Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl19

Photophysical Processes Involved in Creation of Dark Spatial Solitons in Composite Photonic Media

Anna Samoć [a], Marek Samoć [a], Maneerat Woodruff [a] & Barry Luther-davies [a]

[a] Laser Physics Centre, Australian National University, Canberra, ACT 0200, Australia

Version of record first published: 05 Dec 2006.

To cite this article: Anna Samoć, Marek Samoć, Maneerat Woodruff & Barry Luther-davies (1994): Photophysical Processes Involved in Creation of Dark Spatial Solitons in Composite Photonic Media, Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals, 253:1, 133-142

To link to this article: http://dx.doi.org/10.1080/10587259408055252

Mol. Cryst. Liq. Cryst. 1994, Vol. 253, pp. 133–142
Reprints available directly from the publisher
Photocopying permitted by license only

Printed in the United States of America

PHOTOPHYSICAL PROCESSES INVOLVED IN CREATION OF DARK SPATIAL SOLITONS IN COMPOSITE PHOTONIC MEDIA

ANNA SAMOĆ , MAREK SAMOĆ, MANEERAT WOODRUFF AND BARRY LUTHER-DAVIES
Laser Physics Centre, Australian National University, Canberra, ACT 0200, Australia

Abstract Investigations of formation of dark spatial solitons in thermal and photobleachable media are reported.

Keywords: solitons, photobleachable, waveguides, nonlinear, self-trapping, self-guiding, self-defocussing, self-phase modulation

INTRODUCTION

Intense laser beams propagating in optically nonlinear media are subject to various nonlinear interactions involving frequency mixing and phase modulation. A nonlinear process of great practical importance is self-phase modulation, i.e. a modification of the refractive index of a medium by a light beam. This modification, in turn, influences the propagation of the beam. Under certain conditions such interactions can lead to the formation of soliton-like stable structures in the time or space domains[1-7]. An example of such a structure is a self-trapped beam or a bright spatial soliton[3-5]. On the other hand, a *dark* spatial soliton is a *self-guided dip* in the intensity profile of an optical beam propagating in a nonlinear medium. It is a stable spatial structure with a constant shape and size where diffraction of the beam is compensated by the action of the nonlinearity of the medium. A dark soliton requires the medium to have a negative nonlinearity, i.e., the nonlinear refractive index n_2 (defined by $n=n_o+n_2I$ where n is the refractive index and I is the light intensity) or third order susceptibility $\chi^{(3)} <0$ (self-defocussing behaviour). This is opposite to the condition for a bright soliton which can be formed in nonlinear media with $n_2>0$.

An exciting feature of spatial solitons is their ability to write waveguides. A (1+1)D black soliton can be generated by introducing a π phase jump across the centre of a Gaussian beam and propagating the resulting interference pattern through a defocussing medium. A black soliton writes a refractive index profile in which the bright regions on the sides of the soliton have a lowered refractive index due to the negative n_2, while in the centre of the dark soliton the index is unperturbed. The waveguide formed in this way can be used to guide a separate probe beam coaxially along the soliton. Dark spatial solitons can thus be used for guiding light beams by other light beams and this is of great interest to all-optical processing of information[6,7].

