Rejuvenation in scale-free optics and enhanced diffraction cancellation life-time

J. Parravicini,[1,2] **C. Conti,**[1,3] **A. J. Agranat,**[4] **and E. DelRe**[1,2,*]

[1] *Department of Physics, University of Rome "La Sapienza," 00185 Rome, Italy*
[2] *IPCF-CNR, University of Rome "La Sapienza," 00185 Rome, Italy*
[3] *ISC-CNR, University of Roma "La Sapienza," 00185 Rome, Italy*
[4] *Applied Physics Department, Hebrew University of Jerusalem, 91904 Israel*
[*] *eugenio.delre@uniroma1.it*

Abstract: We demonstrate rejuvenation in scale-free optical propagation. The phenomenon is caused by the non-ergodic relaxation of the dipolar glass that mediates the photorefractive nonlinearity in compositionally-disordered lithium-enriched potassium-tantalate-niobate (KTN:Li). We implement rejuvenation to halt aging in the dipolar glass and extend the duration of beam diffraction cancellation.

OCIS codes: (160.2750) Glass and other amorphous materials; (190.4400) Nonlinear optics, materials; (190.5330) Photorefractive optics.

References and links

1. E. DelRe, E. Spinozzi, A. J. Agranat, and C. Conti, "Scale-free optics and diffractionless waves in nanodisordered ferroelectrics," Nat. Photonics **5**, 39–42 (2011).
2. J. Parravicini, F. Di Mei, C. Conti, A. J. Agranat, and E. DelRe, "Diffraction cancellation over multiple wavelengths in photorefractive dipolar glasses," Opt. Express **19**, 24109–24114 (2011).
3. C. Conti, A. J. Agranat, and E. DelRe, "Subwavelength optical spatial solitons and three-dimensional localization in disordered ferroelectrics: Toward metamaterials of nonlinear origin," Phys. Rev. A **84**, 043809 (2011).
4. J. Parravicini, C. Conti, A. J. Agranat, and E. DelRe, "Programming scale-free optics in disordered ferroelectrics," Opt. Lett. **37**, 2355–2357 (2012).
5. E. DelRe, J. Parravicini, A. J. Agranat, and C. Conti, "Kovacs and inverse Kovacs effect in the optical scale-free regime" in *Nonlinear Photonics*, OSA Technical Digest (online) (Optical Society of America, 2012), paper NTu3D.6.
6. J. Parravicini, A. J. Agranat, C. Conti, and E. DelRe, "Equalizing disordered ferroelectrics for diffraction cancellation," Appl. Phys. Lett. **101**, 111104 (2012).
7. V. Folli, E. DelRe, and C. Conti, "Beam instabilities in the scale-free regime," Phys. Rev. Lett. **108**, 033901 (2012).
8. J. D. Jackson, *Classical Electrodynamics*, 3rd ed. (John Wiley & Sons, New York, 1999).
9. Y. S. Kivshar and G. P. Agrawal, *Optical Solitons* (Academic Press, New York, 2003).
10. H. Kosaka, T. Kawashima, A. Tomita, M. Notomi, T. Tamamura, T. Sato, and S. Kawakami, "Self-collimating phenomena in photonic crystals," Appl. Phys. Lett. **74**, 1212–1214 (1999).
11. H. S. Eisenberg, Y. Silberberg, R. Morandotti, and J. S. Aitchison, "Diffraction management," Phys. Rev. Lett. **85**, 1863–1866 (2000).
12. O. Firstenberg, P. London, M. Shuker, A. Ron, and N. Davidson, "Elimination, reversal and directional bias of optical diffraction," Nat. Phys. **5**, 665–668 (2009).
13. A. J. Agranat, R. Hofmeister, and A. Yariv, "Characterization of a new photorefractive material: $K_{1-y}L_yT_{1-x}N_x$," Opt. Lett. **17**, 713–715 (1992).
14. A. Bokov, "Recent progress in relaxor ferroelectrics with perovskite structure," J. Mater. Sci. **41**, 31–52 (2006).
15. P. Ben Ishai, C. E. M. De Olivera, Y. Ryabov, Y. Feldman, and A. J. Agranat, "Glass-forming liquid kinetics manifested in a KTN:Cu crystal," Phys. Rev. B **70**, 132104 (2004).
16. N. Sapiens, A. Weissbrod, and A. J. Agranat, "Fast electroholographic switching," Opt. Lett. **34**, 353-355 (2009).

