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SELF-TRAPPING WITH PICOSECOND LIGHT PULSES

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Molecular mechanisms in liquids, which are responsible for the self-trapping of small-scale filaments of light, have been tested with regard to inertial response. Picosecond light pulses are found to self-trap even in viscous liquids and rigid glass where linear and rotational diffusional motions are frozen out. This and other evidence based on the characteristics of these filaments supports a molecular electronic distortion mechanism.

The physical mechanisms which give rise to the self-trapping of light[1] in molecular systems are examined here for the case of picosecond excitation. On this time scale, it is concluded that residual nuclear motions, involving the molecular orientational Kerr effect and the recently proposed molecular redistribution or clustering mechanism,[2] are not necessarily dominant. For example, the present observation of self-trapped filaments in viscous media, where these motions are frozen out, points to a pure electronic distortion mechanism. Our measurements of the filament's radial intensity profile reveal a similarity in the diameter and the peak electric field for viscous and fluid media alike and suggest that electron distortion may be of general importance for picosecond beam trapping.

Thus our results contrast with earlier studies[3,4] which utilize nanosecond exciting light and which support the molecular orientational Kerr effect[3] or possibly the even slower electrostrictive process.[4] Until now, the question of mechanism on a picosecond scale has not been challenged, although recently the quenching of the orientational Kerr effect has been mentioned to explain partly reduced stimulated Raman scattering.[5]

In suitable media, the orientational Kerr effect and molecular clustering can be distinguished from electronic distortion because of their different inertial character. The first two involve nuclear orientational and translational motions, which are driven by optical electric fields and damped by Brownian movement. The relaxation times[6,7] for these diffusional motions are given approximately by

$$\tau(\text{orientation}) = \tfrac{4}{3}\pi\eta a^3/kT,$$

$$\tau(\text{translation}) = \frac{\langle x^2\rangle}{D} = \frac{6\pi\eta a\langle x^2\rangle}{kT}, \qquad (1)$$

where η is the viscosity and a is the molecular radius. In ordinary liquids, the linear diffusion coefficient $D \sim 10^{-5}$ cm^2/sec, and if clustering is important, the diffusion distance x must be at least an angstrom. This gives τ(translation) $\gtrsim 10^{-11}$ sec which is of the same magnitude as τ(orientation). On the other hand, the response time for electron distortion is about the period of a Bohr orbit, $2\pi a_0\hbar/e^2 \sim 10^{-16}$ sec. Thus, with increasing viscosity and because of (1), a point will eventually be reached where only the electron motions will contribute to beam trapping.

Before discussing these results, we first describe the techniques for characterizing self-trapped filaments in the near infrared. In these studies, self-trapping is induced in a 10-cm liquid cell by the collimated beam of a neodymium glass laser, mode locked[8] by Eastman 9860 dye and with a spectral wavelength of 1.06 μ. The incident beam enters the liquid after passing through a 1-mm circular aperture, and it consists of a train of about 35 pulses, separated by intervals of 3.8 nsec, and with about 20-MW power in each pulse. A laser pulse width of about 1 psec is obtained by the two-photon fluorescence technique[9] using 10^{-3} M Rhodamine B dye in acetone. (The use of the second harmonic of the laser beam as a source would have facilitated these measurements, but pulse powers of ~1 MW in the 1-mm collimated beam were insufficient for producing filaments, except in CS_2.)

Filaments are photographed in the exit plane of the cell through a microscope having a numerical aperture of 0.33, a magnification of 60×, and a resolution of 2 μ at the laser wavelength. Because of dispersion, the microscope focusing adjustment is carried out with a 1.15-μ cw He-Ne laser as source and an infrared image converter as detector (RCA 6914A with a resolution of 35 line pairs/mm). The nonfilamented beam, which would otherwise elevate the photographic background and damage the objective, is largely eliminated by a 1-mm stop placed directly in front of the objective. No more than 10% of the widely diverging filament light is intercepted by the stop.