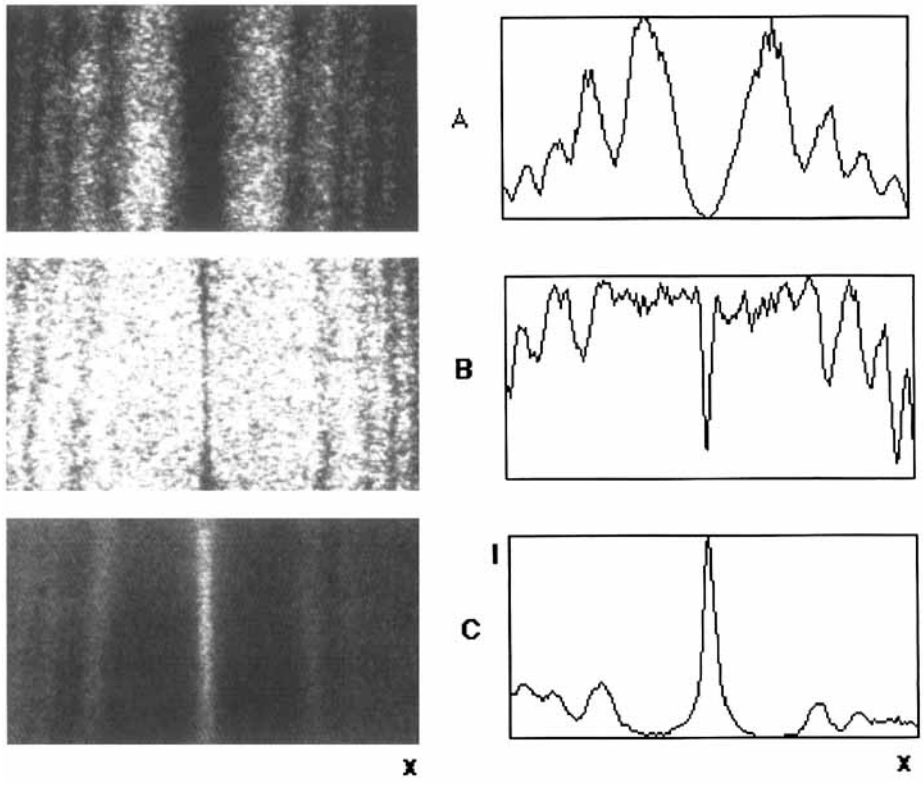


FIGURE 1 Photographs and intensity profiles taken with a digitizing camera for: A) - the interference pattern formed by a phase mask used to create a dark spatial soliton with a 514.5 nm Ar laser beam; B) - a dark soliton in a solution of a dye (methyl orange) in ethanol. $1x10^{-4}$ M solution, 2 cm cell, absorption coefficient = 0.13 cm^{-1}, light intensity = 60 mW/mm^2; C) - the image of a waveguide formed by a soliton as seen by a broad probe 632.8 nm beam

Figure 1 shows an intensity profile caused by a π phase jump in the absence of a nonlinear medium and formation of a dark soliton and the associated waveguide in a thermally defocussing medium.

GENERAL CONDITIONS FOR SOLITON FORMATION

It is very useful to associate properties of a spatial soliton with the waveguide properties of the refractive index profile which it induces. As pointed out by Snyder et al[3-5], *a spatial soliton is a mode of the waveguide which it induces*. In the case of a black soliton, it is the 2nd mode of the refractive index profile exactly at cutoff i.e. when

$$V = \frac{2\pi w}{\lambda}\sqrt{n_0^2 - n_\infty^2} = \sqrt{2} \qquad (1)$$

where V is the characteristic waveguide parameter, w is the waveguide (soliton) halfwidth, n_0 and n_∞ are the maximum and minimum refractive index values and λ is the wavelength.

This means that, in order to generate a soliton of a given dimension, one has to induce a refractive index change in the material of the magnitude necessary to generate a single-mode graded index waveguide of the same dimension. Thus, refractive index changes of the order of 10^{-6} to 10^{-4} will generate spatial solitons of halfwidths ranging from about 100 μm to 10 μm. Such changes can be accomplished by several mechanisms with various time scales of response. Three classes of photophysical processes which may cause refractive index reduction are:

- thermally induced changes in absorbing media. The major component of these changes is due to thermal expansion leading to a defocussing behaviour, however, other processes may also contribute to the total refractive index change.
- photochemical transformations of dye molecules. These can lead to transient or permanent bleaching of absorption, which, through the Kramers-Kroning transformation causes a decrease of the refractive index.
- electronic third-order optical nonlinearity of molecules. As a rule, negative (defocussing) $\chi^{(3)}$ occurs for wavelength between the single-photon and two-photon absorption edges of the material, although there are also materials for which negative zero frequency (nonresonant) $\chi^{(3)}$ has been postulated[8].

