AC FIELD ENHANCEMENT OF DIFFRACTION FROM PERMANENT GRATINGS IN DYE-DOPED LIQUID CRYSTALS

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Permanent gratings can be written in doped liquid crystals with high dye concentration without any externally applied electric field, using low-intensity visible light. The gratings are adaptive as their diffraction efficiency can be easily controlled by an AC field. The diffracted intensity could also be modulated by a low-frequency electric field with the magnitude of modulation decreasing for higher frequencies. The permanent gratings are durable, remaining in the cells for over a year, even after applying high temperatures.

Introduction

Research into photorefractive-like gratings in nematic liquid crystals has opened up new possibilities for their applications in photonics. Photorefractive transient gratings [1, 2] can be an order of magnitude more efficient than those in solid-state photorefractive materials. Liquid crystals with dispersed polymers [3], doped with dyes [2], fullerenes [4, 5] or with added photoconducting polymer layers [6] show, for example, high two-beam coupling coefficients. Permanent gratings [7—9] are also interesting for applications in holographic storage [10—12].

There is typically more than one mechanism involved [13, 14] in a reorientation of liquid crystal molecules: optical and dye-induced torques, a photorefractive space-charge field and photoisomerization [15]. The reorientation is also determined by surface effects [7, 16, 17]. For example, the adsorption of phototransformed dye molecules onto the polymer surface can give rise to a permanent alignment of molecules [16]. In the case of high dye concentrations, this effect of phototransformation of dye molecules and their adsorption on the cell surfaces can significantly contribute to the final orientation of liquid crystal molecules. Moreover, the surface-mediated photorefractive effect, with its electric charges trapped at the liquid crystal and alignment layer interface, can also provide a strong additional contribution [18].

By doping a polymer layer with dye, gratings can be also written directly into that layer [8, 9]. The diffraction efficiency of such doped-polymer-mediated gratings can be varied (but not enhanced) by application of an AC/DC field [9]. Patterns [1, 19], written in cells without special coating of the cell surfaces, were quasipermanent, namely decayed over a period of a few hours [2].

Doping liquid crystals with dyes also increase their light sensitivity and lower the intensity threshold for a molecular reorientation [17, 20]. The dye-induced torque can, in fact, be over two orders of magnitude stronger than a pure optical torque [20].

In our investigations, we concentrated on liquid crystals with high dye concentration and, in particular, on permanent reorientational patterns that can be written in these materials with light, in the absence of any electric field. These permanent patterns dominate over transient and photorefractive gratings. We tested their durability and adaptivity, as well as their temperature and electric field dependence.

1. Experimental Setup and Results

The liquid crystal we used was the mixture of a nematic liquid crystal K15 (5CB) and Methyl Red dye. The concentration of the dye in our cells was high and can be estimated as above 2%. Initial homeotropic alignment was achieved by treatment of the cell windows with a lecithine solution. This was the only layer added to the cell windows, apart from the standard transparent ITO electrodes.

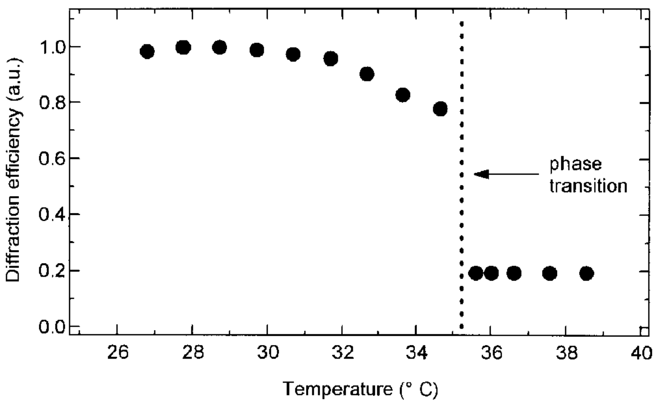

Fig. 1. Intensity of a diffracted beam from a permanent grating and its dependence on temperature

Holographic gratings were written using the conventional two-wave mixing technique with two linearly p-polarized λ=488 nm beams of equal intensity. The angle of incidence of both beams was approximately 30°, and the angle between them was small, in order to achieve grating spacings between 10 and 11 μm. No external voltage was applied during the writing process. The diffraction efficiency was monitored by a λ=633 nm, p-polarized, probe beam incident along the normal to the cell's surfaces.

