Photorefractive Saturable Kerr-Type Nonlinearity in Photovoltaic Crystals

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(Received 16 September 1996; revised manuscript received 18 March 1997)

We deduce the equation describing the refractive index perturbation in photovoltaic photorefractive crystals produced by the incidence of a focused laser beam and an incoherent uniform illumination. Under short-circuit conditions the equation shows a saturable Kerr-type nonlinearity that can be controlled by the intensity of the uniform background illumination. *Z*-scan experiments in an iron doped lithium niobate crystal are carried out using a 532 nm wavelength laser line to evaluate its self-lensing properties and to measure its photovoltaic field. [S0031-9007(97)03237-7]

PACS numbers: 42.65.Hw, 42.65.Jx, 42.65.Tg, 72.40.+w

The action of an optical beam on the refractive index and the self-trapping effects in photovoltaic (PV) photorefractive (PR) crystals were studied recently [1–3]. The theoretical analysis predicts a saturable Kerr-type nonlinearity in this kind of material where the refractive index perturbation in one dimensional form can be written as [1]

$$\Delta n_i = \Delta n_0 \frac{I}{I + I_d}, \tag{1}$$

where Δn_0 is the saturable refractive index perturbation. I is the focused beam irradiance and the saturable intensity I_d is the dark irradiance which is defined as the ratio of the thermal excitation rate to the photoexcitation cross section and can be considered as the irradiance necessary to produce a density of carriers equal to that in the dark. As in the case of PR screening solitons [4–7], this parameter plays an extremely important role in the PV soliton formation and in determining the self-focusing or defocusing strength. In fact, several authors have reported that the soliton width is minimum for a soliton-to-dark irradiance ratio not far from unity [1,5,8]. Similar conclusions were obtained for the screening soliton configuration [6,7]. Because of the extremely low dark irradiance in PV materials (for $LiNbO_3$ it is often of the order of microwatts per square centimeter [1] or even much smaller [9]), soliton formation requires a rather low focused beam irradiance and this results in an incredibly long time (hours or much more) [5], since the soliton formation time is roughly of the order of the dielectric relaxation time, which in turn is inversely proportional to the soliton intensity. In order to obtain faster time response a trapped beam with much higher intensity than the dark intensity is desired. However, in this case, the nonlinearity expressed by Eq. (1) would have inconveniently reached oversaturation. Consequently, the electric field induced by the photorefraction inside the crystal is almost uniformly screened in the illuminated region [2] and the lenslike refractive index variation would never be achieved. Therefore, the extremely low dark irradiance actually restricts the practical PV soliton formation. In order to avoid the limitations arising from the low value of the dark irradiance several authors proposed to use an additional incoherent uniform illumination that is assumed to play the same role as the dark irradiance [1,2]. However, unlike the case of biased crystals, theoretical support for this hypothesis is lacking.

In this Letter, we deduce the formulation of the nonlinear equation in photovoltaic PR materials for a focused laser beam and an incoherent background uniform illumination. We show that the form of the nonlinearity depends on the external circuit conditions of the crystal: Under short-circuit conditions the PV nonlinearity is still a saturable Kerr-type one and the background illumination indeed plays the role of the dark irradiance. Therefore the background illumination can be actually used to control the PV nonlinearity and to optimize the self-focusing or defocusing and the soliton formation. Under open-circuit conditions instead the saturable intensity cannot be controlled by the uniform illumination. Using the *Z*-scan formula that we have derived from the saturable Kerr nonlinearity the PV field in an iron doped lithium niobate crystal is determined from *Z*-scan experiments.

Figure 1 depicts the configuration of the light beam propagation used in our analysis. For the convenience of the analysis, a cylindrical Gaussian laser beam with intensity distribution $I(x, z) = I(0, z)\exp[-2x^2/w^2(z)]$ is assumed to propagate along the z direction. The crystal's c axis is oriented parallel to the gradient direction of the Gaussian beam, i.e., the x axis. The spatial extent of the Gaussian beam along the direction perpendicular to both the c axis and the propagation axis z is assumed to be infinite. An incoherent light with uniform intensity distribution I_R illuminates the total volume of the crystal. For

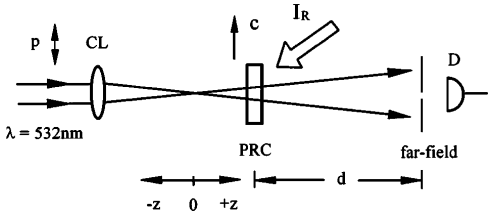

FIG. 1. Gaussian beam propagation configuration.

simplicity, the polarization of the two light beams is assumed to be linear and to be along the crystal's c axis.

