Supporting information

Self-propelled round-trip motion of Janus particles in a static line optical tweezers

Jing Liu[†], *Hong-Lian Guo*[‡,*], *Zhi-Yuan Li*[†,‡,*]

†Laboratory of Optical Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China

‡School of Physics, South China University of Technology, Guangzhou 510640, China

*Address correspondence to lizy@aphy.iphy.ac.cn, phhlguo@scut.edu.cn

- **Experimental setup and Janus particle preparation details**

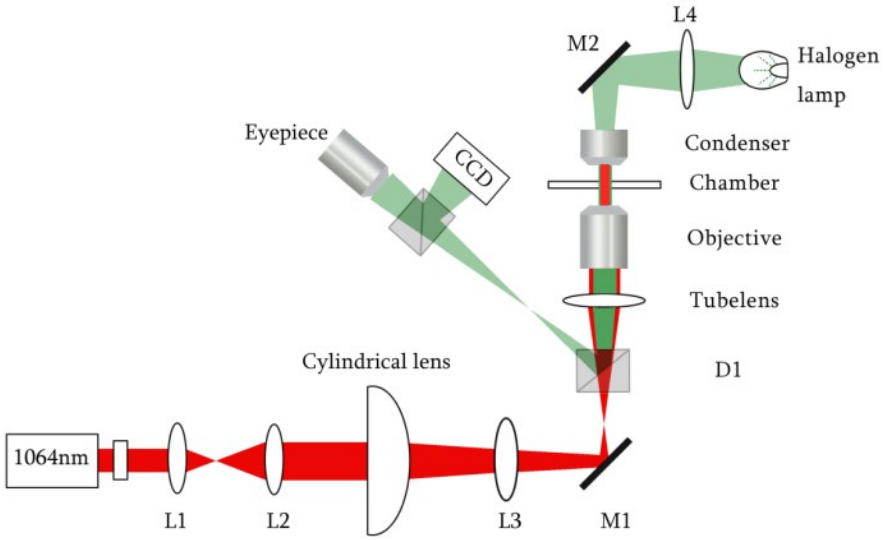


Figure S1. Experimental schematic diagram of the LOT setup.

A. Line optical tweezers setup

The experimental setup for manipulating Janus micro-particles in aqueous solution with the line optical tweezers (LOT) is shown in Figure S1(a). A collimated laser beam (Nd:YAG Coherent Compass 1064nm) was expanded by lenses L1 and L2, and then introduced into cylindrical lens, which converted a circular-shaped Gaussian beam into a line-shape beam. Then the line-shape beam was reflected by a mirror into a water-immersion objective (Leica, $\times$100, NA=1.2) in an inverted microscopy (Leica DMIRB), and focuses into an optical line on the focal plane, forming LOT. Simply speaking, LOT is an asymmetric focused Gaussian beam where the beam size is comparable with a point optical tweezers (200-300 nm) in the two transverse directions and much larger (~50 μm) in the longitudinal line direction. The intensities are lower at the two ends of the line and higher at the center, creating a small yet non-zero gradient force in the longitudinal direction. As a result, the non-uniform intensity LOT allows confinement and

trapping of particles in 3D space, albeit tight in the transverse directions and loose in the line direction.

B. Preparation of Janus particle

In our experiments, we produced the half Au-coated metallo-dielectric Janus particles by magnetron sputtering of a thin gold layer onto polystyrene (PS) microspheres. Janus particles were fabricated in a process illustrated in Figure S1(b). Firstly, a drop of PS (diameter 5 μm) solution was deposited on a clean hydrophilic glass slide. Then a monolayer of polystyrene spheres was self-assembled on the glass surface during evaporation. Secondly, a 3 nm gold layer was sputtered onto dried patches of PS particles with a magnetron sputtering at a rate of 4 nm/min. Finally, the Janus particles were dispersed in deionized water by immersing the substrate in a water-filled beaker and holding it in an ultrasonic bath for 60 s. According to Ref. [1], the nanometer Au film is thickest at the pole region (~3 nm) while thinnest at the equator region (~0 nm), and has a continual variation in thickness.

