

Limiting factor of the diffraction efficiency in azo dye doped polymers

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ABSTRACT

We have used theoretical models to give an account of the photoinduced reorientation of the azo-dye molecules in polymers and the related macroscopic effects that are diffraction efficiency and photoinduced birefringence. Measurements have been carried out for three doped polymers presenting different behaviors according to the writing intensity and the sample temperature. Interpolation of the experimental data reveals that the limiting factor for the amplitude of the diffraction efficiency is principally the temperature of the samples, whereas then the sensitivity of the compounds seems to be only driven by the nature of the dye.

Keywords: azo dye, photoinduced birefringence, diffraction, temperature

1. INTRODUCTION

Holographic support that can reversibly record information as a phase modulation is required in various fields: in data storage as a large capacity volume memory,¹ in opto-electronic for optical computing,² in photonics as conjugated mirror, in non-destructive testing as recordable media,³ . . . According to the application, figures of merit required are quite different, some needing good diffraction efficiency, some high sensitivity and some both. Moreover, the used wavelengths are ranged from the far IR (for telecommunication) to the limit of the blue visible spectrum (for data storage). To satisfy all these requests, it is necessary to develop a large number of different materials. Doped polymers are an interesting way to achieve these goals since they can be tailored by chemical engineering to match the different needs. Polymers have also intrinsic properties which make them interesting: they can be set down in thin layer or in bulk, spread over large surfaces, formed in complex shapes.

One possibility to synthesize such a compound is to use azo-dye doped polymers. In this kind of material, refractive index can be changed by using polarized light. Indeed, azo-chromophores can reorient themselves by multiple trans to cis photo-isomerisations and cis to trans back photo or thermal relaxations. Since the molecules can absorb light only if there is a component of their electric field vector along the isomerisation transition axis, the latter tend to be oriented perpendicularly to the polarization vector of the incident light. According to recent results,⁴ it appears as evident that this orientation process is driven by random walk rather than by constant rotation induced by torque. The refractive index change is a consequence of the reorientation since azo-chromophores are birefringent molecules, i.e. their dipolar momentum along the transition axis is different than the others.

Macroscopic results of the microscopic reorientation are photoinduced birefringence and phase holograms writing. The birefringence can be induced by illuminating the sample with a linearly polarized light beam, and it can be observed by using a weakly absorbed beam linearly polarized at 45° according to the writing beam polarization. The latter will undergo a modification of its polarization due to the birefringence of the sample; the ellipticity of this reading beam can be shown by placing an analyzer behind the sample. The reason of the 45° angle between polarizations of the writing and the reading beams is that, at this angle, the reading polarization axis is the bisector of the sample propagations axes and thus undergoes the maximum of the effect.⁵ The situation is quite different for the holographic recording, where two polarized laser beams interfere into the sample. The polarizations of the writing beams do not need to be parallel,^{5,6} nor linear.⁷ Thus, it is possible to use this kind of media to write

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holograms with orthogonally or circularly polarized beams. To read the holograms, a weakly absorbed laser beam placed at the Bragg angle can be used. The latter can also have a particular polarization in order to enhance some effects.⁸

The advantages to use azo-dye doped polymers are that it already exists a lot of chromophores, that promise an important possibility of tailoring the figures of merit; these molecules can easily be mixed with a great number of polymeric matrices, that allows to avoid light diffusion problems or to change the mechanical properties of the sample; there is no need of high electric field (as it is the case in photorefractive media⁹) and it is possible to store information by different means (polarization, intensity, holography, coherent or incoherent light). Nevertheless, the figures of merit, i.e. efficiency and sensitivity, are generally poor for these materials.

This work come within the scope of a better understanding of the microscopic mechanisms of photo-orientation, in order to overcome the remaining problems. For that purpose, we have studied the diffraction efficiency as well as the photoinduced birefringence according to the writing intensity and the sample temperature of three different polymers doped with the same chromophore. This lets appear the influence of the polymer matrix and the chemical bond on the optical properties. We have also interpolated the experimental data with a theoretical model presented in the next section in order to extract parameters which allow objective comparison between compounds.

