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COMMENTS AND ADDENDA

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Phase-Modulation Spectra in Kerr Liquids

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Spectra of laser fields phase modulated by pulses in Kerr liquids have been calculated, retaining all phase information. For short and long pulses, spectra like those observed in CS_2 and bromobenzene, respectively, are predicted. The CS_2 spectra are quite sensitive to a phase parameter.

Spectral broadening of laser light by phase modulation has been observed to accompany the formation of self-trapped filaments in materials with intensity-dependent refractive indices.[1-8] The molecular orientation mechanism for the index nonlinearity in Kerr liquids is too slow to be driven at optical frequencies, but index modulation at difference frequencies can occur if the optical field is not monochromatic.[2] Broadening in solids due to a fast electronic mechanism has also been observed,[8] and the theory developed here should apply to this case if no second harmonic is generated in the medium. Typical broadened spectra observed in CS_2 appear in the letter by Shimizu.[5] A large fraction of the power is concentrated near the laser frequency (the slit region was attenuated by a factor of 41). Away from the laser frequency, the width, separation, and intensity of the spectral components generally increase with distance from the laser frequency toward either the Stokes or anti-Stokes side. There is considerable variation from filament to filament in intensity and spectral extent. Spectra observed in bromobenzene[6] and also in the $SeOCl_2$ liquid laser[7] have uniformly spaced components that generally decrease gradually in intensity away from the laser line, contrasting sharply with the CS_2 spectra.

The case of two monochromatic incident laser beams with small frequency separation ω_s has been treated by Bloembergen and Lallemand,[2] who calculated the refractive index correction, and by Cheung, Rank, Chiao, and Townes (CRCT),[9] who calculated the spectrum. The theoretical spectrum (Fig. 1) consists of perfectly sharp lines with spacing ω_s and intensity proportional to $J_m^2(\alpha)$, where m is the sideband number and α a parameter depending on the initial fields.

Shimizu has shown that the nonzero breadth of the CS_2 components implies that one of the incident fields is a pulse. Each sideband acquires a width comparable to the pulse bandwidth, and several overlapping sidebands can merge to form one broad spectral component. Cheung *et al.* treat pulsed fields by fitting an envelope of their discrete spectrum to the data. For the case of one incident field much stronger than the other, the envelope is similar to the CS_2 Stokes spectra. Envelope fitting is equivalent to *ad hoc* assignment of a shape, width, and phase to each sideband. For bromobenzene, where individual sidebands are resolved, the CRCT theory predicts more intensity far from the laser line than is observed, and does not predict the intense central region.

Gustafson *et al.*[10] have calculated the spectral development of an ultrashort pulse in a Kerr liquid. The pulse is phase modulated by itself in their model, while the CRCT model assumes phase modulation of a strong, initially monochromatic, field by interaction between itself and a weaker pulse. The Stokes and anti-Stokes spectra calculated by Gustafson *et al.* are essentially determined by the shapes of the leading and trailing edges, respec-

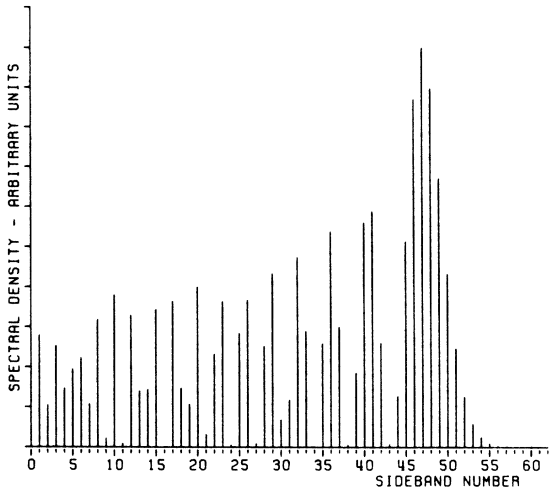

FIG. 1. Discrete spectrum predicted by the CRCT theory. The anti-Stokes side (not shown) is identical with the Stokes side.

