Nonlinear optical Kerr coefficients of disordered media

R. E. de Araujo and A. S. L. Gomes
Departamento de Física, Universidade Federal de Pernambuco, Cidade Universitária, Recife 50739-640, Pernambuco, Brazil
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The degenerate and nondegenerate nonlinear Kerr coefficients n_2 of disordered media composed of submicrometer dioxide titania particles embedded in a dye solution were measured using an interferometric technique. The influence of the scattering particles on the value of n_2 are discussed in the light of stimulated emission in these systems. The results obtained can be useful for applications in nonlinear ultrashort pulse propagation in disordered media. [S1050-2947(98)06402-6]

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I. INTRODUCTION

Disordered media (DM) composed of dielectric particles with characteristic size smaller than the wavelength of the incident radiation have been thoroughly exploited for the understanding of photon propagation in disordered systems. Multiple scattering of light studied in those media has improved dramatically the understanding of fundamental as well as applied phenomena. Photon localization is perhaps the most studied fundamental phenomenon in such media [1–3], whereas biomedical applications, such as identification of hidden objects in biological tissues using optical techniques, have also been exploited [4,5]. One of the most recent features reported was the observation of laser action in DM in a dye solution [6–10], which has led the way to a different field: the propagation of light in disordered media with optical gain [11,12].

For an intense laser beam propagating through a DM, nonlinearities can substantially affect the scattering properties of the media [13]. Two mechanisms are identified as responsible for such alterations: topology variations (changes in the distribution of the scattering particles) or alteration in the refractive index of the components of the disordered media. An important example of the exploitation of the first mechanism can be found in Refs. [14, 15], where the radiation pressure resulting from the optical field gradient induces a force that leads to motion of the scattering particles in the medium.

The second mechanism, variation in the refractive index, can be described by $n=n_0+n_2I$, where n_0 is the linear refractive index, n_2 is the nonlinear Kerr coefficient, and I is the light intensity. The physical origin of n_2 can be, for instance, electronical or thermal. The variation of the refractive index due to the nonlinear Kerr coefficient can lead to suppression of scattering [16].

In this work, the nonlinear Kerr coefficient n_2 of a DM consisting of TiO_2 submicrometer spheres embedded in an optically active medium is experimentally determined. Both degenerate and nondegenerate n_2 were measured in the nanosecond regime. The influence of the presence of the scatterers is analyzed in the light of stimulated emission.

II. MEASUREMENTS OF DEGENERATE NONLINEAR KERR COEFFICIENTS

For homogeneous media, there are several techniques available that can be used to measure the sign and magnitude of the nonlinearity. Examples are four-wave mixing, interferometric methods, Z scans, self-focusing, and self-phase modulation. For DM, because of the spatial scattering suffered by the beam, care must be taken and some of the techniques mentioned above are not easily implemented. In Refs. [14, 15], self-focusing and four-wave mixing have been used to obtain the values for the sign and magnitude of the $\chi^{(3)}$. In both experiments, the particles (1-μm latex spheres) were dissolved in water.

In our experiments, an interferometric technique based on a Twyman-Green interferometer has been employed following the work of Ref. [17]. This technique, used in conjunction with an incoherent light source (laser pulses with coherence time much shorter than the pulsewidth), allows one to measure nonlinear effects on the ballistic component, which suffers less scattering, independent of the more scattered diffuse component [18] propagating through the DM. The experimental scheme is shown in Fig. 1 (the dashed line represents a change in the experimental setup used in Secs. III and IV). A broadband, Nd:YAG pumped dye laser (where YAG denotes yttrium aluminum garnet) (pulsewidth 13 ns, bandwidth 5 nm, center wavelength 618.5 nm, coherence time 250 fs, and repetition rate 5 Hz) was employed as the pump source. The disordered medium consisted of TiO_2 nanospheres with 250 nm diameter (R-900 Ti-pure from DuPont) embedded in a $2\times10^{-3}M$ solution of Rh640 in methanol. The nanospheres concentration used here was 5.7

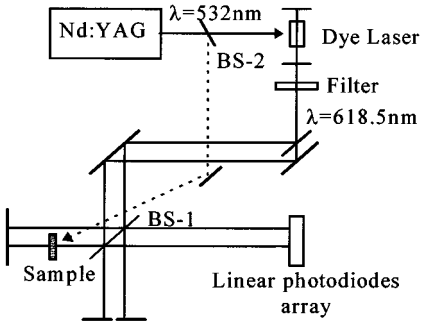

FIG. 1. Experimental scheme used to measure the nonlinear refraction index of the disordered media. For the nondegenerate n_2 measurement a beam splitter BS-2, is introduced to divert the pump beam.

