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Nonlinear optical response of colloidal suspensions

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Abstract: We experimentally probed the nonlinear optical response of aqueous nano-colloidal suspensions to provide a test of the theoretical approaches that have been proposed for the nonlinearity, namely an exponential model, an artificial Kerr medium, and a non-ideal gas model. The best agreement with experiment is found using the non-ideal gas model for the colloidal suspension which in turn can be used to infer values for the second virial coefficient of the medium and the nonlinear coefficients.

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References and links
1. P. W. Smith, P. J. Maloney, and A. Ashkin, “Use of a liquid suspension of dielectric spheres as an artificial Kerr medium,” Opt. Lett. 7(8), 347–349 (1982).
2. P. W. Smith, A. Ashkin, and W. J. Tomlinson, “Four-wave mixing in an artificial Kerr medium,” Opt. Lett. 6(6), 284–286 (1981).
3. 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(6), 276–278 (1982).
4. V. E. Yashin, S. A. Chizhov, R. L. Sabirov, T. V. Starchikova, N. V. Vysotina, N. N. Rozanov, V. E. Semenov, V. A. Smirnov, and S. V. Fedorov, “Formation of soliton-like light beams in an aqueous suspension of polystyrene particles,” Opt. Spectrosc. 98(3), 511–514 (2005).
5. P. J. Reece, E. M. Wright, and K. Dholakia, “Experimental observation of modulation instability and optical spatial soliton arrays in soft condensed matter,” Phys. Rev. Lett. 98(20), 203902 (2007).
6. C. Conti, G. Ruocco, and S. Trillo, “Optical spatial solitons in soft matter,” Phys. Rev. Lett. 95(18), 183902 (2005).
7. R. El-Ganainy, D. N. Christodoulides, C. Rotschild, and M. Segev, “Soliton dynamics and self-induced transparency in nonlinear nanosuspensions,” Opt. Express 15(16), 10207–10218 (2007).
8. R. El-Ganainy, D. N. Christodoulides, Z. H. Musslimani, C. Rotschild, and M. Segev, “Optical beam instabilities in nonlinear nanosuspensions,” Opt. Lett. 32(21), 3185–3187 (2007).
9. R. Gordon, J. T. Blakely, and D. Sinton, “Particle-optical self-trapping,” Phys. Rev. A 75(5), 055801 (2007).
10. M. Matuszewski, W. Krolikowski, and Y. S. Kivshar, “Spatial solitons and light-induced instabilities in colloidal media,” Opt. Express 16(2), 1371–1376 (2008).
11. M. Matuszewski, W. Krolikowski, and Y. S. Kivshar, “Soliton interactions and transformations in colloidal media,” Phys. Rev. A 79(2), 023814 (2009).
12. “Critical collapse beyond a self-focusing power occurs for the case sd = 4, where s is the order of the nonlinearity, s = 2 for a Kerr medium, and d the number of transverse dimensions. Super-critical collapse occurs for sd>4 and tends to be much more abrupt spatially than critical collapse, see, for example, N. E. Kosmatov, V. F. Shvets, and V. E. Zakharov, “Computer simulation of wave collapses in the nonlinear Schrodinger equation,” Physica D 52, 16–35 (1991).
13. M. Sheik-bahae, A. A. Said, and E. W. Van Stryland, “High-sensitivity, single-beam n2 measurements,” Opt. Lett. 14(17), 955–957 (1989).
14. P. M. Hansen, V. K. Bhatia, N. Harrt, and L. Oddershede, “Expanding the optical trapping range of gold nanoparticles,” Nano Lett. 5(10), 1937–1942 (2005).
15. R. El-Ganainy, D. N. Christodoulides, E. M. Wright, W. M. Lee, and K. Dholakia, “Nonlinear dynamic rotation of non-ideal gases of interacting colloidal nano-particles,” to be submitted (2009).
16. J. Junio, E. Blanton, and H. D. Ou-Yang, “The Kerr effect produced by optical trapping of nanoparticles in aqueous suspension,” Optical Trapping and Optical Micromanipulation IV, Edited by Dholakia, Kishan; Spalding, Gabriel C. Proceedings of the SPIE, Volume 6644, pp. 664408 (2007).
1. Introduction

It has been known since the pioneering work of Ashkin and associates [1–3] in the 1980s that liquid suspensions of spherical dielectric nanoparticles can show very large optical nonlinearities. This has been amply demonstrated in the intervening years in a variety of experiments including four-wave mixing [2], self-focusing optical beams [3], optical spatial soliton propagation [4], and modulation instability [5]. Physically, the nonlinearity of dielectric nanoparticle suspensions has its origin in the fact that in the presence of a continuous wave optical field with intensity profile $I(r)$ the dielectric nanospheres experience an optical dipole force $F_{\text{grad}} = \text{grad}(U)$, with $U = \alpha I(r)/4$ the dipole interaction energy, $\alpha$ being the particle polarisability in the liquid. For the case of particles of higher refractive-index than the surrounding liquid, $\alpha > 0$, the particles thus experience an electrostrictive volume force that attracts them into the spatial regions of high intensity thereby increasing the local density and subsequently the local refractive-index. This leads to the model of colloidal suspensions as artificial Kerr media in which the induced change in refractive-index is proportional to the applied light intensity $\Delta n = n_{2K} I$, with $n_{2K} > 0$ the nonlinear Kerr coefficient.