The extent of stimulated Raman scattering in picosecond filaments appears to be no greater than 10%, as determined in preliminary tests. Thus, the spectral wavelength of trapped radiation is largely in the vicinity of 1.06 μ. Since the number of filaments is smaller than for nanosecond excitation, because of a higher threshold, the over-all Raman conversion is considerably less also, in support of recent observations.[5]

For the 1-μ spectral region, hypersensitized spectroscopic Kodak I-Z plates are required. The resolving power of this plate is greater than 50 lines/mm and in no way limits the resolution of the magnified filament. Filament intensities are obtained from a step-wedge calibration and the plate sensitivity at 1.06-μ wavelength. The latter, as determined with the mode-locked laser, gives a sensitivity of 6.5 erg/cm^2, in good agreement with the 8.0-erg/cm^2 value reported by Kodak.[10]

The radial intensity profile of a filament is obtained from a microdensitometer trace as illustrated in Fig. 1 for nitrobenzene. This is the simplest mode structure and the most common, but the next higher order mode[11] which contains an intense additional ring of ~25-μ diam has also been observed in several liquids. Note that in Fig. 1 the intensity scale has been linearized and that such a trace provides the filament energy and the diameter, which is taken as the full width at half-maximum. The observed profile (solid curve) closely follows a Gaussian shape (circles) which has not been predicted. Theory[1] based on an electric-field-dependent refractive index of the form $\Delta n = n_2 E^2$ yields a radial intensity which is significantly broader near the peak and narrower in the wing. However, index saturation which is known to influence the profile in the orientational Kerr effect[12] should now be considered for the case of electron distortion.

Table I gives results for some liquids which illustrate the magnitude of these quantities and their dependence on molecular symmetry and viscosity. The entries are statistical averages, derived usually from a sample of 10-20 filaments, and the ± quantities are average deviations from

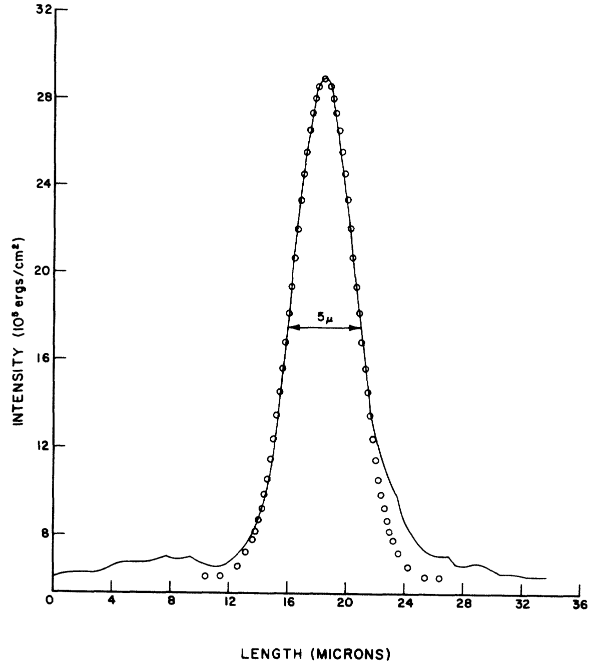

FIG. 1. The radial intensity profile of a self-trapped filament in nitrobenzene at 23°C. The profile is obtained from a microdensitometer trace, and the instrument's resolution of 1.2 μ or less did not alter the shape. The solid curve is experimental while the circles are a Gaussian fit.

Table I. Characteristics of self-trapped filaments.[a]

	Diameter (μ)	Energy (erg)	Power[b] (kW)	Peak electric field[c] (10^7 V/cm)
CS_2	5.9 ± 1.3	1.3 ± 0.6	130	2.0 ± 0.6
Nitrobenzene	5.8 ± 0.8	0.9 ± 0.4	90	1.6 ± 0.4
CCl_4	7.9 ± 0.2	1.2 ± 0.8	120	1.3 ± 0.4
Toluene	10.1 ± 1.0	0.6 ± 0.2	60	0.7 ± 0.1
Ether, ethanol, toluene (23°C)	7.6 ± 1.4	1.2 ± 0.6	120	1.5 ± 0.4
Ether, ethanol, toluene (−140°C)	8.9 ± 1.3	3.8 ± 1.9	380	2.0 ± 0.5
Ether, ethanol, toluene (−165°C)	17.7 ± 1.7	13.1 ± 10.2	1310	2.0 ± 1.0

[a] Liquid temperature is 23°C unless otherwise noted.

[b] Obtained from the preceding column assuming a pulse duration $\Delta t = 1$ psec.

[c] Obtained from the peak intensity of the microdensitometer trace, $cnE_p^2\Delta t/16\pi$ (in erg/cm^2), assuming $\Delta t = 1$ psec. This, therefore, is not the local field but the field of the transmitted beam.

the mean. While the mean values appear to be characteristic of the liquid, variations as large as a factor of 2 in the diameter and 10 in the energy do occur. This behavior would indicate that these are not invariant quantities as previously suggested.[13] Furthermore, our statistics do not yield any obvious correlation of the diameter with energy or with the peak electric field.

