Optically induced photovoltaic self-defocusing-to-self-focusing transition

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We show theoretically and experimentally that the photovoltaic nonlinearity that gives rise to spatial solitons can be switched from self-defocusing to self-focusing (or vice versa) by use of background illumination. This raises the possibility of bright photovoltaic solitons in $LiNbO_3$. © 1998 Optical Society of America

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Optical spatial solitons, in particular photorefractive (PR) solitons,[1–9] have been investigated extensively in the past few years. PR solitons are formed when an optical beam creates a nonuniform charge distribution that gives rise to a space-charge field, E, in the crystal. By means of the Pockels effect this field creates a refractive-index change $\Delta n \propto E$. Here we focus on photovoltaic (PV) solitons, which are formed in unbiased PR media that exhibit a strong bulk photovoltaic effect.[5–8] In these crystals in an open-circuit configuration, the index change is $\Delta n = \Delta n_0 (I/I_d)/[1 + (I/I_d)]$, where $I(x)$ is the beam intensity, I_d is the natural dark irradiance, $\Delta n_0 = -(1/2) n_b{}^3 r_{\mathrm{eff}} E_p$, and $E_p = \kappa_{\mathrm{eff}}/(q\mu\tau_r)$ is the maximum attainable PV field, where κ_{eff} is the effective PV constant, q is the electron charge, μ is the electron mobility, and τ_r is the recombination time.[5,7] Dark PV solitons that arise from such Δn were recently observed[6,9] in $LiNbO_3$, in which $\Delta n_0 < 0$. There are two differences between the PV Δn and the screening soliton Δn. First, for screening solitons the sign of Δn_0 is controlled by the polarity of the applied field.[2] For PV solitons the sign of Δn_0 cannot be changed in a given material because it is determined by the sign of the product $(r_{\mathrm{eff}}\kappa_{\mathrm{eff}})$, which is fixed given the wavelength and the polarization of the beam. Second, adding a background beam of intensity I_b for screening solitons merely increases the background density of free carriers, which is equivalent to an increase in I_d by I_b.[2] (In all bright screening soliton experiments I_b is used to fine-tune the nonlinearity and avoid the necessity of high applied fields.[3,4]) On the other hand, for PV solitons, adding a background beam brings about new effects because it adds a term in the PV current. The problem of using background illumination of the same polarization as that of the focused beam[10] was solved recently.[8] In an open circuit, in which one-dimensional beams lead to $J = 0$ everywhere in the crystal, the nonlinearity saturates to Δn_0 for $I_b \gg I_d$. In a short circuit, in which the crystal is connected to an external load of zero resistance, the background beam effectively increases the I_d by I_b, giving a refractive-index change $\Delta n = \Delta n_0 I/(I_b + I_d + I)$.[8]

Here we show theoretically and experimentally that, if the focused and the background beams are orthogonally polarized, and if the crystal is not connected to an external resistor (open circuit), then the refractive-index change can reverse its polarity (e.g., from self-defocusing to self-focusing). In the short circuit this polarity reversal transition does not occur.

We start with the rate and continuity equations and Gauss's law in a PR medium with electrons as the sole charge carriers, plus the scalar wave equation for the optical field $E_{\mathrm{opt}} = A(x,z)\exp(ikz - i\omega t) + \mathrm{c.c.}$ $(k = 2\pi n_b/\lambda)$. $A(x,z)$ is the slowly varying amplitude. The crystal is illuminated uniformly by a background beam of intensity I_b, which is polarized normally to the soliton. In steady state the equations are[5–8]

$$s(|A|^2 + I_b + I_d)(N_d - N_d{}^i) - \gamma n N_d{}^i = 0\,, \quad (1)$$

$$\overline{\nabla} \cdot \overline{J} = \overline{\nabla} \cdot [q\mu n \overline{E} + K_B T \mu \overline{\nabla} n + \kappa_{ijj} s (N_d - N_d{}^i) \times (|A|^2 + \kappa I_b)]\hat{i} = 0\,, \quad (2)$$