Despite some successes in applying the artificial Kerr medium model to experiments on nonlinear optics in colloidal suspensions there remains a fundamental conflict between theory and experiment regarding the nature of the nonlinearity of this soft condensed matter system [6–11]. In particular, carried to its logical conclusion, the electrostrictive model for the nonlinear optical response leads to a Boltzmann distribution for the particle density [1] and hence to an exponential optical nonlinearity $\Delta n(I) = \Delta n(I = 0)e^{\alpha I/4KW}$ for a colloidal system at temperature $T$ [7–9]. The Kerr nonlinear optical response is therefore simply the leading nonlinearity, linear in the intensity, in an infinite expansion of higher-order nonlinearities. The fundamental issue is whether the exponential model is an accurate representation of the nonlinearity of colloidal suspensions, and whether the higher-order nonlinearities can be accessed experimentally. The answer is not a priori obvious since the exponential model is a single particle model that neglects particle-particle interactions which in turn can limit the compressibility of the liquid suspension, potentially limiting the nonlinear optical response of the soft condensed matter system to the leading Kerr term.

In response to this fundamental issue a variety of theoretical models have appeared in the literature with no resolution to date. Even though the Boltzmann distribution was recognized in the seminal work of Smith et al. [1], all subsequent work carried out in this area (four-wave mixing, self-focusing etc) was analyzed within the context of a Kerr model [2–4]. This is reasonable if the incident intensities are assumed to be small compared to a critical thermal energy, e.g. if $\alpha I/4KW << 1$, for which the exponential model may be safely approximated by a Kerr response. More recently Conti et al. [6] have introduced a nonlocal Kerr nonlinear model for generic soft condensed matter systems that yields stable optical spatial solitons (OSS), but they do not address how this is related, if at all, to the underlying experimental parameters of this problem. In addition, El-Ganainy et al. [7,8] and independently Gordon et al. [9] have derived an exponential model starting from first principles and considered OSS dynamics in colloidal suspensions, but with the caveat that this will only apply at low colloidal densities. However, including the full exponential model renders the OSS highly unstable due to the super-critical self-focusing collapse that arises for higher-order self-focusing nonlinearities [12], in apparent contradiction with recent experimental observations of relatively stable OSS that seem to agree reasonably with the Kerr model [4,5].
Matuszewski et al. [10,11] have shown that by treating the liquid suspension as a hard sphere gas they can include the compressibility of the system and this has the effect of saturating the exponential nonlinearity at high intensities. Yet according to this latter hard-sphere gas model [10,11], such saturation effects only come into play at very high filling factors, much higher than those encountered in experimental studies [2–5]. Thus the actual mechanism behind the nonlinear optical response of colloidal suspensions still remains largely unresolved. The goal of this paper is to introduce and analyze a diagnostic experiment intended to submit the different theories to an experimental test.

The remainder of this paper is organized as follows: In Section 2 we describe our diagnostic experiment starting with an overview to provide motivation. Section 3 describes our physical model and the numerical results and comparison with experiment are given in Section 4. Summary and conclusions are given in Section 5.

2. Experiment and results

In this section we first give a brief overview of our experimental scheme for probing the nonlinear optical response of colloidal suspensions to motivate our study, and then we describe the experiment in more detail and provide results.

2.1 Experimental overview

Our experimental setup for probing the nonlinear optical response of colloidal suspensions is shown in Fig. 1. The details of the experimental scheme will be discussed below, but from the theoretical standpoint the key is that it has, at its core, two oppositely directed and aligned identical single-mode optical fibers that are inserted into the colloidal suspension and separated by a distance D, shown in the inset of Fig. 1. In this scheme the input fiber to the left serves to launch a beam having the well defined beam profile of the single-mode fiber and variable input power directly into the colloidal suspension, and the collecting fiber to the right is used to measure the power coupled into the same single-mode beam profile after propagating the distance D. In our experiments the fiber mode profile was well characterized by a Gaussian field of spot size $w_0$, and the distance D was typically a few times the Rayleigh range of the initial Gaussian beam. We remark that our experimental scheme is reminiscent of the classic Z-scan method for measuring Kerr nonlinearities with the collecting fiber playing the role of the aperture in the Z-scan method [13]. In our case, however, it is the input power that is scanned. The key advantage of this technique is to gain direct access to the nonlinear suspension within the aqueous medium. In addition, this approach eliminates any spherical aberration due to beam propagation through the glass and water interfaces present for cuvette-type geometries, and by using thinner chambers in contrast to more standard rectangular cuvettes, convective roles may be suppressed.

