Optical shock waves in silica aerogel

S. Gentilini,[1,*] **F. Ghajeri,**[2] **N. Ghofraniha,**[3] **A. Di Falco,**[2] **and C. Conti**[1,4]

[1] *ISC-CNR c/o Department of Physics, University of Rome La Sapienza, P.le A. Moro 2, 00185, Rome, Italy*
[2] *School of Physics and Astronomy, University of St.Andrews, KY16 9SS Fife, UK*
[3] *IPCF-CNR c/o Physics Department, University of Rome La Sapienza, P.le A. Moro 2, 00185, Rome, Italy*
[4] *Physics Department, University of Rome La Sapienza, P.le A. Moro 2, 00185, Rome, Italy*

[*] *silvia.gentilini@roma1.infn.it*

Abstract: Silica aerogels are materials well suited for high power nonlinear optical applications. In such regime, the non-trivial thermal properties may give rise to the generation of optical shock waves, which are also affected by the structural disorder due to the porous solid-state gel. Here we report on an experimental investigation in terms of beam waist and input power, and identify various regimes of the generation of wave-breaking phenomena in silica aerogels.

OCIS codes: (190.0190) Nonlinear optics; (190.4400) Nonlinear optics, materials; (190.4720) Optical nonlinearities of condensed matter; (160.4330) Nonlinear optical materials.

References and links

1. C. N. Likos, "Effective interactions in soft condensed matter physics," Phys. Rep. **348,** 267–439 (2001).
2. M. Anyfantakis, A. Koniger, S. Pispas, W. Kohler, H. Buth, B. Loppinet, and G. Fytas, "Versatile light actuated matter manipulation in transparent non-dilute polymer solutions," Soft Matter **8,** 2382–2384 (2012).
3. I.C. Khoo, *Liquid Crystals: Physical Properties and Nonlinear Optical Phenomena* (Wiley, 1995).
4. R.W.Boyd, *Nonlinear Optics* (Academic Press, 2002).
5. C. Conti, N. Ghofraniha, G. Ruocco, and S. Trillo, "Laser beam filamentation in fractal aggregates," Phys. Rev. Lett. **97,** 123903 (2006).
6. W.M. Lee, R. El-Ganainy, D.N. Christodoulides, K. Dholakia, and E.M. Wright, "Nonlinear optical response of colloidal suspensions," Opt. Exp. **17,** 10277–10289 (2009).
7. E. DelRe, E. Spinozzi, R. Agranat, and C. Conti, "Scale free optics and diffractionless waves in nano-disordered ferroelectrics," Nature Photonics **5,** 39–42 (2011).
8. N. Ghofraniha, C. Conti, and G. Ruocco, "Aging of the nonlinear optical susceptibility in doped colloidal suspensions," Phys. Rev. B **75,** 038303 (2007).
9. N. Ghofraniha, 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).
10. A. Ashkin, J.M. Dziedzic, and P.W. Smith, "Continuous-wave self-focusing and self-trapping of light in artificial Kerr media," Opt. Lett. **7,** 276–278 (1982).
11. S. Gentilini, N. Gofraniha, E. DelRe, and C. Conti, "Shock waves in thermal lensing," Phys. Rev. A **87,** 053811 (2013).
12. C. Conti, and E. DelRe, "Optical supercavitation in soft matter," Phys. Rev. Lett. **105,** 118301 (2010).
13. M.A. Aegerter, N. Leventis, and M.M. Koebel, *Advances in Sol-gel Derived Materials and Technologies* (Springer, 2011).
14. J.T. Seo, Q. Yang, S. Creekmore, B.Tabibi, D.Temple, S.Y. Kim, K.Yoo, A. Mott, M. Namkung, and S.S. Yung, "Large pure refractive nonlinearity of nanostructure silica aerogel," Appl. Phys. Lett. **82,** 4444–4446 (2003).
15. J. T. Seo, S. M. Mao, Q. Yang, L. Creekmore, H. Brown, R. Battle, K. Lee, A. Jackson, T. Skyles, B. Tabibi, K. P. Yoo, S. Y. Kim, S. S. Jung, and M. Namkung, "Large optical nonlinearity of highly porous silica nanoaerogels in the nanosecond time domain," J. Korean Phys. Soc. **48**, 1395–1399 (2006).
16. N. Ghofraniha, L. Amato Santamaria, V. Folli, S. Trillo, E. DelRe, and C. Conti, "Measurement of scaling laws for shock waves in thermal nonlocal media," Opt. Lett. **37,** 2325–2327 (2012).

