Longitudinal Optical Binding of High Optical Contrast Microdroplets in Air

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Binding along the beam axis (which we shall call ''longitudinal optical binding'') has been observed between micron-sized oil droplets in a three dimensional optical trap in air. We argue that it is the high optical contrast which is responsible for the exceptionally stable doublet structures observed experimentally. It was also observed that optically bound doublets tend to cling to interference fringes created by the two counterpropagating beams. Our observations are qualitatively discussed in the context of both the ray model (optics) approximation, and in the Rayleigh (dipolar) range. Our observations were consistent with calculations of binding and trapping forces which we carried out by employing an exact multiple-scattering theory.

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Since the pioneering work of Ashkin [1], optical trapping and manipulation of single microparticles has become a commonplace experimental tool. Moreover, in multiple-particle traps, optical interactions between scattering microspheres have been repeatedly demonstrated and have begun to find applications in their own right [2–4]. These optically induced interactions, (commonly known as ''binding forces'') can be attractive or repulsive, and indicate intriguing perspectives in multiple-particle manipulations and spontaneous optical organization [2].

Optical trapping of mesoscopic particles has predominately been studied for particles suspended in liquids. Manipulation of micron-sized particles in rarefied media is notoriously more difficult due to the existence of strong van der Waals sticking forces, and viscosities approximately a thousand times smaller in air than those present in liquids. Nevertheless, air [1,5,6] and vacuum [5,7,8] trapping and levitation experiments have been performed and exhibited strong single-particle trapping efficiencies. The two main solutions to overcome van der Waals forces are, usually, the use of aerosols like oil [8] or water droplets [1,9] and, for solid particles, strongly focused high power laser beams [5,6] and ultrasonic vibrations to free the particles from the solid surfaces on which they tend to cling.

Although optical binding studies in liquid media offer numerous advantages such as heavy damping, the weak optical contrasts between the trapped particles and the liquid in the experimental configurations studied so far tends to weaken these interactions and may limit their applications. In more rarefied media such as air with less damping, the high optical index contrasts can induce higher trapping efficiencies as well allow for a better confinement of whispering gallery modes [10,11] which can, in turn, induce nonlinear scattering studies such as lasing [12] and stimulated Raman spectroscopy [9]. The possibility of creating stable optically bound clusters of particles in air could open up new domains of study in the aforementioned phenomena, as well as introducing others such as the formation of secondary optical traps for nanoscale or molecular particles [13].

In this Letter, we report on the apparently first experimental and theoretical studies of strong longitudinal binding (i.e., binding along the direction of the beam axis) in two and three droplet systems of oil microdroplets in air (see Figs. 1 and 2). These droplets are much smaller than those previously studied in air or vacuum (from 1 to 2.5 μm in diameter), and considerably smaller than those

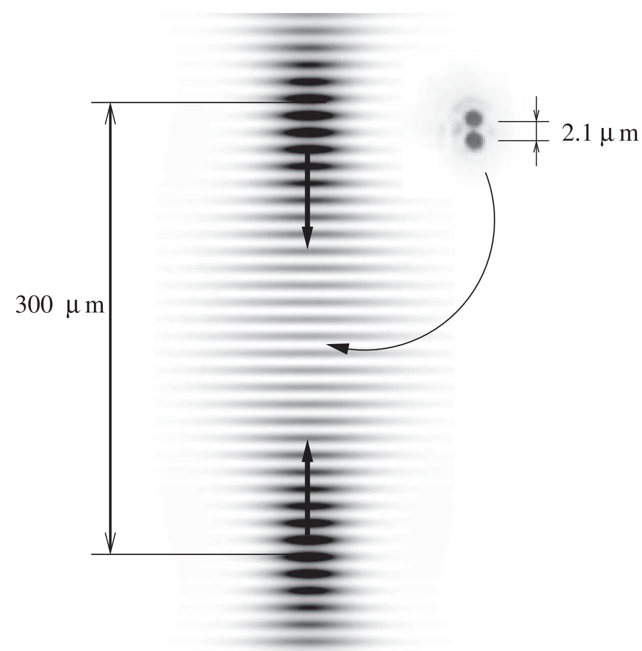

FIG. 1. Principle of the experimental setup for coherent and counterpropagating circularly polarized laser beams. The screen shot of the oil-droplet doublet (most stable observed structure from far) is out of scale.

