

SELF-TRAPPING WITH PICOSECOND PULSES AND "ROCKING" OF MOLECULES*

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We report a study of the spectral properties of self-trapped filaments of light with picosecond excitation. The clean spectra obtained allow the measurement of the relaxation time of the nonlinear refractive index induced by the field; it is a few tenths of a picosecond for many liquids. We suggest that "rocking" of molecules in the field of neighboring ones is the main mechanism for trapping in our case.

The physical mechanisms which are responsible for the self-trapping of light¹ of picosecond duration have already been considered by Brewer and Lee.² These authors reach the conclusion that the trapping phenomenon, in the case of picosecond excitation, is due to molecular electronic distortion. In this Letter a study of the spectral broadening of filaments produced by picosecond laser light pulses is reported. The short exciting pulses which have been used (~5 psec) usually produce very regular spectra through the phenomenon of self-phase modulation.^{3,4} From the interpretation of these spectra the following conclusions are reached: (1) Neither the Kerr effect due to orientational diffusion of molecules nor the mechanism of molecular electronic distortion appears to play a predominant role in self-trapping of picosecond pulses. (2) The measured values of the relaxation time τ for the nonlinear refractive index δn ($\tau=0.3-0.6$ psec for CS₂, toluene, and bromobenzene) appear to be more consistent with the measurements of τ from the extreme wing of the spontaneous Rayleigh line.⁵ According to the interpretation of this wing due to Starunov⁶ and Fabelinskii and Starunov,⁷ we suggest that trapping is in our case mainly due to "rocking" of molecules in the field of the neighboring molecules. The nonlinear refractive index is in this case due to the rocking motion of the molecules driven by the torque exerted by the optical field, the torque resulting proportional to the square of the field's amplitude. This interpretation sheds new light on the trapping phenomenon at least in the case of picosecond excitation.

For our experiments, a TEM₀₀-mode, mode-locked ruby laser with 5-psec pulses has been used.⁸ The unfocused beam of the laser was sent through a 12-cm-long trapping cell containing either CS₂, bromobenzene, or toluene. The apparatus for detecting the spectra of the filaments is very similar to that described elsewhere.³ The theoretical resolving power of the grating was ~ 0.6 cm⁻¹. With these short pulses, clean peri-

odic spectra due to self-phase modulation are obtained. An example of these spectra is shown in Fig. 1. On increasing the laser power above the threshold power for trapping, the extension of the spectra steadily increased.⁹ These observations are in full agreement with the interpretation of such spectra in terms of self-phase modulation due to short pulses as first discussed by Shimizu³ and then more fully by Gustafson *et al.*⁴ When the power was increased by about a factor 3, the longest spectra showed an extension of several hundred wave numbers, and they were no longer so regular. Often, in this case, a fine structure^{10,11} with a period ranging between 7 and 10 cm⁻¹ was observed in all the previous liquids.

According to the works previously mentioned,^{3,4} the spectrum $f(z, \omega)$ of the light pulse after traveling a distance z in the filament can be written as

$$f(z, \omega) = e^{ikz} \int_{-\infty}^{+\infty} A(t, z) e^{i(\gamma \delta n + \omega t)} dt, \quad (1)$$

where $A(t, z)$ is the field amplitude, $\gamma = \omega_0 z / c$, δn is the nonlinear refractive index induced by the field, ω_0 the laser angular frequency, and ω the angular frequency of the spectrum minus ω_0 . As done by Gustafson *et al.*⁴ one can assume a given shape for $A(t, z)$ and a given functional relation between δn and A , and then, by computer, calculate $f(z, \omega)$ to be fitted with the experimental re-

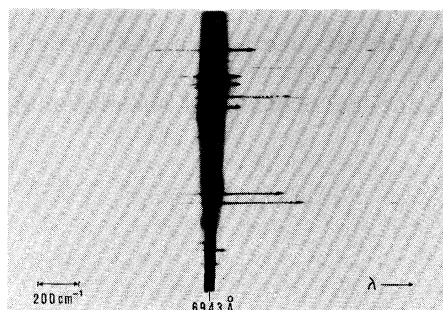


FIG. 1. Typical regular spectra due to self-phase modulation as obtained with picosecond excitation.

