AIP | Journal of Applied Physics

# On the possible correlation between dark conductivity, photoconductivity, and photorefractivity in dye-doped nematic liquid crystals

Prapong Klysubun and Guy Indebetouw

Citation: Journal of Applied Physics **92**, 2528 (2002); doi: 10.1063/1.1495887
View online: http://dx.doi.org/10.1063/1.1495887
View Table of Contents: http://scitation.aip.org/content/aip/journal/jap/92/5?ver=pdfcov
Published by the AIP Publishing

---

**Articles you may be interested in**
Optically switchable biphotonic photorefractive effect in dye-doped liquid crystal films
Appl. Phys. Lett. **85**, 5822 (2004); 10.1063/1.1836864

Reduced photorefraction in hafnium-doped single-domain and periodically poled lithium niobate crystals
Appl. Phys. Lett. **84**, 1880 (2004); 10.1063/1.1687460

Supra-nonlinear photorefractive response of single-walled carbon nanotube- and C 60 -doped nematic liquid crystal
Appl. Phys. Lett. **82**, 3587 (2003); 10.1063/1.1577215

Charge transport due to photoelectric interface activation in pure nematic liquid-crystal cells
J. Appl. Phys. **92**, 4863 (2002); 10.1063/1.1511273

Orientational photorefractive effect in nematic liquid crystal with externally applied fields
J. Appl. Phys. **88**, 1709 (2000); 10.1063/1.1305905

---

MIT LINCOLN LABORATORY CAREERS

Discover the satisfaction of innovation and service to the nation

- Space Control
- Air & Missile Defense
- Communications Systems & Cyber Security
- Intelligence, Surveillance and Reconnaissance Systems
- Advanced Electronics
- Tactical Systems
- Homeland Protection
- Air Traffic Control

LINCOLN LABORATORY
MASSACHUSETTS INSTITUTE OF TECHNOLOGY

LEARN MORE

[This article is copyrighted as indicated in the article. Reuse of AIP content is subject to the terms at: http://scitation.aip.org/termsconditions. Downloaded to ] IP: 128.173.126.47 On: Fri, 01 May 2015 20:15:10

JOURNAL OF APPLIED PHYSICS VOLUME 92, NUMBER 5 1 SEPTEMBER 2002

# On the possible correlation between dark conductivity, photoconductivity, and photorefractivity in dye-doped nematic liquid crystals

Prapong Klysubun[a] and Guy Indebetouw[b]
*Department of Physics, Virginia Tech, Blacksburg Virginia 24061-0435*

(Received 11 February 2002; accepted for publication 3 June 2002)

The dc conductivity of dye-doped nematic liquid crystal cells exhibits a characteristic transition from a cubic current–voltage relationship at applied voltages smaller than about 1 V to a linear one at voltages larger than about 1.5 V. The photoconductivity, and the related photorefractive response of these cells exhibit an apparent threshold at about the same characteristic voltage: An applied voltage larger than about 1.5 V is needed to measure sizeable photocurrents and photorefractive responses. We propose a model that is consistent with all these observations. At low applied voltages, a residual space charge limits the dc current, and prevents the participation of photoexcited charge carriers to photoconductivity and photorefractivity, while at higher applied voltages, the disappearance of the residual space charge allows the manifestation of these photoinduced responses. Experimental results seem to confirm the validity of the model. © *2002 American Institute of Physics.*
[DOI: 10.1063/1.1495887]

## I. INTRODUCTION

Recent renewed interest in liquid crystal systems for information storage and processing stems from the huge enhancement of their coupling to an optical field due to orientational effects.[1] This enhancement is particularly strong in dye-doped liquid crystal systems, which thus exhibit interesting and potentially useful photorefractive properties.[2,3] A broadly accepted theoretical framework explaining the orientational enhancement was proposed by Janossy,[4] and refined by Marrucci.[5] In this conjecture, a necessary condition for the enhancement is that the photoexcitation of a dye molecule must result in a significant change in the guest/host interaction. A number of mechanisms resulting in such changes have been identified,[6,7] resulting in enhancements that can be positive or negative depending on the system's interactions. These mechanisms may involve conformational changes of the dye-molecule (e.g., *cis-trans* isomerization of azo dye), or mechanisms that involve an enhanced dipolar interaction, or hydrogen bonding, of the photoexcited guest with the host. In an attempt to sort out these mechanisms, and to complement published results,[6–9] we have compared the photorefractive responses of a large number of guest/host systems. The result of this study will appear elsewhere.

Three important observations were made in the course of this study.

