Dielectric nonlinearity in photorefractive spatial soliton formation

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We find that the anomalous behavior of optical spatial screening solitons observed in the high-symmetry paraelectric phase is a consequence of nonlinear dielectric effects. These, coupled to space charge in saturated conditions, change the effective optical nonlinearity even far from the phase-transition regime.

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Materials manifesting light-induced electro-optical effects, such as photorefractives, lead to nontrivial optical propagation phenomena that range from wave mixing to self-trapping [1,2]. In a time-independent representation, corresponding to a continuous-wave excitation, model equations are formally analogous to those associated to wave propagation in media with nonlinear susceptibilities. As opposed to direct nonlinear optical effects, these phenomena rest on a temporally nonlocal mechanism, by which the rapidly oscillating light field $\mathbf{E}_{opt}(\omega)$ is partially absorbed and leads to a quasistatic internal space-charge field $\mathbf{E}$, which in turn modulates the material index of refraction n [i.e., the high-frequency linear susceptibility $\chi(\omega)$] through the zero-frequency susceptibility $\chi(0)$. In this interplay between high- and low-frequency responses, which allows the observation of phenomena that would otherwise seem a riddle in a system undergoing direct nonlinear propagation [2], optical self-action is mediated by the static material polarization $\mathbf{P}=\chi(0)\mathbf{E}$, which, being intimately linked to the particular structural state of the material, would have beam phenomenology sensitive to it.

On the contrary, most phenomenology is transparent to the state of the medium. In particular, if we consider photorefractive self-trapping of continuous-wave visible laser beams, leading to photorefractive screening solitons, the light-crystal interaction can be described through a saturated Kerr effect [3]. The physical parameters of the system do not explicitly participate, since they can be taken into account through a renormalization of boundary conditions, soliton full-width-half-maximum (FWHM) $\Delta\xi$ and soliton intensity ratio u_0^2 [2,3]. This feature makes photorefractives a relatively powerful instrument of investigation in nonlinear science, in addition to hosting peculiar phenomena that are both interesting and useful.

This circumstance, fruit of an effective (secondary) interaction, can break down. This occurs blatantly in conditions in which spontaneous polarization plays a significant role, such as for spontaneous self-trapping [4] and soliton fixing [5]. However, these phenomena are not directly related to soliton propagation. A more fundamental manifestation occurs in diffusion-driven self-action, in which boundary conditions actually play *no* role, and solitons form on the sole basis of the crystal temperature T [6]. Yet these beams have, to date, never been observed, and this fascinating prediction finds no place in our present discussion.

We report and describe a distinct manifestation of structural effects that modify in a nontrivial way the behavior of photorefractive screening solitons in the high-symmetry paraelectric phase. In particular, we are able to ascribe anomalies in the existence conditions of these solitons, which occur even relatively far from the Curie temperature T_c, to dielectric nonlinearity [7]. The resulting system is described by a modified nonlinear equation that, apart from depending directly (i.e., in a manner that cannot be factored out in boundary conditions) on crystal temperature T, depends also on the *nature* of the "distant" phase transition.

Screening solitons in paraelectrics are accessibly observed in centrosymmetric potassium-lithium-tantalate-niobate (KLTN) [8] both as slab [9] and needle beams [10]. They are commonly described, in the (1+1)D (D means dimension) slab case, by means of an adaptation of the standard screening model, valid in the noncentrosymmetric case, to the paraelectric case [11]. The difference in the two descriptions is relegated to the relationship connecting the material index of refraction to the static polarization, $\Delta n_{ij}=-(1/2)n^3 g_{ijkl}P_kP_l$ in paraelectrics, as opposed to the linear Pockels effect $\Delta n_{ij}=-(1/2)n^3 R_{ijk}P_k$ in polar samples, n being the unperturbed index, and g_{ijkl} and R_{ijk} the quadratic and linear electro-optic tensors, respectively. For a constitutive relationship between the low-frequency polarization and field $\mathbf{P}=\varepsilon_0(\varepsilon_r-1)\mathbf{E}$, $\varepsilon=\varepsilon_0\varepsilon_r$ being the dielectric constant, this leads to effective saturated Kerr propagation described by $\Delta n \propto 1/(1+I/I_b)^2$, analogous to the nonlinearity $\Delta n \propto 1/(1+I/I_b)$ encountered in noncentrosymmetric ferroelectrics, I being the optical intensity, and I_b the saturating background illumination [2,3]. Thus, the nature of screening in noncentrosymmetric and centrosymmetric samples is generally thought to be the same.