Permanent gratings could be recorded with modest writing beam intensities, namely with as low as 4 mW/cm^2, provided the dye concentration in a liquid crystal was sufficiently high. The gratings were truly permanent as they remained in samples for over a year. In the first stage of illumination, a transient grating was formed that could be erased by uniform illumination. This transient grating would turn into a permanent one if longer writing times and higher intensities were used. We saw no evidence for the two-beam coupling observed previously in similar dye-doped systems [13]. This would suggest that, in our case, the contribution from a phase-shifted photorefractive grating was negligible. The most important difference between our systems and those published earlier [2, 4, 13] was high dye concentration, and it is likely that the strong adsorption of dye molecules on the surface reduced the strength of the photorefractive surface-mediated space-charge field.

The intensities sufficient to write a permanent grating were low; however, the diffraction efficiency increased if longer illumination times were applied. One of the advantages of using a low incident laser intensity is the reduction of a contribution from thermal effects. With intensities we used, there should be less than 0.5°C increase in temperature.

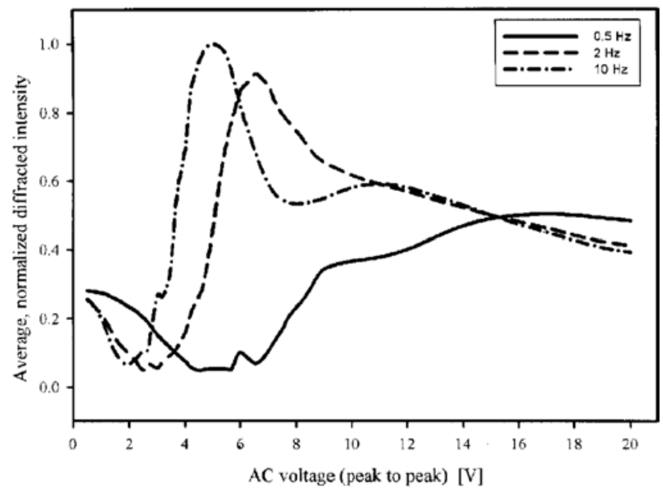

Fig. 2. Average diffraction efficiency versus applied AC field for three different frequencies. The diffraction efficiency has been normalized to the value in the absence of an applied field

In order to test the strength of the permanent gratings, we measured the temperature dependence of diffraction. The cell was heated to reach the nematic/isotropic phase transition. Fig. 1 shows the normalized intensity of the first diffracted order and its dependence on temperature. Increasing the temperature towards the phase transition lowered the diffracted intensity, as expected. At the phase transition, the diffracted intensity experienced a sudden drop. However, in the isotropic phase, a residual diffraction of approximately 20% of the original value remained. When the cell was cooled to room temperature, the diffraction efficiency recovered and, after a few hours at room temperature, the diffraction efficiency returned to its original value. The presence of this residual diffraction indicates that permanent gratings develop on the liquid crystal-ITO interface and that they impose strong anchoring, even at high temperatures.

In the nematic phase, we were able to control the diffraction efficiency by applying a sinusoidal electric field; furthermore, the diffraction efficiency could be either enhanced or reduced. Fig. 2 shows an example of the first diffracted order intensity versus applied AC field, for three different frequencies. The intensity shown has been normalized and has been time-averaged over a number of cycles of the applied field.