(1) At low applied voltages ($<1$ V), the dc dark current increases approximately as the cube of the applied voltage. This behavior then changes to a linear (ohmic) current at higher voltages. In that ohmic region, the dc dark conductivity varies as the square root of the dye concentration.

(2) The photoconductivity of the cell increases linearly with the light intensity, and with the dye concentration. However, virtually no photoconduction is measured below an apparent threshold of about 1.5 V.

(3) The photorefractive response, measured by asymmetric beam coupling in the cell, also exhibits an apparent threshold at approximately the same voltage, above which the beam coupling ratio grows approximately linearly with the applied voltage.

The aim of this article is to explore the possible correlation between these observations, and to propose a simple model that is compatible with all the observed behaviors. The model draws on known or expected behaviors of liquid crystal systems. It assumes that ionic charge carriers are generated primarily near the electrodes by electrochemical reactions activated by the injection of charges.[10] At low voltages, the currents are limited by a residual ionic space charge, as in the double injection model of Lampert and Rose.[11] This predicts a cubic $I-V$ characteristic. At higher voltages, this residual space charge disappears, and the cell behaves as a weak electrolyte with ohmic conductivity, as in the liquid crystal conductivity model of Blinov.[12] These two models, valid in two different ranges of the applied voltage, can thus explain the peculiar dark conductivity of the dye-doped liquid crystal cells, which had been reported by other,[13] but has not been explained. The model can also explain qualitatively the apparent threshold observed in the photoconductivity, and the photorefractive response, as well as the dependence of these responses on the dye concentration, and on the light irradiance level.

In the next section, we describe the experimental results that have triggered our curiosity for further exploration. We discuss, in Sec. III, how the proposed model can explain the peculiar $I-V$ response. Sections IV and V, respectively, show how the model is compatible with the observed photoconductive, and photorefractive responses. The last section

---

[a] Present address: National Synchrotron Research Center, 111 University Avenue, Muang District, Nakhon Ratchasima, 30000 Thailand.

[b] Electronic mail: gindebet@vt.edu

[This article is copyrighted as indicated in the article. Reuse of AIP content is subject to the terms at: http://scitation.aip.org/termsconditions. Downloaded to ] IP: 128.173.126.47 On: Fri, 01 May 2015 20:15:10

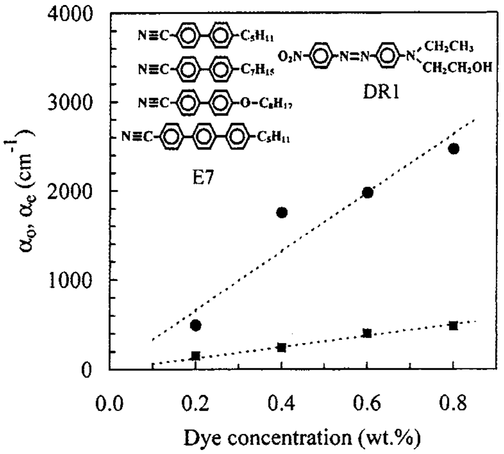

FIG. 1. Absorption coefficients for the mixture E7/DR1 at various dye concentrations. e-(circular dots) and o-(square dots) refer to light polarizations parallel and perpendicular to the molecular axis, respectively.

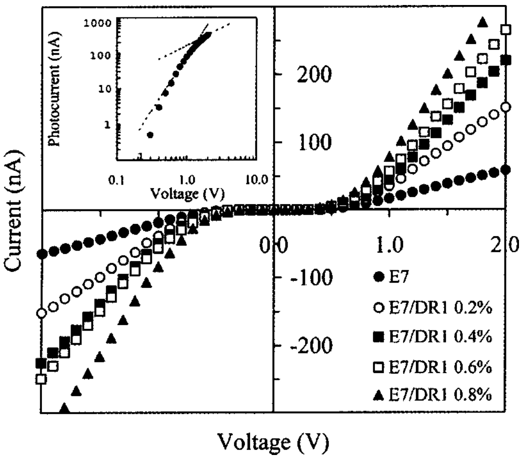

FIG. 2. Current–voltage relationship of 25-$\mu$m-thick cells of E7/DR1 with various dye concentrations. There is no current below about 0.3 V. At low voltages (0.3–1.5 V, approximately), the current grows as the cube of the voltage. Above about 1.5 V, the current is linear in applied voltage). The log–log plot of the inset shows two lines with slopes 3 and 1.

reports some intriguing photorefractive data obtained with very low frequency ac applied voltages. Although more difficult to interpret, these data are shown to be compatible with the proposed model, and seems to confirm its validity.