sults. Actually, interesting results can be obtained by approximately calculating the integral in Eq. (1) with the method of stationary phase¹²: At each angular frequency ω the main contribution to the integral (1) comes from the points at which $\gamma\delta\dot{n} = -\omega$. For what follows, reference will be made to Fig. 2 where a typical spectrum is schematically shown [Fig. 2(a)] and where a bell-shaped curve (not necessarily symmetrical) is assumed for $\delta n(t)$ [Fig. 2(b)]. In Fig. 2(b) the two inflection points 1 and 2 of δn are also indicated. By using the method of stationary phase one can show that (1) the number m of minima of the spectrum [Fig. 2(a)] on the Stokes side is equal to that on the anti-Stokes side and (2) the following relations hold approximately:

$$\delta n_p / \delta \dot{n}_1 = m / \nu_S, \quad (2a)$$

$$\delta n_p / \delta \dot{n}_2 = -m / \nu_{AS}, \quad (2b)$$

where the time derivatives are taken at the inflection points and where ν_S and ν_{AS} are the absolute values of the spectral extensions on the Stokes and anti-Stokes sides, respectively.

To proceed further, the following functional relation between $\delta n(t)$ and $A(t)$ is now assumed:

$$\tau_2^2 \delta \ddot{n} + \tau_1 \delta \dot{n} + \delta n = n_2 A^2, \quad (3)$$

where the equation may describe either one of the following three phenomena: (1) rotational diffusion of molecules, where it is generally assumed that $\tau_2 = 0$ and τ_1 is equal to $\frac{1}{3}$ of the Debye relaxation time; (2) molecular electronic distortion, where one must assume that $\tau_2 = \tau_1 \approx 0$; (3) "rocking" of molecules in the field of neighboring molecules. This phenomenon was first introduced by Starunov^{6,7} to account for the experimentally observed wings of the Rayleigh line. More recent experimental studies on the Rayleigh wing in CS₂

seem to confirm this model.¹³ Since the induced nonlinear polarization can be shown to be proportional to the angular displacement $\Delta\theta$ of the molecules, Eq. (3) follows from Eq. (15), Ref. 6, by putting at the right-hand side of this equation a term due to the torque exerted by the optical field on the molecule. By doing this one gets $\tau_2 = (I/\mu)^{1/2}$, $\tau_1 = \zeta/\mu$, and $(n_2)_{\text{rocking}} / (n_2)_{\text{rot diffusion}} \approx 6KT/\mu$, where I is the moment of inertia, ζ is a coefficient of internal friction, and μ is a coefficient of orientational elasticity. In the case of CS₂ from Ref. 7 one gets $(n_2)_{\text{rocking}} / (n_2)_{\text{rot diffusion}} \approx 0.2$, $\tau_1 \approx 0.46$ psec, and from the slope of part BC of Fig. 3, Ref. 7, $\tau_2 \approx 0.21$ psec. These values can be taken as indicative; it is important, however, to notice that the times involved in this case are of the order of a few tenths of a picosecond.¹⁴

Equation (3) is obviously open to criticism. Firstly it does not describe the saturation of δn at high fields, and secondly it does not present the two phenomena of rotational diffusion and "rocking" of molecules in a unified manner. Therefore it would be difficult to describe the behavior of the liquid at the transition region between the above two phenomena. In the case of rotational diffusion a more exact procedure would be that followed by Herman.¹⁵ Equation (3) is therefore assumed here as a phenomenological one to be compared with the experimental results.

