Observation of Bistability and Hysteresis in Optical Binding of Two Dielectric Spheres

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Using a dual-beam fiber optic trap, we have experimentally observed bistability and hysteresis in the equilibrium separations of a pair of optically bound dielectric spheres in one dimension. These observations are in agreement with our coupled system model in which the dielectric spheres modify the field propagation, and the field self-consistently determines the optical forces on the spheres. Our results reveal hitherto unsuspected complexity in the coupled light-sphere system.

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1. Introduction.—Light-matter interactions may be used to dictate the organization and manipulation of colloidal and biological matter at the microscopic level. An inhomogeneous optical field permits dielectric spheres of higher refractive index than their surrounding medium to be trapped in three dimensions in the field maxima [1,2] primarily through the dipole interaction. This allows physicists, chemists, and biologists to explore a range of fundamental phenomena. This includes many seminal studies involving just one or two trapped objects, namely, thermally activated escape from a metastable state for a single particle exposed to a double well potential [3], studies of optical angular momentum, stochastic resonance [4], and various studies of colloidal behavior in external potentials. Furthermore, hydrodynamic interactions between two particles have been shown to display interesting correlation behavior in their positions [5].

An emergent but poorly understood phenomena is that the interaction of light and matter may lead to self-organization of particles into arrays and create analogues to atomic systems [6,7]. This is termed "optically bound matter." Such self-organized systems are likely to have impact across the biological and colloidal sciences and, indeed, may possibly permit large scale self-assembly in up to three dimensions. In particular, the two-particle interactions between an object and its nearest neighbor create a self-consistent and homogeneous solution that allows an optical geometry to, in principle, create a colloidal array ranging for a few to several hundred objects.

Golovchenko and co-workers [6,7] investigated systems where the interaction of coherently induced dipole moments of the spheres were said to interact to bind matter. In addition, optical organization through interactions of optical scattering in the beam propagation direction has been recently observed and allows interactions between microparticles separated by distances and order of magnitude larger than their individual diameters [8–10]. This latter form of optical binding offers the promise for new studies of hydrodynamic interactions and, indeed, of large scale organization of matter but is presently not well understood. A detailed study of the two-particle optically bound system would offer a new and important step to truly addressing this issue, and we believe it to be a suitable starting point for larger, more complex forms of binding.

In this Letter we present a detailed investigation of a one-dimensional optically bound system of an isolated pair of colloidal microspheres held in a dual-beam, optical-fiber trap. Careful investigation of the equilibria positions in this system reveals a hitherto unsuspected complexity: Namely, we observe bistability in the sphere separations dependent on the refractive-index difference between spheres and host medium, and hysteresis in the trap equilibrium separations as the fiber separation is varied. These observations match well with numerical solutions based on the coupled equations for the light propagation and the forces acting on the spheres, where an expression for the optical forces is employed that is derived from the Lorentz force formula.

Bistability and bifurcation are ubiquitous in several physical and biological systems and are closely linked with the concept of feedback. In the optical domain bistability is usually linked with the notion of nonlinearity but one can observe classical bistability with no explicit nonlinearity, for example, the radiation pressure from the intracavity field on a moving mirror [11]. Competition between parameters such as dispersion and nonlinearity in a wide variety of physical systems can ultimately lead to the coexistence of several stable solutions that may each be energetically favorable. Our observations for a two-particle optically bound system constitute the first realization of bifurcation and bistability that is inherently linked with the coupled nature of the problem and the direct interplay between radiation pressure and the light redistribution by each constituent microsphere with accompanying positive feedback. This results in a novel bistable optically trapped system showing bifurcation of the equilibrium separations and hysteresis of the interparticle separation when we adiabatically vary the fiber separation.

2. Coupled systems model.—Our model comprises two laser fields of wavelength λ counterpropagating (CP) along the z axis that interact with a pair of transparent dielectric spheres of refractive index n_s, diameter d, with centers at longitudinal positions z_j, $j = 1, 2$, and that are immersed in a host medium of refractive index n_h. The CP fields originate from two fibers (see Fig. 1) aligned along the z axis and with ends located at $z = -D_f/2$ for the forward field and $z = D_f/2$ for the backward field, with D_f being the fiber spacing, and the output fields being modeled as identical collimated Gaussians of spot size w_0 and power P_0. By virtue of the symmetry of the applied laser fields, we seek a solution for the configuration of the two spheres, which is also symmetric around $z = 0$, with $z_2 = -z_1 = D/2 > 0$, D being the sphere separation. The spatial evolution of the CP fields for a given configuration of the two spheres is modeled using the paraxial wave theory described in Ref. [9]. The paraxial theory, which fully incorporates the focusing effect of the spheres on the fields, is valid for sphere diameters and Gaussian spot sizes larger than an optical wavelength, meaning that the present theory is specialized to Mie scatterers.

