Quasistatic-electric- and optical-field-induced birefringence and nonlinear-optical diffraction effects in a nematic-liquid-crystal film

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(Received 16 September 1985)

We present theoretical and experimental results on quasistatic-electric- and optical-field-induced birefringence as well as quasistatic-electric-field-enhanced optical diffraction effects in a nematicliquid-crystal film. For an inclined-incident pump beam, the enhancement of the diffraction effect and birefringence values due to the quasistatic electric field does not saturate at high quasistatic electric field strength. Instead, it decreases from the maximum, through zero (i.e., no effect), to a region of negative coupling between the fields. Locally this novel zero-crossing phenomenon occurs at a quasistatic electric field strength for which the nematic director has been reoriented by the electric field to align with the polarization direction of the optical field. The zero-crossing effect manifested by the sample as a whole is the net result of the field-coupling effect on the local director along the beam path, which locally could be positive, zero, or negative. This novel effect is also independent of the pump-laser intensity. Our experimental results are in quantitative agreement with theoretical predictions.

I. INTRODUCTION

Because of its large molecular anisotropy and strong correlation among the molecules, it is well known that the director, i.e., the average molecular orientation of the liquid crystalline material, can be easily reoriented by a static electric and magnetic field,¹ and the birefringence of the sample would change accordingly. This is referred to as the Fréedericksz transition. Electrically controlled birefringence (ECB) has since been used for light valves and other applications.² The optical-field-induced Fréedericksz transition and associated nonlinear optical effects in a nematic-liquid-crystal (NLC) film such as self-focusing, $^{3-6}$ degenerate wave mixing, $^{6-9}$ and optical bistability $^{10-14}$ have also been extensively studied recently. In particular, interesting nonlinear-optical propagation effects, e.g., far-field diffraction rings, 5,15-17 have been observed. This phenomenon has been described as the spatial analog of the well-known self-phase modulation of light as the laser beam traverses the sample. The diffraction effect is shown to be further enhanced by biasing with a quasistatic electric field on the sample.¹⁸ It was also recently shown theoretically that intrinsic optical bistability in a homeotropically aligned NLC cell can be enhanced or suppressed by a low external field.¹⁹ In a previous letter²⁰ we have shown that quasistatic-electricfield-enhanced diffraction effects in a NLC film can be described by a simple physical model as the combined effects of the critical behavior of the NLC just above the quasistatic-electric-field-induced Fréedericksz transition and the nonlinear coupling of the optical and quasistatic electric fields. In this paper we present results of a detailed investigation on quasistatic-electric- and opticalfield-induced birefringence and diffraction effects in a NLC film. In Sec. II we briefly state the essential results from an approximate theory for field-induced molecular

reorientation angles and birefringence. In Sec. III our experimental methods are outlined. In Sec. IV the experimental data are summarized and shown to be in quantitative agreement with theoretical predictions. In particular, we report for the first time a novel zero-crossing effect for an inclined-incident pump beam. A number of potential applications of the observed effects will also be discussed.

II. THEORY

Based on a continuum model of the liquid crystal, previous workers^{6,8} have derived theory for many nonlinearoptical processes in a NLC film. We have in general followed these derivations, except that the effect of a quasistatic electric field has been taken into account. For simplicity, a homeotropically aligned NLC film of thickness *d* is considered. Its unperturbed director \hat{n}_0 is along the *z* axis. This is illustrated in Fig. 1. The incident light beam is in the (x,z) plane with the wave vector **k** making an angle β with the *z* axis. The quasistatic electric field is also biased along the *z* axis. Under the action of the applied fields, the angle between **k** and the local director $\hat{n}(\mathbf{r})$ is



FIG. 1. Configuration for a homeotropically aligned nematic liquid crystal under the action of a quasistatic electric field and a light beam at inclined incidence.

given by $\psi = \beta + \theta$, where θ is the reorientation angle of the director, i.e., the angle between $\hat{\mathbf{n}}(\mathbf{r})$ and $\hat{\mathbf{z}}$ at z. Thus $n_x = \sin\theta(z)$, $n_y = 0$, and $n_z = \cos\theta(z)$.

