**Abstract**—We study the influence of the solvent viscosity in the local irregular intensity fluctuations characteristic of high Fresnel number dye lasers. The relative amplitude of the fluctuations in a flashlamp-pumped dye laser is analyzed. We complete our previous work by using several solvents and different excitation energies above threshold. A decrease of the relative fluctuation amplitude is found as the solvent viscosity or the pumping energy increases. This effect is theoretically analyzed in the framework of the semiclassical Maxwell–Bloch equations, with a model based on the orientation of the dye molecules driven by the laser field in the solvent. These results point out the importance of the orientation of the dye molecules in the intensity fluctuation phenomenon.

**Index Terms**—Dye laser, local fluctuations, viscosity.

I. INTRODUCTION

ACTIVE AND passive devices with optical feedback have deserved a great deal of attention due to their rich dynamics. Most of the interest focuses on their spatial and temporal behavior, including regimes of activity in which the number of degrees of freedom is controllable [1]–[4] or others where more complex states, such as optical turbulence, are present [5]–[9]. Large-aperture (high Fresnel number) photorefractive, OPO, and laser oscillators share this last type of dynamics. Although in low-aperture devices the boundary conditions play a central role in the spatio-temporal dynamics, in large-aperture oscillators a dynamics that is mediated by bulk mechanisms is expected [9], [10]. The nature of the bulk mechanisms is not clear in many cases and, therefore, experimental results are important to support the theoretical concepts. In that context, the observation of local dynamics in transverse patterns may give significant information about the underlying bulk process. In particular, local irregular intensity fluctuations have been found in dye lasers [5]–[7] and transversely excited atmospheric (TEA) CO$_2$ lasers [11]. The main feature of this behavior is the chaotic oscillation observed when the intensity output is measured in small regions of the laser spot. As the observation area becomes larger, the chaotic oscillation amplitude diminishes until it disappears. This extinction happens when the measured area is much smaller than the whole spot, so an interpretation based on transverse mode dynamics can be discarded. Another interesting feature about this performance was the observation of a predominant frequency which did not depend on the geometrical characteristics of the resonator, the pumping or losses, and thus, appears to be determined by the intrinsic dynamics of the phenomenon. In our flashlamp-pumped dye laser, the predominant frequency was placed at 55 MHz with two harmonics at 110 and 165 MHz [5]–[7].

In order to isolate the fluctuations effect, some additional possible factors that may influence the dynamics were investigated. In previous works [6], light polarization dynamics were tested as a possible cause of the phenomenon studied here. The fluctuations were studied using an intracavity Brewster window in order to fix the polarization of the laser light. However, no measurable changes were observed in regard to the randomly polarized case. Then, we assume the laser light to be linearly polarized.

The laser was operated in a single-pulse regime and the measurements were performed at the same temperature (20 °C) which was maintained by a heat exchange circuit. If we estimate the solvent heat absorption rate to be $\approx$30% of the capacitor stored energy, then the effect on the solvent refraction index must be less than 0.5% [12]. The change is monotonically produced during the pulse and it is homogeneously distributed in the solvent pumped volume; therefore, it can be said that it does not play any role in the fast local fluctuations. For this reason also, thermal lensing and heat removal are not problems in the single-pulse regime.

The flux of the solvent through the lasing region is about 6 l/min. A turbulent regime is established in the region due to the solvent velocity. It is well known that dye flow turbulence can induce linewidth instabilities, as reported by Duarte et al. [13]. Several works deal with the stabilization of intensity and frequency temporal characteristics by neutralizing dye flow turbulence. However, the phenomenon analyzed in this paper is not related to this flow turbulence. In fact, to be sure of the influence of the medium turbulence in the fluctuations studied in this paper, we repeated the experiment with no flux at all, finding the same results.

Therefore, the fluctuations analyzed in this paper reveal the existence of local chaotic dynamics with a low transverse correlation.

In a previous publication, a model was proposed to explain the main characteristics of the intensity fluctuations in high-
Fresnel-number lasers [6]. The model, following the idea of Emelyanov and Yukalov [14], assumed the formation of population inversion filamentary structures which were coupled by the nearest neighboring diffracted field. The model predicted a dynamical frequency locking of the filamentary structures, and it gave an almost invariant average Fourier spectrum with several broad peaks, as it had been experimentally observed.

