Singular Behavior of Light-Induced Space Charge in Photorefractive Media under an ac Field

G. F. Calvo, B. Sturman, F. Agulló-López, and M. Carrascosa
Departamento de Física de Materiales, C-IV, Universidad Autónoma de Madrid, Cantoblanco, E-28049 Madrid, Spain
(Received 17 December 1999)

We show that a photoconductive crystal, placed in a rapidly alternating ac field and exposed to nonuniform light, exhibits singularities of the induced space charge and discontinuities of the corresponding space-charge field. The singularities appear at the local intensity maxima when the curvature of the intensity profile exceeds a certain (often very low) threshold value. We analyze the characteristic features of the singular ac response and consider its possible optical manifestations.

PACS numbers: 42.65.Hw, 42.70.Nq

Charge separation owing to the generation of free carriers is the main ingredient of the photorefractive nonlinearity inherent in many semiconductors and dielectrics [1,2]. Most photorefractive phenomena (spatial amplification, phase conjugation, nonlinear scattering, etc.) involve the buildup of space-charge field gratings in response to interfering light beams and diffraction by these gratings via the linear electro-optic effect. Diffusion of photoexcited carriers was initially the only mechanism capable of producing a field profile proportional to the light intensity gradient (gradient response) which is necessary for many applications [1–3]. Unfortunately, the gradient response caused by diffusion is often insufficiently high for practical purposes.

In 1985, an alternating current (ac) technique was proposed in order to enhance the photorefractive response of high-speed materials like the sillenites ($Bi_{12}SiO_{20}$, $Bi_{12}TiO_{20}$, $Bi_{12}GeO_{20}$), semiconductors GaAs, CdTe, etc. [3,4]. This method requires a strong rapidly alternating (often as a square wave) electric field and serves currently as one of the main tools for shaping the photorefractive nonlinearity. Within the linear approximation in the light modulation, it also gives a gradient response which is, however, much greater than that caused by diffusion.

Although it was found [5–7] that the linear approximation in the ac case is restricted to values of the light modulation considerably smaller than 1, the concept of gradient response has been overwhelmingly used for the arrangement and interpretation of a large number of ac-photorefractive experiments; see, e.g., [2,8–10] and references therein. These experiments deal not only with the recording of spatial gratings but also with nonlinear beam propagation [9–11], which has become topical during recent years in connection with photorefractive solitons [2,12] and possible applications [13,14]. In the case of beam propagation, the light contrast, defined as the ratio between the maximum signal and background intensities, is typically much greater than 1. The effects of material nonlinearity lying outside the linear approximation can become decisive in this region. Previous theoretical attempts [5,6,15] to enter the nonlinear domain have shown some modification of the ac response but they have not found its new general qualitative features.

The purpose of this Letter is to reveal new characteristic features of the ac response beyond the small contrast approximation. We show analytically and numerically that above a fairly low threshold for the intensity profile curvature, the main feature of this response is the occurrence of very strong and narrow peaks of the light-induced charge density. These charge singularities, coupled with discontinuities of the space-charge field, arise at the local intensity maxima. Their width is defined by an internal characteristic length of the material which is much smaller than the spatial scale of the intensity variations. We also consider possible optical manifestations of the singular ac response.

Our starting point is the standard one-trap model for electron transport that is justified for the sillenites and useful for many semiconductors. Within this model, the space-charge field E, the concentration of donors N^+, and the free electron density n obey in the 1D case the equations [1,2]

$$E_z = \frac{q}{\varepsilon\varepsilon_0}(N^+ - N_A), \tag{1a}$$

$$N_t^+ = s_i I(N_D - N^+) - s_r n N^+ \tag{1b}$$

$$= -\mu\left[n(E_{\mathrm{ex}} + E) + \frac{k_B T}{q} n_z\right]_z. \tag{1c}$$

Here the subscripts t and z denote differentiations with respect to time and the spatial coordinate, I is the light intensity, E_{ex} the applied field, q the elementary charge, $\varepsilon\varepsilon_0$ the dielectric constant, μ the electron mobility, T the absolute temperature, k_B the Boltzmann constant, s_i and s_r are the ionization and recombination constants, and N_D and N_A the concentrations of donors and acceptors.