#201088 - $15.00 USD Received 8 Nov 2013; revised 22 Dec 2013; accepted 25 Dec 2013; published 16 Jan 2014

17. N. Ghofraniha, S. Gentilini, V. Folli, E. DelRe, and C. Conti, "Measurement of scaling laws for shock waves in thermal nonlocal media," Phys.Rev.Lett. **109,** 243902 (2012).
18. S. Gentilini, N. Ghofraniha, E. DelRe, and C. Conti, "Shock wave far-field in ordered and disordered nonlocal media," Opt.Expr. **20,** 27369–27375 (2012).
19. J.C.Bronsky and D. McLaughlin, *Singular Limits of Dispersive Waves* (Plenum, 1994).
20. M.A. Hoefer, M.J. Ablowitz, I. Coddington, E.A. Cornell, P. Engels, and V. Schweikhard, "Dispersive and classical shock waves in Bose-Einstein condensates and gas dynamics," Phys. Rev. A **74,** 023623 (2006).
21. W. Wan, S. Jia, and J.W. Fleischer, "Dispersive superfluid-like shock waves in nonlinear optics," Nature Physics **3,** 46–51 (2007).
22. C. Barsi, W. Wan, C. Sun, and J.W. Fleischer, "Dispersive shock waves with nonlocal nonlinearity," Opt. Lett. **32,** 2930–2932 (2007).
23. N. Ghofraniha, C. Conti, R. Ruocco, and S. Trillo, "Shocks in nonlocal media," Phys. Rev. Lett. **99,** 043903 (2007).
24. C. Conti, A. Fratalocchi, M. Peccianti, and G. Ruocco, "Observation of a gradient catastrophe generating solitons," Phys. Rev. Lett. **102,** 083902 (2009).
25. A. Armaroli, S. Trillo, and A. Fratalocchi, "Suppression of transverse instabilities of dark solitons and their dispersive shock waves," Phys. Rev. A **80,** 053803 (2009).
26. W. Wan, V. Dylov, C. Barsi, and J.W. Fleischer, "Diffraction from an edge in a self-focusing medium," Opt. Lett. **35,** 2819–2821 (2010).
27. A. Venkateswara Rao and P.B. Wagh, "Preparation and characterization of hydrophobic silica aerogels," Mat. Chem. and Phys. **53,** 13–18 (1998).

1. Introduction

Complex materials as soft-colloidal matter, polymers, and liquid crystals [1], are attracting growing interest for their relevance in fundamental physics studies and for their potential use in applications including biophysics, imaging and sensing. The nonlinear optical properties of complex materials are particularly interesting and result from a variety of competing phenomena, as photo-chemical [2] and re-oriental effects [3], electrostriction [4–6], photo-refraction [7] and aging [8, 9]. Their interplay may sustain strong nonlinear phenomena, but is often accompanied by strong scattering losses, which force to analyze the role of randomness on nonlinear waves and nonlinear optical processes [10, 11]. Additionally, most of the liquid solutions and/or colloidal dispersions are also largely affected by thermally driven diffusion and convection [12]. These are very also detrimental for practical applications and make soft-colloidal samples useless for high power regimes.

Silica aerogel (SA) is a very interesting candidate material in which a convection-free solid-state with complex structure on nanometric-scale sustains strong nonlinearities in the presence of a reduced amount of scattering, because of the very low volume-average refractive index [13]. Recently the nonlinear optical response of SA has been reported [14, 15]. Results indicate that SA are potentially well suited for very high power applications, as optical limiters and optical switching. This suggests to consider the propagation and the generation of non-linear optical waves in SA, with special reference to spatial effects, as they are expected to be very relevant at high power regimes, a topic so far un-explored.

SA presents a thermal de-focusing nonlinearity due to light absorption and temperature dependent refractive index. Aerogel are excellent thermal insulators because of the micro-porous structure, which prevents the formation of convection cells, and hence, allows steep temperature-gradient profiles. This also results in large spatial modulations of nonlinear optical refractive index.

This subtle interplay between the low thermal conductivity and silica absorption allows to envisage that thermal lensing may occur in SA, however the peculiar heat-transport properties and the presence of disorder make such an effect much more complicated than in standard homogeneous materials.

It is known that thermal lenses may be formed in colloidal solutions, and lead to the generation of optical dispersive shock waves (DSWs) [11, 16–18], which may eventually be inhib-

#201088 - $15.00 USD Received 8 Nov 2013; revised 22 Dec 2013; accepted 25 Dec 2013; published 16 Jan 2014

ited when the strength of disorder is high enough. DSWs in optics are generated by an abrupt change in the phase of the optical field, corresponding to a singular mathematical solution of the relevant nonlinear wave equation in the so-called hydrodynamical limit of the model. This singularity is smoothed by the occurrence of a modulated wave-train. DSWs are here characterized by these regularizing fast oscillations (*undular bores*) observable in the intensity profile transverse to the propagation direction [19–26].

