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Multiple trapped states and angular Kramers hopping of complex dielectric shapes in a simple optical trap

J. N. WILKING[1] and T. G. MASON[1,2,3(a)]

[1] *Department of Chemistry and Biochemistry, University of California-Los Angeles - Los Angeles, CA 90095, USA*
[2] *Department of Physics and Astronomy, University of California-Los Angeles - Los Angeles, CA 90095, USA*
[3] *California NanoSystems Institute, University of California-Los Angeles - Los Angeles, CA 90095, USA*

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Abstract – Custom-shaped dielectric colloids in a simple focused laser beam create complex multi-dimensional potential landscapes. For laser powers above the threshold needed to trap spheres, many intricate shapes having holes and arms, as sampled using letters that have thicknesses comparable to the wavelength, can be trapped stably in more than one position and orientation. Particular trapped states can be reproducibly obtained by controlling how a particle enters the trap. By systematically enlarging the central hole of a square toroid, we alter the location, orientation, and existence of a trapped state. Thermal fluctuations of the trapped letter "N" cause Kramers hopping between two preferred orientational states, yielding a double-well angular potential.

Understanding the optical manipulation of complex colloidal shapes is opening up new possibilities for controlling the construction of microscale assemblies [1–3]. "Colloidal alphabet soup", a lithographically designed dispersion containing microscale versions of all 26 capital letters of the modern English alphabet in a viscous liquid [4,5], provides a significant challenge to existing means of micromanipulation. A single-beam field-gradient optical trap [6], known as "laser tweezers", represents a simple method for potentially manipulating this moveable microscopic type. A variety of microscale dielectric objects, such as spheres [6], cubes [7], rods [8], disks [9], and crosses [10] have been trapped and manipulated using laser tweezers, yet the general rules for determining whether or not a particle having a complex shape can be optically trapped, and, if so what its position and orientation would be, are not well understood.

In the absence of optical absorption, dielectric particles near the focal point of a laser beam experience forces and torques arising from photon momentum transfer [11]. Light that is backscattered from the particles creates an effective radiation pressure that pushes the particles in the average direction of propagation, $\boldsymbol{k}$, of the laser beam. However, strongly focused laser light creates very high electric field gradients in all directions around the focal point, resulting in forces that tend to pull a higher dielectric constant material into the region of highest field strength, despite the radiation pressure. Likewise, torques can arise from simple photon momentum transfer [10,12,13] and also from angular momentum transfer for optically anisotropic materials [7,9,14]. If the forces and torques generated by radiation pressure overcome the gradient forces and torques that tend to stabilize the particle in the trap, then the particle will not trap and will be ejected in the $\boldsymbol{k}$-direction away from the focal point. However, if at least one configuration can be found in which the forces and torques arising from radiation pressure and field gradients form a potential well that is significantly deeper than thermal energy $k_{\mathrm{B}}T$, then the particle can be trapped stably in three dimensions.

The sizes and shapes of microscale particles play essential roles in determining their possible stable positions and orientations in an optical trap. Simple rod-shaped particles are known to trap with their symmetry axes aligned along $\boldsymbol{k}$ [8], whereas thin disk-shaped particles and platelets are known to trap "on edge" with their symmetry axes aligned perpendicular to $\boldsymbol{k}$ [9]. In either case, the position and orientation of the particle maximize

(a)E-mail: mason@physics.ucla.edu (corresponding author)