We have used the standard low-intensity approximation exploiting the smallness of the electron lifetime, $\tau = 1/s_r N_A$, in comparison with the dielectric relaxation time $t_d \propto I^{-1}$. We suppose also that $N_A \ll N_D$, which is the case for the sillenites (where $N_A/N_D \approx 10^{-3}$) and for many other crystals [3]. In what follows, we also take into account the dark excitation of electrons by assuming a small effective background illumination.

Let us assume further that $E_{ex} = E_0 p(t)$, where E_0 is the amplitude of the ac field, and $p(t) = \pm 1$ is a periodic function with a zero average value, $\langle p(t) \rangle = 0$, and a period T much smaller than t_d. Correspondingly, we set $E = E_0(e + \tilde{e})$, where e and $\tilde{e}$ are the slow and fast dimensionless components, such that $\langle \tilde{e} \rangle = 0$, $\langle e \rangle = e$, and $|\tilde{e}| \ll |e|$.

Our aim now is to obtain a closed equation for e by averaging over the fast oscillations. The whole derivation procedure is as follows: First, we reduce algebraically Eqs. (1) to a single nonlinear differential equation for E. Then, taking the average, we obtain in the leading approximation in T/t_d a relation between e and $\langle p\tilde{e}_{zt} \rangle$. Neglecting provisionally the diffusion contribution to the electric current density [the last term in Eq. (1c)], we represent this relation as

$$\langle p\tilde{e}_{zt} \rangle = \frac{q s_i N_D I}{\varepsilon \varepsilon_0 E_0} \, e \, . \tag{2}$$

Multiplying the nonlinear equation for E by $p(t)$ and repeating the averaging, we arrive at another relation between e and $\langle p\tilde{e}_{zt} \rangle$. Finally, using Eq. (2), we obtain in steady state,

$$\left[\frac{I(1 - e^2)}{1 + \nu e_\xi} \right]_\xi + eI = 0 \, , \tag{3}$$

where $\xi = z/l_0$ is the normalized coordinate, $l_0 = \mu\tau E_0$ the drift length, and $\nu = \varepsilon\varepsilon_0/q\mu\tau N_A$ a characteristic dimensionless parameter.

Equation (3) is worthy of attention. It is a second-order differential equation with two different nonlinear terms. The term e^2 originates from the drift nonlinearity [product nE in Eq. (1c)], whereas the term νe_ξ in the denominator (equal to $\delta N^+/N_A$, where $\delta N^+ = N^+ - N_A$ is the light-induced charge density) comes from the product nN^+ in Eq. (1b) and describes trap saturation. This term can be larger than 1 but it is assumed to be much smaller than N_D/N_A. In other words, we consider possible saturation of acceptors but not of donors. The field $e(z)$ is obviously invariant to scaling of the intensity profile $I(z)$.

Another important observation is that $\nu = 1/4Q_{\max}^2$, where $Q_{\max}$ is the maximum value of the quality factor for the space-charge waves [7]. The resonant linear and nonlinear phenomena related to these waves and the literature data on N_A and $\mu\tau$ give evidence that $Q_{\max} \gg 1$ and $\nu \lesssim 10^{-2}$ in the sillenites and many semiconductors [2,3,7]. This means that the term νe_ξ in Eq. (3) is important only when $e(z)$ changes considerably within the saturation length $l_s = \nu l_0 = \varepsilon\varepsilon_0 E_0/qN_A \ll l_0$. In the whole region where $\nu e_\xi \ll 1$ [i.e., $e(z)$ is a smooth function] the field profile obeys the nonlinear first-order equation

$$2e_\xi = 1 + I^{-1} I_\xi (e^{-1} - e) \, . \tag{4}$$

Equation (4) carries no information about the traps and implies only a linear recombination for photocarriers and their drift in the electric field with mobility μ.

