SMALL-SCALE FILAMENTS IN LIQUIDS AND TRACKS OF MOVING FOCI*

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Experimental results show that the small-scale filaments in liquid, obtained with a single-mode laser, are composed of continuous series of focal spots. Many related observations are shown to be consistent with the picture of moving foci.

Experimental observation on self-focusing of laser light in liquids has shown that the self-focused beam would eventually break into intense filaments a few microns in diameter.[1,2] The existence of these filaments has recently attracted much attention. They have been attributed to self-trapping predicted by Chiao, Garmire, and Townes.[3] Recent experiments[4] on spectral broadening in filaments, obtained by using an inhomogeneous, multimode laser, seem to support this assertion. Lugovoi and Prokhorov[5] however suggest that in some situations filaments are simply tracks of moving foci, in accordance with time variation of the input laser intensity. In this paper, we would like to present some experimental evidence that filaments obtained with a single-mode laser are actually composed of a continuous time series of focal spots. We also show that many effects inherently related to self-focusing are consistent with the picture predicted by Lugovoi and Prokhorov.

A single-mode ruby laser, Q switched by cryptocyanine, was used in the experiments. The beam was passed through a 0.75-mm pinhole before propagating into the liquid cell in order to assure maximum spatial homogeneity. A typical oscilloscope trace of input pulses is shown in Fig. 1(a) together with the Fabry-Perot pattern in Fig. 1(b). "Filaments" or moving focal spots were observed by focusing a camera at the end of the cell. In most cases only one "filament" appeared [Fig. 1(c)]; occasionally, there were two, when the input laser power was exceptionally high. This is in clear contrast to the results obtained with a multimode laser, where tens or even hundreds of filaments are frequently observed on each picture.[4] As the laser power was increased from below to above the self-focusing threshold, the photograph first showed a bright spot of about 50 μ in diameter, which gradually became more intense and shrank to a more or less limiting "filament" size (10 ± 2 μ in diameter in toluene and 5 ± 1 μ in CS_2). The pulse emitted from the filament was detected by an ITT F4018 photodiode in combination with a Tektronix Model No. 519 oscilloscope. A 1-mm disk was inserted somewhere in front of the photodiode to block off the background of non-self-focused laser light.[1] The pulse duration was then measured by the convolution technique.[6] The results on toluene showed that with increasing input laser power, as the bright spot shrank from 50 μ to the limiting 10-μ "filament" size, the pulse duration changed from 1 nsec to 200 psec, and then as the "filament" size remained unchanged, the pulse duration continued to shorten to less than 100 psec. While the pulse became shorter, the energy content in the pulse decreased accordingly, but the peak intensity in the limiting "filament" remained roughly constant at 30 GW/cm^2. Spectral analysis with spectograph and Fabry-Perot on the "filament" pulses yielded a linewidth of less than 1 cm^{-1}.

These results are consistent with the picture of "filaments" formed by moving foci. On the other hand, it would be rather difficult to ex-

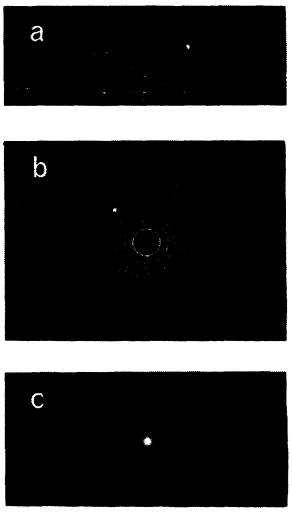

FIG. 1. (a) A typical oscilloscope trace (5 nsec/div) of an input laser pulse. (b) A Fabry-Perot pattern (1.25-cm spacing between plates) of an input laser pulse. (c) A typical "filament" in toluene. The picture was taken by focusing the camera at the end of the cell with a 125× magnification.

plain how a self-trapped filament of such high intensity could last for more than a few millimeters without showing appreciable spectral broadening.[7] A way to help distinguish the two cases is to focus the camera inside rather than at the end of the liquid cell. If the "filament" were indeed a self-trapped filament which terminates at the end of the cell, we would expect to see a blurred defocused image of the filament; otherwise, as the laser power is increased above the self-focusing threshold, we should always see a clear image of the focal spot extended gradually further inside the cell. In our experiments, we took simultaneous photographs with two cameras, one focused at the end of the cell and the other up to a few centimeters inside the cell. For laser power above the self-focusing threshold, we found on both photographs at equivalent positions a bright focal spot of about the same size (10 ± 2 μ in toluene), consistent with the picture of moving foci. The focal spot appeared deeper inside the cell for higher laser power, but not up to the point at which the peak of the laser pulse would self-focus, presumably because stimulated backward Raman and Brillouin scattering effectively terminated self-focusing through depletion of the incoming laser power (see explanation below). One might think that these results could also be interpreted as a self-trapped filament moving along a line. We rule out such a possibility on the following grounds: (1) Calculation[8] shows that a trapped filament of 30-GW/cm^2 peak intensity without appreciable spectral broadening should be depleted by Raman scattering in less than a few millimeters. (2) Focal spots were observed within 1 cm of the point at which the peak of the input pulse should self-focus, when the input peak power was not too far above the self-focusing threshold. This showed that the trapped filaments, if existing, could not be longer than 1 or possibly 0.5 cm. The limit was set by the experimental inaccuracy in determing the self-focusing threshold, assuming the worst case that self-focusing was not terminated by stimulated scattering. Physical results would of course be essentially the same, whether it is a moving focal spot of finite focal region or a moving, short, trapped filament. We also focused the camera up to a few millimeters outside the cell. The observed image was almost an order of magnitude smaller than one would expect from diffraction of a self-trapped filament, indicating some focusing action of the beam extended outside the cell.