We discuss here only the first two processes.

The nonlinear response of the material should be considered in terms of its spatial and temporal behaviour. In general, a change of refractive index of a medium does not

follow immediately the intensity changes but is a nonlocal function of time and spatial variables:

$$\Delta n(t,\mathbf{r}) = \int_{-\infty}^{t}\int_{-\infty}^{\infty} n_2(t-t_1,\mathbf{r}-\mathbf{r}_1)\, I(t_1,\mathbf{r}_1)\, dt_1 d\mathbf{r}_1 \qquad (2)$$

which means that the effective nonlinear index n_2 is a function of both the frequency ω and the spatial frequency of illumination s, i.e $n_2 = n_2(\omega,s)$.

THERMAL PHENOMENA

Formation of dark solitons can readily be observed in absorbing vapours and liquids such as alcohol solutions of dyes with a cw laser[1,6,7]. The underlying mechanism of the refractive index change is without a doubt thermal. Investigation of the thermal response in various media is of interest for several reasons. Changes of refractive index due to a change of temperature are easy to observe with low power cw lasers. Investigations of model dark soliton devices can therefore be easily performed with this kind of nonlinearity. On the other hand, even if materials with other than thermal mechanisms of nonlinearity are used, the thermal response remains as a side effect, since attainment of relatively high nonlinearity without absorption losses is virtually impossible.

Factors that determine the thermal nonlinear response can be conveniently visualised by considering irradiation of an absorbing medium with a spatially nonuniform, time-varying light intensity $I = I(t,\mathbf{r})$ and the heat flow equation:

$$c_p\rho\frac{\partial T}{\partial t} = \alpha I(t,\mathbf{r}) + \kappa\nabla^2 T \qquad (3)$$

where α is the absorption coefficient, c_p is the specific heat, ρ is the density and κ is the heat conductivity coefficient.

The time and spatial dependence of the temperature distribution will depend on the initial and boundary conditions of the problem. Representing $I(t,\mathbf{r})$ in terms of Fourier components of the form $I = I(\omega,s)\, e^{i\omega t} e^{isx}$, one can show that, for one-dimensional heat flow, in terms of the frequency components, the relation is:

$$\Delta T(\omega,s) = \frac{\alpha}{i\omega c_p\rho + s^2\kappa}\, I(\omega,s) \qquad (4)$$

where ω stands for the frequency and s is the spatial frequency. The temperature change may lead to a refractive index change through several mechanisms. A thermochromic change in the absorption spectrum of the medium may lead to accompanying refractive index changes. The temperature change also induces density changes by thermal expansion. In the case of a liquid or gas under steady-state conditions, the change of the refractive index with temperature is therefore:

$$\Delta n = \left(\frac{\partial n}{\partial T}\right)_v \Delta T + \left(\frac{\partial n}{\partial v}\right)\left(\frac{\partial v}{\partial T}\right)\Delta T \cong \left(\frac{\partial n}{\partial T}\right)_v \Delta T - \frac{n^2-1}{2n}\beta\,\Delta T \qquad (5)$$

where v is the specific volume and $\beta = (1/v)\,(\partial v/\partial T)_p$ is the volume expansion coefficient at constant pressure and invariancy of molar refraction is assumed. As an example, one finds from Equation (1) and (5) that a 50 μm diameter soliton formed in the ethanol solution ($\beta = 6.6\times10^{-4}\ ^oC^{-1}$,[9]) in the experiment depicted in Fig.1b corresponded to only about 10^{-2} deg difference of temperature between the dark zone and the surroundingbright regions while the overall temperature increase of the bright region is several degrees. In Eq.(5) only the second term was considered.

