Programming scale-free optics in disordered ferroelectrics

Jacopo Parravicini,1,2 Claudio Conti,1,3 Aharon J. Agranat, 4 Eugenio DelRe 1,2
1 Department of Physics, Università di Roma “La Sapienza”, 00185 Roma, Italy
2IPCF-CNR, Università di Roma “La Sapienza”, 00185 Roma, Italy
3ISC-CNR, Università di Roma “La Sapienza”, 00185 Roma, Italy
4Applied Physics Department, Hebrew University of Jerusalem, 91904 Israel

Compiled May 2, 2014

Using the history-dependence of a dipolar glass hosted in a compositionally-disordered lithium-enriched potassium-tantalate-niobate (KTN:Li) crystal, we demonstrate scale-free optical propagation at tunable temperatures. The operating equilibration temperature is determined by previous crystal spiralling in the temperature/cooling-rate phase-space. © 2014 Optical Society of America

OCIS codes: 160.2750, 190.4400, 190.5330

Optical diffraction can be compensated in waveguides or when solitons form. The waves must have a specific size, shape, and specific intensity, for the solitons. Recent experiments in electromagnetically-induced transparency and nonlinear optics have identified a situation in which diffraction is compensated, for solitons, but without the scale-dependent constraints, a phenomenon termed diffraction cancellation [1, 2]. In photorefractive crystals this has been demonstrated in a regime of so-called “scale-free optics” supported by the formation of a dipolar glass, a phenomenon typical of disordered ferroelectric crystals that host glass-forming polar nanoregions (PNRs) when rapidly cooled [3, 4]. Scale-free optics opens the way to a number of enticing effects, such as wavelength-insensitive propagation [5] and scale-free spatial instability [6]. Moreover, for its range of validity [7], the underlying model predicts a regime that supports subwavelength beam propagation [8].

Experimentally, rapid cooling gives rise to scale-free optics only when the cooling rate is above a threshold value and the final equilibration temperature coincides with the Curie point $T_C$ (in cooling), where the paraelectric-ferroelectric phase transition occurs [2]. In fact, the scale-free model requires the photorefractive diffusion field to cancel diffraction, a phenomenon that entails an enhanced static dielectric response as only observed at $T_C$ [9]. The key is that the dielectric anomaly is rendered accessible by the PNR-driven glass which suppresses long range order and the optical scattering typical of the equilibrium phase-transition [10, 11].

In PNR-driven dipolar-glasses thermal history can actually shift the crystal $T_C$ with its associated anomalous enhancement of response. This property gives rise to a marked thermal hysteresis that delimits what is termed the crystal glassy phase (see Fig. 1a) [4, 12–14]. In this letter we program the temperature at which scale-free optics occurs by exploiting specific sequences of heating and quenching stages. In our results, we are able to span the greater part of the crystal glassy phase.

To grasp the physical underpinnings of our experiments, we recall that photorefraction leads to a diffusive nonlinearity [15, 16], which profoundly alters beam propagation, in that diffraction is governed by an effective refractive index $n_{\text{eff}}$ [2, 5, 8]

$$n_{\text{eff}} = n_0/(1 - (L/\lambda)^2),$$

where $n_0$ is the unperturbed refractive index, $\lambda$ the wavelength and $L = 4\pi n_0^2 \varepsilon_r \sqrt{\chi_{\text{PNR}}/(K_B T/q)}$. Here $g$ is the effective quadratic electro-optic coefficient, $\chi_{\text{PNR}}$ is the effective history-dependent low-frequency dielectric susceptibility of the dipolar glass, $K_B$ the Boltzmann constant, $T$ the crystal equilibration temperature (i.e. the temperature measured at a given instant) and $q$ is the charge of the photoexcited carriers. Eq. (1) is valid for $L \lesssim \lambda$. As $L \to \lambda$, $n_{\text{eff}} \gg n_0$ and diffraction is cancelled, the scale-free regime, independently of beam size and intensity. The condition $L = \lambda$ requires $\chi_{\text{PNR}} \sim 10^5$ (which imposes $T \approx T_C$ for rapid cooling schemes).

We use a $6 \times 3 \times 2.5$mm sample of ferroelectric Li-enriched Cu-doped potassium-tantalate niobate (KTN:Li). In Fig. 1a we report the quasi-static dielectric response $\varepsilon_r$ versus $T - T_C$ ($T_C = 14.5^\circ$C) obtained from capacitance measurements at slow heating and cooling rates ($|\alpha| = |\Delta T/\Delta t| \simeq 0.01^\circ$C/s) [14]. The marked hysteresis in $\varepsilon_r(T - T_C)$ (shaded region in Fig. 1) signals a non-ergodic phase several degrees above and below $T_C$, a well-known complex dielectric response [17, 18] that is analogous to that observed in other relaxors [4]. The Cu doping gives it a greenish tint and a strong photorefraction [2].