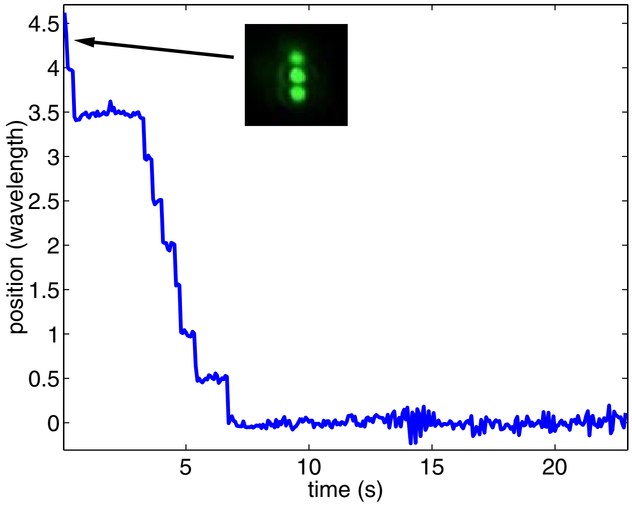

FIG. 2 (color online). Mean position (in units of λ) of a trapped doublet returning to its equilibrium position, taken for reference as zero, after the smaller third droplet (on top) escaped. The droplets are roughly 2.1 μm apart from one another.

studied in simulations of trapping and/or binding before now [11]. We have found that they exhibit behavior [14] quite different from that previously observed in water [4,15,16]. We also report on our preliminary exact multipolar calculations invoking the Maxwell stress tensor in the resonant or "Mie" regime [17] which corroborate our observations.

The experimental configurations for observing longitudinal binding in both air and liquid consist essentially of optical traps formed by two counterpropagating and diverging laser beams (see Fig. 1: not to scale). In our air experiment, the laser beam we use is circularly polarized ($\lambda = 532$ nm, $P \simeq 30$ mW). The counterpropagating beam is generated by a cat's eye retroreflector. The so-built two counterpropagating beams are focused 300 μm apart with a weak numerical aperture (1/15). Trapping occurs in the region where the upward and downward beam have near equal irradiance. The trap is $\sim$20 μm wide and over 300 μm high. Trapping in the direction transverse to the beam propagation is achieved by forces associated with the intensity gradient. Trapped doublets in such an arrangement are exceptionally stable (on the order of hours) and are quite resistant to physical and optical perturbations as well.

Optical trapping of microspheres (of radius R, and refraction index n_s) imbedded in a lower index homogenous medium n_m, has been studied for a long time in the geometric optics (ray model) regime ($kR \gg 1$: $k = n_m \frac{\omega}{c}$) [18], but the particles under study here clearly fall outside of this domain. Nevertheless, one might be tempted to argue based on geometric optics intuition that the optical binding observed here may be due to each sphere acting as a lens which creates an optical trap for the other sphere. This could be a first approximate explanation for the stability of the doublet, as well as explaining why similar phenomenon are not observed in liquid media.

On the other extreme, when the radius of the spheres is such that $kR < 1$, the dipole or "Rayleigh" approximation becomes reasonable (also depending on the relative refractive index). For multiple-particle traps in which the particle separations are orthogonal to both the beam axis and the beam polarization, it has been experimentally observed and theoretically corroborated in the Rayleigh approximation that spheres experience optical potential minima every λ (wavelength in the medium) [3]. When the particle separation is along the beam axis, similar calculations indicate (longitudinal) potential wells roughly every $\lambda/2$. Both of these periodic phenomena are essentially of a wave-interference origin, and one can expect analogous behavior (in the far-field at least) even when the scattering is essentially of a resonant nature ($R \sim \lambda$).

In our experiment, the trapping beams cross a glass cell filled with a sunflower oil-droplet cloud. The glass cell protects droplets from convection currents in the room. Scattered irradiance is projected onto a video camera. The most commonly trapped structure was a doublet, being observed more frequently than even a single droplet. The observation of interference of their respective Airy rings tells us that the droplets in a doublet are phase locked despite sometimes considerable perturbations arising from Brownian motion, intensity fluctuations between the counterpropagating beams, and the speckle pattern (mainly due to the surrounding droplet cloud and dirt on the optics).

It also appears clear that the droplets are not in contact as capillary forces are much larger than optical forces. Droplets coming into physical contact would coalesce rapidly into a single droplet. Despite all of the aforementioned perturbations, trapped doublets were typically stable for hours, implying a deep optical potential barrier separating the particles.

