FREQUENCY SHIFTS IN SELF-FOCUSED LIGHT*

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The stimulated Raman, Brillouin, and Rayleigh-wing scattering of molecular liquids frequently yields narrow lines with unique frequency shifts.[1-3] Under certain conditions, however, an extensive frequency-broadening effect[1,3,4] is noticed. It can be induced by a multimode laser and arises through iteration of stimulated Rayleigh-wing scattering.[5] In this Letter, a new broadening mechanism is described which results in additional and somewhat variable frequency shifts. These are shifts initiated through large temporal variations in the nonlinear refractive index, resulting from the molecular-orientational Kerr effect, and they have now been observed and identified when intense light waves self-focus.[6] The dominant Fourier components also can be amplified by stimulated two- and four-photon[7] Rayleigh-wing scattering, and through iteration, the spectrum can widen into a multitude of Stokes and anti-Stokes orders near the laser and Raman wavelengths.

A theoretical description of these effects is not given here, but clearly frequency broadening in the steady-state regime[5] must be extended to cover nonstationary time variations. Such a theory would necessarily include the related topic of light pulse distortion.[8,9]

Characteristic spectra in the vicinity of the ruby-laser wavelength 6940 Å are shown in Fig. 1 for individual filaments of self-trapped light. These filaments are formed in a 10-cm long cell by the collimated output of a Q-spoiled ruby laser beam which is apertured down to a 500-μ diam and to ~100-kW power. Light from the exit plane of the cell is imaged by a microscope (50×) onto either a camera of a stigmatic-grating spectrograph, with its slits removed, or that of a Fabry-Perot interferometer. In this way, the spectral and spatial content of single filaments of self-trapped light can be monitored simultaneously. Thus, with the grating, the spectrum of each filament tends to appear as a streak whose width is the filament diameter; with the interferometer, the fringes are confined to a circular zone of the same diameter.

The spectra are seen to vary considerably from filament to filament, probably in accordance with the power contained and their stage of development. The shifts to lower frequencies are dominant as expected. In Fig. 1(a), the shift for CS_2 is -11 cm^{-1} and does not correspond to known values for the stimulated Raman (656 cm^{-1}) and stimulated Brillouin (0.2 cm^{-1}) scattering or to the predicted value for stimulated Rayleigh-wing scattering (3 cm^{-1}).[5,10] Appropriate multiples or combinations of these

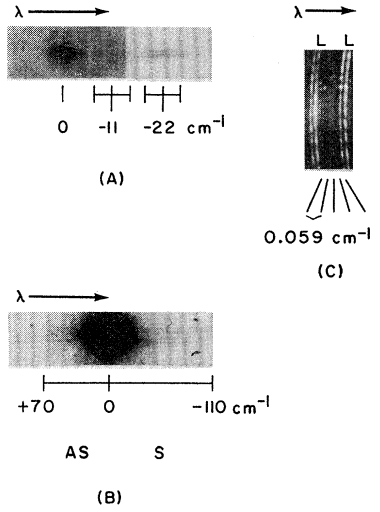

FIG. 1. Spectra of individual filaments of self-focused light in the vicinity of the ruby-laser wavelength 6940 Å. (a) and (b) were obtained with a grating spectrograph (resolution: ~10^5) with its slits removed and (c) with a Fabry-Perot interferometer (resolution: ~3 ×10^6). Wavelength increases to the right in each case. (a) CS_2. Negative copy. Tic marks identify two Stokes orders and image of filament itself (diam: ~4μ), while horizontal lines reproduce linewidth. Nonfilamented laser light to the left outlines entrance aperture of grating spectrograph. (b) CS_2. Negative copy. Unresolved Stokes (S) and anti-Stokes (AS) orders are shown where each streak identifies a single filament, and the extent of broadening is indicated. Central region contains filamented and nonfilamented light, and it is overexposed in reproduction to elevate streaks. (c) Nitrobenzene. Positive copy of interferogram showing five Stokes orders. The two laser wavelengths (L) are separated in time by ~1μ sec and thus are not simultaneous modes; their separation in frequency is 0.092 cm^{-1}. Filament diameter is ~10μ.