Thermal preparation required to morph $\chi_{\text{PNR}}$ is achieved through a computer-controlled Peltier-junction that fixes the temperature versus time $T = T(t)$ schedule (the sequence of quenching and heating preparatory stages). Once the thermal preparation has been completed, we shine laser light into the sample and detect diffraction. We focus a linearly polarized (in the horizontal $x$-direction) $z$-propagating beam onto the input facet and analyze the input and output beam intensity distribution Full-Width-at-Half-Maximum (FWHM) using an imaging system and a CCD camera. The condition $L = \lambda$ is found when in-
put and output FWHM measurements coincide, i.e., the
diffraction of micron-sized beams ceases, independent of
size and peak intensity.

We first carry out experiments using the standard con-
stant \( \alpha \) thermal preparation (rapid cooling). As reported
previously [2], only a small region in the experimentally
available parameter space \((T - T_C, \alpha) = (T - T_C, T)\) gives
rise to \( L \approx \lambda \), tagged in Fig. 1b as “GLASSY”. The region
is evidently smaller compared to the thermal range in
which dielectric hysteresis is detected (shaded region).
A typical result for parameters external to the glassy re-
region is shown in Fig. 2b. Here, the rapid cooling process
is characterized by \((T - T_C, \alpha) = (2.3^\circ C, -0.12^\circ C/s)\).
As seen in the transverse intensity distribution images,
the input intensity distribution with a FWHM of \( \Delta x \approx \Delta y \approx 12 \mu m \) (see Fig. 2a) diffracts and spreads to
\( \Delta x \approx \Delta y \approx 30 \mu m \) after the \( l_\perp = 3 \) mm propagation in
the sample (Fig. 2b). As expected, diffraction cancella-
tion is not observed. Comparing diffraction to expected
Gaussian beam diffraction (\( \Delta x_{\text{out}} \approx 33 \mu m \), given
the crystal \( n_0=2.38 \) and \( \lambda = 633 \) nm), we find \( L/\lambda \lesssim 0.1 \)
\((n_{\text{eff}} \approx n_0)\). We consider paraxial diffraction where a
standard launch/detection scheme is sufficient (see [2]).

We next repeat the experiment reaching the same final
temperature \((T - T_C = 2.3^\circ C)\), but through a different
thermal preparation, characterized by a non-monotonic
thermal path. The set of successive points occupied by
the sample in the \((T - T_C, \alpha)\) plane at the different in-
stants of time is the curve \( S \) in Fig. 1b. This represen-
tation allows to appreciate how, in terms of the rapid
cooling experiments, the crystal is actually exposed in se-
quence to all three “phases”, the paraelectric, the glassy,
and the ferroelectric one. After this preparation, the
output beam intensity distribution, reported in Fig. 2c,
manifests no diffraction, signalling \( L/\lambda \approx 1 \) even though
the final equilibration temperature is external to or-
iginal GLASSY region (but still contained in the shaded
one). We thus conclude that the spiralling trajectory \( S \)
in the \((T - T_C, \alpha)\) plane is able to produce the giant \( \chi_{\text{PNR}} \)
which is necessary to obtain \( L/\lambda \approx 1 \) even though
the final temperature \( T \neq T_C \). For comparative purposes,
in Fig. 1b we also plot the \( R \) curve associated to the rapid
cooling process (of Fig. 2b) [19].

We next extend the use of these “spiralling” trajec-
tories to obtain a scale-free response (i.e., excited glassy be-
havior) throughout the shaded region in Fig. 1. Results
reported in Fig. 3 demonstrate the activation of scale-
free propagation regime at tunable temperatures beyond
the GLASSY region. We underline that our results are as-
associated to aging and are hence transient in time. For a
beam with a peak intensity of \( I_p \approx 6 \) \( \text{W/cm}^2 \), the scale-
free phenomenon occurs after an exposure of \( 20 \) s and
begins decaying after approximately \( 130 \) s. Remarkably,
once the aging has washed out the scale-free regime, the
response can be rejuvenated at the same or at a different
temperature. Repeating the full cycle, i.e., washing out
previous photorefractive charge displacement (resetting
of the dipolar glass), the same trajectory will lead to the

