

Experimental Study of Small-Scale Filaments of Light in Liquids

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We show, with a single-mode Q -switched laser and a single-mode mode-locked laser, that the observed small-scale filaments are actually the tracks of moving foci, but under suitable conditions, light can be trapped over an appreciable distance in these tracks. The results are in rough agreement with theoretical prediction.

It is well-known that self-focusing of a laser beam leads to the formation of small-scale filaments.¹ These filaments can be interpreted by either the self-trapping model² or the moving-focus model.³ The former is supported by experimental results obtained with multimode Q -switched lasers⁴ or mode-locked lasers,⁵ and the latter by results obtained with single-mode Q -switched lasers.⁶ Recently, we have suggested that while self-focusing of an input laser pulse should indeed yield a moving focal spot, a trapped filament of appreciable length can, however, exist in the dielectric waveguide established by the moving focal spot if the velocity of the focal spot approaches the light velocity.⁷ In this paper, we first show, with quantitative experimental results, that self-focusing of a Q -switched laser pulse leads to a moving focal spot. We then show, with a weakly mode-locked laser, that trapped filaments indeed exist as one would predict from the theory.⁷

If the stationary theory of self-focusing is approximately valid for a laser pulse,⁸ then at time t , the focal spot should appear at the self-focusing distance^{8,9}

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

$$K = (n/4)(a^2/f)(c'n/n_2)^{1/2},$$

where $P(t)$ is the input laser power at time t , P_{cr} is the critical power for self-trapping,² a is the beam radius, f is a parameter of the order of 1,⁹ c' is the light velocity in the medium, and n and n_2 are the linear and nonlinear refractive indices, respectively. The focal-spot motion is then completely described by Eq. (1) if K , P_{cr} , and $P(t)$ are known. Note that the self-focusing distance is greatly reduced if, with the same P , the beam radius is decreased.

In our experiments, a single-mode Q -switched ruby laser was used. A typical laser pulse had a peak power of about 100 kW and a duration of around 8 nsec. The spectral width of the laser output is 0.02 cm^{-1} . With such a laser beam

shining into a cell of CS_2 or toluene, we consistently observed only a single "filament" in the self-focused beam. In order to decrease the self-focusing distance, we used an inverted telescope to reduce the beam diameter to $300 \mu\text{m}$. It is seen from Eq. (1) that by measuring the threshold power for self-focusing at various cell lengths, one can find the quantities K and P_{cr} . Then knowing the input laser pulse $P(t)$, one can plot out the position of the focal spot as a function of time. This was done for the beam in toluene. A typical curve of z_f vs t , corresponding to a given input pulse $P(t)$ (measured by the fast detection system composed of an ITT F4018 photodiode and a Tektronix 519 oscilloscope), is given in Fig. 1.

Figure 1 indicates that the focal spot should first appear at a distance of 23 cm inside the cell. Then it should immediately split into two, one moving forward and the other first backward and then forward. The one moving forward al-

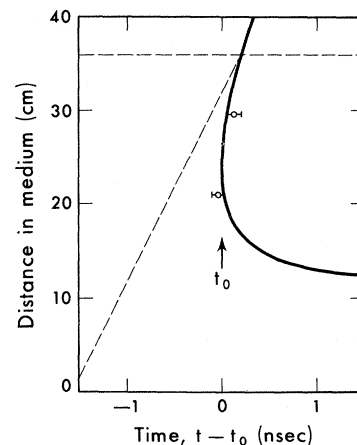


FIG. 1. Position of the focal spot of a self-focused beam as a function of time in a toluene cell of 36 cm long. The solid curve was calculated from Eq. (1) using experimentally determined parameters K , P_{cr} , and $P(t)$. The dots with the error bars at 21 and 29.5 cm are results obtained from direct measurements with respect to the focal spot appearing at the end of the cell. The dashed line with a slope equal to the light velocity is shown for comparison.

