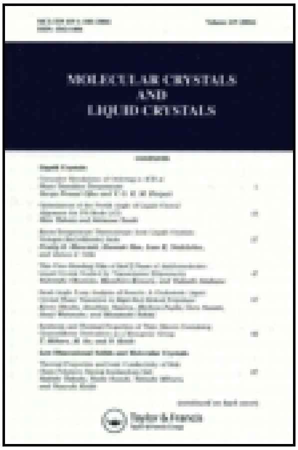

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Optical Phase Conjugation and Wavefront Correction by Thin Nematic Liquid Crystal Cells

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We report a study of optical phase conjugation in thin nematic cells by using low intensity of the incident beams and without any external applied field. The reported effect exploits the colossal optical nonlinearity recently observed in thin nematic liquid crystals films doped with a small amount of methyl red. The high conjugated reflectivity values obtained with the analysed samples, allows successful correction of severely aberrated wavefronts of very weak light beams.

INTRODUCTION

Molecular reorientation is responsible for the nonlinear optical response of many liquids made by anisotropic molecules. Among them, liquid crystals (LCs) have the peculiar feature of keeping the anisotropy on a macroscopic scale due to the long-range molecular orientational order. For this reason they exhibit a collective behaviour that enhances the molecular reorientation and leads to the well known Giant Optical Nonlinearity (GON) [1].

After the first observations of GON, several effects leading to further enhancements of LCs optical nonlinearity, have been reported as, for instance, the Jannossy effect [2], the reorientation driven by azo-dye photoisomerization [3] or the photorefractive-like effect [4].

More recently, an extraordinarily large nonlinear response has been observed in Methyl-Red (MR) doped nematics with homeotropic

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alignment [5]. After the first paper by Khoo and co-workers reporting a nonlinear coefficient up to $6\,cm^2/W$, other experiments, including some performed by the authors of the present manuscript, confirmed such a "supranonlinear" response. In particular, by acting on the sample pre-alignment and thickness, a colossal nonlinear coefficient overcoming $10^3\,cm^2/W$ has been recently observed in these materials [6].

Since a light-induced photovoltage is usually associated to light absorption in these compounds, a tentative explanation for the huge optical nonlinearity was to consider it as a sort of photorefractive-like phenomenon originated by the internal photo-induced electric field which acts as the external dc field of the Carr-Helfrich effect [7]. However, the low value of the photo-voltage ($\approx$10 mV) cannot account in a simple way for the huge nonlinear response observed; moreover the effect has been reported to be local [7].

The MR photo-isomerisation has also been indicated as the effect on the basis of the phenomenon [8]. Photo-isomerization of the azo-dye is certainly involved in the process, but it does not appear to be the main effect leading to the observed nonlinearity. In fact the reorientation driven by photo-isomerisation has the very distinctive feature to occur always outwards the linear polarization of the exciting beam, and this is not the case for the effect described.

A different interpretation has been recently suggested by the authors of the present manuscript claiming that the origin of the effect is the light-induced modification of the surface conditions, which in turn gives rise to a bulk reorientation through the LC elasticity. This phenomenon named as SINE (Surface Induced Nonlinear Effect) can occur without a direct optical or electric torque on the director in the bulk [9,10].

The question with the huge optical nonlinearity of MR-doped nematics is whether or not the phenomenon can be used for applicative purposes. As we have recently shown, the answer is positive and samples showing the colossal optical nonlinear response can be used in Optical Phase Conjugation (OPC) experiments and in the correction of severely aberrated wavefront of weak light beams [11].

We report in this manuscript details on OPC and Wavefront Correction (WFC) experiments performed on different kind of cells. The effects of thickness, surface prealignment and doping on the detected signal are discussed on the light of the SINE effect. A brief qualitative description of this latter is also given in the next section.

Surface Induced Nonlinear Effect (SINE)

As it is well established, in MR-doped nematics it is possible to modify the anchoring conditions by "writing" an easy axis that affects the sample producing a new steady orientation. The formation of such an easy axis can be explained by taking into account the adsorption and desorption of dye molecules at the cell surface irradiated by the incident light [12,13]. The basic idea of SINE is that these light-induced processes are already active before a new permanent orientation sets in in the sample.