Wave propagation in the NLC film is governed by Maxwell's equations. An incident light field would experience an effective dielectric constant,

$$\epsilon_{\rm eff} = \frac{\epsilon_{\perp} \epsilon_{\parallel}}{\epsilon_{\perp} \cos^2 \psi + \epsilon_{\parallel} \sin^2 \psi} , \qquad (1)$$

where $\epsilon_{||}$ and ϵ_{\perp} are dielectric constants along and perpendicular to the director, respectively. Equation (1) is field dependent due to field-induced reorientation of the molecules. The angle $\theta(z)$ can be calculated as usual by minimizing the total free energy of the sample, $F = \int \mathcal{F} dv$. The free energy density \mathcal{F} is given by

$$\frac{K}{2} \{ [\nabla \cdot \hat{\mathbf{n}}(\mathbf{r})]^2 + [\nabla \times \hat{\mathbf{n}}(\mathbf{r})]^2 \} - \frac{\Delta \epsilon_{\rm ac}}{8\pi} [\mathbf{E}_{\rm ac} \cdot \hat{\mathbf{n}}(\mathbf{r})]^2 - \frac{\Delta \epsilon_{\rm ac}}{8\pi} [\mathbf{E}_{\rm ac} \cdot \hat{\mathbf{n}}(\mathbf{r})]^2 = \frac{K}{2} \left[\frac{d\theta}{dz} \right]^2 - \frac{\Delta \epsilon_{\rm op}}{8\pi} E_{\rm op}^2 \sin^2 \psi - \frac{\Delta \epsilon_{\rm ac}}{8\pi} E_{\rm ac}^2 \cos^2 \theta .$$
(2)

In Eq. (2), K is the elastic constant of the NLC in the socalled one-constant approximation;⁴ E_{ac} and E_{op} are quasistatic and optical electric fields inside the sample; and $\Delta \epsilon_{ac}$ and $\Delta \epsilon_{op}$ are the dielectric anisotropies, $\Delta \epsilon = \epsilon_{||} - \epsilon_{\perp}$, due to the quasistatic electric and optical fields, respectively. This minimization procedure yields the usual Euler-Lagrange equation, which can be rewritten as

$$\left(\frac{d\theta}{dz}\right)^2 = C - 2(A\sin^2\psi + B\sin^2\theta), \qquad (3)$$

where

$$A = (\Delta \epsilon_{\rm op} / 8\pi K) E_{\rm op}^2, \quad B = -(\Delta \epsilon_{\rm ac} / 8\pi K) E_{\rm ac}^2,$$
$$C = 2[A \sin^2(\beta + \theta_m) + B \sin^2 \theta_m],$$

and θ_m is the maximum reorientation angle at the center of the sample cell. With the boundary condition $\theta = 0^\circ$ at z = 0 and z = d, integration of Eq. (3) in the region 0 < z < d/2 yields

$$z \left[2A \sin^2(\beta + \theta_m) + 2B \sin^2\theta_m \right]^{1/2}$$

= $\int_0^{\theta(z)} \left[1 - \frac{A \sin^2(\beta + \theta') + B \sin^2\theta'}{A \sin^2(\beta + \theta_m) + B \sin^2\theta_m} \right]^{-1/2} d\theta' .$
(4)

The value of θ_m is obtained by setting $\theta = \theta_m$ at z = d/2 in Eq. (4).

Typically, numerical integration of Eq. (4) is required for solving $\theta(z)$. The effect of superposed quasistatic electric and optical fields on molecular reorientation is most evident by examining Eq. (3) in the limit of weak fields, i.e., minor distortion of the director. In this limit, Eq. (3) can be rewritten as

$$2\xi^{2}\frac{d^{2}\theta}{dz^{2}} + \left[2\cos(2\beta) - 2\left[\frac{\Delta\epsilon_{ac}}{\Delta\epsilon_{op}}\right]\left[\frac{E_{ac}}{E_{op}}\right]^{2}\right]\theta + \sin(2\beta) = 0, \quad (5)$$

where $\xi^2 = 4\pi K / \Delta \epsilon_{\rm op} E_{\rm op}^2$. Let us define

$$\phi \equiv \left[2\cos(2\beta) - 2\left(\frac{\Delta\epsilon_{\rm ac}}{\Delta\epsilon_{\rm op}}\right) \left(\frac{E_{\rm ac}}{E_{\rm op}}\right)^2 \right] \theta + \sin(2\beta) \ . \tag{6}$$