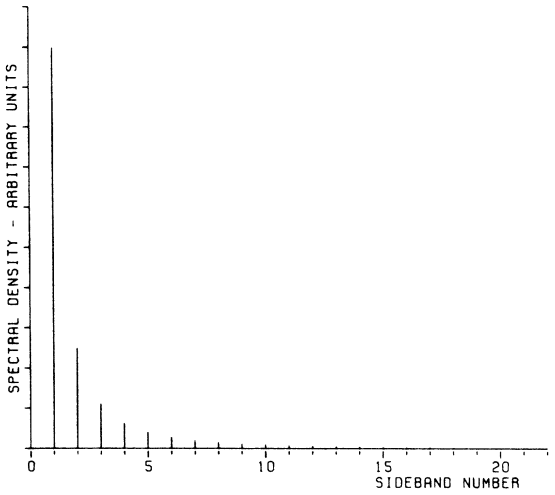

FIG. 3. Spectra in the long-pulse limit $\Gamma \to 0$. The anti-Stokes side (not shown) is identical with the Stokes side.

tively, of the pulse envelope. The calculated spectra always extend further to the Stokes side, in contradiction to some of the experiments, and the large intensity observed near the laser frequency by Shimizu is not predicted. The authors speculate that the filament-to-filament variation in spectral extent is due to variation in pulse shape, with spectra favoring the anti-Stokes side generated by pulses with sharp trailing edges. This interpretation requires at least two dissimilar pulse-forming mechanisms operating alternatively in quite similar filaments.

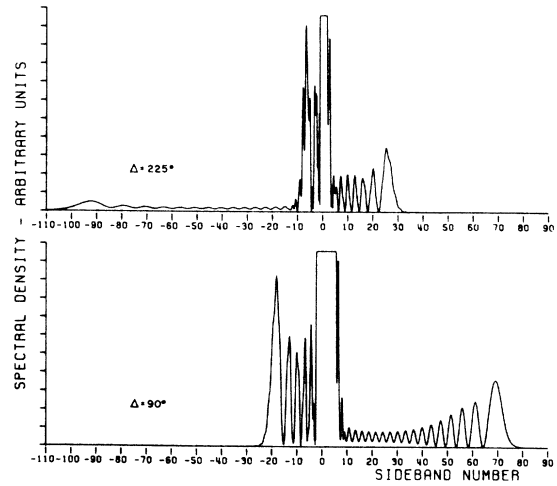

FIG. 2. Spectra predicted by the present theory for $\beta = 100$, $\Gamma = 1$, $\tau = 0$. The laser frequency corresponds to $\mu = 0$. The spectrum has been truncated in the region near $\mu = 0$, where the spectral density reaches values as large as 60 times the truncation level. Various degrees of asymmetry between Stokes (at right) and anti-Stokes spectra are predicted for different values of Δ.

We have calculated broadened spectra for a strong monochromatic laser field of frequency ω_0 and a weak Lorentzian pulse of center frequency $\omega_0 + \omega_s$ incident on a Kerr liquid. A practical laser field may be treated as monochromatic if its bandwidth is much smaller than ω_s. The origin of the pulse is not important; it can enter the liquid from outside or be generated inside by a nonlinear process. The light propagates in the z direction and the Kerr liquid fills the region $z > 0$. At $z = 0$, the field is given by

$$E(0, t) = E_0 \cos(\omega_0 t + \delta) + E_1 \eta(t) \sin[(\omega_0 + \omega_s) t] e^{-\Gamma t}, \tag{1}$$

where $E_0 \gg E_1$ and $\eta(t)$ is the unit step function. We have solved the system of equations for $E(z, t)$ and the refractive index $n(z, t)$ using a perturbation expansion in the nonlinear index coefficient n_2. To first order, the time-dependent index correction is

