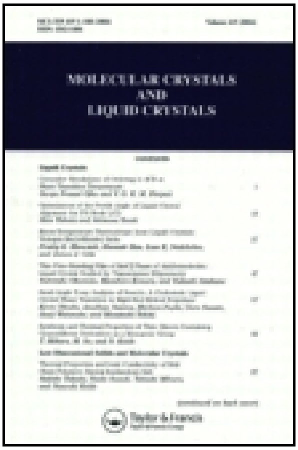

Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl20

Space-Charge Field Induced Refractive Index Modulation in Chiral Smectic A and C Mesophases: A Simple Model

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Version of record first published: 18 Oct 2010

Mol. Cryst. Liq. Cryst., Vol. 398, pp. 107–116, 2003

ISSN: 1542-1406 print/1563-5287 online
DOI: 10.1080/15421400390221484

SPACE-CHARGE FIELD INDUCED REFRACTIVE INDEX MODULATION IN CHIRAL SMECTIC A AND C MESOPHASES: A SIMPLE MODEL

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The photorefractive properties of chiral smectic A and C phases were studied using a model we had previously developed. The space-charge field induced reorientations considered are due to the electroclinic effect, in the case of the chiral smectic A, and to the reorientation of the spontaneous polarization for the chiral smectic C. The model takes into account both geometrical parameters, like for example light direction and polarization, and material parameters, like birefringence, spontaneous tilt angle or electroclinic coefficient. Results are extremely useful in order to optimize material parameters.

Keywords: photorefractivity; smectics; non-linear optics; liquid crystal

INTRODUCTION

The need of optical devices in the field of the information processing technologies has prompted the development of new materials with interesting optical properties and constantly improving performance. Among these, photorefractive materials are widely studied and characterized because of their peculiar properties. Photorefractivity is observed in materials where the space redistribution of photogenerated charges induces an electric field which, in turn, affects the refractive index. The photogeneration is usually obtained under conditions of non-uniform illumination. Mobile charges, under the effect of diffusion and electric fields, migrate macroscopically within the sample, giving rise to an internal space-charge electric field $\vec{E}_{sc}$, which is phase shifted with respect to the light pattern. If the refractive index of the material depends on the electric field, then $\vec{E}_{sc}$ will modulate the refractive index: the resulting hologram is a spatially shifted replica of the initial non-uniform illumination. The photorefractive effect was first observed in inorganic salts [1] but the experimental and synthetic difficulties encountered in optimizing the performance of single crystals, led researchers to synthesize and study photorefractive materials based on organic solids [2–4]. In organic media there are two different mechanisms that contribute to the field-dependence of the refractive index. One of them is the non-linear Pockels effect and the second one is birefringence. In this last case the electric dipoles on the cromophores are oriented by an electric field and such induced birefringence is at the origin of the modulation of the refractive index.

Because of their high and spontaneous birefringence, liquid crystals soon became extensively studied for applications in photorefractive devices. The first photorefractive liquid crystals were nematic mesophases [5–9], where the director reorientation is due to dielectric anisotropy. More recently, the first studies of photorefractive smectic mesophase have also appeared [10–12]. While in nematics the reorientation is an effect which is quadratic in the electric field, in smectics it is linear. In particular, in ferroelectric S_C* phases the reorientation is achieved through the field interaction with the spontaneous polarization, while in S_A* mesophases the director reorientation is due to the electroclinic effect.

In this paper we use a simple model that we recently developed [13] in order to illustrate the potential of chiral smectic phases as photorefractive materials. In the model, the director distribution in S_C* and S_A* mesophases under the influence of a sinusoidal electric field is first calculated. The refractive index pattern due to this director distribution is then obtained as a function of light direction and polarization and of sample orientation.

INDEX MODULATION IN S_A* PHASES

In this section we will first briefly summarize the main features of the model that we will use to calculate the refraction index modulation in photorefractive samples of S_A* mesophases [13]. We will then show and comment on the most important results. In the model we consider a space-charge field $\vec{E}_{sc}$ created within a photoconducting medium by two coherent overlapping interfering laser beams. $\vec{E}_{sc}$ can be described as:

$$\vec{E}_{sc} = E_{sc} cos \frac{2\pi\xi}{\Lambda} \hat{\xi} \quad (1)$$

where Λ is the spacing of the interference pattern along the coordinate ξ. In organic materials the photogeneration process usually requires the application of an external field $\vec{E}_0$ and, if E is the component of $\vec{E}_0$ in the grating direction, our model assumes that:

$$E_{sc} = kE \quad (2)$$

where and $k \leq 1$. The space-charge field will modulate $\vec{E}_0$ and, via the electroclinic effect, it will also modulate the refractive index. The model neglects the elastic energy associated with such reorientations, which is acceptable for small deformations over large Λ's or for dispersed systems. The model also considers the orientation of the smectic layers with respect to the interference pattern.