Experiments project a somewhat different story [9]. They indicate that in KLTN this analogy is well founded except for the anomalous existence conditions of centrosymmetric self-trapping. Apart from the known mismatch between the experimental and theoretical curves, observed both in centrosymmetric and noncentrosymmetric samples, and attributed in part to the coupling between soliton and background fields [12], first observations indicated a peculiar temperature dependence of the existence curve that could not possibly find an explanation in the standard screening model. In particular, it was found that in the range of the values of the sample temperature T in which screeners were observable, higher values of T (farther from T_c) gave rise to an existence curve that was progressively shifted towards higher values of soliton normalized FWHM $\Delta\xi$ in the $(\Delta\xi, u_0)$ parameter plane [9].

This effect, observed *outside* the hysteretic cycle, suggests that we are not involving spontaneous polarization. However, being that screening solitons are supported by strong, spatially modulated, space-charge fields, a less dramatic manifestation of the dielectric anomaly appears in the form of dielectric nonlinearity. This structurally modifies the low-frequency constitutive relationship, introducing a nonlinear T-dependent behavior that does not factor out, as does the dependence of ε, g, n, on T. Thus, whereas the mechanism at the basis of self-trapping is the modulation of the local crystal polarization induced by the light generated space-charge field [3], the very presence of this field locally modifies the thermodynamic potential of the lattice, inducing a point-dependent modification of the dielectric response (dielectric nonlinearity).

In the tractable $(1+1)$D case, i.e., for slab solitons whose optical polarization is along the x direction, and for which $\mathbf{E}$ is a scalar x-dependent function, a description of dielectric nonlinearity can be obtained starting from the free energy of the crystal $a(\mathbf{P})$ [7]. A phenomenological description, in the stress-free case, as a function of the crystal polarization P along the cubic direction x, is given by the relationship

$$a(P)=\tfrac{1}{2}\sigma P^2+\tfrac{1}{4}\overline{\xi}P^4+\tfrac{1}{6}\zeta P^6, \tag{1}$$

where σ, $\overline{\xi}$, and ζ are material-dependent, possibly temperature-dependent constants. In this context, the dependence of the dielectric constant $\varepsilon \simeq (\partial P/\partial E)$ on the electric field $E \equiv (\partial a/\partial P)$ is approximately described by [neglecting the P^6 term in Eq. (1)]

$$\varepsilon(E)=\frac{\varepsilon(0)}{1+3\overline{\xi}\varepsilon(0)^3E^2}, \tag{2}$$

where

$$\varepsilon(0)=\frac{1}{\sigma}=\frac{\varepsilon_0 C}{T-T_0} \tag{3}$$

follows the Curie-Weiss relationship. ε_0 is the vacuum dielectric constant, and C and T_0 are phenomenological constants. The electro-optic response mediating optical self-action is, therefore,

$$\Delta n=-\tfrac{1}{2}n^3 g_{eff}\varepsilon_0^2\varepsilon_r^2(x)E^2(x), \tag{4}$$

where g_{eff} is the effective electro-optic coefficient (that depends on the geometry of the scalar setup [9]) and ε_r is x dependent through the modulation of $E(x)$ and Eq. (2). From Eq. (2), this relationship can be approximated by

$$\Delta n=-\tfrac{1}{2}n^3 g_{eff}\varepsilon(0)^2[1-6\overline{\xi}\varepsilon(0)^3E^2(x)]E^2(x), \tag{5}$$

in conditions in which $3|\overline{\xi}|\varepsilon(0)^3E^2(x)\ll 1$ [in our conditions $[3|\overline{\xi}|\varepsilon(0)^3E^2(x)]_{max}\sim 0.1$].

Indeed this is not the only change in the self-consistent standard approach. There is the more subtle appearance of polarization charges $\rho_P=\mathbf{E}\cdot\boldsymbol{\nabla}\varepsilon$ alongside the normal separated charge $\rho_E=\varepsilon\boldsymbol{\nabla}\cdot\mathbf{E}$. However, as in the standard approach, we are concerned with experimental configurations in which the direct influence of charge density can be neglected [3]. In fact, ρ_E and ρ_P involve the same scales, and, in conditions in which the screening theory is valid, its extension to include dielectric nonlinearity does not involve ρ_P.

