Spontaneous Self-Trapping of Optical Beams in Metastable Paraelectric Crystals

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(Received 9 November 1998)

We report on the observation of a new mechanism for self-trapping of optical beams: self-trapping that stems from spontaneous creation of ferroelectric crystalline clusters, seeded by a weak photorefractive diffusion field. This is an evident observation of the highly nonlinear aspects of propagation in a thermodynamically metastable system, including optically driven crystalline ordering in a medium undergoing a phase transition.

PACS numbers: 42.65.Jx, 77.84.–s

Spontaneous symmetry breaking in systems undergoing a phase transition is one of the most intriguing subjects of statistical physics. In this regime, fluctuations are exhalted and ultimately allow for a macroscopic dynamical change in the properties of a system. In this Letter, we investigate light propagation in a metastable system undergoing a phase transition, and show that, when the propagating entity (light) is coupled to the out-of-equilibrium host (matter), the spontaneous material response can lead to considerable nonlinear propagation effects and the light induces crystalline ordering. We launch a very narrow optical beam into a photorefractive potassium-lithium-tantalate-niobate (KLTN) crystal [1] and study its diffraction. When the crystal is at a temperature slightly above the paraelectric-ferroelectric transition, the beam diffracts and is initially strongly distorted. After a temporal transient, the beam self-focuses and eventually self-traps: exhibiting nondiffracting propagation. This occurs in both one $(1 + 1)$D and two $(2 + 1)$D transverse dimensions. The resultant self-trapping mechanism of the beam is extremely *insensitive* to the beam's parameters, and is attributed to spontaneous alignment of dipolar clusters induced by the weak photorefractive diffusion field. We refer to this mechanism as "spontaneous self-trapping" to underline the fact that the interaction is a seeded thermodynamic relaxation.

Optical beam propagation at the vicinity of the Curie temperature (T_c) in media undergoing a phase transition has been recently addressed in the context of photorefractive media [2]. However, no observation has been carried out in the highly nonlinear metastable phase-transition regime itself. Typically, the medium is optically opaque at $T = T_c$, as the strong density fluctuations severely scatter light. Numerous other effects were observed in such systems. For example, in supersaturated aqueous urea solutions, light induces crystallization and prenucleation [3]. In some atomic/molecular systems, laser induces clustering [4]. In paraelectric KLTN, the screening of space-charge separation induces metastable ferroelectric clustering [5]. In nematic liquid crystals, laser heating generates isotropic "holes," giving rise to strong self-phase modulation [6], and in metals at the melting point, an enhanced cubic nonlinearity has been reported [7]. Thus far, optical *propagation* effects have been studied only in critical binary liquid mixtures [8], and even there only theoretically. Here we study nonlinear beam propagation in very close proximity to the thermodynamic phase transition: at a temperature where the system is stable with respect to noise-induced phase fluctuations, but metastable with respect to optically induced fluctuations. This is the first work we are aware of that studies experimentally optical propagation effects in materials that are exactly at their *optically induced* phase transition point.

Our experiments are carried out with a setup typical of spatial soliton studies [2]. A $\lambda = 514$ nm linearly polarized $(\|x)$ laser beam is focused onto the input face of a zero-cut KLTN sample, oriented with one principal axis (henceforth the z axis) parallel to the propagation direction. We first launch a 1D beam (narrow in x) by means of a cylindrical lens, and investigate spatial effects. We then repeat the experiment with a circular beam. The sample is kept at a temperature T by means of a current-controlled Peltier element and a feedback stabilizing driver. A second beam is split after the laser, which illuminates the crystal uniformly while copropagating with the (first) focused beam. Unlike other experiments with photorefractive solitons (e.g., Refs. [2,9]), here this beam is used to investigate the physical process, and *does not* participate in the nonlinear interaction (unlike the "background beam" [2,9]); it is blocked during the self-trapping process.