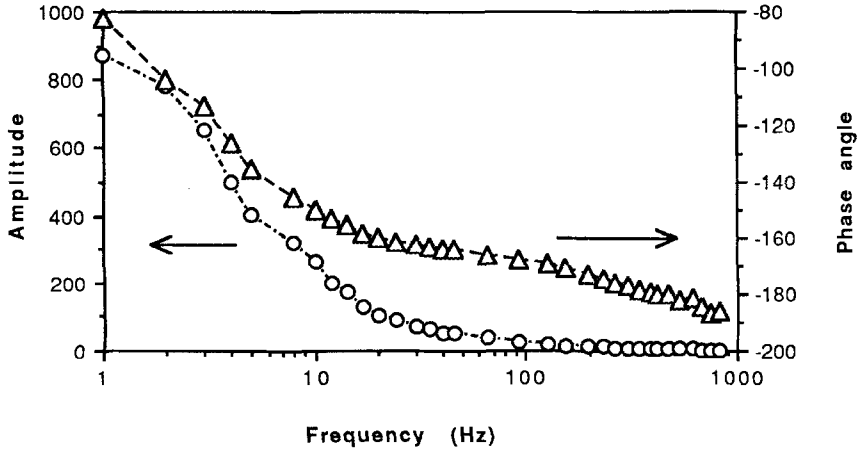
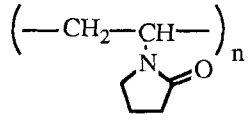


FIGURE 2 Amplitude and phase of the nonlinear thermal response obtained by chopping the soliton forming beam (514.5 nm) and detecting the variation of intensity of a probe beam (632.8 nm) guided by the soliton-formed waveguide. Measurement was performed in a 1 cm cell of copper chlorophyllin solution (2.84×10^{-5} M) in 1-methyl-2-pyrrolidinone.

The speed of response of the thermal nonlinearity is limited by the thermal conductivity of the medium and the geometry of the experiment, but, additionally, there also is a time lag between the temperature change and the change of density of the material. Fig. 2 shows the result of a measurement of a speed of response of a bulk thermal medium (solution of dye) in which a soliton is formed by light chopped at various frequencies and a probe beam is used to detect refractive index changes (waveguide formation) associated with the soliton formation. It can be seen that the response is, indeed, very slow. However, it has been predicted that in optimized thin film structures the response speed can be increased to microsecond range[10].

Our more practical interest was in the thermal nonlinear response in solid composite media such as doped polymers and sol-gel silica glasses, which can be processed into planar waveguides.We therefore investigated nonlinear effects and soliton formation in such materials. The host polymer for most work described here was polyvinylpyrrolidone (PVP):



which has an advantage of accepting high concentrations of various guest molecules without the loss of good optical properties (low scattering). We found that the thermal response in polymers can lead to a confusing behaviour. The reasons for these complications can be explained as follows: A spatial distribution of temperature in a solid leads to formation of a complicated stress-strain pattern. If a medium cannot increase its total volume, then the areas of higher temperature can only decrease their density by straining the neighbouring areas of lower temperature. This generates stress at the boundaries between hot and cold areas. The refractive index will then change due to density changes and, also due to the stress induced birefringence[11] (elastooptic effect). For a viscoelastic solid such as a polymer or a polymer-solvent system, there are also additional phenomena which may contribute to the total refractive index change, especially at longer times or low frequencies. The existence of a constant stress leads to a time-dependent strain (creep) while a constant strain produces a time-dependent stress (stress relaxation). These phenomena are important for the formation of solitons in polymers. Finally, however, another phenomenon appears which further complicates the process. Temperature gradients may also induce thermodiffusion of components in a medium[12]. This Soret effect is especially important if, for example, a low refractive index solvent is used to cast a film of a higher refractive index polymer. The diffusion of the solvent remaining in the polymer changes local composition of the mixture thereby inducing local refractive index changes.