After a doublet has been trapped, many other droplets coming from the cloud continue to enter the trap, and interact with the trapped doublet. These interactions sometimes lead to trapped triplet structures (see Fig. 2). These structures have a much more limited stability than doublets and typically finish their existence with the third droplet either collapsing onto the doublet or escaping. The newly trapped droplet of a triplet structure is always observed to be smaller than the two previously trapped particles. This is due to the fact that a stable, readily imaged, doublet is the end result of the merging of several particles initially present in the cloud. We also remark that the center to center separation distance of the doublet increases as particles grow in such a manner that the particles never touch. The increase in the doublet size finishes when the entire oil cloud in the cell has descended to the floor of the chamber. The radii of the droplets initially present in the cloud can be estimated from a Stokes model and the observed terminal velocities to lie within the range of 0.5 μm to 0.8 μm.

When a small third droplet hangs on the doublet, the equilibrium position in the trap is different from that of a doublet. Although we do not analyze this effect quantitatively here, we believe that geometric dissymmetry and optically induced interactions between the particles leads to a dissymmetry in the radiation pressure. In Fig. 2, we illustrate the time evolution of the mean position of a doublet returning to its equilibrium position. The previous structure was a bound triplet for which the third droplet finally escaped. Taking a mean of the images (which cover several pixels) allows us to determine the position of the doublet to subpixel precision. The doublet is seen to cling to fringes created by the two counterpropagating coherent beams. We conclude that the interference fringes are capable of creating shallow $\lambda/2$ traps for the doublet as a whole which lead to a measurable dwell time for the doublet with thermal energy being sufficient to make transitions to more stable minima, henceforth ensuring that the doublet as a whole attains a potential energy minimum.

Up until this point in our observations, one could imagine that the binding effect could be described purely as a lens effect in the geometric optics approximation. From time to time, we observed rapid changes in the doublet appearance. This phenomenon may be due to a collision with an nonvisible droplet coming from the descending cloud. The low resolving power of the imaging microscope objective ($0.25N.A.$) creates an interference pattern between particles' images preventing a precise measurement of the distance separation. However, the difference of distance separation could be estimated to be of the order of 200 nm. The observed phenomenon might alternatively correspond to a switching between two longitudinal optical potential wells, separated by $\approx \lambda/2$. This second hypothesis is further supported by the observations of the interference of Airy rings (see Fig. 3). We can observe that in the first picture, the Airy rings interfere in a manner as to give a dark fringe between the droplets while on the other, bright dots can be seen on the symmetry plane. This tends to confirm that in one case, the nonresolved particles are emitting in phase and in the other case, they are emitting in phase opposition. This interpretation, inspired by the Rayleigh approximation is also consistent with the rigorous calculations presented further.

FIG. 3. Two consecutive pictures of a doublet. The quick change ($<1/15$ s) is attributed to a switching between two different equilibrium positions which is corroborated by the qualitatively distinct interference patterns. Droplets are ~ 2 μm in diameter.

In our case, the radii of the spheres are comparable in size to the wavelength of the trapping radiation (typically referred to as the resonant or Mie regime), so that neither Rayleigh nor geometric optics approximations are valid, and one must invoke full electromagnetic calculations in order to obtain quantitative force predictions. An advantage of our quasiexact calculations is that they describe not only the direct wave-particle interactions, but all of the multiple-scattering effects as well. Our calculations were performed by coupling an analytic calculation of the optical force [17] with analytic multiple-scattering calculations (formulated in terms of a multiple-scattering T matrix [19,20]).

We present here results of simulations concerning initial doublet formation and stability (force simulations for the larger doublets will be presented elsewhere). We define the binding force, $F_{b,\parallel} \equiv \frac{1}{2}\hat{\mathbf{z}} \cdot (\mathbf{F}_2 - \mathbf{F}_1)$, where $\mathbf{F}_1$ and $\mathbf{F}_2$ are the optical forces on droplets 1 and 2 labeled such that $z_2 > z_1$ so that $F_{b,\parallel} < 0$ corresponds to attraction. In Fig. 4, we show the results of our calculations of $F_{b,\parallel}$ on a pair of $1.2\lambda \simeq 0.6$ μm in radius oil droplets ($n_s = 1.48$). The force is calculated as a function of particle separation, d/λ. The curve (a) corresponds to $F_{b,\parallel}$ for a single plane wave. For curves (b) and (c) the binding force is calculated when a circularly polarized plane wave is coherently reflected back onto itself. The curve (b) corresponds to the case where one particle is artificially "frozen" to be centered on one of the interference maxima, while for curve (c), one particle is frozen to be centered on an intensity minimum. The force is calculated in picoNewtons for an incident beam irradiance of 1 mW per μm^2. The beam irradiance used in the experiment being $\sim$0.3 mW μm^{-2}, forces should simply be multiplied by this factor, the doublet geometry is irradiance independent as long as optical forces dominates thermal forces.