An upper limit for τ_1 can be readily obtained by considering Eq. (3) at time t_2 [Fig. 2(b)] and by taking into account Eq. (2b). Since the right-hand side of Eq. (3) is positive (which would be also true even if higher order terms in the fields were added to the right-hand side to take into account saturation¹⁶) one gets $\tau_1 < (m/\nu_{AS})$. From a large number of spectra of the type shown in Fig. 1 the results listed in Table I have been obtained. It is worth emphasizing that the results have been obtained only from those spectra in which both the Stokes and anti-Stokes sides could be clearly measured. From the minimum values of m/ν_{AS} in Table I one sees that the upper limit for τ_1 is approximately the same for the three liquids, ~ 0.7 psec. One therefore reaches the first conclusion that the phenomenon of rotational diffusion cannot be the predominant one in our case since the times involved are approximately 2, 4, and 10 psec for CS₂, toluene, and bromobenzene, respectively.¹⁷

This result alone would not eliminate molecular electronic distortion as a possible mechanism. An approximate value of τ_1 can however be obtained by considering Eq. (3) at the two times t_1

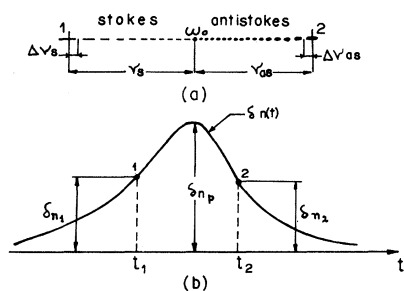


FIG. 2. (a) Schematic drawing of a typical spectrum. (b) Bell-shaped curve (not necessarily symmetrical) which is assumed for $\delta n(t)$ and which is used in the discussion of the test.

Table I. Values for the relaxation time τ_1 of the non-linear index of refraction δn induced by the field as measured from the spectra.

CS ₂						
n	ν_S (10 ¹² Hz)	ν_{AS} (10 ¹² Hz)	m	$\Delta\nu_S/\Delta\nu_{AS}$	σ	τ_1 psec
1	5.1	10	7	0.5	4	0.36
2	6.2	10.8	9	0.6	7	0.4
3	7	10.9	17	0.65	4	0.6
4	7.5	11.2	13	0.8	4	0.45
5	7.5	10.9	16	0.6	6	0.65
6	10.2	11.9	20	0.77	11	0.85
7	10.2	11.4	12	0.9	8	0.5
8	12.2	16	22	0.82	11	0.7
9	13.1	16	21	0.9	14.5	0.7
10	14.2	17.1	28	0.87	12	0.83
11	15.8	18.1	24	0.83	5.6	0.51
12	18.4	20.4	38	0.75	9	0.9

Toluene						
n	ν_S (10 ¹² Hz)	ν_{AS} (10 ¹² Hz)	m	$\Delta\nu_S/\Delta\nu_{AS}$	σ	τ_1 psec
1	4.4	4.3	3	1.22	24	0.39
2	8.6	7.35	6	1.25	9	0.38

Bromobenzene						
n	ν_S (10 ¹² Hz)	ν_{AS} (10 ¹² Hz)	m	$\Delta\nu_S/\Delta\nu_{AS}$	σ	τ_1 psec
1	1.4	2.36	4	0.57	2	0.37
2	2.3	3.3	4	0.55	2	0.22
3	4.16	5.5	4	0.62	2.3	0.19
4	4.16	6.6	6	0.5	1.5	0.2
5	10.6	13.4	15	0.74	5.5	0.49

and t_2 [Fig. 2(b)], by taking the ratio of the two expressions and by using Eqs. (2). One easily gets

$$\frac{\tau_1(\nu_S/m)\delta n_p + \delta n_1}{-\tau_1(\nu_{AS}/m)\delta n_p + \delta n_2} = \frac{A_1^2}{A_2^2}, \quad (4)$$

where A_1^2 and A_2^2 are the square of the field amplitudes at times t_1 and t_2 , respectively. From the method of stationary phase applied to the integral (1) at the angular frequencies $\omega_S = 2\pi\nu_S$ and $\omega_{AS} = 2\pi\nu_{AS}$ one gets $A^2(t_1)/A^2(t_2) = |f_1|^2(\delta\ddot{n}_1)^{2/3}/|f_2|^2(\delta\ddot{n}_2)^{2/3}$, where $|f_1|^2$ and $|f_2|^2$ are the intensities of the spectra at the points 1 and 2 of Fig. 2(a) and the third derivatives of δn are taken at the times t_1 and t_2 . It can be further shown that $(\delta\ddot{n}_1)^{1/3}/(\delta\ddot{n}_2)^{1/3} = \Delta\nu_S/\Delta\nu_{AS}$, where $\Delta\nu_S$ and $\Delta\nu_{AS}$ are frequency separations between the first maximum and minimum of the spectrum on the Stokes and anti-Stokes sides, respectively [Fig. 2(a)].