As follows from Eqs. (3) and (4), an even intensity distribution $I(z)$ corresponds to an odd distribution of $e(z)$. This symmetry property is compatible with the concept of a gradient response.

The small contrast approximation means for Eqs. (3) and (4) that $e \simeq -I_\xi/I \ll 1$. It is in agreement with the formulas for the ac response obtained within the linear approximation [3,4].

Let us return to the general nonlinear case described by Eq. (3) and consider two important particular cases, (a) and (b). They correspond to Gaussian and periodic intensity profiles that are specified by the relations

$$I \propto 1 + f_0 \exp(-4z^2/d^2) \, , \tag{5a}$$

$$I \propto 1 + m \cos(2\pi z/\Lambda) \, . \tag{5b}$$

The values of f_0 relevant to experiment are much greater than 1 (a weak background excitation) whereas the modulation of the interference light pattern, m, is less than 1. In case (a) we have $e(\pm\infty) = 0$ as the boundary conditions for Eq. (3); they can be replaced by $e(0) = e(\infty) = 0$ in view of the symmetry properties. In case (b) we can use the boundary conditions $e(0) = e(\Lambda/2) = 0$.

For our numerical calculations we accept the following parameters representative for experiments with $Bi_{12}SiO_{20}$ crystals: $N_A = 10^{16}$ cm^{-3}, $N_D = 10^{19}$ cm^{-3}, $\varepsilon = 56$, $\mu\tau = 4 \times 10^{-7}$ cm^2/V, and $E_0 = 25$ kV/cm. They correspond to $l_0 = 60$ μm, $l_s \simeq 0.4$ μm, and $\nu \simeq 7 \times 10^{-3}$. Comparable values of ν are typical of other sillenites.

Figure 1 shows the distribution $e(z)$ induced by a Gaussian beam of width $d = 0.6 l_0$ for several values of f_0. For $f_0 \gtrsim 0.5$ the field profile is characterized by a very pronounced discontinuity at $z = 0$. Outside the narrow discontinuity region, the function $e(z)$ obeys the first-order Eq. (4). Its broad maximum occurs far from the discontinuity; for $f_0 \gtrsim 0.5$, an increase of f_0 shifts the position of the maximum to the right and the corresponding maximum field $e_{\max}(f_0)$ saturates, approaching 1. Only for $|z|/d \gtrsim 1.5$, where $|e| \ll 1$, the shown field profiles

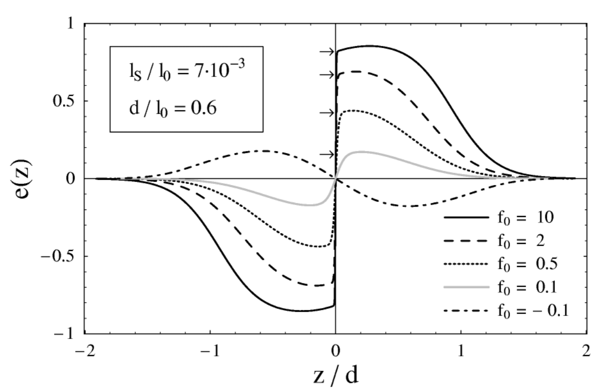

FIG. 1. Normalized space-charge field $e(z)$ for $d/l_0 = 0.6$ and $f_0 = -0.1$, 0.1, 0.5, 2, and 10. The arrows indicate the corresponding values of $e(0)$ for Eq. (4).

correspond to the low-contrast approximation. The arrows in Fig. 1 indicate the values of $e(0)$ for the first-order Eq. (4). For $f_0 \gtrsim 0.5$ the ratio $e(0)/e_{\max} \simeq$ const whereas for $f_0 \lesssim 0.1$, it becomes smaller and the discontinuity less and less pronounced. Negative values of f_0 give no discontinuity of $e(z)$. The steepening of the field profile is a nonlinear feature that appeared in some previous numerical calculations [6,15].