To illustrate our experimental scheme we consider the simplified model in which the colloidal suspension may be treated as a self-focusing Kerr medium with a critical power $P_{crK}$ for self-focusing or self-trapping. Then for input powers much less than the critical power, the power at the collecting fiber $P_{coll}$ will be a fixed fraction less than unity of the input power, the fraction being determined by the linear beam spreading and wavefront curvature that accumulate over the distance D. However, as the input power is increased towards the critical power from below the beam spreading will be reduced, and the fraction of power at the collecting fiber will increase with input power. For the special case that the input power equals the critical power for self-trapping the input beam should propagate with unchanging beam profile between the fibers leading to, in principle, perfect power coupling. Increasing in the input power beyond the critical power leads to further contraction of the propagating beam spot size between the fibers so that the fraction of power coupled into the collecting fiber should therefore decrease with increasing input power. Thus for a Kerr medium, the critical power for self-focusing can be estimated experimentally by looking for a peak or rollover in the plot of the measured power $P_{coll}$ at the collecting fiber versus input power $P_{in}$. 

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Fig. 1. shows the experimental setup. Two oppositely directed and aligned identical single-mode optical fibers (SMF) that are inserted into the colloidal suspension and separated by a distance D, shown in the inset. The input fiber to the left launches a well defined Gaussian beam (wavelength, \( \lambda = 1090\,\text{nm} \), spot size \( w_0 = 3.4\,\mu\text{m} \) at variable input power (monitored through the second output from the 50/50 fiber splitter and power meter (PM)) directly into the colloidal suspension, and the collecting fiber to the right is used to measure the power coupled into the same single-mode beam profile after propagating the distance D using a photodetector (PD). An orthogonal differential interference contrast (DIC) imaging (with LP – linear polarizer, NP – Nomarski prism) system is used to detect subtle refractive index variations (from the accumulated nanoparticles) within the sample. An imaging microscope objective (imaging MO) and a tube lens (TL) relay the DIC image onto a high speed digital camera (DC).

2.2 Experimental details and results

We now turn to a more detailed description of the experimental setup in Fig. 1. A linearly polarized laser (10 W, 1090 nm, SPI laser) is coupled into a 50/50 fiber splitter (FC1064-50-FC- 2x2 SM Coupler) using a three positioning fiber coupling stage (MDE122, Elliot scientific) with a coupling MO (10X, 0.25NA, Comar). The beam profile of the SMF is well characterized as a Gaussian with spot size \( w_0 = 3.4\,\mu\text{m} \) at the wavelength \( \lambda = 1.09\,\mu\text{m} \). One end of the fiber splitter (50/50) is directed onto a PM (Melles Griot) to monitor the power fluctuations in the laser and fiber coupling stage (< 2%). The other end of the splitter is coupled, via FC connector, into a single ended cleaved SMF (1060XP, Thorlabs, mode field diameter to 6.8 \( \mu\text{m} \)). The cleaved end of the SMF is then inserted into a capillary of inner diameter of 200 \( \mu\text{m} \) (Invitrocom) as shown in the inset. The sample is mounted onto an X-Y translations stage (H117, Prior Scientific) within an inverted microscope platform (TE2000E, Nikon). An imaging MO (20X, NA DIC 0.50 NA, Nikon) is used to image the sample onto a DC (A622f Basler). A condenser assembly to support DIC illumination is used. The same polarization optics is placed in the imaging path so as to pick up the small phase difference.
within the sample. The DIC illumination technique has previously been used to visualize single nanoparticles [14].

The cleaved end of a second identical SMF (1060XP, Thorlabs, mode field diameter) is inserted into the other end of the capillary and brought to a known distance away from the input face of fiber. Once the two fibers are aligned, the whole chamber is subsequently sealed with epoxy to reduce fluctuations. The output of the second fiber illuminates PD (PDA10CS-EC, Thorlabs), which in turn measures the coupled power. A set of calibrated neutral density filter (ND) is used so as to prevent over exposure onto the PD. A custom Labview program is used to remotely control the input power and capture the coupled power at the PD using a data acquisition device (National instruments NI USB-6009). The nanoparticles used here are polystyrene plain spheres of diameter \(2a = 0.099 \mu m\) (10% coefficient of variation), \(a\) being the radius (PS02N/6391, Bangs Lab). The spheres are treated with a sulfate groups: 0.1% SDS (sodium dodecyl sulfate) and 0.05% sodium azide anti-microbial agent. The initial concentration is \(1.921 \times 10^{14}\) particles/cm\(^3\). For each dilution step, heavy water (\(D_2O\)) is used as the diluting agent so as to reduce thermal convective flow.