Therefore, from Eq. (4),

$$\frac{\tau_1(\nu_S/m)\delta n_p + \delta n_1}{-\tau_1(\nu_{AS}/m)\delta n_p + \delta n_2} = \frac{|f_1|^2\Delta\nu_S^2}{|f_2|^2\Delta\nu_{AS}^2} = \sigma. \quad (5)$$

To calculate τ_1 from Eq. (5) one must know the ratios $\delta n_1/\delta n_p$ and $\delta n_2/\delta n_p$. These ratios are both assumed to be approximately 0.6. This is in fact true for any Gaussian pulse, whereas for a Lorentzian pulse the ratio would be 0.75. We therefore believe that this assumption will not drastically affect our results. It is worth mentioning that the above ratios can also be obtained from the spectra (in a rather lengthy way however) and that the values which have been measured for a few spectra range from 0.4 to 0.6.

From Eq. (5) the values of τ_1 can now be obtained by measuring σ , ν_S/m , and ν_{AS}/m from the spectra. To measure σ , the contrast factor of our film (Kodak spectroscopic film 1N) under picosecond-pulse excitation was determined. The values of τ_1 obtained in this way are listed in Table I. One sees that for each liquid the values are quite consistent, irrespective of the extension of the spectra, and that they are of the order of a few tenths of a picosecond. One therefore reaches the second conclusion that the molecular electronic distortion is not the predominant mechanism in our case.

From Table I one sees that the average value for τ_1 is 0.6 psec for CS₂, 0.39 psec for toluene, and 0.3 psec for bromobenzene. These values appear to be in agreement with the values of τ obtained by measurements at the far portion of the spontaneous Rayleigh wing. In fact, although values of τ for toluene and bromobenzene are not, to our knowledge, available, measurements on CS₂ and other liquids¹⁴ indicate that τ is always of the order of a few tenths of a picosecond. Hence, according to the interpretation of the far portion of the Rayleigh line due to Starunov we suggest that molecular "rocking" plays a predominant role in self-trapping with picosecond pulses. It is worth noting that the present interpretation does not appear to be in contrast to the temperature experiments reported by Brewer and Lee² since even in a glass state the molecules can undergo oscillations.

Since measurements of filament diameter and energy in the case of CS₂ are in agreement with those obtained with nanosecond excitation,¹⁶ it is suggested that in this case too the "rocking" mechanism may play an important role.

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¹R. Y. Chiao, E. Garmire, and C. H. Townes, *Phys. Rev. Letters* **13**, 479 (1964).

²R. G. Brewer and C. H. Lee, *Phys. Rev. Letters* **21**, 267 (1968).

³F. Shimizu, *Phys. Rev. Letters* **19**, 1097 (1967).

⁴T. K. Gustafson, J. P. Taran, H. A. Haus, J. R. Lifshitz, and P. L. Kelley, *Phys. Rev.* **177**, 306 (1969).

⁵I. L. Fabelinskii, *Molecular Scattering of Light* (Plenum Press, Inc., New York, 1968), Table 32, p. 413.

⁶V. S. Starunov, *Opt. i Spektroskopiya* **18**, 300 (1965) [translation: *Opt. Spectry. (USSR)* **18**, 165 (1965)].

⁷I. L. Fabelinskii and V. S. Starunov, *Appl. Opt.* **6**, 1793 (1967).

⁸R. Cubeddu, R. Polloni, C. A. Sacchi, and O. Svelto, "Picosecond Pulses, TEM₀₀ Mode, Mode-Locked Ruby

Laser" (to be published).

⁹M. M. Denariez-Roberge and J.-P. E. Taran, *Appl. Phys. Letters* **14**, 205 (1969).

¹⁰A. C. Cheung, D. M. Rank, R. Y. Chiao, and C. H. Townes, *Phys. Rev. Letters* **20**, 786 (1968).