According to our interpretation the easy axis can be accounted for by considering that, after the cell filling, a layer of dark-adsorbed dye molecules grows over the boundary surfaces, as demonstrated by previous experiments on the same materials [13]. This layer is expected to be anisotropic in the direction of filling. The subsequent cell irradiation with polarised light, affects the dark-adsorbed layer in a way that depends on the polarisation direction. If the incident light is linearly polarised parallel to the filling direction, the majority of the adsorbed dye molecules absorbs the radiation and desorb from the irradiated surface. In these conditions the main effect of pump irradiation is the depletion of the dark-adsorbed layer in a direction parallel to the pump polarisation, which produces a LC reorientation outward this latter direction. As soon as pump irradiation stops, the "motor" responsible for the depletion disappears and the system relaxes to the condition of dynamic equilibrium existing before irradiation. If the incident light is linearly polarised perpendicular to the filling direction, the initial situation is different. Now desorbtion is not the favourite effect since there are only few dye molecules able to absorb the incident light and to desorb from the surface. The main effect is thus the light-induced adsorbtion of the few molecules close to the irradiated surface, whose dipole moment has a non-vanishing component along the optical field. This produces an increase of adsorbed dye molecules parallel to the pump polarisation, which gives rise to a LC reorientation toward this direction. When irradiation stops, the system relaxes again to the pre-existing condition. This is true as long as the pump intensity or the exposure time are small enough to avoid memory effects. In this way, in the homeotropic configuration, dye adsorption can give rise to both transient and permanent modifications of the surface conditions in the azimuthal plane, namely a transient or permanent increase of the azimuthal anchoring energy. As discussed in more detail elsewhere [9], this effect produces a weakening of the homeotropic anchoring that leads to an increase of the pre-tilt angle describing the actual director orientation on the irradiated surface.

The generation of an easy axis in the azimuthal plane changes the surface conditions, thus making unstable the initial alignment. In this way a new director orientation is established through the cell due to the elasticity of the medium. Within this scheme it is easy to explain the observed nonlinear optical behaviour. Since the bulk orientation of a liquid crystal is strongly affected by the anchoring conditions, any change of these induced by a lightwave will produce a bulk reorientation. This reorientation affects the light propagation itself with the consequent onset of a nonlinear optical response. Moreover, since only a thin layer near the surface has to be excited in order to get the elastic reorientation of the whole sample, a very low intensity will be required to induce the nonlinear response.

EXPERIMENTAL DETAILS

The samples used are 1 μm and 3 μm cells containing a mixture of the nematic pentyl-cyano-biphenyl and of the azo dye Methyl Red (MR). Two different dye weight concentrations have been analysed: 0.1% and 1%. Homeotropic alignment of the LC cells has been obtained by using DMOAP as aligning agent. Samples with both the surfaces covered by DMOAP and with a single surface coated, have been analysed.

The experimental set-up (Fig. 1) is a standard one for degenerate four-wave mixing experiments. The used source, a frequency doubled continuous wave Nd:YVO_4 laser, is splitted into three beams of about 3 mm in diameter. Two of them, the pump beams, have equal intensity and are counterpropagating with respect to the cell; the third one, the signal beam indicated by I_3, is six times weaker than the others and crosses I_1 at an angle of 0.02 rad. The signal beam power is of submilliwatt level. The back reflection of the sample toward the direction of observation is eliminated by allowing a very small tilt of the cell. This does not affect the magnitude of the detected signal. No external field is applied.

RESULTS AND DISCUSSION

The OPC reflectivity R, defined as the ratio I_4/I_3, easily reaches 1.5-2% for a signal intensity I_3 of 8.6 mW/cm^2, as shown in Figure 2

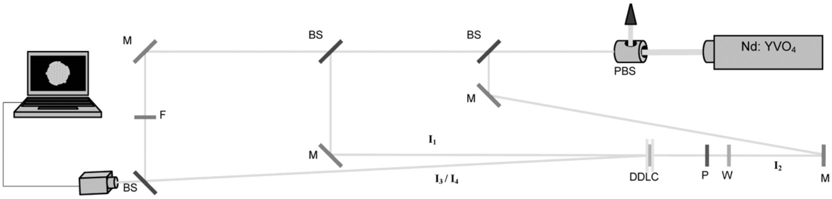

FIGURE 1 Experimental set-up. (See COLOR PLATE XLV)

