

## Phase-Transition-like Properties of Double-Beam Optical Tweezers

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We report on double-beam optical tweezers that undergo previously unknown phase-transition-like behavior resulting in the formation of more optical traps than the number of beams used to create them. We classify the optical force fields which produce multiple traps for a double-beam system including the critical behavior. This effect is demonstrated experimentally in orthogonally polarized (noninterfering) dual-beam optical tweezers for a silica particle of  $2.32\ \mu\text{m}$  diameter. Phase transitions of multiple beam trapping systems have implications for hopping rates between traps and detection of forces between biomolecules using dual-beam optical tweezers. It is an example of a novel dynamic system with multiple states where force fields undergo a series of sign inversions as a function of parameters such as size and beam separation.

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Since their inception, it has been known that optical tweezers have nonconservative force components in their force fields; this is commonly expressed by the division of the optical force into the gradient force (conservative) and the scattering force (nonconservative) [1]. When single Gaussian beam traps are used to measure forces, for example, in biophysics [2], the trap is usually considered to be a harmonic potential. That is, the gradient force is assumed to be described by Hooke's law, and the scattering force is ignored. In this case, the trap can be described by a single parameter, the trap stiffness  $k$ . This approximation is useful as it provides quantitative results of sufficient accuracy, especially when considering other sources of uncertainty in the system. While a more complete picture of the optical force field is possible [3,4], this is not required for such applications. However, with two trapping beams, the approximation of any single linear response function fails and nonequilibrium effects must be considered [5].

While in general the optical force field for a double-beam trap cannot be modeled as a linear spring, or as a pair of linear springs at the limits of small and large separations of the two beams, such models can be used for small displacements of trapped particles. For small separations there should be one trap, which can be modeled as harmonic, and for large separations, there are two noninteracting traps. Thus, at some intermediate separation of the beams, there must be a transition from the single trap case to the multiple traps case; this would constitute phase-transition-like behavior. Here, we investigate the transition from a single trap to multiple traps, using an accurate electromagnetic computational model [6], and demonstrate experimentally a particular case in a dual-beam optical tweezers apparatus.

We assume that our two beams are completely uncorrelated, which could be experimentally obtained by using beams from independent sources, and can be approximated by using beams with orthogonal polarizations. Previously,

unusual behaviors of dual-beam optical tweezers may have been misattributed to an interference between the two beams. The experiment greatly reduces interference by using orthogonal polarization.

We calculate [6] single-beam force fields and add them with a varying lateral displacement to obtain the total optical force field. We assume the beams are uncorrelated, but otherwise identically linearly polarized in a high numerical aperture ( $\text{NA} = 1.25, 1.3$ ) aberration free optical tweezers, trapping silica ( $n_p = 1.45$ ) and polystyrene ( $n_p = 1.59$ ) particles with radii of  $0\text{--}2.66\lambda_{\text{med}}$  in water, where  $\lambda_{\text{med}}$  is the wavelength units in the medium (water).

We show two examples of such force fields in Fig. 1, using a line integral convolution with white noise [7]. In the case of a  $0.66\lambda_{\text{med}}$  radius particle shown in Fig. 1(a), beam separation has resulted in the formation of two traps. Between the two equilibrium points, the particle is still axially trapped, and the potential barrier between the traps is smaller than the energy required to escape completely. Therefore, it should be possible to observe hopping between the two traps, driven by Brownian motion.

Different size regimes result in manifestly different behavior. For larger particles, such as the  $2\lambda_{\text{med}}$  particle shown in Fig. 1(b), the force fields from a single beam have a more complex spatial variation, and the superposition of two such force fields has intricate alternating attractive and repulsive regions. In the case shown, this has resulted in *three* stable equilibrium positions. Whether or not the third equilibrium point on the beam axis is stable or unstable depends on the size and relative refractive index of the trapped particle, since the axial forces, and hence the axial equilibrium position, can vary rapidly with size and refractive index due to interference effects [3,4].