In Figure 1 we show a schematic illustration of sample orientation. The plane, which contains the Figure, is the p-plane, which also contains the laser beams and the sample normal $\hat{N}$, while θ is the angle between the two writing beams and β is the angle between the sample normal and the beams bisector. The smectic planes in the sample are considered to be homogeneously oriented so that the director $\hat{n}$, in the absence of fields, is perpendicular to the sample normal. We choose a set of axes so that the $\hat{x}$ axis is parallel to the normal $\hat{N}$, the $\hat{z}$ axis is perpendicular to the smectic layers, and then also parallel to the director, and the $\hat{y}$ axis is determined considering a right-handed system. The $\hat{z}$ axis, as illustrated in Figure 2, can be oriented at an angle γ with respect to the p-plane: a variation of γ corresponds to a rotation of the whole sample around $\hat{x}$.

Besides geometric parameters, the model also takes into account some material parameters. In particular, it is necessary to consider the electroclinic coefficient e_c and the ordinary (n_o)and extraordinary (n_e) refractive indices of the mesophase. In order to calculate the director orientation, we need to find the intensity of total field $\vec{E}_{TSL}$ within the smectic layers, given by:

$$E_{TSL} = E_0\left[\left(\pm 1 + k\sin^2\beta\cos\frac{2\pi\xi}{\Lambda}\right)^2 + k^2\sin^2\beta\cos^2\beta\sin^2\gamma\cos^2\frac{2\pi\xi}{\Lambda}\right]^{\frac{1}{2}} \quad (3)$$

with the $\pm$ sign depending on whether the direction of E_0 is along $+\hat{x}$ or $-\hat{x}$. We can also define the direction of $\vec{E}_{TSL}$, deriving the angle Ω between

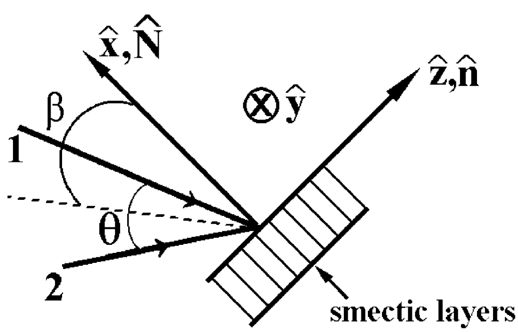

FIGURE 1 Illustration of sample and axes orientation. The incoming beams are labelled 1 and 2.

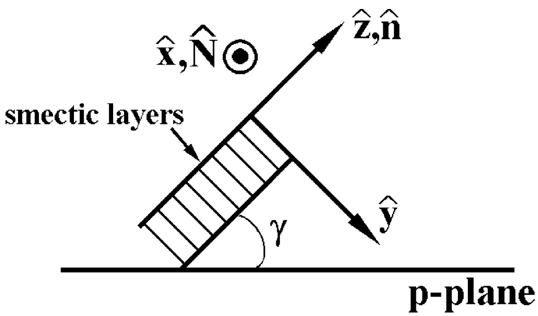

FIGURE 2 Different view of sample and axes orientation.

the $\hat{x}$ axis and the field:

$$tag\,\Omega = \frac{k\sin\beta\cos\beta\sin\gamma\cos\frac{2\pi\xi}{\Lambda}}{\pm 1 + k\sin^2\beta\cos\frac{2\pi\xi}{\Lambda}} \tag{4}$$