frequencies are unlikely as well. Furthermore, this shift is not unique but can vary by a factor of ~5 or more from one filament to the next in the vicinity of the Rayleigh wing. It is also significant that the line shape of the scattered light does not join onto the exciting line; instead, the two are separated by a clear gap, indicative of a shift. This result also excludes iterative stimulated Rayleigh-wing scattering excited by different laser modes.[5] However, the exciting and scattered light wave can generate higher order Stokes shifts (of 11 cm^{-1}, for example) by this same two-photon process, as shown in Fig. 1(a). In this manner, the spectrum near the laser wavelength can extend as far as ~100 cm^{-1} for some filaments in CS_2 and to a lesser extent in other liquids.

Figure 1(c) shows similar results for self-trapped light in nitrobenzene, but at a much higher resolution as the shifts are smaller. The Fabry-Perot interferograms show nonfilamented laser light as well, which appears as a background and serves as a reference wavelength. It is found that Raman light does not contribute to this spectrum. Five orders of stimulated Rayleigh-wing scattering are shown in which the interorder shift is 0.059 cm^{-1}. The spacing between Fabry-Perot orders is 0.505 cm^{-1}, and there is no evidence for overlapping orders in the Rayleigh spectrum. It is worth mentioning that under lower resolution these multiorder shifts would merge, and the effect could be misinterpreted as line broadening. Other filaments give different shifts such as 0.032, 0.057, 0.093, and 0.125 cm^{-1}, which are iterated also. In general, these are not related in any obvious way to either the mode spacing of the laser (0.010 cm^{-1}) or the known shifts for stimulated light scattering. The last value is an exception apparently as it agrees with the stimulated Rayleigh shift $\Delta\nu = -1/(2\pi\tau)$,[3] where τ is the molecular orientational relaxation time. The magnitude of these shifts is seen to be considerably less than in CS_2, partly because the index change may be less and partly because the orientational relaxation time of nitrobenzene is about 30 times larger.

Evidence also exists for stimulated four-photon Rayleigh-wing scattering under self-focusing conditions. This is indicated by the presence of both Stokes and anti-Stokes components as shown in Fig. 1(b) for CS_2. While the fundamental shift is not resolved here, sharp-line spectra of Stokes and anti-Stokes components have been observed in nitrobenzene. (The spectra are not suitable for reproduction, however.) Again, the shift varies from one filament to the next and for nitrobenzene over the approximate range 0.03 to 0.12 cm^{-1}.

Thus, both two- and four-photon stimulated Rayleigh-wing scatterings are observed in self-focused beams. In each case, however, the frequency dependence predicted from steady-state theories is usually not obeyed under self-focusing conditions. The two-photon stimulated Rayleigh-wing scattering, for example, is expected to exhibit maximum gain when the shift is $\Delta\nu = -1/(2\pi\tau)$.[3,5] For the four-photon case, the gain is expected to be a maximum for zero-frequency shift and to decrease continuously with increasing shift for any angle in near forward scattering.[7] What is neglected in these theories is the temporal dependence and thus the possibility of generating new frequencies through self-focusing itself. When the initial intensity of these components exceeds that for spontaneous Rayleigh scattering, amplification by two- or four-photon scattering will be favored at the new shifts, causing the spectrum to deviate from the steady-state behavior as observed.

Stimulated Raman light which is self-trapped is observed to undergo a similar behavior, although the Raman broadening can be about two to three times more extensive than at the laser wavelength, and sometimes more than one fundamental shift is operative making the spectrum rather complex. These effects are not well understood although multiple self-focusing is a likely possibility for there being more than one shift.

The evidence presented here reveals for the first time that frequency broadening in stimulated light scattering is not a line broadening at all but actually has its origin in multiorder frequency shifts which in some filaments may be very closely spaced. The observation of these shifts solely in self-focused light beams and their variability points strongly to self-focusing as the causative mechanism.