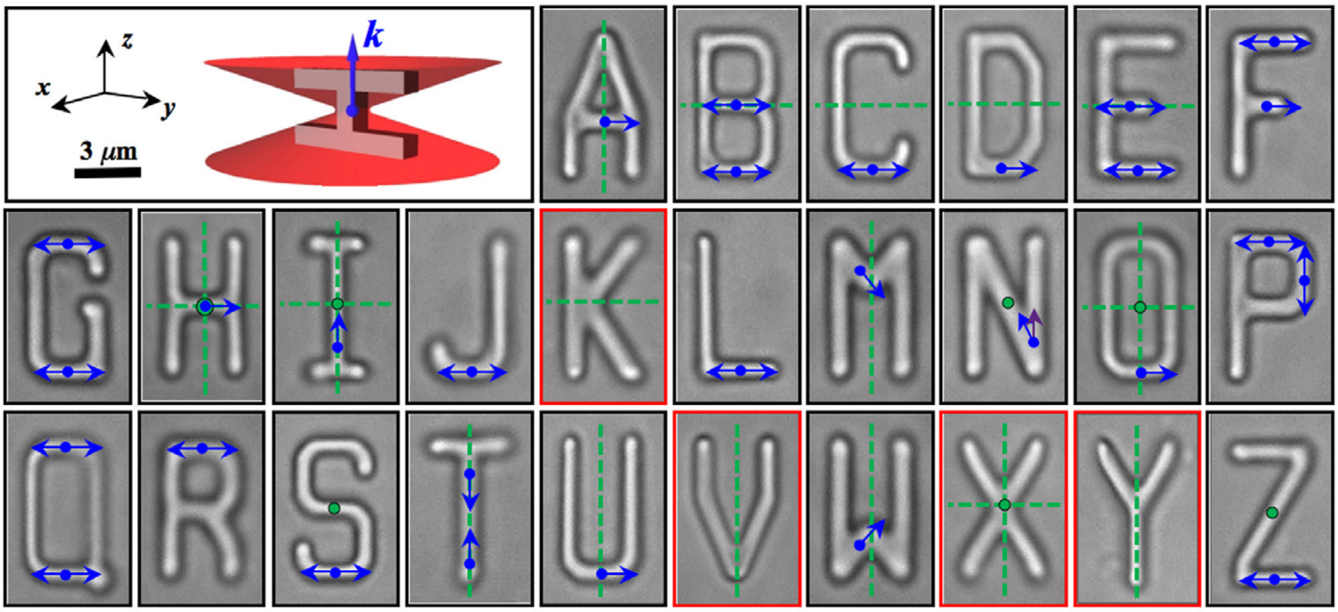

Fig. 1: (Color online) Positions and orientations of dielectric colloidal letters that are stably trapped by laser tweezers (black borders). Upper left: a schematic of a letter in a highly focused Gaussian beam. "H" traps on edge with its crossbar aligned along the optical axis. For each stably trapped state, the focal point of the optical trap is marked by a blue circle and the blue arrow indicates the direction of the average light propagation vector $\boldsymbol{k}$. Panels: measured stably trapped states of the colloidal alphabet (capital letters: $4\,\mu\text{m} \times 7\,\mu\text{m} \times 1\,\mu\text{m}$) at fixed laser power $P = 17\,\text{mW}$ in the yz-plane obtained by varying the approach of the particle to the trap. For clarity, stably trapped states of symmetric particles are shown only once; additional stable states can be inferred from a particle's mirror planes (green dashed lines) and twofold rotation axes (green circles). When trapped, the letter N alternates between two different orientations (blue and purple arrows). The letters K, V, X and Y (red borders) do not stably trap in any configuration at this P.

the highest dielectric constant material in the region of the strongest electric field, while overcoming the radiation pressure. These differences in the trapping of symmetric anisotropic objects point to a more general, beautiful, and important physical-mathematical question: what is the complete set of closed surfaces which define all three-dimensional homogeneous dielectric objects that can be stably trapped using focused single-beam laser tweezers? Beyond this, can one uniquely predict the position and orientation of such objects if they are trappable? Although limited data are available for a few cases, the general answers remain unknown. Indeed, in some ways, this physical puzzle resembles certain classical mathematical problems, such as defining the complete set of prime numbers. Although much recent activity has been centered on patterning light in more complex ways for optical micromanipulation [2,3,15–18], much still remains to be understood about the behavior of complex shapes in a simple gradient trap.