Equation (5) becomes

$$(\xi')^2 \frac{d^2 \phi}{dz^2} + \phi = 0 , \qquad (7)$$

where

$$(\xi')^2 = 2\xi^2 / [2\cos(2\beta) - \Delta\epsilon_{\rm ac} E_{\rm ac}^2 / \Delta\epsilon_{\rm op} E_{\rm op}^2]$$

The solution of Eq. (5), subject to the boundary condition $\theta = 0^{\circ}$ at z = 0 and z = d, is

$$\phi = \sin(2\beta) \left| \cos(z/\xi') + \frac{1 - \cos(d/\xi')}{\sin(d/\xi')} \sin(z/\xi') \right|.$$
(8)

In the limit of weak fields, i.e., $E_{\rm ac} \ll E_{\rm op} \ll E_c$, where E_c is the critical field for the Fréedericksz transition, one can easily show that $z/\xi' < d/\xi' \ll 1$. The reorientation angle $\theta(z)$ can be obtained by using Eqs. (5) and (8) upon expansion of the sine and cosine terms:

$$\theta(z) \sim \frac{\sin(2\beta)}{4\xi^2} \left[\frac{z^4}{12(\xi')^2} - \frac{z^3 d}{6(\xi')^2} - z^2 + zd \right]$$

= $\frac{\Delta\epsilon_{\text{op}} \sin(2\beta)}{16\pi K} E_{\text{op}}^2 \left[(zd - z^2) + \left[-\frac{z^3 d}{6} + \frac{z^4}{12} + \frac{zd^3}{12} \right] \left[\frac{2\cos(2\beta)\Delta\epsilon_{\text{op}} E_{\text{op}}^2 - 2\Delta\epsilon_{\text{ac}} E_{\text{ac}}^2}{8\pi K} \right] \right]$ (9)

and

$$\theta_m \sim \frac{\Delta \epsilon_{\rm op} d^2 \sin(2\beta)}{64\pi K} E_{\rm op}^2 \ . \tag{10}$$

Similar expressions can be derived for $E_{op} \ll E_{ac} \ll E_c$.

Both terms enclosed in the large parentheses in Eq. (9) are positive. For the sample we investigated, MBBA [*p*-methoxy-benzylidene-*p*-(*n*-butyl)aniline], $\Delta \epsilon_{op} > 0$, whereas $\Delta \epsilon_{ac} < 0$. Thus the quasistatic electric field should help reorient the director toward the direction of polarization of the light field.

The effective dielectric constant becomes

$$\boldsymbol{\epsilon}_{\text{eff}} \sim \boldsymbol{\epsilon}_{\perp} \left[1 + \frac{\Delta \boldsymbol{\epsilon}_{\text{op}}}{\boldsymbol{\epsilon}_{\parallel}} \boldsymbol{\beta}^2 \right] + \delta \boldsymbol{\epsilon}(\boldsymbol{\theta}) \tag{11}$$

and

$$\delta \epsilon(\theta) = 2\epsilon_{\perp} \Delta \epsilon_{\rm op} \beta \theta(z) , \qquad (12)$$

where $\theta(z)$ is given by Eq. (9) in the weak-field limit. $\delta \epsilon(\theta)$ gives rise to nonlinear-optical propagation effects through self-phase modulation of the laser beam traversing the NLC film. Since $\delta \epsilon(\theta) > 0$, self-focusing results for an incident beam with a typical Gaussian profile. At higher laser intensities $(I \ge 1000 \text{ W/cm}^2 \text{ for a } 75\text{-}\mu\text{m}\text{-}$ thick nematic MBBA sample),⁶ the director reorientation can induce phase changes greater than 2π across the beam profile and far-field diffraction rings can occur. In a first approximation, the number of rings observed can be estimated by maximum phase change across the beam profile, $N \simeq (\text{phase change})/2\pi$. It is not immediately obvious, however, that applying a quasistatic electric field would increase the number of laser-induced rings because such a field alone only produces a uniform phase change across the laser-beam profile. Equation (9) does indicate a nonlinear coupling between the two fields. It is also expected through ECB data that phase changes should show a typical critical behavior and increase dramatically just above the quasistatic-electric-field-induced Fréedericksz transition. Thus quasistatic-electric-field-enhanced optical diffraction effects can be explained by a combination of the above two effects. Field-induced birefringence and hence $\theta(z)$ can be measured by a normally incident (along the z axis) probe beam through the beam center of the pump beam. Its extraordinary component then experiences a phase shift with respect to the ordinary component:

$$\delta = 2 \times \frac{2\pi}{\lambda_p} \int_0^{d/2} \left[\frac{n_0 n_e}{(n_0^2 \cos^2 \theta + n_e^2 \sin^2 \theta)^{1/2}} - n_0 \right] dz ,$$
(13)

where n_0 and n_e are, respectively, the ordinary and maximum extraordinary refractive indices of the NLC. λ_p is the wavelength of the probe laser. The factor 2 is due to symmetry of the sample. For $E_{\rm ac} \ll E_{\rm op} \ll E_c$,

$$\delta \sim \frac{\pi n_0 \Delta \epsilon_{\rm op}}{\lambda_p \epsilon_{||}} \left[\frac{\Delta \epsilon_{\rm op} \sin(2\beta)}{16\pi K} E_{\rm op} \right]^2 \\ \times \left[\frac{1}{30} d^5 + \frac{17}{2520} d^7 \left[\frac{1}{(\xi')^2} \right] \\ + \frac{31}{630} d^9 \left[\frac{1}{12(\xi')^2} \right]^2 \right].$$
(14)

III. EXPERIMENTAL

A block diagram of our experimental apparatus is shown in Fig. 2. MBBA films of 68, 90, and 138 μ m thick maintained at 23 ± 0.1 °C are used. They are sandwiched between glass windows coated first with indium oxide as transparent electrodes and then obliquely evaporated with SiO to prevent $E_{\rm ac}$ -induced space charges in the nematic medium. Finally, the cell windows are treated with octadecyldimethyl[3-(trimethoxysilyl)-propyl] ammoniumchloride (DMOAP) for homeotropic alignment. The alignment is checked with conoscopy. It is found that the samples show a pretilt of 2°-3° at most. A quasistatic electric field at 1 kHz is applied normal to the glass window. An unfocused Ar⁺ laser (beam diameter ~2.25 mm) incident at 30° provides the pump beam.

The field-induced birefringence at the pump-beam center is measured by a tightly focused He-Ne probe laser using a modulation technique originally devised by Lim and Ho.²¹ Briefly, the probe beam is split and the main measurement beam passes through, successively, the sample, a $\lambda/4$ plate, and a rotating polaroid before being detected by photodiode A. The projection of E_{op} on the same plane is at an angle of 45° to the probe laser polarization direction. The axis of the $\lambda/4$ plate is aligned with the probe laser polarization direction. A polaroid is



FIG. 2. Block diagram of the experimental setup.

IV. RESULTS AND DISCUSSION

Through quasistatic-electric-field-induced birefringence measurements, it is determined that the Fréedericksz transition critical potential $V_{\rm th} = 3.72 \pm 0.04$ V. From the expression $V_{\rm th} = \pi (4\pi K / |\Delta\epsilon|)^{1/2}$ and $\Delta\epsilon = -0.5$ for MBBA, we obtain $K = 6.2 \times 10^{-7}$ dyn. This is in good agreement with values for K_1 and K_3 in the literature.¹ The phase change due to the quasistatic electric field only and that due to the superposed fields at a pump-laser intensity of ~10 W/cm² (400 mW) are illustrated for the region around the critical field in Fig. 3. At this intensity, Eq. (14) predicts a phase change of 1.28° without $E_{\rm ac}$ and 2.36° at 1 V. These are in good agreement with experimentally measured values (not shown in the figure) of 1°±0.3° and 2.7°±0.4°, respectively. For the above calculation, we have taken into account the fact that the ray direction is different from **k**. Using the refractive index data of MBBA (Ref. 22) and the relation

$$\epsilon_{\parallel} \tan\beta' = \epsilon_{\perp} \tan\beta , \qquad (15)$$

we obtain the actual angle of incidence inside the sample, $\beta' - 14.2^{\circ}$.



FIG. 3. ECB and superposed quasistatic-electric- and optical-field-induced birefringence in the Fréedericksz transition region. The solid curves are drawn freehand as a guide to the eye.