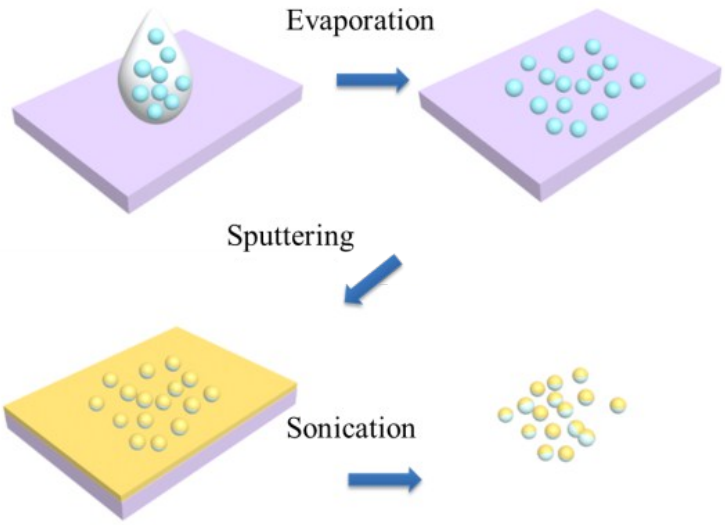


Figure S2. Schematic illustration of the fabrication process of Janus particles.

- **2. Description of the supplementary movies provided**

All movies are real time and in AVI format. These movies was recorded by a CCD camera (OK-AM1110) mounted on the microscope at the 45 frames/second. In the experiments, Kohler illumination is used to reduce image artifacts and provide high sample contrast. Illumination lights are incident on the sample and then collected by the objective lens, reflected by the dichroic prism, finally received by the CCD camera. In order to differentiate the Au-PS separation line, the lamp voltages in the movie S1 and S2 are 5 V, which is higher than the 2.5 V lamp voltages in the movie S3. As the gold transmits less light than the PS, the Au-coated hemisphere is darker.

Video S1: Supplementary Movie 1_5umJanus_40mW.avi

A 5 μm in diameter Janus particle makes round-trip motion in LOT at the laser power of 40 mW.

Video S2: Supplementary Movie 2_5umJanus_60mW.avi

A 5 μm in diameter Janus particle makes round-trip motion in LOT at the laser power of 60 mW.

Video S3: Supplementary Movie 3_2umPS_40mW.avi

As a comparison, the motion of PS microspheres (diameter 2 μm) in the LOT at the laser power of 40 mW was recorded. An increasing number of PS microspheres were trapped and self-organized into a line at the position where the line optical trap was located.

- **3. Ray optics method used to calculate the optical force and torque**

The ray optics method[1] has been used to handle a dielectric microscopic particle and metallo-dielectric Janus microscopic particle trapped in a point optical tweezers[2,4]. In the ray optics method applied to the current LOT system, the focused beam is decomposed into a large number of individual rays, each with appropriate intensity, direction, and state of polarization. These rays propagate in straight lines in media of uniform refractive index. As illustrated in Figures S3(a) and (b), the Janus particle is the polystyrene (PS) particle with radius r_s and half-coated with gold film of thickness h. The thickness of gold film is maximal at the top surface of the PS sphere, and reduces gradually to zero at the position of equator, approximately following the relation of $h = h_0 \sin\psi$, where $h_0 = 3\text{nm}$ is the maximum thickness at the top of the PS particle, while ψ is the inclination angle of a particular point at the PS surface with respect to the *y*-axis.

The simple ray optics model of the LOT used here for calculating the trapping forces on a Janus particle is illustrated in Figures S3(c) and (d). The incident ray is noted by a vector $\vec{k}_1(k_x, k_y, k_z)$. The algorithm first determines the point on the surface where the ray enters the sphere ($\vec{a}_1$), then calculates the reflection (R), transmission (T) and absorption of the incident ray based on the local surface properties[3] (incident angle, coating thickness etc.). Multiple reflections and transmissions of a single ray are accounted for until all reflected and diffracted components from the initial ray have exited the sphere. At each point where the total momentum of the ray changes, a force is calculated. Thus the optical force from ray $\vec{k}_1$ is