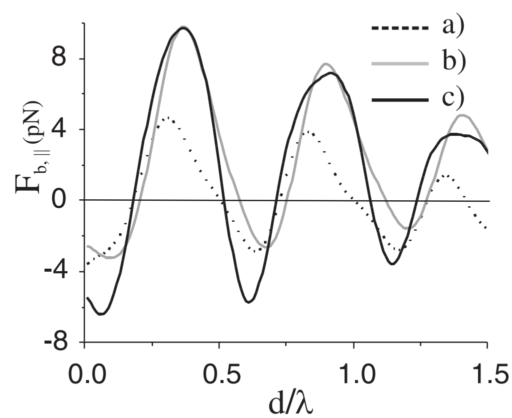

FIG. 4. Binding force: (a) $F_{b,\parallel}$ for a single plane wave, (b) $F_{b,\parallel}$ for counterpropagating circularly polarized plane waves with one particle on an interference maxima, (c) $F_{b,\parallel}$ in the same situation as for (b) except that one particle lies on an interference minima.

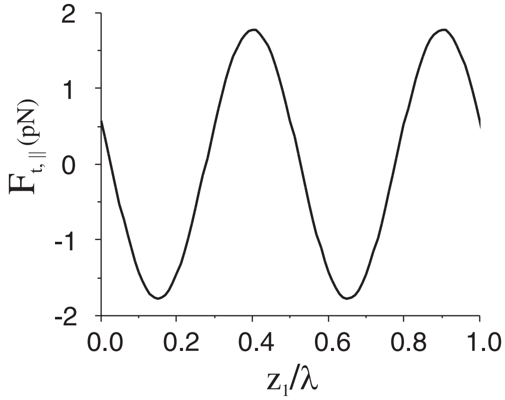

FIG. 5. Total longitudinal force, $F_{t,\|}$, on a doublet with a separation distance of $d = 0.55\lambda$ as a function of the position, z_1/λ, of the "first" particle.

An important observation of the force in Fig. 4 is the existence of a local stable equilibrium position ($F_{b,\|} = 0$ and $dF_{b,\|}/dz < 0$) which occur every $\sim\lambda/2$, even for a single plane wave, which can explain the high stability of the doublet despite strong speckle perturbations. It is also important to remark that the calculations indicate that the optical forces in this experiment are on the order of several picoNewtons and should consequently dominate other forces present (gravitational and electrostatic) on the doublet to yield rather deep optical potential wells (which are centered on the equilibrium positions) in agreement with our experimental observations. We also remark that the positions of the local equilibrium positions vary only slightly when the position of the frozen particle is moved with respect to the interference fringes.

The total longitudinal force on the doublet, $F_{t,\|} \equiv \hat{\mathbf{z}} \cdot (\mathbf{F}_1 + \mathbf{F}_2)$, is plotted in Fig. 5. Although this plot is the result of our full multipole calculations, the total force on the doublet can be qualitatively understood in terms of the "gradient" force and the intensity maxima and minima of the interference fringes. This figure indicates that the total trapping forces on a doublet originating from interference fringe effects are comparatively weak with respect to the forces associated with binding. This observation is consistent with experimental observations where the mean remaining time in fringes is much smaller than the doublet lifetime. More precise comparisons between theory and experiment could be made by modeling the longitudinal variations in the beam irradiance.

In conclusion, binding effects have been studied both experimentally and theoretically in air for particles in the resonant or Mie domain. The strong relative index contrast is the main difference between these experiments and those carried out in water. Potential minima separated by $\lambda/2$ may have been observed in longitudinal binding in analogy with the λ periodicity observed in optical binding transverse to the beam propagation [3]. It also seems likely that there is an analogy between our observations and the recent longitudinal bistability observed in liquid media experiments [21]. The strong binding behavior that we have observed in air opens opportunities to build self-structured photonic crystals or opals. It can also be a useful tool to study mode coupling between microcavities and the creation of secondary, high gradient optical traps.

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