## II. EXPERIMENTAL OBSERVATIONS

The results reported in this section are for a system consisting of the nematic mixture E7 (from Merck), and the dye disperse red 1 (DR1 from Aldrich). Their molecular structures are shown in the inset of Fig. 1. A number of different dye-liquid crystal combinations were found to exhibit behaviors similar to these reported here. The samples were prepared between two indium tin oxide-coated glass plates held 25 $\mu$m apart by Mylar spacers. The plates were treated with octadecyltrichlorosilane, and baked to induce a homeotropic alignment of the nematic director. The homogeneity of the cell was checked under a polarizing microscope. The cells' area is about 2 cm$^2$. The absorption coefficients for the o- and e-waves (respectively, polarized perpendicular and parallel to the molecular axis) are shown in Fig. 1 for different dye concentrations. $\alpha_e$ was calculated from measurement at oblique incidence, which leads to a sizeable uncertainty due to error propagation. There is also an uncertainty in measuring the exact amount of dye dissolved in the host. Nevertheless, the expected linear relationship is obtained.

Figure 2 shows the dc current–voltage response of cells with different concentrations. Voltages were applied to the cell from a stabilized dc power supply (Hewlett-Packard 6235A), measured with a digital multimeter (Keithley 177), and the dc current was monitored with a picoammeter (Keithley 485). For small voltages (<1 V), the current grows as the cube of the voltage. This is shown more explicitly on the log-scale plot in the insert. At higher voltages (>1.5 V approximately), the current grows linearly with the applied voltage. The transition between the cubic and linear currents is continuous, but the two limits correspond to two distinct regimes in the model. For this reason, they are referred to as, respectively, the "low voltage" or "space charge-limited current" regime, and the "high voltage" or "ohmic" or "weak electrolyte" regime. The dark dc current and the total (dark and photo) current of a cell E7-DR1 0.6% (a different cell from that of Fig. 1) are shown in Fig. 3. The photocurrent, calculated as the difference between the two curves, is shown in the inset. The photocurrent resulted from a steady illumination of the sample by a 30 mW cw laser beam (wavelength 532 nm, cross-section area 4 mm$^2$). The photocurrent is about one order of magnitude smaller than the dark current, which is typical for organic polymeric materials. The salient feature of this data is that a measurable photocurrent appears only for applied voltages higher than about 1.5 V. Below that threshold, the photocurrent is below the sensitivity of our apparatus. Photoconductivity is thus observed in the ohmic regime of the cell, but not in its space charge-limited regime. Figure 4 illustrates the photorefractive response of a cell E7-DR1 0.2% (different from that of Fig. 2). Two laser beams of equal intensity were made to overlap with a small angle in the cell, with an average incidence angle of 45°. The photo-

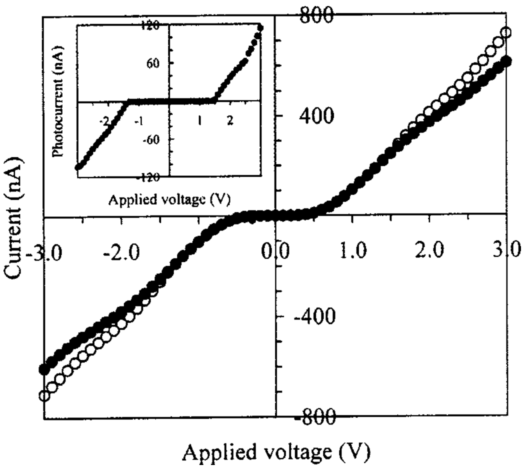

FIG. 3. Dark current (filled dots) and total (dark plus photo) current (open dots) for a cell E7/DR1 0.6%. The inset shows the linear photocurrent above 1.5 V, for an illumination by a 30 mW laser beam (wavelength 532 nm, cross-section area 4 mm$^2$).

[This article is copyrighted as indicated in the article. Reuse of AIP content is subject to the terms at: http://scitation.aip.org/termsconditions. Downloaded to ] IP: 128.173.126.47 On: Fri, 01 May 2015 20:15:10

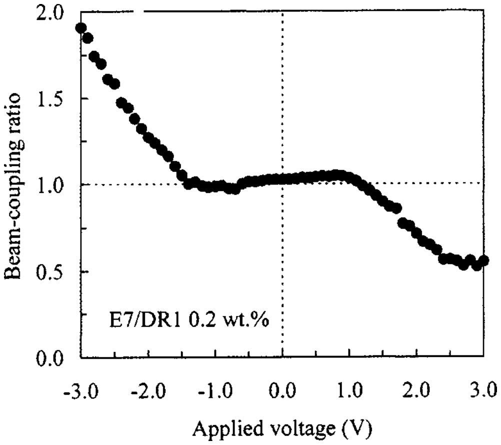

FIG. 4. Photorefractive response of a cell E7/DR1 0.2%, characterized by the asymmetric energy exchange in a two-beam coupling experiment, and showing an apparent threshold at about 1.5 V.

