

The role of the solvent viscosity on the spatiotemporal instabilities of large aperture dye lasers

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Local intensity fluctuations in a large aperture dye laser have been measured for several solvent viscosities. A decrease on the relative fluctuations amplitude as the solvent viscosity increases is found. From a theoretical approach, we show that the intensity fluctuations follow the same behavior as the molecular polarization orientation driven by the laser field. © 1998 American Institute of Physics. [S0003-6951(98)00131-4]

High energy flash-lamp pumped dye lasers are characterized by a large beam diameter (≈ 1.5 cm), so the Fresnel number of the resonator is also large ($F \approx 100$). Such a big transverse dimension favors the development of extremely local irregular spatiotemporal instabilities.¹⁻⁵ These instabilities appearing in systems with optical feedback and large aperture are supposed to be mediated by bulk mechanisms in the active medium.⁵⁻⁸ The observation of local dynamics in transverse patterns may give significant information about the underlying bulk process.

The main feature of this behavior was the chaotic oscillation observed when the intensity output was measured in small regions of the laser spot. This fact indicated the existence of a local chaotic dynamics with a low transverse correlation.

In a previous publication, a model has been proposed to explain the main characteristics of the intensity fluctuations in high Fresnel number lasers.³ The model, following the idea of Emelyanov and Yukalov,⁹ assumed the formation of population inversion filamentary structures which were coupled by the nearest neighboring diffracted field.

In an earlier work using the same dye laser, we observed a strong reduction in the amplitude of local irregular fluctuations in a Rhodamine 6G solid copolymer solution.¹⁰ That means that the local irregular fluctuations present in liquid solutions (Rhodamine 6G in methanol), practically disappear in solid solutions. In the Emelyanov and Yukalov model a coupling between the induced dipoles oriented by the laser field takes place. Being the polarized molecules inside a material host, the achieved orientation during the time of radiation of the laser pulse will depend on the solvent viscosity. In very viscous solvents, the orientation of the dye molecules will become difficult, decreasing the mutual interaction average. It leads to a substantial reduction of the filamentation capability and superradiant coordination length,¹¹ which is proportional to the polarization decay time.

The object of our study is to test experimentally this prediction by measuring the influence of the solvent viscosity on the relative amplitude of the intensity fluctuations.

Our main measurement system consists of a coaxial flash-lamp pumped untuned dye laser having a simple reso-

nator made up of two flat mirrors (reflectivities $R_1 = 1$ and $R_2 = 0.3$).¹⁰ The measurements were made with Rhodamine 6G solved in several polar solvents, whose viscosities are quoted in Table I. The viscosities vary in a wide range from the smallest one available as dye solvent (methanol) to practically infinite. The same Rhodamine 6G solid copolymer solution P (HEMA-MMA) described in a previous work¹⁰ was used as a limit of very high viscosity.

The laser was operated in a single pulse regime and the measurements were even performed at the same temperature (20 °C) maintained by a heat exchange circuit. If we guess that the solvent heat absorption rate is around 30% of the capacitor stored energy, then the effect on the solvent refraction index is less than 0.5%.¹² The change is produced monotonously during the pulse and homogeneously distributed in the solvent pumped volume, therefore can be said it does not play any role in the very fast local fluctuations.

The flux of the solvent through the lasing region is about 6 l min^{-1} . To be sure about the influence of the medium turbulence in the fluctuations, we repeated the experiment with no flux at all, finding the same results.

The dye concentration was varied between $2.5 \times 10^{-5} \text{ mol/l}$ and $7 \times 10^{-5} \text{ mol/l}$, but this parameter did not show any influence in the fluctuation amplitude. Anyway, the reported experimental results were obtained with identical concentration in all the cases and solvents: $7 \times 10^{-5} \text{ mol/l}$.

Liquid dye solutions were circulated through the coaxial flash-lamp inner tube, whose diameter was 12.7 mm. The overall discharge length was 200 mm. At the usual excitation energies the laser output energy (~ 200 mJ) is very repetitive. Small areas of the laser beam in the near field were selected with two pinholes with diameters of 20 and 30 μm .