For the coupling power measurements, we prepared two samples with colloidal densities of \(\rho_0 = 1.921 \times 10^{13}\), \(1.921 \times 10^{12}\) particles/cm\(^3\). The collecting fiber is placed at a distance \(D = 110 \pm 10 \mu m\) away from the input fiber. For each run, a total of 200 data points are taken and averaged over a 1 second acquisition time. At lower input powers (<400 mW), the collected power varies linearly with the input power (the difference in the power plots is due to minor misalignment in \(z\)).

Figure 2 shows the experimental results for the power measured at the collecting fiber (collected power, \(P_{coll}\)) versus input power (\(P_{in}\)) for the two concentrations above. For the lower density (dash-dot line) the input power remains well below the critical power (5 W) and the collected power scales linearly with the input power. For the higher density (solid line) we see that that the collected power scales linearly for lower input powers, rises above the low concentration linear plot for higher powers, and finally there is a peak in the plot of \(P_{coll}\) versus \(P_{in}\) akin to that discussed above for the case of a Kerr medium, the rollover occurring at \(P_{coll} \approx 0.5\) W. Due to the thin capillary walls, we are also able to observe the alignment of the nanoparticles under DIC illumination for the concentration of \(1.921 \times 10^{13}\) particles/cm\(^3\). In the inset of Fig. 2, we show two DIC images for input powers of 0 W and 0.514 W. When the input is at 0.514 W, we can see a fine line rack denoting the refractive index change (with the DIC imaging) within the sample. This corresponds to the formation of a fine channel of increased nanoparticle density.

Before moving onto the analysis of our experimental results, for reference we measured the nonlinear Kerr coefficient \(n_2\) using a standard Z-scan measurement [13] for the sample concentration of \(\rho_0 = 1.9 \times 10^{13}\) cm\(^{-3}\). In order to satisfy the Z-scan requirements (sample thickness less than beam’s Rayleigh length (in air)), we increased the input beam spot size to 15 \(\mu m\) (measured in air). The beam was directed into a sealed sample cell of 100 \(\mu m\) thickness that is scanned in 10 \(\mu m\) steps using a motorized stage (T-LS Series, Zaber Technologies). The transmitted power is collected and measured after passing through a 3 mm aperture. The measured nonlinear was approximately \(n_2 = 2 \times 10^{-9}\) cm\(^2\)/W. We comment that the nonlinear coefficient is comparable with previous measured values, and the scattering loss (measured at lower power) in the sample at wavelength of 1.09 \(\mu m\) is 3.91 cm\(^{-1}\) is much lower than that reported in the visible [1].

The remainder of this paper shall be devoted to the analysis of these results and what can be extracted from them about the nature of the nonlinear optical response of colloidal suspensions.
3. Physical model and numerical results

3.1 Field propagation equation

For our theoretical and numerical study we adapt the recently developed theory of El-Ganainy et al. [7] for field propagation in a colloidal suspension. In particular, we consider propagation of a linearly polarized field of frequency $\omega$ and free-space wavevector $k_0 = \omega/c = 2\pi/\lambda_0$ along the z-axis in a colloidal suspension at temperature T composed of a host liquid of refractive-index $n_b$, colloids of radius $a$, volume $V_p$, and refractive-index $n_p$, and relative-index $m = (n_p/n_b)$. We denote the electric field envelope as $\varphi(x,y,z)$ in such a way that $|\varphi|^2$ is the field intensity. Then the paraxial wave equation governing field propagation between the fibers over the distance $D$ may be written as

$$\frac{\partial^2 \varphi}{\partial z^2} = \frac{i}{2k_0 n_b} \left( \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \right) \varphi + \alpha_0 \left( i\beta - \frac{1}{2} \right) r \varphi,$$

where $\alpha_0 = \sigma_0$ is the linear (intensity) absorption coefficient accounting for low power Rayleigh scattering losses, $\rho_0$ being the equilibrium colloidal density in the absence of any illumination and

$$\sigma = \frac{128\pi^2 a^2 n_b^4}{3} \left( \frac{a}{\lambda_0} \right) \left( \frac{\beta}{m^2 + 2} \right),$$