Figure 2 shows the beam separation at which multiple traps form and its dependence on particle size. The

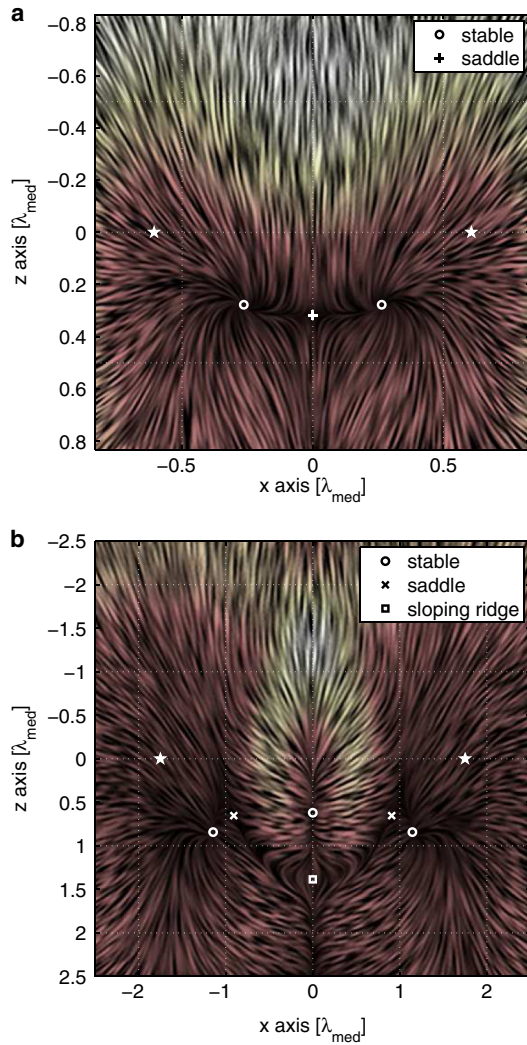


FIG. 1 (color online). Force field visualization for two particles. The centers of the double beams which produce these force fields are denoted with stars. (a) Force field of a trapped subwavelength-sized microsphere in dual-beam optical tweezers. The  $x$ - $z$  plane pictured (with the beam propagating in the  $+z$  direction, and the  $x$  axis is the separation axis) contains the expected features for a simple double Gaussian potential past a critical separation, creating two traps. The stable equilibria (circles) are the locations where the particle spends the most time. Dynamics requires a change in the sign of curvature, in the plane seen in the figure, and thus a saddle (cross) appears. (b) Force field for a sphere with radius larger than the wavelength. Simple Gaussian potentials do not describe the energy landscape the particle experiences. A third stable point and an upward sloping ridge (square) are now present.

difference between the small and large particle cases can be seen clearly. For small particles, the important factor is the separation of the beams compared to the beam width. For large particles, the separation of the beams compared to the particle size is most important. The refractive index affects the separation, since it affects the axial position at which particles are trapped, and hence the width of the beams at the trapping positions.