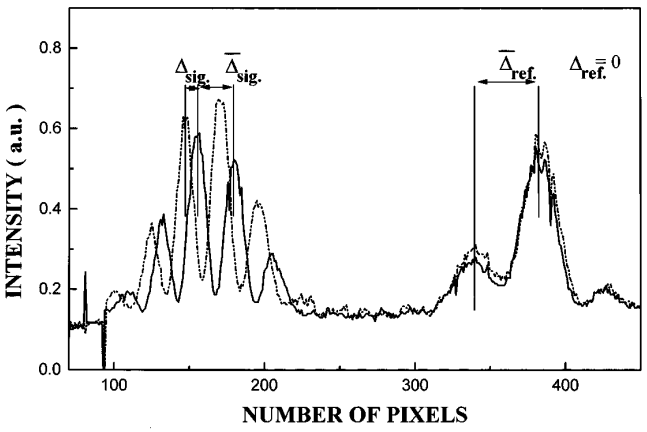

FIG. 2. Experimental result of the displaced fringes for two different beam intensities.

$\times 10^{10}$ cm^{-3}. This is the same disordered system in which laser action has been observed previously [6,12]. The double collimated beams entering the Twyman-Green interferometer are the signal (which passed through the sample) and reference beams. The signal beam was focused upon a 200-μm-thick cell containing the disordered medium to a beam diameter of 120 μm, yielding a maximum intensity of 24 MW/cm^2. The outcoming beams from the interferometers formed double interference fringes pattern, spatially separated, which were detected by a 1024 pixel linear photodiodes array and displayed on a digital oscilloscope linked to a personal computer for analysis. As the intensity of the signal beam through the sample was varied, the nonlinear index change $\Delta n = n_2 I$ changed the phase wave front propagating through the DM. This phase variation ϕ was determined by the fringe displacement induced in the signal beam. To ensure that acoustic vibrations or external thermal variations do not contribute to the fringe displacement, the reference beam was used. Thus the phase change due to the nonlinear effects was determined by

$$\phi = 2\pi \left(\frac{\Delta_{\mathrm{sig}}}{\bar{\Delta}_{\mathrm{sig}}} \pm \frac{\Delta_{\mathrm{ref}}}{\bar{\Delta}_{\mathrm{ref}}} \right) \tag{1}$$

(in radians), where Δ_{sig} (Δ_{ref}) is the observed fringe shift and $\bar{\Delta}_{\mathrm{sig}}$ ($\bar{\Delta}_{\mathrm{ref}}$) is the separation between the maxima of the zeroth and first fringe orders in the signal (reference) beam. The phase variation ϕ is known to be related directly to the optical path change through the DM ($\phi = 2\pi \Delta x/\lambda$). Shown in Fig. 2 is a typical experimental result obtained for different beam intensities. The displaced fringes represent the signal beam, whereas the superimposed fringes are due to the reference beams. To obtain the value of the nonlinear refractive index, several measurements were performed and the data were averaged. A simple analysis of the beam propagation through the DM can be performed, taking into account that the intensity of the beam decreases exponentially through the DM, according to $I = I_0 \exp(\alpha x')$, where $\alpha = \sigma\rho$ is the scattering depth σ is the cross section calculated through Mie theory, and ρ is the scatterers' density. The variation in the optical path in the DM due to the intensity change ΔI leads to an effective nonlinear refractive index given by

$$n_{2\,\mathrm{eff}} = \frac{\pm \phi\lambda}{2\pi(\Delta I)T(1+T^2 e^{-\alpha L})} \left[\frac{\alpha}{1-e^{-\alpha L}} \right], \tag{2}$$