is the Rayleigh scattering cross section, and $\beta = k_0(n_p - n_b) V_p / \sigma$ is a dimensionless constant that reflects the strength of the self-focusing nonlinearity to the nonlinear Rayleigh scattering losses.
The factor \( r = \frac{f}{f_0} \) is the ratio of the volume filling factor of particles \( f \) to the field-free background value \( f_0 = V_p \rho_0 \), so that \( r = 1 \) for zero optical intensity. Different models for the nonlinear optical response colloidal suspensions correspond to different choices for the factor \( r \) and we shall survey these below. From Eq. (1) the nonlinear change in the refractive-index \( \Delta n \) is related to the ratio \( r \) via the relation \( k_n \Delta n = \alpha_o \beta r \), and the intensity loss coefficient due to Rayleigh scattering is \( \alpha_r = \alpha_o r \). For comparison with the experiment the following parameters are used: \( \lambda = 1.09 \mu m, \rho_{m} = 1.57, n_{b} = 1.33, \rho_{b} = 1.9 \times 10^3 \text{cm}^{-3}, a = 50 \text{nm}, f_0 = 0.01 \), and \( D = 110 \mu m \), corresponding to polystyrene spheres in water. In this wavelength region, the Rayleigh scattering losses due to the colloidal particles is expected to be very small, and we have verified this in numerical simulations with and without the losses.

We shall employ Eq. (1) for propagation between the input and collecting fibers along with the initial Gaussian field representing the field launched by the input fiber at \( z = 0 \)

\[
\varphi(x, y, 0) = \frac{2P_{\text{in}}}{\pi w_0^2} e^{-\left(x^2+y^2\right)/w_0^2},
\]

with input spot size \( w_0 \) and input power \( P_{\text{in}} \). Once the propagated field \( \varphi(x, y, D) \) is calculated we can calculate the power at the collecting fiber via the integral

\[
P_{\text{coll}} = \left( \frac{2}{\pi w_0^2} \right)^{3/2} \int dx dy e^{-\left(x^2+y^2\right)/w_0^2} |\varphi(x, y, D)|^2,
\]

which is the modulus squared of the propagated field projected onto the collecting fiber mode. For comparison with the experiment we shall use the spot size \( w_0 = 3.4 \mu m \).

### 3.2 Medium equations

From Eq. (1) the nonlinear change in the refractive-index \( \Delta n \) of the colloidal medium is related to the ratio \( r \) via the relation \( k_n \Delta n = \alpha_o \beta r \), leading to the expansion

\[
\Delta n(I) = \left( \frac{\alpha_o \beta}{k_0} \right) r(I) = (n_p - n_b) V_p \rho_o \left( 1 + r' I + \frac{I^2}{2} + \ldots \right)
\]

\[
= \Delta n(0) + n_2 I + n_4 I^2 + \ldots,
\]

where \( r' = (dr/dI)|_{r=0} \). In general the ratio \( r \) must be determined from the equation of state of the soft condensed matter system exposed to the laser field. Here following a recent paper by El-Ganainy et al. [15] we treat the colloidal suspension as a non-ideal “Van der Waals” gas to allow for compressibility of the colloidal suspension, and by keeping the second and third virial coefficients \( B_2 \) and \( B_3 \), the relation between the optical intensity \( I \) and the ratio \( r \) can be written as

\[
\frac{\alpha I}{4k_n T} = \ln(r) + 2 \left( \frac{B_2}{V_p} \right) f_0 (r-1) + 2 \left( \frac{B_1}{V_p^2} \right) f_0^2 \left( r^2 - 1 \right),
\]

where the particle polarizability is given by \( \alpha = 3V_p \varepsilon_0 n_b^2 \left( m^2 - 1 \right) / \left( m^2 + 2 \right) \). Equation (4) is valid when the virial expansion converges

\[
\frac{P}{k_n T} = \rho + B_2 \rho^2 + B_4 \rho^4 + \ldots \approx \rho \left( 1 + B_2 \rho + B_4 \rho^2 \right),
\]

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with \( p \) and \( \rho \) the pressure and density of the gas, which is valid if \((B_2/V_p)f_{\text{max}}^2 < 1\), and \((B_3/V_p^2)f_{\text{max}}^2 < 1\). Matuszewski et al. [11,12] have considered the case that the colloidal suspension is treated as a hard sphere gas which allows for the treatment of the virial coefficients to all orders. Our treatment though limited to the second and third virial coefficients allows, in principle, for the inclusion of arbitrary inter-particle interaction potentials.