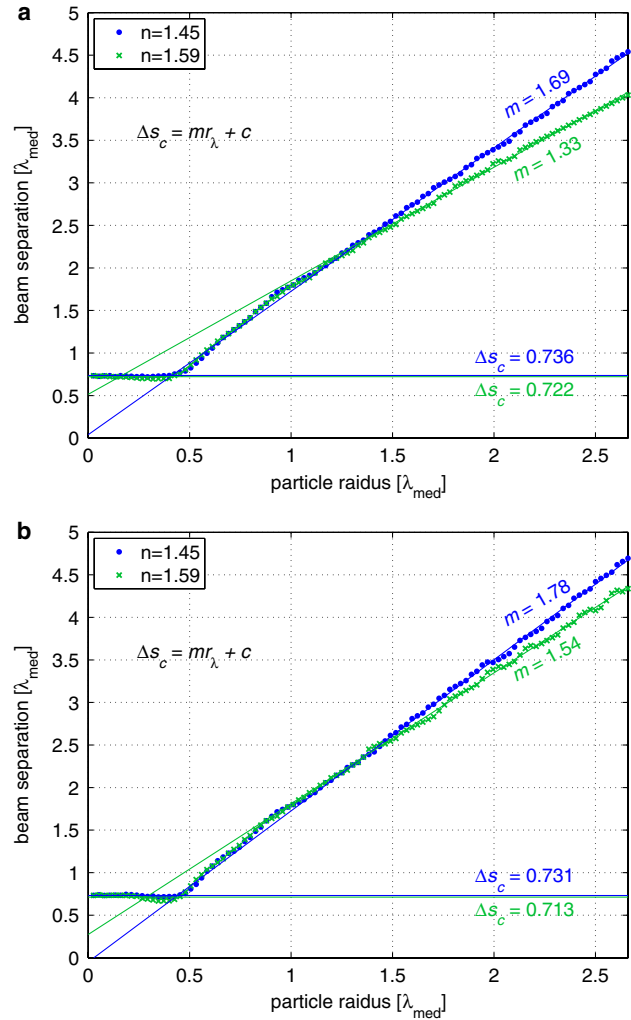


FIG. 2 (color online). (a) Critical beam separation ( $\Delta s_c$ ) required to produce two or more traps as a function of particle radius for silica (dots) and polystyrene (crosses) in  $NA = 1.25$  objective tweezers. (b) The same for a  $NA = 1.3$  objective lens. For small particles, the beam separation required to produce multiple traps is dependent only on the beam waist size. For wavelength-sized or larger particles, there is a linear dependence on the size; the best-fit gradients are shown. At larger separations the silica and polystyrene curves diverge since the polystyrene particles are trapped farther from the focus, and thus experiencing stronger influence from both beams.

Noting that three stable equilibria occur in Fig. 1(b), we investigated the number of traps that exist by calculating the transverse forces along the zero axial force contour [4]. Figure 3 shows three different trap formation behaviors. The number of traps formed varies from (a) two, (b) three, to (c) more. Only the two and three trap variations are likely to be observed; the explanation for this can be constructed with an argument demonstrated in Fig. 4. The two particles pictured correspond to Figs. 1(a) and 1(b). For a small particle, the stiffness (gradient of force with respect to displacement) is uniform near the beam axis, and decreases smoothly as the particle moves far from the beam axis. As a

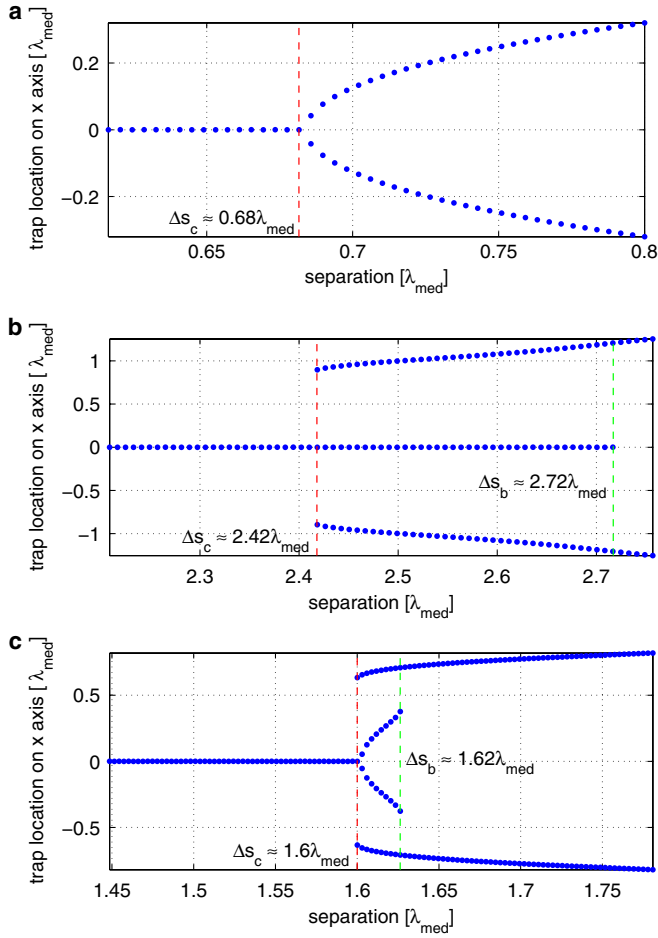


FIG. 3 (color online). Trap locations along the beam separation axis ( $x$  axis) along the zero axial force contour. (a) Small particles, smaller than a wavelength in radius form two traps. For a  $0.4\lambda$  radius polystyrene particle, a single trap changes to a double trap at a separation of  $\Delta s_c = 0.68\lambda_{med}$ . (b) Particles of radius larger than the wavelength form three traps. At the critical separation for a  $1.33\lambda_{med}$  radius particle, two new traps form some distance from the original, central, trap. The strength of the central trap reduces until  $\Delta s_b = 2.72\lambda_{med}$ , where that point becomes a saddle. (c) For particular configurations of double-beam optical tweezers many traps can form at particular particle sizes. Here, using a  $NA = 1.3$  objective lens, a  $0.88\lambda_{med}$  polystyrene particle forms four traps over a small separation range due to ripples in the radial force curve.