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 [15]. That means that the local irregular fluctuations observed in liquid solutions (Rhodamine 6G in methanol) practically disappear in solid solutions. This result pointed out an influence of the solvent viscosity on the intensity fluctuations. The Emelyanov and Yukalov model is based on the superradiant dipole–dipole interaction driven by the laser field. In this model, a coupling between the induced dipoles oriented by the laser field takes place. The main shortcoming of that calculation (as it is described in the original paper) is the stationary approach posed on it, being explicitly conceived for application in CW lasers. In the case of pulsed lasers, it is possible in principle that the same coupling takes place, but the dipole orientation must be produced during the laser pulse. Since the polarized molecules are 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, a substantial reduction of the superradiant coordination length and the filamentation capability being foreseeable [16].

In a recent paper, we tested the influence of the solvent viscosity on the fluctuations amplitude. We observed a monotonous decrease of the relative fluctuations amplitude as the solvent viscosity increased [17]. Based on these results, in this paper we have continued our research to get a deeper insight into this effect. We have performed additional measurements in which new experimental parameters have been considered. The current results have led us to a more realistic interpretation of the whole phenomenon. Now we follow a quantum mechanical approach to model the matter behavior. In addition, we guess a nonlinear relationship between the solvent viscosity and the laser electric field (electrorheological effect).

The paper is organized as follows. In Section II, we describe the experimental setup. Section III presents the experimental results. In Section IV, we develop a model to explain the general behavior found in our experiments. Finally, we briefly conclude in Section V.

### II. Experimental Setup

Our main measurement system consists of a coaxial flashlamp-pumped untuned dye laser having a simple resonator made up of two flat mirrors, a broad-band multilayer total reflector and a multilayer partial reflector (30% reflectivity). The measurements were made using Rhodamine 6G in several polar solvents, the viscosities of which are quoted in Table I. A Rhodamine 6G solid copolymer solution was also used as the limit of high viscosity. The dye concentration was varied from $2.5 \times 10^{-5}$ mol/l to $7 \times 10^{-5}$ mol/l. Liquid dye solutions were circulated through the coaxial flashlamp inner tube, the ends of which have been closed by two flat uncoated laser grade polished fused silica windows. The flashlamp inner diameter was 12.7 mm and the overall discharge length was 200 mm. The excitation was carried out using a 0.5-$\mu F$ capacitor, which allows a maximum of 30 kV. The laser was operated in a single shot regime, leaving enough time between two shots to avoid accumulative radial thermal gradients. The solution was changed every one hundred shots to prevent the effects of dye photolysis. Small areas of the laser beam were selected with two pinholes with diameters of 20 and 30 $\mu$m. The light intensity was recorded by an optical fiber connected to a photodiode (risetime $\leq 1$ ns) and linked to a transient programmable digitizer having 1-GHz bandwidth in real time. The entire measurement system was placed inside a Faraday cage.

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 [5]. 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 all individual signals were averaged. 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. We performed two series of measurements: series I using an excitation energy far from threshold (68 J), and series II, near threshold (14 J). We were not able to measure some points in series I because the necessary excitation energy exceeded the system capability.

### III. Experimental Results

We observe in both series a monotonic decrease of the relative fluctuations amplitude percentage as the solvent viscosity increases. This is in agreement with our previous results. Pulse temporal profiles corresponding to two different solvents, methanol and glycerin, are shown in Fig. 1(a) and (b), respectively. We must remark that the influence of the viscosity depends on the range of fluctuation frequencies analyzed. Fig. 2 shows the relative fluctuations amplitude as a function of the mathematical filter cutoff frequency for series I. We can see that below 20 MHz the fluctuations show a