The material response of a PR medium in which the electrons are the sole charge carriers is governed by the following set of equations [10]:

$$\frac{\partial N_D^+}{\partial t} = [s(I + I_R) + \beta_T](N_D - N_D^+) - \gamma n N_D^+ , \quad (2)$$

$$\frac{\partial \rho}{\partial t} + \nabla \cdot \vec{J} = 0 , \quad (3)$$

$$\rho = e(N_D^+ - N_A - n) , \quad (4)$$

$$\nabla \cdot (\epsilon \varepsilon_0 \vec{E}) = \rho , \quad (5)$$

$$\vec{J} = e\mu n\vec{E} + \mu k_B T \nabla n + \kappa s(N_D - N_D^+)(I + I_R)\vec{c} . \quad (6)$$

Here N_D, N_D^+, N_A, and n are, respectively, the volume density of total donors, ionized donors (acceptors), nonphotoactive ions (that compensate the ionized donors N_D^+ in the dark), and photoelectrons in the conduction band, s is the photoexcitation cross section, β_T is the thermal excitation rate of the electrons, ρ is the charge density, $\vec{J}$ is the electric current density, and $\vec{E}$ is the space-charge field. The other parameters are the recombination coefficient γ, the electric charge e, the electron mobility μ, Boltzmann's constant k_B, the absolute temperature T, the photovoltaic constant κ for the polarization used in this configuration, the dielectric constant ϵ of the crystal, and the permittivity of the vacuum ε_0. $\vec{c}$ is the unit vector pointing to the c-axis direction.

Steady state for short-circuit condition.—In steady state, $\partial N_D^+/\partial t = 0$ and $\partial \rho/\partial t = 0$. For a short-circuit condition the current (along the crystal's c axis direction) is constant. In typical PR materials $N_D, N_D^+, N_A \gg n$ for cw laser irradiance. Thus Eqs. (2), (4), and (5) yield the following results:

$$n = \frac{[s(I + I_R) + \beta_T]}{\gamma f}\left(1 + l_D \frac{\partial}{\partial x}\frac{E}{E_t}\right)^{-1} \times \left(1 - f l_D \frac{\partial}{\partial x}\frac{E}{E_t}\right), \quad (7)$$

$$N_D^+ = N_A\left(1 + l_D \frac{\partial}{\partial x}\frac{E}{E_t}\right), \quad (8)$$

where $f = N_A/(N_D - N_A)$, $l_D = (\epsilon \varepsilon_0 k_B T/e^2 N_A)^{1/2}$ is the Debye length, and $E_t = k_B T/e l_D = e N_A l_D/\epsilon \varepsilon_0$ is the limiting space-charge field corresponding to the Debye wave number $k_D = 1/l_D$. If along the c-axis direction the spatial extent $w(z)$ of the Gaussian laser beam is much less than the width of the crystal d, at $x \to \pm d/2$, $E(x, z) = E_0$ (constant), $\partial E/\partial x = 0$, and $N_D^+ = N_A$. Consequently, the current can be expressed by

$$J_0 = e\mu n_0 E_0 + \kappa s(N_D - N_A) I_R , \quad (9)$$

where $n_0 = n(x \to \pm d/2) = (sI_R + \beta_T)/\gamma f$ is the photoelectron density at $x \to \pm d/2$. The constant E_0 will generally depend on the external circuit condition and the illumination. From Eqs. (6) to (9), one can get the space-charge field