Theoretically, knowing the time variation of the input laser power, we can calculate how the focal spot moves along the line. We assume that for a certain laser power P the focal spot appears at the self-focusing distance[9]

$$Z_f = K/[(P/P_{cr})^{1/2} - 1], \quad (1)$$

where P_{cr} is the critical power for self-trapping[3] and K is a constant depending on the geometric factors of the input beam and the nonlinear refractive index of the medium. By measuring the threshold power for self-focusing at various cell lengths we can find K and P_{cr}. The motion of the focal spot can then be determined from Eq. (1), knowing the time variation of the input laser power and taking into account the fact that light propagates with finite velocity. Figure 2 shows the position of the focal spot in toluene

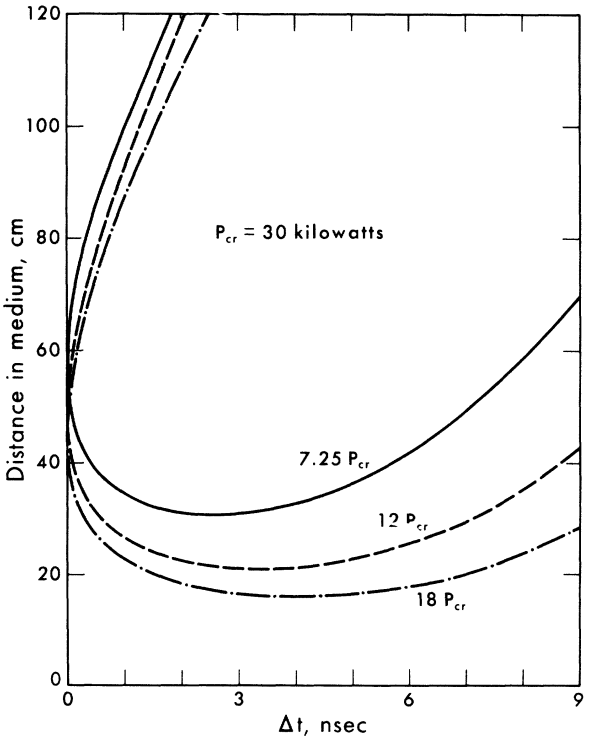

FIG. 2. Theoretical curves indicating the position of the focal spot inside toluene as a function of relative time Δt for several peak powers of a Gaussian laser pulse with 7.6-nsec full width at half-maximum. Here $\Delta t = 0$ refers to the instant the first focal spot is formed. Curves are calculated using Eq. (1) and experimental values of self-focusing threshold at different cell lengths. Note that since light travels with finite velocity, peak of the laser pulse would focus earlier with shorter self-focusing distances than the leading part of the pulse focusing with longer self-focusing distances. This also explains why, at a given Δt, two focal distances can be obtained from Eq. (1).

as a function of time for a set of input Gaussian pulses with various peak powers.[10] These pulses were chosen to match approximately the observed laser pulses in the experiments. However, there could be some residual spatial inhomogeneity in our laser beams; so the curves in Fig. 2 only describe approximately the actual experiments.

One consequence is immediately obvious from the picture of moving foci shown in Fig. 2. For a given cell length, the focal spot would spend more time at the end of the cell when the input laser power is just at the self-focusing threshold than in the case where the input power is considerably above the self-focusing threshold. In the former case, we would expect to see a brighter focal spot at the end of the cell. This is in fact what was observed. We noticed in addition, with the aid of motion pictures, that whenever the input power was near the self-focusing threshold, there appeared a bubble of about 100-μ diam out of the focal region at the end of the cell. Rough estimate shows that while an intense field is necessary to initiate the bubble, an energy of a few ergs is needed to create the bubble. This can therefore happen only if the focal spot stays at a local 10-μ region for more than 10 psec. Most of our experiments were done on toluene with a cell length of 33 cm and an input laser pulse of peak power between $6P_{cr}$ and $12P_{cr}$. From Fig. 2, we expect that the focal spot would first appear in the liquid medium at the end of the cell and then move inward. For higher input power, smaller pulse width, and longer cells the focal spot could first appear inside the cell and then split into two focal spots, one moving towards the front and the other towards the end of the cell. This would happen only at $P > 100\ P_{cr}$ for the 33-cm toluene cell we used. Then the focal spot could also move with a speed greater than the velocity of light.

