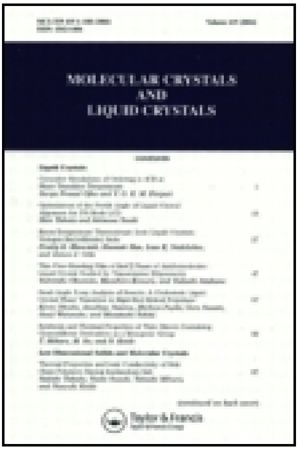

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Absorptive Dynamic Diffraction Gratings in a Dye-Doped Nematic Liquid Crystal

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Absorptive Dynamic Diffraction Gratings in a Dye-Doped Nematic Liquid Crystal

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Dynamic diffraction grating formation by crossed pump beams with wavelength 514 nm in a mixture of 5CB and azodye R4 was investigated by a probe beam with wavelength 633 nm. The dependence of the diffraction efficiency on the pump and probe polarization is consistent with the dye bleaching and corresponding buildup of the cis species as the main mechanism for the optical grating formation. Diffraction at 633 nm is absorptive and is caused by the periodic distribution of the cis species which absorbs in the red. This observations are in quantitative agreement with simutaneous measurements of absorption at 514 nm and at 633 nm. A rate equation model of the grating formation is presented.

INTRODUCTION

The research on light induced changes in the optical properties of nematic liquid crystals mixed with dyes has mostly been focused on the very large changes of the effective index of refraction which are due to the reorientation of nematic director. In dye-doped nematics optically induced reorientation is greatly enhanced through the changes in the orientational distribution of the dye molecules in the ground and excited states, that is, by the well-known Janossy effect [1–3] . Lately, optical nonlinearities which are similar to photorefractivity in solid crystals have also been observed and studied [4] . Both these effects depend on nematic director reorientation and are therefore tensorial, that is, the main change occurs in the off-diagonal components of the dielectric tensor.Scalar changes of the optical properties in nematic - dye systems are usually attributed to the rise in local temperature due to light absorption. In this paper we report on light induced dynamic grating diffraction measurements in a mixture of 5CB and azo dye 4'-dimethylaminophenyl-[1,4-phenylenebis(azo)]-3-chloro-4-heptyloxy benzene (R4). R4 in its ground state trans configuration absorbs

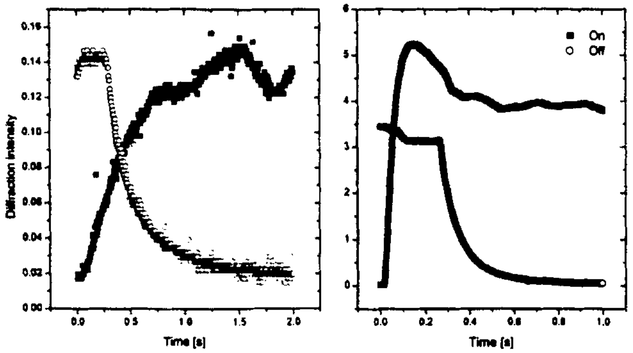

Figure 1: Probe diffraction intensity after switching on and off the pump beam for low (1 mW) and high (47 mw) pump power.

in green and blue and upon absorption can switch to the metastable cis configuration, from where it thermally relaxes back to the trans state in about 10 s [7] . In the diffraction experiment we create a grating with two crossed laser beams at 514 nm and observe it with a He-Ne probe beam. Similar experiments have been performed in some other azo dyes in nematic liquid crystal [5,6] . In order to explain our findings, we also performed measurements of the changes in absorption and linear dichroism at 633 nm that occur as the pump at 514 nm is switched on. These measurements are similar to the experiments in the dye population dynamics performed by Janossy and Szabados [7] . We show that the grating diffraction can be fully explained by a periodic distribution of the cis species of R4, generated by the two pump beams, that absorbs at 633 nm.

EXPERIMENT

The experiments were carried out on a 0.5 % mixture of R4 in 5CB at room temperature. The orientation of the sample, achieved with rubbed polyimide, was planar. The thickness was 24 μm. The experimental setup for the diffraction experiment was as follows. Argon laser beam was divided by a beam splitter into two partial unfocused beams with diameter 3 mm which crossed in the plane of the sample at an angle ranging from 0.007 to 0.033 radians, so that the resulting grating had a wavenumber from $1.46 \cdot 10^5$ m^{-1} to $6.9 \cdot 10^5$ m^{-1}. Most data were taken at 0.007 radians where the diffraction signal is strongest. The polarization of each pump beam could be changed. One or both of the beams were modulated with a mechanical chopper which allowed us to measure the growth and decay of the dynamic diffraction gratings. The power in each beam was up to 30 mW. The gratings were probed by a low power (around 1 mW) He-Ne laser beam with adjustable polarization. The incidence angel of the probe beam was close to zero so that Bragg condition was not satisfied and multiple diffraction orders could be observed when the amplitude of the grating was large.