2. THEORY

The orientation of the chromophores, induced by light, is perturbed by the thermal agitation which tends to randomize the organization. The angular distribution of the molecules can then be described by a Maxwell-Boltzmann distribution:

$$nb_{(\theta)} = \frac{N \exp \left[\frac{-A_{(T)} \varepsilon \cos^2(\theta)}{kT} \right]}{\int_0^\pi \exp \left[\frac{-A_{(T)} \varepsilon \cos^2(\theta)}{kT} \right] \frac{\sin(\theta)}{2} d\theta} \frac{\sin(\theta)}{2}, \quad (1)$$

where θ is the angle formed by the polarization axis of the writing beam and the isomerisation axis of the chromophore (see figure 2(a)); N is the number of chromophores; ε is the writing beam electric field amplitude and $A_{(T)}$ describes the ability for a molecule to rotate. $A_{(T)}$ is a global parameter which contain physical constants as isomerisation cross section, polymer viscosity, relaxation probability, ...

We must now determine the behavior of the parameter $A_{(T)}$ versus temperature. According to the free volume theory, molecules included in a polymer matrix are encapsulated in a cavity which diameter decreases with temperature.¹⁰ At a threshold temperature T_0 , this cavity reaches the chromophores size, preventing the latter to rotate freely. under these conditions, all the dye molecules are perfectly oriented by the writing beam and decreasing furthermore the temperature does no longer influences the chromophores order. Equation 1 of the angular distribution becomes:

$$nb_{(\theta)} = \frac{N \exp \left[\frac{-A \varepsilon \cos^2(\theta)}{k(T-T_0)} \right]}{\int_0^\pi \exp \left[\frac{-A \varepsilon \cos^2(\theta)}{k(T-T_0)} \right] \frac{\sin(\theta)}{2} d\theta} \frac{\sin(\theta)}{2} \quad \text{for } T \geq T_0. \quad (2)$$

By comparing equation 1 with equation 2, we found the following formula for $A_{(T)}$:

$$A_{(T)} = A \frac{T}{T - T_0}. \quad (3)$$

This formalism raises the problems that the behavior according to the temperature is discontinuous. In figure 1, we have plotted the inverse of the term contained in the exponential versus temperature with and without the introduction of the temperature threshold.

It is clear that equation 3 has been obtained phenomenologically and does not come from a microscopical theory of the photoinduced reorientation process. However, it is interesting to discuss the shape of $A_{(T)}$ plotted as insert of figure 1 in order to justify its behavior as molecular reorientation. $A_{(T)}$ describes the ability of the molecules to rotate either by the influence of the writing beam or by the thermal agitation. The function starts from the infinity at T_0 to drop towards A when temperature increases. This means, in microscopic term, that near T_0 , the alignment

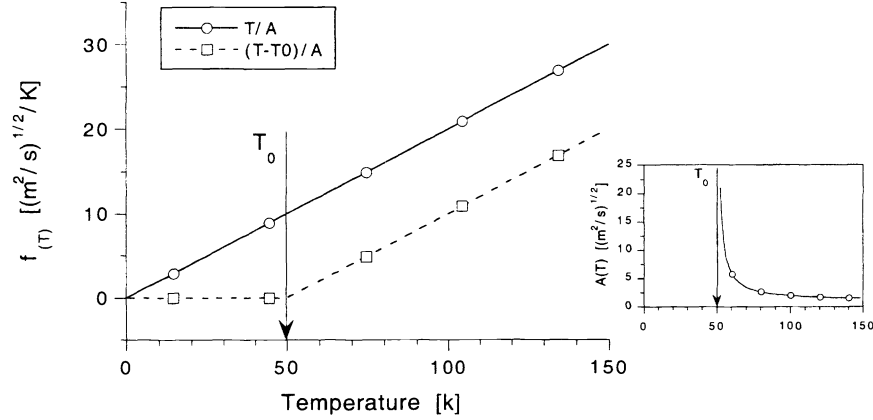


Figure 1. Inverse of the term contained in the exponential of the angular distribution of the chromophores. Insert: function $A(T)$ versus temperature (see text for more details).

of the chromophores by the writing beam is still under completion whereas the rotation by thermal agitation is no longer occurring. At higher temperatures, both processes (isomerization and random reorientation) occur at a constant rate (A).