To explore these questions, we fabricate an aqueous dispersion of microscale capital letters using an ultraviolet i-line reduction stepper (Ultratech XLS 5 : 1) that cross-links regions of a thin layer of epoxy polymer photoresist, SU-8, deposited on the surface of a flat substrate [4]. As opposed to earlier methods of micromachining [13,19–21], we mass-produce high-fidelity particles with features down to $0.3\,\mu\text{m}$ without etching. The letters are highly uniform with a typical polydispersity less than 5%, have a fixed aspect ratio ($4\,\mu\text{m}$ wide, $7\,\mu\text{m}$ tall, $1\,\mu\text{m}$ thick), a fixed pen width of $1\,\mu\text{m}$, and do not contain serifs. Although the shapes are limited to only a single font of a single alphabet, they nevertheless provide a wide range of morphologies and a good means of sampling geometrical diversity. Since the epoxy polymer is clear, optical absorption at visible wavelengths is negligible. The refractive index of the crosslinked SU-8 resist is $n \approx 1.6$, higher than that of water. The laser tweezers (fig. 1) consist of an expanded TEM_{00} Gaussian beam from a linearly polarized helium-neon laser (wavelength $\lambda = 633\,\text{nm}$) that enters a high numerical aperture (NA) microscope objective lens (Nikon PlanApo CFI60 100× 1.4 NA). Very dilute volume fractions of particles ($< 10^{-5}$) ensure that only one particle interacts with the optical trap at any given time. All particles are tweezed at least $10\,\mu\text{m}$ away from the glass walls and at the laser's maximum power, $P = 17\,\text{mW}$.

We attempt to trap each letter by moving it near, but below, the focal point using the microscope's translational stage. In order to provide a significant sampling of initial particle positions and orientations relative to the beam, we have repeated this procedure over thirty times for each letter, seeking to vary the direction and angle of approach of the particle relative to the focal point (fig. 1).

Strikingly, many particles can be trapped in several different stable configurations that can be accessed by controlling the avenue of approach. The final trapped state is not random; reproducible trajectories to particular trapped states can be selected by choosing particular routes of approach. In all cases, as with other thin particles that are comparable in thickness to the wavelength of visible light [9], the letters trap "on edge" with their surface normals perpendicular to $\boldsymbol{k}$. Trapped letters tend to rotate about the z-axis until they align along the plane of polarization of the laser beam. For all letters, as P is lowered, stably trapped states become metastable and ultimately disappear as Brownian motion prevails and the particles escape.

At $P = 17\,\text{mW}$, we determine the position and orientation of each trapped configuration by observing the particle entering the trap and then moving the trapped particle toward the surface of the coverslip until it touches and is noticeably perturbed by the solid wall. When perturbed, the particle rotates and translates, or in some cases, as with the letter I, an oscillatory switchback instability can occur [22]. In all cases, the measured focal points of stably trapped states lie inside the letters.

Our observations indicate that plate-like letters having thicknesses comparable to λ, lateral dimensions larger than λ, and lateral aspect ratios of 7 : 4, usually trap with the shorter of the two lateral dimensions along the optical axis. This orientation causes most of the dielectric material to extend outside the region of significant laser intensity, reducing the radiation pressure that can lead to instability. It is more unusual for letters to orient with their longer dimensions along $\boldsymbol{k}$. Two exceptions are: 1) mirror-symmetric letters having rod-like dielectric segments running lengthwise down the middle of the particle and at least one crossbar at the top or bottom (*e.g.* I and T), and 2) letters with short dielectric segments aligned along the long lateral dimension but offset, with no additional dielectric material above or below the trapping segment (*e.g.* P). Trapping points that lie on symmetry axes are generally more stable, as deduced from the amplitude of motion driven by thermal fluctuations. For example, the letter B traps more stably along the central crossbar than along the upper or lower crossbars. Sinuous shapes, such as S, when approached from the center, cannot be stably trapped there, but instead rotate until a stable configuration at the extremities is achieved. Letters that contain holes can be trapped, but never in configurations that place the focal point outside the particle.