In the limit of an “ideal gas” in which particle-particle interactions are neglected, \((B_2/V_p)f_{\text{max}}^2 \rightarrow 0\), \((B_3/V_p^2)f_{\text{max}}^2 \rightarrow 0\). Eq. (6) yields \( r(I) = \exp(I/I_c) \), where \( I_c = 4k_B T/\alpha \), giving the limit of the exponential model for the nonlinearity

\[
\Delta n(I) = (n_p - n_h)V_p \rho_0 e^{I/I_c} \approx n_{2K} I_c + n_{2K} I,
\]

where we identify the nonlinear coefficient \( n_{2K} = (n_p - n_h)V_p \rho_0 / I_c \) in the limit of an artificial Kerr medium. In contrast, combining Eq. (5) and Eq. (6) for the non-ideal gas case with \( B_2 \) and \( B_3 \) non-zero yields

\[
n_2 = \frac{n_{2K}}{1 + 2(B_2/V_p)f_0 + 3(B_3/V_p^2)f_0^2},
\]

\[
n_4 = \frac{2n_{2K}^2}{2I_c} \left[ \frac{1 - 3(B_3/V_p^2)f_0^2}{1 + 2(B_2/V_p)f_0 + 3(B_3/V_p^2)f_0^2} \right].
\]

These formulae reveal that including the virial coefficients softens the effective Kerr nonlinear optical response with respect to the exponential model since \( n_2 < n_{2K} \), but that higher-order nonlinearities are also present yielding a super-Kerr nonlinear response, though we limit our attention to \( n_4 \) here. We further see that the relative role of the exponential nonlinearity, represented by the \( \ln(r) \) term in Eq. (6), depends on the magnitude of the first virial coefficients. Junio et al. have previously described the necessity of including higher-order nonlinearities in describing colloidal suspensions [16].

In addition to the parameter values already quoted, to facilitate comparison with the experiment we shall set \( B_3 = 0 \) and vary the scaled second virial coefficient \( (B_2/V_p) \) which we expect to be dominant. Figure 3 shows the variation of the scaled nonlinear index change \([\Delta n(I)/n_{2K}I_c - 1]\) versus scaled intensity \((I/I_c)\) for \( (B_2/V_p) = 25 \) using both the non-ideal gas model in Eq. (6) (red line) and the exponential model in Eq. (8) (blue line), and a considerable softening of the exponential model is clearly seen in the fact that the index change increases far less rapidly with intensity for the non-ideal gas. Inclusion of the virial coefficients is therefore an important factor in understanding how the exponential model can give way to the notion of an artificial Kerr medium or a super-Kerr nonlinear response.
Fig. 3. shows the variation of the scaled nonlinear index change $\frac{\Delta n(I)}{n_{2k} I_c} - 1$ versus scaled intensity $\left(\frac{I}{I_c}\right)$ for $(B/\nu_p) = 25$ using both the non-ideal gas model in Eq. (6) (red line) and the exponential model in Eq. (8) (blue line).

4. Numerical results

To analyze the experimental results in Fig. 2 we have used two numerical approaches, namely Beam Propagation Method (BPM) simulations based on Eqs. (1)-(4), and also a variational Gaussian (VG) model based on the non-ideal gas model in Eq. (9) using $n_2$ and $n_4$. Although the BPM results are ultimately more accurate we concentrate on the VG model as it offers some insights into the underlying physics and it is important as a check on the purely numerical BPM results. In all cases we checked that the same trends occur in both the BPM and VG results. First we describe our VG model and then we turn to our numerical results. Throughout our numerical study the parameters used are those previously quoted unless otherwise stated.

4.1 Variational Gaussian model

To analyze propagation of the initial Gaussian beam (3) according to Eq. (1) we employ the variational approach of Anderson and Bonnedal [17]. In particular, we use the results for $n_2$ and $n_4$ for the non-ideal gas in Eq. (1) by replacing $\alpha_0 r = k_0 n_2 I + k_0 n_4 I^2$, and we set the Rayleigh scattering losses to zero. In the variational approach a suitable Lagrangian is introduced that yields the paraxial wave Eq. (1) when the Euler-Lagrange equations are evaluated. Although this makes an exact solution no easier it permits and approximation based on a prescribed trial function, which we choose here as a Gaussian

$$\varphi(x, y, z) = \frac{2P_m}{\pi w^2(D)} e^{\mathcal{R}(z)} \exp\left(-\frac{z^2}{R(z)} + \frac{x^2 + y^2}{w(z)^2} + ik(x^2 + y^2)/2R(z)\right),$$

where $\mathcal{R}(z)$ is the on-axis phase of the field, $w(z)$ is the Gaussian spot size of the propagating field, and $R(z)$ is the radius of curvature of the field. Here we have the initial conditions $w(0)$
= w₀ and 1/R(0) = 0 since the initial Gaussian is collimated. Evaluating the Euler-Lagrange equations we find the equations of motion for the Gaussian beam spot size and radius of curvature [17,18]

\[
\frac{d^2 w}{dz^2} = \frac{4}{k^2 w^3} \left( 1 - \frac{P_c}{P_r} - \frac{8w_0^2}{9w^2} \left( \frac{n_3 I_{in}}{n_2} \right) \left( \frac{P_c}{P_r} \right) \right), \quad \frac{1}{R(z)} = \frac{1}{w} \frac{dw}{dz},
\]