$$n_{ac} = (c/\omega_0 z)\, \beta [\sin(\omega_s T - \Delta) e^{-\Gamma T} + \sin(\Delta) e^{-T/\tau}], \tag{2}$$

where

$$T = t - n_0 z/c,$$

$$\beta = n_2 E_0 E_1 (\omega_0 z/c)\, [(1 - \Gamma\tau)^2 + \omega_s^2 \tau^2]^{-1/2},$$

and

$$\Delta = \delta + \arctan [\omega_s \tau/(1 - \Gamma\tau)].$$

Apart from a small amplitude-modulation term and an unimportant over-all phase factor, the phase-

modulated strong field is given by the real part of

$$E_c(z, t) = E_0 \exp[i\omega_0 T - i\omega_0 z n_{ac}/c]. \quad (3)$$

The spectral density $S(\omega_0 - \mu\omega_s)$ is evaluated in the usual manner in terms of the Fourier transform of $E_c(z, t)$. The sideband number μ is not restricted to integer values; positive μ corresponds to the Stokes spectrum. It can be shown analytically that, independent of all parameters, (a) the Stokes and anti-Stokes spectra are interchanged if the phase angle Δ changes by π, and (b) the total integrated powers into the Stokes and anti-Stokes spectra are the same. The latter result does not contradict the apparent favoring of one side by the observed spectra away from the laser line, since the intense region near the laser frequency contributes heavily to the powers.

For the case of pulse bandwidth Γ comparable to ω_s, the spectra are quite sensitive to Δ. The filament-to-filament spectrum variation is probably largely due to stochastic variations in this phase. The spectra of Fig. 2, computed using $\Gamma = 1\ \mathrm{cm}^{-1}$, $\beta = 100$, $\tau = 0$, and the CRCT value $\omega_s = 2.5\ \mathrm{cm}^{-1}$, compare favorably with some of Shimizu's CS_2 observations. His other observations can be reproduced for different values of β and Δ. Variation of τ has little effect except through Δ.

The spectrum of Fig. 3 represents the long-pulse limit $\Gamma \gg \omega_s$. In this limit the spectra are independent of Δ, and the sideband width, although small, increases with μ. The peak spectral density decreases monotonically, but it can be shown that the integrated *power* in the kth sideband is proportional to the square of $J_k(\beta)$, as in the CRCT theory. A spectrometer with insufficient resolution will perform the integration and produce a spectrum like that of Fig. 1 rather than that of Fig. 3.

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Vacancies and ^{4}He Impurities in Solid ^{3}He†

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Hamiltonians to describe vacancies and ^{4}He impurities in solid ^{3}He derived from a Hubbard-like-model starting point are presented and discussed.

Giffard and Hatton[1] used NMR techniques to measure the exchange-lattice relaxation rate[2] in solid ^{3}He in the temperature range 0.4–0.6 °K in the presence of ^{4}He impurity atoms. They found an extra contribution to the rate that is of the form $\eta(x)\, T^{n(x)}$, where x is the ^{4}He concentration and T is the temperature; $n(x)$ varies from 8.3 ± 2 (for $x < 5\times10^{-7}$) to 8.7 ± 0.1 (for $x \simeq 3\times10^{-4}$). Guyer and Zane[3] argued against the interpretation of this effect as being the result of a two-phonon process, as was initially expected; they suggested that it may be due to tunneling processes in the presence of vacancies and ^{4}He atoms in the lattice. In a recent letter[4] (hereafter GZ) they have used a Hubbard-like model Hamiltonian for solid ^{3}He with a single ^{4}He atom present as an impurity, to construct an effective Hamiltonian which describes the exchange of positions of atoms caused by the tunneling motion of the atoms through the lattice in the absence of vacancies. They have found the concentration dependence and the temperature dependence of the relaxation rate in the above case. In this paper, we develop a unified approach for the study of tunneling processes that occur when vacancies are present in the lattice in addition to the ^{4}He atom.

The model Hamiltonian used in GZ may be written in the form

$$H = H_0 + V, \quad (1)$$