Spatial solitons and light-induced instabilities in colloidal media

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Abstract: We study nonlinear light propagation in colloidal nanosuspensions. We introduce a novel model for the nonlinear response of colloids which describes consistently the system in the regimes of low and high light intensities and low/large concentrations of colloidal particles. We employ this model to study the light-induced instabilities and demonstrate the formation of stable spatial solitons as well as the existence of a bistability regime.

OCIS codes: (190.1450) Bistability, (190.3970) Microparticle nonlinear optics, (190.6135) Spatial solitons.

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1. Introduction

The effect of nonlinear interaction of light with suspensions of nanometer-sized colloidal dielectric particles has been known for over two decades [1–4]. Yet, very recently the interest in these systems has been revived [5–7], with several attempts to tackle the physics beyond the simple Kerr limit [8–11]. The origin of relatively high optical nonlinearity, first predicted by Palmer [1] in the case of aerosols of dielectric spheres, is due to the optical gradient force acting on the dielectric particles. The effect of this force is to change the concentration of colloidal particles, increasing the refractive index contrast in regions of higher light intensity. It was shown experimentally that the resulting self-focusing nonlinearity can lead to the beam self-focusing, formation of solitons, four-wave mixing or optical phase conjugation [2–7].

To describe mesoscopic processes taking place in these systems, one has to employ the formalism of statistical physics. In the simplest case, the balance between the optical gradient force and particle diffusion due to Brownian motion leads to the propagation equation in the form of nonlinear Schrodinger equation (NLS) with Kerr nonlinearity [1–4]. It was shown recently that inclusion of particle interactions can lead to spatially nonlocal equation describing light propagation [8, 9]. However, all these models treat the light force as a small perturbation, resulting in a linear dependence of the nonlinear index change on the light intensity. The generalization of the noninteracting particles model for the case of higher light intensities has been proposed very recently by two independent groups [10, 11]. The resulting form of the nonlinearity was found to be exponential leading, for instance, to unphysical catastrophic collapse phenomenon even in the one-dimensional case. It is hence obvious that for high light intensity the noninteracting particles approximation is no longer valid and better, physically justified model is required.

In this paper, we introduce, what we believe, is the first model of light propagation in colloids which takes into account particle interactions and therefore correctly describes colloidal system in the regimes of low and high optical intensity and low and large particle concentrations. We consider the case of particles interacting through the hard sphere potential [12]. We show that our model correctly reproduces results of the exponential nonlinearity [10, 11] in the relevant light intensity regime but also predicts the phenomenon of soliton bistability which can be present in these systems in the one-dimensional geometry. This is confirmed by our semi-analytical and numerical calculations. We also demonstrate soliton switching when slightly perturbed solution from the unstable branch switches to the stable one which corresponds to higher particle concentration.

2. Model

We assume that the dielectric colloidal particles interact with each other through a hard sphere potential. In the steady state the colloidal particles satisfy the Maxwellian velocity distribution, which follows from the phase space density in the canonical ensemble $\rho \sim \exp(-E/k_{\mathrm{B}}T)$. The pressure exerted by colloidal particles can be obtained from the equation of state in analogy with the hard sphere gas [12]

$$\frac{\beta P}{\rho} = Z(\eta)\,, \tag{1}$$

where $\beta = 1/k_{\mathrm{B}}T$, P is the pressure, ρ is the colloidal particle density, $Z(\eta)$ is the compressibility, and $\eta = \rho/\rho_0$ is the packing fraction. In the case of ideal gas we have $Z = 1$. For a hard sphere gas the Carnahan-Starling formula $Z \approx (1+\eta+\eta^2-\eta^3)/(1-\eta)^3$ gives a very good approximation up to the fluid-solid transition at $\eta \approx 0.5$ [12]. This phenomenological formula is in agreement with exact perturbation theory calculations as well as molecular dynamics simulations.