#175570 - $15.00 USD Received 5 Sep 2012; revised 20 Oct 2012; accepted 20 Oct 2012; published 19 Nov 2012

17. E. DelRe and M. Segev, "Self-focusing and solitons in photorefractive media" in *Topics in Applied Physics* vol. 114 (Springer, Berlin, 2009) pp. 547–572.
18. G. A. Samara, "The relaxational properties of compositionally disordered ABO_3 perovskites," J. Phys. Condens. Matter **15**, R367 (2003).
19. E. Donth, *The Glass Transition* (Springer-Verlag, Berlin & Heidelberg, 2001).
20. L. Leuzzi and T.M. Nieuwenhuizen, *Thermodynamics of the Glassy State* (Taylor & Francis, New York & London, 2008).
21. J. P. Bouchaud, P. Doussineau, T. de Lacerda-Arôso, and A. Levelut, "Frequency dependence of aging, rejuvenation and memory in a disordered ferroelectric," Eur. Phys. J. B **21**, 335–340 (2001).
22. K. Jonason, E. Vincent, J. Hamman, J. P. Bouchaud, and P. Nordblad, "Memory and chaos effect in spin glasses," Phys. Rev. Lett. **81**, 3243–3246 (1998).
23. R. Mathieu, M. Hudl, and P. Nordblad, "Memory and rejuvenation in a spin glass," Eur. Phys. Lett. **90**, 67003 (2010).
24. S. Mossa and F. Sciortino, "Crossover (or Kovacs) effect in an aging molecular liquid," Phys Rev. Lett. **92**, 045504 (2004).
25. G. Parravicini, M. Campione, F. Marabelli, M. Moret, and A. Sassella, "Experimental assesment of nonergodicity in tetracene single crystals," Phys. Rev. B **86**, 024107 (2012).
26. A. Gardchareon, R. Mathieu, P. E. Jonsson, and P. Nordblad, "Strong rejuvenation in chiral-glass superconductors," Phys. Rev. B **67**, 052505 (2003).
27. V. V. Shvartsman, S. Bedanta, P. Borisov, W. Kleemann, A. Trach, and P. M. Vilarinho, "(Sr, Mn)TiO(3): A magnetoelectric multiglass," Phys. Rev. Lett. **101**, 165704 (2008).
28. N. Gofraniha, C. Conti, and G. Ruocco, "Aging of the nonlinear optical susceptibility in doped colloidal suspensions," Phys. Rev. B **75**, 224203 (2007).
29. N. Gofraniha, C. Conti, G. Ruocco, and F. Zamponi, "Time-dependent nonlinear optical susceptibility of an out-of-equilibrium soft material," Phys. Rev. Lett. **102**, 038303 (2009).
30. C. Conti and E. DelRe, "Optical supercavitation in soft matter," Phys. Rev. Lett. **105**, 118301 (2010).
31. P. Doussineau and A. Levelut, "Memory against temperature or electric field sweeps in potassium niobio-tantalate crystals," Eur. Phys. J. B **26**, 13–21 (2002).
32. S. Sahoo, O. Petracic, W. Kleemann, P. Nordblad, S. Cardoso, and P. P. Freitas "Aging and memory in a superspin glass," Phys. Rev. B **67**, 214422 (2003).
33. B. Crosignani, E. DelRe, P. Di Porto, and A. Degasperis, "Self-focusing and self-trapping in unbiased centrosymmetric photorefractive media," Opt. Lett. **23**, 912–914 (1998).
34. B. Crosignani, A. Degasperis, E. DelRe, P. Di Porto, and A. J. Agranat, "Nonlinear optical diffraction effects and solitons due to anisotropic charge-diffusion-based self-interaction," Phys. Rev. Lett. **82**, 1664–1667 (1999).

1. Introduction

Supercooled photorefractive relaxors support a regime of undistorted beam propagation termed "scale-free optics" [1–7]. In distinction to linear waveguide modes [8] and spatial solitons [9], this occurs without the scale-dependent constraints typical of linear and nonlinear diffraction compensation [1], a diffraction cancellation that is reminiscent of effects observed in linear periodic systems [10, 11] and self-induced transparency [12]. Scale-free optics possesses considerable applicative potential because, in principle, it can greatly increase the resolution of image transmission [1, 3, 7]. The effect emerges in lithium-enriched potassium-niobate-tantalate (KTN:Li) [13], a photorefractive ferroelectric crystal with compositional disorder that manifest pronounced relaxor behavior [14, 15]. At lower levels of Li content, the crystals (i.e., KLTN) support electroholography [16] and optical spatial solitons [17]. When the relaxor is in a nonequilibrium state [1, 4–6], i.e., when the hosted glass of polar nanoregions (PNRs) is rapidly cooled near the ferroelectric room-temperature Curie point T_C [1, 14, 18], the PNRs display an anomalously large static susceptibility which activates the scale-free optics [14].