The narrow central peak and the broad symmetric profile in Fig. 2 illustrate the relationship between the charge and intensity distributions for $d/l_0 = 0.6$ and $f_0 = 10$. The size of the charge singularity is dramatically much smaller than the beam width. The dotted shifted peak shows the charge singularity induced by a slightly Gaussian-like asymmetric beam (the dotted broad profile) that has its maximum at $z = 0$ and the light intensity weight center $w_c = \int xI(x)\,dx / \int I(x)\,dx$ at $z/d = w_c \simeq -0.19$. We see that the singularity is pinned exactly to the intensity maximum only in the symmetric case. In the case of a slightly asymmetric profile, it moves towards $z/d = w_c$.

The central part of the charge distribution induced by a Gaussian beam is depicted in Fig. 3 for several values of f_0. One sees that the size of the core is of the order of l_s. The singularity is not only very narrow but it is also very strong. One sees that the acceptors become saturated already for $f_0 \simeq 2$. The wide wings of the charge profile, where $|\delta N^+/N_A| \ll 1$, cannot be resolved in the scale of Fig. 3.

Now we touch on the influence of the beam width on the above features. Decreasing d/l_0 makes the singular behavior more pronounced. On the other hand, increasing d/l_0 results in decreasing $e(0)/e_{\max}$, i.e., shifts the singular behavior towards large beam amplitudes. For $d/l_0 \gtrsim 6$ our numerical calculations have shown no singular behavior.

Let us turn now to the grating recording [case (b)]. Figure 4 exhibits the field distribution $e(z)$ produced by a periodic light pattern of a period $\Lambda = 0.4 l_0$ for several values of m. It has the same discontinuity up to very low modulations. For $\Lambda/l_0 \gtrsim 8$ the singularity disappears.

The results found become more clear if we look at proper physical analogies. The essence of Eq. (3) is the presence of a small coefficient ν before the highest-order (second) derivative. Such a situation is typical, e.g., for the shock waves described by Burgers equation [16], where a narrow wave front is controlled by a small viscosity, and for the domain walls in the phase transition theory [17].

Furthermore, the inevitability of the singular behavior follows directly from Eq. (4). Let us assume that the singularity is absent. Then this first-order differential equation possesses (for an arbitrary smooth intensity profile) a continuous solution that simultaneously meets two boundary conditions. This is impossible in general.

To make our assertion more specific, we obtain a criterion for the existence of a linear solution, $e(\xi) = e'(0)\xi$, of Eq. (4) at the origin for the Gaussian light profile given by Eq. (5a). By equalizing the zero-power terms (in ξ) on the left- and right-hand sides of Eq. (4), we arrive at a quadratic equation for $e'(0)$ that has real solutions only if $f_0 < f_0^{\text{th}}$, where

$$f_0^{\text{th}} = (64 l_0^2 d^{-2} - 1)^{-1}. \quad (6)$$

Above the threshold, any solution of Eq. (4) vanishing at infinity has to be nonzero at $z = 0$. Note that for $d \ll 8l_0$ the threshold amplitude f_0^{th} becomes very small whereas for $d > 8l_0$ a linear solution in the vicinity of zero exists for any f_0.

The found criterion can be generalized for an arbitrary smooth intensity distribution. If we write $I \propto (1 - z^2/2R^2)$ in the vicinity of a maximum, the generalized criterion for the radius of curvature of the intensity profile is $R < R_{\text{th}} = 2\sqrt{2}\,l_0$.

Our numerical calculations have shown that decreasing R from R_{th} to $R_{\text{th}}/2$ is accompanied by an increase of $e(0)$ from zero to a value comparable with $e_{\max}$, which corresponds to a developed singularity.