| Table I: Viscosity at $T = 20 \, ^\circ\text{C}$ of the Solvents Used. Percentages Given are Relative to Weight of Components. Data Extracted from [24] |
|---|---|
| Solvent | Viscosity (cP) |
| Methanol | 0.006 |
| Isopropyl alcohol | 0.025 |
| Ethylene glycol | 0.2 |
| Glycerin 88% + H$_2$O 12% | 1.5 |
| Glycerin 92% + H$_2$O 8% | 3.8 |
| Glycerin | 14.9 |
| Copolymer P (HEMA-MMA) | $\infty$ |
different dependence on the viscosity. This seems to point out that only the high-frequency fluctuations ($\nu > 20$ MHz) have their origin related to the orientation of the molecules, and, therefore, the solvent viscosity will affect their relative fluctuations amplitude. It can be also observed that the relative amplitude of the fluctuations is lower as the pumping is higher (see Fig. 3). This last result will be related to the dynamical behavior of the filamentary structures, so it will only be explained using dynamical models of the filamentary structures like the one referenced previously [6]. We do not observe appreciable changes in the relative fluctuations amplitude percentage either with the variation of dye concentration or with the pinhole used to select small areas of the laser beam (20–30 $\mu$m in diameter).

IV. THEORETICAL INTERPRETATION

A. The Model

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. We start our calculation with the slowly varying amplitude Maxwell–Bloch equations for material variables [18]

$$\frac{\partial \vec{p}_0}{\partial \mathcal{H}} = -\left(\gamma_\perp + i(\omega_{12} - \omega)\right)\vec{p}_0 - i\left(\frac{\vec{p}_{12}}{\eta}\frac{\mathcal{E}}{\eta}\right)\vec{p}_{12}d \tag{1}$$

$$\frac{\partial d}{\partial \mathcal{H}} = \gamma_\parallel (d_0 - d) - \frac{2i}{\eta}(\mathcal{E} \cdot \vec{p}_0 - \mathcal{E} \cdot \vec{p}_{12}) \tag{2}$$

where $\vec{p}_0$ is the polarization per molecule induced by the field $\mathcal{E}$, $d$ is the population inversion per molecule, $\gamma_\perp$ the depolarization time, $\omega_{12}$ the transition frequency, $\omega$ the electromagnetic field frequency, $\vec{p}_{12}$ the dipole moment of the transition, $\gamma_\parallel$ the lifetime of the population inversion, and $d_0$ the stationary population inversion per molecule in the absence of laser field. We choose the $z$ axis as the direction of the (linearly polarized) electric field, and $\mathcal{E} = E\hat{e}_z$. $\theta$ is the angle between the laser field $\mathcal{E}$ and the induced polarization $\vec{p}_0$ (or the dipole moment $\vec{p}_{12}$), i.e., $\theta$ is the angle of the dipole moment of the transition with the $z$ axis. We redefine the polarization $\vec{p}_0$ as $\hat{p}_0$. If we consider perfect resonance ($\omega_{12} - \omega \approx 0$), we can take the polarization and the electric field like real variables. In the quasi-stationary regime, we...
obtain for the matter variables

\[ d \simeq \frac{d_0}{1 + \frac{E^2}{E_0^2} \cos^2 \theta} \]  

(3)

\[ \vec{p} \simeq \frac{\alpha_0 E \cos \theta}{1 + \frac{E^2}{E_0^2} \cos^2 \theta} \frac{\vec{p}_{12}}{\hbar/2} \]  

(4)

where \( \alpha_0 = \frac{\mu^2_1 d_0}{\hbar \gamma_{1 \perp}} \) is a molecular polarizability and

\[ E_0 = \sqrt{\frac{\hbar^2 \gamma_{1 \perp}}{(4 \mu^2_1)}} \]

is a saturation field.

We see that the polarization induced by the laser field depends on the angle \( \theta \) between the laser field and the dipole moment of the transition. The molecule will try to rotate to make its dipole moment parallel to the laser field. This rotation follows the equation

\[ \frac{d^2 \theta}{dt^2} + \kappa \alpha^2 \frac{d \theta}{dt} + E_p \sin \theta = 0 \]  