In the next step the CP fields for a given configuration of the two spheres are in turn used to calculate the optical force F_z acting along the z axis on each sphere using the Lorentz force based approach of Ref. [12], assuming that the two CP beams are mutually incoherent, thereby neglecting any interference between them. A detailed account of this force calculation will be given in a forthcoming publication. For each CP field the calculated optical force acts along the direction of that field. Optical binding arises from the fact that the force acting on a given sphere is composed of two components along the z axis, one from the laser field whose beam waist is closest to the given sphere and a second oppositely directed force arising from the CP laser field that is partly refocused onto the given sphere by the other sphere. Balancing of these two forces by the refocusing of the spheres provides an intuitive explanation of optical binding [8].

We have numerically solved the coupled equations for the CP fields and the optical forces acting on the spheres to find the equilibrium sphere spacings where the net force F_z acting on each sphere is zero. An equilibrium spacing is stable when $dF_z/dD < 0$ at the zero crossing. In a previous paper we employed a simplified approach to calculating the equilibrium sphere positions [9] in which the force was assumed proportional to the laser intensity via a scattering coefficient. Although this approximate model gave qualitative agreement with previous experiments, it fails to account for the observed bistability in optical binding. Finally, we note that in our model we assume that the spheres remain well confined in the plane transverse to the z axis by virtue of the optical forces provided by the transverse structure of the CP fields, so that the sphere motion is confined to the z axis. With this caveat the equilibrium spacings do not depend on the laser power, which agrees with our experimental observations and previous studies [10].

The dashed lines in Figs. 2(a) and 3 show the numerically predicted sphere equilibrium separations. For a laser wavelength of 1070 nm the fiber mode spot size was chosen as $w_0 = 3.4$ μm, and $d = 3$ μm diameter spheres were used with refractive index $n_s = 1.41$. In the experiment the host index was varied (see below), giving a controllable index mismatch $\Delta n = n_s - n_h$ between the spheres and host medium. The dotted line in Fig. 2(a) shows that for a fixed fiber separation $D_f = 90$ μm, when $\Delta n < 0.076$ only one equilibrium is present, whereas for $\Delta n > 0.076$ three solutions appear, the middle solution being found to be unstable. Thus, the coupled light-matter system exhibits regions of bistability, namely, two stable solutions for a given set of parameters. Physically, bistability in the optical binding is possible in the coupled system due to feedback: Changing the sphere separation alters the electromagnetic field distribution via the focusing properties of the spheres, which in turn alters the forces on the spheres. (Optically bound matter is thereby nonlinear in a manner analogous to Einstein's theory of gravity in which "matter tells space how to curve and space tells matter how to move.") Because of this feedback the forces on the spheres, viewed as a function of sphere separation, can become highly nonlinear, and give rise to bistability. We note that the bistability predicted here for two spheres is wholly distinct from the "bistable trapping" found by Lyons and Sonek for a single sphere [13]. That refers to the existence of two stable trapping positions for a *single* object in a trap rather than any interplay between two or more adjacent trapped objects. We note too that Singer and colleagues [10] observed a bistability between an optically bound state and a standard linear trapped array (with touching particles). The dashed line in Fig. 3 shows the predicted equilibrium separations as a function of fiber separation D_f for fixed index mismatch $\Delta n = 0.0924$ in the bistable region, with the negative sloped middle branch being found unstable. Here we see an upper switch point at $D_f = 120$ μm beyond which there is only one stable solution, and a lower switch point at $D_f = 65$ μm. Based on this, we expect that if the fiber separation is slowly cycled from below the lower switch point to above the upper switch point and this process reversed, the sphere separation should trace out a hysteresis loop as the sphere separation follows the local stable equilibrium.