$$\vec{F}_{k_1} = \frac{P_{k_1} n_1}{c}\left(\vec{k}_1 - \vec{k}_{1r} R_1 - \vec{k}_{2t} T_1 T_2 - \sum_{n=3}^{\infty} \vec{k}_{nt} T_1 T_n (R_2 \cdot R_3 \cdots R_{n-1})\right) = \frac{P_{k_1} n_1}{c} \vec{Q}_{k_1}, \qquad (1)$$

where $P_{k_1} n_1 / c$ is the incident momentum per second for this ray of light, $\vec{k}_1(k_x, k_y, k_z)$ is the initial vector of light, $\vec{k}_{1r}$ is the first reflection vector of light, and $\vec{k}_{nt}$ is the *n-th* transmission

vector of light, T_n and R_n represent the *n-th* Fresnel transmission and reflection coefficients respectively. The total optical force that the focused beam exerts on the Janus particle is the sum of forces from each individual ray,

$$\vec{F}_{optical} = \sum_{i=1}^{N} W_{k_i} \vec{F}_{k_i} = \sum_{i=1}^{N} \frac{P_{k_i} n_1}{c} W_{k_i} \vec{Q}_{k_i} . \tag{2}$$

Here W_{k_i} is the weight of contribution parameter of the k_i ray of light, which is proportional to the intensity profile of the incident Gaussian beam in the entrance pupil of the high-NA lens, and N is the total number of rays considered in the calculation. In the above equations, the force and torque are related with the power P_{k_i} of each ray of light comprising the incident laser beam. In the experiment, this parameter can be connected with the power *P* of the laser beam entering the pupil of objective lens by the following equation

$$P_{k_i} = P \frac{r \Delta\varphi \cdot \Delta r}{\pi r_{\max}^2} \exp(-\frac{r^2}{r_{\max}^2}) \tag{3}$$

for the ray of light (with directional vector $\vec{k}_i$) hitting the objective lens at the position with polar coordinate $\vec{r}_i = (r, \varphi)$. And $r_{\max}$ is the maximum radius of the objective pupil. The total optical force $\vec{F}_{optical} = \left[F_x, F_y, F_z \right]$ can be written alternatively as $\vec{F}_{optical} = \vec{Q} \cdot P n_1 / c$, where $\vec{Q} = \left[Q_x, Q_y, Q_z \right]$ denotes the linear momentum exchange efficiency (or the trapping efficiency) of the LOT on the particle.

Similarly, the elemental torque with respect to a point in space $\vec{S} = (S_x, S_y, S_z)$ can be calcualted by

$$\vec{\tau}_{k_1} = \frac{P_{k_1} n_1}{c}[(\vec{a}_1 - \vec{S}) \times \vec{k}_1 - (\vec{a}_2 - \vec{S}) \times \vec{k}_{1r} R_1 - (\vec{a}_3 - \vec{S}) \times \vec{k}_{1r} \vec{k}_{2t} T_1 T_2 - \sum_{n=3}^{\infty} (\vec{a}_n - \vec{S}) \times \vec{k}_{nt} T_1 T_n (R_2 \cdot R_3 \llcorner R_{n-1})] = \frac{P_{k_1} n_1}{c} \vec{M}_{k_1}. \tag{4}$$

$$\vec{\tau}_{optical} = \sum_{i=1}^{N} W_{k_i} \vec{\tau}_{k_i} = \sum_{i=1}^{N} \frac{P_{k_1} n_1}{c} W_{k_i} \vec{M}_{k_i}. \tag{5}$$

The total optical torque $\vec{\tau}_{optical} = \left[\tau_x, \tau_y, \tau_z\right]$ can be written alternatively as $\vec{\tau}_{optical} = \acute{M} \cdot P n_1 / c$, where $\acute{M}$ denotes the angular momentum exchange efficiency (or the torque efficiency) of the LOT on the particle.

