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Spontaneous symmetry breaking and circulation by optically bound microparticle chains in Gaussian beam traps

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It has been known for some time that simple “optically bound” chains of dielectric microparticles can form in a counter propagating Gaussian beam optical trap. Here we report experimental observations of more complex trapped states, which do not reflect the underlying symmetry of the optical beam trap they are confined in. We discuss both stationary off-axis trapping and dynamic motion. We confirm the results using a rigorous Mie scattering model and also give a physical explanation for these static and dynamic off-axis trapped states.

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I. INTRODUCTION

“Optically bound” [1] chains of dielectrical microparticles in a counterpropagating Gaussian beam optical trap were first reported by two independent groups [2,3], and a number of groups have since investigated the behavior of such optical traps [4–7]. In all but one of these papers the particles are confined to the beam axes, leading to a one-dimensional problem. In Ref. [7], circulating modes were reported, but these were caused by a deliberate misalignment of the trapping beams.

In this paper we report experimental observations of self-sustaining circulation within an optical trap, which is *not* caused by beam misalignment, but arises spontaneously from the physics of the optical binding interaction, despite the rotational symmetry of the perfectly aligned Gaussian beam trap. The particles move in and out of the common beam axis as they circulate, in a driven harmonic motion. We show that this type of behavior is predicted by our rigorous Mie scattering model, and we give a physical explanation for both static and dynamic off-axis trapped states.

Our experiment involves a trap formed from two orthogonally polarized counterpropagating Gaussian beams of vacuum wavelength 1064 nm, focused using a pair of 50 mm focal length lenses to a beam waist radius of around 3 μm , with the beam foci around 180 μm apart along their common axis. This is an experimental configuration which is commonly referred to as a “counterpropagating optical trap” or “dual-beam trap” [2–4,7]. A schematic diagram of the trapping region is shown in Fig. 1. This low numerical aperture configuration contrasts with single-beam high numerical aperture “optical tweezers” systems. The latter is almost exclusively used for trapping and manipulation of a single particle per beam, whereas “optical binding” of multiple particles is normally studied using low numerical aperture systems like the one we discuss here [1,8–11].

Figure 2 shows a series of frames from a video of particles circulating in this optical trap. 3.0 μm diameter silica beads are suspended in heavy water in the trapping region. It can be seen that the beads are circulating within the trap,

with a period of approximately 10 s. The images are a composite of both a backlit white-light transmission image of the particles, and direct imaging of the laser light scattered from the particles (the imaged scattered light is slightly offset relative to the white-light image due to chromatic aberrations in the optics and coherent scattering effects from the microspheres). The white-light illumination is purely to aid with the viewing of the particles, and is nowhere near intense enough to affect the interparticle interactions. It can be seen from the change in brightness of the scattered light that the particles are on the beam axis as they move to the right, but are on the edge of the Gaussian beam, where they are exposed to a weaker light intensity as they move to the left.

We have reproduced behavior similar to that shown in Fig. 2 using a model based on Mie scattering theory, as we will see later in Fig. 9, but we will begin by considering Mie scattering calculations for smaller numbers of particles in order to understand the mechanisms underlying this behavior.

II. OFF-AXIS TRAPPED STATES

We modeled a counterpropagating beam trap, similar to the one described above but containing only two particles. Figure 3(a) plots the lateral force on the first of the two particles and Fig. 3(b) the force on the second of the particles, in both cases just considering the effects of right-going of the two trapping beams, when both particles are slightly

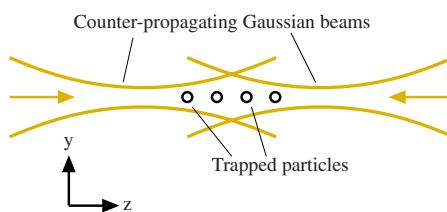


FIG. 1. (Color online) Schematic diagram showing the trapping region of a counterpropagating beam trap. Two low numerical aperture Gaussian beams are focused to points a few hundred microns apart, with a common beam axis (the z axis). Microparticles are trapped in the region between the two focal points, traditionally forming a chain along the common beam axis.