result, a small particle exhibits behavior where the local stiffness of the combined traps smoothly changes between being dominated by one trap to two. Larger particles typically have a stiffness which rapidly increases near the edges of the trap and sharply falls off as the particle moves even farther away. Because of this increase in stiffness, the two new traps form around the central equilibrium before the two individual traps are completely separated. The two new traps appear while the original central trap still exists, typically at a higher stiffness than the original single-beam trap.

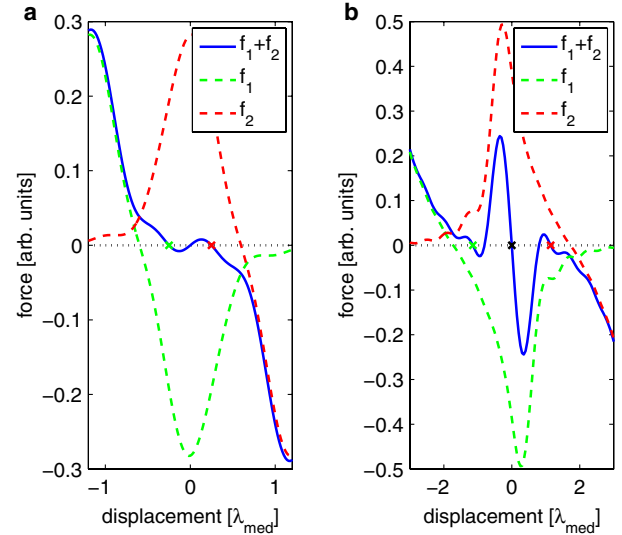


FIG. 4 (color online). Component ( $f_1, f_2$ ) and total forces ( $f_1 + f_2$ ) experienced along the separation direction at the stable trap height corresponding to Figs. 2(a) and 2(b). The resulting traps are denoted with crosses. The horizontal dotted line corresponds to zero force in the separation direction. (a) Subwavelength particles such as the one depicted here gain two traps past the critical separation. The gradient of force is fairly constant until the peak where it falls off. The falloff enables a double trap to form. (b) Larger than wavelength radius particles initially produce three traps past the critical separation. The increase in gradient enables the trap at the center to be sustained past the critical separation until the traps no longer overlap at which point it becomes a double trap system.

More than three traps around the critical separation are possible, but would be difficult to observe experimentally, for three reasons. The first reason is that the formation of more than three traps was only observed in calculations for a  $NA = 1.3$  lens and high refractive index polystyrene particles, which are only marginally trapped and can easily escape from the trap through Brownian motion. The second reason is that the depth of the traps is so low that they could be easily missed in experiments as the ensemble could be mistaken for a single, floppy, loose trap. Third, the stability of many double-beam optical tweezers is limited to a few nanometers in the focal plane due to instability in the motions of the optical apparatus.

If we are to observe the presence of multiple traps experimentally, through hopping of a particle between the traps due to Brownian motion, it is important to realize that past a certain separation either no zero axial force contour exists or the maximum axial restoring force gives a smaller barrier than between the two beams, and thus a trapped particle will be more likely to escape completely than to hop between the traps. This is especially the case for particles with refractive indices greater than 1.5. Therefore, a silica particle of  $2.32 \mu\text{m}$  was selected to be trapped. Using a 1070 nm laser source (YLM-5-LP, IPG Photonics) two beams with orthogonal linear polarization