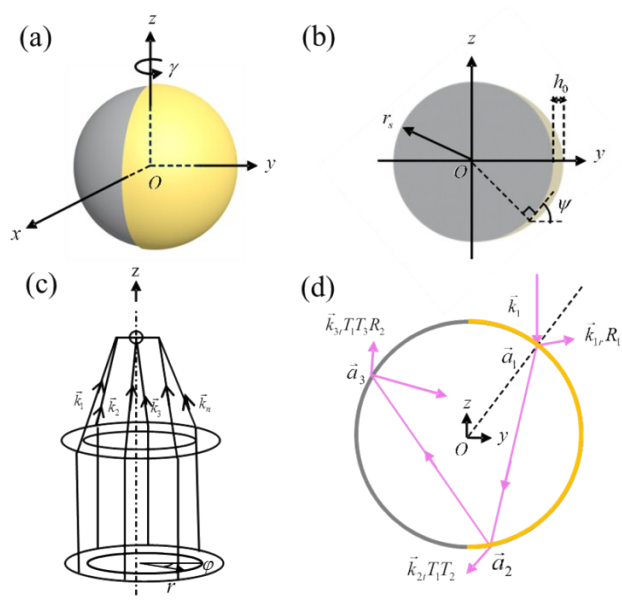


Figure S3. Schematic diagram of ray-optics model used to handle trapping and motion of Janus particles in the LOT. (a) 3D stereogram of the Janus particle made from a polystyrene (PS) bead half coated with a thin gold film. (b) Schematic diagram of the cross sectional geometry of the Janus particle in the plane of $x = 0$. The gold thin film thickness reduces gradually from the top of the PS bead (the y-axis direction) to the separation plane (the x-z plane). (c) Schematic diagram of a LOT and illuminating the Janus particle for optical trapping and manipulation. In

the ray-optics model, the laser beam is decomposed into a large amount of rays of light denoted by their directional unit vectors $\vec{k}_1$, $\vec{k}_2$, $\vec{k}_3$, L , $\vec{k}_n$, L . The $\vec{k}_i$ ray intersects with the objective pupil at the position with polar coordinate $\vec{r}_i = (r, \varphi)$. (d) Schematic diagram of the ray tracking for a specific ray $\vec{k}_1$ within the Janus particle, where a multiple events of ray reflection and refraction will take place at the positions $\vec{a}_1$, $\vec{a}_2$, $\vec{a}_3$, etc..

- **4. Equilibrium position calculation**

In this section we will show through detailed theoretical calculations and analyses that the turning points A and B in the experimentally observed motion dynamics of Janus particle correspond to the equilibrium positions of the LOT for the Janus particle and calculate the quantitative value of these two positions by using the ray-optics algorithm discussed in Sec. 3. The details and parameters for the calculation are as follows.

The line center of LOT is set to be the coordinate origin, and the collimated Gaussian beam at a wavelength of 1064 nm propagates along the +*z*-axis and is compressed to a focal line by a microscope objective. The length in the longitudinal direction of the line beam is 48 μm. The numerical aperture of water-immersion objective is $\mathrm{NA} = 1.2$, the refractive index is $\hat{n} = 0.10444 + 6.8635i$ for gold at the wavelength of 1064 nm. The Janus particle is a PS particle with radius $r_s = 2.5\mathrm{um}$ and half-coated with gold film 3 nm in maximum thickness, i.e., $h_0 = 3\mathrm{nm}$. The position of Janus particle is changed and scanned along the *x*-, *y*-, and *z*- axes in search of a position where the net optical force on the particle is zero. This obviously spans over a large amount of parameter space and thus consumes a large scale numerical calculation. However, the ray-optics algorithm is very efficient and fast to handle this task. During the position scan, the orientation angle of the particle is fixed. Different from the PS bead, the force

felt by a Janus particle strongly depends on the orientation angle and position in LOT. The orientation angle denoted by γ is the rotational angle of the Janus particle around the *z*-axis, which is shown in Figure S3(a).

