Experimental Observation of Modulation Instability and Optical Spatial Soliton Arrays in Soft Condensed Matter

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In this Letter we report observations of optically induced self-organization of colloidal arrays in the presence of unpatterned counterpropagating evanescent waves. The colloidal arrays formed along the laser propagation axis are shown to be linked to the breakup of the incident field into optical spatial solitons, the lateral spacing of the arrays being related to modulation instability of the soft condensed matter system.

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Optical spatial solitons (OSS) are spatially localized, nondiffracting modes that are supported in nonlinear optical media. They are the result of a balance between diffraction and self-phase modulation which results in self-focusing under intense illumination [1,2]. For a plane-wave incident field, small wave front perturbations cause the optical field to breakup into periodic arrays of OSS [3,4] or more complex patterns [5], an effect known as modulation instability (MI) [6–8]. Both OSS and MI are generic properties of wave propagation governed by the nonlinear Schrödinger equation. In addition to nonlinear optics, there are numerous examples in nature where such solitary waves and related phenomena are observed, particularly in pattern formation in granular systems [9] and complex fluids [10].

Recently Conti *et al.* suggested the possibility of OSS formation and MI in soft condensed matter systems, where local concentrations of constituent particles provide localized intensity dependent refractive index variations [11,12]. They developed a nonlocal theory which relates the Kerr response to a static structure factor, specific to the constituent materials. Ashkin and co-workers indicated the potential of bulk colloidal suspensions to act as artificial Kerr medium in a number of nonlinear optical experiments, including self-focusing, optical bistability, and four-wave mixing [13–15]. The nonlinearity is an electrostrictive effect arising from optical gradient forces experienced by the dielectric particulates, which cause them to aggregate at regions of high intensity, thereby locally increasing the refractive index and leading to a self-focusing effect [16].

In this Letter we present an experimental system, based on an optical waveguide geometry, to explore the nonlinear optical properties of large-scale colloidal aggregates. We show that the nonlinear behavior of the system leads to observation of optically induced self-organization and particularly that colloidal arrays formed along the laser propagation axis are linked to the breakup of the incident field into OSS. The lateral spacing of the arrays is related to modulation instability of the nonlinear coupled light-matter system. These results represent the first conclusive experimental realization of both MI and OSS arrays in colloidal suspensions [11,12]. We note Yashin *et al.* have indicated the formation of solitonlike beams in bulk colloidal samples, but no conclusive demonstration of spatial solitons or any inference of MI was forthcoming in that study [16].

For the experimental work a prism-coupled resonant dielectric waveguide was used to generate counterpropagating (CP) waveguide modes with an evanescent wave (EW) component extending in to the colloidal solution (see Fig. 1). Transverse optical gradient forces due to the EW acting on particles in proximity to the supporting surface result in the accumulation of particles at the center of the illuminated area, where the scattering forces due to the CP fields along the propagation (x) axis are balanced [17]. The waveguide constrains the field profile perpendicular to the waveguide to be the lowest order mode along the z axis:

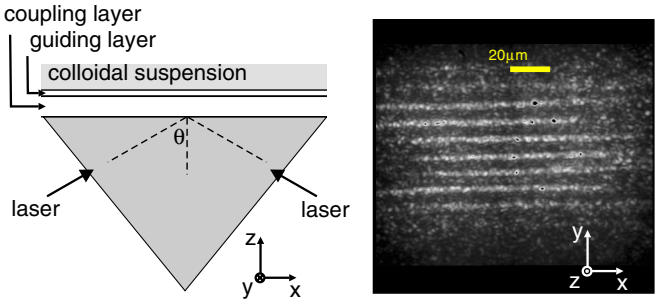

FIG. 1 (color online). A schematic diagram outlining the geometry of the experimental setup and a light scattering image of spontaneous ordering of 410 nm polymer colloids seen under typical experimental conditions. The top left diagram indicates the orientation of coupling prism; the propagation direction of the two counterpropagating modes are along the x direction and the observed array formation is aligned along the x direction with a periodic spacing in the y direction (top right). The bottom diagram shows the structure of the nonlinear prism-coupled slab waveguide used for the experiments.

this effectively eliminates one transverse dimension (z) leaving the propagation axis x and transverse dimension y. This particular design maximizes the nonlinear effects of the colloidal dispersion by using a waveguide design with a mode that has an enhanced field strength in the sample solution [18].