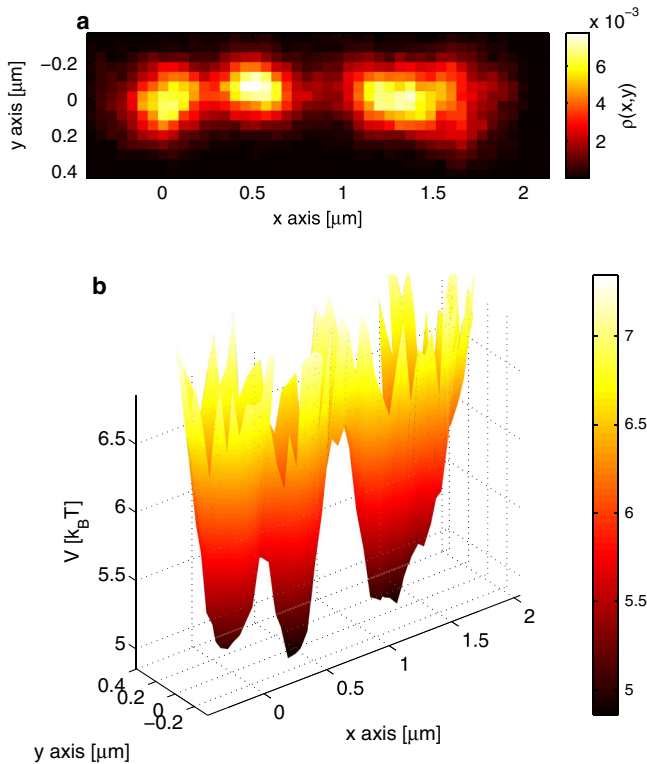


FIG. 5 (color online). (a) Histogram of normalized probability of particle position for a  $2.32 \mu\text{m}$  silica particle trapped in  $\text{NA} = 1.25$  dual-beam optical tweezers. Lighter regions denote regions of higher occupation probability. The stable trapping points for the particle are approximately the highest probability regions. The separation of the two farthest traps is about  $\Delta x = 1.35 \mu\text{m}$ . (b) The pseudopotential resulting from taking  $-\log[\rho(x, y)]$  of (a). The displacement of the two outer traps in medium wavelength units is  $1.7\lambda_{\text{med}}$ . The energy differences between the three traps is on the order of  $k_B T$ .

were focused by a  $\text{NA} = 1.25$  (PLAN 100×A/1.25, Nikon) into a sample of water containing the silica particles. By means of a gimbal mounted mirror one of the beams was moved with respect to the other [8] to a known region where three traps would form. A movable 1:1 telescope was required to ensure the beams were focused in the  $xy$  plane. The particle was observed to hop between three stable trapping locations. This was observed by means of a camera (Proscilica GE680, Allied Vision Technologies GmbH) operating at 500 frames per second. Figure 5(a) depicts the histogram of position data obtained with a center of mass algorithm similar to [9], where accurate measurements of particle positions in optical tweezers are made using high-speed video microscopy. It was determined that three traps were present using the transverse occupation probability in Maxwell's statistical distribution for distinguishable particles; a representation of the pseudopotential surface in this plane appears in Fig. 5(b).

We have shown that double-beam optical tweezers often do not display trapping behavior dictated by a simple harmonic trapping model of optical tweezers. Particles smaller than the beam waist follow this intuitive picture. Closely paralleling the intensity gradient of the light trapping the particle, two beams produce two traps at twice the beam waist. In choosing a particle larger than the wavelength in radius, we break the double well potential approximation for two beams. Three traps form at a critical separation proportional to approximately twice the sum of beam waist and radius in particles with radius about a wavelength in size and larger. It is possible but unlikely to observe even greater numbers of traps near critical separations. This has implications for experiments where dual-beam optical tweezers are used to detect forces as the addition of an extra pseudopotential between the extrema can affect force and position measurements. This could also have applications in biophysics in the situation where the bandwidth of the trap needs to be strictly controlled without increasing, or decreasing, the optical power traveling the beam path. One of the best descriptors of the behavior of double-beam traps is that of a phase transition, in much the same manner as a phase transition between states of matter. Double-beam optical tweezers display first-order transitions and a (classical) second-order phase transition. Previously only second-order phase-transition-like behavior had been observed, for example, [5]; here we report that double-beam optical tweezers display first-order transition behavior.

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