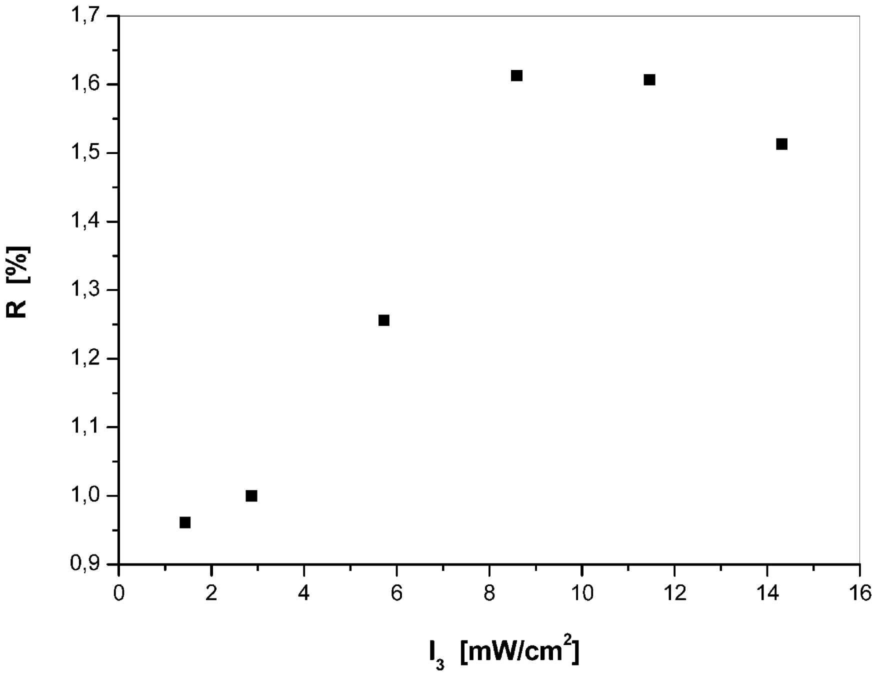

FIGURE 2 Conjugated reflectivity R vs. signal beam intensity I_3 obtained at room temperature with a 1 μm thick cell with only one surface coated by DMOAP. The dye concentration is 1%.

reporting the typical behaviour of R as a function of I_3. Note that a signal wave of 1.7 mW/cm^2 gives rise to about 1% of reflectivity. In these conditions the conjugated wave has approximately the same divergence and spatial quality of the signal wave.

The characteristic times of build-up and decay of the conjugated signal can be evaluated by blocking and opening the light beams incident on the sample (the conjugated signal is expected to disappear when any of the beams, I_1, I_2 or I_3 is blocked). Figure 3 reports the situation corresponding to shutting and opening I_3 with a frequency of 0.04 Hz. The build-up of the signal is instantaneous while the relaxation takes a few seconds increasing with the irradiation time from 3 to 10 s. A similar behaviour of I_4 has been observed shutting and opening the two pump beams. In this case the relaxation is faster, especially shutting I_2. The build up and decay times are strongly dependent on the prealignment conditions. In samples with both the surfaces treated to get strong homeotropic anchoring, they are two or even three times

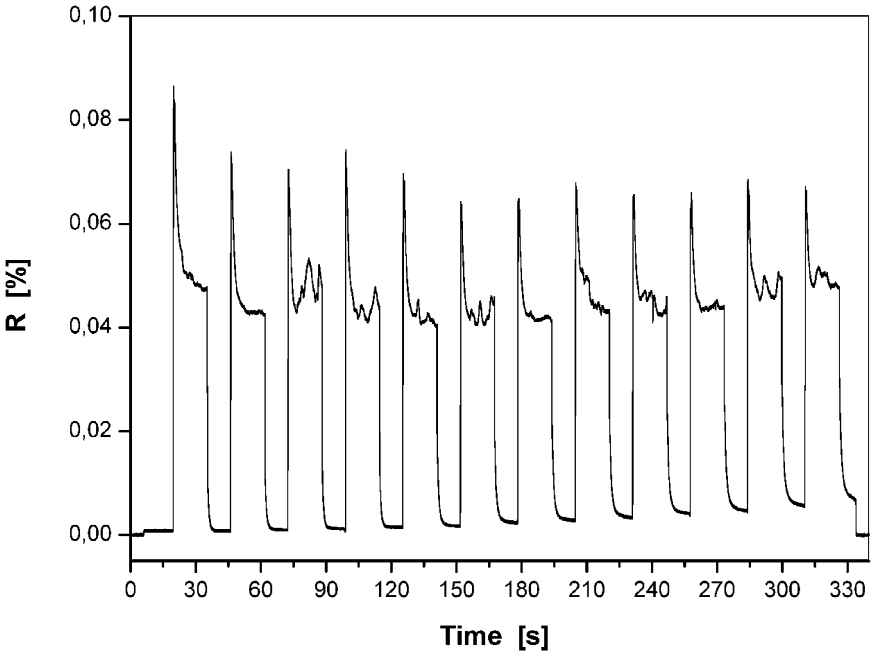

FIGURE 3 Conjugated reflectivity R vs. time recorded while shutting and opening the signal light beam I_3.

slower than those measured in samples with one surface left without surfactant.