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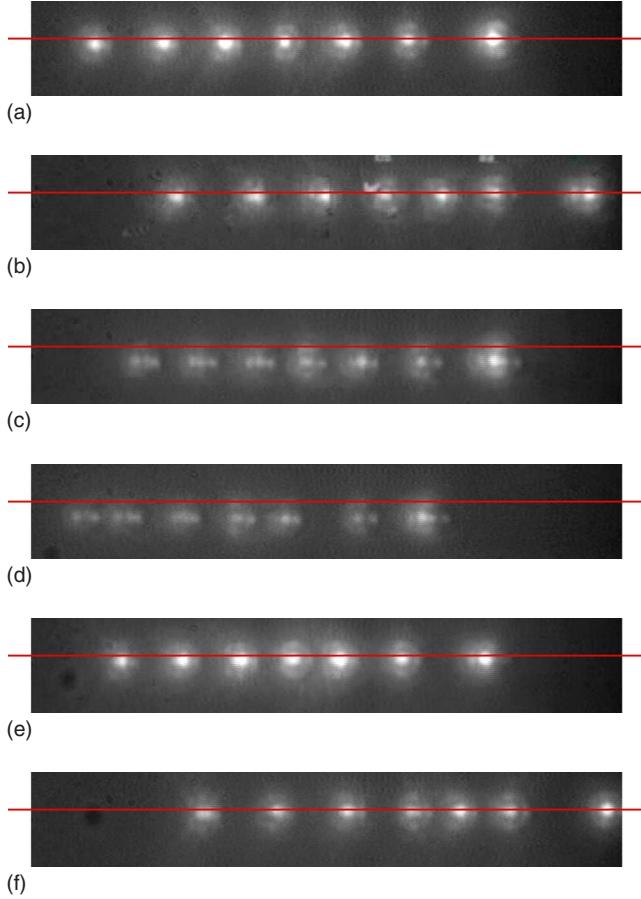


FIG. 2. (Color online) Video frames from an experiment showing off-axis circulation of particles in an aligned Gaussian beam trap, taken over a 10 s period. The horizontal line indicates the approximate position of the beam axis. Frames a–b show the chain moving right, on the beam axis, c–d show the chain moving left, approximately 4 μm below the beam axis on the screen, and e–f show the chain moving right again, on the beam axis (all six frames have the same field of view). When on the beam axis the particles are exposed to a higher beam intensity, the particles move faster and scatter the light more strongly (the directly scattered laser light in the image is slightly offset due to chromatic aberrations in the optics and coherent scattering effects from the microspheres). The position of the beam axes was determined to under 1 μm by observation of the trajectories of isolated particles exposed to a single beam at a time. Similarly, the beam alignment in the x direction (into the page) was determined to within 2 μm based on the change in focus of the particle images as a function of x coordinate (but, as implied by the results in [7], we would not expect misalignment along the x axis to lead to circulation in the yz plane anyway).

offset from the beam axis. Because, under the experimental parameters for those plots, the particles interact with the field largely through forward scattering (as discussed in [8]), the first particle behaves very similarly to how an isolated single particle would behave in the trap. Through the gradient force, it is pulled back onto the beam axis from its initial small offset. In contrast, the graph shows that the second particle is pulled *further* off axis by the focusing effects of the first particle on the field. As shown in the intensity map