According to the angular distribution of the chromophores previously introduced; we will calculate, in the next two sections, the diffraction efficiency and the transmission efficiency coming from the photoinduced birefringence. These parts have been developed in order to compare directly the theory of reorientation to the measurements we have undertaken (section 3). More detailed developments can be found elsewhere.¹¹

2.1. Diffraction efficiency

As described in the introduction, holograms can be recorded in this kind of compounds by the modification of the refractive index in the illuminated zones. However, since the absorption coefficient of the polymer is quite high at the writing wavelength, the writing intensity decreases throughout the film thickness. Thus, the refractive index modulation varies according to the sample depth. To take into account this phenomena, we have used a matrix formalism to describe the diffraction efficiency.^{12,13} Transmitted (R_{out}) and diffracted (S_{out}) waves are given by:

$$\begin{pmatrix} R_{out} \\ S_{out} \end{pmatrix} = \prod_{k=1}^N M_k \begin{pmatrix} 1 \\ 0 \end{pmatrix}. \quad (4)$$

According to our experimental conditions (see chapter 3), M_k , the diffraction matrix of the k^{th} layer, is given by the following expression:

$$M_k = \exp\left(\frac{-\alpha_r d}{2 \cos \theta_B}\right) \begin{pmatrix} \cos\left(\frac{\pi d}{\lambda \cos \theta_B} n_1\right) & -i \sin\left(\frac{\pi d}{\lambda \cos \theta_B} n_1\right) \\ -i \sin\left(\frac{\pi d}{\lambda \cos \theta_B} n_1\right) & \cos\left(\frac{\pi d}{\lambda \cos \theta_B} n_1\right) \end{pmatrix}, \quad (5)$$

where α_r is the absorption coefficient at the reading wavelength; d is the layer thickness; θ_B is the Bragg angle at the reading wavelength; λ is the reading wavelength and n_1 is the refractive index modulation between illuminated and dark zones of the holographic grating.

In order to calculate the refractive index modulation (n_1), we assume that, in average over the thickness of the sample and over the height of the holographic grating, the propagation modes of the light are parallel and perpendicular to the polarization axis of the writing beams. This hypothesis is in agreement with the orientation mechanism. The refractive indices effectively encounter by the reading beam are obtained by carrying out a statistical

average over the index ellipsoid, balanced by the angular distribution of the molecules. In agreement with the experimental geometry shown in figure 2(a), refractive indices according to the y and z polarization and according to the dark or illuminated zones of the hologram are given by the following equations:

$$n_{y\text{dark}} = \iint_0^\pi \frac{\sin \theta}{2\pi} \sqrt{\frac{1}{\frac{\sin^2 \varphi + \cos^2 \theta \cos^2 \varphi}{n_o^2} + \frac{\sin^2 \theta \cos^2 \varphi}{n_e^2}}} d\theta d\varphi, \quad (6a)$$

$$n_{z\text{dark}} = \iint_0^\pi \frac{\sin \theta}{2\pi} \sqrt{\frac{1}{\frac{\sin^2 \theta}{n_o^2} + \frac{\cos^2 \theta}{n_e^2}}} d\theta d\varphi, \quad (6b)$$

$$n_{y\text{illuminated}} = \iint_0^\pi \frac{\exp\left[\frac{-A\varepsilon_{(d)} \cos^2 \theta}{k(T-T_0)}\right] \frac{\sin \theta}{2}}{\int_0^\pi \exp\left[\frac{-A\varepsilon_{(d)} \cos^2 \theta}{k(T-T_0)}\right] \frac{\sin \theta}{2} d\theta} d\varphi \sqrt{\frac{1}{\frac{\sin^2 \varphi + \cos^2 \theta \cos^2 \varphi}{n_o^2} + \frac{\sin^2 \theta \cos^2 \varphi}{n_e^2}}} d\theta d\varphi, \quad (6c)$$

$$n_{z\text{illuminated}} = \iint_0^\pi \frac{\exp\left[\frac{-A\varepsilon_{(d)} \cos^2 \theta}{k(T-T_0)}\right] \frac{\sin \theta}{2}}{\int_0^\pi \exp\left[\frac{-A\varepsilon_{(d)} \cos^2 \theta}{k(T-T_0)}\right] \frac{\sin \theta}{2} d\theta} \sqrt{\frac{1}{\frac{\sin^2 \theta}{n_o^2} + \frac{\cos^2 \theta}{n_e^2}}} d\theta d\varphi. \quad (6d)$$