We thus impose self-consistently that the slowly varying amplitude $A(x,z)$ of the optical field $E_{opt}=A(x,z)e^{ikz-i\omega t}$ (and $I=|A|^2$) of wavelength λ and wave vector $k=2\pi n/\lambda$ be a soliton eigenfunction of the form

$$A(x,z)=u(x)e^{i\Gamma z}(I_b)^{1/2}, \tag{6}$$

z being the direction of propagation, Γ the propagation constant eigenvalue, and I_b the aforementioned background illumination. The internal normalized space-charge field $Y \equiv E/(|V|/L)$, where V is the field applied to the crystal x electrodes, and L is the width of the sample along x, is related to the normalized soliton amplitude u through the relationship [2,3]

$$Y=-\frac{1}{[1+u(\xi)^2]}, \tag{7}$$

where $\xi=x/d$ is the transverse coordinate normalized to the spatial scale $d=(-2kb)^{-1/2}$, and $b=(k/n)\Delta n_0$ is the scale of the optical nonlinearity, where $\Delta n_0 = -(1/2)n^3 g_{eff}\varepsilon(0)^2(V/L)^2$ ($\Delta n_0<0$, being $g_{eff}>0$ in KLTN) is the index of refraction change in the absence of light and without phase-transition effects.

Imposing Eq. (6) and inserting the index modulation obtained from Eqs. (7) and (5) into the parabolic wave equation for $A(x,z)$, the $(1+1)$D case is described by the following nonlinear wave equation:

$$\frac{d^2u(\xi)}{d\xi^2}=-\left[\delta-\frac{1}{[1+u(\xi)^2]^2}\left(1+\rho\frac{1}{[1+u(\xi)^2]^2}\right)\right]u(\xi), \tag{8}$$

where $\delta=\Gamma/b$ is determined by the boundary conditions and $\rho=-6\overline{\xi}\varepsilon(0)^3(V/L)^2$ is the dimensionless scale of the dielectric nonlinearity. Whereas, for $\rho=0$ Eq. (8) reduces to the one at the basis of the standard screening model [11], a finite value of ρ gives rise to a different nonlinear equation. Without considering the change in the saturation of the nonlinearity, the overall effect can be seen as a higher-order "focusing" one for $\rho>0$, i.e., for an anomaly associated

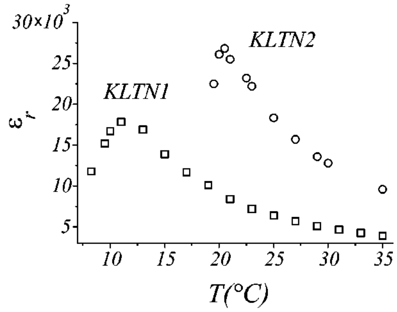

FIG. 1. Linear dielectric anomaly of soliton-supporting samples KLTN1 (squares □) and KLTN2 (circles ○).

with a first-order phase transition, where $\bar{\xi}<0$, and a "defocusing" one for a second-order transition, where $\bar{\xi}>0$ and hence $\rho<0$.

In order to close the self-consistent approach, we can relate the eigenvalue δ to boundary (launch) conditions. To do this, we note that, as occurs for the standard screening theory [2,3], no dissipative term containing $u'(\xi)$ appears in Eq. (8), and it can be integrated once by quadrature. Finally, imposing the boundary conditions pertaining to bright solitons (the only species observed in KLTN [9,10]), and defining $u_0 \equiv u(0)$, we obtain the expression for δ:

$$\delta=\frac{1}{1+u_0^2}+\frac{\rho}{3}\frac{(1+u_0^2)^3-1}{u_0^2(1+u_0^2)^3}. \tag{9}$$

