of this distortion on the major features of the spectra is insignificant for the cases treated here. This is in contrast to the results given in a previous paper,[9] in which the spectra were found during strong shock development.

Pulse envelopes, whose widths are of the order of the relaxation time or less in extent, may result from the self-focusing process or from stimulated scattering processes occurring in the backward direction. In such cases the lagging edge of the phase $\delta\phi(z,t)$ will possess an exponential tail with a decay time equal to the relaxation time τ. Since this portion of the phase curve determines the upshifted Fourier components, we can immediately conclude that these components will be most affected by the relaxation. Figure 3(a) shows a computer calculation of the pulse development at two distances including the effects of relaxation, while Fig. 3(b) shows the phase development at the same distance (the initial phase was assumed to be zero). The relaxation time is taken to be 2

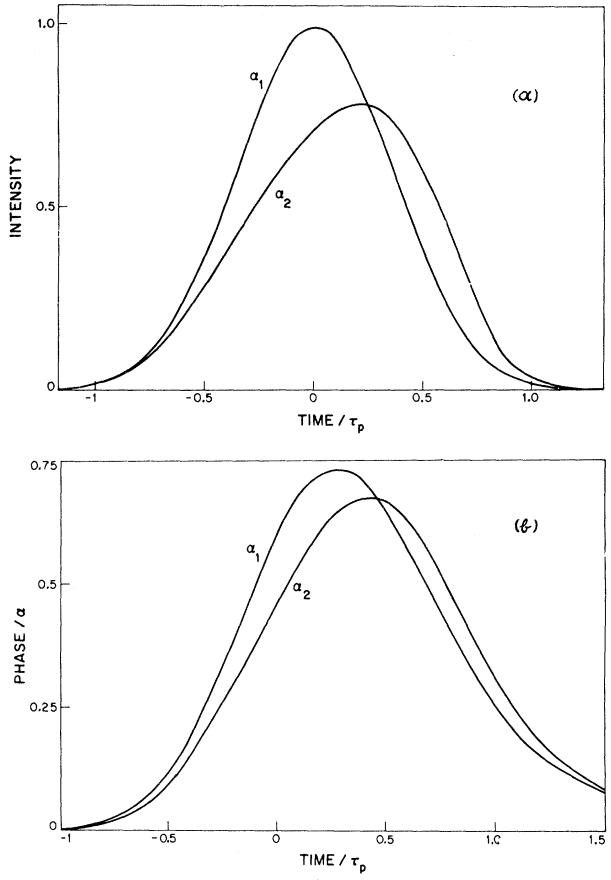

FIG. 3. Influence of relaxation upon the pulse development. The initial intensity is assumed Gaussian with full $1/e$ width, τ_p, given by $\omega_0\tau_p = 1.4 \times 10^4$ radians. The ratio of the relaxation to the pulse width, τ/τ_p, is 0.36. (a) Intensity profiles normalized to the initial maximum intensity of the Gaussian plotted for two values of the phase parameter, $\alpha_1 = 117$ and $\alpha_2 = 2396$. At the shock distance z_s (not shown) α is equal to $\alpha_s = 2635$. (b) Corresponding phase curves, normalized with respect to α, assuming the initial phase to be zero.

psec corresponding to CS_2.[20]

The frequency spectrum corresponding to a propagation distance z_1 (Fig. 3) is shown in Fig. 4, and illustrates the influence of relaxation, which is particularly noticeable on the anti-Stokes side. Of the two contributions from the envelope $\mathcal{A}(z,t)$ to any upshifted Fourier component, the one nearest the envelope peak dominates more than in the relaxationless case, since the other contribution will be out in the exponential tail of the phase curve where the envelope is smaller. Thus, the interference pattern shows less contrast for the upshifted part of the spectrum. However, as in the relaxationless case, this interference effect increases with increasing upshift, since the ratio of the two envelope amplitudes determining each frequency component tends to one.

In contrast, the entire Stokes spectrum exhibits a pronounced interference effect similar to that

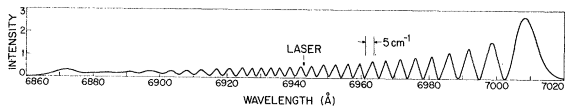

FIG. 4. Spectrum of the pulse of Fig. 3 at α_1 with ω_0 at the ruby frequency ($2\pi c/\omega_0 = 6943$ Å). τ_p is 5 psec, and $\tau = 2$ psec is the orientational relaxation time for CS_2. The intensity is plotted in arbitrary units.

observed in the relaxationless case. We also notice that there is a compression of the frequency spectrum, since the height of the phase curve is decreased due to the effects of relaxation. Frequency compression, however, is even greater on the anti-Stokes side due to the exponential tail.