The measured signals were numerically processed. In order to separate the high frequency fluctuations from the low frequency temporal evolution, we have implemented a high pass mathematical filter whose cutoff frequency can be varied.² Once the experimental data were filtered by this method, we calculated the relative fluctuations amplitude percentage, i.e., the ratio between the amplitudes of the high frequency fluctuations and the low frequency temporal evolution. The process was repeated with several pulses (20–30) in the same experimental conditions, and then the results of

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TABLE I. Viscosity at $T=20^\circ\text{C}$ of the solvents used. Data extracted from *Handbook of Chemistry and Physics*, 53rd ed. (CRC, Cleveland, 1972) and Ref. 10.

| Solvent | Viscosity (P) |
|------------------------|---------------|
| Methanol | 0.006 |
| Isopropyl alcohol | 0.025 |
| Ethylene glycol | 0.2 |
| Glycerin | 14.9 |
| Copolymer P (HEMA-MAA) | ∞ |

all individual signals were averaged. The threshold energy is different to each solvent (e.g., around 42 J in methanol but 100 J in glycerin for our measurement system). Therefore in order to compare the relative fluctuations amplitude obtained with the different solvents, it is necessary to measure each solvent with the same excitation energy above threshold. In the experiment that we report, that energy was 68 J.

We observe a monotone decrease of the relative fluctuations amplitude percentage as the solvent viscosity increases. This fact is illustrated in Figs. 1(a) and 1(b) where the temporal profiles corresponding to two different solvents (methanol and glycerin) can be seen. We must remark that the influence of the viscosity depends on the range of fluctuations frequencies analyzed. Figure 2 shows the relative fluctuations amplitude as a function of the mathematical filter cutoff frequency. We can see that below 20 MHz the fluctuations show a different dependence on the viscosity. This seems to point out that only the high frequency fluctuations ($\nu > 20$ MHz) have their origin related with the orientation of the molecules, and therefore the solvent viscosity will affect their relative fluctuations amplitude. We do not observe appreciable changes in the relative fluctuations amplitude percentage neither with the variation of dye concentration nor with the pinhole used to select small areas of the laser beam (20–30 μm of diameter).

As we mentioned previously, the filamentation process is assumed to depend on the orientation of the induced dipoles by the laser field. Now we study the behavior of one dye molecule in the presence of the laser field taking into account the friction experienced due to the solvent. The magnitude of the induced momentum in the polarization axis direction of the molecule is $p = \alpha E \cos \theta$, where θ is the angle between the electric field \mathbf{E} and the polarization axis, and $\alpha = e^2 \lambda / (4 \pi m c \gamma_{\perp})$ is the classical resonant polarizability of the dye molecule,¹³ being e the electron charge, m its mass, λ the electric field wavelength, and γ_{\perp} the depolarization rate. The molecule spins to put its dipole moment parallel to the electric field. The rotation follows the equation:¹⁴

$$I \frac{d^2 \theta}{dt^2} + r \frac{d\theta}{dt} + E p \sin \theta = 0. \quad (1)$$

We have called $r = \kappa a^3 \eta$, with η the solvent viscosity. The effective rotation radius of the molecule a is estimated as an average of its inertial radii, regarding to the molecular geometry. κ is a Stokes-like constant¹⁵ and I is the inertia moment. The inertial term in this Eq. (1) is negligible. Using the induced polarization we integrate the Eq. (1) during the time of radiation of the laser pulse (t_{pulse}), under the simplifying assumption of a constant value of E :