3. Experiment.—Light at 1070 nm from a ytterbium fiber laser was split into two equal beams and coupled into two single mode fibers (measured mode field diameter at 1070 nm of 6.8 μm) to form a dual-beam fiber trap. Prior to launch within the fibers, two variable neutral density filters ensured equal CP beam intensities in the trap region with the power from each fiber variable in the range from 100 to 200 mW. Both fiber ends were mounted face to face on a glass cover slip. One fiber ($F1$) was kept stationary and the opposing ($F2$) was adjustable through a motorized micropositioning stage to vary the fiber separation (see Fig. 1). The emitted light fields were overlapped transverse to the beam propagation axis in the (x-y) plane

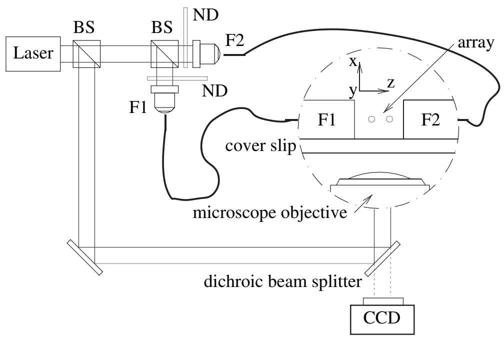

FIG. 1. Experimental setup: Laser and first beam splitter (BS) for optical tweezers coupled through a dichroic beam splitter in a 60× microscope objective. Second beam splitter (BS) for fiber coupling ($F1$ and $F2$) with neutral density filters (ND). The magnified inset shows the side view of both fibers mounted on the cover slip, $F1$ stationary and $F2$ adjustable via a micropositioner. The array is formed between the two fiber faces and observed from underneath the setup through the microscope objective and the dichroic beam splitter with a CCD camera.

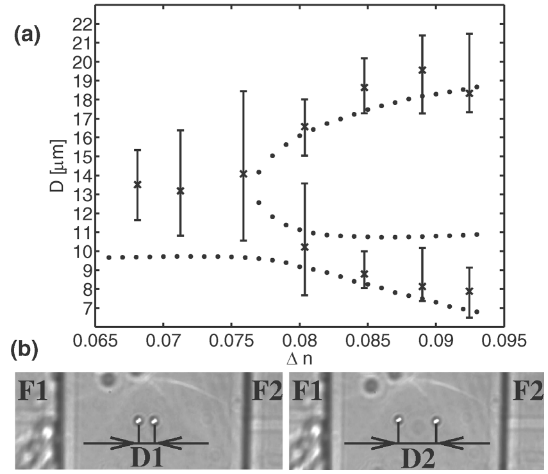

FIG. 2. (a) Plot of numerical data (dots) and experimental results (crosses with error bars) at a power of 100 mW. The crosses indicate the overall mean separation D, error bars represent the distribution of the mean values with their standard deviations (typically 12 data sets with about 3 different sphere combinations) of different measurements (typically 300) for a fiber separation of 90 ± 3 μm. For a refractive-index difference Δn from 0.095 down to 0.08 two stable positions can be observed. Between $\Delta n \sim 0.008$–0.07 the bistability of the systems ceases, resulting in a larger fluctuation in the data and in only one stable position for Δn smaller than 0.07. (b) Two stable positions with a separation $D1(\Delta n) > D2(\Delta n)$ within a dual-beam fiber trap ($F1$ and $F2$; the picture shows part of the cladding of each fiber).

and could be separated along the z axis. By carefully choosing an optical path difference well above the laser coherence length (<1 mm), the beams were rendered mutually incoherent so that standing wave effects were eliminated [10].

Silica microspheres of $d = 3$ μm (with an estimated refractive index, related to information provided by Bangs laboratories and [14], of $n = 1.41$) in a host solution were added on the cover slip between the two fibers. The refractive-index difference Δn of the host solution with respect to the sphere was successively varied in our experiment by using D_2O or a deionized water and sucrose solution, which was measured with a refractometer [10]. A key parameter in the formation of optically bound arrays is the refractive-index difference Δn between the sphere and the host medium.

A 60× microscope objective and CCD camera were used to capture images onto a computer. Using a LABVIEW program, we tracked the positions of the particles with a relative accuracy of better than ± 0.5 μm and the fiber separations with an absolute accuracy of better than ± 3 μm and extracted the experimental data presented. Additionally, optical tweezers were incorporated through the observation microscope in order to load the fiber trap and initialize the sphere positions, and were then turned off.