In the presence of slowly varying external potential, such as that induced by the presence

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of optical beam, the particle velocity distribution is locally Maxwellian. The gradient of the density $\rho(\mathbf{r})$ is assumed to be locally parallel to $\hat{x}$, and we consider a small box of volume $\mathrm{d}V = \mathrm{d}x\mathrm{d}S$, with length $\mathrm{d}x$ and normal surface $\mathrm{d}S$. The difference in pressure exerted on the right and left surface $\mathrm{d}P$ gives rise to an effective force acting on the colloidal particles F_{int}. It is equal to the external force that is necessary to sustain the density gradient, and $\mathrm{d}P = -F_{\mathrm{int}}/\mathrm{d}S = -f_{\mathrm{int}}\rho\mathrm{d}V/\mathrm{d}S = -f_{\mathrm{int}}\rho\mathrm{d}x$, where f_{int} is the average force acting on a single particle. Using Eq. (1) we get $\mathrm{d}(\rho Z)/\mathrm{d}x = -f_{\mathrm{int}}\rho\beta$. The particle current density is equal to

$$\vec{j} = \rho\mu(\vec{f}_{\mathrm{ex}} + \vec{f}_{\mathrm{int}}) = \rho\mu\vec{f}_{\mathrm{ex}} - D\nabla(\rho Z)\,, \tag{2}$$

where μ is the particle mobility, and $D = \mu/\beta$ is the diffusion constant. In the ideal gas limit, this equation becomes Eq. (3) of [11]. Let $m = n_{\mathrm{p}}/n_{\mathrm{b}}$ be the ratio of the colloidal particle refractive index to the background refractive index. Polarizability of a sphere is given by

$$\alpha = 3V_{\mathrm{p}}\varepsilon_0 n_{\mathrm{b}}^2\left(\frac{m^2-1}{m^2+2}\right), \tag{3}$$

where V_{p} is the sphere volume. If we look for the steady state ($\vec{j} = 0$) in the presence of optical field gradient ($\vec{f}_{\mathrm{ex}} = (\alpha/4)\nabla I$, where $I = |E|^2$) we obtain

$$\rho\frac{\alpha\beta}{4}\frac{\mathrm{d}I}{\mathrm{d}x} = \frac{\mathrm{d}(\rho Z)}{\mathrm{d}x}\,, \tag{4}$$

which can be solved analytically to give the dependence $I(\eta)$

$$\frac{\alpha\beta}{4}I(\eta) = g(\eta) - g(\eta_0)\,, \tag{5}$$

where $g(\eta) = (3-\eta)/(1-\eta)^3 + \ln\eta$, and η_0 is the background packing fraction. For small η this result is equivalent to an exponential dependence derived in Refs. [10, 11]

$$\eta = \eta_0\exp\left(\frac{\alpha\beta}{4}I\right). \tag{6}$$

Our model neglects nonlocal effects that were considered earlier by other authors [8, 9]. It was pointed out that in the case of interaction trough the hard sphere potential the nonlocality is negligible [9].

The effect of Rayleigh scattering, although always present, can be greatly reduced in the case of small colloidal particles, since it is proportional to the third power of the particle radius for a given packing fraction [4].

Assuming relatively low packing fraction, the corresponding nonlinear refractive index change can be approximately calculated using the Maxwell–Garnett formula [13]

$$\varepsilon_{\mathrm{eff}} = \varepsilon_{\mathrm{b}} + \frac{3\eta\varepsilon_{\mathrm{b}}\left(\varepsilon_{\mathrm{p}}-\varepsilon_{\mathrm{b}}\right)}{\left[\varepsilon_{\mathrm{p}}+2\varepsilon_{\mathrm{b}}-\eta\left(\varepsilon_{\mathrm{p}}-\varepsilon_{\mathrm{b}}\right)\right]}\,. \tag{7}$$