(11)

where the critical power for self-focusing due to the Kerr effect alone (n₂) for a Gaussian beam is given by

\[
P_c = \frac{1.8962 \lambda^2}{4\pi n_2 n_0},
\]

(12)

and \( I_{in} = 2P_c / \pi w_0^2 \). These equations can be solved to find the Gaussian spot size and radius of curvature at the collecting fiber, and the measured power calculated using

\[
P_{coll} = \left( \frac{4P_w}{\pi^2 w_0^2 w^2(D)} \right)^2 \left( \int dx dy e^{-(x^2+y^2) / w_0^2} e^{-(x^2+y^2) / w^2(D)} e^{i(kx^2+y^2) / 2R(D)} \right)^2
\]

\[
= \frac{4P_w}{\pi^2 w_0^2 / w^2(D)} \left( 1 + \frac{w_0^2}{w^2(D)} - \frac{ikw_0^2}{2R(D)} \right)^2,
\]

(13)

which is physically the modulus squared of the propagated Gaussian field projected onto the collecting fiber mode. Note that both the propagated beam spot size \( w(D) \) and the beam curvature \( R(D) \) affect the power measured at the collecting fiber.

For a given set of parameters we solve Eq. (11) numerically with the initial condition \( w(0) = w_0, dw(0) / dz = 0 \) for an incident collimated beam (1/R = 0), and then calculate the collected power using Eq. (13). Physical insight into the self-focusing dynamics may be obtained by recognizing that Eq. (11) is analogous to Newton’s equation for a unit mass particle moving in one dimension with \( w \) playing the role of the particle coordinate, \( z \) the role of time, and the right-hand-side being the force. Self-focusing contraction of the beam occurs when the force is negative meaning that the particle is attracted to the origin (\( w = 0 \)). For an artificial Kerr medium (\( n_2 > 0, n_4 = 0 \)) the force is negative and self-focusing collapse occurs for \( P_w > P_c \). In contrast for the non-ideal gas model with both \( n_2 \) and \( n_4 > 0 \) present the force can be negative and self-focusing collapse phenomena can occur even for \( P_w < P_c \) if the higher-order term is large enough. Thus we anticipate that for the non-ideal gas model the critical power based on the artificial Kerr nonlinear coefficient alone is not generally sufficient to understand the beam propagation.

### 4.2 Exponential nonlinearity

One general finding from the numerical study of our experiment in Fig. 1 is that BPM simulations based on Eq. (1) and the exponential nonlinearity (7) could under no amount of fine tuning of parameters be made to resemble the experimental results in Fig. 2. The reason is very simple, namely, since the exponential nonlinearity includes arbitrarily high orders of self-focusing nonlinearity, once self-focusing collapse starts it is immediately super-critical [12], leaving no room for the relatively smooth rollover seen in Fig. 2. Rather, using the exponential nonlinearity the plot of the power measured by the collecting fiber versus the input power is initially linear with input power, then it increases very sharply with increasing input power, and finally drops abruptly to zero above a given power. (This plot is not included...
since it was virtually impossible to maintain numerical accuracy once the collapse initiated). We conclude that the pure exponential model is therefore not a good representation of the nonlinear optical response of our experiment.

### 4.3 Artificial Kerr medium

We next consider the numerical results treating the colloidal suspension as an artificial Kerr medium for which we truncate the exponential nonlinearity to the first nonlinear term shown in Eq. (8). Using the previously quoted numerical parameters we obtain $n_{2K} = 2.3 \times 10^{-9}$ cm$^2$/W, which yields the critical power $P_{crK} = 0.6$ W from Eq. (11) with $n_2$ replaced by $n_{2K}$ (We remark that the critical power for the lower concentration example in Fig. 2 calculated in this manner is 5 W). Figure 4 shows a plot of the power measured by the collecting fiber versus the input power using the artificial Kerr medium model, both powers being normalized to $P_{crK}$. The red line is obtained using the VG model and the blue line is obtained using the BPM with good overall agreement between the models, though there is a small but discernible shift in the position of the rollover power. In particular, we see that the rollover occurs at an input power $P_{roll} \approx 0.6$ W for both models, in reasonable agreement with the experimental value of 0.5 W. Indeed, fine tuning of the numerical parameters can easily reduce the difference in the rollover powers between the experiment and models, and it is tempting to do so. However, this still begs the questions of why the exponential model can be successfully truncated to the artificial Kerr medium model or super-Kerr model, what is the rationale for this? We shall see next that the non-ideal gas model provides a good physical explanation.

![Graph of power measured by the collecting fiber versus input power using artificial Kerr medium model](image)

**Fig. 4.** shows the power measured by the collecting fiber versus the input power using the artificial Kerr medium model, both powers being normalized to the critical power for self-focusing $P_{crK}$. The red line is obtained using the VG model and the blue line is obtained using the BPM.