For SA, it is important to determine the conditions for the suppression of the DSW, relevant for any kind of nonlinear optical device. A key difference with respect to previous studies on shock waves in random media is the fact that in SA disorder does not change with time because of the absence of material fluctuations (e.g., Brownian motion of colloidal beads). In addition SA may be irradiated by very high power levels with tightly focused beams without melting or boiling; the resulting phenomenology can be very rich and SA turns out to be very useful for the investigation of highly nonlinear dynamics in the continuous wave (CW) regime.

In the following we report on an experimental investigation of thermal lensing in SA and show that DSW may be inhibited by disorder. Different regimes for the shock dynamics are determined, and we found that the sample may sustain different nonlinear optical phenomenology.

2. Sample preparation and experimental set-up

The SA samples are prepared following a base-catalyzed sol-gel procedure [27]. We mix equal amounts of a 1 : 1 solution of of tetramethoxysilane (TMOS) and methanol (MeOH) and a 2 : 1 solution of methanol and deionised water enriched with NHO_4H (in a ratio 1000 : 5.4). The solution is then poured in polymethylmethacrylate (PMMA) cuvettes with a square section of 1mm$\times$1mm and left to gel at room temperature. After gelling, the samples are immersed in acetone to dissolve the PMMA mold and promote the ageing process. The acetone is eventually removed by supercritical drying of the alcogels in a critical point drier.

The sample used in the nonlinear experiment has matter density $\rho = 0.215$ g/cm^3 and refractive index n$=$ 1.074: both these parameters determine the degree of scattering. Experiments are done at varying input laser power P_{in} and impinging point on the sample. A continuous-wave (CW) solid state TEM_{00} laser at wavelength $\lambda = 532$ nm is focused on the input SA facet, with sample length $L = 8.5$ mm along the propagation direction Z (the aerogel shrinks during the drying process). A photograph of typical SA sample illuminated with a collimated beam is shown in Fig. 1.

The experimental setup is shown in the inset of Fig. 1(a). We vary the input beam waist w_0 and record the transmitted intensity distribution $I(X,Y)$ by a CCD camera. Figure 1(b) shows the transmission versus input power for the various explored beam waists. The transmission is calculated as the ratio between the incident laser power P_{in} and the output power P_{out}. Each curve is obtained averaging over three realizations of disorder. The reported curves reveal that most of the input beam is transmitted in regimes with and without DSW, and that the transmission is nearly independent of the input power and beam waist.

3. Experimental results

Figure 2 shows the intensity distribution profiles as collected by the CCD camera at the output of the sample by varying the incident laser power, P_{in}, and the laser beam size, w_0.

In correspondence of regions of the SA sample displaying low enough disorder, a transition from scattering dominated regimes to nonlinear regimes is present: at moderate powers DSWs are not observed because of scattering losses, at high powers DSWs can be generated. Notably, this behaviour is not found in the case of liquid samples [18], where increasing power at high disorder largely enhances convective and thermophoretic effect, and further suppresses DSW

#201088 - $15.00 USD Received 8 Nov 2013; revised 22 Dec 2013; accepted 25 Dec 2013; published 16 Jan 2014

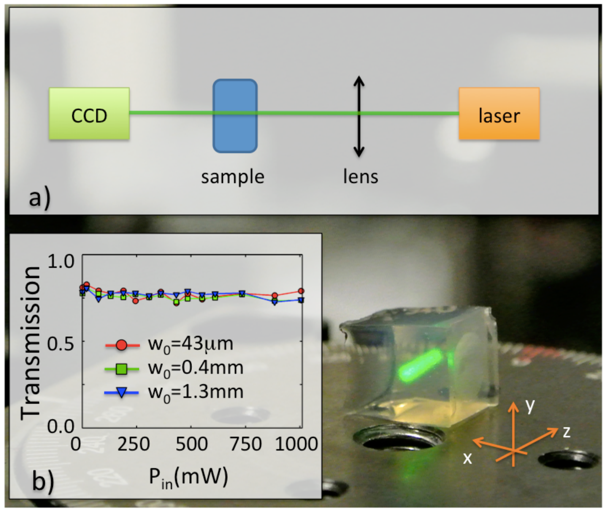

Fig. 1. Picture of a typical aerogel sample. a) Sketch of the experimental setup. b) Measured optical transmission versus input power, P_{in}, obtained by impinging on the SA sample with different laser beam diameter.