(5)

with \( \eta \) the solvent viscosity, \( \alpha \) the effective rotation radius of the molecule, \( \kappa \) a Stokes-like constant, and \( I \) is the inertia moment. The inertial term in (5) is negligible. Using the polarization (4), we integrate (5) over the time of radiation of the laser pulse \( t_{\text{pulse}} \)

\[ \int_{\theta_0}^{\theta} \frac{1 + \frac{E^2}{E_0^2} \cos^2 \theta'}{\cos \theta' \sin \theta'} \, d\theta' = - t_{\text{pulse}} \frac{\alpha_0 E^2}{\kappa \alpha^2 \eta} \]  

(6)

Hence, we obtain

\[ \left( \frac{\tan \theta}{\tan \theta_0} \right) \left( \frac{\sin \theta}{\sin \theta_0} \right)^{E^2/E_0^2} = \exp \left( - \frac{\alpha_0 E^2 t_{\text{pulse}}}{\kappa \alpha^2 \eta} \right). \]  

(7)

The polarization in the laser field direction is \( p_z = p \cos \theta \), which depends on the initial condition (initial angle \( \theta_0 \)); therefore, we must average over all possible initial angles to obtain the mean polarization induced in the \( z \) axis

\[ \bar{p}_z = \int_0^{\pi/2} d\theta_0 \sin \theta_0 p_z. \]  

(8)

To calculate an expression for \( p_z \), we suppose that during the laser pulse radiation time the rotated angle is smaller than the initial one, i.e., \( \Delta \equiv \theta - \theta_0 \ll \theta_0 \). Using this approximation in (7), we obtain

\[ \Delta \simeq \frac{\sin 2 \theta_0}{2 \left( 1 + \frac{E^2}{E_0^2} \cos^2 \theta_0 \right)} \left( \exp \left( - \frac{\alpha_0 E^2 t_{\text{pulse}}}{\kappa \alpha^2 \eta} \right) - 1 \right). \]  

(9)

In the same way, we approximate the polarization in the laser field direction

\[ p_z \simeq \frac{\alpha_0 E \cos^2 \theta_0}{1 + \frac{E^2}{E_0^2} \cos^2 \theta_0} \left[ \frac{\alpha_0 E \sin^2 2 \theta_0}{2 \left( 1 + \frac{E^2}{E_0^2} \cos^2 \theta_0 \right)^3} \right. \]

\[ \cdot \left( \exp \left( - \frac{\alpha_0 E^2 t_{\text{pulse}}}{\kappa \alpha^2 \eta} \right) - 1 \right). \]  

(10)

We introduce (10) into (8) to perform the integration over the initial angles \( \theta_0 \)

\[ P \equiv \frac{\bar{p}_z}{\alpha_0 E_0} = P_c - P_l \left( \exp \left( - \frac{\alpha_0 E^2 t_{\text{pulse}}}{\kappa \alpha^2 \eta} \right) - 1 \right). \]  

(11)

This is the dimensionless mean induced polarization in the laser field direction reached at \( t = t_{\text{pulse}} \). The value of the parameters \( P_c \) and \( P_l \) are

\[ P_c \equiv \frac{E_0}{E} \left( 1 - \frac{E_0}{E} \arctan \left( \frac{E}{E_0} \right) \right) \]  

(12)

\[ P_l \equiv \frac{1}{4} \left( \frac{E_0}{E} \right)^3 \left[ 1 + \frac{2 - 1 + \frac{E^2}{E_0^2}}{E_0^2} \right. \]

\[ \left. + \left( 1 - 3 \frac{E^2}{E_0^2} \right) \arctan \left( \frac{E}{E_0} \right) \right]. \]  

(13)

Equation (11) follows the general behavior we can behold in our measurements (see Fig. 4). The function decreases as the viscosity increases, and both limits \( \eta \to 0 \) and \( \eta \to \infty \) are finite. If the filamentation model holds, it can be expected that the relative fluctuations amplitude is a function of the polarization \( P \). Exponential dependencies between the system variables often appear in amplification regimes in lasers, and in our case it also seems to be the closest behavior to the experimental results. Hence, we take the following tentative function representing the relative fluctuations amplitude:

\[ F \propto \exp \left( \eta P \right) \]  