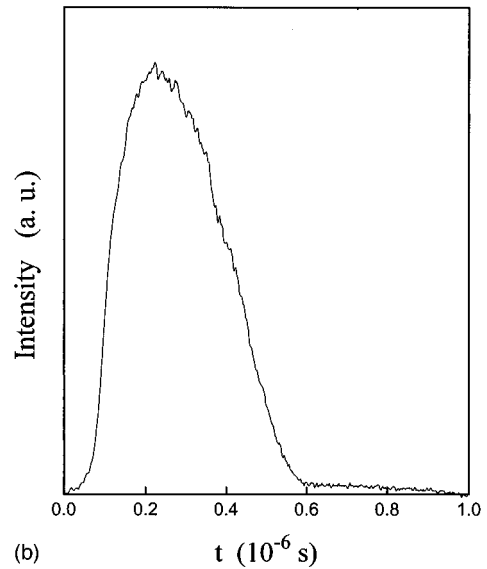
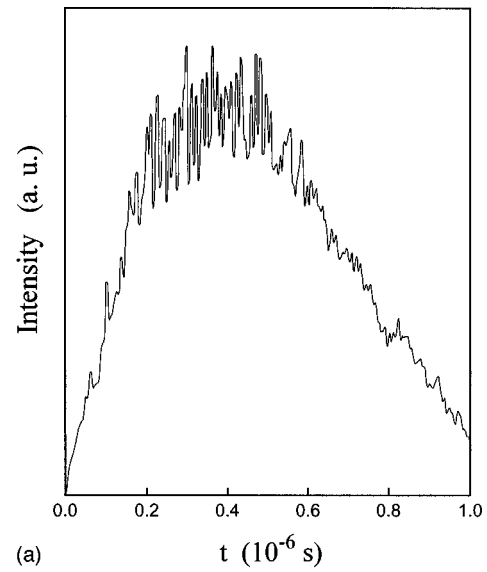


FIG. 1. Experimental signal using a solution of Rhodamine 6G in (a) methanol, (b) glycerin.

$$\tan \theta = \tan \theta_0 \times e^{-\frac{\alpha E^2 t_{\text{pulse}}}{r}}. \quad (2)$$

If we choose the z axis as the direction of the (linearly polarized) electric field, $\mathbf{E} = E \mathbf{e}_z$, then the polarization in the laser field direction is:

$$p_z = p \cos \theta = \frac{\alpha E}{1 + \tan^2 \theta_0 \times e^{-\frac{\alpha E^2 t_{\text{pulse}}}{r}}}. \quad (3)$$

It depends on the initial condition (initial angle θ_0), so we must average over every possible initial angle to obtain the mean polarization induced in the z axis ($\overline{p_z} = \int_0^{\pi/2} p_z \sin \theta_0 d\theta_0$). Hence we get:

$$\overline{p_z} \equiv \frac{\overline{p_z}}{\alpha E} = \frac{1}{1 - e^{-\beta/\eta}} \left[1 - \frac{1}{\sqrt{e^{\beta/\eta} - 1}} \tan^{-1} (\sqrt{e^{\beta/\eta} - 1}) \right], \quad (4)$$

where $\beta \equiv 2\alpha E^2 t_{\text{pulse}} / (\kappa a^3)$. Equation (4) follows the general behavior we can behold in our measurements (see Fig.

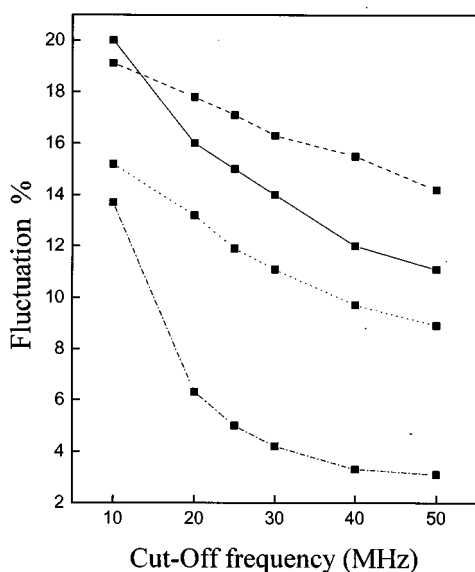


FIG. 2. Relative fluctuations percentage as a function of the mathematical filter cutoff frequency for different solvents: methanol (dashed line), isopropyl alcohol (solid line), ethylene glycol (dotted line), and glycerin (dashed and dotted line).