Our KLTN sample measures $3.7^{(x)} \times 4.6^{(y)} \times 2.4^{(z)}$ mm and has a pale green color. It is doped with vanadium and copper, and exhibits strong photorefraction in the visible [10]. It has a refractive index $n = 2.4$ (at $\lambda = 514$ nm) and quadratic electro-optic coefficients $g_{11} = 0.12\ \mathrm{m^4\,C^{-2}}$ and $g_{12} = 0.02\ \mathrm{m^4\,C^{-2}}$ [2]. The

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low frequency dielectric constant ε_r exhibits a dielectric anomaly at $T_c = 10\,^{\circ}\mathrm{C}$, indicating a first-order (from paraelectric to ferroelectric) phase transition. In the absence of external elements, the onset of the noncentrosymmetric phase and macroscopic ordering is seeded by imperfections and local strain, and can be viewed visually due to the electro-optic response to the spontaneous polarization. Fitting the ε_r values in the paraelectric region with the Curie-Weiss law $\varepsilon_r = C/(T - T_0)$ gives $C = 1.5 \times 10^5\,^{\circ}\mathrm{C}$ and $T_0 = 6.2\,^{\circ}\mathrm{C}$. The transition manifests temperature hysteresis [11] for $T < 14\,^{\circ}\mathrm{C}$ characteristic of first-order transitions, indicating that the nucleation is affected by the presence of long range dipolar domain forces (domain-nucleation interaction). Our measurements refer to the decreasing temperature branch.

In the (1 + 1)D configuration, a cylindrical lens generates a 1D Gaussian "sheet of light" at the input, as shown in Fig. 1. The left column shows the input beam at $T = 34\,^{\circ}\mathrm{C}$ (deep in the paraelectric phase). The confined direction has a 11 μm input full width at half maximum (FWHM). After propagating 2.4 mm in the crystal, the beam diffracts to a 26 μm FWHM output (middle). When the temperature is lowered into the metastable regime at $T = 11\,^{\circ}\mathrm{C}$, after a strong "spikelike" transient beam displacement (occurs $\ll$1 sec), the output beam resembles the input, and no diffraction is observed (right column in Fig. 1). This result is fully reproducible in a whole range of peak beam intensities, from 0.1 to about 10 $\mathrm{W\,m^{-2}}$ *without* observable changes, apart from the duration of the initial transient regime (typically, a few seconds in this range of intensities).

In the (2 + 1)D configuration, the focusing lens is spherical. Typical results are shown in Fig. 2: the circular 11 μm FWHM input beam (left), the diffracted 24 μm FWHM output beam when the crystal is deep in the paraelectric phase (middle), and the 11-μm-wide self-trapped output beam at $T = 11\,^{\circ}\mathrm{C}$ (right). The final nondiffracting "needle" beam is observed after a transient dominated by strong beam deformation and rapid sideways switching. We repeat this experiment with intensities varying through the range investigated in the (1 + 1)D configuration, and, again, we observe no difference in the steady-state results apart from the duration of the transient.

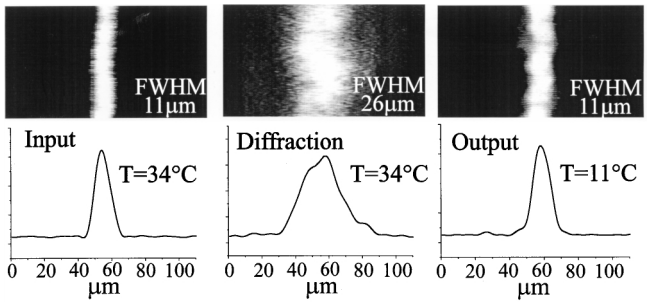

FIG. 1. (1 + 1)D case: the 11 μm beam self-traps at $T = 11\,^{\circ}\mathrm{C}$.

We then investigated the decay dynamics of these "spontaneous self-trapped beams" by first observing self-trapping (as in Fig. 2), blocking the focused beam, and then allowing the collimated beam to uniformly illuminate the crystal, keeping the crystal temperature constant. Adjusting the uniform beam intensity so as to make it comparable to the focused beam peak intensity, and allowing the space-charge field to relax (erasure occurs within a few seconds, for the intensities used), the collimated beam was blocked and the focused beam was again launched. Diffractionless propagation *without observable transient dynamics* was observed. When the same procedure was repeated after increasing the crystal T from the initial 11 °C to approximately 14 °C, no noticeable effects were observed. The trapping disappeared only at $T > 14\,^{\circ}\mathrm{C}$. Independent measurements of ε_r show that this temperature coincides approximately with the high temperature of the hysteresis loop.