A key feature is that scale-free optics appears in an out-of-equilibrium phase and manifests aging [19, 20], a phenomenon that is well known also in other relaxor ferroelectrics [14, 21] as well as in a great variety of other systems, such as spin-glasses [22, 23], polymers [24], organic semiconductors [25], superconductors [26], multiferroics [27] and soft matter optical nonlinearity [28–30]. Aging causes diffraction cancellation to persist only for a finite life-time which depends on the specific experimental conditions. The effect disappears as the glassy state

#175570 - $15.00 USD Received 5 Sep 2012; revised 20 Oct 2012; accepted 20 Oct 2012; published 19 Nov 2012

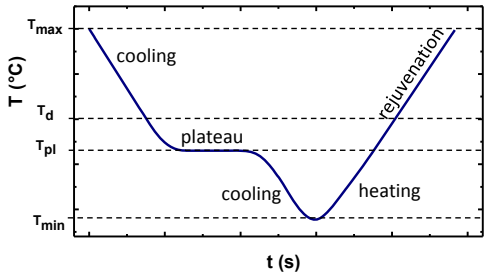

Fig. 1. Basic temperature variation protocol for rejuvenation experiments (as described e.g. in [21]): the sample is cooled at a constant rate from $T_{\max}$ to an intermediate temperature $T_{\rm pl}$ below the (dynamic) glass transition temperature T_d, where cooling is interrupted and an isothermal evolution takes place, for a given time interval (plateau); cooling then resumes down to $T_{\min}$ and is immediately followed by a steady heating at an opposite rate from $T_{\min}$ to $T_{\max}$.

of the PNRs relaxes [6]. Methods to forestall this relaxation and extend the life-time of scale-free optics appear crucial to its development into a practical high-resolution imaging technique.

Interestingly, in some systems that age, even after the physical parameters of the disordered system have been allowed to freely evolve and relax, the use of a specifically tailored thermal protocol $T = T(t)$ can cause the "aged" parameters to be restored to the value they had before the relaxation: the system is said to be "rejuvenated" [31]. In its basic realization, rejuvenation is achieved using the thermal trajectory $T = T(t)$ illustrated in Fig. 1.

As a manifestation of history-dependence, rejuvenation is, along with the so-called cross-over effect, a consequence of the chaotic nature of the glassy phase [32]. Most importantly, aging and rejuvenation is observed in the dielectric and susceptibility of relaxors ferroelectrics [21, 23, 31].

In this work we harness rejuvenation of the dielectric susceptibility of the dipolar glass hosted in relaxor KTN:Li to achieve rejuvenation in the scale-free optical response. The effect is used to extend the life-time of the diffraction cancellation regime, increasing the time stability of the scale-free dynamics. Through rejuvenation, we are able to reverse the decay of the scale-free regime and lengthen the diffraction cancellation regime by more than three times (from an original time interval of 120 s to more than 400 s in our experiments).

2. Methods

The scale-free optical regime exploits the diffusive nonlinearity of the photorefractive effect [33, 34]. The propagation of a beam in the z direction with wavelength λ is governed by [1–3]

$$2ik\frac{\partial A}{\partial z} + \nabla_{\perp}^2 A - \left(\frac{L}{\lambda}\right)^2 \frac{(\nabla_{\perp} I)^2}{4I^2} A = 0 \tag{1}$$

where k is the wave vector, $I = |A|^2$ is the intensity of the field envelop A and the parameter L is defined by the relationship

$$L = 4\pi n_0^2 \varepsilon_0 \sqrt{g}\, \chi_{\rm PNR}(K_B T/q). \tag{2}$$

Here g is the effective quadratic electro-optic coefficient, $\chi_{\rm PNR}$ is the effective history-dependent low-frequency dielectric susceptibility of the dipolar glass (due to PNRs), K_B the