3). The function decreases as the viscosity increases, and both limits, $\eta \rightarrow 0$ and $\eta \rightarrow \infty$, are finite. If the filamentation model holds,⁸ the molecules become coupled by dipole-dipole interaction and a filamentation of the gain happens.⁹ Those gain filaments are also coupled by the diffracted field giving place to a local dynamics.³ Then it may be expected that the relative fluctuations amplitude is a function of the polarization \mathcal{P}_z . So, we are going to suppose a linear relationship between the relative fluctuations amplitude percentage F and the polarization \mathcal{P}_z .

It is remarkable that $\lim_{\eta \rightarrow \infty} \mathcal{P}_z = 1/3$. This limit represents the induced molecular polarization when the rotation is not possible, and it is the origin of the fluctuations when the viscosity approaches to infinite. Taking into account that

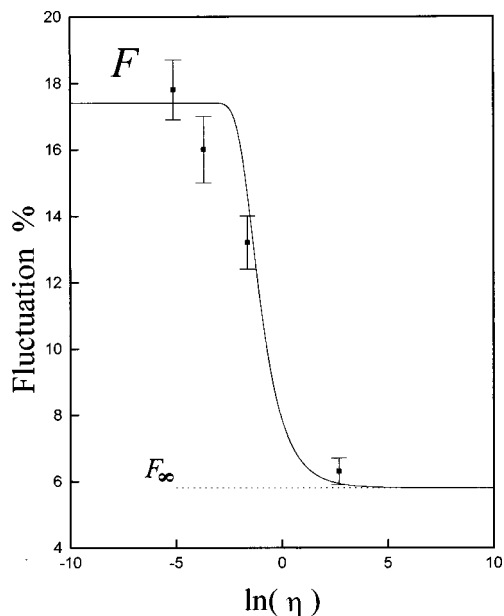


FIG. 3. Fit F (solid line) as a function of the logarithm of the viscosity when cutoff frequency is 20 MHz. Experimental limit measured in copolymer solid solution (dotted line). The viscosity is given in poises.

limit, we use the following function to represent F :

$$F = 3F_{\infty} \mathcal{P}_z, \quad (5)$$

where $F_{\infty} \equiv F(\eta \rightarrow \infty)$ is the relative fluctuations amplitude percentage when the viscosity goes to infinity. This value will be equivalent to the fluctuations in a solid solution, that means, the measurements obtained with the solid copolymer solution can be identified with this limit. We fit the function (5) to our experimental results. The estimated parameters for our dye laser are:^{3,4} $\lambda = 600 \times 10^{-9}$ m, $t_{\text{pulse}} = 0.5 \times 10^{-6}$ s, $\gamma_{\perp}^{-1} = 1 \times 10^{-12}$ s,¹⁶ $a = 3.5 \times 10^{-10}$ m. The value of κ depends on the shape of the solid which is rotating in a fluid,¹⁵ we choose in our case $\kappa \approx 3.5$. The output energy was 240×10^{-3} J. With these values we obtain $E = 1.7 \times 10^6$ V m⁻¹, $\alpha = 4.5 \times 10^{-36}$ C² m² J⁻¹, and $\beta = 0.85$ poises, which are very reasonable values. We use the measured residual fluctuations data in the copolymer solid solution $F_{\infty} \approx 5.8\%$. It is shown in Fig. 3 the fit of the proposed function (5) to the experimental data. As it can be seen, the fit obtained is quite accurate to the experimental results. The error bars on experimental data were obtained by statistically averaging over a large number of pulses (~ 20 – 30).

The above described simple model shows the similarity between the fluctuations dependence on the solvent viscosity and on the average dipole orientation. Thus our present experimental results enlighten the role played by the molecular orientation in the spatiotemporal laser dynamics. They also support models of gain filamentation based on the orientation and coupling of induced molecular dipole moments.³

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