For low refractive index contrast ($\varepsilon_{\mathrm{p}}/\varepsilon_{\mathrm{b}} \approx 1$) we have

$$\varepsilon_{\mathrm{eff}} \approx \varepsilon_{\mathrm{b}} + \frac{3\varepsilon_{\mathrm{b}}\left(\varepsilon_{\mathrm{p}}-\varepsilon_{\mathrm{b}}\right)}{\left(\varepsilon_{\mathrm{p}}+2\varepsilon_{\mathrm{b}}\right)}\eta = \varepsilon_{\mathrm{b}} + \delta\eta\,. \tag{8}$$

Note that signs of α and δ are the same. Substituting this formula to the Helmholtz equation $\nabla^2 E + k_0^2 n_{\mathrm{eff}}^2 E = 0$, we obtain propagation equation for slowly varying envelope of the electric

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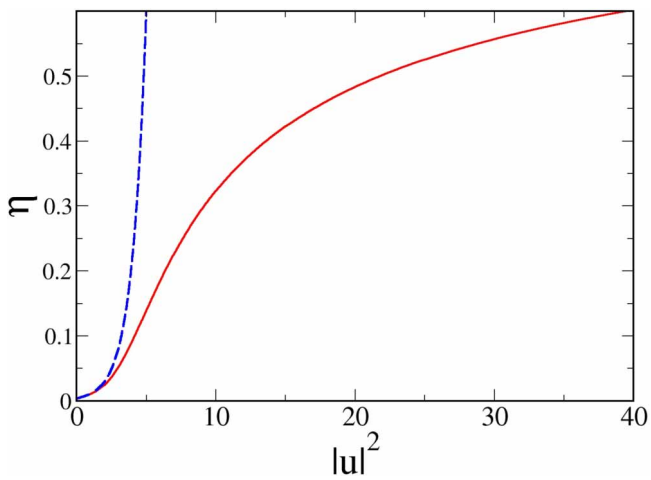

Fig. 1. Packing fraction η of colloidal particles vs. the light intensity (solid line). The dashed line shows the dependence of the exponential model, see Eq. (6).

field defined by $E = u \exp\left[ik_0 (\varepsilon_b + \delta\eta_0)^{1/2} z\right]$

$$i\frac{\partial u}{\partial z} + \frac{1}{2}\nabla_\perp^2 u \pm (\eta - \eta_0)u = 0\,, \tag{9}$$

in normalized units, where $\pm$ correspond to the case of positive or negative $(m-1)$, and

$$|u|^2 = \pm[g(\eta) - g(\eta_0)]\,. \tag{10}$$

Notice that since $g(\eta)$ is monotonically increasing, the nonlinearity is always self-focusing independently of whether $m > 1$ or $m < 1$ [11]. In the following, we will focus on the case of $m > 1$ (+), which was realized in number of experiments with polystyrene colloids [2–7].

Typical dependence $\eta(|u|^2)$ in this case is shown in Fig. 1. In the low intensity limit, the nonlinear index change is Kerr-like (proportional to intensity). For higher intensities, it is well described by the exponential model of [10, 11]. Finally, for higher densities the particle hard-sphere interactions become significant and the nonlinearity saturates as the exponential model breaks down. One can show that for high background packing fraction η_0 the exponential regime can be absent, with direct transition from the Kerr-like to saturated regime.

3. Modulational instability

Continuous wave solutions to the Eq. (9) have the form $u = |u_s|\exp(i\eta_s z + i\phi)$, where the relation (10) holds between u_s and η_s. We calculate the growth rate of linear perturbations around this solution by substituting

$$u = \left\{|u_s| + a(t)\mathrm{e}^{ikx} + b(t)\mathrm{e}^{-ikx}\right\}\mathrm{e}^{i\eta_s t}\,, \tag{11}$$

and expanding $\eta \approx \eta_s + \eta'(|u_s|^2)(|u|^2 - |u_s|^2)$. The perturbations $a(t)$ and $b(t)$ grow as $\exp(i\omega t)$, where

$$\omega^2 = k^2\left(\frac{k^2}{4} \mp \eta'(|u_s|^2)|u_s|^2\right)\,. \tag{12}$$