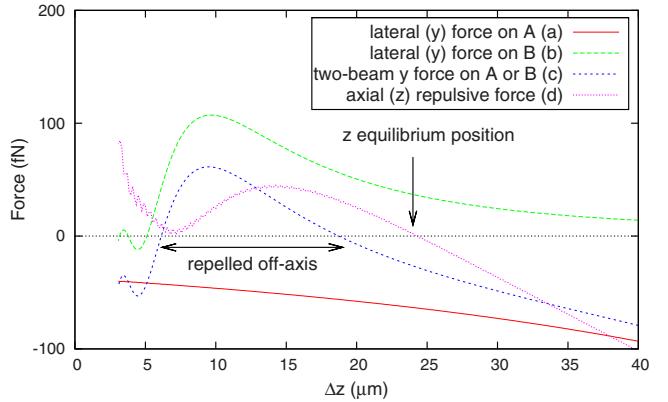


FIG. 3. (Color online) Forces acting on a pair of trapped 3 μm diameter silica particles as a function of particle spacing Δz along the z axis. The particles are very slightly offset from the beam axis (by approximately 10 nm). Positive forces represent repulsion. If we first just consider the right-going beam, particle A is drawn on axis (a), but B is pushed off axis for a large range of particle spacings (b). When the effects of both beams are considered (c), both particles are pushed off axis for z spacings between 6 and 19 μm . In this case that range does not coincide with the equilibrium particle spacing in z (d).

in Fig. 4, the “plume” of light focused by the first particle is angled slightly off-axis due to the diverging nature of the incident beam at this point and the lenslike behavior of the sphere. Through the same gradient force, this causes the second particle to be pulled further off axis.

When the effects of both counterpropagating beams are considered, there is a range of interparticle spacings, for which the lateral (y) force (which through symmetry is the same for both particles) acts to repel the particles from the beam axis. In the case of Fig. 3(c) this range is between 6 and 19 μm . For this range of interparticle spacings the particles are in a state of unstable equilibrium on the beam axis, and a small perturbation (due for example to Brownian motion) will be amplified and cause the particles to move away from the beam axis. However, Fig. 3(d) shows the z force (force parallel to the beam axes) as a function of interparticle spacing, demonstrating that in this particular scenario, *when the particles are at their equilibrium separation* in the z direction, the particles are stably trapped on the beam axis, as is conventionally the case in optical trapping, and stable off-axis trapping does not occur in this two-particle case.

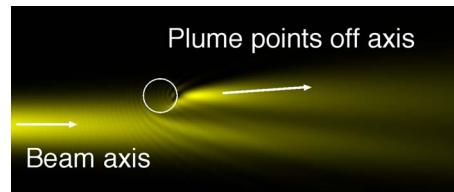


FIG. 4. (Color online) Field intensity around a single off-axis particle, showing the “plume” of light focused by the particle (in this case a 3 μm diameter silica sphere). Due to the diverging nature of the beam, this is angled slightly off-axis, and so a second particle will be drawn *even further* away from the axis through the gradient force.

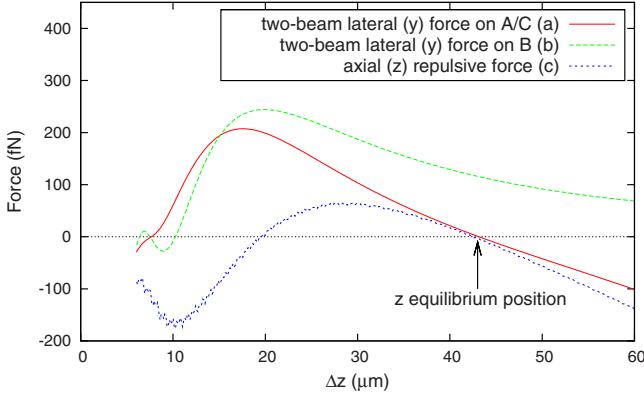


FIG. 5. (Color online) Forces acting on three trapped $3\text{ }\mu\text{m}$ diameter silica particles (labeled A, B, and C from left to right) as a function of particle spacing in z . At the equilibrium interparticle spacing (determined from curve (c)), the end particles feel very little lateral force (a), but the central particle is pushed off axis (b). The net result is that the three particle chain is pushed off axis.