Fig. 1. (a) KTN:Li static dielectric constant \( \varepsilon_r \) for slow
cooling (red curve) and slow heating (green curve) as a
function of \( T \). (b) Representation of the parameter plane
\((T - T_C, \alpha)\) of the sample preparation. Both regions
of paraelectric and ferroelectric behavior, which are unable
to support scale-free propagation through rapid cooling,
are indicated, together with the glassy region (dashed
line indicates the approximate phase separation). The
shaded region indicates the range of hysteresis, while
the “GLASSY” region indicates the temperature-cooling
rate range where standard constant \( \alpha \) preparation (rapid
cooling) can give rise to \( L \approx \lambda \) (see text). The red
(R) and blue (S) curves represent respectively the rapid-
cooling (\( \alpha = \) constant) and the spiralling trajectories
with initial \((T_i = 24.5^\circ C)\) and final \((T_f = 16.8^\circ C, \quad \text{dot-
dashed line) temperatures that refer to the experimental
data in Fig. 2.}

In terms of soft-matter physics, our experiments
demonstrate a method to outdo the strong temperature
selectivity required in rapid cooling experiments through
the complexity of the dipolar glass in the region of the
non-ergodic phase, identified by thermal hysteresis in
the dielectric susceptibility [3, 4]. Our technique, based
on the use of non-monotonic thermal preparation, ap-
pears analogous to the cross-over or Kovacs effects in
soft-matter [12, 13]. Triggered by non-monotonic ther-
mal preparation, cross-over is known to lead to effects
that are absent through rapid cooling [14].
The points lie in a temperature range of more than 4 \degree C, representing, Fig. 3. Programming scale-free response at temperatures $T \neq T_C$. (Top right) Representation in the $(T - T_C, \alpha) = (T - T_C, T)$ parameter plane of the different conditions of scale-free optics reported. Activation, testified by the diffraction-free output intensity distribution (top) reported in the series of images $S_1$–$S_5$, is achieved through spiralling thermal trajectories (bottom). Programmed temperatures $T \approx 14.8$ °C ($S_1$), 15.7 °C ($S_2$), 16.8 °C ($S_3$), 17.7 °C ($S_4$), 18.7 °C ($S_5$). In the parameter plane representation, $\alpha$ is the mean value along the trajectory. The points lie in a temperature range of more than 4 °C, simplifies further experimental endeavors, such as the all-important search for sub-micron beam propagation [7,8], where spatial solitons intrinsically break down [20].

We thank M. Deen Islam for technical assistance. This work was supported by funding from the Italian Ministry of Research through the FIRB grant PHOCOS-RBFRO8E7VA and from the ERC under the European Community 7th Framework Program (FP7/2007-2013)/ERC grant agreement no. 201766. Partial funding was received by the SMARTCONFOCAL of the Regione Lazio and by the PRIN no. 2009P3K7Z2 projects. A.J.A. acknowledges the Peter Brojde Center for Innovative Engineering.

References

1. O. Firstenberg, P. London, M. Smuker, A. Ron, N. Davidson, Nat. Physics 5, 665 (2009).
2. E. DelRe, E. Spinozzi, A.J. Agranat, C. Conti, Nat. Photonics 5, 39 (2011).
3. G.A. Samara, J. Phys. Condens. Matter 15, R367 (2003).
4. A. Bokov, J. Mater. Sci. 41, 31 (2006).
5. J. Parravicini, F. Di Mei, C. Conti, A.J. Agranat, E. DelRe, Opt. Express 19, 24109 (2011).
6. V. Folli, E. DelRe, C. Conti, Phys. Rev. Lett. 108, 033901 (2012).
7. The model breaks down for scales at which the space charge saturates. This is a sample-dependent limit associated to the acceptor density $N_A$ and the static $\varepsilon_r$. In typical KTN:Li samples $N_A \approx 2 \cdot 10^{18}$ cm$^{-3}$, so that even in the crytical regime with $\varepsilon_r \sim 10^5$, the model breaks down for beam widths $l \sim 0.1$ µm.
8. C. Conti, A.J. Agranat, E. DelRe, Phys. Rev. A 84, 043809 (2011).
9. F. Jona, G. Shirane, Ferroelectric crystals (Dover 1993).
10. E. DelRe, M. Tamburrini, M. Segev, R. Della Pergola, A.J. Agranat, Phys. Rev. Lett. 83, 1954 (1999).
11. W. Kleemann, R. Lindner, Ferroelectrics 199, 1 (1997).
12. L. Leuzzi, T.M. Nieuwenhuizen, Thermodynamics of the glassy state (Taylor & Francis, 2008).
13. S. Mossa, F. Sciortino, Phys Rev. Lett. 92, 045504-1 (2004).
14. J. Parravicini, A.J. Agranat, C. Conti, E. DelRe, submitted to Phys. Rev. Lett. (2012).
15. B. Crosignani, E. DelRe, P. Di Porto, A. Degasperis, Opt. Lett. 23, 912 (1998).
16. B. Crosignani, A. Degasperis, E. DelRe, P. Di Porto, A. J. Agranat, Phys. Rev. Lett. 82, 1664 (1999).
17. P. Ben Ishai, C.E.M. De Oliveira, Y. Ryabov, Y. Feldman, A.J. Agranat, Phys. Rev. B 70, 132104 (2004).
18. A. Gumennik, Y. Kurzwell-Segev, A.J. Agranat, Opt. Mater. Express 1, 332 (2011).
19. The final equilibration temperature is held through small-amplitude rapid adjustments that are omitted in the $S$ and $R$ curves.
20. E. DelRe, A. Ciattoni, E. Palange, Phys. Rev. E 73, 017601 (2006).
References