To find the equilibrium position, we display the calculated total force as a function of the position and orientation of the Janus particle in the LOT. According to the experiment results, we focus to analyze two main orientations of the Janus particle in the LOT, which is $\gamma = 30°$ and $\gamma = 210°$. The trapping efficiency $\vec{Q}(\vec{r}) = (Q_x, Q_y, Q_z)$ of the Janus particle in the LOT at the orientation $\gamma = 30°$ and $\gamma = 210°$ are respectively presented in Figure S4 and S5, for clarification of comparison and discussion. It can be obtained that the equilibrium position of the Janus particle is $[-0.14, -10.0, 1.3]$μm at $\gamma = 0°$ and $[0.14, 10.0, 1.3]$μm at $\gamma = 210°$. Obviously these two positions are symmetric with respect to the line trap center. Furthermore, following the same procedures we can also find different equilibrium positions when varying the orientations of the Janus particle other than these two specific orientations. It is worth noting that the stiffness coefficient of the Janus particle in the LOT in *y*-axis (longitudinal direction) is much smaller than that in *x*- and *z*- axes (transverse direction). To calibrate the LOT to perform quantitative measurements, we need to measure the trap stiffness $\vec{\kappa} = [\kappa_x, \kappa_y, \kappa_z]$. If a particle is not far from the trap center (so-called Hookean region), the optical force a particle feels is directly proportional to its displacement $\Delta\vec{S}$ from the equilibrium position $\vec{S} = (S_x, S_y, S_z)$, i.e. $\vec{F} = \kappa_x \Delta S_x \hat{x} + \kappa_y \Delta S_y \hat{y} + \kappa_z \Delta S_z \hat{z}$. Through linear fitting the data in Hookean region in Figures S4 (b)-(d), the stiffness coefficients in *x*-, *y*-, *z*- axes of a Janus particle in a LOT are respectively $\kappa_x = 2.36 \times 10^{-3}$ pN/nm, $\kappa_z = 2.05 \times 10^{-3}$ pN/nm, and $\kappa_y = 4.20 \times 10^{-5}$ pN/nm for a laser power of 60 mW (at a wavelength of 1064 nm) in the specimen. This is a natural result because the optical

confinement (focus beam size) is much tighter (smaller) in the transverse direction than in the longitudinal direction.