where L is the sample length, λ is the wavelength of the beam, and T is the fraction of the intensity transmitted by the cell's wall. The sign of the nonlinearity is determined by comparing the fringe displacement due to the intensity variation with respect to the change in position due to one of the interferometer's arm displacement. This system was tested using a material with a known nonlinearity (semiconductor-doped glass, Corning filter 3-69) and a value of n_2 similar to that of Ref. [17] was obtained. For the system used (Rh640+TiO_2 spheres), the scattering depth for $\lambda = 618.5$ nm was calculated to be 68.5 cm^{-1}. From the measurements and with the help of Eq. (2), the value of $n_{2\,\mathrm{eff}} = -(8\pm 2)\times 10^{-11}$ cm^2/W was determined. We have also measured the value of $n_{2\,\mathrm{eff}}$ for the dye alone and a value of $-(7\pm 2)\times 10^{-11}$ cm^2/W. The negative sign of the measured nonlinearity is expected from the Liouville equation assuming a simple two-level system for the dye. Therefore, we can conclude that for the scatterers density employed ($\rho = 10^{11}$ cm^{-3}) the contribution for the nonlinearity is determined solely by the dye. This is understood due to the fact that the nonlinear refractive index of TiO_2 is of the order of 10^{-13} cm^2/W [19]. We will point out later the role played by the scatterers.

III. MEASUREMENTS OF THE NONDEGENERATE NONLINEAR KERR COEFFICIENT

In order to measure the nondegenerate nonlinear Kerr coefficient, the only change required in the experimental setup was the introduction of a second beam of different wavelength (dashed line in Fig. 1). The second harmonic of the Nd:YAG laser at 532 nm was used for this purpose, and the signal beam was kept at the same wavelength (618.5 nm). The beams were overlapped at the front wall of the cell providing an interaction region determined by the penetration depth of the green beam. Again, a theoretical analysis was performed, taking into account the penetration depth of the beam at 532 nm and, more importantly, the gain effect that it can introduce. The resulting expression for $n'_{2\,\mathrm{eff}}$ is

$$n'_{2\,\mathrm{eff}} = \frac{1}{(I_f - I_0)(1+T^2 e^{gL' - 2\alpha L''})} \left(\frac{g}{e^{-gL'} - 1} \right) \times \left[\pm \frac{\phi\lambda}{2\pi} - n_{2\,\mathrm{eff}}(\Delta I) T e^{gL'} (1 + T^2 e^{-\alpha L''}) \times \left(\frac{1 - e^{\alpha L''}}{\alpha} \right) \right], \tag{3}$$

where $n_{2\,\mathrm{eff}}$ is the effective nonlinear refractive index measured in Sec. II, L' is the penetration depth of the green

beam, $L''=L-L'$, and $\exp(gL')$ is the amplification factor of the signal beam. The value of g was experimentally determined by measuring independently the transmitted signal intensity at 618.5 nm in the absence and in the presence of the strong green beam.

The values obtained were $n'_{2\,\mathrm{eff}}=-11\times10^{-10}$ cm^2/W for the DM ($2\times10^{-3}M$ of Rh640 in methanol with a concentration of 5.7×10^{10} cm^{-3} of TiO_2) and -8×10^{-10} cm^2/W for the dye alone (same solution). These results took into account the values of $g=35$ cm^{-1} for the DM and $g=50$ cm^{-1} for the dye alone. As in the degenerate case, the value of the nonlinear Kerr coefficient is determined by the dye alone. However, the presence of the green beam increases the modulus of the nonlinear nondegenerate Kerr coefficient compared to the degenerate case by one order of magnitude. This is due to the cross phase modulation effect induced by the green beam on the red beam, which can arise physically from the electronic or thermal effect or a combination of both. As the sign of the measured nonlinearity was negative and the pulse duration is long (10 ns), thermal effects may be playing a major role even though a low repetition rate was employed. Experiments using shorter (a few picoseconds) pulses should be performed in order to clarify the origin of the cross phase modulation effects.

IV. THE INFLUENCE OF THE SCATTERERS AND CONCLUSIONS

One question then arises: What is the influence of the scatterers on the nonlinearity of the DM? To answer this question, another set of experimental data was collected, where the intensity of the excitation (green) beam was varied and the signal beam intensity was kept constant at 23 MW/cm^2. The phase variation versus intensity was then plotted for both dye alone and the DM. The results are shown in Figs. 3(a) and 3(b). In Fig. 3(a), for dye alone, it can be seen that the maximum phase shift is obtained at a maximum excitation beam intensity and it varies softly. On the other hand, in the DM, there is an abrupt phase shift reaching the maximum at a very small intensity. According to Figs. 3(a) and 3(b), a phase shift of 6 rad can be achieved with 8 times less power in the DM than in the dye alone.