### 4.4 Non-ideal gas model

We next present VG results based on the non-ideal gas model with values for the nonlinear coefficients $n_2$ and $n_4$ based on Eq. (9) for the previous quoted parameter values and varying values of the second virial coefficient ($B_2/V_p$) (for this study we set $B_3 = 0$). From Eq. (9) it is...
already clear that increasing \((B_2/V_p)\) past zero will reduce the Kerr nonlinear coefficient \(n_2\) below the value \(n_{2K}\) for an artificial Kerr medium, and this will cause the critical power \(P_{cr}\) in Eq. (12) to be even greater than that for the artificial Kerr medium \(P_{crK} = 0.6\ W\). This would seem to be a step backwards in terms of improving the agreement with the experiment. However, this argument ignores the presence of the next order nonlinearity that is proportional to \(n_4\). Figure 5 shows the power measured at the collecting fiber versus the input power according to the VG model for \((B_2/V_p) = 10\) (blue line \(P_{roll} = 0.4\ W\)), \((B_2/V_p) = 20\) (red line \(P_{roll} = 0.5\ W\)), and \((B_2/V_p) = 30\) (green line \(P_{roll} = 0.6\ W\)). (Using the BPM we find \(P_{roll} = 0.4, 0.5, 0.62\ W\) for \((B_2/V_p) = 15, 25, 35\), respectively). In all three cases we see that by properly including both the nonlinear coefficients \(n_2\) and \(n_4\) the rollover power is less than 0.6 W in all cases, the rollover power being \(P_{roll} = 0.5\ W\) for \((B_2/V_p) = 20\) (red line) which gives the best fit estimate to the experiment and values for the nonlinear coefficients \(n_2 = 1.66 \times 10^{-9}\ cm^2/W\), \(n_{2K} = 4.2 \times 10^{-16}\ cm^4/W^2\). This value for \(n_2\) is in reasonable agreement with the previously quoted value \(n_2 = 2 \times 10^{-9}\ cm^2/W\) measured by the Z-scan method.

Junio et al. [16] have previously commented on the necessity of incorporating higher-order nonlinear effects to account for dipole-dipole interactions between the spheres. This type of interactions is accounted here (along with other many body effects) through the second virial coefficient. As noted earlier, the non-ideal gas model accounts for the compressibility of the colloidal suspension via the virial coefficients, and their inclusion softens the nonlinear optical response from the exponential nonlinearity model (see Fig. 3). In this way the exponential model can be tamed to produce an artificial Kerr medium, but we also see the necessity of including the higher-order nonlinearity proportional to \(n_4\) thus yielding a super-Kerr nonlinear response. We can also compare with the hard sphere gas model of Matuszewski et al. [10,11] and we find the value of the second virial coefficient \((B_2/V_p) = 4\), which is too small to explain our experiment.
We finish by noting that we have used the rollover power as the fit parameter between theory and experiment, but it must be noted that there is some discrepancy between the functional form of the experimental and theoretical plots of the variation of collected power versus input power in Figs. 2 and 4. In particular, the theoretical plots vary far more abruptly past the rollover point in comparison to the experiment. However, the shot-to-shot variations in the experimental collected power, reflected in the vertical error bars, are also greater past the rollover. The abrupt drop off past the rollover in the theory is most certainly a consequence of super-critical collapse due to the \( n_4 \) nonlinearity [12] which translates into the sensitivity to small variations in the input power in the experiment beyond the rollover. This makes the detailed comparison of theory and experiment past this rollover point more difficult.

4. Summary and conclusions

In summary, we have introduced an optical fiber-based diagnostic experiment that can be used to probe the nonlinear optical response of a colloidal suspension and provide a test of the theoretical approaches that have been proposed. The exponential nonlinearity model was found to have the least correlation with our experiment, followed by the artificial Kerr medium approach. Including the effects of compressibility via the second virial coefficient in the non-ideal gas model was found to yield good agreement with the experiment, and in turn can be used to infer values for the second virial coefficient \( (B_2/\rho) \), and the nonlinear coefficients. Based on the experience of these findings we plan to extend the approach in the future to more accurately measure the nonlinear optical response, and also probe the value of the higher virial coefficients. A theoretical issue that deserves more attention is how the granularity of the colloidal suspension enters the nonlinear optical response. In particular, for the concentration used in our experiment the mean distance between colloids is \( \ell \approx \rho_0^{-1/3} \approx 0.4 \, \mu m \), and we expect physically that the nonlinear refractive-index change will saturate in value for an optical beam whose size approaches this value. This is perhaps where a non-local nonlinear model could enter the theoretical treatment and lead to a further softening of the nonlinear optical response in comparison to the exponential model.

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