Optical trapping was achieved using two CP waveguide modes excited through the prism coupler. TE polarized light from a 1064 nm, Yb-doped fiber laser was weakly focused onto the coupling layer-prism interface at an angle of approximately 60°; the spot size in at the sample surface in lateral direction was 100 μm. A second beam, coupled through the opposite prism face, was used to excite the opposing mode; the two beams where not temporally coherent. The relative power in each arm was also adjusted to achieve stable trapping at the center of the trap; all powers quoted refer to the accumulative power in both arms.

Submicrometer diameter sized colloids accumulated in the trap at high incident powers. They formed regularly spaced linear arrays oriented along the direction of the incident EW wave vector with a lateral separation of several micrometers. The presence of large numbers of particles was necessary to observe the pattern formation. Video stills of the array formation are shown in Fig. 2 for a solution of 410 nm colloidal particles (concentration equivalent to 0.1% solid) accumulated in the counterpropagating cavity-enhanced evanescent wave trap (total incident power = 600 mW). Linear arrays, formed along the propagation direction of the evanescent waves, are regularly separated in the lateral direction with a periodicity of approximately 10 μm. We observe linear arrays under a range of experimental conditions including, different concentrations of colloids and different sized particles (200–600 μm diameter). Whilst particles within the array were arranged in a disordered manner and exhibit strong Brownian fluctuations, we observed no evidence of thermal gradient based convective or thermophoretic motion [19], indicating that the effect is predominantly an optical one.

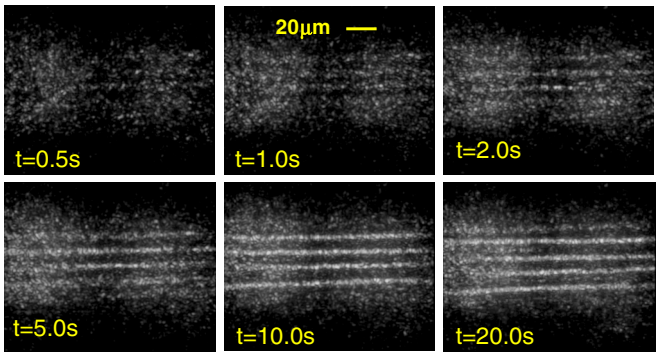

FIG. 2 (color online). The temporal evolution of the OSS formation for incident powers above the threshold. Each panel is a CCD image of the illuminated region at different times after the light is coupled to the system. Bright field imaging also confirms the presence of ordered colloids in regions of high intensity.

We associate and explain this new type of optically-induced self-organization due to arrays of OSS formed by the breakup of the waveguide mode through MI. (Because of the presence of waveguide losses and external pumping via the prism coupler we are strictly dealing with solitary waves as opposed to solitons.) Qualitatively, the observed ordering exhibits a number of properties that are suggestive of the presence of OSS. First, a power threshold exists below which particles are accumulated but no ordering is observed. Above this power threshold, stable arrays are formed with a characteristic spacing, which extend beyond the footprint of the illuminated area along the propagation axis. The spacing between the arrays in the lateral direction decreases with increasing power, until arrays become unstable and spontaneously evolve leading to merging and branching of arrays. From a simple analysis of the image the spacing between linear arrays is found to be 8.7 ± 0.6 μm. This characteristic spacing is similar for a range of different sample preparations, e.g., diluted samples (0.01% packing fraction) and smaller sized colloids (200 nm): only the power threshold for observing the arrays changes significantly.

We now give a précis of the underlying theory for comparison with the experiment. For a prism-coupled nonlinear planar waveguide the amplitudes $a_{\pm}(x, y)$ for the guided wave fields counterpropagating along the x axis may be described by the coupled nonlinear Schrödinger equations (NLSEs) [20,21]

$$\pm \frac{\partial a_{\pm}}{\partial x} = \frac{i}{2k_0\beta_0}\frac{\partial^2 a_{\pm}}{\partial y^2} + t_m a_{in}^{\pm}(x, y) - \alpha_{\text{eff}} a_{\pm} + ik_0\Delta\beta_0 \int_{-\infty}^{\infty} dy' G(y - y')(|a_{+}(x, y')|^2 + |a_{-}(x, y')|^2)a_{\pm}, \quad (1)$$

where k_0 is the incident wave vector and β_0 is the effective index of the guided wave. The first term on the right-hand side of the NSLEs describes beam diffraction in the transverse dimension (y axis), the second term describes excitation of the guided wave by the incident CP fields $a_{\text{in}}^{\pm}(x, y)$ through the prism with transmission coefficient t_m, and the third term proportional to α_{eff}describes losses due to both medium absorption and reradiation from the guided wave back into the prism. Detailed expressions for the above coefficients are given by Liao *et al.* [19,20]. The second line of Eq. (1) describes nonlinearities in the structure layers due to the colloids, with $G(y)$ a kernel that accounts for the nonlocality of the nonlinear response and $\Delta\beta_0$ the Kerr nonlinear coefficient for a local response with $G(y) = \delta(y)$ [12]. Here we have taken account of the fact that the CP fields are mutually incoherent by neglecting any standing wave effects in the nonlinear optical response.