FIG. 4. Theoretical fit of the superposed-field-induced birefringence data as a function of V_{ac} .



FIG. 5. Maximum phase change across the pump-beam profile as a function of pump-laser intensity, with and without $V_{\rm ac}$ at 4 V. The solid curve is drawn freehand as a guide to the eye.

The difference in phase change with and without the optical field increases dramatically to 210° at 3.8 V, just above the quasistatic-electric-field-induced Fréedericksz transition critical field, illustrating the strong enhancement in the transition region (Fig. 3). To compare with theory, we numerically solved Eq. (4) and substituted into Eq. (13). This is drawn as the solid curve in Fig. 4, which fits the experimentally measured birefringence quite well.

In Fig. 5 we have plotted the maximum phase difference across the pump-beam profile, $\Delta \delta$, as a function of pump-laser power. As we mentioned earlier, there is a nonlinear coupling between \mathbf{E}_{op} and \mathbf{E}_{ac} . This is also expected to enhance the phase difference besides the critical behavior of the nematics. In particular, the nonlinear coupling would cause the enhancement to be nonuniform because of the Gaussian beam profile. For example, $\Delta \delta$ increases from 2.8° at 400 mW (~10 W/cm²) to 192° when a superposed quasistatic electric potential of 4 V is applied. This physical picture is shown in the inset of Fig. 5. An enhanced phase difference of multiples of π , presenting itself as alternating bright and dark rings in the far-field pattern, could thus be realized with superposed optical and quasistatic electric fields.

Figure 6 shows the number of far-field rings of a 90- μ m-thick sample as a function of pump-laser intensity with \mathbf{E}_{op} only and with a superposed quasistatic electric potential of 4 V. Photographs of the far-field pattern for a 138- μ m-thick sample are shown in Fig. 7. As many as 12 rings are observed for $V_{ac} = 4.1$ V and a pump-laser intensity as low as 600 W/cm². This is a six-fold enhancement in the number of diffraction rings compared with



FIG. 6. Number of far-field rings for a $90-\mu$ m-thick sample is plotted as a function of pump-laser intensity. The solid circles show data with a biased quasistatic electric potential of 4.1 V. The solid curves are drawn freehand as a guide to the eye.

the case without the quasistatic electric field. No ring has been observed at high electric potential (e.g., $V_{ac} \ge 5$ V) and a very low laser intensity of 5 W/cm². This verifies that the far-field pattern shown in Fig. 7 is not due to diffraction by domains.²³

It is expected that the enhancement effect due to director reorientation in the nematics by the superposed fields should increase drastically just above the Fréedericksz transition critical field and then gradually saturate to a maximum value at high field strengths. To examine this, we have subtracted ECB from superposed-field-induced birefringence for four different pump intensities and plotted the phase difference $\Delta\delta$ as a function of biased voltage $V_{\rm ac}$ in Fig. 8. It can be seen that $\Delta\delta$ reaches a maximum value at 3.9±0.2 V, just above the Fréedericksz critical field. This corresponds to the point where $d\theta/dV_{\rm ac}$ or $d\delta/dV_{\rm ac}$ is a maximum. With further increases in $V_{\rm ac}$, the gradients decrease. The phase difference $\Delta\delta$, however, does not approach a saturated value as expected. In fact, the $\Delta\delta$ vs $V_{\rm ac}$ curve goes through zero and then becomes negative for $V_{\rm ac}$ larger than 5 V. We also note that the field strength for which zero crossing occurs is independent of the pump-laser intensity.

This novel phenomenon can be understood by examin-







(b)

FIG. 7. Far-field pattern of a $138-\mu$ m-thick sample at a pump-laser intensity of 600 W/cm², (a) with and (b) without a superposed quasistatic electric potential of 4 V.