$\vec{E}_{TSL}$ will induce a rotation of the director, due to the electroclinic effect, by an angle χ_A:

$$\chi_A = e_c E_{TSL}$$
$$= e_c E_0\left[\left(\pm 1 + k\sin^2\beta\cos\frac{2\pi\xi}{\Lambda}\right)^2 + k^2\sin^2\beta\cos^2\beta\sin^2\gamma\cos^2\frac{2\pi\xi}{\Lambda}\right]^{\frac{1}{2}} \tag{5}$$

from which the components of the director can be obtained:

$$n_x = sin\chi_A\, sin\,\Omega \tag{6a}$$

$$n_y = -sin\chi_A\, cos\,\Omega \tag{6b}$$

$$n_z = cos\,\chi_A \tag{6c}$$

In order to calculate the refractive index modulation, we also need to consider the polarization of the incoming light. Defining α as the angle between the polarization versor $\hat{p}$ and the p-plane, then the components of $\hat{p}$ will be:

$$p_x = \cos\alpha\sin(\beta - \theta/2) \tag{7a}$$

$$p_y = -\sin\alpha\cos\gamma + \cos\alpha\sin\gamma\cos(\beta - \theta/2) \tag{7b}$$

$$p_z = \sin\alpha\sin\gamma + \cos\alpha\cos\gamma\cos(\beta - \theta/2) \tag{7c}$$

It is now possible to calculate the effective refractive index at any position along ξ as:

$$n_{eff} = \frac{n_o n_e}{\sqrt{n_e^2 \sin^2 \psi + n_o^2 \cos^2 \psi}} \tag{8}$$

where ψ is the angle between the polarization vector p̂ and the director $\hat{n}$ given by:

$$\cos \psi = p_x n_x + p_y n_y + e_z n_z \tag{9}$$

If we now substitute the expressions derived in Eqs. (6), (7) and (9) in Eq. (8), we obtain the following expression for n_{eff}:

$$n_{eff} = \frac{n_o n_e}{\left\{ n_e{}^2 + [\sin \chi_A (a + b - c + \cos \chi_A (d + e)]^2 (n_o{}^2 - n_e{}^2) \right\}^{\frac{1}{2}}} \tag{10}$$

where for brevity we indicated

$$\begin{aligned} a &= \sin \Omega \cos \alpha \sin(\beta - \theta/2) \\ b &= \cos \Omega \sin \alpha \cos \gamma \\ c &= \cos \Omega \cos \alpha \sin \gamma \cos(\beta - \theta/2) \\ d &= \sin \alpha \sin \gamma \\ e &= \cos \alpha \cos \gamma \cos(\beta - \theta/2) \end{aligned}$$

We can now use the values of χ_A and Ω from Eqs. (4,5) to obtain n_{eff}. If we do this twice, for ξ = 0 and for ξ = Λ/2, i.e. for the maximum and minimum values of E_{sc}, respectively, we can obtain the refractive index modulation $\Delta n = n_{eff}(\xi = 0) - n_{eff}(\xi = \Lambda/2)$.

To summarize, the refractive index modulation depends on the material parameters k, e_c, n_o and n_e and on the geometrical parameters β, θ, γ and α.

The angles β and θ have a restricted variation range, since they are tied to photogeneration efficiency and to grating spacing. The effects of the variation of the parameters α and γ are instead very interesting to study because by suitably changing these parameters we can maximize Δn, which is the difference between the effective refractive index n_{eff} calculated at the maximum and at the minimum value of $\vec{E}_{sc}$.

In Figure 3 we show that there is a large variation of Δn as a function of α and γ. It is interesting to note that Δn can be positive or negative, giving two different signs in two-beam coupling experiments, and that, for some values of α and γ, Δn vanishes.

Considering material parameters, both an increase of k and of the quantity $|n_o - n_e|$ obviously produce an increase of refractive index modulation, in the first case through an increase of the space-charge field

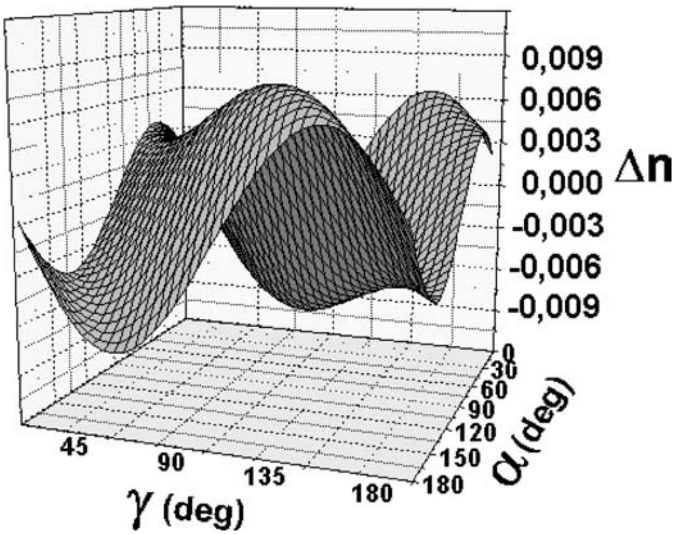

FIGURE 3 (see COLOR PLATE XXXVII) Modulation of the refractive index as a function of γ and α from Eq. 4. In the calculations we used $k = 0.7$, $\theta = 1°$, $\beta = 31°$, $n_e = 1.6$, $e_c = 0.19$ deg µm/V, $E_0 = 40$ V/µm, $n_o = 1.5$.