refractive response to the incident interference pattern results in an asymmetric energy exchange between the two beams. This asymmetry is a signature of the nonlocal nature of the photorefractive effect, and the beam ratio at the exit face of the cell was taken as a measure of the magnitude of the effect. It was shown elsewhere[9] that the induced photorefractive grating is a phase grating (refractive index modulation) shifted by $\pi/2$ relative to the interference pattern creating it. It is thus thought to be a grating formed by a static space charge modulation, due to trapped charges in the cell. Figure 4 shows that the photorefractive response exhibits also an apparent threshold around 1.5 V, the voltage at which the cell enters its ohmic regime, and photoconduction appears.

## III. DARK CURRENT MODEL

It is generally accepted that conductivity in liquid crystals is ionic.[12] The charge carriers can be intrinsic ionic impurities, geminate ion pairs generated in the bulk by background ionization under the effect of an applied field, or generated in the bulk by photoionization, and ions can also be created near the electrodes by photochemical reactions activated by the injection of electrons and holes. This latter mechanism has been shown to dominate the conductivity in a number of liquid crystal systems.[10] We speculate that this is the case in our system, because it actually predicts all the observations exposed in the previous section.

In the model, ions are produced near the electrodes by photochemical reactions activated by the injection of holes at the anode, and electrons at the cathode. Cations generated near the anode diffuse rapidly through a thin ''injection layer'' and then drift toward the cathode. Anions from the cathode move in the opposite direction, and bilinear recombination takes place in the bulk. For simplicity's sake, a symmetrical behavior of anions and cations is assumed. This assumption may not be correct (in a number of systems, anions, and cations have very different mobilities, for example), but as far as conductivity is concerned, the validity of the assumption may affect the detailed balance, but not the general current behavior.

In the low voltage regime, the charge carriers generated near on electrode migrate toward the opposite electrode, and recombine with counter ions on their way. If recombination is incomplete, there will be excess ionic charges in the cell (excess of cations on the anode side, and excess of anions on the cathode side).[14] This residual space charge is what limits the current in this regime.[11] The total number of ions per unit area generated near the electrodes is $N_i$, so that the total surface charge density there is $qN_i$, where $q$ in the elementary charge. For simplicity's sake, as already stated, anions and cations are assumed to have the same mobility $\mu$, and the same lifetime $\tau$. The cell can then be divided into three regions. Two regions of width $\delta$ each, adjacent to the electrodes where there is a residual excess charge density (these are dubbed the ''space charge regions''), and a central, nearly neutral region of width $d-2\delta$ ($d$ is the thickness of the cell). As in other injection systems,[11] it is reasonable to assume that diffusion currents are significant only in the thin, so-called injection layers. The bulk currents are thus drift currents, and they are assumed to be volume (as opposed to electrode) limited (i.e., the electric field vanishes at both electrodes). The problem has two characteristic times, the carrier lifetime $\tau$, and the transit time $T$ for a carrier to cross the space charge region from the electrode to the central neutral region. The amount of excess charge in the space charge region depends on the ratio of these two characteristic times. A long transit time inhibits recombination and results in a larger residual excess charge, while a long lifetime increases the recombination probability and results in less excess charge.[11] The conjecture is that the excess charge per unit area in the space charge region is about

$$Q \approx qN_i T/\tau. \tag{1}$$

Since the excess charge is approximately confined in a region of width $\delta$, the charge density in that region of the cell is $\approx Q/\delta$. The electric field in the cell vanishes at the anode, increases in the positive space charge region up to a maximum $E_0$ in the central region, and decreases in the negative space charge region to vanish at the cathode. Poisson's equation in the space charge region ($0<x<\delta$) gives $dE/dx \approx E_0/\delta \approx Q/\epsilon\delta$, where $\epsilon$ is the dielectric constant. Thus

$$E_0 \approx Q/\epsilon. \tag{2}$$

Using an average electric field $E_0/2$ in the space charge region, and integrating Poisson's equation gives

$$\int_0^d E\,dx \approx (d-\delta)E_0 = V_0, \tag{3}$$

where $V_0$ is the voltage applied across the cell. Using the same average electric field $E_0/2$, the transit time through the space charge region is found to be

$$T \approx \delta(\mu E_0/2)^{-1}, \tag{4}$$

where $\mu$ is the carrier mobility. Finally, using Eqs. (1)–(4), the current density can be written as

$$J \approx qN_i/T \approx \varepsilon\tau\mu^2\delta^{-2}(d-\delta)^{-3}V_0^3, \tag{5}$$

where a numerical constant has been omitted. This is the first main result of the model. It shows that, as long as there