The fine structure of the discontinuity is described by the second-order Eq. (3) if we put $I(\xi) =$ const in the vicinity of $z = 0$. Then we find

$$e(z) \simeq c \tanh[cz/l_s(1 - c^2)], \quad (7)$$

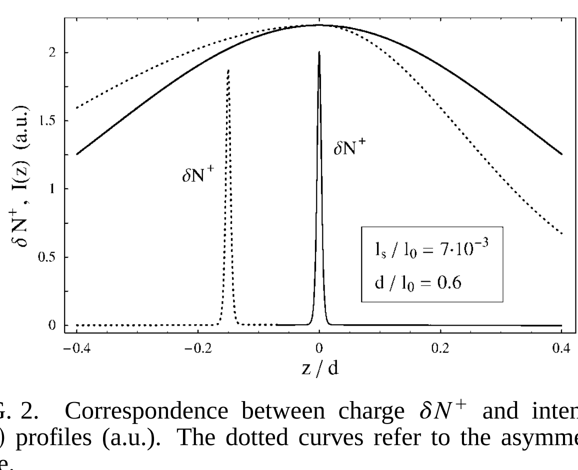

FIG. 2. Correspondence between charge δN^+ and intensity $I(z)$ profiles (a.u.). The dotted curves refer to the asymmetric case.

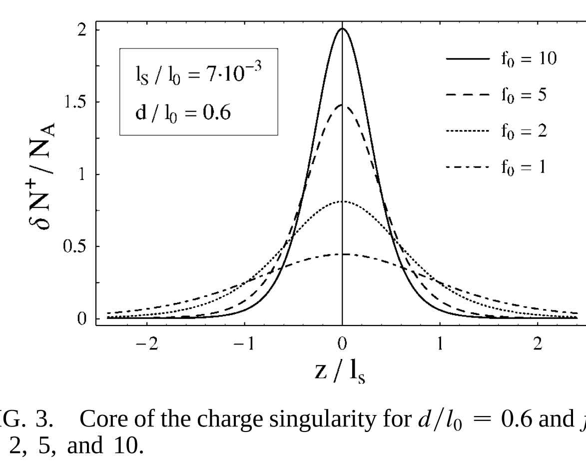

FIG. 3. Core of the charge singularity for $d/l_0 = 0.6$ and $f_0 = 1$, 2, 5, and 10.

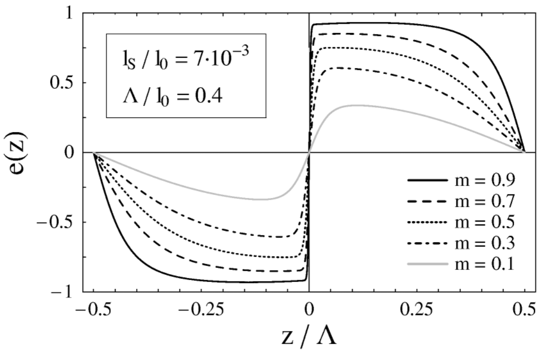

FIG. 4. Normalized space-charge field $e(z)$ for the grating recording; the period is $\Lambda = 0.4l_0$ and the modulation is $m = 0.1$, 0.3, 0.5, 0.7, and 0.9.

where c is an integration constant. Matching the singular and smooth components, we have $c \simeq e(0)$. Obviously, c grows with f_0 remaining smaller than 1. Correspondingly, the width of the singularity decreases with increasing f_0 and it becomes somewhat smaller than l_s. The described features are seen in Fig. 3.

Now we consider the role of the diffusion contribution to the current density, omitted when deriving Eq. (3). Retaining this contribution leads to the appearance of additional differential high-order terms on the right-hand side of Eq. (3). However, if the diffusion length, $l_D = (\mu\tau k_B T/q)^{1/2}$, is much smaller than $(l_0 l_s)^{1/2}$, i.e., $E_0 \gg (k_B T N_A/\varepsilon\varepsilon_0)^{1/2}$, these additional terms are negligible not only outside the singularity but even in its core. Using the accepted values of N_A and ε, we obtain the following numerical condition of validity of Eq. (3), $E_0 \gg 3$ kV/cm, that is always fulfilled in ac experiments.