We also see that the total intensity of the anti-Stokes components is less than that of the Stokes side due to the phase delay and the exponential tail.

The relative importance of the above effects depends upon the detailed shape of the pulse and its length with respect to the relaxation time. However, many self-modulated filaments display the type of Stokes–anti-Stokes asymmetry illustrated in Fig. 4.[8,14,21]

Variation in observed spectra could be due to variation in pulse shape.[22] A significant fraction of the filaments observed by various authors show weak upshifted wings which are more extensive than the intense downshifts. Such spectra might arise from pulses whose tails drop sharply. Stimulated scattering in the backward direction as well as self-steepening would produce a sharp tail.

For the spectrum of Fig. 4, dispersive effects have been neglected since the propagation distance is small compared to the steepening distance, as well as the distance for exponential gain to be significant. We also have obtained the spectrum for the same conditions as in Fig. 4, but with dispersion included. Because of the change in group velocity the spectral spread was about 4% smaller, while the asymmetry was about 3% greater. Since without dispersion and assuming no pulse distortion, the less the spread the smaller the asymmetry, this result may, therefore, indicate the onset of exponential gain effects discussed in Sec. III. Noncollinear gain effects, not included in the present plane-wave calculation, will also have an effect on asymmetry. It has been shown that relaxation inhibits but does not prevent the steepening of the envelope[9] and the buildup of the phase. If lifetimes and propagation distances associated with the filaments are great enough, dispersion will ultimately become important, provided other spreading mechanisms are negligible.

In Fig. 5, a spectrum is shown for a case of sinusoidal modulation, where the modulation frequency is chosen to give the same number of Stokes peaks and the same total Stokes shift as in the Gaussian case (Fig. 4). A modulation depth of 100% was chosen; less modulation would give correspondingly less asymmetry. Note that for the periodic case, the extent of the anti-Stokes is comparable to the Stokes spectrum.

Figure 6(a) shows an experimental spectrum

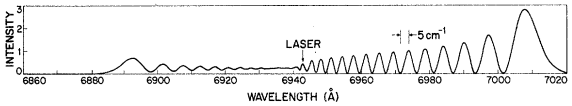

FIG. 5. Spectrum of a light beam, whose intensity was 100% sinusoidally modulated, plotted in arbitrary intensity units. The period is 10.2 psec, and the relaxation time is 2 psec. The phase parameter α is equal to 1.46.

taken with a mixture of two thirds CS_2 and one third benzene by volume. The spacing of the peaks close to the laser was about 3.5 cm^{-1}. A similar spectrum was obtained theoretically [Fig. 6(b)] using a Gaussian pulse of width 5.4 psec and a relaxation time of 9 psec. Perhaps the CS_2 orientational relaxation time is altered by the presence of the benzene. Deviation from Gaussian pulse shape might modify somewhat the parameters used.

VI. CONCLUSION

Frequency broadening in small-scale trapped filaments can be attributed to either an initial modulation of the light within the filament or to the propagation of ultrashort pulses within filaments. In the former case, or in the case of a sequence of pulses, one expects a discrete underlying structure to accompany the envelope of the spectrum. Experimentally, it is not yet clear how often and under what conditions such a structure is to be found.

Short pulses might be generated by stimulated scattering or in the focusing process. If it is found experimentally that a single filament produces a continuous spectrum, then the individual pulse length in the filaments can be of the order of 5 to 10 psec in CS_2.

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APPENDIX A

The Nondispersive Energy and Phase Equations

We wish to show that Eqs. (10a) and (11), in the absence of dispersion and to lowest order in the nonlinearity, reduce to the energy and phase equations employed previously[9] in a study of self-steepening. Therefore, dropping dispersion, we write (10a) as

$$O^+\left(\mathcal{E}+\frac{\delta\epsilon}{4\epsilon_0}\mathcal{E}\right)=-\frac{\omega_0}{2i}\frac{\delta\epsilon}{\epsilon_0}\mathcal{E}-\frac{1}{2}\frac{\partial}{\partial t}\left(\frac{\delta\epsilon}{\epsilon_0}\mathcal{E}\right)\ . \quad (A.1)$$

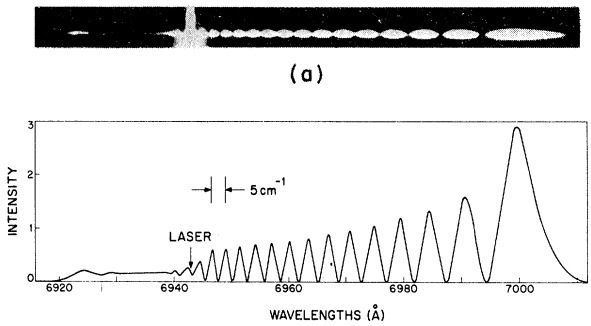

FIG. 6. (a) Experimental spectrum and (b) theoretical fit using a Gaussian pulse of width 5.4 psec, a relaxation time of 9 psec, and $\alpha = 265$.