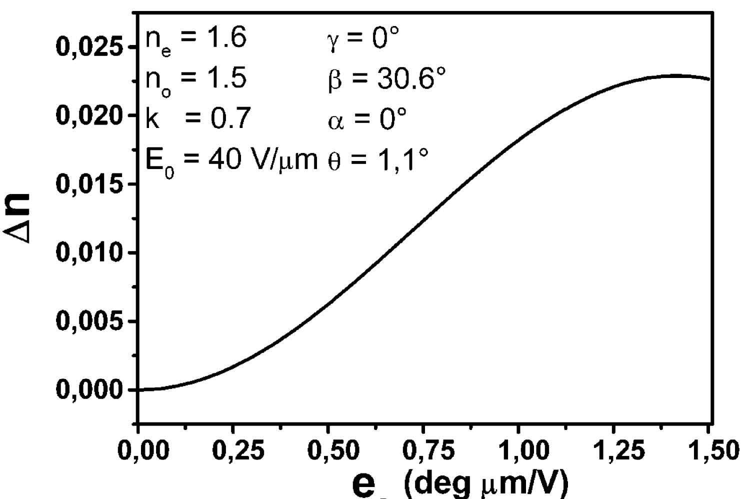

FIGURE 4 Modulation of the refractive index as a function of the electroclinic coefficient.

magnitude. It is more interesting to follow the influence of the electroclinic coefficient e_c on the refractive index modulation.

In Figure 4 we show the calculated values of Δn versus e_c and we can note that increasing e_c we obtain an increase of Δn. It is important to underline that all the parameters used in the simulation are realistic and that, at 40 V/µm, an electroclinic coefficient $e_c \sim 0.5$–1 deg µm/V, which is typical of most electroclinic materials, is sufficient to obtain a large index modulation.

INDEX MODULATION IN S_C* PHASES

The model can be easily extended to the S_C* phase by considering the interaction between the spontaneous polarization and the total electric field as the reorienting mechanism [13]. In order to calculate the refractive index modulation, it is still possible to use the equations obtained for the S_A* model, but replacing the field dependent tilt χ_A with a constant tilt χ_C. In this case the modulation of refractive index is entirely due to the different directions that the total electric field assumes along the grating and not, as in the S_A*, also to the modulation of the tilt angle.

In Figure 5 we illustrate, for a S_C* phase and for realistic values of the different parameters, the calculated dependence of Δn on light polarization

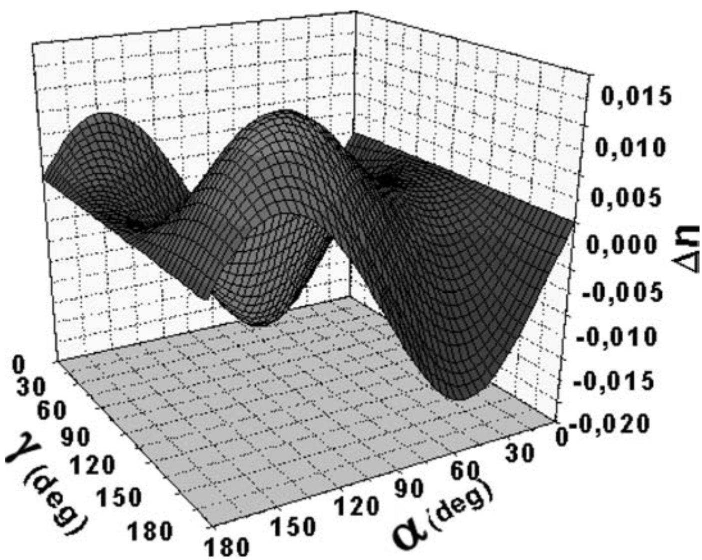

FIGURE 5 (see COLOR PLATE XXXVIII) Modulation of the refractive index in S_C* as a function of γ and α from Eq. 4. In the calculations we used $k = 0.7$, $E_0 = 40$ V/µm, $n_o = 1.5$, $n_e = 1.6$, $\beta = 31°$, $\theta = 1°$, $\chi_C = 19°$.