In the first experiments the stable equilibrium of two 3 μm silica spheres were determined experimentally for a fixed fiber separation of 90 ± 3 μm and varying refractive-index mismatch Δn. Figure 2(b) shows an example observation of the two fibers $F1$ and $F2$ at the same spacing with two distinct sphere spacings $D1$ and $D2$. The overall mean values of the sphere separation taken over on average 12 data sets of about 300 measurements each with typically 3 different sphere pairs are shown as crosses in Fig. 2(a), and the associated spread in the measurements are indicated by vertical bars. We see that there is good overall agreement between the experimental data and the stable equilibria obtained from the theory, and, in particular, we see that for $\Delta n > 0.075$ bistability is evident in the experiment. Furthermore, as expected, the fluctuations are largest closest to the critical point where the new solutions appear. It is clearly seen that the deviation between theory and experiment is largest for smaller index mismatches Δn. This is understood by realizing that as the index mismatch decreases, the net optical forces acting on the spheres get smaller, so the equilibria are created by cancellation of ever smaller forces due to the CP fields. In this situation the numerical equilibria become more and more sensitive to the precise material parameters, whereas for larger index mismatches the equilibria are more robust against the precise parameters. Nevertheless, there is very good correlation between the numerics and experiment and excellent evidence for bistability. We note that the detailed structure of the bistability depends on the parameter values. For example, for a fiber separation of 100 μm an upper branch persists and the new solutions have equilibrium spacings less than the upper branch. This inverted case compared to Fig. 2(a) has also been seen and shows also bistability in good agreement with the coupled systems model.

In a second experiment we investigated the hysteresis predicted in the numerical results (dots) in Fig. 3. For this experiment the separation between the fiber ends was

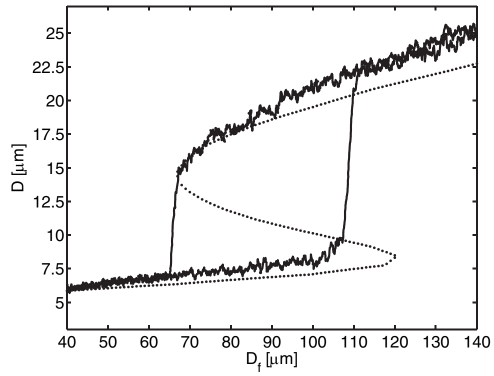

FIG. 3. Parametric plot of the equilibrium sphere separation D with varying fiber separation D_f showing clear hysteresis. The dots are the results of the numerical simulations, and the solid line is the experiment.

varied from 140 to 40 μm and back to 140 μm. If the fiber spacing is changed too quickly, the hysteresis loop washes out and eventually vanishes. For a relatively slow velocity, 1 μm/sec, the spheres were found to adiabatically follow the changing fiber separation in the system.

The solid line in Fig. 3 shows the sphere separation plotted parametrically as a function of fiber separation as the fiber separation is slowly cycled, and there is excellent overall agreement between the numerics and experiment. The agreement is best for smaller fiber separations but dwindles for larger separations. The explanation for this is that for large fiber separations the optical forces acting on the spheres are getting ever smaller, meaning that vibrations and imperfections in the system, and not included in the model, can play a bigger role and deviations between theory and experiment are not unexpected. To put this in context, the Rayleigh range for the light fields emitted by the fibers is around 30 μm so that for a fiber separation of 100 μm the fields are considerably reduced compared to the input. Thus, at the upper switching point we see that the system switches early, which can be attributed to enhanced sensitivity to external perturbations and noise around the switching point. In particular, the data are clearly noisier on the upper branch for fiber separations between $D_f = 110$–140 μm in comparison to the lower branch for fiber separations between $D_f = 40$–60 μm. We note that the detailed numerical hysteresis loop is sensitive to the parameters used, in particular, the upper switching point can change by many microns with a small change in refractive index. This is also reflected in our experimental observations, where the hysteresis loop for different pairs of spheres is sensitive to the nominal size and refractive-index differences within one batch. Nonetheless, this experiment clearly demonstrates that hysteresis can occur in optically bound systems.

Our model predicts that the sphere equilibria should be independent of power, and we do observe this in our experiments over a range of 60 to 200 mW. At the lowest powers the system is far noisier, as mechanical vibrations, evaporative flow in our open sample cell, and other external perturbations are more able to induce a premature transition between the stable branches, or even (at large fiber separations) a total loss of particles from the trap. For this reason, the experiment shown in Fig. 3 was performed at the relatively high power of 200 mW, allowing us access to data at very large fiber separations.

4. Conclusion.—In this Letter we have reported observations and simulations of bistability in an optically bound array. In particular, using a dual fiber trap, we mapped out the region of bistable solutions as a function of the refractive-index difference between the spheres and the host solution. By slowly cycling the fiber separation, we showed that the system exhibits hysteresis as the array adiabatically follows the local equilibrium separation. Our simulations based on the spheres coupled to the light field were shown to yield good agreement with the experimental results.

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