To understand the pattern forming properties of the nonlinear prism coupler we consider the stability properties of the high power plane-wave solution with respect to spatial modulations along the lateral dimension [6–8]. To proceed we consider perturbed plane-wave solutions of the form $a_{\pm}(x, y) = a_0 + \mu_{\pm}\exp(2\pi i y/L) + \nu_{\pm}\exp(-2\pi i y/L)$, where we have added weak spatial sidebands to the strong plane-wave solution $a_{\pm} = a_0$ of Eq. (1), obtained by choosing $a^{\pm}_{\mathrm{in}}(x, y) = a_{\mathrm{in}}$ and setting the spatial derivatives to zero. If the spatial sidebands grow they will create a spatial modulation of the intensity, and hence colloid density, of period L along the lateral or y dimension. Substituting the perturbed plane-wave solutions into the NLSEs (1), and linearizing in the weak spatial sidebands, one obtains four-wave parametrically coupled equations for the sidebands. By demanding that the four-wave parametric interaction is phase matched we obtain the condition on the spatial period $L = \pi/(k_0\sqrt{2\beta_0\Delta\beta_0 S(0)|a_0|^2})$ of the MI, with $S(q)$ the static structure factor which is the Fourier transform of the kernel $G(y)$. The gain of the parametric interaction may be found by seeking a solution for the spatial sidebands of the form $\mu_{\pm} = \alpha_{\pm}\exp(gx)$, $\nu_{\pm} = \beta_{\pm}\exp(gx)$, which yields for the parametric gain $g = k_0\Delta\beta_0|S(4\pi/L)||a_0|^2 - \alpha_{\mathrm{eff}}$, so the power in the guided wave needs to exceed the threshold value $(|a_0|^2)_{\mathrm{th}} = \alpha_{\mathrm{eff}}/(k_0\Delta\beta_0|S(4\pi/L)|)$ for the growth of arrays to be possible. Thus, at threshold the lateral array spacing will be

$$L_{\mathrm{th}} = \frac{\pi}{k_0\sqrt{2\beta_0\Delta\beta_0 S(0)(|a_0|^2)_{\mathrm{th}}}} = \frac{\pi}{\sqrt{2\beta_0 k_0 \eta\alpha_{\mathrm{eff}}}} \qquad (2)$$

with $\eta = S(0)/S(4\pi/L)$.

By experimentally determining the loss coefficient α_{eff} we may predict the lateral array spacing (2) at threshold that can be compared with the experiment for internal consistency. From a fit of the experimentally measured variation of the low power reflectivity of the prism-coupled waveguide versus incident angle (not shown) we estimate the effective loss coefficient as $\alpha_{\mathrm{eff}} = 110\ \mathrm{cm}^{-1}$. Using Eq. (2) with $\eta = 1$ we obtain a threshold periodicity $L_{\mathrm{th}} = 7\ \mu$m, in reasonable agreement with the experimental value $8.7 \pm 0.6\ \mu$m. This implies that for our current experiment nonlocality is not a key factor, since for a local response $G(y) = \delta(y)$ we have $S(q) = \mathrm{const}$ and $\eta = 1$. This is also consistent with the low packing fraction of the colloidal suspension used in our experiment, 0.1%, which implies that the static structure factor for the colloidal system, viewed as a system of hard spheres, will be largely constant, see Fig. 1 of Ref. [11].