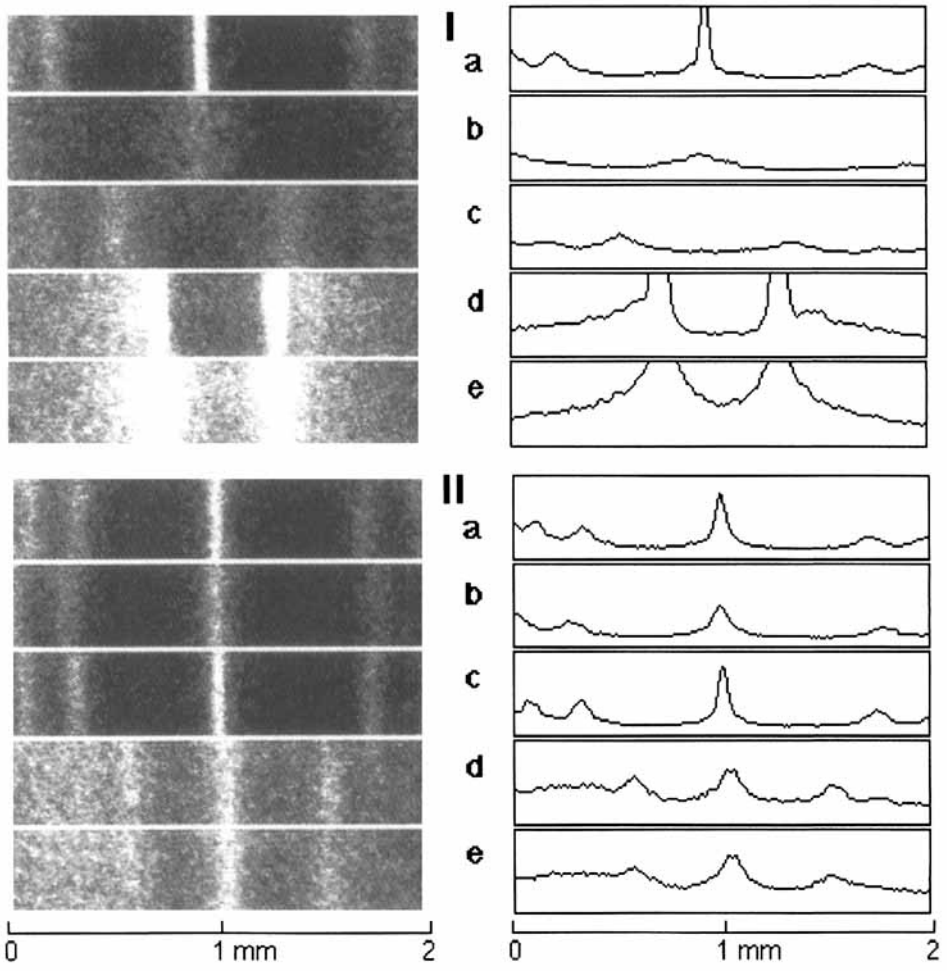


FIGURE 3 Photographs and intensity profiles obtained using 632.8 nm probe beam for experiments in which a dark soliton was formed in a 2 cm long cell containing a dye-polymer-solvent mixture (I - 60%PVP in ethanol, II- 60% PVP in 1-methyl-2-pyrrolidinone), the dye in both cases was methyl orange . a) 10 s after the 514.5 nm soliton-forming beam was turned on, b) 60 s, c) 10 min. The soliton forming beam was then turned off and the photographs taken after d) 20 s and e) 5 min.