The generation of charge singularities with the aid of large-size light beams is a new feature of the ac technique attractive for semiconductor and optical applications. This feature goes, in fact, beyond the scope of the photorefractive effect because it has nothing to do with the linear electro-optic effect. For photorefractives, the most interesting question concerns optical manifestations of the charge singularity. For applications based on weak optical nonlinearity (like displacement sensing [11,13]) the steepening of the field profile is a positive effect because it increases the sensitivity of measurements.

The influence of the singularities on nonlinear beam propagation in thick samples is a more challenging subject. Strong transverse phase gradients near the singularity can result in spatial instabilities of the wave front and lead to irregular light structures. A similar effect has been reported recently as "beam collapse" [10]. We expect also that the ac nonlinearity strongly modifies (because of high-intensity gradients) the speckle structure of laser irradiation. It is not excluded that the found ac response is compatible with localized solutions describing the trapping of light by the singularity and similar to the solitary solutions for the gradient photorefractive response [18].

In conclusion, we have shown that beyond the small-contrast limit the response of a photoconductive crystal, placed in a rapidly alternating ac field and exposed to nonuniform light, strongly differs from the expected diffusionlike response. The striking feature of the ac response is the presence of very narrow and strong charge singularities inseparable from discontinuities of the space-charge field at the local intensity maxima. This suggests new optical effects in photorefractive crystals.

This work was supported through project No. PB-98-0056. Also, G. F. C. acknowledges financial support from the UAM and B. S. thanks an IBERDROLA visiting professorship.

[1] *Photorefractive Materials and Their Applications*, edited by P. Günter and J. P. Huignard (Springer-Verlag, Berlin, 1988).
[2] L. Solymar, D. J. Webb, and A. Grunnet-Jepsen, *The Physics and Applications of Photorefractive Materials* (Clarendon Press, Oxford, 1996).
[3] M. P. Petrov, S. I. Stepanov, and A. V. Khomenko, *Photorefractive Crystals in Coherent Optical Systems* (Springer-Verlag, Berlin, 1991).
[4] S. I. Stepanov and M. P. Petrov, Opt. Commun. **53**, 292 (1985).
[5] J. E. Millerd *et al.*, J. Opt. Soc. Am. B **9**, 1449 (1992).
[6] G. A. Brost, J. Opt. Soc. Am. B **9**, 1454 (1992).
[7] B. I. Sturman *et al.*, J. Opt. Soc. Am. B **10**, 1919 (1993).
[8] A. A. Kamshilin, E. Raita, and A. V. Khomenko, J. Opt. Soc. Am. B **13**, 2536 (1996).
[9] A. V. Khomenko *et al.*, Opt. Commun. **150**, 175 (1998).
[10] C. A. Fuentes-Hernandez and A. V. Khomenko, Phys. Rev. Lett. **83**, 1143 (1999).
[11] A. A. Kamshilin *et al.*, Opt. Lett. **24**, 832 (1999).
[12] M. Shih, M. Segev, and G. Salamo, Phys. Rev. Lett. **78**, 2551 (1997).
[13] A. A. Kamshilin *et al.*, Appl. Phys. Lett. **83**, 1143 (1999).
[14] M. Sheik-Bahae *et al.*, IEEE J. Quantum Electron. **26**, 760 (1990).
[15] S. M. Shandarov *et al.*, Appl. Phys. B **68**, 1007 (1999).
[16] G. B. Whitham, *Linear and Nonlinear Waves* (Wiley-Interscience, New York, 1974), Chap. 4.
[17] M. E. Lines and A. M. Glass, *Principles and Applications of Ferroelectrics* (Claredon Press, Oxford, 1977), Chap. 4.
[18] D. N. Christodoulides and T. H. Coskin, Opt. Lett. **21**, 1460 (1996).