Multiplying this by $\mathcal{E}^*$ and adding the result to $\mathcal{E}$ times the complex conjugate of Eq. (A. 1) gives

$$O^+\left(1+\frac{\delta\epsilon}{4\epsilon_0}\right)\mathcal{E}^*\mathcal{E}+\frac{\mathcal{E}^*\mathcal{E}}{4}O^+\left(\frac{\delta\epsilon}{\epsilon_0}\right)$$

$$=-\frac{1}{2}\frac{\partial}{\partial t}(\delta\epsilon\mathcal{E}^*\mathcal{E})-\frac{\mathcal{E}^*\mathcal{E}}{2}\delta\epsilon\ . \quad (A.2)$$

By taking the two terms on the right-hand side to the left-hand side and then adding and subtracting $\frac{1}{4}O^+[(\delta\epsilon/\epsilon_0)\mathcal{E}^*\mathcal{E}]$ to the latter, this becomes

$$\frac{\partial}{\partial z}(\rho v)+\frac{\partial}{\partial t}\rho+\frac{\rho}{2\epsilon}\frac{\partial\epsilon}{\partial t}-\frac{1}{4}\delta\epsilon O^+\left(\frac{\rho}{\epsilon}\right)=0 \quad (A.3)$$

with ρ equal to $\epsilon\mathcal{E}^*\mathcal{E}/8\pi$, and v equal to $c/\epsilon^{1/2}$ for ϵ equal to $\epsilon_0+\delta\epsilon$, assuming $\epsilon_0 \gg \delta\epsilon$.

It follows from Eq. (11) (neglecting terms arising from $j=1$ and higher) and (A. 1) that

$$\mathcal{H}\cong\sqrt{\epsilon_0}\left(1+\frac{\delta\epsilon}{2\epsilon_0}\right)\mathcal{E}\cong\sqrt{\epsilon}\,\mathcal{E}\ . \quad (A.4)$$

Therefore ρ is identical with the energy density ρ of Ref. 9 and (A. 3) describes the propagation of energy in the nonlinear medium. Finally, this equation can be reduced to Eq. (6) of Ref. 9 by dropping the term $\delta\epsilon O^+(\rho/4\epsilon)$, which is of order $(\delta\epsilon)^2$.

To obtain the phase equation we note that for $\mathcal{E}$ equal to $\mathcal{A}e^{+i\delta\phi}$, the imaginary part of (A. 1) gives the propagation equation for $\delta\phi$:

$$\frac{c}{\sqrt{\epsilon}}\frac{\partial\delta\phi}{\partial z}+\frac{\partial\delta\phi}{\partial t}=-k_0\left(\frac{c}{\sqrt{\epsilon}}-\frac{c}{\sqrt{\epsilon_0}}\right)\ , \quad (A.5)$$

which, when only zero- and first-order terms in $\delta\epsilon$ are kept, becomes Eq. (10) of Ref. 9.

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[‖]Operated with support from the U. S. Air Force.

[1]N. Bloembergen and P. Lallemand, Phys. Rev. Letters 16, 81 (1966).

[2]P. Lallemand, Appl. Phys. Letters 8, 276 (1966).

[3]K. Shimoda, J. Appl. Phys. (Japan) 5, 615 (1966).

[4]H. P. H. Grieneisen and C. A. Sacchi, Bull. Am. Phys. Soc. 12, 686 (1967).

[5]R. G. Brewer, Phys. Rev. Letters 19, 8 (1967).

[6]F. Shimizu, Phys. Rev. Letters 19, 1097 (1967).

[7]A. C. Cheung, D. M. Rank, R. Y. Chiao, and C. H. Townes, Phys. Rev. Letters 20, 786 (1968).

[8]For a comprehensive discussion of small scale self-trapped filaments see: R. G. Brewer, J. R. Lifsitz, E. Garmire, R. Y. Chiao, C. H. Townes, Phys. Rev. 166, 326 (1968).