In a first set of experiments, we investigated the dielectric nonlinearity of the soliton-supporting samples, in the range of temperatures where self-trapping is observed. In order to highlight the role of dielectric nonlinearity, which as mentioned is strongly dependent on the nature of the transition, our studies are carried out on *two* samples of KLTN, KLTN1, and KLTN2, which undergo two substantially *different* phase transitions. KLTN1 experiences a second-order-like transition at $T_{c1}\simeq 11\,°\mathrm{C}$, whereas KLTN2 undergoes a first-order-like transition at $T_{c2}\simeq 20\,°\mathrm{C}$ (see Fig. 1). The samples, which measure, respectively, $3.7^x \times 4.3^y \times 2.4^z$ mm and $2.6^x \times 2.1^y \times 9.2^z$ mm, are zero-cut with respect to their principal axes of the high-symmetry paraelectric phase. They are different nonstechiometric compositions of KTN and KLN, this accounting for their qualitatively and quantitatively different behavior, and have a perovskite-like ferroelectric structure. They are doped with copper and vanadium atoms, and these give rise to a substantial photorefractive response for light up to 600 nm in both samples.

The low-frequency electric field is delivered to each sample by means of gold electrodes deposited on the x facets, $L_1=3.7$ mm and $L_2=2.6$ mm apart. A static voltage V is applied to the facets by a low-current programmable high-voltage supply, whereas an LCR meter superimposed an uncoupled low-frequency (<100 KHz) oscillation for simultaneous capacitance measurement. The sample temperature T is fixed by a Peltier junction, driven by a stabilization circuit whose output is the junction current and input is the attained sample temperature, measured by means of a thermocouple in contact with the sample.

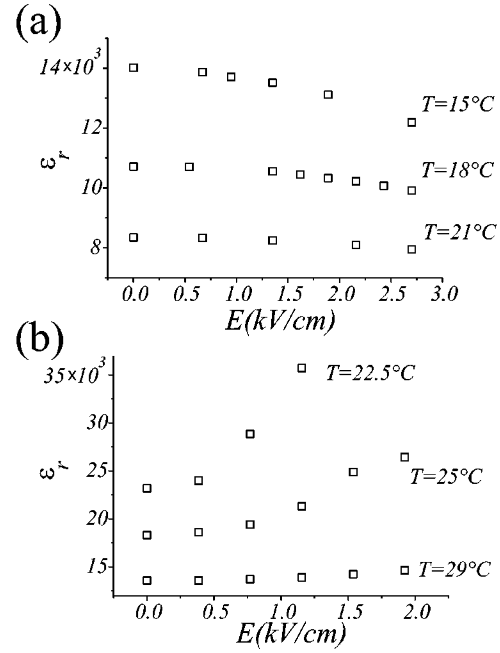

FIG. 2. Dielectric nonlinearity of (a) KLTN1 and (b) KLTN2 for various temperatures of interest where solitons are observed. Note the opposite behavior of the samples.

For a zero-applied external field, $E_0=V/L=0$, the measured dielectric anomaly, i.e., the behavior of ε_r for different values of crystal T, is shown in Fig. 1. The curves refer to decreasing temperature trajectories, and the hysteretic cycle occurs for temperatures below $T\simeq 13\,°\mathrm{C}$ for KLTN1 and $T\simeq 22\,°\mathrm{C}$ for KLTN2. The phenomena we wish to interpret refer to values above this limit, where the approximate description of dielectric nonlinearity contained above holds. Where spontaneous polarization exists, the perturbative approach is not valid, and an appropriate theory has not been developed. Fitting the curves with the phenomenological mean field Eq. (3) gives $C1\simeq(1.05\times 10^4)\,°\mathrm{C}$, $T_{01}\simeq 9\,°\mathrm{C}$, and $C2\simeq(2.11\times 10^4)\,°\mathrm{C}$, $T_{02}\simeq 13\,°\mathrm{C}$. Note how the different nature of the two transitions emerges in the different values of the mismatch $T_{c1}-T_{01}\simeq 2\,°\mathrm{C}$ and $T_{c2}-T_{02}\simeq 7\,°\mathrm{C}$, higher for the first-order one.

To characterize dielectric nonlinearity of the samples, approximated by Eq. (2), we superimposed on the low-frequency oscillation a static electric field (uncoupled to the LCR meter) through a controllable high-voltage supply. Results are shown in Fig. 2. As can be seen directly from this data, neglecting dielectric nonlinearity is a useful, but hardly defensible, approximation.