A dramatic increase of the conjugated signal is observed for temperature values just below the nematic-isotropic transition temperature $T_{N/I}$. This phenomenon could be connected with the decrease of the order parameter due to temperature increase and to consequent modification of the dielectric anisotropy and of the elastic constants of the LC. This hypothesis is supported also by the lowering of the threshold power necessary to get the nonlinear effect, similar to the behaviour observed in the optical Fredericks transition [14]. The pre-transitional change of correlation strength could also play some role. A systematic study of the effects of temperature in optical phase conjugation in thin samples, is currently under way.

Preliminary measurements seem to indicate that the build-up and decay times of the conjugated signal do not depend on temperature.

Figure 4 shows the conjugated reflectivity versus the signal beam intensity I_3 for 1 μm thick cell. The maximum R values are one order of magnitude higher than those reported in Figure 2 obtained at room

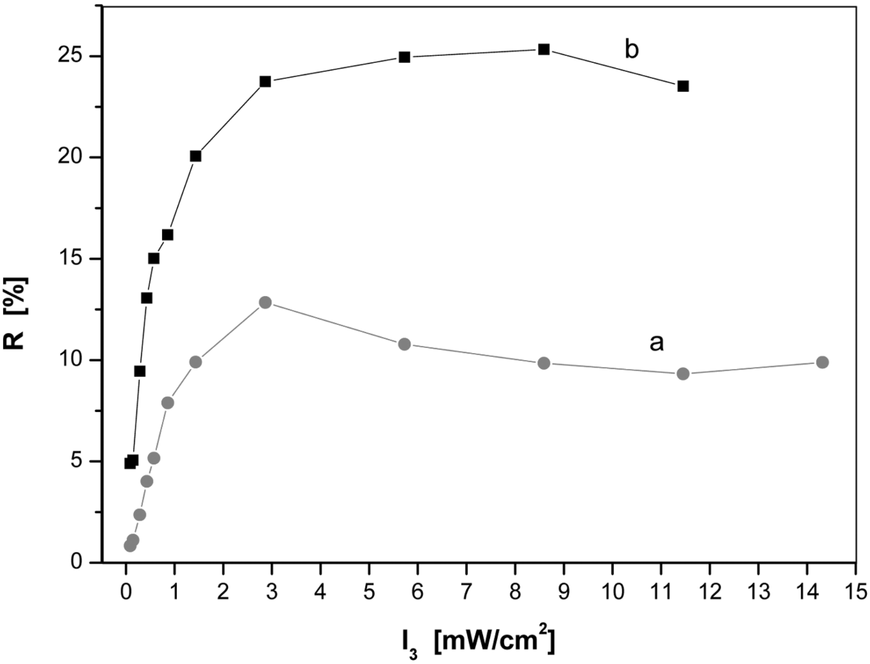

FIGURE 4 Typical conjugated reflectivity R vs. signal beam intensity I_3 for 1 μm thick cells with 1% MR weight concentration. Curve *a* and *b* refers to samples obtained by covering with DMOAP both the substrates and just one substrate, respectively. The cell temperature is just below the phase transition temperature $T_{N/I}$, optically monitored. (See COLOR PLATE XLVI)

temperature, the rise is steeper and the sensitivity is higher (a reflectivity of about 10% can be reached with less than 0.35 mW/cm^2 signal intensity (see in particular curve *b*)). The typical shape of the temperature curve is reported in Figure 5. As it can be seen, R rises to a peak and then decays to zero when T overcomes $T_{N/I}$. The dynamics is slow and the rise and decay times are of the order of seconds.