$$E = \left(E_0 \frac{I_R + I_d}{I_t} - E_p \frac{I}{I_t} + E_p \frac{I + I_R}{I_t} f l_D \frac{\partial}{\partial x}\frac{E}{E_t}\right)\left(1 + l_D \frac{\partial}{\partial x}\frac{E}{E_t}\right)\left(1 - f l_D \frac{\partial}{\partial x}\frac{E}{E_t}\right)^{-1} - \frac{k_B T}{e} \times \left\{\frac{\partial}{\partial x}(\ln I_t) - \left[\left(1 + l_D \frac{\partial}{\partial x}\frac{E}{E_t}\right)^{-1} + f\left(1 - f l_D \frac{\partial}{\partial x}\frac{E}{E_t}\right)^{-1}\right] l_D \frac{\partial^2}{\partial x^2}\frac{E}{E_t}\right\}, \quad (10)$$

where $E_p = \kappa \gamma N_A/e\mu$ is the photovoltaic field constant, $I_d = \beta_T/s$ is the so-called dark irradiance, and $I_t = I + I_R + I_d$. The last term in Eq. (10) originates from the diffusion term. If the intensity $I(x, z)$ varies smoothly with respect to x the diffusion effects in typical PR media may be neglected relative to the PV effects and the dimensionless term $l_D \partial E/\partial x = (\epsilon \varepsilon_0/e N_A)(\partial E/\partial x)$ is expected to be much less than unity [1]. The constant field E_0 (or the current density J_0) can be computed from the potential condition $\oint \vec{E} \cdot d\vec{l} = 0$. For a short-circuit condition and a Gaussian-like intensity distribution with characteristic scale $w(z)$ much smaller than the sample's size and under the condition $I_R \approx I(0, z)$, a numerical calculation predicts an approximate relation

$$E_0 \approx 0.8 \frac{w(z)}{d} E_p \ll E_p . \quad (11)$$

Therefore the expression (10) for the space-charge field can be simplified to

$$E \approx -E_p \frac{I}{I + I_R + I_d} . \quad (12)$$

Through the linear electro-optic effect, the crystal's refractive index perturbation follows the saturable Kerr-type nonlinearity

$$\Delta n_i = \Delta n_0 \frac{I}{I + I_R + I_d} , \quad (13)$$

where $\Delta n_0 = n_b^3 r E_p/2$ is the saturable refractive index perturbation, n_b is the unperturbed background index of refraction, and r is the effective electro-optic coefficient. Equation (13) is, formally, similar to Eq. (1) but with $I_R + I_d \approx I_R$ as the saturable intensity. The possibility of using I_R to increase the effective dark irradiance is

clearly supported now by Eq. (13). According to the theoretical results by Valley *et al.* [1] one can directly conclude that by keeping the background and soliton irradiance at a similar higher value, fast PV spatial soliton formation without oversaturation is possible. By the way, the nonlinearity expressed by Eq. (13) relies strongly on the short-circuit condition. We will show next that such nonlinearity cannot be obtained for an open-circuit crystal.

Steady state for open-circuit condition.—In this case there is no steady-state current ($J = 0$). Following the same derivations as above it can be shown that the refractive index perturbation has now the form

$$\Delta n_i = \Delta n_0 \frac{I + I_R}{I + I_R + I_d}. \tag{14}$$

Apparently, the nonlinearity represented by Eq. (14) suffers from the same issue of oversaturation as Eq. (1). The physical reason for this is that the charge carriers photoexcited by the uniform illumination will accumulate on the two end surfaces of crystal normal to the c axis and this will result in an almost saturated electric field equal to $E_0 = -E_p I_R/(I_R + I_d) \approx -E_p$ [see Eq. (9)]. This saturated field would prohibit the index perturbation induced by any additional irradiance.

Transient regime for short-circuit condition.—By using the same boundary condition for a short-circuit condition and a quasi-steady-state approximation ($\partial n/\partial t = 0$), through a tedious derivation the following dynamic equation can be obtained from Eqs. (2) to (6):