We found that, regardless of the frequency and the angle of incidence, the diffraction efficiency had the same characteristic features (Fig. 2). At first, with increasing AC field, the diffraction efficiency decreased

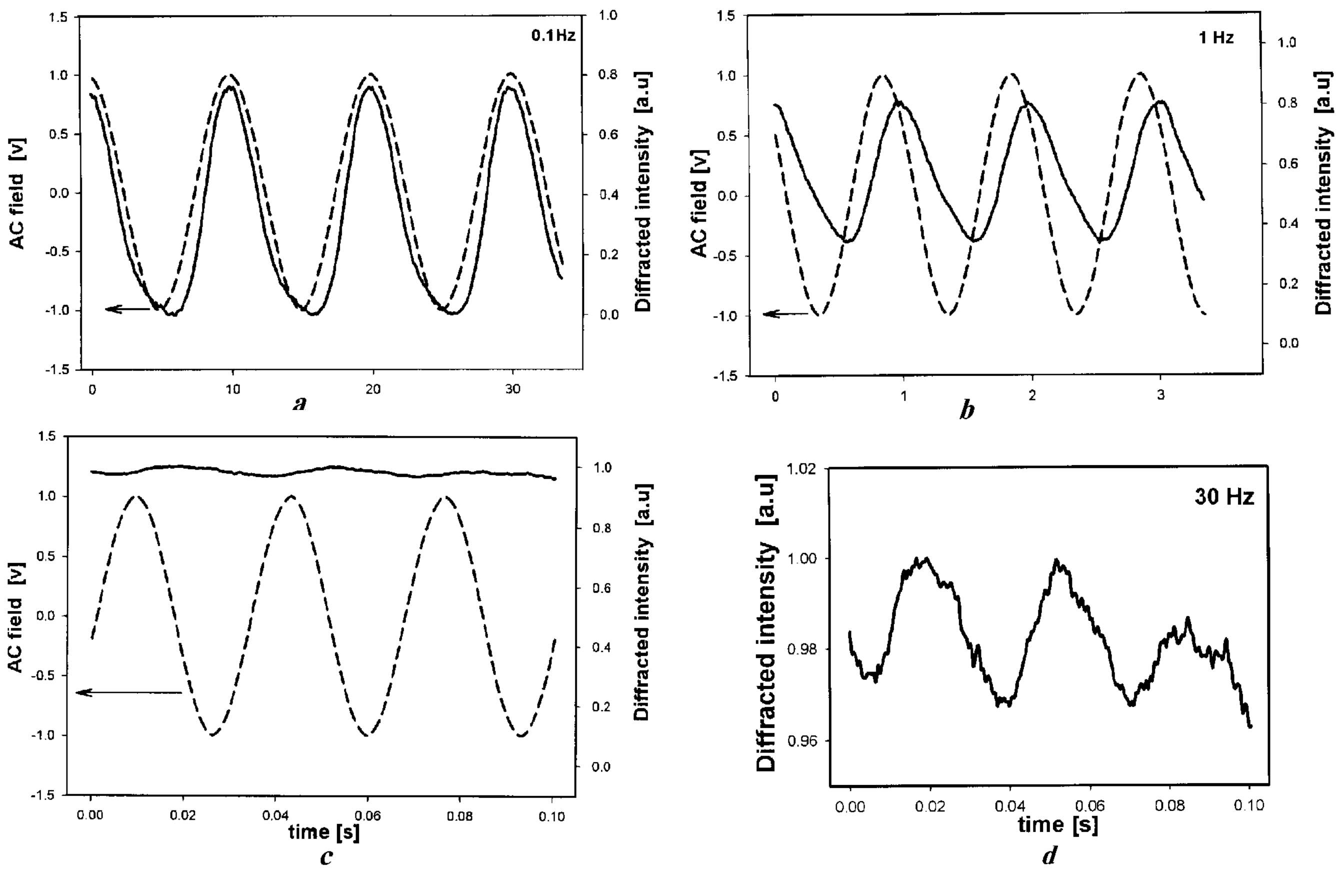

Fig. 3. Modulation of the diffracted signal by AC field. AC field of different frequencies: 0.1 Hz (*a*), 1 Hz (*b*), 30 Hz (*c*), 30 Hz (*d*) — with higher resolution. AC field shown as a dashed line

significantly, almost reaching zero and then increased, reaching a maximum which was 3—4 times higher than the efficiency measured at zero AC field value. The value of AC field at which a maximum and a minimum of the diffraction efficiency are observed depended on the angle of incidence and on the frequency of the applied field. The same dependence on AC field existed for a wide range of frequencies. We applied up to 300 kHz AC field and observed a similar enhancement, though the optimum position of peak diffraction changed. For example, at 300 kHz, the maximum diffraction is observed at 21 V of AC field. This enhancement of diffraction with AC field voltage provides an exciting option for a flexible control of diffraction — a function that could prove invaluable in switching applications.