A few letters (K, V, X, Y), all of which have arms that do not intersect at right angles, cannot be trapped regardless of the approach trajectory and are ejected. The letter N can be trapped but alternates between two different orientations as thermal energy perturbs it. Although topologically similar to N, the letter Z has a different aspect ratio, traps in a different location relative to the focal point along a single direction. Likewise, H and I are also topologically similar, and they both trap with their crossbars along $\boldsymbol{k}$ despite their different aspect ratios.

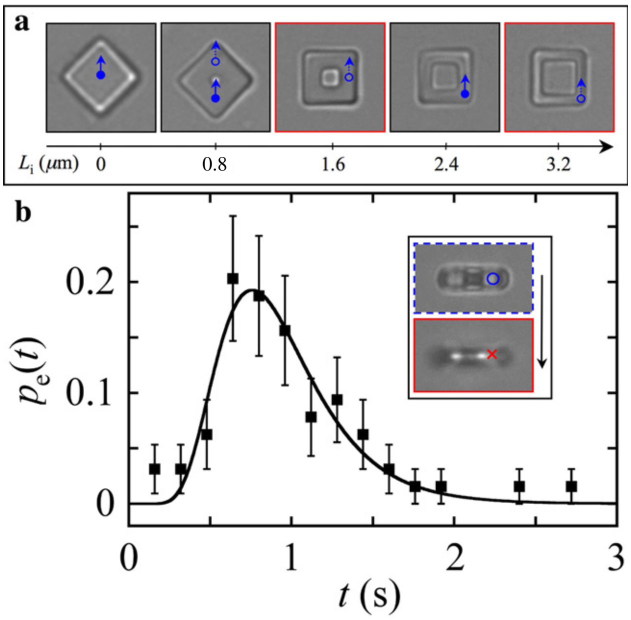

Fig. 2: (Color online) Controlling the inner edge length L_i of a square hole in a $1\,\mu$m thick square plate-like particle with fixed outer edge length $L_o = 4.5\,\mu$m greatly affects its stability in an optical trap. Stably trapped states (solid blue circles and solid arrows) and short-lived metastable states (open blue circles and dashed arrows) are shown in the yz-plane. a) $L_i = 0\,\mu$m: stably trapped with diamond-like orientation. $L_i = 0.2\,\mu$m: stably trapped with focal point below the central hole in the particle; metastable trapping point above the central hole. $L_i = 1.6\,\mu$m: unstable with short-lived metastable trapping state in a flag-like orientation (always escapes trap). $L_i = 2.4\,\mu$m: square frame stably traps in a flag-like orientation. $L_i = 3.2\,\mu$m: thin square frame cannot be permanently trapped. b) Measured distribution of escape times, $p_e(t)$, for square-frame particles ($1.6\,\mu$m hole) that are only transiently trapped. The solid line is a fit to a log-normal distribution with a zero intercept at time $t = 0$; the most probable residence time is $t_{mp} = 0.76 \pm 0.03\,$s. Inset: view (xy-plane) of a metastably trapped particle (open blue circle —upper panel) escaping the trap (red cross —lower panel).

Toroidal shapes are a particularly interesting case, since a higher dielectric constant material is absent in the particle's core, and the gradient force is significantly reduced when the focal point is near its center of mass. It is not obvious that particles containing holes can be trapped using single-beam Gaussian tweezers. To explore the stability and configurations of a symmetric toroidal shape, we have created a series of square platelets that have identical exterior dimensions, with progressively larger central square holes having inner edge lengths L_i (fig. 2a). For the limiting case of a square prism, the platelet does trap with two of its corners lying along $\boldsymbol{k}$, like a diamond with the focal point very close to the particle's center. When a small hole is placed in the center, the single