This lack of coincidence between y repulsion and the equilibrium spacing in z applies over all the parameter space we have explored for two particles, and there is a reason for this. As stated earlier, the y repulsion relies on the bright “focal plume” of one particle drawing the other particle off axis. However it is this same bright focal plume, which acts to repel the particles in z , pushing them apart to a greater equilibrium spacing [8]. However, if we introduce more particles into the trap, the particles are forced closer together [2,7,8], and it is possible for the y repulsion mechanism to act at the equilibrium chain spacing. Figure 5 shows the force on a chain of three trapped particles, for which the net y force on the chain will act to push it off axis.

III. OFF-AXIS DYNAMICS

Figure 6(a) shows the trajectory of three particles which are initially positioned very close to the beam axis. It can be seen that the particles settle into a stable trapped configuration away from the beam axis (arrows represent stable particle positions). The state has not recovered from the tiny initial perturbation, but has switched from its initial unstable equilibrium on axis to a completely different state which does not reflect the symmetry of the trap.

Figure 6(b) shows a similar trajectory, but for particles with a slightly higher refractive index, which causes them to interact more strongly with the beams. The particles are pushed off axis as before, but instead of settling into an entirely stationary condition, their trajectory stabilizes into a closed orbit. Such *limit cycle* behavior was previously reported by Ng *et al.* [9] for clusters of particles trapped in the plane perpendicular to coherent counterpropagating plane waves, but in our experiment the motion is in a plane parallel to the beam, and does not reflect the symmetry of the trap. We emphasize that the particles are in a heavily overdamped regime in which free harmonic oscillation cannot be supported. It is the continual input of energy into the system by the trapping beams which drive this harmonic motion.

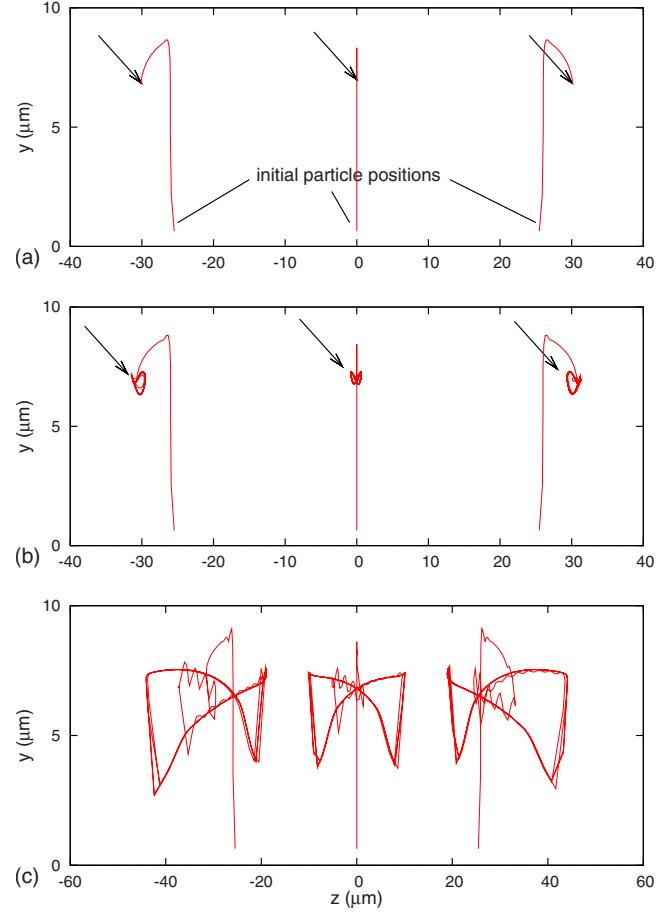


FIG. 6. (Color online) Simulated trajectories of three $1.9\text{ }\mu\text{m}$ diameter silica particles in the trap (refractive index of surrounding medium is 1.35). The particles start close to the beam axis at the start of the simulation. In the top plot, they are rapidly pushed off-axis and then stabilize at the positions marked by the arrows. The particles are stationary, but are positioned away from the beam axis rather than being trapped on axis in accordance with the symmetry of the trap. In the middle plot, where the particles have a slightly larger refractive index, the particles attain similar positions but are not completely stationary. They follow small oscillatory trajectories. In the bottom plot, where the refractive indices are still higher, the particles eventually stabilize into wide-ranging closed trajectories in which they circulate around the trap indefinitely. If alternatively the particles are positioned at a small negative y coordinate, instead of a small positive one as shown in these figures, then the resultant trajectories are, as expected, identical mirror images of those shown here, reflected in the z axis.