[This article is copyrighted as indicated in the article. Reuse of AIP content is subject to the terms at: http://scitation.aip.org/termsconditions. Downloaded to ] IP: 128.173.126.47 On: Fri, 01 May 2015 20:15:10

exists a sizeable region of excess charge so that the resulting residual space charge limits the current, one may expect a cubic $I-V$ relationship. As the voltage increases, however, one expects the carrier lifetime to become increasingly field dependant (for injection in dielectrics, for example, the carrier lifetime is found to follow $\tau \propto V^{-1}$, approximately),[15] so that the cubic behavior is expected only at very low voltages.

Geminate ion pairs can also be generated in the bulk through field ionization. At such low field, however, the process in not expected to be significant.[14] Furthermore, if an anion-cation pair is created in the positive space charge region, for example, where there is an excess of cations, we expect an immediate recombination to take place, leaving the residual space charge distribution unchanged. Consequently, the carriers created by field driven ionization, even if in substantial number, are not expected to contribute to the dark current as long as a sizeable residual space charge is present.

Clearly, this simple scenario will breakdown at some point. As the applied voltage increases, the width of the regions of excess charge shrinks until the entire cell is neutral with uniform recombination throughout (except for the so-called injection layers). The electric field is then constant in the cell (except near the electrodes), $E_0 \approx V_0/d$, and the current is now limited by the ions generation rate in the injection layers, and the recombination rate in the cell. The cell then behaves as a weak electrolyte with ohmic conduction. If $n_0$ is the number density (per unit volume) of unrecombined anions and cations in the cell, $\gamma_D$ is the dissociation rate of the species producing the ions (the dye in our system), $c$ is the dye concentration, and $\gamma_R$ is the bilinear recombination rate, we have, in steady state[12]

$$\gamma_D c \approx \gamma_R n_0^2. \tag{6}$$

Furthermore, if $N_i$ is the number of ions per unit area created in the injection layers, and $n_0$ is the number of ions per unit volume in the cell, $N_i \approx n_0 d$, and the current density becomes, using Eq. (6):

$$J \approx qN_i/T_0 \approx (q\mu/d)(\gamma_D/\gamma_R)^{1/2} c^{1/2} V_0, \tag{7}$$

where $T_0 \approx d(\mu E_0)^{-1}$ is the transit time through the entire cell. In conclusion, when the applied voltage is high enough and the excess space charge vanishes, the current is expected to be ohmic, with conductivity proportional to the square root of the dye concentration. In this regime, the additional carriers generated in the bulk by field ionization is still expected to be small, but now they will contribute to the current, since there is no excess charge to immediately neutralize them. This current contribution from carriers generated in the bulk is expected to be proportional to the dye concentration, so that the total conductivity is expected to increase more rapidly than as $c^{1/2}$. Furthermore, the departure from the square root behavior may give a measure of the ratio of these two current contributions. The dark conductivity of cells with different concentrations is shown in Fig. 5. Conductivity was measured as the slope of the linear region of the $I-V$ curves of Fig. 2. The fit gives $\sigma \approx c^{0.54}$, which is compatible with the model, and confirms the assumption that the process of carrier generation near the electrodes (due to charge injection) largely dominates the process of geminate generation in the bulk.

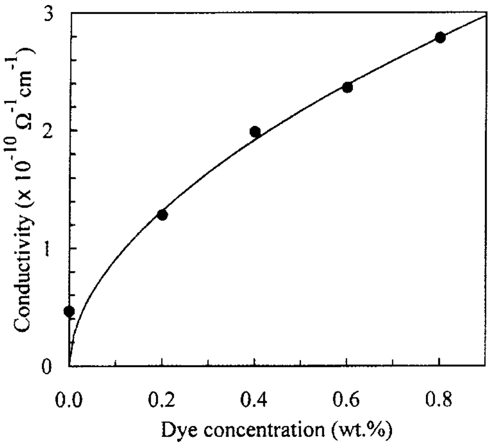

FIG. 5. dc dark conductivity of 25-mm-thick cells of the E7/DR1 mixture with various dye concentrations. The solid line is a best fit giving a relationship $\sigma \approx c^{0.54}$.