$$\overline{\nabla} \cdot \overline{E} + (q/\varepsilon_s)(n + N_A - N_d{}^i) = 0\,, \quad (3)$$

$$\left(\frac{\partial}{\partial z} - \frac{i}{2k}\frac{\partial^2}{\partial x^2}\right) A(x,z) = \frac{ik}{n_b}\Delta n(E) A(x,z)\,, \quad (4)$$

where $\Delta n = -n_b{}^3 r_{iij} E/2$ is the refractive-index change, z is the propagation axis, and x is the transverse coordinate. Here n is the electron density, $N_d{}^i$ is the density of ionized donors, $\overline{J}$ is the total current density, $\overline{E}$ is the space-charge field in the crystal. Relevant crystal parameters are N_d, the total donor density; N_A, the density of negatively charged acceptors; s, the photoionization cross section; γ, the recombination coefficient ($\tau_r = 1/\gamma N_A$); ε_s, the low-frequency dielectric constant; k_B, Boltzmann's constant; T, the temperature; μ; and r_{eff}. In Eqs. (1)–(4) we assume that the polarization of the focused beam is in the $\hat{j}$ crystalline direction, giving rise to a PV current in the $\hat{i}$ direction via κ_{ijj}, and that the polarization of the background beam is in the $\hat{i}$ direction, giving rise to a current in the $\hat{i}$ direction as well but via κ_{iii}. The ratio $\kappa = \kappa_{iii}/\kappa_{ijj}$ can attain any value when the crystalline $\hat{i}$ and $\hat{j}$ axes are not related by symmetry. In our $LiNbO_3$ crystal, at $\lambda \approx 0.5\ \mu$m, $\kappa_{iii} > \kappa_{ijj}$.[11] In Eqs. (1)–(4), $\overline{J} = J\hat{i}$ and $\overline{E} = E\hat{i}$. Ohm's law yields $V = -\int_{-1/2}^{+1/2} E \mathrm{d}l = RSJ$, where V is the potential between the crystal's electrodes separated by l, S is the surface area of the electrodes, and R is the external resistance.[7]

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We seek stationary solutions of $A = u(x)\exp(i\Gamma z)\sqrt{I_d + I_b}$ (Γ = real), where $u(x)$ is real. The analysis is simplified because in typical PR crystals $n << \{N_A, N_d{}^i\} << N_d$.[2,4–8] As is justified in Ref. 7, we neglect the $\overline{\nabla} \cdot \overline{E}$ term from Eq. (3) and the diffusion term $K_B T \mu \overline{\nabla} n$ from Eq. (2) (which we can verify after obtaining E and the soliton solution for A). Thus we obtain $N_d{}^i \approx N_A$ from Eq. (3) and use it and $N_d - N_A \approx N_d$ in Eq. (1) to get $n \approx (sN_d/\gamma N_A)(I + I_d + I_b)$. Substituting for n in Eq. (2), we obtain

$$E \approx \frac{J - \kappa_{ijj} s N_d (I + \kappa I_b)}{q \mu s \frac{N_d}{N_A \gamma} (I + I_b + I_d)} \cdot \qquad (5)$$

This expression for E and the corresponding Δn apply not only for solitons but also to evolving beams as long as the z variation of A is much slower than the x variation. We now examine solutions for open and short circuits separately.