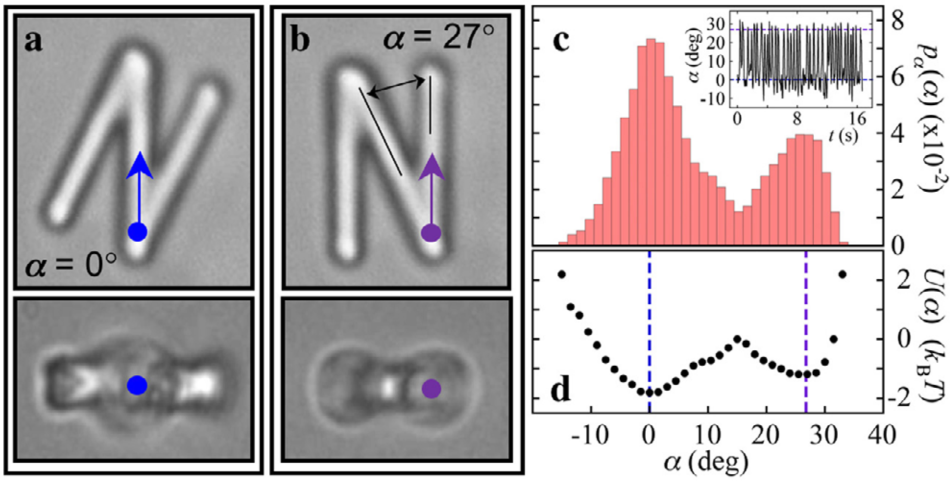

Fig. 3: (Color online) Orientational Kramers hopping of the letter N held in a single-beam Gaussian trap. Thermal driving switches the letter between two preferred orientations α with $\boldsymbol{k}$ along: a) $\alpha = 0°$, the longer linear diagonal segment (blue) and b) $\alpha = 27°$, the shorter side segment (purple) (top panel: yz-plane; bottom panel: xy-plane). c) The time-averaged angular probability distribution, $p_\alpha(\alpha)$, obtained from a time trace of α (inset), exhibits two peaks at 0° and at 27°. d) The shape of the double-well angular potential, $U(\alpha)$, extracted from $p_\alpha(\alpha)$ shows two asymmetric minima separated by a barrier that is slightly larger than thermal energy k_BT.

stable trapping point bifurcates into two possibilities —both above and below the hole. The upper point is only metastable; thermal buffeting and the radiation pressure rapidly drive the particle into the lower point where it is stably trapped. As L_i is further increased, the lower trapping location becomes metastable, and the donut cannot be permanently trapped. However, when the hole is made large enough that the particle resembles a square frame with a thickness comparable to λ, stable trapping re-appears, albeit in a different position and orientation with $\boldsymbol{k}$ along an edge. Interestingly, this corresponds to the behavior of the oblong letter O, which traps stably, but with $\boldsymbol{k}$ pointing along the shorter dimension. As the frame is made yet thinner, the volume of dielectric material in the brightest part of the focused beam becomes small enough, while the radiation pressure remains large, so the tweezers cannot hold the particle. This example illustrates the richness of trapping phenomena that can occur when systematically varying the geometry of a simple symmetric shape.

We have measured the distribution of escape times, $p_e(t)$, of the square-frame particle in the metastably trapped state (fig. 2b); this is an example of the classic problem of thermally activated escape over a potential barrier [23,24]. Although simulations have predicted an asymmetric distribution [25] that is similar to our observations, no simple analytical prediction is known, so we empirically fit our data to a log-normal distribution that captures the prominent tail. The most probable escape time is $t_{mp} = 0.76 \pm 0.03$ s. We estimate the thermal attempt frequency of the particle in a diffraction-limited potential well to be $\omega_0 \approx 2\pi D/(\lambda/2)^2 \approx 2\,\mathrm{s}^{-1}$, where D is the diffusion coefficient. For simple Kramers escape, $t_{mp} \approx \omega_0^{-1} \exp(\Delta U/k_BT)$, and we deduce that the barrier height ΔU of the metastable trap is only slightly larger than k_BT; particles almost always leave after a few attempts. When the square-frame particles leave the trap, they usually rotate with significant angular momentum.