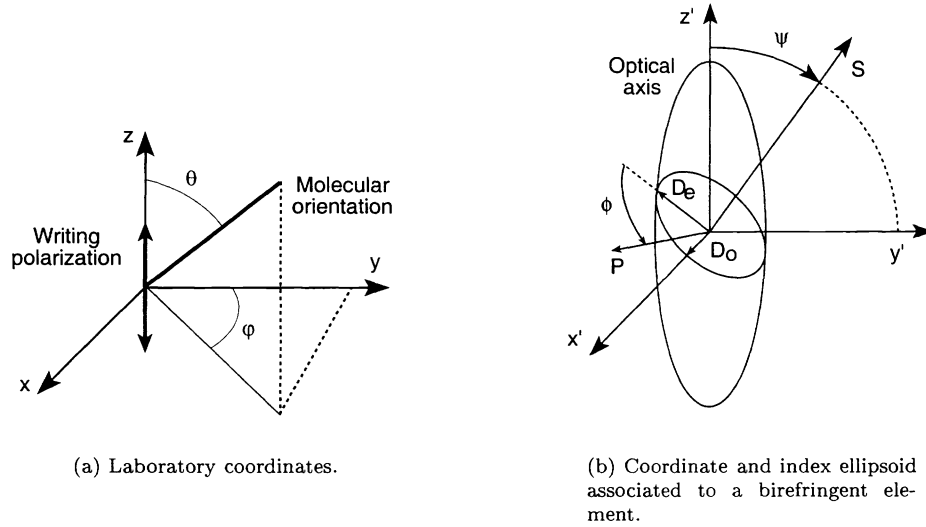


Figure 2. The two coordinates systems used in the theoretical models. z is along the writing polarization, x is along the reading beams propagation. The z' axis is along the optical axis, x' is along the polarization of the writing beam, S is the direction of propagation of the reading beam and P is the reading polarization axis.

2.2. Photoinduced birefringence

When chromophores are reoriented by the light polarization field, the polymer becomes birefringent. As shown by the angular distribution equation (number 2), the alignment (and consequently, the birefringence) is proportional to the intensity of the light electric field. As for the diffraction efficiency, the latter is not constant over the sample thickness and therefore, we must take into account the optical absorption of the polymer. In this case, one can consider the sample as a stake of birefringent layers each one having a particular orientation of its principal axes and phase delay. By using the Jones formalism, the sample can be described as a product of matrices acting on the electric vector of the incident light:

$$\begin{pmatrix} E'_x \\ E'_y \end{pmatrix} = \exp\left(-\frac{N\alpha_{HeNe}d}{2}\right) \frac{1}{2} \begin{pmatrix} 1 & -1 \\ -1 & 1 \end{pmatrix} \times [R_{(-\phi^N)} \times W_0^N \times R_{(\phi^N)}] \times \dots \times [R_{(-\phi^1)} \times W_0^1 \times R_{(\phi^1)}] \times \frac{1}{2} \begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix} \times \begin{pmatrix} E_x \\ E_y \end{pmatrix}. \quad (7)$$

The unitary matrices refer to the polarizer and the analyzer in the experimental set-up. $R_{(\phi)}$ is the rotation-matrix and W_0 is the Jones matrix describing the phase delay. Both ones are defined as follow:

$$R_{(\phi)} = \begin{pmatrix} \cos \phi & \sin \phi \\ -\sin \phi & \cos \phi \end{pmatrix}, W_0 = \begin{pmatrix} e^{-i\frac{\Gamma}{2}} & 0 \\ 0 & e^{i\frac{\Gamma}{2}} \end{pmatrix},$$

where ϕ is the angle between the reading polarization and the vector D_e (see figure 2(b)) and Γ is the phase delay expressed in equation 8.

$$\Gamma = \frac{2\pi}{\lambda} d (D_o - D_{e(\psi)}) \quad (8)$$

D_o and $D_{e(\psi)}$ are the projections of the ordinary and extraordinary axes of the indices ellipsoid onto the plan perpendicular to the reading beam propagation axis. Representation of these values as the angle ϕ can be found in figure 2(b).

By introducing the angular distribution of the birefringent elements (equation 1) into the equation 7 of the light propagation in the sample, it is possible to carry out a numerical simulation by using the Monte-Carlo technique.

3. EXPERIMENTS

3.1. Polymers design

In order to show the influence of the polymeric matrix over the photoinduced orientation process, we have synthesized three compounds where the same chromophore has been included by different ways. Azo-dye is based on the 2,5-dimethyl-4-(p-nitrophenylazo)anisole (DMNPAA), the amount of chromophore in the matrix is 10 weight percent for all the materials.