ways moves with a velocity faster than the light velocity. In order to verify this, we set up an experiment to measure the focal-spot movement directly. A 36-cm toluene cell was used and a beam splitter (a microslide of 100 μm thick) was inserted in the cell at a distance d away from the end of the cell. Focal spots of about 10 μm in diameter were observed, both at the beam splitter and at the end of the cell. The defocused light from both focal spots was now collected with appropriate optical delay by the same photodiode and two pulses with duration less than 0.1 nsec showed up on the oscilloscope. The input laser pulse was also monitored simultaneously. With our fast detection system, the time lag Δt between the two short pulses can be measured to within ± 0.08 nsec. Thus, for $d = 6.5$ cm and 15 cm, we found $\Delta t = 0.08$ nsec and 0.25 nsec, respectively. These values agree well with the values 0.15 and 0.20 nsec obtained directly from the curve in Fig. 1. If the two pulses had come from a short light pulse traversing the cell (e.g., from a light pulse propagating in a trapped filament), we would have found $\Delta t = 0.32$ and 0.75 nsec, respectively. Our experimental results showed clearly that the pulses came from a focal spot which was moving forward with a velocity faster than the light velocity.

We also recorded the spectrum of light emitted from the focal spot at the end of the cell with a Fabry-Perot interferometer. We found that the spectral width was about 0.3 cm^{-1} . It increased slightly with a longer cell. In comparison with the laser spectral width of 0.02 cm^{-1} , this is a definite spectral broadening. However, from the theory of Gustafson *et al.*,¹⁰ this amount of spectral broadening should correspond to a trapped filament of length less than 2 mm. We are therefore safe in saying that, in the present case, the observed filament was the result of a moving focal spot rather than a trapped filament. In fact, the small spectral broadening is what one would expect from the moving-focus model.⁷

Since light was not trapped in the filament, we expected that we could also detect the focal spot by focusing the camera inside the cell.⁶ Photographs of the beam cross section inside the cell indeed showed a Raman spot of $\sim 10 \mu\text{m}$ in diameter, but showed no clear laser focal spot. We believed that the disappearance of the laser spot was due to depletion of the laser radiation by the stimulated Raman process in the focal region. This was possible if the focal spot extended over a distance of more than 1 mm.¹¹ In the earlier

investigation with the focal spot moving backward,⁶ we did find the laser focal spot inside the cell, but in that case, the focal spot moved much more slowly. Consequently, there was enough undepleted laser energy emitted from a local focal spot for it to be detectable.

We would also expect to see a focal spot start at 23 cm inside the cell and move backward (see Fig. 1). However, this focal-spot movement was quickly terminated when self-focusing was terminated by the backward stimulated Raman and Brillouin scattering through depletion of the incoming laser power. This was seen from comparison of the oscilloscope traces of the incoming and the transmitted laser power.¹²

As we mentioned earlier, light diffracted from a focal spot can be partially trapped in the temporary channel of Δn set up by the focal spot moving ahead of it. The trapping can be over a long distance if the focal-spot velocity v_f approaches the light velocity c' . In order to have $v_f \approx 1.1c'$ (for light to be trapped over a maximum length of a few centimeters in toluene) for the case in Fig. 1, we must have a cell length of ~ 100 cm.⁷ On the other hand, this condition can be reached with a cell length of 35 cm if the duration of the input laser pulse is changed to ~ 1.5 nsec. Such a laser pulse can easily be obtained by weakly mode-locking a ruby laser.