Thermally driven hopping of the letter N between two different angular positions around one stable trapping point is an interesting example of orientational bi-stability. We have measured the temporal dependence of the angle, α, about the x-direction perpendicular to $\boldsymbol{k}$, and have extracted the time-averaged angular probability, $p_\alpha(\alpha)$ (fig. 3 and supplementary information given online by N_4x_slower.avi[1]). This distribution shows a more prominent peak at $\alpha = 0°$, corresponding to the alignment of the longer diagonal connector of N along $\boldsymbol{k}$, and a smaller peak centered around $\alpha = 27°$, corresponding to the alignment of a shorter side segment of N along $\boldsymbol{k}$. Assuming that p_α can be described by a Kramers hopping problem in the limit of Gaussian noise with zero mean [24], we determine the angular potential, $U(\alpha) = -k_BT \ln(p_\alpha(\alpha))$, where we fix $U = 0$ at the top of the barrier between the wells. The potential has two asymmetric wells [26] with minima at $\alpha = 0°$ and 27°, and the central barrier is larger than k_BT when approached from either well. This double-well angular potential provides a simple one-dimensional illustration of complexity in the potential in the six-dimensional space given by the position of the particle's center of mass relative to the focal point and its three angles of orientation. This observation

[1]Movie of the angular fluctuations of the letter N in a single-beam optical trap at laser power $P = 17$ mW. The view from the bottom is over a real duration of 5 s, adjusted to play at one quarter of the actual speed; the frame width is 14 μm.

of angular bistability of a particle in a simple optical trap contrasts with the translational thermal hopping resulting from complex potentials using two laser beams [27] or more sophisticated diffractive optical elements [2].

We find that the addition of a short dielectric rod to the structure of a particle that we could not trap can effectively provide a "handle" that permits trapping (*e.g.* A does trap, yet V does not). Although many particles having holes significantly larger than λ can be trapped, the addition of a small hole comparable to λ may destabilize a particle. The stability of a given particle depends on the laser power; some particles that can be trapped at higher P cannot be trapped at lower P. Likewise, it is possible that the letters that we could not trap at the laser's maximum P can actually be trapped at higher P, since the optical potential well may become significantly deeper than k_BT. It would be interesting to explore how the stably trapped states of dielectric letters change when written with a narrower pen width that is smaller than λ.

These experiments provide an overview of many important new trapping phenomena that can arise when particles having complex shapes interact with a simple gradient optical trap: multiple stable trapping configurations can exist for a single shape, initial conditions of particles entering a trap determine the first accessible stable configuration, thermally driven angular fluctuations of orientations of trapped particles that have bi-stable angular potential wells, and re-entrant trapping of toroidal shapes depending on the size of the center hole. Since the particles we have investigated all have uniform thickness, we have only investigated a relatively small subset of shapes and sizes. Other interesting possibilities may exist with particles that have non-uniform thickness or even greater levels of complexity, such as lithographic "Janus" particles that have two differently shaped faces [4]. Although the question of which particle shapes and sizes can be trapped using a simple focused laser beam remains open, our experiments have sampled a significant diversity of morphologies and will guide theoretical work.

For all its richness, the trapping behavior that we have described has been obtained only for a single set of trapping parameters. Varying the numerical aperture, the laser power, and the laser wavelength, as well as the size and width of the letters, will provide interesting extensions to this work. We anticipate that ray-optics simulations, similar to those performed for spheres [28], will provide a rich multi-dimensional potential landscape that will confirm our findings of multiple trapped states and double-well potentials for larger complex particle shapes. For smaller shapes closer to the diffraction limit, going beyond the ray-optics approximation may be necessary. Altering the structure of the tweezing optical field will facilitate trapping, manipulation, and control of the entire alphabet in three dimensions. Introducing optical anisotropy in the polymer material will yield additional possibilities for manipulation. Our results provide insights that will facilitate the optically directed assembly of complex three-dimensional shapes composed of two or more lithographic particles.

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