We observe the strongest diffraction when both pump beams and the probe beam are polarized parallel to the director n, that is, when all polarizations are extraordinary (e). We also observe relatively strong diffraction when the pump beams are ordinary and the probe is extraordinary and a smaller signal for e-polarized pump and o-polarized probe. All other choices of polarization give about 100 times smaller signal than for e-polarizations. In particular, the diffraction is weak for polarization choices that are most sensitive to director reorientation, for example when all the beams are polarized at 45° to n.

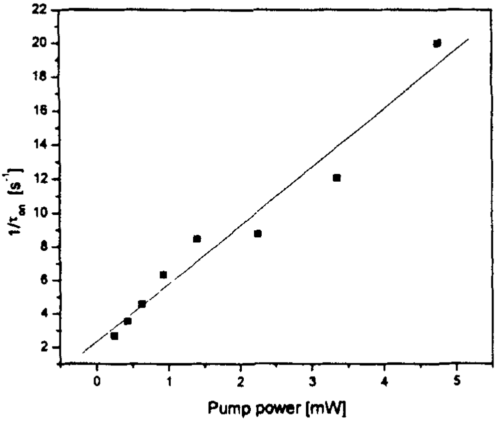

Figure 2: Reciprocal rise-time of the diffraction intensity vs. pump power.

Figure 1 shows the dependence of the probe 1st order diffraction intensity

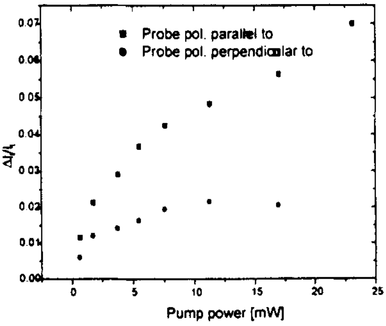

Figure 3: Relative decrease in probe transmission at 633 nm vs. pump power for extraordinary and ordinary probe polarization. Pump is e-polarized.

I_d as a function of time after the pump beams are switched on and off. All beams are ordinary. Two pairs of curves are shown, one for small and one for large pump power. For low power the signal increases monotonously after the pump is switched on, while for high power the signal first has a peak and then decreases. The inverse rise-time of the diffraction grating is shown in Figure 2. The dependence is approximately linear, as can be seen from the fit to the data.

The fact that I_d is largest when all polarizations are extraordinary suggests that the observed grating is either thermal or absorptive. We will discuss thermal effects below. In order to clarify the role of absorption we carried out simultaneous measurements of absorption of the pump beam at 514 nm and of probe at 633 nm. The setup used was the same as for the diffraction experiment except that only one of the pump beams was used. The pump beam was again chopped so that time dependence of the probe absorption could be studied. An experiment of this kind has already been performed by Janossy and Szabados [7], except that they only used Ar laser in their measurements.

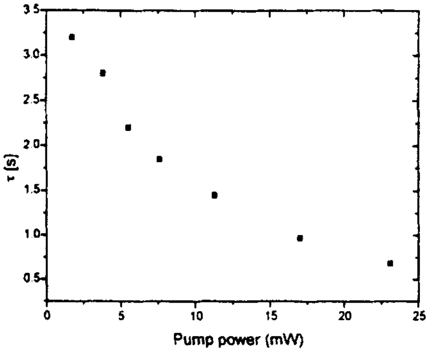

Figure 4: Rise-time of the probe absorption vs. pump power after switching on the pump.

For the purpose of explaining diffraction the most important result of the absorption experiment is that while there is nearly no absorption at 633 nm with the pump off, that is, with all the dye in the trans configuration, absorption at 633 nm increases with increasing pump power. This shows that the cis conformer absorbs at 633 nm. Figure 3 shows the change in sample transmissivity at 633 nm as a function of the e-polarized pump power for both e- and o- probe polarizations. The rise-time of absorbance at 633 nm vs. pump power after it is switched on is shown in Figure 4. In a simple model that we will present below, one would expect this rise-time to be inversely proportional to P. The dependence in Figure 4 is in fact weaker which is probably due to the fact that the sample is optically thick at 514 nm so that the pump power is not homogeneous across the sample. The decay time of the cis absorbance after the pump was switched off was around 3 s. This is considerably shorter than the cis-to-trans thermal relaxation rate of 9 s, reported in [7] . The increase in the relaxation rate is due to the photoexcitation by the probe beam.

DISCUSSION

The fact that the trans species of the dye does not absorb at 633 nm while the cis species does, implies that in our diffraction experiment we get an absorptive grating at 633 nm due to the periodic distribution of the cis species, created by the two pump beams. From the results presented in Figure 3 we can calculate the expected diffraction efficiency at a given pump power. At pump power 5 mW the relative change in the transmitted e-polarized probe intensity is 0.04. The diffraction efficiency, calculated in the thin grating Fraunhoffer approximation should then be around 10^{-3}, so the agreement is quite good.

In the geometry used in the diffraction experiment, where all the beams are e-polarized, there can be no orientation contribution to the diffraction efficiency. There can be, however, thermal contribution. For the purpose of estimating the periodic part of the temperature increase in the sample, we can assume that heat is conducted only in the direction of the grating. Inclusion of heat conduction to the sample walls would make the temperature variation only smaller. Then we get that the amplitude of the temperature grating is $\delta T = I_{abs}/h\lambda q^2$, where h is the sample thickness and λ is the heat conductivity of 5CB. This gives $\delta T \sim 5$ mK, from where the change in the extraordinary index of refraction is $\sim 10^{-4}$, much too small to contribute appreciably to the observed I_d. Also, thermal relaxation rate should be about 1000 times faster than observed.