For the first compound, we have mixed the birefringent molecule to Poly[N-vinyl carbazole] (PVK) which is a well-known compound for photorefractive experiments.¹⁴ The two other matrices are copolymers of [ω -(N-carbazolyl)alkylmethacrylate] and [4-(11-methacryloylalkyloxy)-2,5-dimethylphenyl](4-nitrophenyl)diazene. The length of the two alkyl spacers has been set to be hexyl and undecyl for the C6-C11-DMNPAA and undecyl and hexyl for the C11-C6-DMNPAA. The chemical structure of these compounds is presented in figure 3. Glass transition temperatures (T_g) have been measured by DSC to be equal to 288 K for PVK:DMNPAA, 329 K for C6-C11-DMNPAA and 253 K for C11-C6-DMNPAA. Polymers and chromophore synthesis can be found elsewhere.¹⁵

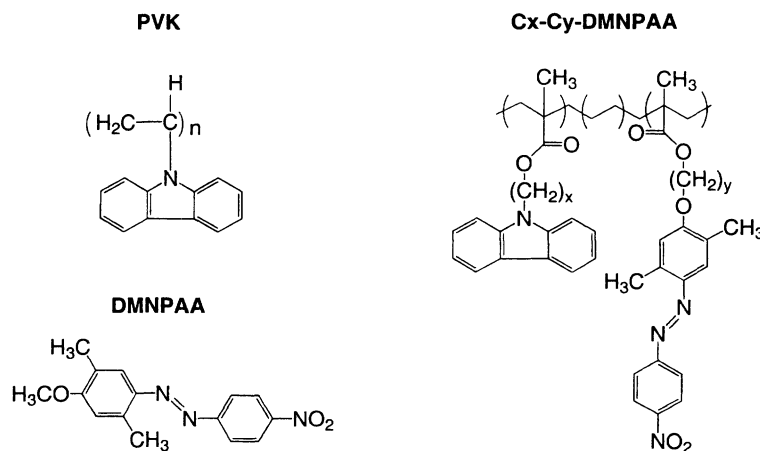


Figure 3. Chemical structure of the different compounds used, with their acronym.

3.2. Measurements

Four kinds of measurement have been performed: diffraction efficiency and photoinduced birefringence versus writing intensity and versus sample temperature. Set-up descriptions are schematically presented at figures 4. In brief, we use a 514 nm argon laser to induce orientation of the chromophores. Writing beams are linearly polarized. The reading beam come from an HeNe laser at 633 nm and is linearly polarized. For photoinduced birefringence (fig. 4(b)), the direction of the reading beam polarization is at 45° according to that of the writing beam. An analyzer is placed behind the sample in such a way that only the component polarized perpendicularly to the initial polarization is transmitted to the detector. For diffraction efficiency experiments (fig. 4(a)), the argon beams make a 5° angle which bisector is perpendicular to the sample. The HeNe beam is polarized according to the ones of the writing beams and it reaches the sample at Bragg angle. The reading intensity has been fixed low enough to prevent any molecular orientation to come from this beam. In experiments versus temperature, the writing intensity was set to 1 mW/cm^2 for photoinduced birefringence and 1.2 mW/cm^2 per beam for diffraction measurement.

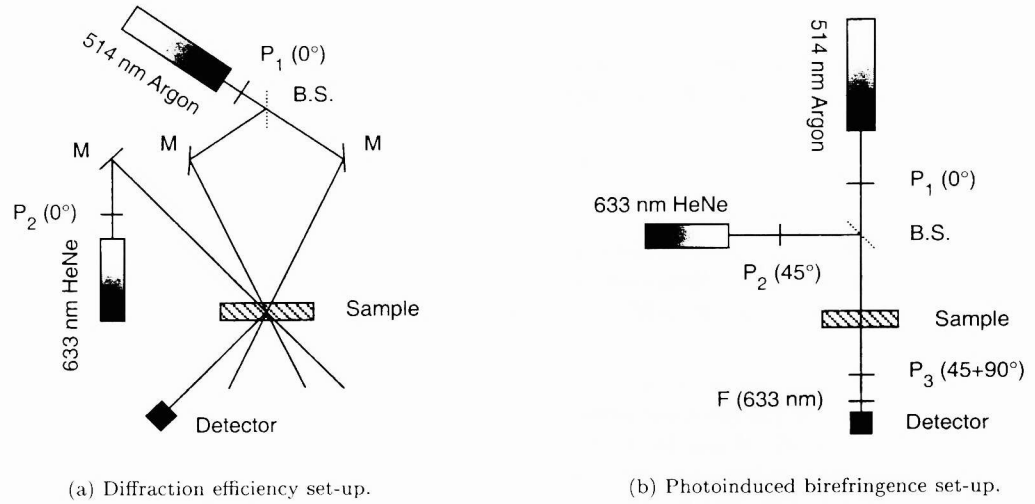


Figure 4. Experimental set-up. Legend: M: mirror, B.S.:beamsplitter, P: polarizer, F: transmission filter.