Figure 2 shows that for input power larger than the self-focusing threshold, if self-focusing were not terminated by other processes, then the duration of a "filament" pulse would be much longer (>1 nsec for $P > 7P_{cr}$) than what was observed (~150 psec) and would increase with increase of input power, opposite to what was observed. However, various stimulated scattering processes can be initiated at the focal region. The backward stimulated Raman and Brillouin scattering would deplete effectively the incoming laser power[11] and consequently terminate the "filament" by depleting the later part of the input laser pulse to a level below the self-focusing threshold. The self-focused light diffracted from the focal spots could also be depleted by forward Raman scattering. Because of longer interaction length, depletion would of course be more appreciable for focal spots deep inside the cell. In all respects, the focal spots near the end of the cell should be less affected by stimulated scattering. Photographs indeed showed that focal spots inside the cell were much less intense than those close to the end of the cell. From Fig. 2, we conclude that to yield the observed "filament" pulse duration, the major part of the pulse must be emitted from a short section of the "filament" presumably within 1 cm towards the end of the cell. This also explains why the pulses were shorter and weaker for higher input power.

In conclusion, we believe that under conditions similar to ours, the so-called "filaments" are actually the result of moving foci. The size of a "filament" should then be the size of the focal spot. Machine calculations,[12] with a simple model of saturable-refractive index, indicate that a self-focused beam would defocus and then refocus again. If the laser power is being depleted by stimulated scattering in the focusing process, then the self-focused beam after defocusing would not have enough self-focusing strength to refocus. For an input laser pulse which is nonhomogeneous and multimoded, self-trapped filaments may still exist[4] because of very different propagation conditions. However, a question yet to be answered is why the size of the observed "filaments" in a given liquid seems to remain roughly constant irrespective of the input pulses.

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SIMPLE MODEL FOR SEMICONDUCTOR-METAL TRANSITIONS: SmB_6 AND TRANSITION-METAL OXIDES

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We propose a simple model for a semiconductor-metal transition, based on the existence of both localized (ionic) and band (Bloch) states. It differs from other theories in that we assume the one-electron states to be essentially unchanged by the transition. The electron-hole interaction is responsible for the anomalous temperature dependence of the number of conduction electrons. For interactions larger than a critical value, a first-order semiconductor-metal phase transition takes place.

Many substances, including SmB_6[1] and a number of transition-metal oxides,[2] exhibit semiconductor-metal transitions.[3] The transitions are in many cases first-order phase transitions (e.g., in V_2O_3); however, they can also result from a gradual but anomalously large increase in conductivity over a range of temperatures (e.g., in SmB_6 and Ti_2O_3). In addition, measurements of large magnetic susceptibilities with anomalous temperature dependences suggest that in many of these materials localized magnetic moments exist and that they are intimately connected with the transition. As an example, it has been hypothesized[1] that in SmB_6 the conduction electrons and the localized moments are produced simultaneously by the promotion of a single localized electron from the spherically symmetric Sm^{++} ion ($J=0$) into a conduction band. The Sm^{+++} ion left behind ($J=\frac{7}{2}$) acts as a localized moment.

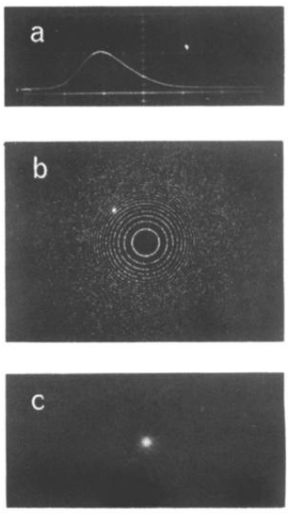
We present here a simple theory of the semiconductor-metal transition based on a model having both localized and itinerant interacting quasiparticle states. The relevant single-electron states consist of (a) bands of extended Bloch functions and (b) a set of localized states centered at the sites of the metallic ions in the crystal. As $T\to 0$ the localized states are lower in energy than the band states and are fully occupied by electrons. Therefore the quasiparticle excitations are either localized holes or itinerant electrons. In the language of second quantization and in the spirit of the Landau theory of Fermi liquids, we write the one-particle terms as

$$H_0=\sum_{\nu\vec{k}\sigma}\epsilon_\nu(\vec{k})a_{\nu\vec{k}\sigma}^{\dagger}a_{\nu\vec{k}\sigma}+\sum_{i\sigma}Eb_{i\sigma}^{\dagger}b_{i\sigma}, \tag{1}$$

where $a_{\nu\vec{k}\sigma}^{\dagger}$ creates an electron in state $\vec{k}$, band ν, with spin σ, and $b_{i\sigma}^{\dagger}$ creates a hole with spin σ at site i. The energies $\epsilon_\nu(\vec{k})$ and E are positive definite and such that

$$\Delta\equiv\min[E+\epsilon_\nu(\vec{k})]>0 \tag{2}$$

is the energy gap for the formation of an electron-hole pair. We further assume that the quasiparticle interaction is screened, and its range short enough so that only intra-atomic terms need be considered. In this case the interaction



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