We observed features of formation of dark solitons in composite media which indicate that all the above mechanisms may be operative. In particular, we observed formation of long-lived structures in polymers and polymer solutions (Fig.3). For certain compositions of the mixtures the long time refractive index changes in these structures

were opposite to those needed by the dark soliton. Therefore, dark solitons initially formed were then destroyed on prolonged irradiation (I in Fig.3). On the other hand, in other compositions, permanent waveguiding structures could be formed by dark solitons (II in Fig.3). Long-lived refractive index structures of type I were also observed in PVP mixtures with ethylene glycol, water and benzyl alcohol and type II in mixtures with 1-vinyl-2-pyrrolidinone. Our measurements on various polymer-solvent compositions lead us to conclude that both the thermodiffusion caused refractive index changes (caused by flow of one component of the mixture against the other) and those due to creep and stress relaxation (flow of the medium as a whole) are operative and separating these two contributions is very difficult. Both processes exist within the same time range. For example the diffusion coefficient for the PVP-methanol system is 9.6×10^{-7} cm^2/s [13] which leads to the mean diffusion path $\langle r \rangle = \sqrt{Dt}$ of about 80 μm in 1 minute - the time and distance scale of the observed effects.

BLEACHING PHENOMENA

A negative refractive index change can also be obtained if absorption of a medium is bleached: either in a permanent or in a reversible way. Permanent bleaching is of interest because it provides a way for using dark solitons to "write" waveguides and waveguide structures such as junctions. Also, slow bleaching by a cw laser beam can be considered a reasonable model of a fast process of saturation of absorption by a short laser pulse.

We have investigated a large number of dyes of various chemical structures in various media (solution, molecularly doped polymer, sol-gel silica glass) and found that the refractive index changes necessary to create a dark soliton and an associated waveguide can be obtained by bleaching. In particular, many xanthene dyes have been found to be suitable for writing permanent waveguide structures by bleaching. As shown in Fig.4, a polymer film doped with a xanthene dye can be bleached to almost complete transparency in the visible.

The refractive index changes accompanying the bleaching are shown in Fig. 5. There is an approximate linear relation between the content of the unbleached dye and the refractive index, showing that a simple equation based on Lorentz model:

$$\frac{n^2-1}{n^2+2} = \frac{4}{3}\pi \left(N_{polym}\alpha_{polym} + N_s \alpha_s + N_p \alpha_p\right) \qquad (6)$$

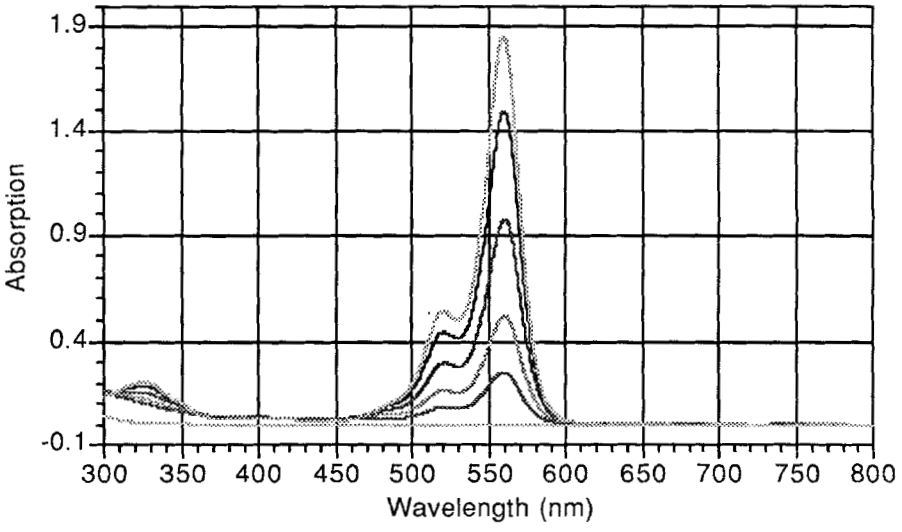


Figure 4 Decay of absorption in a phloxine B doped (4.82 %), 2.6 mm thick polyvinylpyrrolidone (PVP) film bleached by 514.5 nm light (250 mW/cm^2). The absorption curves are for the bleaching time equal to 0,5,17,40 and 77 min (from the topmost curve down). The lowermost curve is the absorption spectrum of pure PVP.