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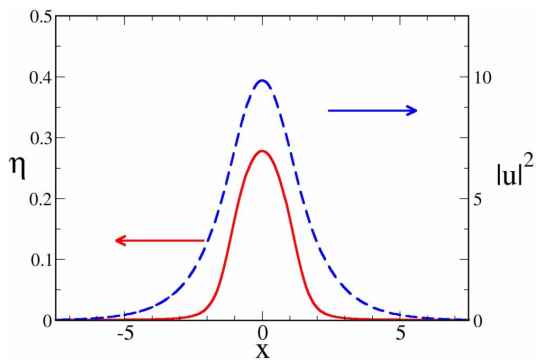

Fig. 2. Profile of colloidal particle packing fraction (solid) and light intensity (dashed) of a soliton solution for parameters $\eta_0 = 10^{-3}$, $\kappa = 0.1$, $P = 35$, $m > 1$.

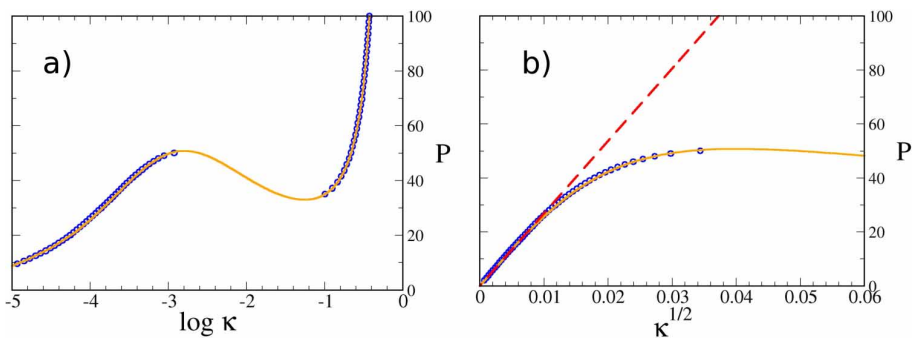

Fig. 3. Soliton power against the propagation constant for $\eta_0 = 10^{-3}$ in (a) logarithmic scale (b) square root scale up to $\kappa^{1/2} = 0.06$. Solid line shows the value of the integral (15) and circles correspond to stable numerical solutions of the Eq. (9). The dashed line in (b) is a linear fit to the Kerr-like dependence for low κ.

The CW solution will be modulationally unstable if $\pm\eta'(|u_s|^2) > 0$. We can calculate this derivative using (10)

$$\pm\frac{\mathrm{d}\eta}{\mathrm{d}|u|^2} = \pm\left(\frac{\mathrm{d}|u|^2}{\mathrm{d}\eta}\right)^{-1} = \left[\frac{1}{\eta} + 2\frac{4-\eta}{(1-\eta)^4}\right]^{-1}. \tag{13}$$

It is easy to see that instability always occurs for both $m > 1$ and $m < 1$. The wavevector corresponding to maximum growth rate $\mathscr{I}(\omega_m) = \eta'|u|^2$ is $k_m = \sqrt{2\eta'|u|^2}$. Notice that for low light intensity regime this formula leads to the result of El-Ganainy [14].

