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STUDY OF SPECTRAL BROADENING IN A FILAMENT OF LIGHT

George K. L. Wong and Y. R. Shen

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We have measured with nanosecond laser pulses the spectral broadening of light emitted from a filament under controlled input conditions. Our results are in semi-quantitative agreement with predictions from the moving focus model.

It has been shown recently that at least for nanosecond pulse excitation, moving foci are responsible for the observation of small-scale filaments in Kerr liquids.^{1,2} Most observed characteristics of the filaments can be explained by the moving focus model. In particular, spectral broadening of light from a filament can be interpreted as due to phase modulation acquired by the self-focused light in traversing the nonlinear medium.³ However, no controlled, systematic experiment on spectral broadening has yet been performed to check the validity of the interpretation. Such experiment is important in view of the fact that spectral broadening has long been used as evidence to support the self-trapping model.⁴⁻⁶ In this paper, we would like to report the results of the first experiment on spectral broadening with nanosecond input pulses under controlled input conditions. We show that there exists, in fact, semi-quantitative agreement between theory and experiment. Induced partial trapping or nonlinear diffraction of light in a filament can occur,^{1,3} but its effect is only on the detailed structure of the broadened spectrum.

In our experiment, we used a single-mode ruby laser pulse switched out from a train of weakly mode-locked pulses. The pulse width was about 1.2 nsec, the beam diameter was about 300 μm , and the maximum peak power used in the experiment was about 40 kW in CS_2 and 120 kW in toluene. Such a pulse consistently yielded a single filament in self-focusing. The spectrum of light emitted from a filament at the end of the cell was observed by imaging the filament with a 10 $\times$ magnification onto the widely opened entrance slit of a Jarrell-Ash 1.5-m Fastic spectrograph. Kodak 1-N plates were used to record the

spectra. For each shot, we monitored simultaneously the input laser pulse by an ITT F4018 fast photodetector in connection with a Tektronix 519 oscilloscope.

We have performed the experiment at various input power levels on CS_2 with four different cell lengths, 5, 10, 15, and 20 cm, and on toluene with two different cell lengths, 10 and 15 cm. The results are shown in Figs. 1 and 2 as discrete data points. We plot in Fig. 1 the extent of spectral broadening on the Stokes side, $\Delta\omega_{\text{max}}$, as a function of the input peak power for two different cell lengths in CS_2 and in toluene, and in Fig. 2, $(\Delta\omega)_{\text{max}}$ as a function of the cell length for an input peak power of about 28 kW in CS_2 . We also show in Fig. 3a a typical spectrum of a filament obtained in our experiment.

According to the moving focus model,³ spectral broadening on the Stokes side is given approximately by

$$\Delta\omega_{\text{max}} \cong (\omega_0/c)(n/c - 1/v)^{-1}(\Delta n_{\text{max}}^*)_{\ell} \quad (1)$$

where ω_0 is the incoming laser frequency, c is the light velocity in vacuum, v is the velocity of the moving focus at the end of the cell ($z = \ell$), n is the refractive index of the medium, and $(\Delta n_{\text{max}}^*)_{\ell}$ is the maximum effective field-induced refractive index at the end of the cell.⁷

We can determine v in Eq. (1) from measurements as follows. We assume that for nanosecond input pulses in Kerr liquids, the position of the moving focus obeys the equation⁸

$$z_f(t) = (K/P_{\text{cr}}^{1/2}) / [(P(t')/P_{\text{cr}})^{1/2} - 1] \quad (2)$$

with $t' = t - nz_f(t)/c$, where $P(t)$ is the power of the input pulse varying with time, and K and P_{cr} are coefficients which depend on the characteristics of the medium and of the input laser beam. We can obtain $K/P_{cr}^{1/2}$ and P_{cr} experimentally using the method of Wang.⁹ In our experiment, we found $K/P_{cr}^{1/2} = 2.2$ cm and $P_{cr} = 8$ kW in CS_2 , and $K/P_{cr}^{1/2} = 2.0$ cm and $P_{cr} = 30$ kW in toluene. Knowing K and P_{cr} , we can plot z_f vs t from Eq. (2). The resulting U curve describes very well the motion of the focus as has been shown experimentally.¹ Then, from $(\partial z_f / \partial t)_{z=l}$, we can obtain v . We notice that for a given input pulse shape, v is a function of the input peak power and the cell length.

It is more difficult to find $(\Delta n_{max}^*)_l$ since $(\Delta n_{max}^*)_l$ depends not only on the actual intensity of the focus at $z = l$, but also on the detailed focusing geometry. However, from the moving focus model,³ we expect that as long as the focus is moving towards the end of the cell, $(\Delta n_{max}^*)_l$ should decrease with the cell length since the beam self-focused at a longer distance has a smaller power, but for a fixed cell length, $(\Delta n_{max}^*)_l$ is nearly independent of the input peak power. If we assume instantaneous response of the medium to the field, then the field-induced refractive index at the focus is $\Delta n_o = (8/ca^2)n_2P$ where a is the $(1/e)$ radius of the focus, and $n_2 = 1.1 \times 10^{-11}$ esu for CS_2 and 2.5×10^{-12} esu for toluene. For $P = 10$ kW in CS_2 , we have $\Delta n_o = 2.5 \times 10^{-3}$. Experimental measurements suggested that the induced refractive index in the focus should be around $1-2 \times 10^{-3}$.¹⁰ Since the response of a medium is never instantaneous, and we do not expect all the power in the beam to self-focus into a filament, the quantity $(\Delta n_{max}^*)_l$, which also contains a negative contribution from

diffraction, should certainly be smaller than the maximum Δn_0 at $z = \ell$.