We next investigate the refractive index pattern that supports the self-trapped beam at $T = 11\,^{\circ}\mathrm{C}$. In the (1 + 1)D case, a microphotograph is taken of the crystal output face with the self-trapped beam blocked and the uniform beam (polarized along the y axis, at 90° with respect to the x axis) illuminating the sample. The uniform illumination "repels" from the region in which self-trapping has formed. Repeating this procedure in the (2 + 1)D configuration gives the results shown in Fig. 3. In Fig. 3(a), we show the crystal output face with the self-trapped beam blocked and the uniform beam polarized along the y axis. As in (1 + 1)D, light is repelled from the region that gives rise to self-trapping. In Fig. 3(b) the polarization of the uniform beam was rotated by 45°. Here, some of the light is guided in the region where self-trapping occurs (central intensity hump). Figures 3(a) and 3(b) indicate that dipolar clusters have formed with a prevalent orientation at 45° with respect to the cubic axes: This is clearly visible from the #-type structure with the 45° inclination in these figures. Heating the crystal (in the absence of illumination) to 13 °C (just below the hysteresis loop high temperature) and returning to a y-polarized uniform beam reveals that a structure persists even though the characteristic dipolar clusters are no longer observable [Fig. 3(c)]. The clusters diminish in size and the crystal response is due to a less ordered clusterization (outside the central hump). Finally, in Fig. 3(d) the crystal is heated to 15 °C. Effects due to the initial self-trapping are no longer observable. Note that, without first going through the self-trapping process, the crystal does not exhibit any metastable ferroelectric structures

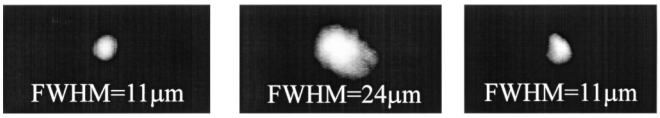

FIG. 2. (2 + 1)D case: the 11 μm beam self-traps at $T = 11\,^{\circ}\mathrm{C}$.

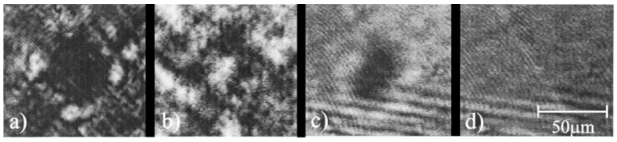

FIG. 3. Crystal output face ($T = 11\,^{\circ}\mathrm{C}$) illuminated by a plane wave polarized at 90° with respect to the polarization of the previously formed self-trapped beam (a); at 45° (b); at 90° after heating the sample to $T = 13\,^{\circ}\mathrm{C}$ (c); and at 90° heating it to $T = 15\,^{\circ}\mathrm{C}$ (d).

at $T = 11\,^{\circ}\mathrm{C}$, and the microphotograph is almost identical to that of Fig. 3(d) (which was taken at $T = 15\,^{\circ}\mathrm{C}$).