In each experiments, the diffracted or the transmitted intensity has been recorded according to the writing time. Efficiency data, defined as the ratio between output and incident HeNe intensity, have been fitted by the following equation:

$$\eta(t) = \sin^2 [A (1 - \exp(-t/\tau_A)) + B (1 - \exp(-t/\tau_B)) + \phi] . \quad (9)$$

In photoinduced experiments, the phase parameter ϕ is defined by the transmitted intensity before the writing beam was switched on. This come from the sample making process which aligns chromophores.⁵ Inversely, when measuring the diffraction efficiency, no light could be diffracted without writing beams. So, in this case, the phase parameter is always equal to zero.

In figures 5, we have plotted the maximum efficiency of the phenomenon. It is given by the efficiency at saturation minus the efficiency at $t = 0$.

$$\eta_{\max} = \sin^2 [A + B + \phi] - \frac{\phi}{|\phi|} \sin^2 (\phi) . \quad (10)$$

We can see in figures 5 that temperature and intensity have a considerable influence on the efficiency since there is variation over several orders of magnitude. There is also a large difference of behavior according to the polymer used. The C6-C11-DMNPAA has the greatest values and the C11-C6-DMNPAA shows the most important variation versus temperature. We will see further that the saturation of the efficiency according to the writing intensity is an artifact coming from the samples heating.

