Subwavelength optical spatial solitons and three-dimensional localization in disordered ferroelectrics: towards metamaterials of nonlinear origin

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We predict the existence of a novel class of multidimensional light localizations in out-of-equilibrium ferroelectric crystals. In two dimensions, the non-diffracting beams form at arbitrary low power level and propagate even when their width is well below the optical wavelength. In three dimensions, a novel form of subwavelength light bullets is found. The effects emerge when compositionally disordered crystals are brought to their metastable glassy state, and can have a profound impact on super-resolved imaging and ultra-dense optical storage, while resembling many features of the so-called metamaterials, as the suppression of evanescent waves.

Introduction — Out-of-equilibrium materials display remarkable features, most of them still to be understood. Recent experiments in supercooled photorefractive crystals have allowed the observation of “scale-free” optical solitons [3,4] supported by an extremely weak diffusive nonlinearity [2,3], which becomes active through the emergence of a dipolar glass with anomalously enhanced susceptibility. These nonlinear beams have a truly remarkable feature: they are independent of size and intensity. Size-independence spurns a very basic and potentially ground-breaking exploration: are these scale-free optical beams capable of propagating even when their size is noticeably smaller than the optical wavelength?

In this Letter we predict that glassy photorefractive ferroelectrics [1,2] support a novel kind of multidimensional light localization at scales below the optical wavelength. The effect requires a huge susceptibility that can be harnessed in the out-of-equilibrium, or non-ergodic, phase, by acting on the previous history of the sample [1]. The finding is thus part of a still infant field of investigation that focuses on using out-of-equilibrium optical materials to achieve novel and hereto unexplored effects [1,2], a non-ergodic nonlinear optics that is rooted in statistical mechanics, material science, and nonlinear wave propagation. Subwavelength propagation overcomes a basic limit to optical imaging and microscopy, i.e., that a light field can only propagate components of its spatial spectrum within the diffraction limit. High frequency components that correspond to details comparable to and smaller than the wavelength normally form evanescent waves that simply do not propagate. In stark contrast, in our predictions light leads to self-trapped beams of arbitrary intensity and widths, for which no diffraction limit holds. These predictions are based on an intensity-independent nonlinearity, which can be interpreted as the consequence of a widely tunable refractive index accompanied by a transmission of evanescent fields. Put differently, a nonlinearity-based meta-material [10,11].

Intensity-independent PNR nonlinearity — Our model system is a compositionally disordered and impurity-doped photorefractive relaxor ferroelectric (e.g., the KLTN [3]). When it is rapidly cooled below the characteristic Bruns temperature $T_B$, it exhibits polar nano-regions (PNR). These are highly polarizable randomly-distributed ferroelectric-like regions that form a dipolar glass and provide an enormous enhancement of the photorefractive nonlinear optical response but retain very limited scattering losses [1]. The clue to the whole matter, originally discussed by Burns [15], is that the optically-induced index of refraction change depends on the spatial average of the square of the crystal polarization $P$. When the crystals display a zero polarization due to disorder (i.e., $\langle P \rangle \approx 0$), averaging over disorder leads to a non-vanishing effect that depends exclusively on the mean-square of the polarization (i.e., $\langle |P|^2 \rangle$). Optical response is thus directly correlated to the underlying nature of the crystal fluctuations, that become anomalously large and dependent on the crystal history in the out-of-equilibrium state. Specifically, averaging over the randomly oriented PNR [3,7], the index of refraction perturbation is