To investigate the preference of this self-trapping mechanism to the optical polarization, we launch a circularly polarized circular beam [by inserting a $\lambda/4$ plate in the beam path before the sample, in the (2 + 1)D configuration]. The circularly polarized beam never self-traps, as a whole, for any value of near-transition temperature. At $T = 11\,^{\circ}\mathrm{C}$, the beam always splits into two distinct parts. The first part is trapped (as in the linearly polarized case) and is itself linearly polarized either in the x or y direction. The second part, polarized orthogonal to the trapped beam, is always distorted and is repelled from the central beam region, as shown in Fig. 4. The actual direction of the linear polarization of the guided portion of the beam is random, with the probability of having a self-trapped x-polarized beam almost identical to that of having a y-polarized one. Thus, the polarization of the self-trapped component is chosen in tandem with the growth of the ferroelectric clusters, which are themselves affected by the optical polarization. This self-trapping mechanism is not based on thermal effects or on strain-induced photovoltaic processes [12] as intensity does not play any significant role, i.e., the self-trapping is not affected by intensity variations over a large intensity range. The fast dynamics during the transient phase indicates that spontaneous nucleation of ferroelectric clusters in the undercooled system is involved [13]. We believe that the underlying mechanism is based on light-induced diffusion space-charge fields, which are approximately intensity independent. Such fields, in a periodic structure, can induce *periodic* ferroelectric domain reversal at the ferroelectric phase [14], or metastable periodic dipolar clusters at the paraelectric phase [5]. In what follows, we explain our spontaneous self-trapping as driven by a light-seeded crystalline ordering.

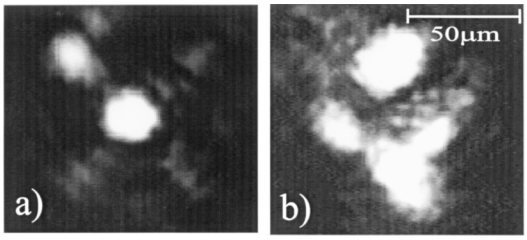

FIG. 4. Nonlinear splitting of a circular polarized beam into a trapped linearly polarized beam (a) and a diffracted orthogonally polarized beam (b).

In the (1 + 1)D case, the initially highly diffracting beam is exciting electrons from impurities into the conduction band, where they diffuse to less illuminated regions, being retrapped there by other ionized donors or acceptors. Since the donors left behind are positively charged, this leads to charge separation and to a space-charge diffusion field in the x direction (the direction of beam confinement). Near the phase transition, the noncentrosymmetric ferroelectric configuration begins competing with the unpolarized (paraelectric) phase, and any field larger than the coercive field E_c (at T) can seed metastable ferroelectric clusters [5]. The diffusion field is $\mathbf{E}_d = -(k_B T/q)\nabla I/(I + I_d)$ (k_B is the Boltzmann constant, q is the electron charge, I is the intensity distribution, I_d is the dark irradiance, and the gradient is over spatial coordinates). For an input Gaussian beam, E_d is very small in the central region (where the gradient is zero) and far from the beam, whereas in the region surrounding the beam (at a distance of the order of the beam radius σ from the beam's center) it reaches $E_d \approx (k_B T/q)2/\sigma \approx 50\ \mathrm{V/cm}$. Near the phase transition, $E_c < E_d$, and E_d induces domain formation. Thus, on both sides of the propagating 1D beam, two counterpolarized domains form, leaving the central region of the paraelectric phase [Fig. 5(a)]. In the center, the refractive index is unaffected ($n_p = 2.42 \pm 0.06$) as this region always remains in the paraelectric phase, whereas at the ferroelectric region the crystal is birefringent with $n_e < n_p < n_o$. Thus, when the beam is x polarized, it corresponds to extraordinary polarization in the ferroelectric region, which has a lower refractive index

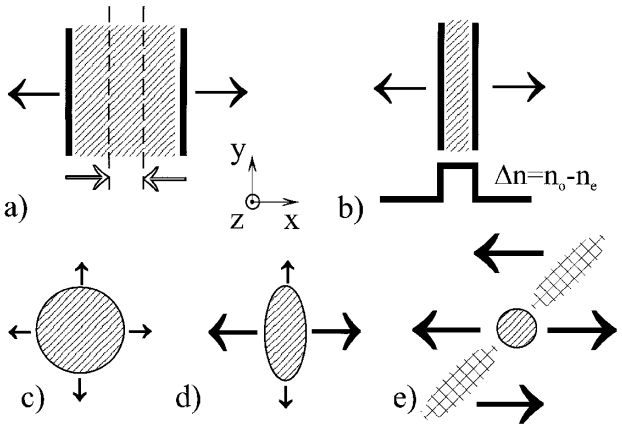

FIG. 5. Spontaneous self-trapping: (a) Diffracting (1 + 1)D beam (shaded region) seeds two counterpolarized domains with spontaneous polarization. Light is concentrated on a smaller area, thus moving the maxima of the diffusion fields inwards, which make the walls drift inwards. (b) Stationary index structure that traps the beam. (c) Initial diffusion-induced domains in the (2 + 1)D case. (d) Prevailing domains for an x-polarized beam (shaded). (e) Final domain structure with prevalent x-directed domains and residual screened regions with 45° walls. Filled arrows indicate average polarization of clusters.