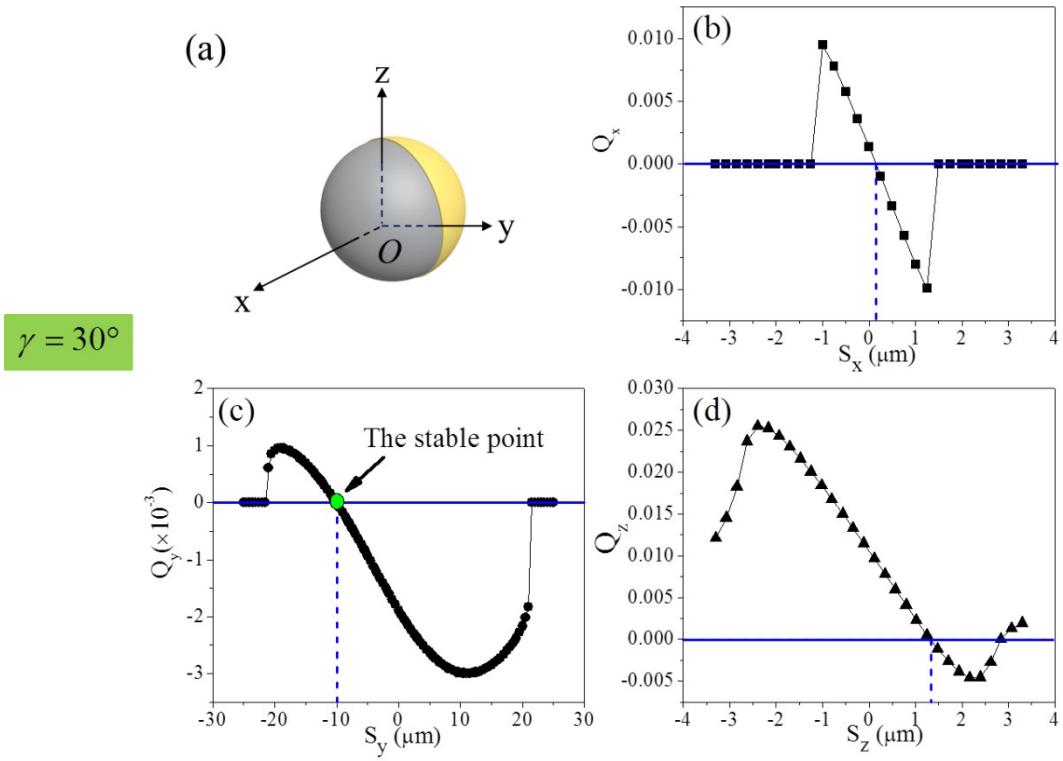


Figure S4. The trapping efficiencies of a Janus particle at the orientation $\gamma = 30°$ imposed by the LOT. (a) 3D stereogram of the asymmetric Janus particle and (b) the sketch of Q_x : S_x (c) the sketch of Q_y : S_y (d) the sketch of Q_z : S_z of a Janus particle.

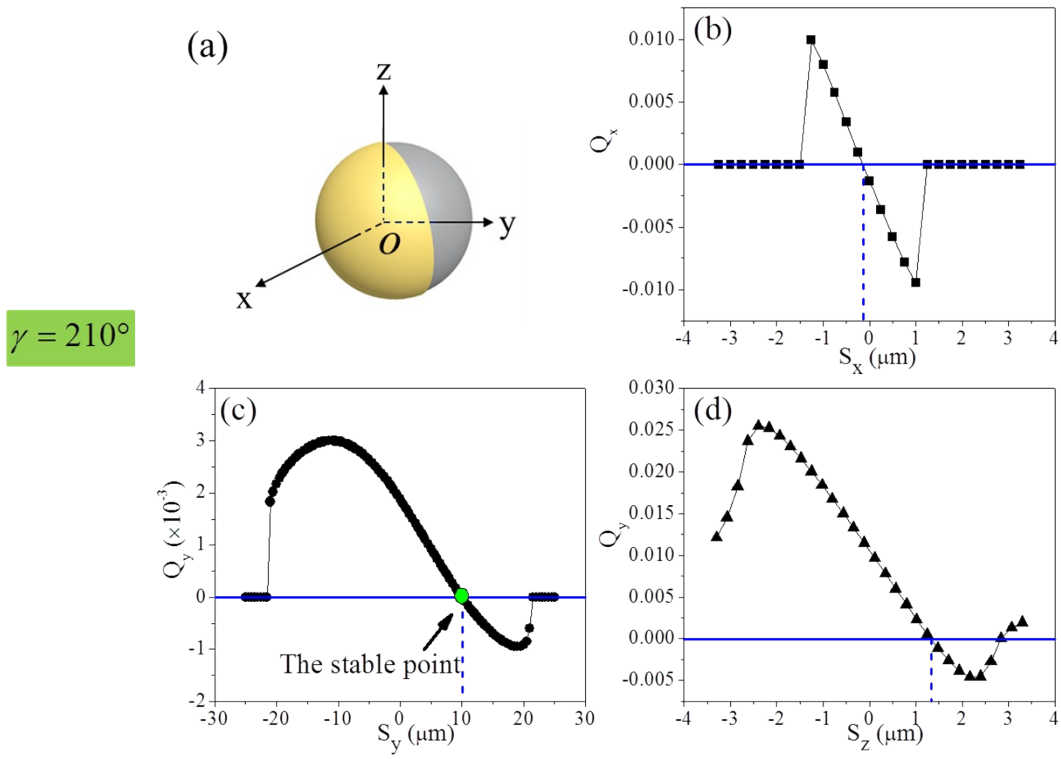


Figure S5. The trapping efficiencies of a Janus particle at the orientation $\gamma = 210°$ imposed by the LOT. (a) 3D stereogram of the asymmetric Janus particle and (b) the sketch of Q_x : S_x (c) the sketch of Q_y : S_y (d) the sketch of Q_z : S_z of a Janus particle.

- **5. Transverse fluctuation of Janus particle and consequent optical torque**

In this section we will show that in practical experiment the Janus particle does not follow a 1D rectilinear motion, namely, move in a precise straight line along the focal line of LOT, but rather is subject to random fluctuation motion in the transverse direction. We focus on the positions of the Janus particle at both ends of its trajectory in the experiments. The recorded positions of a Janus particle in *x*-axis in the experiments are shown in Figures S6(a) and (b) at laser power 40 and 60 mW, respectively.