Open Circuit. When $J = 0$, Eq. (5) reduces to

$$E = -\frac{N_A \gamma \kappa_{ijj} (I + \kappa I_b)}{q \mu (I + I_b + I_d)} \cdot \qquad (6)$$

From Eq. (6) one can retrieve Eq. (14) of Ref. 8 (obtained for both beams with the same polarization) by setting $\kappa = 1$. In that case E and therefore Δn saturate to a constant value that does not depend on x if $I_d \ll I_b$. Essentially, the two beams are equivalent to one beam whose intensity is shifted upward by a constant value (leaving no dark areas). As long as the total intensity, $I(x) + I_b$, is much larger than I_d everywhere in the crystal, it sets up a uniform electric field equal to E_p, irrespective of the beam profile. On the other hand, when $I_b = 0$ we retrieve $\Delta n = \Delta n_0 I(x)/[I(x) + I_d]$.[5,7] Setting $\kappa = 1 + \delta$ in Eq. (6) yields

$$E = -\frac{N_A \gamma \kappa_{ijj}}{q \mu} \left(1 + \frac{\delta I_b - I_d}{I + I_b + I_d}\right) \cdot \qquad (7)$$

We neglect I_d (because typically $I_b \gg I_d$) in Eq. (7) to obtain an index change that contains a constant term and a term that depends on $I(x)$:

$$\Delta n = \Delta n_0 + \Delta n_0 I_b \delta / [I(x) + I_b], \qquad (8)$$

where $\Delta n_0 = n_b{}^3 r_{iij} N_A \gamma \kappa_{ijj} / (2q\mu)$. To explain these effects we plot $\Delta n = \Delta n_0 I(x)/[I(x) + I_d]$ (the $I_b = 0$ case) and Δn of Eq. (8) for the Gaussian-like $I(x)$ of Fig. 1(a) and for $\Delta n_0 < 0$. The $I_b = 0$ [Fig. 1(b)] case gives rise to self-defocusing, which turns into self-focusing [Fig. 1(c)] on the addition of an orthogonally polarized background beam. Notice that only the shape and not the sign of Δn changes; i.e., $\Delta n(x) < 0$ for all x in both cases. This is the expected result for LiN_bO_3, in which $I_b = 0$ always results in self-defocusing and $\Delta n_0 < 0$. However our result is general and applies also to the opposite case: If the nonlinearity is of the self-focusing type for $I_b = 0$ (i.e., $\Delta n_0 > 0$), then adding the background beam turns Δn into a self-defocusing nonlinearity. This transition is possible only when $\kappa > 1$ ($\delta > 0$) and $\delta I_b > I_d$, as is evident from Eq. (7). In principle, one can always choose the two polarizations such that $\kappa > 1$ as long as the two PV constants differ from each other.

We substitute Δn of Eq. (8) into Eq. (4), solve for bright solitons (as in Ref. 7), and obtain the soliton existence curve of Fig. 2. The existence curve shows $\Delta\xi$, the intensity FWHM normalized by $d = [kn_b(|r_{iij}E_p|)]^{-1}$, as a function of $u_0 \equiv u(x = 0)$. Notice that this curve gives a unique relation between $\Delta\xi$ and u_0 that resembles that of the screening solitons.[2] Intuitively, I_b establishes a uniform electric field across the crystal that is screened nonuniformly, giving rise to bright solitons. The inset in Fig. 2 shows the calculated soliton profile for $u_0 = 0.1$.

Experimentally, we launch a 14-μm FWHM ordinarily (o) polarized beam ($\lambda = 488$ nm, $n_b = 2.27$) into our 1-cm-long Fe-doped LiN_bO_3 crystal, in which we measure (by interferometry) $E_p = 66$ kV/cm. We add an extraordinarily (e) polarized background beam (I_b) that copropagates with the focused beam. To eliminate fanning on I_b we use a fast-rotating diffuser to make it spatially incoherent. Typical results are shown in Fig. 3. The input beam of Fig. 3(a) diffracts to a FWHM of 72 μm at the output [Fig. 3(b)] at $t = 0$ (before the space-charge field is formed). With $I_b = 0$, the beam becomes self-defocused when E reaches steady state [Fig. 3(c)], broadening to ~4 times its regular-diffraction output. When we add the e-polarized I_b and repeat the experiment we observe that the focused beam goes from self-defocusing to self-focusing: First it narrows to its regular diffraction size and then it further self-focuses to a steady state of 35 μm FWHM shown in Fig. 3(d). The left-hand portion of the beam in Fig. 3(d) is diffusive, and its structure is not fully symmetric, as expected from solitons. The peak intensity of the incident focused beam, I_0, is 0.8 mW/cm^2, and $I_b = 0.7$ mW/cm^2. We repeat the experiment with $I_0 = 0.4$ mW/cm^2 and observe that the output beam breaks into two, of which the FWHM of the stronger beam is 29 μm [Fig. 3(e)]. The data points of Figs. 3(d) and 3(e) are marked on Fig. 2. We conducted numerous experiments in which u_0 is fixed at the value of Fig. 3(d) and the width of the input beam varies between 14 and 25 μm (these points are marked by the crosses in Fig. 2). The point corresponding to Fig. 3(d) is expected to support ~20-μm solitons but, experimentally, points with widths of