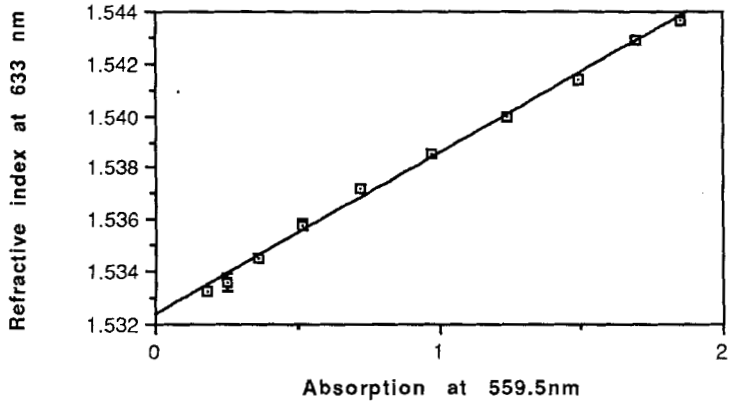


FIGURE 5 Changes of refractive index during bleaching for a film containing 4.82 % Phloxine B in PVP versus absorption of unbleached Phloxine B.

describes reasonably well the bleaching induced refractive index changes. In the above equation α_s and α_p stand for linear polarizabilities of the photochemical reaction substrate (Phloxine B) and product, respectively, and the substrate and product concentration sum is $N_s+N_p=N_{0s}$, i.e. equal to the initial concentration of Phloxine B. N_s can be determined assuming that the film absorption is proportional to it. Our experiments on films containing 4.8% Phloxine B give the difference of polarizabilities at 632.8 nm for Phloxine B and the photobleaching product equal to $\alpha_s-\alpha_p=34$ Å^3. The mechanism of bleaching in this family of dyes and the associated kinetics are complicated[14,15].

We found that it is possible to "write" permanent waveguides using these bleachable materials and propagation of dark soliton-like structures. The processes involved in creation of the waveguides involve both the thermal nonlinearity and bleaching induced refractive index changes, being modified, however, by the presence of spatially nonuniform and time-dependent absorption.

REFERENCES

1. G.A. Swartzlander,Jr, D.R. Andersen, J.J. Regan, H.Yin and A.E. Kaplan, Phys.Rev.Lett. 66,1583 (1991).
2. G.R. Allan, S.R. Skinner, D.R. Andersen and A.L. Smirl, Optics Letters, 16,156 (1991)
3. A.W. Snyder, D.J. Mitchell, L. Poladian and F. Ladouceur, Optics Letters, 16, 21 (1991)
4. A.W. Snyder, L. Poladian and D.J. Mitchell, Optics Letters, 17, 789 (1992)
5. A.W. Snyder and D.J. Mitchell, Optics Letters, 18, 101 (1993)
6. B. Luther-Davies and Y.Xiaoping, Optics Letters, 17,496 (1992)
7. B. Luther-Davies and Y.Xiaoping, Optics Letters, 17,1755 (1992)
8. e.g. C.W. Dirk, L-T. Cheng and M.G. Kuzyk, Int.J.Quantum Chemistry, 43,27 (1992)
10. D. Fotheringham and B. Luther-Davies, to be published
11. L.R.G. Treloar, The Physics of Rubber Elasticity, Clarendon Press, Oxford 1967
12. H. Tyrrel, Diffusion and heat flow in liquids, Butterworths, London 1961
13. J. Brandrup and E.H. Immergut, Polymer Handbook, 3rd ed., J. Wiley and Sons, NY, 1989
14. M. Koizumi and Y. Usui, Mol.Photochem. 4, 57 (1972)
15. D.C. Neckers, J. Photochem.Photobiol. A47, 1 (1989)