The two curves *a* and *b* in Figure 4 describe R vs I_3 for a cell doped by 1% of MR and obtained by covering with DMOAP both the substrates (curve *a*) and only one substrate (curve *b*). It is evident that when only one substrate is treated with the aligning agent, the efficiency of the phase conjugation effect increases, in agreement to what expected according with the surface-induced reorientation. The difference is not so evident for 3 μm thick cells. In this case samples with two or only one substrate coated by DMOAP show approximately the same maximum conjugated reflectivity, as shown in Figure 6.

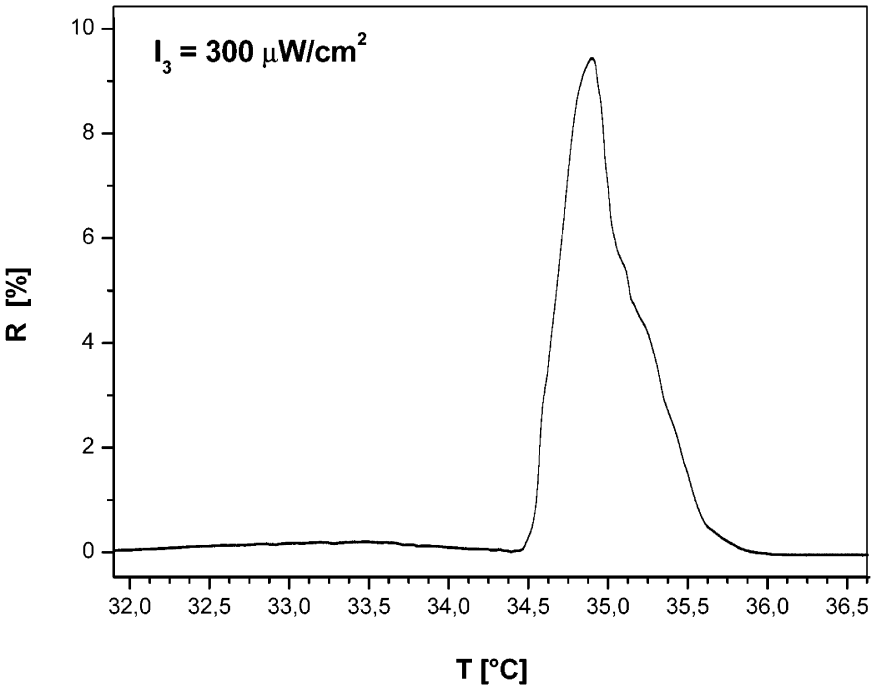

FIGURE 5 Typical shape of the R vs. time curve during cell heating.

The weak anchoring of the untreated surface, does not produce an increase of the efficiency. It is also worth noting that the higher thickness does not give rise to a higher conjugated signal.

The effect of MR concentration is showed in Figures 7 and 8, where the comparison between the R vs I_3 curves obtained by doping the cells with 1% and 0.1% of MR, is reported. The samples, 1 and 3 μm cells, respectively, have only one substrate coated by the surfactant. Once again, the difference is stronger in case of thinner cells, for which the decrease of the dye concentration by one order of magnitude brings to a sensitive drop of the conjugated reflectivity. For 3 μm thick cells, the decrease of the dye content reduces the signal only by about one half.

To summarise, 1 μm cells give the best results when doped with 1% of dye and when one substrate offers weak anchoring to the LC molecules. On the other hand, the thicker cells are not so sensitive to the prealignment conditions nor to the dye concentration, giving high reflectivity values also with 0.1% of MR. These observations point out the important role of prealignment and dye doping in the high nonlinearity of thin samples. In 3 μm cells the interaction length of the