## IV. PHOTOCONDUCTION

The model discussed in the previous section can also explain qualitatively the existence of an apparent threshold in the photocurrent (more precisely, a transition between a regime of no photoconductivity at low voltages to a regime of ohmic photoconductivity at higher voltages). The contribution to the current of carriers created in the bulk by photoionization is similar to that of carriers created there by field ionization. At low voltages, when there exists a sizeable residual space charge in the cell, one of the ions of a photogenerated pair immediately recombines with an excess counter ion, leaving the excess space charge distribution unchanged. Consequently, these charges do not contribute to the current, and there is no (or negligible) photoconductivity. At higher voltages, when the cell behaves as a weak electrolyte, and the dark conductivity is ohmic, the photogenerated carriers add to the uniform distribution of unrecombined ions in the bulk, and thus contribute to the current. The total number density of ions in the bulk is then $n = n_0 + sIc$, where $n_0 \approx (\gamma_D c/\gamma_R)^{1/2}$ is the contribution due to charge generation at the electrodes, $s$ is the photoionization constant (per unit light intensity), $I$ is the light intensity, and $c$ is the dye concentration. In the ohmic regime, the total current is of the form $i \approx (\sigma_0 + \sigma_{\mathrm{PH}})V_0$, with $\sigma_0 \propto c^{1/2}$, and $\sigma_{\mathrm{PH}} \propto Ic$. Figure 6 shows the photocurrents of cells with different concentrations as functions of the illumination irradiance (the symbols are the same as in Fig. 2). The predicted linear relationship is clearly confirmed. The inset of Fig. 6 shows the photocurrent as a function of the concentration for an illumination of 1 W/cm$^2$. Allowing for some uncertainty in determining the exact amount of dye in the cell, the concentration dependence of the photocurrent is also linear, as predicted.

[This article is copyrighted as indicated in the article. Reuse of AIP content is subject to the terms at: http://scitation.aip.org/termsconditions. Downloaded to ] IP: 128.173.126.47 On: Fri, 01 May 2015 20:15:10

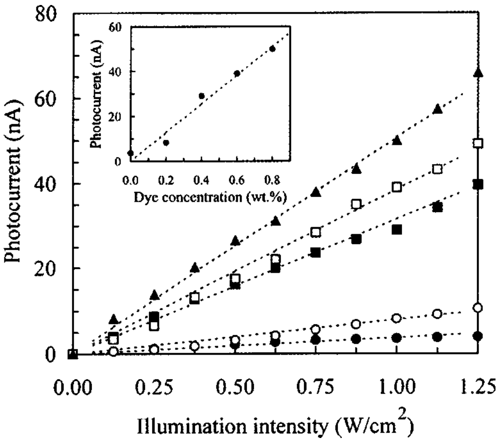

FIG. 6. Photocurrent vs illumination intensity for 25-mm-thick cells of the E7/DR1 mixture with various dye concentrations (the symbols are the same as in Fig. 2). The inset shows the linear dependence of the photocurrent on the dye concentration.

## V. PHOTOREFRACTIVITY

The photorefractive response of our samples (Fig. 4) shows, as the photoconductivity, an apparent threshold at an applied voltage marking the transition between the low-voltage, space charge-limited regime, and the high-voltage, weak ohmic electrolyte regime. Photorefraction, however, requires not only the photogeneration of charge carriers, but also the trapping of charges by immobile sites to form a static space charge. The resulting space charge field, the spatial distribution of which follows that of the light irradiance distribution that creates it, results in a refractive index modulation via an electro-optic or an orientational response in the material. The formation of a stationary space charge requires that the symmetry between anions and cations, assumed in the discussion of the conductivity, be broken. Various mechanisms could be responsible for breaking that symmetry, e.g., different ionic mobility, different trapping probability, or different probability of charge transfer from the ions to immobile sites, for example. We assume that one such mechanism is at play in our system, since it exhibits a photorefractive response. When the irradiance is a spatially periodic interference pattern of two coherent laser beams, the result is a photorefractive grating that induces a coupling between the interfering beams. In such experiments, charge carriers are photoexcited nonuniformly in a spatial pattern having the same distribution as the interference pattern. The fate of these carriers varies.

In the low voltage regime, the carriers created by photoionization appear in regions of the cell where a residual excess of anions or cations exists. As before then, one of the ions recombines with an excess counter ion, and the residual space charge remains unchanged. Of course these ions have a certain probability of being trapped by, or transfer their charge to an immobile site, but since the ionic distribution does not acquire a periodic modulation, no spatially modulated static space charge is formed, and no photorefractive response is measured. In the high voltage, ohmic regime, the residual space charge distribution of excess ionic carriers vanishes. Photoexcited pairs created in the region of high irradiance then add to the already present uniform distribution of unrecombined ions, and the ionic distribution acquires a spatial modulation that follows the irradiance distribution. As a result, more charges are trapped by immobile sites in the high irradiance regions, and a static, modulated space charge distribution is formed, resulting in a photorefractive response.