From the model we also find the array spacing above threshold is power dependent. The relation $L = \pi/(k_0\sqrt{2\beta_0\Delta\beta_0 S(0)|a_0|^2})$ relates the lateral array spacing to the guided wave power $|a_0|^2$. Then using the strong plane-wave solution a_0 of Eq. (1), we find the relations

$$\frac{L}{L_{\mathrm{th}}} = \frac{1}{\sqrt{\xi}}, \qquad \frac{(1+4\eta^2\xi^2)}{(1+4\eta^2)}\xi = \frac{P_{\mathrm{in}}}{P_{\mathrm{th}}}, \qquad \xi = \frac{|a_0|^2}{(|a_0|^2)_{\mathrm{th}}}, \qquad (3)$$

where P_{in} is the input power, and P_{th} the threshold input power for array formation which can be determined experimentally. Plotting L/L_{th} versus $P_{\mathrm{in}}/P_{\mathrm{th}}$ parametrically as a function of ξ according to Eqs. (3) we have a prediction of how the lateral array spacing varies with input power. Within the confines of our Kerr nonlinear model the lateral array spacing decreases monotonically with increasing input power beyond the threshold. Figure 3 shows a plot of average array spacing as a function of the input power along with the theoretical prediction obtained with a local nonlinearity $\eta = 1$. There is good overall agreement between the theory and experiment, more so close to threshold where our linearized approximation is more appropriate, and, in particular, that the array spacing decreases with increasing power as expected in the array formation is due to MI.

OSS and MI arise here because we have employed a continuum approximation to the colloidal system, that is, we have approximated the colloidal system as soft matter and ignored its granular structure. This importance of the granular nature of the nonlinear medium, particularly near the limit of the continuum approximation, is highlighted by our observation that the lateral spacing between the arrays begins to decrease with increasing particle size and the emergence of more complicated pattern formation when colloidal particles begin to cluster.

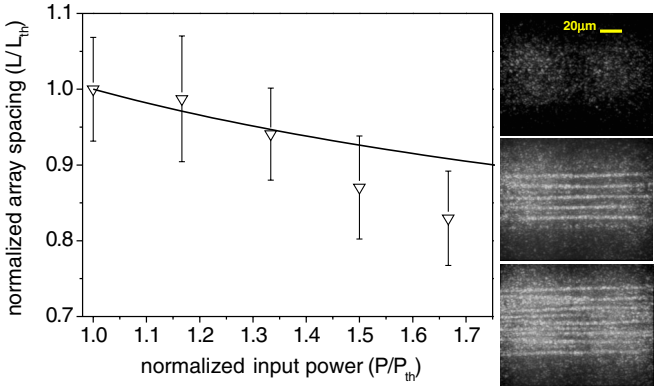

FIG. 3 (color online). The graph on the left shows in the period of the array spacing as a function of the input power above threshold. The experimental data have been normalized with respect to the threshold array spacing and threshold input power. The period (L) is normalized with respect to the period at threshold (L_{th}) and the input coupling power (P) is normalized to the power at threshold (P_{th}). The solid line indicates the variation as predicted by the continuum model. The three panels on the right show the array formations for different illumination intensities. Below the threshold the OSS are not observed (top), even after extended periods of illumination; just above threshold stable arrays of OSS are formed.

We further contend that even for colloidal systems for which the granular nature is more important, the ideas of pattern formation are still of use for intuiting the underlying physics. In particular, continuum models have previously been of great utility in understanding the pattern forming properties of granular media [22]. MI in nonlinear systems may also lead to more complex pattern formation, such as two-dimensional square lattices, hexagonal and spiral type formation; this may be achieved by driving the system well above the threshold [22]. It is also known that interaction forces do exist between solitons, such as attractive forces between in-phase coherent OSS [2]; these forces may play a role in the ordering of higher order solitons. Furthermore by driving the system externally, different types of localized waves, such as oscillons, may be observed [9].

An interesting parallel may be drawn between the OSS/MI interpretation of optical force induced self-organization of colloidal dispersions and optical force mediated particle-particle interactions observed in optical binding, particularly in light of the recent reports of two-dimensional checkerboardlike array formation in a trapping geometry, similar to the one described here [23]. While longitudinal binding (i.e., binding along the propagation axis) has been well characterized, the origin of lateral binding is still not well established. A nonlinear systems approach may provide further insights into the observed pattern formation for such lateral binding, although in this case the granularity of the systems would become increasingly important.

In conclusion we have presented experimental evidence for the observation of modulational instability and formation of optical spatial solitons arrays in soft condensed matter. Our experiments performed on submicron colloidal dispersions offer the first conclusive demonstration in such media of these nonlinear phenomena and the observations are supported by theoretical calculations. The experimental system that we have developed here is an excellent platform for exploring more general properties of optically induced nonlinear instability in colloidal systems. Finally the light-matter interaction has also been noted for smaller numbers of colloid to organize into equilibrium positions due to the gradient and scattering forces: our work may also link to such lateral (or transverse) ''optical binding'' which remains a promising but not well understood method for optically controlled self assembly of particles [23,24].

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