We now consider the physical mechanisms which contribute to an intensity-dependent refractive index and to self-trapping of picosecond light. The present observation of filaments in CCl_4, which cannot exhibit an orientational Kerr effect because of symmetry, clearly shows that other mechanisms[2] can play a significant role. However, this evidence alone does not discriminate between the two likely possibilities of molecular clustering or electron distortion. Note also that nitrobenzene and CS_2 have similar characteristics in Table I even though their orientational relaxation times are 50 and 1.9 psec, respectively, and this too indicates that mechanisms other than the orientational Kerr effect are involved.

To illustrate the importance of electron distortion, an organic liquid mixture was selected which yields self-trapped filaments even when it is cooled and becomes a rigid glass. The liquid, whose composition by volume is ether (49%), ethanol (7%), and toluene (44%), is contained in a 10-cm cell which is thermally isolated in an optical Dewar. The self-trapping is due to toluene as the other two liquids are ineffective alone. Viscosities[14] of 0.4 cp (23°C), 10^3 cp (−140°C), and in excess of 10^6 cp (−150°C or less) were utilized, and the corresponding filament characteristics are given in Table I. For the glass, which forms at about −150°C, the relaxation times of (1) are longer than 10^{-5} sec, i.e., they are at least seven orders of magnitude longer than a picosecond pulse. The critical power per pulse required to initiate filaments throughout a 1-mm cross section is about 10 MW, almost the capability of our laser, and does not appear to depend significantly on the viscosity. This magnitude compares favorably with the estimate of ~2 MW from steady-state trapping theory[1] and the use of the experimental nonlinear electronic polarizability coefficient for xenon, $\gamma_{\rm Xe} = 39 \times 10^{-37}$ esu/atom.[15] However, if clustering or the orientational Kerr effect are dominant, the simplest estimate would suggest that the critical power increases directly with an increasing relaxation time or viscosity, and this behavior is clearly not observed.

As might be expected for the case of electron distortion, the filament diameter changes only slightly when the viscosity increases by three orders of magnitude. For the glass, however, with an additional three orders of magnitude or more in the viscosity, the diameter doubles. Whether this is the result of an intrinsic change in the electron polarizability, which reflects a radical change in state, or is due to the influence of optical distortions from strains is not evident. In support of the former, ~20-μ-diam filaments were observed (in a few instances) in a uniform rod of polymethylmethacrylate[16] at 23°C, which similarly is another rigid medium exhibiting the electronic distortion mechanism.

While the exciting power needed to initiate self-focusing[17] with picosecond light is about 10 MW, the power in a single filament is but a few tens to a few hundreds of kilowatts. Furthermore, filaments usually occur in a group, indicative of a common origin from a larger filament at an earlier stage. Both results suggest that the critical-power requirement is relaxed with increasing field strength, as would occur with a rapidly increasing polarizability. The detailed polarizability behavior of molecular liquids in very strong electric fields is in general unknown. However, calculations for the CO molecule, which are given in the accompanying Letter,[18] show that the polarizability increases sharply as ionization is approached, at fields near 10^9 V/cm. Larger, more polarizable molecules should behave similarly and ionize more easily, but the peak radiation fields in filaments (Table I) are no larger than 3×10^7 V/cm, including the Lorentz local field factor $\frac{1}{3}(\epsilon+2)$. This is at least an order of magnitude below that for ionization, and we conclude that a quantitative understanding of the electron distortion mechanism, as well as saturation in the index, is still lacking. It may be that the Lorentz local-field correction is inadequate at high fields and that intermolecular interactions, which enhance the polarizability, cannot be ignored as in the CO calculation. Recent measurements[15] of the Kerr electro-optic effect in the rare gases show that these interatomic interactions are quite significant. In addition, should the filament duration be less than 1 psec due to pulse sharpening,[19] the powers and fields of Table I would be even larger and thus subject to revision.

A comparison of the diameters and peak electric fields for the liquids in Table I reveals that they are quite similar not only to each other but to small-scale filaments[3] produced by nanosecond excitation from a ruby laser. Note that when index saturation occurs, it is the field which is to be compared and not the power. For nanosecond exciting light, the orientational Kerr effect is surely responsible for the initial self-focusing as the exciting power requirement is but a few tens of kilowatts[3] (compared with a few tens of megawatts for picosecond light) and is in reasonable agreement with theory.[1,3] In the latter stages, however, when the beam has collapsed to many small filaments, the above comparison indicates that electron distortion could play a role and thus could be of importance in small-scale filaments in general.

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