#175570 - $15.00 USD Received 5 Sep 2012; revised 20 Oct 2012; accepted 20 Oct 2012; published 19 Nov 2012

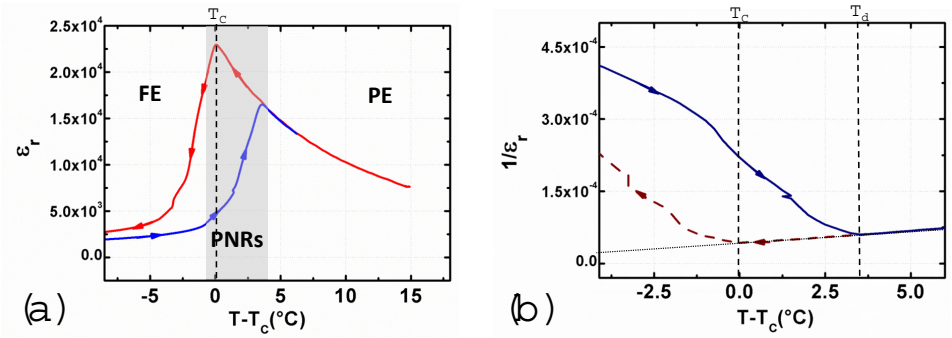

Fig. 2. (a) Identification of the region where dielectric response is dominated by PNRs. KTN:Li static dielectric constant ε_r for slow cooling (red curve) and slow heating (blue curve) as a function of temperature ($T_C = 14.5$ °C). The regions of paraelectric (PE) and ferroelectric (FE) behavior are unable to support scale-free propagation. The glassy region where PNRs can support scale-free propagation is indicated with the shading, where marked thermal hysteresis is detected. (b) $1/\varepsilon_r$ vs. $(T - T_C)$ plot identifying $T_d \simeq 18.2$°C as the dynamic glass transition temperature when the mean-field Curie-Weiss law (dotted line) breaks down [14, 20].

Boltzmann constant, T the crystal equilibration temperature (i.e. the temperature measured at a given istant) and q is the charge of the photoexcited carriers. The regime of diffraction cancellation is obtained when $L = \lambda$, so that for Gaussian beams diffraction ceases independently of beam size or intensity (scale-free regime).

To investigate the rejuvenation phenomenon in scale-free optics we employ a $6\times3\times2.5$mm sample of KTN:Li doped with Cu [1, 15]. The optical setup is similar to that described in [1]: a TEM_{00} x-polarized light beam from a He-Ne laser ($\lambda = 632.8$ nm) propagating in the z direction is focused onto the input facet of the sample. The intensity distribution of the beam at the output of the crystal is imaged and then recorded by a CCD camera. The peak intensity is $I_p \simeq 7$ W/cm^2 and the input beam intensity distribution Full-Width-at-Half-Maximum (FWHM) $\Delta r_{\rm in} = \Delta x_{\rm in} \simeq \Delta y_{\rm in} \simeq 11$ μm. The temperature T is fixed by a computer-controlled Peltier-junction.

3. Results and discussion

Equations (1) and (2) predict a strong dependence of optical propagation on the static $\chi_{\rm PNR}$. The requirement of $L = \lambda$ implies an unusually high value of $\chi_{\rm PNR} \sim 10^5$, as observed only in proximity of T_C. Furthermore, the crystal must be in a non-ergodic phase where PNRs form, so as to avoid critical opacity. In Fig. 2(a) we report the quasi-static dielectric response ε_r versus $T - T_C$ ($T_C = 14.5$°C) obtained from capacitance measurements at slow heating and cooling rates ($|\Delta T/\Delta t| \simeq 0.01$ °C/s) [5, 6]. The marked hysteresis in $\varepsilon_r(T - T_C)$ (shaded region in Fig. 2(a)) signals the glassy non-ergodic phase near T_C, where the formation of PNRs takes place, and the dynamic glass transition temperature $T_d \simeq 18.2$°C is identified with the breakdown of the Curie-Weiss law (Fig. 2(b)) [14, 20].