This allows an evaluation of $\bar{\xi}$ as a function of sample T using the expression of Eq. (2) approximated as in Eq. (5). Results are summarized in Table I. Sample KLTN1 has positive values of $\bar{\xi}$, which increase as T increases, for the temperatures studied, leading to an overall "defocusing" effect. For KLTN2, the measured dielectric nonlinearity parameter $\bar{\xi}$ decreases linearly in magnitude as sample T increases. The

TABLE I. Summary of measured dielectric nonlinearity parameters.

Sample	T (°C)	$\overline{\xi}$ in (S.I.)
KLTN1	15	0.31×10^{9}
	18	0.40×10^{9}
	21	0.62×10^{9}
KLTN2	22.5	-1.64×10^{9}
	25	-0.99×10^{9}
	29	-0.41×10^{9}

negative values indicate a ‘‘focusing effect’’ associated to a first-order transition.

Inserting the values of Table I into Eqs. (8) and (9), allows a prediction of the generalized existence curve by solving numerically the one-dimensional nonlinear equation and finding the value of the self-trapped normalized FWHM $\Delta\xi$ as a function of u_0 and ρ.

To compare the theory to experiments, we carried out a map of soliton existence curves in both samples. This was done launching in the z direction a diffracting $(1+1)$D Gaussian beam from a visible laser source into the crystals, and observing for which values of applied external voltage V and intensity ratio u_0^2 the beam undergoes nondiffracting propagation. Experiments were repeated for several values of sample T. The apparatus, which involves a beam preparation, launching, and detection system, is reported in an exhaustive manner in literature [2]. We might underline that soliton phenomenology we seek is of the steady-state type, i.e., a self-trapped condition that remains so in time as long as the soliton parameters are maintained in proximity of the existence conditions [2,3].

In Fig. 3 we show the experimental data taken for KLTN1 [9] and KLTN2 compared to theory. In particular, in Fig. 3(a) experimental existence points fan out in a manner that is well described by the theory, indicated by the dashed curves. Branching indicates that a stronger nonlinearity is required to trap solitons than would be expected from standard theory [11], indicated by the single solid curve. An opposite behavior is found in Fig. 3(b) that shows results obtained in KLTN2. The solid curve, which is the same as that in Fig. 3(a), is substituted by the branched dashed curves that emerge considering dielectric nonlinearity, and indicate that trapping occurs for lower nonlinear responses. Note the diametrically opposite behaviors of the two samples. Experimental data only marginally match quantitative predictions,

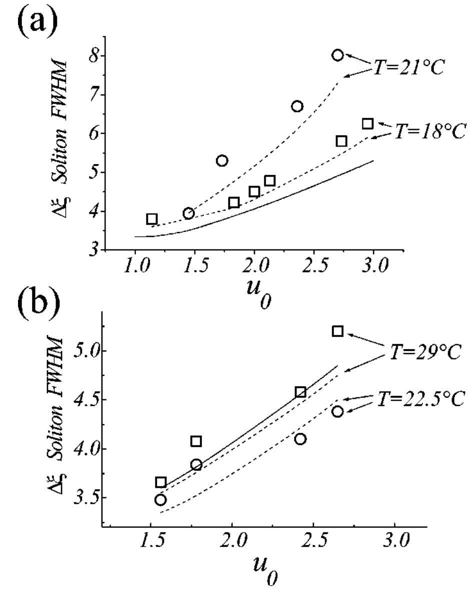

FIG. 3. Comparison between experimental existence points in (a) KLTN1 and (b) KLTN2 for the values of T considered. We note the characteristic branching of the curve due to dielectric nonlinearity, the defocusing effect in the second-order transition that characterizes KLTN1 (a), and the focusing effect in the first-order transition of KLTN2 (b). Dashed curves are the predictions based on the numerical integration of Eq. (8). Full curve corresponds to the standard model, where no branching is present.

as commonly occurs in these experiments [12]. However, this being the central product of the present study, the branching and the specular behavior of the two samples finds an explanation in the modified dielectric response of the samples, which leads to the modified nonlinear propagation regime of Eq. (8).

In conclusion, investigating anomalous self-trapping behavior, we have identified dielectric nonlinearity as one of the basic mechanisms leading to optical soliton formation in photorefractive paraelectric crystals. This allows a more knowledgeable design of electro-holographic soliton-based circuitry, a promising optical device-oriented effort [13,14].

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