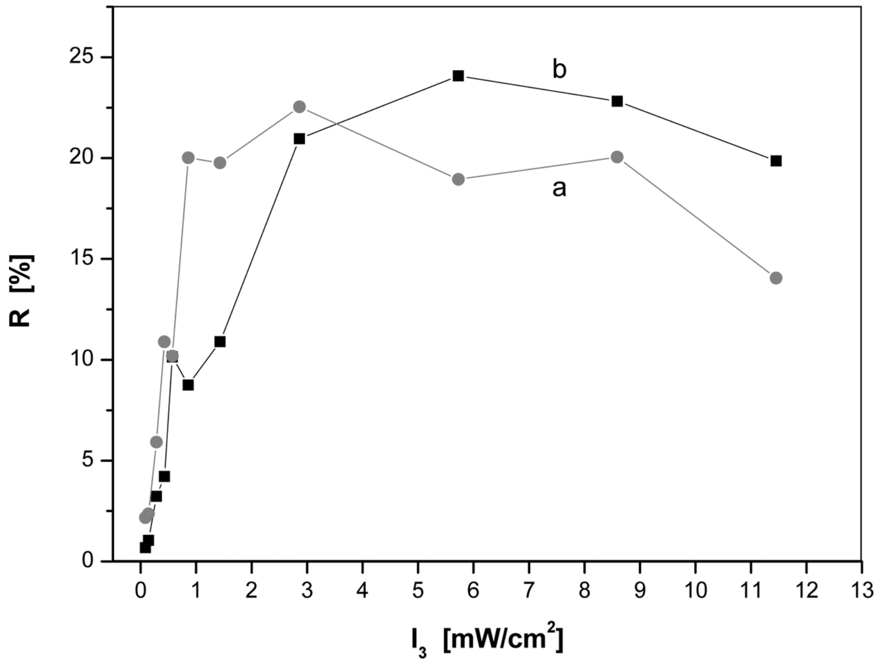

FIGURE 6 Typical conjugated reflectivity R vs. signal beam intensity I_3 for 3 μm thick cells with 1% MR weight concentration. Curve *a* and *b* refers to samples obtained by covering with DMOAP both the substrates and just one substrate, respectively. The cell temperature is just below the phase transition temperature $T_{N/I}$, optically monitored. (See COLOR PLATE XLVII)

waves inside the sample is higher than in thinner ones and this produces a stronger conjugated signal for a given incident intensity, prealignment condition and dye content. For this reason the reflectivity values observed with thicker cells are higher than those obtained with thinner ones in case of low dye concentration and strong anchoring.

Optical phase conjugation has been exploited for wavefront correction of weak light beams. An aberrating medium is inserted in the common path of the signal and the conjugated waves. In these conditions, the signal wavefront emerges distorted from the aberrating medium and impinges on the LC cell. Due to the phase-reversal properties of the OPC process, one should expect the wavefront of the conjugated wave to be corrected from the aberration after passing back through the aberrator. Experiments of this kind have been performed on each of the analysed cells. A typical result, obtained with a 3 μm cell with a single surface coated by DMOAP, is reported in

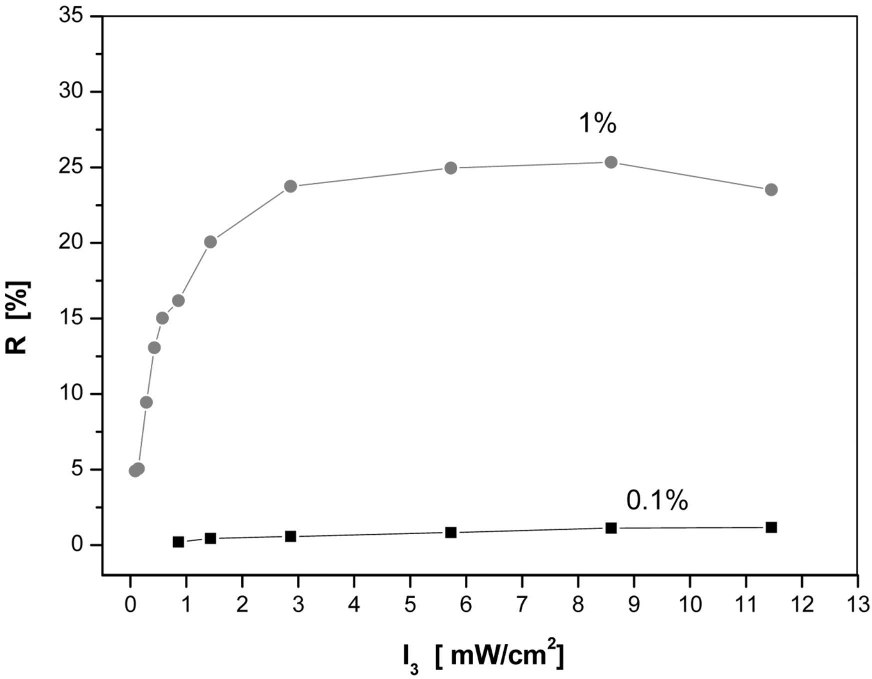

FIGURE 7 Typical conjugated reflectivity R vs. signal beam intensity I_3 for 1 μm thick cells with only one substrate coated by DMOAP. The two curves correspond to two different dye concentration: 0.1% and 1%. The cell temperature is just below the phase transition temperature $T_{N/I}$, optically monitored. (See COLOR PLATE XLVIII)

Figure 9. The undistorted signal beam, the aberrated beam and the conjugated one are showed. This latter is perfectly corrected from the aberration. The signal beam intensity I_3 was 3.5 mW/cm^2.