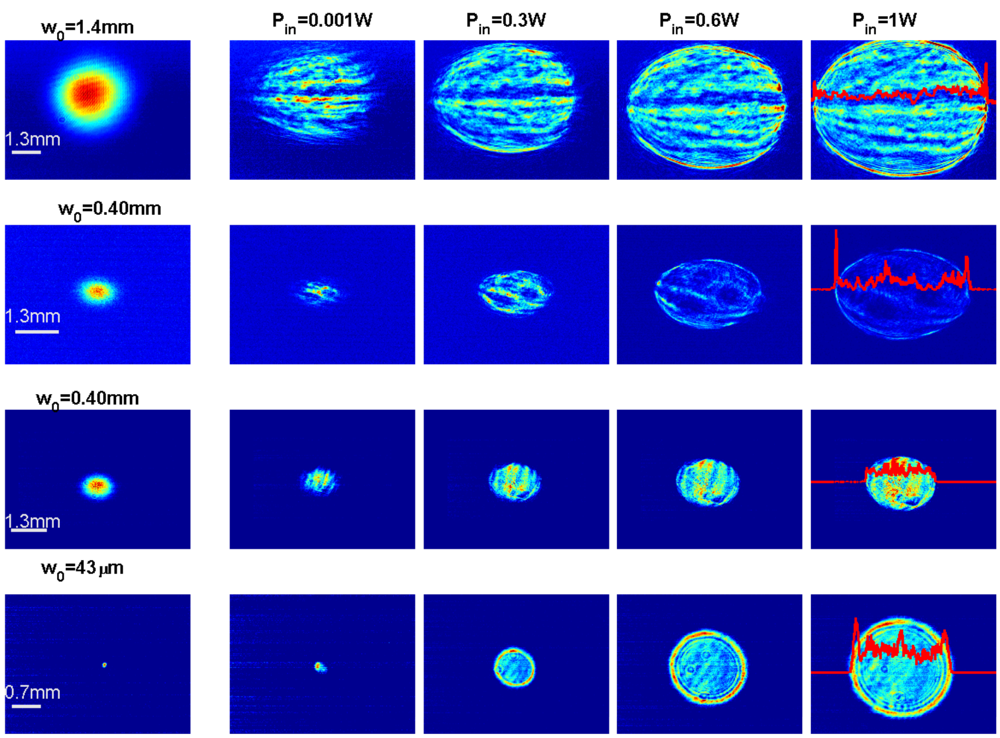

Fig. 2. Far field intensity profiles at the output of the SA for various input conditions: laser power P_{in} ranging from 1 mW to 1 W, and input beam waists w_0 as indicated over the images of the first column, where are reported the far field intensity profiles of the input beam. Note that the images in the second and third rows correspond to the same experimental conditions (in term of incident laser power and beam size), for different positions of the incident laser beam.

generation.

As shown in top panels of Fig. 2, for beam waist $w_0 = 1.4$ mm, at low power P_{in}, we do not observe DSW; but when increasing the input fluence, rings appear accompanied by an intensity dependent beam-spreading; this is a regime allowing DSW but disorder dominated at low intensity.

At smaller beam waists, the competition between nonlinearity and randomness is such that the specific disorder realization determines the occurrence of the shock. In the second raw the occurrence of DSW is observed, while the third raw shows, at the very same power and beam waist (i.e. $w_0 = 0.40$mm), a case in which disorder is dominant. The two regimes are only distinct because they correspond to different spatial position in the same sample.

The last raw, for strongly focused beams ($w_0 = 43\ \mu$m), corresponds to a regime in which disorder is totally frustrated by the nonlinear response, which always prevails.

As also previously demonstrated [11], in the far field, the optical shock waves result into an angular aperture θ that varies linearly with power. In order to further support the observation of DSW in the disordered SA we measure the angular aperture θ versus input power P_{in}, after the radial average of the $I(x,y)$ profiles summarized in Fig. 2. Figure 3 shows θ vs P_{in}, for $w_0 = 0.40$mm on three different input-beam spatial locations of the SA sample. For the considered $w_0 = 0.40$mm, there is a pronounced dependence of the angular aperture with respect to specific disorder realization (input beam position). Retaining the linear growth of θ with P_{in} a signature of the DSWs formation, we note that at the very same power, material density and volume-averaged refractive index, different disorder realizations may produce very different propagation regimes. In fact for two of the three investigated SA sample regions (squares and circles), the angular aperture, θ, takes a constant value for increasing P_{in} as long as, reached a threshold value, it starts to linearly increase. The slope of such a linear growth, as evidenced by the linear fits (continuous and dashed lines), depends on the amount of disorder and, hence, it can be regarded as a measurement of the scattering strength encountered by the propagating beam. In the other explored region of the sample, represented by the triangles, a linear growth with P_{in} does not emerge, signature of the fact that the scattering suffered by the laser beam in that point is enough to totally inhibit the occurrence of the wave breaking phenomenon.