and sample orientation, respectively defined by α and γ. As is the case for the S_A* phase, the index modulation can change sign but the areas where Δn is very small are much wider for the S_C* phase, since the Δn surface is less steep around $\Delta n = 0$. It is also interesting to see the results of model in the case of a change of sign of the applied field. In traditional photorefractive crystals and polymers, the only consequence would be a change of sign in Δn. In liquid crystals this may not hold true, since the reorientation mechanisms are more complex when compared to simple dipolar reorientations.

In Figure 6 we show the results for Δn obtained using the same parameters as in Figure 5, but with an applied field of different sign. As can be seen comparing the two Figures, the main effect is still basically a change of sign of Δn, although other second order differences are present.

Another important check that is possible to perform with the model is the dependence of Δn on tilt angle. In Figure 7 we illustrate the dependence of the maximum possible absolute value of Δn versus the tilt angle χ_C, obtained from data as the ones in Figure 6. We can see how increasing χ_C we obtain an increase in Δn. This parallels what observed for an increase of Δn with e_c in S_A* mesophases (see Fig. 4), but this time the dependence

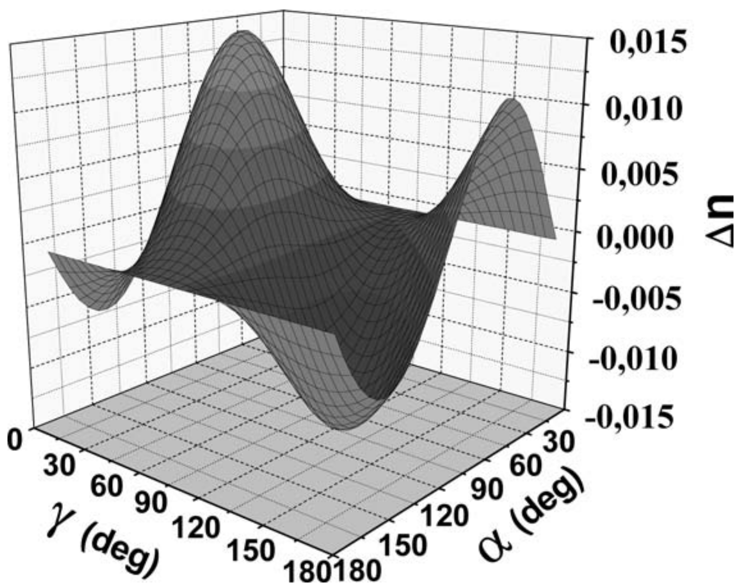

FIGURE 6 (see COLOR PLATE XXXIX) Modulation of the refractive index in S_C* as a function of γ and α from Eq. 4. In the calculations we used $k = 0.7$, $\chi_C = 19°$, $E_0 = -40\,V/\mu m$, $n_o = 1.5$, $n_e = 1.6$, $\beta = 31°$, $\theta = 1°$.

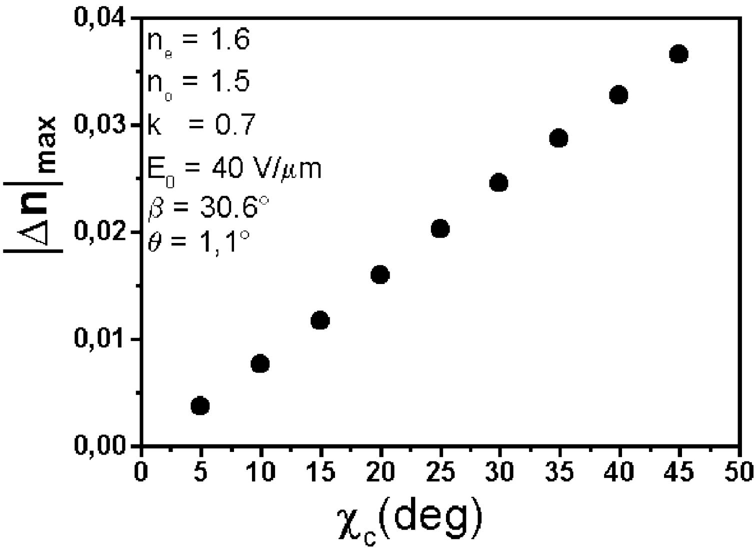

FIGURE 7 Maxima of Δn as function of spontaneous tilt angle.