(14)

where \( \eta \) is a constant for each series which is measured with the same excitation energy above threshold. The relative fluctuations amplitude percentage when the viscosity goes to infinite \( (F_{\infty} \equiv F(\eta \to \infty)) \) will be equivalent to the fluctuations in a solid solution. Therefore, the measurements obtained with the solid copolymer solution can be identified with this limit \( F_{\infty} \). Using this value, we rewrite function (14) as

\[ F = F_{\infty} \exp \left[ -\eta P \left( \exp \left( - \frac{\alpha_0 E^2 t_{\text{pulse}}}{\kappa \alpha^2 \eta} \right) - 1 \right) \right]. \]  

(15)
We must bear in mind that we are applying very high power electrical fields to polar solvents, which can produce electrorheological effects, i.e., instantaneous reversible changes in the apparent viscosity when the fluid is subjected to an externally applied electric field [19]. This type of effect was reported as early as 1896 [20]. In the late 1940’s, this effect was also observed, but this time in suspensions, where the change in the apparent viscosity was very large [21]. The relationship between the apparent viscosity and the applied electric field is quadratic. So, we write the apparent viscosity as

\[ \eta = \eta_0 (1 + \psi E^2) \]  

(16)

where \( \eta_0 \) is the viscosity in absence of the electric field (see Table I), and \( \psi = 2.0 \times 10^{-12} \text{ V.m}^{-1} \) is constant. We have chosen the constant value \( \psi \) taking into account the measured data in [22], which showed approximately a 25% relative increase in the viscosity of the polar fluid when it was exposed to an electric field of \( E \approx 3 \times 10^5 \text{ V.m}^{-1} \). Finally, the function \( \eta \) which yields

\[ \eta = \eta_0 (1 + \psi E^2) \]  

(16)

represents a polarizability field transverse radius of the resonator, \( \gamma_1^{-1} = 1 \times 10^{-12} \text{ s}, \) \( \alpha_\parallel^{-1} = 4 \times 10^{-9} \text{ s}, \) \( \alpha = 3.5 \times 10^{-10} \text{ m}, \) and \( \eta_0 \approx 1 \). The value of \( \kappa \) depends on the shape of the solid which is rotating in a fluid [23]. We choose in our case \( \kappa \approx 2.0 \). Since the quantum efficiency in Rhodamine 6G is very high, we can approximate the lifetime of the population to the spontaneous lifetime. In this way, we get a value for the dipole moment \( \mu^2 \approx 3 \times 10^{-30} \text{ C.m}^2 \) and \( \kappa \approx 2.0 \) which yields a polarizability \( \alpha_0 \approx 3 \times 10^{-30} \text{ C.m}^2 \). The output energies were 240 \( \mu \text{J} \) and 50 \( \mu \text{J} \) for series I and II. These values are proportional to their respective excitation energies above threshold. We use the measured residual fluctuations data in the copolymer solution \( F_{\infty} \approx 5.8 \% \) and \( F_{\infty} \approx 13.0 \% \), which act as limits of \( F_{\infty} \) and \( F_{\infty} \). The fitting parameters take on the values \( \eta_{\infty} = 2.16 \times 10^4 \) and \( \eta_{\infty} = 2.2 \times 10^3 \). In Fig. 3, the fits of the proposed functions (17) to the experimental data are shown for both series (\( F_{\infty} \) and \( F_{\infty} \)). It can be seen that the fits obtained are good in both series. The error bars on the experimental data were obtained by statistically averaging over a large number of pulses. (\( \sim 20-30 \)).

V. CONCLUSIONS

The effect of the solvent viscosity in the local irregular intensity fluctuations characteristic of high Fresnel number dye lasers is analyzed. We perform the measurements using different solvents (see Table I) and two excitation energies above threshold. The relative amplitude of the fluctuations is lower as the viscosity is higher, in agreement with our previous work. About the excitation energy, it is found that the relative amplitude of fluctuations decreases as the pumping increases. We derive a model based on the Maxwell–Bloch equations and the molecular orientation of the dye molecules driven by the laser field in the solvent. An electrorheological effect in the solvent is also considered. The model gives a good fit to the experimental data.

These results provide a clear evidence of the role played by the molecular orientation in the spatio-temporal laser dynamics. They also support models of gain filamentation based on the orientation and coupling of induced molecular dipole moments [6].

REFERENCES


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