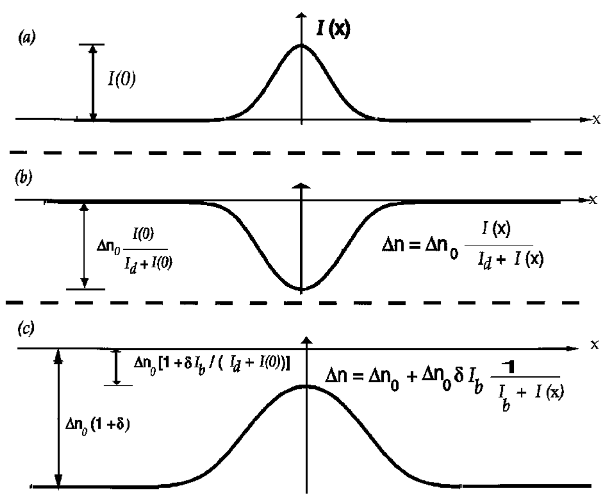

Fig. 1. (a) Beam profile. (b) Self-defocusing Δn for $I_b = 0$. (c) Self-focusing Δn when an orthogonally polarized background beam is added. Here $\Delta n_0 < 0$ (as in $LiNbO_3$).

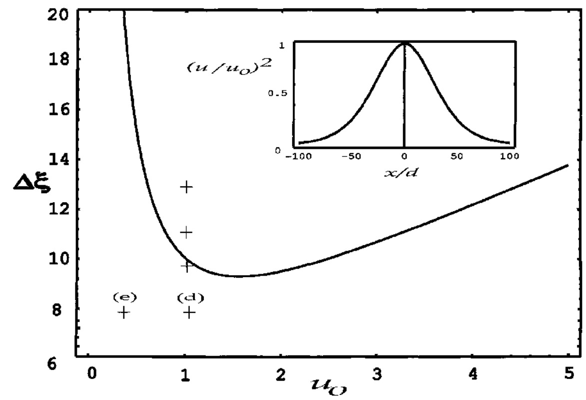

Fig. 2. Soliton existence curve for open-circuit bright photovoltaic solitons, showing the normalized FWHM as a function of u_0. Inset, beam profile for $u_0 = 0.1$. Crosses, experimental data points.

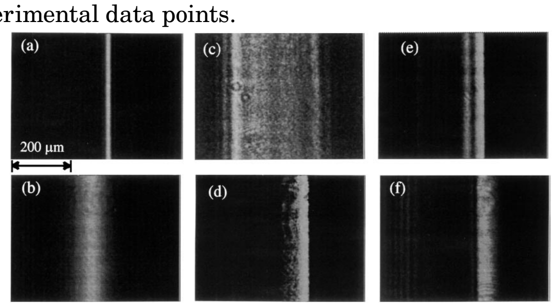

Fig. 3. (a) 14-μm FWHM input beam. (b) Regularly diffracting 72-μm output beam at $t = 0$ (before E forms). (c) Self-defocused output with $I_b = 0$. (d) Self-focused 35-μm output beam with an orthogonally polarized background beam; $I_0 = 0.8$ mW/cm^2 and $I_b = 0.7$ mW/cm^2. (e) Same as (d), except that $I_0 = 0.4$ mW/cm^2. (f) Output beam when both beams are e polarized; $I_0 = 0.9$ mW/cm^2 and $I_b = 0.7$ mW/cm^2. The output beam in (f) shows little change from that in (b) (it self-focuses to 55 μm).