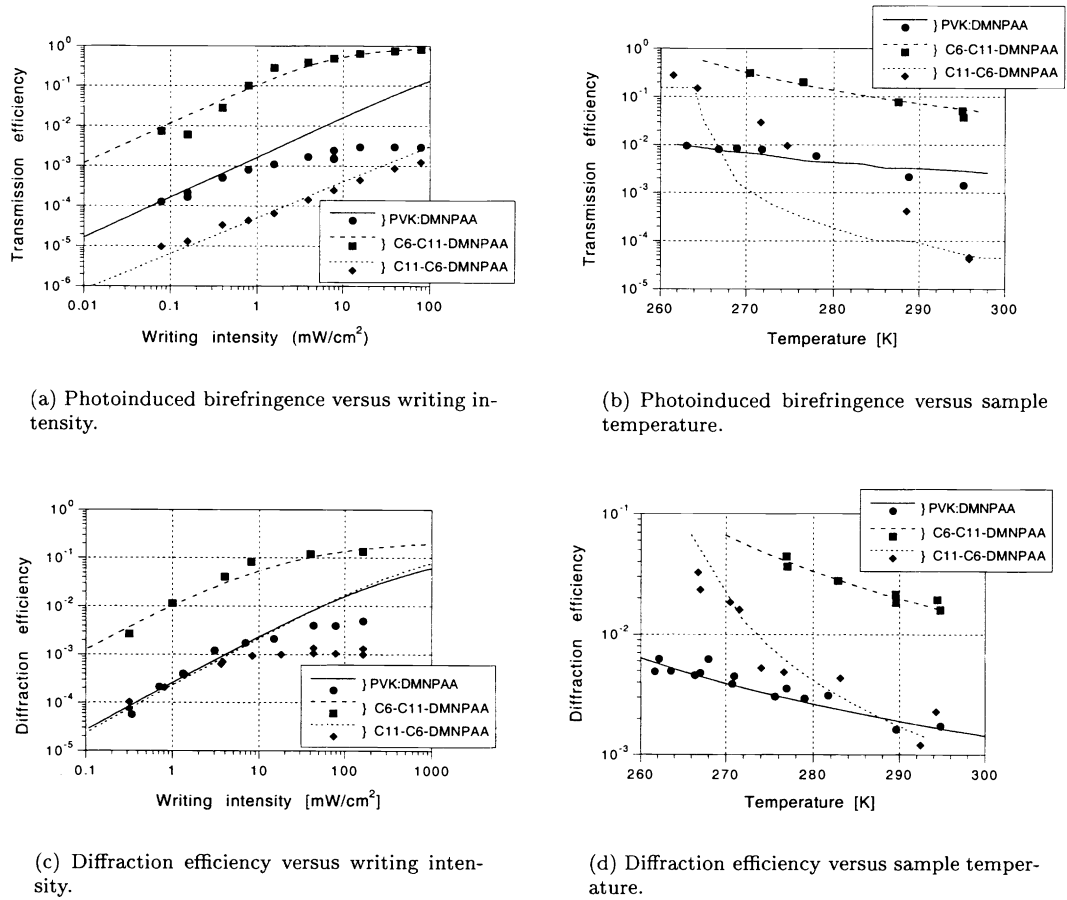


Figure 5. Behavior of the maximum photoinduced birefringence and diffraction efficiency according to the writing intensity and the sample temperature for three azo-dye polymers. Interpolations come from theoretical model.

It is also interesting to analyze the behavior of the sensitivity versus the writing intensity and the sample temperature (figures 6). Sensitivity is commonly related to the slope of the refractive index variation (itself proportional to the chromophore photo-orientation) at the origin time divided by the writing intensity. This is represented by the argument of the square sine of equation 9. Sensitivity is thus given by:

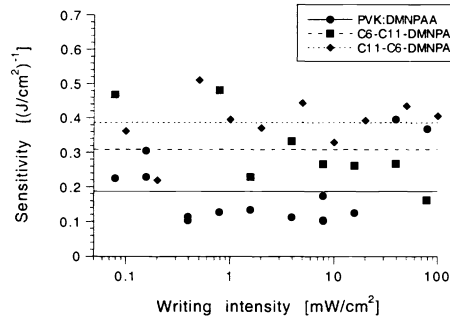
$$S = \frac{\frac{A}{\tau_A} + \frac{B}{\tau_B}}{I_{writing}}, \quad (11)$$

For holographic experiments, we have defined the writing intensity $I_{writing}$ as the maximum of the light intensity in the modulation pattern. According to our experimental conditions where the writing beams are balanced, $I_{writing}$ is so equal to four times the intensity of one argon beam. Since we are discussing about *orientation sensitivity* and not about that of diffraction, this definition seems more logical.

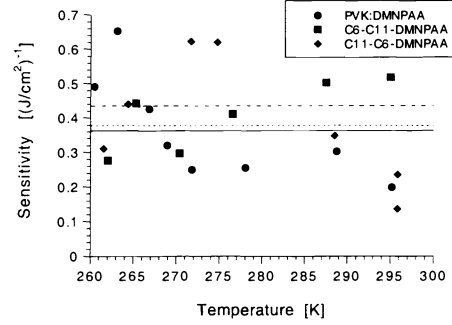
Theoretical models developed in the previous sections are static and could not give an account of the sensitivity. A dynamic theory of the photoinduced orientation can be found elsewhere.^{4,16} According to our measurements, no variation versus the writing intensity (fig. 6(a) and 6(c)), nor the sample temperature (fig. 6(b) and 6(d)), have to be notified. Moreover, the sensitivity is quite similar for the three compounds which have yet very different efficiencies.

4. DISCUSSION AND CONCLUSIONS

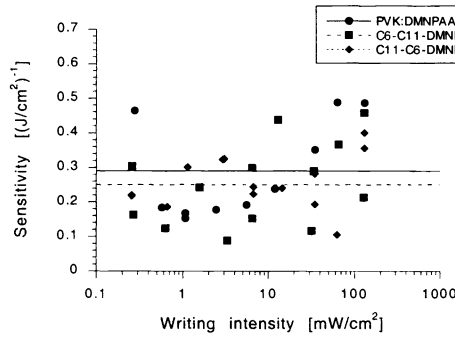
In figure 5, we have interpolated the experimental data with theoretical models. Only the parameters A and T_0 have been adjusted to fit data, all the remaining variables introduced in the model have been fixed according to sample



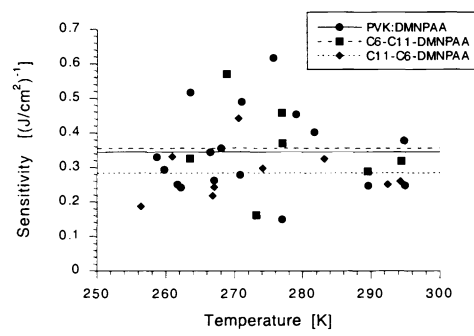
(a) Orientation sensitivity versus writing intensity. Birefringence measurement.