In order to follow the trend of diffraction cancellation as a function of time we introduce the dimensionless parameter S

$$S = \frac{\Delta r_{\rm d} - \Delta r}{\Delta r_{\rm d} - \Delta r_{\rm in}} \tag{3}$$

where $\Delta r_{\rm d}$ is the FWHM of the exiting Gaussian beam due to the standard linear diffraction,

#175570 - $15.00 USD Received 5 Sep 2012; revised 20 Oct 2012; accepted 20 Oct 2012; published 19 Nov 2012

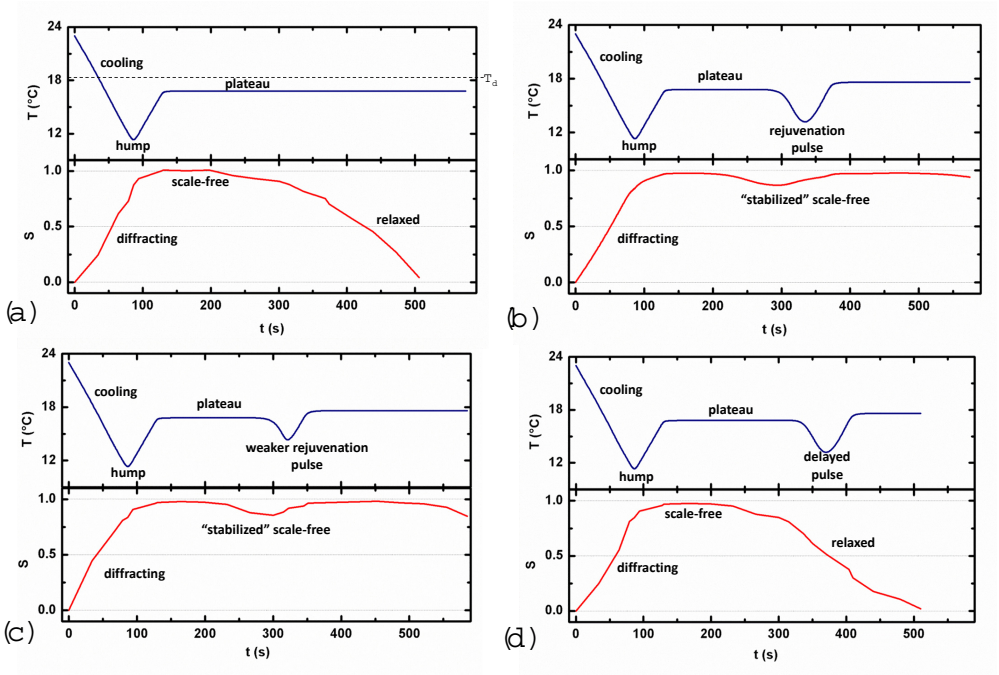

Fig. 3. Optical rejuvenation. (a) Standard thermal protocol $T = T(t)$ to activate scale-free propagation with the corresponding trend of diffraction cancellation S as a function of time; (b) rejuvenation thermal protocol and stabilization of S to its maximum value for a $\Delta T \simeq 3.5°$C pulse activated when $S \simeq 0.85$; (c) rejuvenation for a $\Delta T \simeq 2.5°$C pulse activated when $S \simeq 0.86$; and (d) absence of rejuvenation for a $\Delta T \simeq 3.5°$C activated when $S \simeq 0.73$.

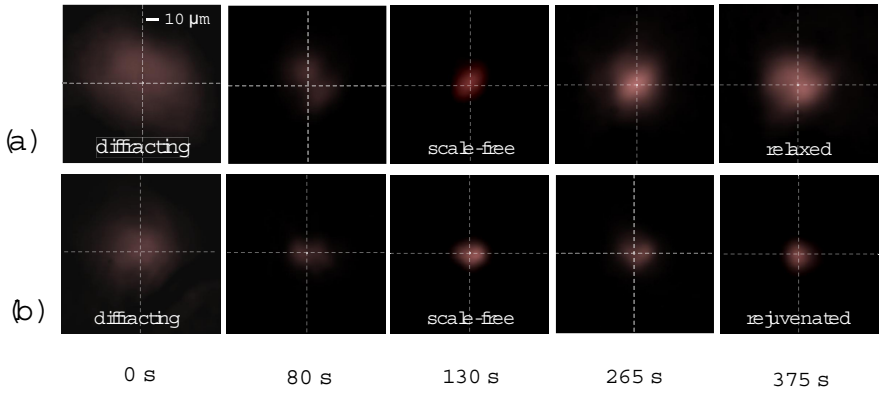

Fig. 4. Intensity distribution at the output facet of the crystal for standard (a) and rejuvenating (b) thermal protocols, as in respectively Figs. 3(a)-(b) and 3(e)-(f).