To verify our prediction, we used a mode-locked ruby laser with a single transverse mode. The length of the cavity was 100 cm and the beam diameter was $\sim 250 \mu\text{m}$ at the entrance window of the cell. Typical mode-locked trains are shown in Fig. 2, where each mode-locked pulse has a full width at half-maximum of 1.6 nsec and the peak power of the highest pulse is about 120 kW. The spectral width of the laser output was always equal to the inverse of the pulse duration. Photographs taken at the end of the cell showed a single filament for each laser shot. However, the single filament on the photograph was in fact a superposition of several (often two or three) filaments created consecutively by several pulses in the mode-locked train. This was seen from the oscilloscope trace of the filament pulses shown in Fig. 2(a). That self-focusing or filament formation was terminated after a few pulses was probably due to thermal or acoustic effect resulting from stimulated light scattering in the liquid. In Fig. 2(b), we recorded simultaneously the Raman pulses emitted from the filaments. The stimulated Brillouin scattering was not observed, presumably being suppressed because

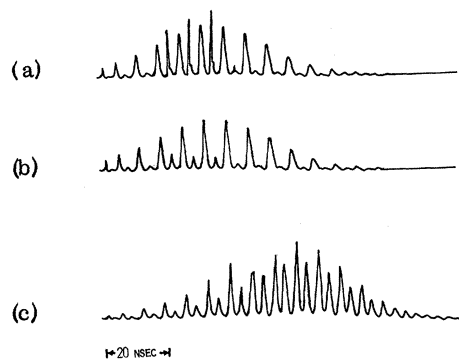


FIG. 2. (a) Interleafed input laser-pulse train and filament pulses. The laser pulses were optically delayed by 6 nsec with respect to the filament pulses. Three filament pulses appeared in this shot. (b) Interleafed input laser-pulse train (optically delayed by 6 nsec) and Raman Stokes pulses from the filaments recorded simultaneously with (a). (c) Interleafed input and transmitted laser-pulse trains showing depletion of laser energy. The input pulses were optically delayed by 6 nsec with respect to the transmitted pulses. They correspond to the train with lower amplitude.

of the transient effect. Whenever the filament pulses were present, the laser pulses were clearly depleted, as shown in Fig. 2(c). Comparison of the energy in the Raman pulses with the depleted energy in the laser pulses showed that the laser power was mainly depleted by stimulated Raman scattering.¹³

In order to study the spectrum of light emitted from the filament, we used a Jarrell-Ash 1.5-m Fastie spectrograph with its entrance slit open wide. The magnified ($\times 10$) image of the filament at the end of the cell was recorded. A typical spectrum of the filament is shown in Fig. 3(a). The appreciable spectral broadening and its semiperiodic structure are manifestations of significant phase modulation of light emitted from the filament.¹⁴ The semiperiodic structure is somewhat smeared because the spectrum is actually

a superposition of the spectra of several filaments as we mentioned earlier. A dot with no spectral broadening is also visible at the laser frequency on the spectrum. It corresponded to the image of the $\sim 10\text{-}\mu\text{m}$ filament, and apparently came from the nontrapped part of the self-focused beam. The Stokes Raman spectrum from the filament is also shown in Fig. 3(a). It has the same characteristics as the spectrum around the laser frequency and shows that the Raman radiation from the filament is even more strongly phase-modulated.

From our theoretical analysis,⁷ we expected to find, in our case, a broadened spectrum of the order of 100 cm^{-1} if the peak power of a mode-locked pulse was above 100 kW. For lower peak power, the broadening would be less, and for peak power less than 60 kW, we expected to see no appreciable spectral broadening at both laser and Raman frequencies. This was roughly what we observed. At high peak power, the superposition of the spectra of several filaments and the effects of the filaments on one another made quantitative analysis difficult. Denariez-Roberge and Taran⁴ showed that spectral broadening increased with the filament length. This can also be shown to be in qualitative agreement with our model.