¹¹J. R. Lifshitz and H. P. H. Grieneisen, *Appl. Phys. Letters* **13**, 245 (1968).

¹²E. T. Copson, *Asymptotic Expansions* (Cambridge University Press, Cambridge, England, 1965), Chap. IV.

¹³S. L. Shapiro and H. P. Broida, *Phys. Rev.* **154**, 129 (1967).

¹⁴See Fabelinskii, Ref. 5. It is worth noting that the quantity there listed is given in our notation by $\tau = (\tau_1^2 - 2\tau_2^2)^{1/2}$.

¹⁵R. M. Herman, *Phys. Rev.* **164**, 200 (1967).

¹⁶R. G. Brewer, J. R. Lifshitz, E. Garmire, R. Y. Chiao, and C. H. Townes, *Phys. Rev.* **166**, 326 (1968).

¹⁷M. F. Vuks and A. K. Atakhodzhaev, *Dokl. Akad. Nauk SSSR* **109**, 926 (1956) [translation: *Soviet Phys. -Doklady* **1**, 496 (1956)].

EXPERIMENTAL EVIDENCE OF THE "CRITICAL REGION" OF A BINARY MIXTURE BY MEANS OF INELASTIC SCATTERING OF LIGHT

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The "critical region" of a binary mixture is found to be characterized by the appearance of an asymptotic dependence such as $\Gamma = AK^3$.

As is well known, the formula giving the spectral linewidth Γ of the light scattered by a binary mixture in the hydrodynamic region is

$$\Gamma = DK^2(1 + \xi^2 K^2)^{1-\eta/2},$$

with D = mass diffusion coefficient, K = scattering vector, and ξ = correlation length.

This formula, however, is no longer valid when $\xi K \geq 1$, as we have shown in a previous paper.¹ In order to obtain the relationship between Γ and K in this region, we have performed systematic measurements in the immediate vicinity of the critical temperature of a cyclohexane-aniline binary mixture. To avoid the difficulties arising from eventual thermal fluctuations of the thermostatic bath, the photocurrent due to the scattered light was recorded on an Ampex tape recorder, which demanded a relatively short time during which the temperature was stable to better than 10^{-3} °C. The rather long spectral analysis was performed at a later time.

Figure 1 shows the observed angular dependence of the spectral linewidth Γ , for different

values of $T - T_c$. This angular dependence (for $T - T_c = 6 \times 10^{-3}$ °K, 5.5×10^{-3} °K, 4×10^{-3} °K, 3.5×10^{-3} °K, and 1.5×10^{-3} °K) is consistent with a law such as $\Gamma = AK^3$ (with $A = 1.3 \times 10^{-13}$ cm³ sec⁻¹ and independent of $T - T_c$). This result is not surprising, because for all these values of $T - T_c$, $\xi K \geq 1$, even for the smallest recorded value of the scattering angle $\theta = 30^\circ$.

At temperatures this close to T_c one might expect spurious effects due to multiple scattering and concentration gradients with height. However a separate experiment conducted on three cylindrical cells of different diameters (14, 10, and 5 mm) has shown that, in spite of variable amounts of multiple scattering (as shown by the observed beating signal-to-shot-noise ratios) the linewidth of the scattered spectrum does not depend on the cell diameter. This experiment shows that while multiple scattering strongly affects the intensity of the scattered light, it does not affect its linewidth. Thus the observed $\Gamma \sim K^3$ behavior cannot be ascribed to a spurious multiple-scattering effect.

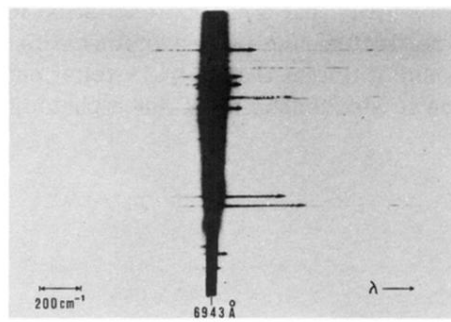


FIG. 1. Typical regular spectra due to self-phase modulation as obtained with picosecond excitation.