1. O. Firstenberg, P. London, M. Snuker, A. Ron, N. Davidson, “Elimination, reversal and directional bias of optical diffraction”, Nat. Physics 5, 665 (2009).
2. E. DelRe, E. Spinozzi, A.J. Agranat, C. Conti, “Scale-free optics and diffractionless waves in nanodisordered ferroelectrics” Nat. Photonics 5, 39 (2011).
3. G.A. Samara, “The relaxational properties of compositionally disordered ABO₃ perovskites”, J. Phys. Condens. Matter 15, R367 (2003).
4. A. Bokov, “Recent progress in relaxor ferroelectrics with perovskite structure”, J. Mater. Sci. 41, 31 (2006).
5. J. Parravicini, F. Di Mei, C. Conti, A.J. Agranat, E. DelRe, “Diffraction cancellation over multiple wavelengths in photorefractive dipolar glasses”, Opt. Express 19, 24109 (2011).
6. V. Folli, E. DelRe, C. Conti, “Beam Instabilities in the Scale-Free Regime” Phys. Rev. Lett. 108, 033901 (2012).
7. The model breaks down for scales at which the space charge saturates. This is a sample-dependent limit associated to the acceptor density N_A and the static ε_r. In typical KTN:Li samples N_A ≃ 2·10¹⁸ cm⁻³, so that even in the critical regime with ε_r ~ 10⁵, the model breaks down for beam widths l ~ 0.1 µm.
8. C. Conti, A.J. Agranat, E. DelRe, “Subwavelength optical spatial solitons and three-dimensional localization in disordered ferroelectrics: Toward metamaterials of nonlinear origin”, Phys. Rev. A 84, 043809 (2011).
9. F. Jona, G. Shirane, Ferroelectric crystals (Dover, 1993).
10. E. DelRe, M. Tamburrini, M. Segev, R. Della Pergola, A.J. Agranat, “Spontaneous self-trapping of optical beams in metastable paraelectric crystals” Phys. Rev. Lett. 83, 1954 (1999).
11. W. Kleemann, R. Lindner, “Dynamic behavior of polar nanodomains in PbMg₁/₃Nb₂/₃O₃”, Ferroelectrics 199, 1 (1997).
12. L. Leuzzi, T.M. Nieuwenhuizen, Thermodynamics of the glassy state (Taylor & Francis, 2008).
13. S. Mossa, F. Sciortino, “Crossover (or Kovacs) Effect in an Aging Molecular Liquid”, Phys Rev. Lett. 92, 045504-1 (2004).
14. J. Parravicini, A.J. Agranat, C. Conti, E. DelRe, “Kovacs and inverse Kovacs effect in the optical scale-free regime”, submitted to Phys. Rev. Lett. (2012).
15. B. Crosignani, E. DelRe, P. Di Porto, A. Degasperis, “Self-focusing and self-trapping in unbiased centrosymmetric photorefractive media”, Opt. Lett. 23, 912 (1998).
16. B. Crosignani, A. Degasperis, E. DelRe, P. Di Porto, A.J. Agranat, “Nonlinear optical diffraction effects and solitons due to anisotropic charge-diffusion-based self-interaction”, Phys. Rev. Lett. 82, 1664 (1999).
17. P. Ben Ishai, C.E.M. De Olivera, Y. Ryabov, Y. Feldman, A.J. Agranat, “Glass-forming liquid kinetics manifested in a KTN:Cu crystal”, Phys. Rev. B 70, 132104 (2004).
18. A. Gumennik, Y. Kurzweil-Segev, A.J. Agranat, “Electro-optical effects in glass-forming liquids at dipolar nano-clusters embedded in a paraelectric environment”, Opt. Mater. Express 1, 332 (2011).
19. The final equilibration temperature is held through small-amplitude rapid adjustments that are omitted in the S and R curves.
20. E. DelRe, A. Ciattoni, E. Palange, “Role of charge saturation in photorefractive dynamics of micron-sized beams and departure from soliton behavior”, Phys. Rev. E 73, 017601 (2006).