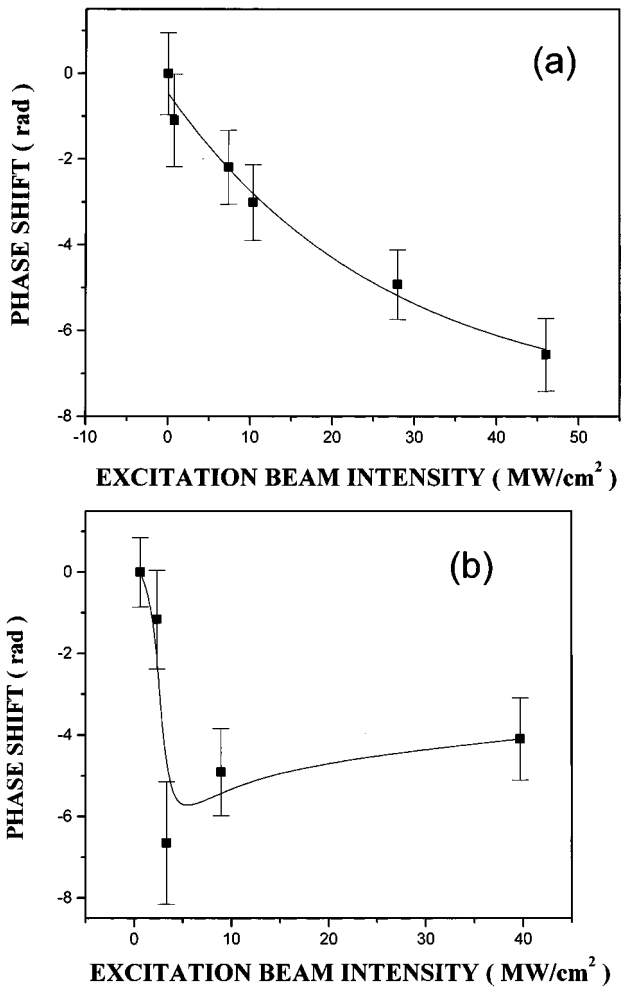

FIG. 3. Measured results of the phase-shift behavior for different excitation beam intensities, when the signal beam crosses (a) a dye solution and (b) a disordered media. The solid lines are best fits of the experimental points.

The main reason for the abrupt phase shift change in the DM is related to stimulated emission in the DM solution. In an independent measurement we have observed that, for the concentration used here, laser emission occurred in the DM, as in Ref. [7], with the same threshold value of Fig. 3(b). Therefore, above the threshold a strong alteration in the dye population exists, inducing a drastic change in the material's susceptibility, and this was transferred, through cross phase modulation, to the signal beam.

Although the results reported here were performed for a single-particle size, optical density, and volume fraction, two values of titania concentration were employed (different by a factor of 5). For the two concentrations used, the behavior of the energy value for which the phase shift changed abruptly followed that of the lasing threshold in the laser paint [22]. Even though more experimental data are required using different particle parameters, one could observe the effects of the feedback due to the scatterers in lowering the ''threshold'' power for the maximum phase shift to occur. Qualitatively, one can argue that all the behavior already shown for the laser paint by several authors, regarding the particle parameters mentioned above, should lead to similar behavior in the threshold for the nonlinear response of the DM in the presence of gain. Even issues such as critical sample size may follow the same behavior as for the laser paint.

Several conclusions can be drawn from this work. First, the value of the nonlinear optical susceptibility of disordered medium can be directly and conveniently measured using an interferometric technique. Second, the role of the particles in the nonlinearity can be identified, as well as the host medium, and artificial nonlinear media can be engineered for further studies. Third, stimulated emission in DM can be of importance for lowering the threshold for the observation of nonlinear effects.

As a further and important conclusion, the system studied here can be exploited for the studies of nonlinear ultrashort pulse propagation. It is known that an ultrashort pulse propagating through a nonlinear medium can be described by the nonlinear Schrödinger equation. A similar equation for DM has already been analyzed [20,21]. From the data obtained

here, it is clear that a random gain medium provides a test medium for nonlinear pulse propagation, analogous to an optical fiber, where the nonlinear contribution and dispersionlike effects (in this case due to multiple scattering) can all occur in a short path cell, whose DM can be conveniently manipulated regarding pump beam characteristics.

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