In conclusion, the proposed model can explain the peculiar dark conductivity of the cell, and the apparent threshold in photoconduction, and photorefraction. The low frequency experiments described in the next section, although more difficult to interpret, seem to confirm the proposed scenario.

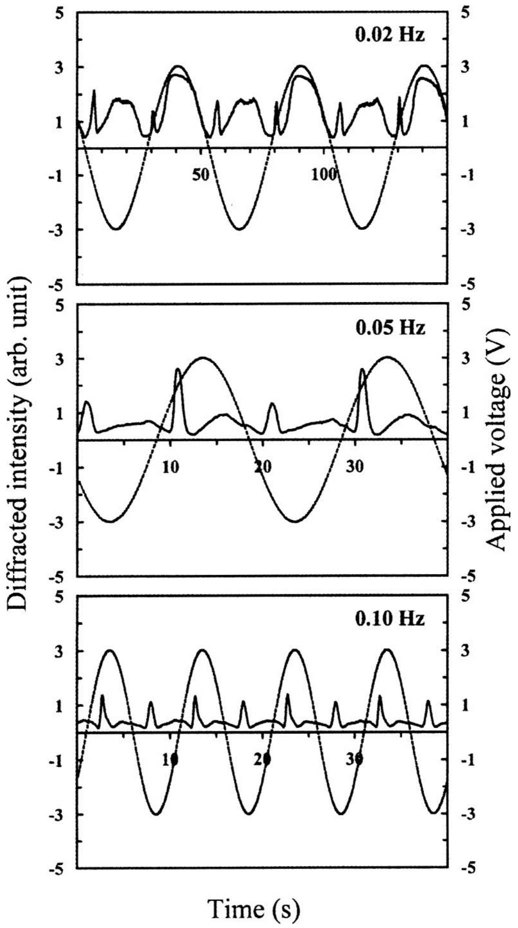

FIG. 7. Dynamic photorefractive response of a cell E7/DR1 0.4%, characterized by the intensity of one of the self-diffracted beams, with low frequency ac voltages applied to the cell.

## VI. LOW FREQUENCY ac RESPONSE

Some photorefractive experiments were carried out with low frequency ac applied voltages, in order to explore the dynamics of the photorefractive response. Typical data are shown in Fig. 7. These data were obtained by illuminating the cell with a spatially modulated polarization pattern obtained by superposing two laser beams with opposite circular polarizations, and intersecting with a small angle in the cell. The resulting polarization pattern in the cell is everywhere linear, but with a direction that varies periodically between ordinary and extraordinary (i.e., a so-called polarization grating). Due to the strong dichroism of the dye (illustrated in

[This article is copyrighted as indicated in the article. Reuse of AIP content is subject to the terms at: http://scitation.aip.org/termsconditions. Downloaded to ] IP: 128.173.126.47 On: Fri, 01 May 2015 20:15:10

Fig. 1), the absorption rate is spatially modulated, with the same period as the polarization grating. As far as charge generation concerned, the situation is thus similar to what is obtained with an intensity-modulated interference pattern. Consequently, one expects the photorefractive response to be manifest only when there is no residual ionic space charge in the cell. A low frequency ac voltage with a peak value of 3 V was applied to the cell. Here, the photorefractive response was monitored by measuring the intensity of one of the self-diffracted beams that appear as a result of the nonlinear optical interaction in the cell. The interpretation of this complicated data is not simple, but the proposed model provides some clues. At very low frequency (0.02 Hz for the first trace of Fig. 7), one observes a transient response that follows, with a delay of a few seconds, the change of polarity of the applied voltage. Another response then appears at a higher voltage, and follows the applied voltage monotonically. A possible interpretation of this data is the following. When the voltage is low ($<1$ V), before a change of polarity, the cell is in the low voltage, space charge limited regime where no photorefractive response is measured for the reason explained in the previous section. When the applied voltage switches polarity, the ions created near the electrodes have a charge opposite to that of the ions of the nearby residual space charge. The residual space charge is thus relatively quickly neutralized by these ions, leaving the cell in a neutral state where a photorefractive grating can be formed. This, however, can only last for a short time, until a residual space charge of opposite sign is formed, and washes out the response. It is only when the applied voltage reaches values above about 1.5 V, that the residual space charge disappears, and the photorefractive response reappears. The response then follows the applied voltage monotonically. At higher frequencies (0.05 Hz for the second trace of Fig. 7), the transient response is seen to increase, and the monotonic response to decrease. The first observation can be explained by the fact that the rate of production of the counter ions that cancel out the residual space charge just after a polarity change of the voltage is higher at higher frequency. Making this cancellation more effective will enhance the transient response. The decrease of the monotonic response is presumably due to the fact that, at higher frequency, the residual space charge does not have time to disappear entirely (as the voltage keeps the same polarity in this part of the cycle, there is no creation of counter ions to cancel the residual space charge). There is a time delay of a few seconds between the polarity change of the voltage, and the appearance of the transient response, as well as between the time at which the voltage reaches a value sufficiently high to eliminate the residual space charge, and the appearance of the monotonic response. When this delay approaches values close to one quarter of the temporal period of the applied voltage modulation, we expect the monotonic response to disappear, as seen at 0.10 Hz in the third trace of Fig. 7. Thereon, the transient response decreases, and eventually disappears at higher frequencies.