FIG. 8. Difference of superposed-field-induced birefringence and ECB as a function of biased voltage for a $138-\mu$ m-thick sample at four different pump-laser intensities. The solid curves are drawn freehand as a guide to the eye.

ing the local picture of a director under the action of the applied fields. The geometry has been shown in Fig. 1. Because $\Delta \epsilon_{op} > 0$, the optical field \mathbf{E}_{op} will always reorient the director toward the direction of polarization. If the director reorientation caused by \mathbf{E}_{ac} , $\theta_{ac}(z)$, is less than $(\pi/2) - \beta$, the two effects will enhance each other, and $\Delta\delta$ is positive. When the quasistatic electric field reorients the director such that it is exactly aligned with \mathbf{E}_{op} , the optical field can reorient the director no further and hence $\Delta \delta = 0$. The negative coupling case, $\Delta \delta < 0$, corresponds to $\theta_{ac}(z) > (\pi/2) - \beta$. The senses of reorientation of the director for \mathbf{E}_{op} and \mathbf{E}_{ac} are opposite to each other in this regime. It is also evident from the above discussions that the zero-crossing effect is peculiar to the inclinedincidence geometry. For a normally incident pump beam, i.e., $\beta = 0$, the effect will always be enhancement. Because the direction of director reorientation is independent of the pump-laser intensity, the zero-crossing potential should be likewise.

The liquid-crystal cells investigated are of finite thickness. For a given quasistatic electric field strength, the effect of the applied field on the local director along the beam path could be positively coupled, noncoupled, or negatively coupled. Thus the zero-crossing effect manifested by the sample as a whole is the net result of the local contributions along the beam.

Our diffraction experiments also correlate with the birefringence experiments discussed above. The number



FIG. 9. Number of far-field rings at a pump-laser intensity of 600 W/cm² for 90- and 138- μ m-thick samples are plotted as a function of bias voltage $V_{\rm ac}$. The solid curves are drawn free-hand as a guide to the eye.

of far-field rings at $I \simeq 600 \text{ W/cm}^2$ is plotted for samples 90- and 138- μ m thick as a function of V_{ac} in Fig. 9. Each of these curves exhibits a peak corresponding to $(d\theta/dV_{ac})_{max}$. The zero-crossing and negative coupling effects manifest themselves as decreasing ring numbers at higher voltages.

Finally, we briefly comment on the heating effect. MBBA is not transparent at the pump-laser wavelength. The molecules can absorb light, and the birefringence of the NLC film would be changed because of the temperature-dependent indices of refraction. It is estimated that the temperature of sample does not rise by more than 2° during the experiment. We have therefore neglected the heating effect. The long-term degradation of the samples due to heating was avoided, however, by reducing as much as possible the irradiating time of the liquid crystal by the pump laser.

V. CONCLUSIONS

We have investigated both theoretically and experimentally quasistatic-electric- and optical-field-induced director reorientation and birefringence in a NLC film. These studies help shed light on nonlinear-optical propagation effects such as far-field diffraction rings. It is found that a properly biased quasistatic electric field can enhance diffraction effects in a NLC film. The enhancement effects are attributed to the critical behavior of the NLC film just above the quasistatic-electric-field-induced Fréedericksz transition and nonlinear coupling between the optical and quasistatic electric fields. Experimental results are in quantitative agreement with theoretical predictions.

For an inclined-incident pump beam, the enhancement does not saturate at a high-bias field. On the contrary, birefringence measurements show that the optical-fieldinduced phase difference with a superposed quasistatic electric field goes from positive through zero to negative. Maximum enhancement occurs just above the quasistatic-electric-field-enhanced Fréedericksz transition critical field, at a point where $d\theta/dV_{ac}$ is maximum. Locally, the zero-crossing phenomenon occurs at a quasistatic electric field strength for which the NLC director has been reoriented to an exact alignment with the polarization direction of the optical field. The macroscopic zero-crossing effect manifested by the NLC film as a whole is the net result of the applied fields on the local directors along the beam path.

There are a number of potential applications for the observed effects. First of all, the number of laser-induced rings, i.e., beam divergences of the laser, can be controlled by a properly biased NLC cell. The novel zero-crossing effect can be viewed as stabilization and maintenance of ECB by an inclined-incident light beam. One could also envision fine adjustment of ECB utilizing this effect. Our results are also of interest for the enhancement of nonlinear-optical effects such as optical bistability and wave-front conjugation in NLC cells by external fields.

ACKNOWLEDGMENT

This work was partially supported by a grant from the National Science Council, Republic of China.

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(a)



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FIG. 7. Far-field pattern of a 138- μ m-thick sample at a pump-laser intensity of 600 W/cm², (a) with and (b) without a superposed quasistatic electric potential of 4 V.