To see whether light is trapped in the filament, we focused the image of the beam cross section at several centimeters inside the cell on the spectrograph. No extended spectral broadening was ever seen at either the laser or the Raman frequency. The fact that only light emitted from filaments at the end of the cell showed spectral broadening gave clear evidence that it was the trapped part of the light which gave rise to the broadened spectrum. Occasionally, we observed clear images of the focal spot at both laser and Raman frequencies, as shown in Fig. 3(b). They corresponded to the case where the peak power

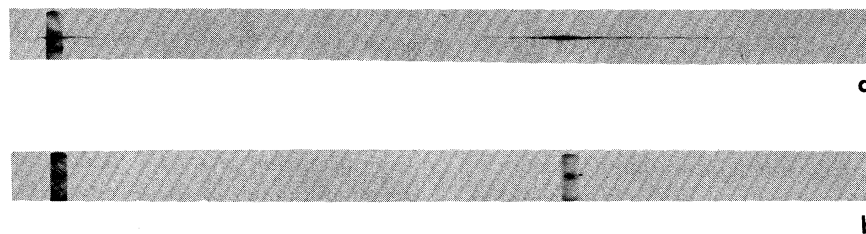


FIG. 3. Laser and Raman Stokes spectra of a filament created by self-focusing of 1.6-nsec mode-locked pulses in a 37-cm toluene cell (a) taken with the spectrograph focused at the end of the cell, (b) taken with the spectrograph focused inside the cell (at 1.5 cm from the end of the cell). The laser spectra are on the left with the slit images centered at $14\,402\text{ cm}^{-1}$, and the Stokes spectra on the right with the slit images centered at $13\,400\text{ cm}^{-1}$.

of the input pulse was low, so that the self-focused light was not being trapped over any appreciable distance, with the result of little spectral broadening.

Although the results presented here are all on toluene, we have performed similar experiments on CS₂. There was no qualitative difference between the two cases, except that spectral broadening in CS₂ was several times more appreciable. We therefore conclude, from our experimental results, that the observed small-scale filaments are actually composed of moving focal spots, but under suitable conditions, the self-focused light can be partially trapped in the dielectric channels established temporarily by the moving focal spots. The condition for trapping can be easily fulfilled with the use of a mode-locked laser or a multimode Q-switched laser.

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Thermal Periodicity of Filament Formation and Elastic Lattice Vibrations

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The mechanism of filament formation, consisting in resonant enhancement of elastic lattice vibrations, is responsible for thermal periodicity both of number of filaments and of intensity of stimulated Raman scattering. Mechanical tuning and some test experiments are discussed.

Lately, new experiments in the field of self-trapping effects have been made. Namely, the filament formation is shown to occur with picosecond pulses,^{1,2} in ionic crystals^{3,4} and in liquid argon.⁵ These experiments point to the fact that the generally admitted mechanism based on the optical Kerr effect, as well as other mechanisms proposed *ad hoc*, are not suitable to explain light trapping. Instead, a resonant enhancement of elastic lattice vibrations has been proposed by present author.⁶

Quite recently a thermal periodicity of the num-

ber of filaments produced has been found,⁷ preceded by the discovery of thermal periodicity of the stimulated Raman scattering (SRS) intensity.^{7,8} The period for both of these phenomena for different liquids (benzene, toluene, CS₂, etc.) is not very different from 1°. It seems that the primary phenomenon is the periodicity of filament number resulting in a similar periodicity of the SRS intensity. This basic phenomenon of filament formation is to be understood in terms of resonant interaction between self-modulated light pulse and thermal lattice vibration in liquids.⁹

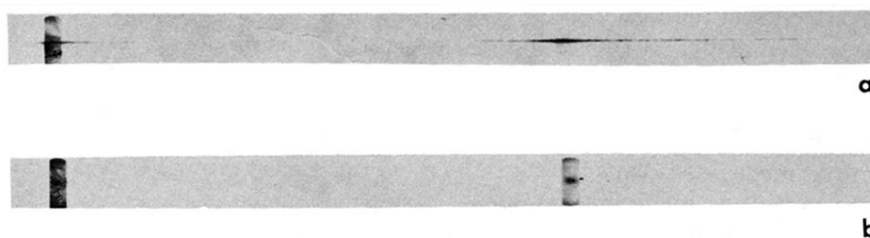


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