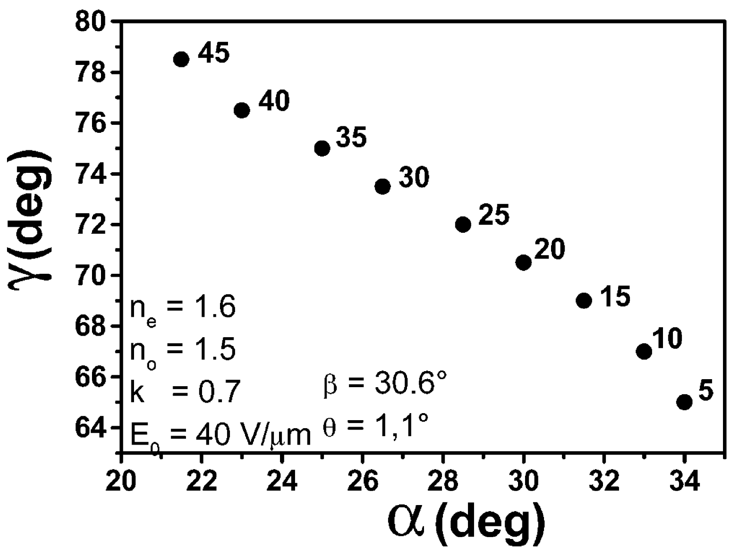

FIGURE 8 Values of α and γ corresponding to Δn_{max} for different values of χ_C. Near each point we report the value of χ_C (in degrees) used in the calculations.

is almost linear. In Figure 8 we show the values of α and γ where we find the maximum absolute values of Δn for different values of χ_C. We can see how both light polarization and sample orientation for maximum performance do not change appreciably. This can be extremely important in selecting the correct values of α and γ in order to obtain the best efficiencies.

CONCLUSIONS

In this paper we presented results obtained from a simple model which derives the refractive index modulation in S_A* and in S_C* phases in the presence of an applied and a sinusoidal electric field, a situation typical of photorefractive materials. In the model, the refractive index can vary depending both on geometrical and on material parameters. For S_A* phases the considered field induced reorienting mechanism is the electroclinic effect while for S_C* phases it is the spontaneous polarization reorientation. We presented results showing how Δn varies as a function of different geometrical and material parameters.

REFERENCES

[1] Ashkin, A., Boyd, G. D., Dziedzic, J. M., Smith, R. G., Ballman, A. A., Levinstein, J. J., & Nassau, K. (1966). ***Appl. Phys. Lett.***, ***9***, 72.
[2] Sutter, K. & Günther, P. J. (1990). ***Opt. Soc. Am. B***, ***7***, 2274.
[3] Ducharme, S., Scott, J. C., Twieg, R. J., & Moerner, W. E. (1991). ***Phys. Rev. Lett.***, ***66***, 1846.
[4] Meerholz, K., Volodin, B. L., Sandalphon, Kippelen, B., & Peyghambarian, N. (1994). ***Nature***, ***371***, 497.
[5] Rudenko, E. V. & Sukhov, A. V. (1994). ***JETP Lett.***, ***59***, 142.
[6] Khoo, C., Li, H., & Liang, Y. (1994). ***Opt. Lett.***, ***19***, 1723.
[7] Wiederrecht, G. P., Yoon, B. A., & Wasielewski, M. R. (1995). ***Science***, ***270***, 1794.
[8] Ono, H. & Kawatsuki, N. (1997). ***Opt. Lett.***, ***22***, 1144.
[9] Golemme, A., Kippelen, B., & Peyghambarian, N. (1998). ***Appl. Phys. Lett.***, ***73***, 2408.
[10] Wiederrecht, G. P., Yoon, B. A., & Wasielewski, M. R. (2000). ***Adv. Mater.***, ***12***, 1533.
[11] Termine, R., De Simone, B. C., & Golemme, A. (2001). ***Appl. Phys. Lett.***, ***78***, 688.
[12] Sasaki, T., Kino, Y., Shibata, M., Katsuragi, A., Ishikawa, Y., & Yoahimi, T. (2001). ***Appl. Phys. Lett.***, ***78***, 4112.
[13] Termine, R. & Golemme, A. (2002). ***J. Phys. Chem. B***, ***106***, 4105.