CONCLUSIONS

In conclusion, we have shown the possibility of obtaining high conjugated reflectivity values in thin nematic LC cells at low total incident intensity and without any external applied field. The study of the effects of the sample thickness, prealignment and dye content indicates that the main responsible for the observed conjugated reflectivity is the colossal optical nonlinearity of thin nematics due to the SINE effect. This is also confirmed by the evaluation of the light-induced average birefringence reported elsewhere [11], that is in agreement with that typical of the colossal optical nonlinearity recently observed in thin LC cells.

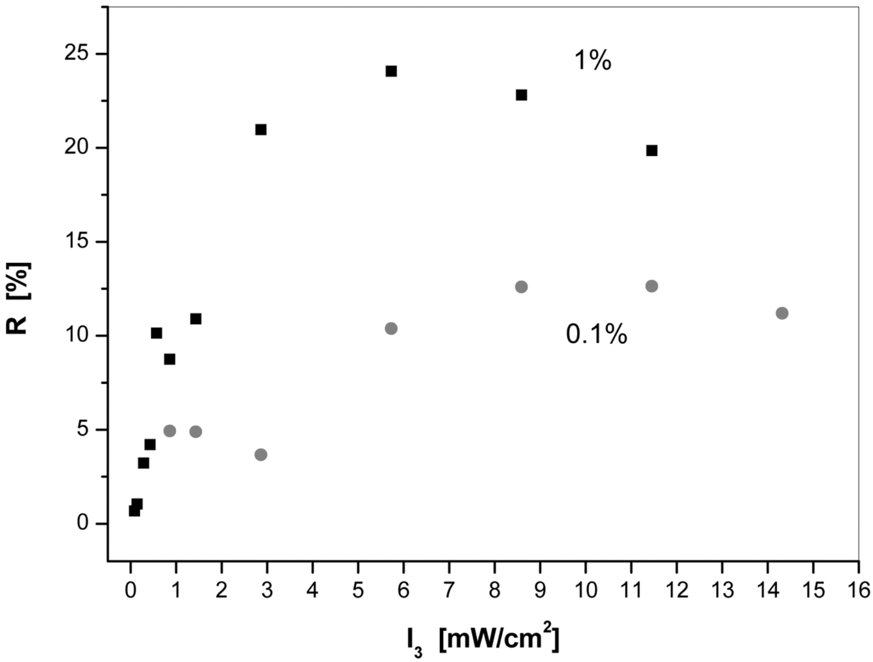

FIGURE 8 Typical conjugated reflectivity R vs. signal beam intensity I_3 for 3 μm thick cells with only one substrate coated by DMOAP. The two curves correspond to two different dye concentration: 0.1% and 1%. The cell temperature is just below the phase transition temperature $T_{N/I}$, optically monitored. (See COLOR PLATE XLIX)

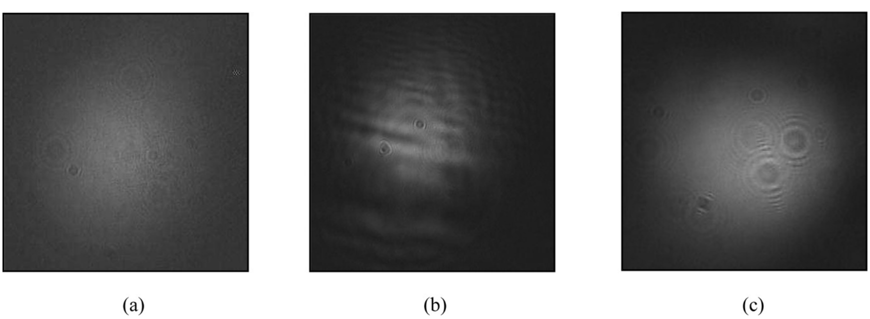

FIGURE 9 CCD images of the signal beam (a), the aberrated signal beam (b) and the conjugated beam corrected from the aberration (c). (See COLOR PLATE L)

The high conjugated reflectivity values obtained with the analysed samples, allow successful correction of severely aberrated wavefronts of low intensity light beams. This is a clear demonstration that highly nonlinear LC thin films can be successfully used for image processing operations.

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