## VII. SUMMARY

Dye-doped liquid crystal cells exhibit peculiar dark and photoconductivities. At low voltages ($<1$ V), the dc dark current is cubic in applied voltage, then becomes linear (ohmic) at voltages above about 1.5 V. In the ohmic regime, the dark conductivity varies approximately as the square root of the dye concentration. Photoconductivity appears only for voltages above about 1.5 V, and varies linearly with the dye concentration, and with light intensity. The photorefractive response also shows a transition from no, or negligible, response to a linear response above about 1.5 V. All these experimental data lead us to think that there may exist a correlation between these observations.

We propose a model that is compatible with, and predicts, these behaviors. Ionic charge carriers are produced primarily near the electrodes by electrochemical reactions activated by charge injection. The cell then behaves as a weak electrolyte. As long as there exists a residual ionic space charge limiting the current in the cell (at low applied voltages), the dark current is cubic in applied voltage, and carriers produced in the bulk by field-induced or photoinduced ionization do not contribute to the current. In this space charge limited regime, photogenerated charges in the bulk are also prevented from forming the static space charge necessary to observe photorefractivity. At higher applied voltages, the residual ionic space charge disappears. The cell is now approximately neutral throughout, and exhibits ohmic conduction. In these conditions, the photogenerated charges produce a photocurrent. In that regime, a spatially modulated illumination results in a spatially modulated ionic charge distribution, which, in turn, creates a modulated static space charge leading to a photorefractive response.

The complicated photorefractive dynamics of the cell observed with low frequency ac applied voltages is compatible with the model, which appears to confirm its validity.

[1] G. P. Wiederrecht and M. R. Wasielewski, J. Nonlinear Opt. Phys. Mater. **8**, 107 (1999).
[2] G. P. Wiederrecht, Annu. Rev. Mater. Sci. **31**, 139 (2001).
[3] N. V. Tabiryan, A. V. Sukhov, and B. Ya. Zel'dovich, Mol. Cryst. Liq. Cryst. **136**, 1 (1986).
[4] I. Janossy, Phys. Rev. E **49**, 2957 (1994).
[5] L. Marrucci and D. Paparo, Phys. Rev. E **56**, 1765 (1997).
[6] L. Marrucci, D. Paparo, P. Maddalena, E. Massera, E. Prudnikova, and E. Santamato, J. Chem. Phys. **107**, 9783 (1997).
[7] E. Santamato, G. Abbate, P. Maddalena, L. Marrucci, D. Paparo, and E. Massera, Mol. Cryst. Liq. Cryst. **302**, 111 (1997).
[8] L. Marrucci, D. Paparo, M. R. Vetrano, M. Colicchio, E. Santamato, and G. Viscardi, J. Chem. Phys. **113**, 10361 (2000).
[9] P. Klysubun and G. Indebetouw, J. Appl. Phys. **91**, 897 (2002).
[10] S. A. Pikin, *Structural Transformations in Liquid Crystals* (Gordon Breach, New York, 1991), Chap. 7.
[11] M. A. Lampert and A. Rose, Phys. Rev. **121**, 26 (1961).
[12] L. M. Blinov and V. G. Chigrinov, *Electro-Optical Effects in Liquid Crystals* (Wiley, New York, 1983), p. 57.
[13] I. C. Khoo, IEEE J. Quantum Electron. **32**, 525 (1996).
[14] L. M. Blinov, *Electro-Optical and Magneto-Optical Properties of Liquid Crystals* (Wiley, New York, 1983), p. 207.
[15] M. A. Lampert, Rep. Prog. Phys. **27**, 329 (1964).

[This article is copyrighted as indicated in the article. Reuse of AIP content is subject to the terms at: http://scitation.aip.org/termsconditions. Downloaded to ] IP: 128.173.126.47 On: Fri, 01 May 2015 20:15:10