The data in Figure 3 also allow us to calculate the order parameter of the dye in the cis configuration. Using $S_{cis} = (\alpha_{\parallel} - \alpha_{\perp})/(\alpha_{\parallel} + 2\alpha_{\perp})$ and assuming that the absorption dipole orientation axis remains parallel to $\mathbf{n}$, we get $S_{cis} = 0.25$ which agrees well with the measurements reported in [7]

The assumption that in our case diffraction is mainly caused by dye absorption can be further tested by comparing the observed behavior of the diffraction signal with the predictions of a rate equation model which is an extension of the models presented in [6, 7]. Let us denote the concentration of the trans and cis molecules by N_t and N_c. The trans molecules are pumped into cis state via optical absorption to an excited state which is short-lived. The cis molecules decay back to the trans state thermally and by photoexcitation. We must also include translational diffusion terms for both dye species. The pump intensity $I = I_0(1 + \cos qx)$ has a periodic spatial dependence, so we write N_t and N_c in the form $N_t = N_{t0} + N_{tq} \cos qx$. This leads to the following system of equations:

$$\begin{aligned}
\frac{dN_{t0}}{dt} &= -\left[(a+b)\,I_0 + \frac{1}{\tau}\right] N_{t0} - \frac{1}{2}a\,I_0 N_{tq} + \frac{1}{2}b\,I_0 N_{cq} + \left(\frac{1}{\tau} + b\,I_0\right) N \\
\frac{dN_{tq}}{dt} &= -(a+b)\,I_0 N_{t0} - \left(a\,I_0 + D_t\,q^2\right) N_{tq} + \left(\frac{1}{\tau} + b\,I_0\right) N_{cq} + b\,I_0\,N \\
\frac{dN_{cq}}{dt} &= (a+b)\,I_0 N_{t0} + a\,I_0\,N_{tq} - \left(\frac{1}{\tau} + b\,I_0 + D_c\,q^2\right) N_{cq} - b\,I_0\,N
\end{aligned}$$

Here $a\,I_0$ and $b\,I_0$ are the photoinduced rates of transition from the trans to cis state and vice-versa due to pump, $1/\tau$ is the thermal relaxation rate from cis to trans, which can also include the constant contribution of the probe photoexcitation, N is the total concentration of the dye, and D_t and D_c are the translational diffusion constants of both dye species. N_{c0} has been eliminated through $N = N_{t0} + N_{c0}$.

We do not posses sufficient data to determine the coefficients of the model, although more extensive measurements of the kind presented here should al-

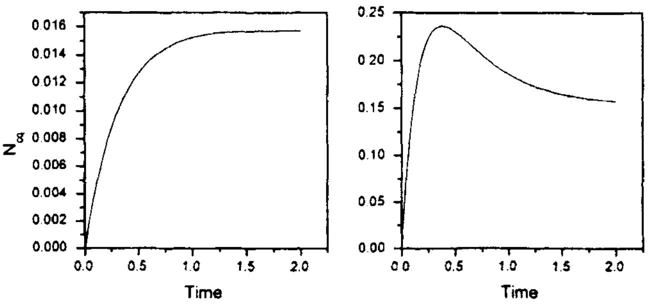

Figure 5: Model calculation of $N_{cq}(t)$ for two pump powers. The ratio of pump power is the same as in Figure 1.

low one to do so, in particular, it should be possible to determine D_t and D_c. The ratio a/b and τ were already determined in [7]. In Figure 5 we only present time dependence of the diffraction signal after the pump has been switched on in case of low and high pump power. The model calculation shows the same behavior as the experimental data in Figure 1 and so lends further support for our explanation of the diffraction mechanism.

It should be noted that the cis absorption contribution to the diffraction signal is also present when the pump and/or probe beams are o-polarized or when **n** is at some angle with respect to pump and probe polarizations. That means that it will contribute also in situations where director reorientation is important. This must be taken into account when studying director reorientation in dye-doped nematic systems.

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References

[1] I. Janossy and A. D. Loyd, *Mol. Cryst. Liq. Cryst.* **203,** 77 (1991).
[2] I. Janossy and T. Kosa, *Opt. Lett.* **17,** 1183 (1992).
[3] I. Janossyand L. Szabados, *Phys. Rev. E* **58,** 4598 (1998).
[4] I. C. Khoo, S. Slussarenko, B. D. Guenther, Min-Yi-Shih, P. Chen, M. V. Wood, *Opt. Lett.* **23,** 253 (1998).
[5] A. G. Chen and D. J. Brady, *Opt. Lett.* **17,** 441 (1992).
[6] B. Saad, M. M. Denariez-Roberge, and T. V. Galstyan, *Opt. Lett.* **23,** 727 (1998).
[7] I. Janossy and L. Szabados, J. Nonlin. *Opt. Phys Mat.* **7,** 539 (1998).