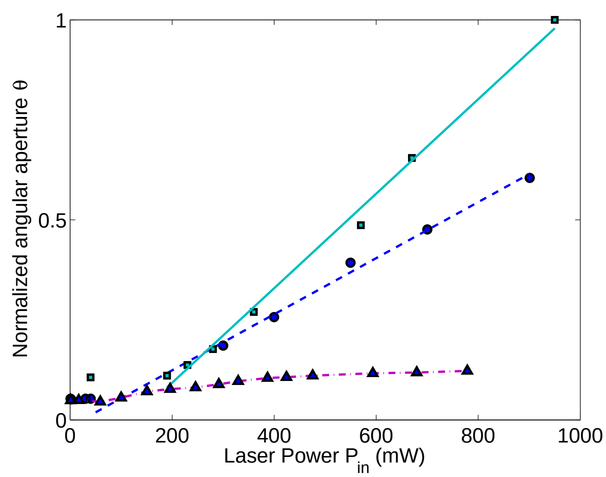

Fig. 3. Angular aperture of the transmitted pulse at the exit face of the sample after impinging with an input laser waist $w_0 = 0.4$mm in different points of the sample.

The whole set of possible spatial responses is achievable in SA by varying the disorder real-

#201088 - $15.00 USD Received 8 Nov 2013; revised 22 Dec 2013; accepted 25 Dec 2013; published 16 Jan 2014

ization and the degree of beam focusing. This is also related to the complex thermal response of the SA.

To show that the nonlinear effect has a thermal origin we study the time dynamics of the formation of the shock waves; in Figs. 4(a)–4(c) we show three snapshots at different instants of the shock formation due to the impinging of a $w_0 = 50\mu m$ and $P_{in} = 0.5W$ laser beam. The very low time-dynamics due to the low thermal conductivity of SA allows to follow the dynamics of the DSW formation in the presence of disorder. From the images we extract the far-field standard deviation given in Fig. 4(d) for different realizations of the measurements obtained by varying the beam waist. The two curves show that the characteristic time scale is of the order of ten seconds, afterward a steady state is reached.

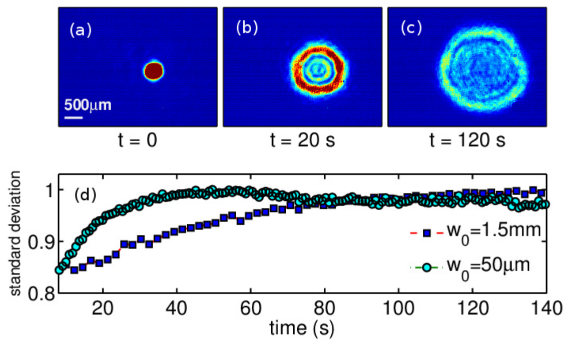

Fig. 4. Time resolved dynamics of the shock waves: (a-c) different intensity profiles at given instants; (d) time dynamics of the far-field width for $P_{in} = 200\ mW$ and two different input beam waists.

It is important to remark the origin of the nonlinear defocusing effect. Around room temperature the thermal nonlinearity of bulk silica is focusing. On the contrary, as also found by other authors [15], we observe a defocusing action in SA due to a decrease of the refractive index with laser power. This response is, hence, to be attributed to the specific porous structure of SA: the thermal induced expansion of the silica scaffold locally reduces the SA density and, hence, the refractive index of the material. The Kerr coefficient of SA is $n_2 \sim -4 \times 10^{-14} m^2/W$ [15], in our experiment this corresponds to a variation of $\Delta n = 10^{-4}$.

4. Conclusion

We reported on the first experimental investigation of spatial shock waves in silica aerogel. We found that this kind of material is very robust with respect to high power input beam in continuous wave regime and does not show any boiling or melting phenomena. This property permits to study high intensity nonlinear waves in the presence of disorder in regimes typically unaccessible by liquid materials. The subtle interplay between disorder and nonlinearity shows that proper material properties may largely hamper the generation of self-phase modulation effect, thus favoring the fabrication of high power devices. SA are hence very promising materials for nonlinear optics.

Acknowledgments

Andrea Di Falco acknowledges support from EPSRC (EP/I004602/1 and EP/J004200/1).