In our experiment, when the cell length ℓ of CS_2 was increased from 5 cm to 20 cm, the corresponding input power $P(z_f = \ell)$ which focused at ℓ changed from 16 kW to 9.8 kW. We assume

$(\Delta n_{\max}^*)_{\ell} = (2.4 - 8.5 \times 10^{-2}\ell) \times 10^{-3}$ for CS_2 . We can then calculate from Eq. (1) the spectral broadening $\Delta\omega_{\max}$ using this expression of $(\Delta n_{\max}^*)_{\ell}$ as well as the value of v obtained from Eq. (2) with the

experimentally measured input laser pulse. The results are shown as solid curves in Figs. 1 and 2. They appear to fit the experimental data points fairly well. We also assume for toluene $\Delta n_{\max}^* = 4 \times 10^{-4}$ and 2.5×10^{-4} for $\ell = 10$ and 15 cm respectively. The calculated curves of $\Delta\omega_{\max}$ vs the input peak power for two different cell lengths again describe the data points fairly well as shown in Fig. 1.

We noticed that the typical spectrum of a filament we obtained in our experiment had a strong central peak superimposed on the relatively weak semiperiodic broadened structure, as shown in Fig. 3a. This central peak shows that part of the light emitted from the filament has experienced little phase modulation. In the actual self-focusing process, the portion of light coming from the periphery of the beam self-focuses and diffracts abruptly, and is essentially phase-unmodulated because of its short path in the region with large Δn . The central portion of the beam, on the other hand, stays in the region of large Δn much longer and is more strongly phase-modulated. In a longer cell and with a shorter input pulse, the focusing is more gradual and the relative intensity of the phase-unmodulated part should decrease. For the phase-modulated part, partial trapping or nonlinear diffraction

of light in the dielectric channel induced by the moving focus may give rise to a stronger phase modulation. In the limit of total trapping, one would expect to see only the broadened, semiperiodic spectrum without the central peak.

Thus, we believe that light emitted from the filament is a coherent superposition of a phase-modulated part and a phase-unmodulated part.

$$E \cong [\mathcal{E}_1(t)e^{i\Delta\phi(t)} + \mathcal{E}_2(t)]e^{i\phi_0 - i\omega_0 t} \quad (3)$$

where $\mathcal{E}_1$ and $\mathcal{E}_2$ are the amplitudes of the fields with and without phase modulation respectively. As long as the spectral widths of $\mathcal{E}_1(t)$ and $\mathcal{E}_2(t)$ are much less than $\Delta\omega_{\max}$, the extent of the spectral broadening is essentially independent of $\mathcal{E}_1(t)$ and $\mathcal{E}_2(t)$. However, the detailed structure of the broadened spectrum is actually governed by the functions of $\mathcal{E}_1(t)$ and $\mathcal{E}_2(t)$, which can only be determined from the actual self-focusing dynamics.

As an example, we have shown in Fig. 3b a power spectrum calculated from Eq. (3) by assuming

$$\begin{aligned} \mathcal{E}_1(t) &= C(t/1.6)^{1/2} && \text{for } 0 \leq t \leq 1.6 \\ &= C && \text{for } 1.6 \leq t \leq 3.2 \\ &= C \exp[-(t - 3.2)/11.2] && \text{for } t \geq 3.2 \end{aligned}$$

with the corresponding $\Delta\phi(t)$ evaluated as in Ref. 3, and $\mathcal{E}_2(t) = C' \exp[-(t - 2.4)/0.8]$, where $C/C' = 1/3$ and t is in psec. The two spectra in Figs. 3(a) and (b) show qualitative resemblance, but a better approximation of $\mathcal{E}_1(t)$ and $\mathcal{E}_2(t)$ is necessary in order to achieve a more

quantitative agreement.

In conclusion, we should mention that Denariez-Roberge and Taran⁵ have performed a somewhat similar experiment. However, they used in their experiment a multimode Q-switched ruby laser, and more than 50 filaments appeared in each shot. Consequently, they were not able to specify the input conditions for the formation of individual filaments. They have interpreted their results using the self-trapping model. We believe that with appropriate assumptions on the input conditions, we can also interpret their results with the moving focus model (which allows induced partial trapping as we have suggested).³ Clearly, for the results to be quantitatively meaningful, experiments with controlled input conditions are necessary. The semi-quantitative agreement of our experimental results with the theoretical predictions gives us yet another support to the moving focus model at least in the case of nanosecond input pulses.

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FIGURE CAPTIONS

Fig. 1. Maximum Stokes broadening vs input peak power for given cell lengths. $\circ$ — 15 cm of CS_2 ; Δ — 10 cm of CS_2 ; ∇ — 15 cm of toluene; $\square$ — 10 cm of toluene. The solid curves are calculated from Eq. (1) with an input pulse of 1.2-nsec full width at $1/e$ points, and with $(\Delta n_{\text{max}}^*)_{\ell} = (2.4 - 8.5 \times 10^{-2}\ell) \times 10^{-3}$ for CS_2 and $(\Delta n_{\text{max}}^*)_{\ell} = (7 - 0.3\ell) \times 10^{-4}$ for toluene.

Fig. 2. Maximum Stokes broadening vs cell length of CS_2 for an input peak power of about 28 kW. Each point is an average of three to four shots. The solid curve is calculated from Eq. (1) with an input pulse of 1.2 - nsec full width at $1/e$ points and with $(\Delta n_{\text{max}}^*)_{\ell} = (2.4 - 8.5 \times 10^{-2}\ell) \times 10^{-3}$.

Fig. 3. (a) Micro-densitometer trace of a typical observed spectrum of light emitted from a filament corresponding to a CS_2 cell length of 10 cm and an input peak power of 27 kW.

(b) Corresponding calculated spectrum (see the text).

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Fig. 1

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Fig. 2

Fig. 3

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