$[n_e \cong (2.29 \pm 0.06)]$ than that of the central paraelectric region (n_p). Thus, this beam "sees" a waveguide structure [Fig. 5(b)] and is guided in the central region. This causes the intensity to be distributed over a smaller area and moves the region in which E_d is large inward towards the center of the beam. This process stops at the innermost region upon the beam, where E_d cannot surpass E_c. If the input beam is approximately the fundamental mode of this waveguide pattern, a condition which depends on the value of E_c at the given T ($E_c = 0$ at $T = T_c$), self-trapping occurs [Fig. 5(b)]. On the other hand, if the beam is y polarized, it sees a lower refractive index in the central paraelectric region, because n_o is slightly larger than n_p ($n_o = 2.45 \pm 0.06$), and is antiguided (repelled) from the central region, as observed in the experiments.

The (2 + 1)D case is more complex. Figures 3(a) and 3(b) indicate that, as in the (1 + 1)D case, at the center of the beam the crystal remains in a paraelectric state, whereas outside the beam domains form and are prevalently polarized along the direction of the polarization of the beam. Figure 4 indicates that the prevailing structure is formed *during* the nonlinear process. Consider first a beam polarized linearly along the x axis. Initially, the diffusion field induces ferroelectric clusters at the margins of the diffracting beam oriented parallel with the electric field direction. Each separate cluster can only be polarized along a principal axis [Fig. 5(c)]. Optical confinement (waveguiding), however, is efficient only for clusters polarized parallel to the beam polarization ($n_e < n_p$). Thus, of the various clusters initially seeded, only those polarized parallel (and antiparallel) to the x axis "grow" [Fig. 5(d)]. Because of strong dipolar interactions, these two domains prevail, giving rise to intermediate screening of y-polarized domains with walls at 45°, which is the final structure observed in Fig. 3 and illustrated in Fig. 5(e). This interpretation is supported by results with a circularly polarized beam: In this case, diffraction compensation is efficient for both polarizations; however, at the end only one type of domain prevails. The prevailing spontaneous polarization coincides with the polarization of the guided beam. The other component is repelled from the guiding central region and diffracts, distorted by the complex domain structure.

This mechanism of spontaneous self-trapping is unique in that the index modulation Δn is not functionally related to the propagating optical beam [15]. Also, the hysteresis nature implies that the self-trapped beam is *not self-supported*: Once a stable cluster pattern has been formed, the dynamics of the interaction passes from a transient statistical nonlinear evolution to a linear propagation regime that is identical to that of a fabricated waveguide. In this respect, the only related system we know of is self-induced permanent waveguides in photosensitive polymers [16]. Yet, our observation leaves several questions unanswered, as we do not know the microscopic details of this complicated self-trapping mechanism via spontaneous formation of clusters, and cannot yet formulate it in a rigorous theoretical form. Certainly, further investigation into the statistical nature of the phenomena, interactions between two such self-trapped beams, plus self-induced ordering characterization, will lead to even more interesting observations where pattern formation occurs right at a natural phase transition. For example, the very fact that our technique allows transmission of light in a narrow channel while the entire region around it strongly scatters light has few equivalents in optics [17]. We believe that our observations are not material specific or even optics specific, but rather belong to a very general class in which light directly influences the state of matter.

The research of E. D. and M. T. was carried out in the framework of an agreement between the Fondazione Ugo Bordoni and the Italian Communications Administration. The research of A. J. A. and M. S. was supported by the Israeli Ministry of Science.

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