$$\tau_m \frac{\partial E}{\partial t} + E\left(1 + l_D \frac{\partial}{\partial x}\frac{E}{E_t}\right)^{-1}\left(1 - f l_D \frac{\partial}{\partial x}\frac{E}{E_t}\right) = -\frac{k_B T}{e}\left(1 + l_D \frac{\partial}{\partial x}\frac{E}{E_t}\right)^{-1}\left(1 - f l_D \frac{\partial}{\partial x}\frac{E}{E_t}\right)\left\{\frac{\partial}{\partial x}(\ln I_t)\right.$$
$$\left. - \left[\left(1 + l_D \frac{\partial}{\partial x}\frac{E}{E_t}\right)^{-1} + f\left(1 - f l_D \frac{\partial}{\partial x}\frac{E}{E_t}\right)^{-1}\right] l_D \frac{\partial^2}{\partial x^2}\frac{E}{E_t}\right\} + E_0 \frac{I_R + I_d}{I_t} - E_p \frac{I}{I_t} + E_p \frac{I + I_R}{I_t} f l_D \frac{\partial}{\partial x}\frac{E}{E_t}, \tag{15}$$

where $\tau_m = \epsilon\varepsilon_0\gamma N_A/[e\mu s I_t(N_D - N_A)]$ is the Maxwell relaxation time. Under the assumption $N_D \gg N_A$ ($f \ll 1$) and without uniform illumination I_R the dynamic equation (15) is simplified to that obtained by Zozulya *et al.* [11]. However, using the same approximation for the short-circuit condition, neglecting the diffusion term and assuming that $l_D/w(z) \ll 1$ for a Gaussian beam Eq. (15) can be simplified to

$$\tau_m \frac{\partial E}{\partial t} + E = -E_p \frac{I}{I + I_R + I_d}. \tag{16}$$

Here we also neglect the contribution from E_0 to the saturable space-charge field due to the short-circuit condition [Eq. (11)]. Equation (16) has a single exponential solution

$$E(t) = E_s[1 - \exp(-t/\tau_m)], \qquad E_s = -E_p \frac{I}{I + I_R + I_d}. \tag{17}$$

The refractive index perturbation, through the electro-optic effect, has the same form,

$$\Delta n_i(t) = \Delta n_0[1 - \exp(-t/\tau_m)]. \tag{18}$$

Experiment.—The perturbation of the refractive index in Eq. (13) can be experimentally measured using the so-called Z-scan technique. In this technique the crystal is placed at a distance z from the focal point of the focused beam along its propagation direction. The intensity at the center of the far-field diffracted beam is measured without any refractive index perturbation and after steady-state focusing (or defocusing) has been achieved. The ratio of the latter over the former value is the normalized transmittance T that is measured for different distances z (scanning for z). The dependence of T upon z has been already formulated using Eq. (13) for a one-dimensional Gaussian beam [12]

$$T(Z, \Delta\phi_0) = \frac{\sqrt{1 + Z^2}(\sqrt{1 + Z^2} + \beta_0)^2}{\sqrt{[\sqrt{1 + Z^2}(\sqrt{1 + Z^2} + \beta_0)^2 + 2\beta_0\Delta\phi_0 Z]^2 + 4\beta_0^2\Delta\phi_0^2}}, \tag{19}$$

where $Z = z/z_0$, z_0 is the confocal parameter of the Gaussian laser beam, $\beta_0 = I_0/I_R$, and I_0 is the intensity of the Gaussian beam at the focus. $\Delta\phi_0 = k\Delta n_0 L$ is the saturable phase perturbation through the crystal sample with the thickness L along the beam propagation direction.

For carrying out the Z-scan experiment, a laser beam from a diode pumped frequency doubled Nd:YAG cw laser with wavelength of 532 nm is divided into two beams. One of them is focused by a cylindrical lens (focal length = 11.5 cm) to provide the one-dimensional Gaussian beam. The measured minimum radius w_0 at the focus of the Gaussian beam is about 16 μm. The confocal parameter z_0 is 1.4 mm. The Gaussian laser beam is extraordinarily polarized and has an intensity $I_0 = 693$ mW/cm^2 at focus. The total power is 0.24 mW. The other beam is expanded and collimated into a diameter of 5 cm to provide a uniform illumination for the total volume of the sample and this uniform illumination is also extraordinarily polarized. In order to avoid recording a reflective PR grating the uniform illumination beam is frequency shifted (400 Hz, which is much larger than the inverse response time of self-lensing effect) by a piezoelectric mirror (PZT).