20 μm and larger exhibit breakup (at this u_0 value). In this sense the fact that the self-focusing of the 14-μm beam of Fig. 3(d) does not fully compensate for diffraction agrees with the theory, but the breakup of a broader beam at this u_0 value does not. We attribute this result to the fact that the e-polarized I_b experiences self-focusing (which we observe in all our experiments) that is much stronger than that of the o-polarized focused beam, because $r_{33} \cong 3r_{13}$, thereby deviating from the theoretical assumption that I_b is uniform. Therefore we believe that bright photovoltaic solitons in $LiNbO_3$ can be generated only in crystals in which $\kappa_{311} > \kappa_{333}$,[11] for which the focused beam should be e polarized and the o-polarized I_b is almost unaffected by the induced lens, as for screening solitons in strontium barium niobate.[4] We now test the prediction that, when both beams have the same polarization, Δn becomes a constant. We launch both beams as e polarized with $I_b = 0.7$ mW/cm^2 and $I_0 = 0.9$ mW/cm^2. The steady-state result shown in Fig. 3(f) reveals that the output beam is slightly self-focused (from 72-μm regular diffraction to 55 μm). A similar result is obtained when both beams are o polarized. The small self-focusing with parallel polarization cannot be explained from Eq. (8) with $\delta = 0$ ($\kappa = 1$) but seems related to the partial guidance of I_b by the lens induced by $I(x)$.

Short Circuit. For a short circuit a net current is flowing through the crystal. Equation (6) yields $E = -(\gamma N_A \kappa_{iii}/q\mu)\{1 + [(\delta I_b - J/(\kappa_{ijj} s N_d)]/(I + I_b)\}$. Comparing this equation and Eq. (7) reveals that the transition to self-focusing can occur only for current densities lower than a threshold current $J_{\text{th}} = s\delta N_d I_b \kappa_{jjj}$. We evaluate the current by substituting E of relation (5) in Ohm's law and setting $R = 0$ (short circuit). Approximating the beam profile to be a square[7] gives $J = \kappa_{iii} s N_d I_b$. Using J in relation (5) yields $\Delta n = \Delta n_0 I/(I_b + I_d + I)$. This is a self-defocusing Δn, giving rise to dark PV solitons (with the effective dark irradiance increased from I_d to $I_b + I_d$). Thus the self-defocusing-to-self-focusing transition is not possible for a short circuit. On the other hand, the increase in the effective I_d facilitates control over Δn in a manner similar to that of screening solitons.[2,4]

In conclusion, we have shown that a strong ($\delta I_b > I_d$) uniform-background beam copropagating with the focused beam in an open-circuit photovoltaic crystal induces a polarity reversal of the nonlinearity when the beams are orthogonally polarized. The result is self-focusing in PV $LiNbO_3$ and should support bright PV solitons in $LiNbO_3$.

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10. The term "focused" is used to distinguish the narrow beam focused to the input face of the crystal from the uniform-background beam. It should not be confused with the terms "self-focused" and "self-defocused," which describe the output beam at steady state.
11. We measure interferometrically $\kappa_{333} = 1.14\kappa_{311}$, but cases in which $\kappa_{311} > \kappa_{333}$ have been reported in, e.g., H. Festl, P. Hertel, E. Kratzig, and R. Baltz, Phys. Status Solidi B **113,** 157 (1982); B. I. Sturman and V. M. Fridkin, *The Photovoltaic and Photorefractive Effects in Non-Centrosymmetric Materials* (Gordon & Breach, Philadelphia, Pa., 1992).