[9]F. DeMartini, C. H. Townes, T. K. Gustafson and P. L. Kelley, Phys. Rev. 164, 312 (1967).

[10]R. Y. Chiao, P. L. Kelley, and E. Garmire, Phys. Rev. Letters 17, 1158 (1966).

[11]The phase-matching angle for interacting plane waves is given from the relations $\vec{k}_1+\vec{k}_2=2\vec{k}_0$ and $\omega_1+\omega_2=2\omega_0$. This angle forms a cone about the forward direction, and is of the order of a few degrees for the stimulated Raman effect. Another phase-matching condition may be found for the propagation of light in filaments [see A. Szöke, Bull. Am. Phys. Soc. 9, 490 (1964)].

[12]L. A. Ostrovskii, Zh. Eksperim. i Teor. Fiz. 51, 1189 (1966) [English transl.: Soviet Physics – JETP 24, 797 (1967)].

[13]For a Gaussian pulse $\mathcal{A}_0^2$ is the maximum initial squared amplitude $|\mathcal{E}_0|^2$ and for a sinusoid it becomes $m|\mathcal{E}_0|^2$, where m is the depth of modulation. Note that for a sinusoid our α is twice that defined in Ref. 7.

[14]J. R. Lifsitz, and H. P. H. Grieneisen, to be published.

[15]It should be noted that the pulses involved here could have flattened tops and hence be longer than 7 psec. However, the rise times must be of the order of half this duration. Such squared pulses would give large spectral contributions at or near the laser frequency. However, it is found experimentally that in many cases the most intense part of the Raman-Stokes light is shifted well away from the frequency which is given by the laser frequency minus the vibrational frequency. This indicates very little intensity in the filament at the laser frequency. (It is difficult to measure the intensity at the laser frequency ω_0 directly because of the strong untrapped background).

[16]J. H. Marburger, W. G. Wagner, J. Quant. Elec. 3, 415 (1967).

[17]G. Bret drew our attention to this point.

[18]N. M. Kroll, J. Appl. Phys. 36, 34 (1965).

[19]N. M. Kroll and P. L. Kelley, to be published.

[20]S. L. Shapiro and H. P. Broida, Phys. Rev. 154, 129 (1967).

[21]D. M. Rank, A. C. Cheung, R. Y. Chiao, and C. H. Townes, in Proceedings of the Quantum Electronics Conference, Miami, Florida, 1968 (unpublished), paper 5E-8.

[22]Suggested by C. H. Townes.

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Charged Bose Gas*

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Green-function techniques are applied to the charged Bose gas at the absolute zero of temperature. The procedure permits some general remarks to be made concerning the expansions of the ground-state energy and depletion parameter in terms of r_s, the ratio of the interparticle spacing to the Bohr radius. It is found that these series contain not only all integer powers of $r_s^{3/4}$ as had been conjectured, but additional functions of r_s as well. The first-two terms in the expansion of the ground-state energy are calculated exactly as are the first-four terms in the expansion of the depletion parameter. The former expansion agrees with the results of Lee and Feenberg. The latter, containing a noninteger power of $r_s^{3/4}$, is new. It is shown that the ground-state energy diagrams which must be summed in the Bose gas are characterized by equal numbers of variables of integration. It is emphasized that these are not the most divergent diagrams as in the electron-gas calculation of Gell-Mann and Brueckner. This difference leads us to reformulate the diagrammatic procedure so as to emphasize its self-consistency. The underlying similarity of the charged Fermi and Bose-gas calculations is then made apparent.

I. INTRODUCTION

The charged Bose gas is a particularly interesting many-body system owing to the fact that the result of a conventional perturbation calculation of its ground-state energy is divergent. Moreover,

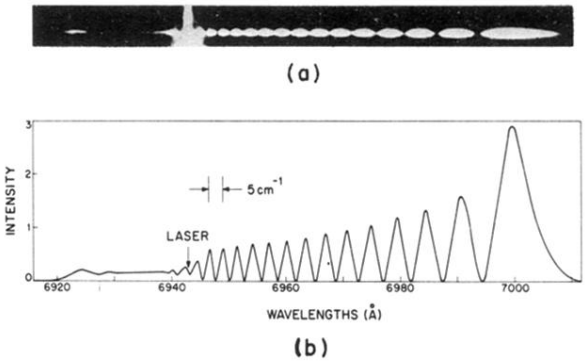

FIG. 6. (a) Experimental spectrum and (b) theoretical fit using a Gaussian pulse of width 5.4 psec, a relaxation time of 9 psec, and $\alpha = 265$.