4. Soliton solutions and bistability

We are looking for soliton solutions in the form $u(\mathbf{r}) = A(\mathbf{r}_\perp)\exp(i\kappa z)$ in the case $m > 1$. In the one-dimensional geometry, which was realized in the recent experiment [6], this leads to

$$-\kappa A + \frac{1}{2}\frac{\mathrm{d}^2 A}{\mathrm{d}x^2} + (\eta - \eta_0)A = 0\,. \tag{14}$$

This equation can be immediately integrated once leading to the first-order differential equation for $\eta(x)$, which can be subsequently solved numerically for given η_0 and κ. In Fig. 2, we

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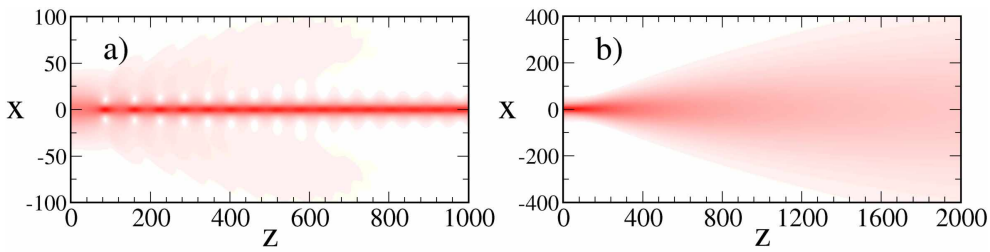

Fig. 4. Propagation of a perturbed unstable soliton for $\eta_0 = 10^{-3}$, $\kappa = 9 \times 10^{-3}$, $P = 41$, $m > 1$: (a) increasing the soliton power by 1% results in switching to a stable soliton after losing about 10% of the beam power to radiation; (b) decreasing the soliton power by 1% leads to diffraction rather than switching to the first stable branch.

plot an example of the soliton profile for the parameters $\eta_0 = 10^{-3}$, $\kappa = 0.1$, and $m > 1$. For experimental parameters $\lambda = 1064$ nm, particle diameter $d = 40$ nm, $n_p = 1.56$, $n_b = 1.33$, this soliton peak intensity reaches ≈ 200 MW/cm^2, and the total beam power is about 1W. At the same time, Rayleigh losses after 1mm of propagation are equal to 12%, while the characteristic nonlinear length is about 10 μm due to high nonlinear index contrast. The soliton power can be calculated as

$$P = \int_{-\infty}^{+\infty} A^2 \mathrm{d}x = 2 \int_{\eta_0}^{\eta_{\max}} A^2(\eta) \left(\frac{\mathrm{d}\eta}{\mathrm{d}x} \right)^{-1} \mathrm{d}\eta \,, \tag{15}$$

where $\eta_{\max}$ is the value at which $\mathrm{d}\eta/\mathrm{d}x = 0$, and it corresponds to the soliton maximum. An example of the dependence $P(\kappa)$ for $\eta_0 = 10^{-3}$, $m > 1$ is shown in Fig. 3.

For comparison, numerical solutions of Eq. (9) obtained with the numerical imaginary-time method are depicted by circles. Using the Vakhitov-Kolokolov criterion for the soliton stability [15], namely $\mathrm{d}P/\mathrm{d}\kappa > 0$, we conclude that there exists a region of bistability for the beam powers corresponding to $P \approx 35 - 50$ [16]. This observation is confirmed by direct numerical simulations of Eq. (9) by monitoring stability of slightly perturbed soliton solutions. This bistability phenomenon is present only for the background packing fraction $\eta_0 < \eta_\mathrm{b} \approx 0.5\%$.

The two branches of stable solitons are separated by an unstable branch. We investigate a possibility of switching of unstable solitons by slightly perturbing its amplitude. The corresponding results are shown in Fig. 4. If the soliton amplitude is slightly increased, the soliton evolves into a stable solution corresponding to the larger values of the propagation constant of a higher branch. On the other hand, decreasing the soliton amplitude leads to the beam diffraction rather than its switching to a lower branch of stable solitons.

5. Conclusions

We have analyzed the light localization in colloidal suspensions composed of dielectric nanospheres. We have derived a new model of the nonlinear response of colloids which takes into account particle interactions through the hard-sphere potential. We have demonstrated that this model leads to nonlinearity saturation at higher light intensities or large particle concentrations. We have revealed that this nonlinear response leads to bistable soliton solutions.

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