The maximum diffraction efficiency, defined as the ratio of the first diffracted order intensity versus total incident probe beam intensity, reached 0.56% and was measured for 5 Hz sine AC field of 6 V. This value compares well with the efficiency observed [21] in a similar system (0.4 %).

Fig. 3 presents the example of a modulated diffracted signal and an applied AC field. For low frequencies and low voltage such as, for example, 0.1 and 1 Hz with 1V of AC field (Fig. 3,*a* and 3,*b*), the liquid crystal's signal response follows the applied field. However, higher applied voltages and/or higher frequencies, such as 30 Hz, give much more complex response. This case is shown on Fig. 3,*c*, where the diffracted signal showed a low modulation, although its intensity remained high. Some modulation, nevertheless, exists, as shown in the inset in Fig. 3,*d*, where the diffracted signal modulated at 30 Hz is plotted at higher resolution. Fig. 4 summarizes these results, demonstrating a decrease of the diffracted signal modulation with increasing AC frequency. Such a dependence is expected, as the reorientation of nematic liquid crystal molecules typically occurs on a timescale of milliseconds, so the molecules are unable to follow the higher frequency driving field.

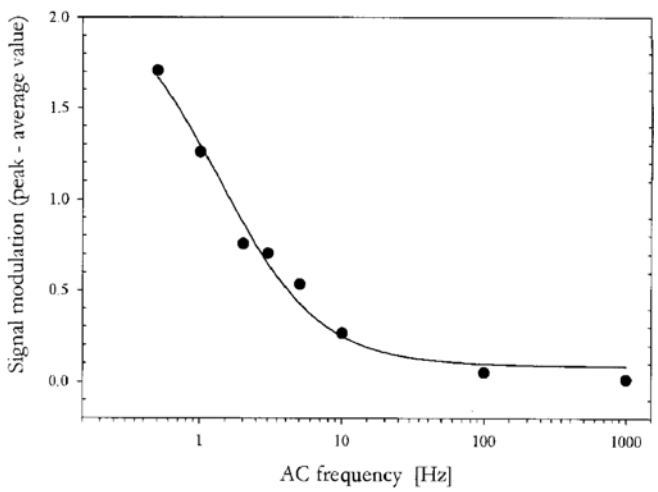

Fig. 4. Diffracted signal modulation dependence on the frequency of applied AC field

Investigating the characteristic features of a diffracted signal, we observed that it followed a hysteresis with the applied AC field (Fig. 5). The measured diffracted intensity was different if AC field was increasing than that for the case of decreasing AC field. The higher the frequency, the more pronounced was the difference. However, in general, it is clear that this hysteresis of the response is present even for very low frequencies, as low as 0.001 Hz. These results indicate some "memory" of the system to a previous light and electric field exposure. The memory effect in similar dye-doped systems has been observed and reported earlier [22].

We have also tested the effect of applying other types of electric field: AC field with a square profile and DC field. In case of square AC field, a similar modulation of diffraction was observed as for the case of a sine field. With DC field, some increase of the diffracted intensity was measured, but much smaller than that in the case of AC fields.

The results on the persistence of gratings in the isotropic phase, as well as controlling their diffraction with an electric field in the nematic phase, confirms earlier reports that surface-mediated effects and charges trapped at surfaces play an important role in the alignment of a whole bulk liquid crystal [18, 21, 22]. The reorientation of liquid crystal molecules was explained [14, 17] by two main mechanisms: light-induced reorientation of dye molecules that exerts a bulk torque on the director, aligning it in the direction perpendicular to the incident light polarization, and phototransformation of dye molecules that adsorbed on the cell surfaces impose a surface torque aligning the director in the direction parallel to the incident polarization. The final orientation of the molecules is a result of the interplay and strength of both mechanisms.