As the refractive index is increased still further, the scale of the limit cycles grows, until a macroscopic circulation within the trap develops [as shown in Fig. 6(c)]. The particles circulate in a figure-of-eight pattern around the trap. This motion cannot be described in terms of particles moving subject to a single conservative potential—indeed in the overdamped case, sustained motion cannot occur in such a model—but we can describe the competing effects which give rise to the macroscopic circulation. A general feature of the motion is that movement in the y direction, perpendicular to the beam axis, is much more rapid than movement in the

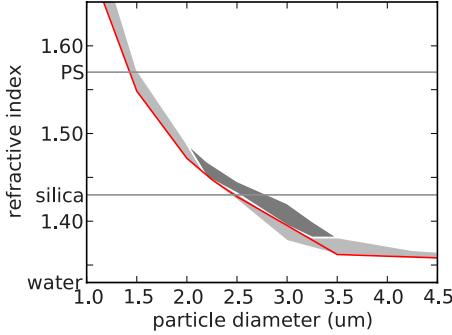


FIG. 7. (Color online) Simulation results showing the range of parameters for which off-axis trapping is seen for three particles. The lightly shaded region indicates stationary off-axis trapping. For all particle sizes investigated (between 1 and 5 μm in diameter) it is possible to see off-axis trapping if the refractive index is selected appropriately. In the range of diameters 2 to 3.5 μm , dynamic off-axis states are seen (the heavily shaded region). Below the red line, stationary on-axis trapping is supported, and it can be seen that there is a region of bistability where both on- and off-axis stationary trappings are supported (the red line overlaps with the lightly shaded region). Refractive indices for water, silica, and polystyrene (PS) are marked for reference. The data in this plot was generated for a beam waist diameter of 5 μm , and beam foci 180 μm apart. The results shown ignore Brownian motion; for sufficiently weak beam powers its effects will be to reduce the extent of the various trapping regimes.

z direction, parallel to the beam axis. Broadly speaking, this is because there is a strong gradient to the laser field in the y direction, leading to strong gradient forces in that direction. The net forces in the z direction are weaker because the forces due to each of the two beams act in opposing directions, and it is only the *difference* between those forces (of similar magnitude to each other) that leads to motion of the particles in the z direction (see [8] for further discussion).

If the particles begin in the center of the trap, on the beam axis, then as we have seen they will be repelled from the beam axis (in response to a small initial perturbation from the unstable on-axis position, caused by Brownian motion), stabilizing at some radius away from the axis. For sufficiently large particle size and refractive index, though, this configuration is in turn unstable in z . If one of the end particles is perturbed slightly away from the center of the chain, that motion will be amplified, and the center of mass of the chain will move in the direction of that initial perturbation. Thus the chain moves away from the $z=0$ position. This motion continues until such a point as the interparticle spacings and the position of the particles relative to the beam waists has changed enough that our analysis of Fig. 5, which assumes the center of mass of the particles is at $z=0$, no longer applies, and the particles are no longer repelled from the beam axis. This means that the particles are rapidly pulled back on axis by the gradient force, at which point they are pushed back toward the center of the trap ($z=0$) by the imbalance of the radiation pressure experienced from the two beams. The process then repeats and the cycle continues.