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OBSERVATION OF PLASMA ION OSCILLATIONS IN A LASER-PRODUCED PLASMA*

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Experiments with 6943-Å light scattered from a high-density carbon plasma have resulted in the spectroscopic observation of ion wings produced by plasma ion oscillations. For an energy density of 200 J/cm^2 and a pulse half-width of 90 nsec, the maximum separation of the ion wings was found to be 0.9 Å. This corresponds to a plasma electron temperature of more than 25 eV and a density of 10^{19} electrons/cm^3.

Since the invention of very intense, monochromatic light sources in the form of lasers, the scattering of light from plasmas has received much attention because of its potential as a plasma diagnostic tool. It is, for instance, possible to obtain most of the important plasma parameters, such as the electron temperature (T_e), ion temperature (T_i), and electron density (n_e) from the spectral intensity distribution of the scattered light.

Consideration of the electron and ion density fluctuations in a plasma has shown that the resulting spectrum of the scattered light consists of a superpositioning of two spectra.[1-3] One of these is due to the high-frequency plasma electron oscillations and is characterized by a satellite peak on either side of ω_0, where ω_0 is the frequency of the incident light. The other spectral component is due to the lower frequency plasma ion oscillations and should manifest itself by splitting the ω_0 line into two symmetric components (ion wings) separated from each other by ~1 Å for a laser-produced high-density plasma. In recent months a number of authors have reported the observation of the spectrum caused by the plasma electron oscillations.[4-6] Depending on the electron density, these broadened satellite lines are generally found around 6900 and 6980 Å if a ruby laser is used. However, because of insufficient resolution, too much stray light, or a too low plasma density, the splitting of the 6943-Å line due to plasma ion oscillations has so far evaded detection.

Recent experiments conducted in this laboratory with light scattered from a ruby-laser-produced plasma have now resulted in the observation and measurement of the spectrum produced by the plasma ion oscillations.

The plasma was created by focusing the beam of a high-power oscillator-amplifier, ruby-laser system onto the flat end of a $\frac{1}{8}$-in.-diam rod of pyrolitic graphite. The Q-switched oscillator was triggered with a rotating dielectric reflector and gave an output of about 2 J with a pulse width of 90 nsec. The gain through the amplifier ruby was about fourfold.

The target was mounted on a stand inside a vacuum chamber. All experiments were conducted at a pressure of 10^{-5} Torr. A positive

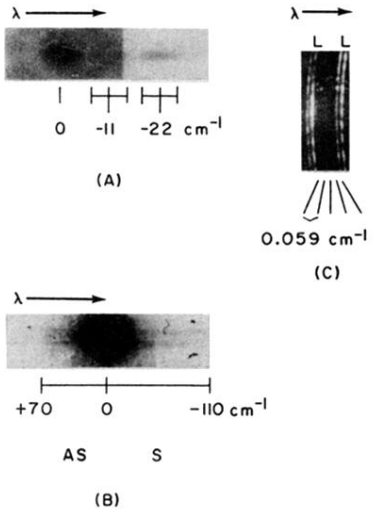

FIG. 1. Spectra of individual filaments of self-focused light in the vicinity of the ruby-laser wavelength 6940 Å. (a) and (b) were obtained with a grating spectrograph (resolution: $\sim 10^5$) with its slits removed and (c) with a Fabry-Perot interferometer (resolution: $\sim 3 \times 10^6$). Wavelength increases to the right in each case. (a) CS_2. Negative copy. Tic marks identify two Stokes orders and image of filament itself (diam: $\sim 4\mu$), while horizontal lines reproduce linewidth. Nonfilamented laser light to the left outlines entrance aperture of grating spectrograph. (b) CS_2. Negative copy. Unresolved Stokes (S) and anti-Stokes (AS) orders are shown where each streak identifies a single filament, and the extent of broadening is indicated. Central region contains filamented and nonfilamented light, and it is overexposed in reproduction to elevate streaks. (c) Nitrobenzene. Positive copy of interferogram showing five Stokes orders. The two laser wavelengths (L) are separated in time by $\sim 1\mu$ sec and thus are not simultaneous modes; their separation in frequency is 0.092 cm^{-1}. Filament diameter is $\sim 10\mu$.