(b) Orientation sensitivity versus sample temperature. Birefringence measurement.



(c) Orientation sensitivity versus writing intensity. Diffraction measurement.



(d) Orientation sensitivity versus sample temperature. Diffraction measurement.

Figure 6. Behavior of the chromophores orientation sensitivity for three azo-dye polymers. Interpolation is a guide for the eyes and do not come from any model.

and set-up characteristics, i.e. absorption coefficient, sample thickness, writing intensity, . . . Since the chromophore used in all the compounds is the same, we have fixed the difference between the ordinary and extraordinary refractive indices to be 10^{-2} , which is a reasonable value.

We can see in figures 5(a) and 5(c) that, if at low intensity the values and the slope of the data is well represented, models do not give account of the diffraction efficiency at higher writing intensities. This is due to the heating of the sample by the writing beams which is not introduced in our models. This fact, measured experimentally, reveal that it is the temperature which limits the efficiency more than the intensity of the writing beams. This is emphasized by the measurements according to the temperature (figures 5(b) and 5(d)) which show the large dependance of the efficiency to this parameter.

By adjusting parameters A and T_0 in the diffraction efficiency model, we have fitted the very different behaviors according to the temperature of the three polymers studied. The values of these parameters are:

- PVK:DMNPAA, $A/k = 27 \text{ K (mW/cm}^2\text{)}^{-\frac{1}{2}}$ and $T_0 = 229 \text{ K}$;
- C6-C11-DMNPAA, $A/k = 75 \text{ K (mW/cm}^2\text{)}^{-\frac{1}{2}}$ and $T_0 = 260 \text{ K}$;
- C11-C6-DMNPAA, $A/k = 10 \text{ K (mW/cm}^2\text{)}^{-\frac{1}{2}}$ and $T_0 = 264 \text{ K}$.

For the PVK:DMNPAA and the C6-C11-DMNPAA, these values have been reused with success to fit the other curves: transmission and diffraction efficiency versus writing intensity and temperature. This proves the coherence between both models, the parameters can so be used to define the polymer and, with the data coming from one experiment, we can recover the behavior in the others.

In the case of the C11-C6-DMNPAA, we were not able to fit all the experiments with the same set of parameters. Indeed, the value of the parameter A/k has been reduced to $1 \text{ K (mW/cm}^2\text{)}^{-\frac{1}{2}}$ in order to interpolate photoinduced birefringence data. This means that, *in this compound*, diffraction efficiency cannot come only from the photoinduced birefringence. Due to the low Tg of this polymer, it should exist another writing mechanism that enhance the diffraction efficiency without inducing birefringence. Experiments are planned to determine the nature of the holographic grating(s) in this polymer. Moreover, we could see in figure 5(b) that models do not give an account of measurements: amplitude is underestimated and shape is different. We think this comes from the behavior of the parameter $A_{(T)}$ that should be different for this compound, but we presently have no reason to explain this discrepancy. In order to remedy to these problems, we are trying to find a more adequate function $A_{(T)}$ that is related to the internal physics of the phenomenon.

The measurement of the sensitivity (figures 6) has revealed that it is no longer influenced by the temperature nor the writing intensity. Since the sensitivity is quite the same for all the compounds studied here, this lets suppose that it is related to the chromophore rather than to the polymer matrix. This opens large perspectives for the optimization of the figures of merit that are efficiency and sensitivity. Indeed, the first one is influenced by the guest polymer and the second one by the host dye; so, we should be able to optimize both independently. Further experiments must be carried out in order to confirm this hypothesis.

In conclusion, we have developed theoretical models to give an account of the molecular photo-orientation according to the writing intensity and the sample temperature. Experiments performed on three different polymers have been fitted with these models and have given good accuracy for two compounds. Concerning the measurements according to the writing intensity, deviations between experimental data and model have been explained in term of temperature supplied by the laser beams. This implies that the limiting factor of the efficiency is essentially the sample temperature. In the point of view of the figures of merit, it has to be noted that by modifying the polymeric matrix and using the same chromophore, we have been able to increase the efficiency (in diffraction and transmission) by several magnitude orders without decreasing significantly the sensitivity.

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