$$\Delta n_{PNR} = -\frac{3}{2}g^2\chi_{PNR}^2E_{DC}^2,$$

where $n_0$ is the bulk refractive index of the isotropic (disordered) crystals, $g$ is the relevant component of the second-order electrooptic tensor, $\chi_{PNR}$ is the low-frequency electric response due to the PNR. The low-frequency electric field $E_{DC}$ is the space-charge field expressed in terms of the optical intensity $I$ as $I = I_0 e^{gE_{DC}^2}$.
mentally investigated in [1] if condition 
\[ L > \lambda \] is satisfied. Note that this 
the key point is that more general solutions exist for 
the diffusive nonlinearity, which only depends on the lo-
cal intensity and not on the beam polarization [2, 3]. The 
Helmholtz model reads as 
\[ \nabla E + \left( \frac{\omega n}{c} \right)^2 E = 0, \] 
whose propagation invariant solution is written as 
\[ E = a \exp(ik_2z) \] where \( k_2 \) is the overall wave-vector in the 
z direction (different from its nonlinear perturbation \( \beta \) in 
the paraxial model above). The scale-free Gaussian is 
also solution of Eq. (7) with arbitrary amplitude \( A_0 \) and 
waist \( w_0 \), with 
\[ k_2 = \sqrt{\left( \frac{2\pi n_0}{\lambda} \right)^2 - \frac{4\gamma^2}{w_0^2}} \] 
that is, for a beam waist that is (within multiplicative 
constants) greater than the wavelength \( \lambda/n_0 \).
The key point is that more general solutions exist for 
\( L > \lambda \), analogous to what occurs in the paraxial case. 
These are still given by Eq. (5) where, however, the wav-
ector is 
\[ k_z = \sqrt{\left( \frac{2\pi n_0}{\lambda} \right)^2 - \frac{4\gamma^2}{w_0^2}} \] 
The corresponding lower limit for the waist is hence 
\[ w_0 > \gamma \frac{\lambda}{n_0}. \] 
The factor \( \gamma \) plays a role similar to a Lorentz contraction 
term in special relativity, even if with a different mean-
ing.
Specifically, for $L > \sqrt{2}\lambda$ the lower limit for the waist is scaled by a factor $\gamma < 1$, and correspondingly beams with a width smaller than the wavelength can propagate in the medium without distortion. Note that for this solution the evanescent waves are completely inhibited by the intensity independent scale-free nonlinearity, which leads to the conclusion that arbitrarily-low power beams with size below the wavelength can propagate.

**Non-paraxial regime** — To investigate the formation of the two-dimensional (2D) soliton in the nonparaxial model we consider the forward propagating projection of the Helmholtz equation, which is written as

$$i\partial_\xi E + k\sqrt{1 + \frac{\nabla^2}{k_0^2} + 2\frac{\Delta n}{n_0}} E = 0. \quad (12)$$

Equation (12) reduces, under suitable limits, to the well-known unidirectional propagation equations (see [10,17] and references therein) for the description of nonlinear optics beyond the paraxial model. The basic difference here is that the nonlinear refractive index $\Delta n$ is retained under the square root, since our nonlinearity is intensity-independent and hence of the same order of the Laplacian even in the low intensity regions of the beam. After introducing the optical carrier with $E = A \exp(ikz)$ and the diffusive nonlinearity, the normalized dimensionless model reads as the non-paraxial normalized model

$$i\partial_\xi \psi + \frac{1}{2} \left[ -1 + \sqrt{1 + \epsilon \nabla^2_{\xi,\eta} - 2\sigma(\mathbf{v} \cdot \mathbf{v})} \right] \psi = 0,$$

$$\mathbf{v}|\psi|^2 + \nabla_{\xi,\eta}|\psi|^2 = 0, \quad (13)$$

where we introduce the dimensionless variables $\xi = x/w_0$, $\eta = y/w_0$, and $\zeta = z/z_0$ with $z_0 = kw_0^2$ the Rayleigh length. $\psi = A/A_0$ is the normalized optical field with $A_0$ an arbitrary constant, and $\mathbf{v} = E_{DC}/(KBT/q)$ is the normalized space-charge field. In Eq (2) only two parameters appear, the degree of non-paraxiality $\epsilon$, which vanishes in the paraxial limit, and the strength of the scale-free nonlinearity $\sigma$ ($\sigma = 1/8$ as $L = \lambda$).

Equation (2) can be numerically solved by Taylor expanding the square root, giving

$$\partial_\zeta \psi = i \sum_{n=1}^{N} \left( \frac{i}{n} \right) \epsilon^{n-1} \left[ \nabla_{\xi,\eta}^2 - 2\sigma(\mathbf{v} \cdot \mathbf{v}) \right]^n \psi \quad (14)$$

where $N$ is the order of approximation selected (for a fixed $\epsilon$) in order to a have a given precision.

To assess the existence of self-trapped scale-free solitons beyond the paraxial regime, we start by considering the linear diffraction regime ($\sigma = 0; \ L << \lambda$) in Fig.2a, which shows the spreading of the waist of a Gaussian beam for an increasing order in the the solution of Eq. (14). For $\epsilon > 0$ non-paraxial terms provide a more pronounced spreading if compared to the paraxial model (i.e., to $N = 1$), and it is shown that for the case of $\epsilon = 0.05$ corrections become inconsequential for $N > 3$.