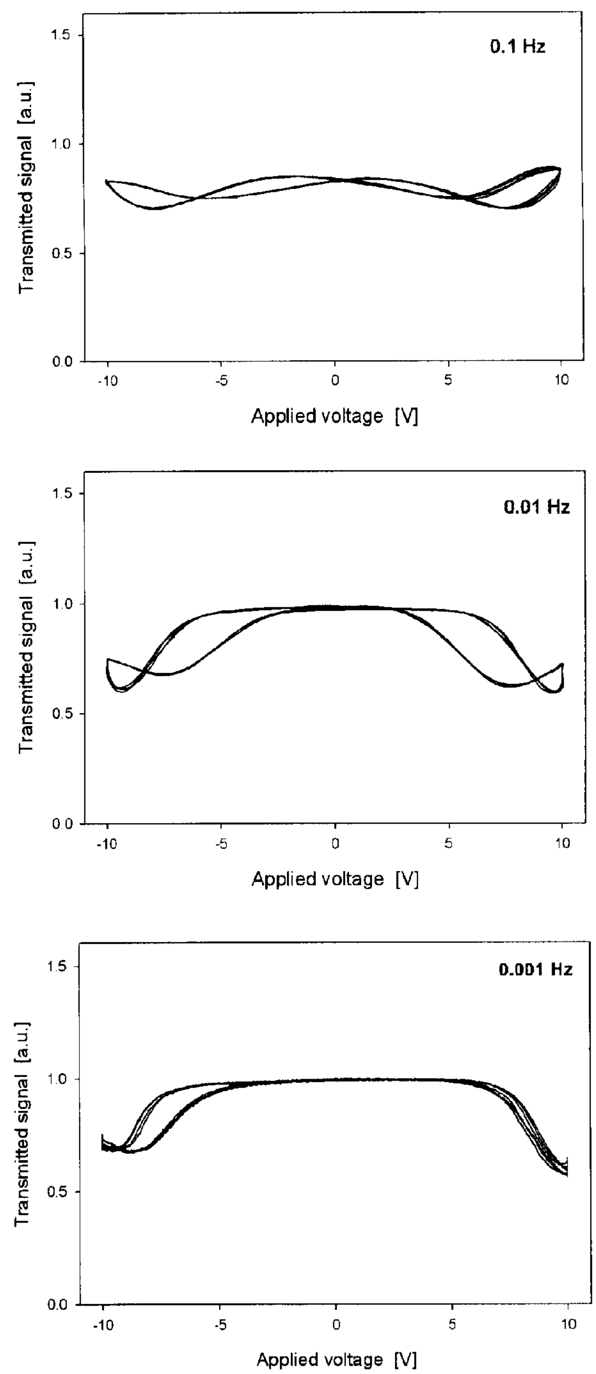

Fig. 5. Hysteresis of the diffracted signal intensity with the varying magnitude of AC field for three AC field frequencies

Moreover, the surface-localized electric field can also provide a strong overall contribution [18]. It can be particularly pronounced if there are ions and impurities in liquid crystals. Photocharges that are generated in the bulk can load the surfaces. Surface charge modulation can also produce an electric field component, normal to the substrates, which can drive ions towards the boundaries where they get adsorbed and trapped. These processes [21, 22] affect not only the orientation of liquid crystals, but also the anchoring conditions — enabling AC field to strengthen or weaken the light-induced diffraction gratings.

As the exact mechanism for the behaviour of permanent gratings we observed is not yet clear, further experimental and theoretical studies will need to be carried out to explain and optimize this process in full.

Conclusions

In conclusion, we demonstrated a method of enhancing or reducing the diffraction from permanent gratings in highly dye-doped nematic liquid crystals. The gratings develop on the liquid crystal-substrate interface and they are robust and durable. Residual diffraction remains even in the isotropic phase. The diffracted signal efficiency reached 0.56%, a value that could be optimized with further experimental investigations. The diffracted signal was easily modulated by either a sine or square AC field, with the depth of the signal modulation dependent on AC frequency. The hysteresis of the diffracted signal response was observed even for very low frequency AC driving fields.