To reduce the possible fanning effect caused by the uniform illumination itself the incidence angle of this beam onto the crystal is slightly periodically varied by a pendular mirror with a frequency of 4.3 Hz. The intensity of the uniform illumination in the total volume of the sample is kept constant, $I_R = 180\ \mathrm{mW/cm^2}$. The estimated intensity ratio β_0 is 3.9. The sample used in our experiment is an iron (0.015 wt%) doped $LiNbO_3$ crystal. The dimensions are 0.85 mm $\times$ 3 mm $\times$ 10 mm, where the crystal thickness along the laser propagation direction is 0.85 mm ($L \ll nz_0$) and the crystal width normal to the crystal *c*-axis direction is 3 mm. The Gaussian beam covers entirely the sample in the direction normal to the crystal *c* axis. The crystal faces perpendicular to the *c* axis are painted with silver paste and short circuited. The absorption coefficient α of this crystal at $\lambda = 532$ nm is $1.9\ \mathrm{cm^{-1}}$.

In the experiment, the scattering (or fanning) effect is found to be small and can be neglected during the formation of the self-lensing structure. The reason for this is probably due to the following two facts: On the one hand, the gain for the scattering light amplification is much lower because of the small thickness of the sample in the beam propagation direction [13]. On the other hand, the uniform illumination beam with a periodically varied direction also functions as an erasing light for the fanning grating formation. The fact that no reflection grating is recorded is verified by the following test: when the uniform illumination is suddenly blocked the transmitted intensity does not change. The normalized transmittances are calculated with the initial and the steady-state value of the transmitted intensity and presented in Fig. 2, which shows the characteristics of the self-defocusing effect. Because of the characteristic large photovoltaic effects of $LiNbO_3$, the *Z*-scan curve shows a strong asymmetry. We use a two parameter (β_0 and $\Delta\phi_0$) fit to consider the uncertainty of the estimated β_0 from the size calculation of the Gaussian beam. The best theoretical fitting to Eq. (19) represented by the solid curve in Fig. 2 gives the parameter $\Delta\phi_0 = 4.9$ and $\beta_0 = 4.2$. From the former, a PV field of $E_p = 3.5 \times 10^4\ \mathrm{V\,cm^{-1}}$ is deduced for this crystal ($n_0 = 2.25$, $r = 30.8\ \mathrm{pm\,V^{-1}}$). This value is in good agreement with that measured from a holographic technique using the same sample [14]. The parameter β_0 obtained by fitting also approaches the estimated one. Song *et al.* have also measured self-lensing effects in $LiNbO_3$ crystal using a *Z*-scan experiment [15], but the shape of their normalized transmittance is quite different from ours, probably because they used different experimental conditions. For example, they used a circular symmetric Gaussian beam (the steady-current distribution inside the sample is consequently different from that in our configuration), much stronger intensity than ours, no background illumination, and no short-circuited sample. For such conditions the oversaturation of the nonlinearity would have occurred.

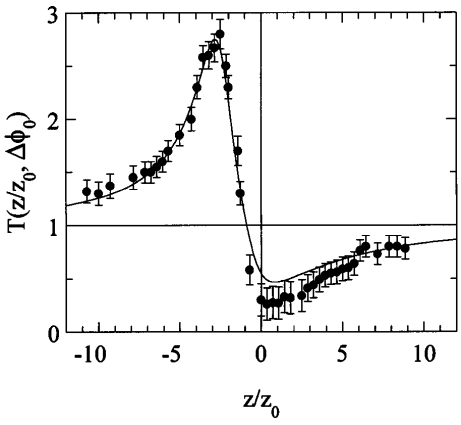

FIG. 2. *Z*-scan experimental results and the best theoretical fit (solid curve) with the parameters $\Delta\phi_0 = 4.9$ and $\beta_0 = 4.2$.

In conclusion, we derived the nonlinear equation for a photovoltaic PR material to include the effect of an incoherent uniform background illumination. The theoretically derived saturable Kerr-type refractive index nonlinearity for the short-circuit condition is accurately verified in a *Z*-scan experiment. The control of nonlinearity by the background illumination is responsible for the good data fitting and the reasonably good value computed for the Fe-doped $LiNbO_3$ crystal used in these experiments.

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