In Fig.2b we show the propagation of the beam for $\sigma = 1/8 \ (L = \lambda)$, including higher order diffraction. In such a nonlinear case, diffractionless propagation is achieved at any order and for any intensity. Note that the resulting beam is propagation invariant at any order $N$ independently on the scale $w_0$ and on the intensity level, thus showing the fact that the proposed scale free solutions are indeed stable and exist for ultra-thin beams.

**Enhanced visibility** — Next we numerically investigate conditions allowing super-resolution. We consider the propagation of two parallel beams for $\epsilon = 0.01$ (order $N = 3$) for various values of the ratio $L/\lambda$. For a given input pattern we find that there exist an optimal value corresponding to minimal input intensity distribution (i.e., image) distortion. In Fig.3 we consider the propagation of two parallel beams (shown in Fig.3c) for different values of $\sigma$. We compare the (Fig.3b) linear propagation regime ($\sigma = L/\lambda = 0$) and the case $\sigma = 0.15$ (Fig.3c); for $\sigma > 0$ (corresponding to high cooling rates see [1]) the visibility radically increases. In Fig.3d we show the fringe visibility versus $\sigma$, calculated as the ratio between the intensity peak value and the value at the center of the beam: the contrast increases (ideally diverges) when the visibility is higher. This result shows that even for beam sizes in the non-paraxial regime, at sufficiently high cooling rate, it is possible to propagate images without loss of information. We stress that these dynamics are attained at any intensity level, and hence represent a completely new regime in optical propagation, where the laws of diffraction and interference are largely modified; the strength of nonlinearity being determined by $\sigma$.

For $L >> \lambda \ (\sigma >> 1/8)$, a Gaussian beam experiences a strong intensity-independent self-focusing, even below the non-paraxial regime (not reported).

**Three-dimensional subwavelength localization** — A notable property is the existence of three-dimensional localized light bullets. Specifically the three-dimensional (3D) Helmholtz equation for the diffusive nonlinearity...
can be cast as a "nonlinear" eigenvalue problem:

\[-\frac{\nabla^2 E}{E} + \left(\frac{L}{\lambda}\right)^2 \left(\frac{\nabla|E|^2}{|E|^2}\right)^2 = k^2\]  

Eq. (15) admits an exact 3D Gaussian solution \(A = A_0 \exp\left[-(x^2 + y^2 + z^2)/w_0^2\right]\) when \(L = \lambda\), for any \(A_0\) and when \(w_0 = \sqrt{3/2\pi\lambda/n_0}\). In distinction to previous results, these solutions are not spatially scale-free and may only have a fixed waist, comparable with the wavelength in the medium. For \(L > \lambda\) a solution exists (more general ones exist and will be reported elsewhere) and is given by

\[A = A_0 \left[\cosh(\sqrt{2}x/w_0) \cosh(\sqrt{2}y/w_0) \cosh(\sqrt{2}z/w_0)\right]^{-\gamma^2}\]  

with \(\gamma\) as above (Eq. (10)) and the waist \(w_0 = \gamma \sqrt{3/2\pi\lambda/2\pi n_0}\). These 3D localized solutions have a waist smaller that the wavelength when \(\gamma < 1\) \((L > \sqrt{2}\lambda)\). They represent a novel form of light localization at any intensity level and with size comparable or smaller than the wavelength. We are not aware of other known light localizations, which can be described by an exact solution; it is a novel kind of bound state between the photo-induced charges and light which may be used to store information.

Metamaterials of nonlinear origin — In standard optics, a Gaussian beam with waist \(w_0\) has a spectral bandwidth of the order of \(1/w_0\), and the minimum waist such that the spectrum is contained in the Ewald circle (i.e., without evanescent waves) is given by \(\lambda/n_0\); in the scale-free regime here considered, due to the non-ergodic phase of glassy ferroelectrics, such a minimum waist is given by

\[\frac{\gamma \lambda}{n_0}.\]  
The medium hence exhibits (for \(L \gg \lambda\)) an effective refractive index

\[n_{\text{eff}} = \frac{n_0}{\gamma^2} = n_0 \sqrt{\left(\frac{L}{\lambda}\right)^2 - 1} \approx n_0 \frac{L}{\lambda} \gg n_0.\]  

The PNR effect is therefore equivalent to an (intensity independent) refractive index, which can be largely tuned and increased, such that beams propagate without evanescent waves and without relevant scattering and absorption losses. This shows that the specific nonlinearity here considered is able to provide those features that are the building blocks for the modern research on metamaterials, from a completely different perspective.

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