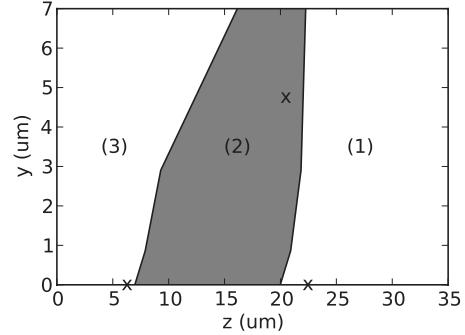


FIG. 8. Simulation results showing what happens when three equally spaced particles are placed at positions $(y, -z)$, $(y, 0)$ and (y, z) and then allowed to move under the effects of the trapping beams. The particles are 3 μm in diameter and the refractive index ratio between the particles and the surrounding medium is approximately 1.044 (which can be achieved by immersing silica particles in a weak sucrose solution). The beam parameters are the same as in Fig. 7. Starting positions in region (1) end up in the on-axis trapping configuration indicated by a cross ($z=22.4 \mu\text{m}$). Starting positions in the shaded region (2) end up in the stationary off-axis trapping configuration indicated by a cross ($y=4.7 \mu\text{m}$, $z=20.4 \mu\text{m}$). The system in fact supports a second, closer on-axis trapped state, which particles starting in region (3) will end up in. If alternatively the refractive index ratio is increased to 1.05 then, over the *whole range* of initial positions shown in the plot, the particles will end up in a circulating off-axis trapped configuration.

IV. DISCUSSION

Figure 7 shows simulated results giving the range of particle diameters and refractive indices for which on- and off-axis trappings are possible for three particles. That plot does not indicate how probable it is that these states will be produced by the natural “wandering” of particles into the trapping region one by one. Figure 8 gives an indication of the stability of a stationary off-axis trapped state, showing the range of starting positions which end up in the off-axis state.

In the results we have shown up to now the circulation has been shown in the yz plane, but it might appear that there is no preferred orientation of the plane of circulation (for example circulation could equally well occur in the xz plane depending on the initial conditions). In a real experiment a preferred direction tends to be introduced by residual convection currents in the surrounding liquid. It is for this reason that the circulation in Fig. 2 takes place in the plane of the video screen. Even in a numerical simulation, without any convective effects, it turns out that there is a slightly preferred direction introduced by the polarization of the beams: although circulation will begin in a plane determined by the initial conditions, in the absence of external influences the plane of circulation will gradually rotate over around ten periods of oscillation until the plane is perpendicular to the polarization of the beams.

The description we have given in the previous paragraphs explains the physical mechanisms behind the circulating modes in the trap, where for ease of understanding we have chosen the simplest case of three trapped particles. We note that parameters such as the particle properties and the beam