Δr_{in} is the FWHM of the Gaussian beam at the input facet of the crystal and Δr is the FWHM of the exiting beam measured as a function of time during the experiment. $S = S(\Delta r)$ allows us

#175570 - $15.00 USD Received 5 Sep 2012; revised 20 Oct 2012; accepted 20 Oct 2012; published 19 Nov 2012

to pinpoint the occurrence of scale-free regime, because it varies continuously from 0, i.e., for standard linear diffraction ($L = 0$, $\Delta r = \Delta r_{\rm d}$), to 1, when diffraction is fully cancelled ($L = \lambda$, $\Delta r = \Delta r_{\rm in}$).

Figure 3 reports optical rejuvenation of diffraction cancellation. Figure 3(a) shows the thermal protocol to activate scale-free propagation used and the resulting $S(t)$. The sample is cooled at a constant rate $\alpha = \Delta T/\Delta t \simeq -0.13$ °C/s below T_C and then heated at $-\alpha$ up to an equilibration temperature of $T = 16.8$ °C which is maintained constant.

At $t = 0$ diffraction is linear ($S = 0$); after a transient of $\simeq 130$ s it comes to a complete deletion ($S \simeq 1$, $L \simeq \lambda$). This regime is maintained for 100-150 s. Then it suffers aging, and relaxes to the original diffractive regime ($S \to 0$).

Figure 3(b) reports the thermal protocol and $S(t)$ for a rejuvenation process. The sample is cooled at a constant rate $\alpha \simeq -0.13$ °C/s below T_C and then heated at $-\alpha$ up to an equilibration temperature $T = 16.8$ °C and kept at that T. The scale-free regime arises as in Fig. 3(a) at $t \simeq 130$ s and remains steady for a time interval of about 110 s, when it begins to relax and disappear, with a time scale of tens of seconds. At $t \simeq 300$ s, S has decreased by 15%. A rejuvenation pulse is thus applied (a cooling at the rate α, followed by heating at $-\alpha$) up to an equilibration temperature of 17.6 °C. The beam stops to broaden and focuses again: at $t = 370$ s diffraction cancellation is fully restored. The rejuvenation depends on the amplitude and time position of the thermal pulse, as shown in Fig. 3(c) and (d). Specifically, a thermal pulse with an amplitude less than $\Delta T \simeq 2$°C and activated after the beam has decayed to below $S \simeq 0.75$ fails to produce a rejuvenation back to $S \simeq 1$. We note that, in distinction to the basic standard rejuvenation protocols of Fig. 1, our protocol has a "hump" before the plateau, necessary to activate the scale-free regime [5, 6]. For comparison, in Figs. 4(a) and 4(b) we report the transverse intensity distribution of the beam in the two conditions of respectively the standard scale-free formation of Fig. 3(a) and the rejuvenation protocol of Fig. 3(b). Even though the time dynamics depend on the specific value of the input intensity, as is known to occur for all photorefractive phenomena, the rejuvenated beam is observed to consistently reach the maximum value $S \simeq 1$ irrespective of the actual value of I_p in the range of 3-20 W/cm^2, as compatible with scale-free propagation [1].

The causal relationship between the details of the protocol and its effect on glassy state is still a matter of study [20]: we are here interested in demonstrating the possibility of activating an optical rejuvenation effect harnessing the nonequilibrium features of the material.

4. Conclusions

In conclusion, we have demonstrated how rejuvenation can be observed in a disordered ferroelectric and used to stabilize scale-free propagation. The phenomenon involves the glassy nature of dielectric polar-nano-regions inside the crystal. Results are a direct evidence of how optics can make use of out-of-equilibrium solid-state mechanisms.

Acknowledgments

We thank G.B. Parravicini for dielectric measurements and M. Deen Islam for technical assistance. This work was supported by funding from the FIRB grant PHOCOS-RBFR08E7VA of the Italian Ministry of Research, from the E.R.C. under the European Community 7th Framework Program (FP7/2007-2013)/ERC grant agreement no.201766, from SMARTCONFOCAL project of the *Regione Lazio* and from the PRIN project no.2009P3K72Z. A.J.A. acknowledges the support of the P. Brojde Center for Innovative Engineering.

#175570 - $15.00 USD Received 5 Sep 2012; revised 20 Oct 2012; accepted 20 Oct 2012; published 19 Nov 2012