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1. *Rudenko E. V., Sukhov A. V.*//Mol. Cryst. Liq. Cryst. Science & Techn. A.— 1996.— **282**.— P.125—137.
2. *Khoo I. C.*//IEEE J. Quant. Electron.— 1996.— **32**.— P.525—534.
3. *Bartkiewicz S., Miniewicz A., Kajzar F., Zagorska M.*//Mol. Cryst. Liq. Cryst.— 1998.— **322**.— P.9—20.
4. *Khoo I. C., Guenther B. D., Wood M. V. et al.*//Opt. Lett.— 1997.— **22**.— P.1229—1231.
5. *Ono H., Kawatsuki N.*//Jpn. J. Appl. Phys.— 1997.— **36**.— P.6444—6448.
6. *Bartkiewicz S., Miniewicz A., Kajzar F., Zagorska M.*//Appl. Optics.— 1998.— **37**.— P.6871—6877.
7. *Francescangeli O., Slussarenko S., Simoni F. et al.*//Phys. Rev. Lett.— 1999.— **82**.— P.1855—1858.
8. *Gibbons W. M., Shannon P. J., Sun S.-T., Swetlin B.J.*//Nature.— 1991.— **351**.— P.49—50.
9. *Sun S-T., Gibbons W. M.*//Appl. Phys. Lett.— 1994.— **65**.— P.2542—2544.
10. *Lucchetti L., Simoni F., Resnikov Y.*//Opt. Lett.— 1999.— **24**.— P.1062—1064.
11. *Simoni F., Francescangeli O., Reznikov Y., Slussarenko S.*//Ibid.— 1997.— **22**.— P.549—551.
12. *Gibbons W. M., Kosa T., Palffymuhoray P. et al.*//Nature.— 1995.— **377**.— P.43—46.
13. *Khoo I. C., Slussarenko S., Guenther B. D. et al.*//Opt. Lett.— 1998.— **23**.— P.253—255.
14. *Simoni F., Francescangeli O.*//J. Phys: Condens. Mater.— 1999.— **11**.— P.R439—R487.
15. *Chen A. G., Brady D. J.*//Opt. Lett.— 1992.— **17**.— P.441—443.
16. *Sun S-T., Gibbons W. M., Shannon P. J.*//Liquid Cryst.— 1992.— **12**.— P.869—874.
17. *Voloshchenko D., Khyzhnyak A., Reznikov Y., Reshetnyak V.*//Jpn. J. Appl. Phys.— 1995.— **34**.— P.566—571.
18. *Zhang J., Ostroverkhov V., Singer K. D. et al.*//Opt. Lett.— 2000.— **25**.— P.414—416.
19. *Khoo I. C.*//Mol. Cryst. Liq. Cryst.— 1996.— **282**.— P.53—66.
20. *Janossy I., Kosa T.*//Opt. Lett.— 1992.— **17**.— P.1183—1185.
21. *Komorowska K., Miniewicz A., Parka J.*//Synthetic Mater.— 2000.— **109**.— P.189—193.
22. *Tabiryan N., Umeton C.*//J. Opt. Soc. Amer. B.— 1998.— **15**.— P.1912.

ЗБІЛЬШЕННЯ ДИФРАКЦІЇ НА СТАЦІОНАРНИХ ГРАТКАХ В РІДКИХ КРИСТАЛАХ З ДОМІШКОЮ БАРВНИКІВ ПРИ ПРИКЛАДАННІ ЗМІННОГО ЕЛЕКТРИЧНОГО ПОЛЯ

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Р е з ю м е

Перманентні гратки можуть бути записані в рідких кристалах з домішками барвників високої концентрації без прикладання будь-якого зовнішнього електричного поля за допомогою видимого світла невисокої інтенсивності. Ці гратки є адаптивними, оскільки їхня дифракційна ефективність може бути легко керована змінним полем. Інтенсивність дифрагованого променя може також бути модульована електричним полем низької частоти з амплітудою модуляції, що зменшується для вищих частот. Такі стаціонарні гратки є стійкими, не стираються більше року навіть під впливом високої температури.