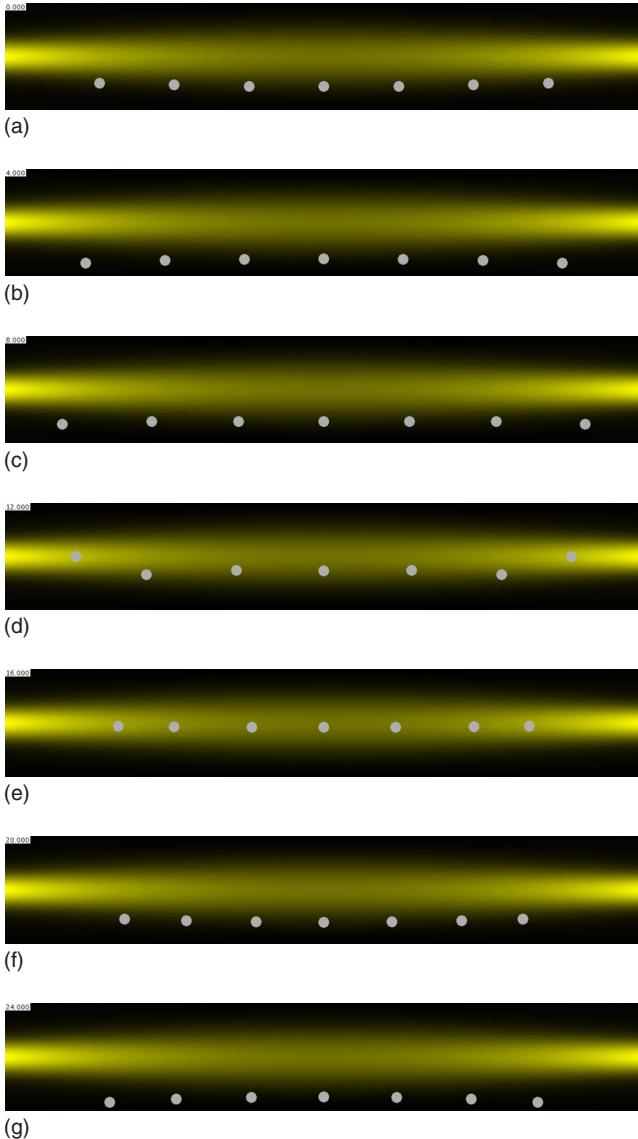


FIG. 9. (Color online) Simulated video frames at four second intervals (for a beam power of 25 mW) showing off-axis circulation with a larger number (seven) of $1.9 \mu\text{m}$ polystyrene beads. (a)–(c) show the chain expanding off axis, (d) and (e) show the chain moving on axis again and being compressed, and (f) and (g) show the chain pushed back off axis. In addition to showing that circulation occurs in simulations with large numbers of particles there are two interesting points about this example. First, the motion is symmetrical about the plane $z=0$ (contrary to the case discussed earlier, this chain is stable against perturbations in z). Second, in this case the mode is not completely stable, and after a few periods of circulation the chain collapses until the particles are touching (which is an effect often observed in experiments as well). Behavior such as that shown in this simulation may be behind the “breathing” modes reported in [2].

waist size were carefully selected to display this symmetry breaking (repulsion from the beam axis) in a short three-particle chain. For the majority of parameters, the effect is not strong enough to repel the three particles from the beam axis. However, as larger numbers of particles join the chain,

the repulsion is enhanced because the particles are pushed closer together as more of them are confined within the fixed trapping region (see [2,12] for experimental results and [8] for a theoretical discussion of this effect), and a critical point will be reached at which the symmetry of the trapped state can be broken. In the case of our experiment this occurred for chains of between seven and ten particles (as illustrated in the video frames shown in Fig. 2).

Finally, to demonstrate that our theoretical model can be extended to larger numbers of particles, Fig. 9 shows a simulation of seven trapped polystyrene particles exhibiting circulation around the trap. As described in the figure caption, this simulation shows some interesting features which are very reminiscent of the experimental results reported in [2]. In the experiments we have performed (an example of which was shown in Fig. 2), it is difficult to be certain whether we are observing truly stable circulation, such as that simulated in Fig. 6, or unstable circulation, such as that simulated in Fig. 9. The circulatory modes we observe tend to either collapse, or eject some particles from the trap, within a few periods of oscillation, but it is not possible to determine for certain whether this is due to external perturbations to the system or due to inherent instability in the observed modes of oscillation.

V. CONCLUSIONS

We have shown experimental and theoretical examples of spontaneous symmetry breaking in a Gaussian beam trap, leading to asymmetric circulatory motion within the trap. We have shown that this is predicted by a Mie scattering model, and we have explained the mechanisms behind this behavior in simple physical terms. Critical to this effect is the concept that the presence of trapped microspheres within the trap strongly modifies the electromagnetic field within the trap, such that the evolution of the system is governed predominately by the light-mediated interparticle interactions rather than the background trapping potential. Our discussion has focused on a low numerical aperture configuration, which is standard in optical binding experiments, but there is potential for further investigation into whether these effects have an impact in high numerical aperture configurations with multiple trapped particles, such as that discussed in [13]. We have demonstrated that off-axis trapping can occur with as few as three particles, and this shows that, even with small numbers of trapped particles, the full interparticle interactions must be considered in order to be able to correctly predict the behavior of the system. This dramatic change in character as more particles are added to the system may for example have significant implications for microassembly applications using optical tweezers.

ACKNOWLEDGMENTS

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