We calculate the torque efficiency M_z of a Janus particle when it moves through the focus along the *x*-axis via the ray optics algorithm, and the result has been plotted in Figures 4c and 4f of the main body of this paper. In the numerical calculation, the optical torque exerted on a Janus particle can be written by

$$\tau_{optics} = M_z \frac{Pn_1}{c}, \tag{6}$$

where P is the laser power, n_1 is the refractive index of water ($n_1 = 1.33$), c is the speed of light in vacuum ($c = 3\times10^8$ m/s). According to the offset in *x*-axis about 0.33 μm in Figure S6(b) for the laser power $P = 60\text{mW}$, M_z is approximately $2\times10^{-4}\,\mu\text{m}$. So the calculated optical torque is $53.2\,\text{pN}\cdot\text{nm}$, and it is this optical torque that supports the rotation of a Janus particle around the *z*-axis.

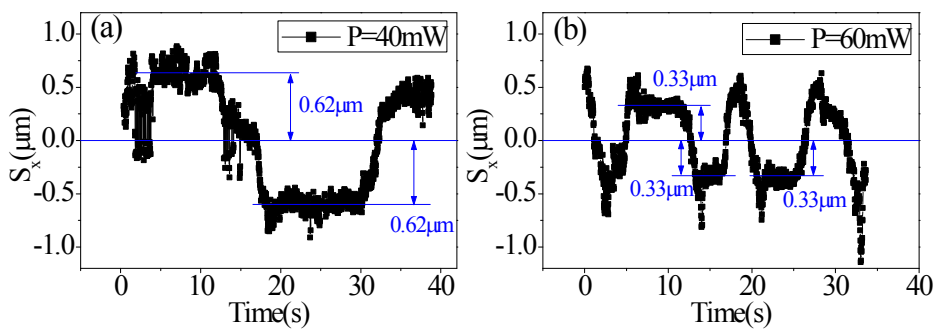


Figure S6. (a) The experimentally recorded positions in *x*-axis of the Janus particle moving along the line optical field at the laser power 40 mW and (b) at the laser power 60 mW.

- **6. Numerical analysis of dynamic processes**

In order to completely understand the physical mechanism underlying the experimentally observation of cyclic round-trip motion, we need to specify the dynamic evolution of a Janus particle in LOT theoretically. As the Janus particles basically move along *y*-axis in the LOT, we

suffice to only discuss the rectilinear motion within one-dimensional space. Following Newton's law, the differential equation of the Janus particle translational motion can be written by

$$m\ddot{r} + 6\pi r_s \eta \dot{r} - F_{optical}(r, \gamma, P) = 0 . \tag{7}$$

In Eq. (7), m represents the particle mass, r stands for the position of the particle with a radius of r_s, and the viscosity of the deionized water is denoted by $\eta = 8 \times 10^{-4}\,\mathrm{Pa \cdot s}$. $F_{optical}$ is the optical force, which is summation of the propulsion force and gradient force, and $6\pi r_s \eta \dot{r} = F_{drag}$ is the viscous drag force due to water.

As the Janus particle basically moves along the *y*-axis in the LOT, we discuss the motion within one-dimensional space, focusing on the rectilinear T_{AB} and T_{BA} process and neglecting the transverse fluctuation. As Eq. (7) cannot be solved analytically, so a fourth-order Runge-Kutta method for solving the differential equation is utilized. The second-order equation can be converted to two first-order equations

$$\begin{aligned} \dot{v} &= \frac{F_{optical}(r) - 6\pi r_s \eta \dot{r}}{m} = a(r, v), \\ \dot{r} &= v, \end{aligned} \tag{8}$$

where v and a are the velocity and acceleration of the particle, respectively. By far Eq. (8) can be solved by the classical fourth-order Runge-Kutta formula.

In numerical analysis, the Runge-Kutta methods are a family of implicit and explicit iterative methods used in temporal discretization for the approximate solutions of ordinary differential equations. The method is a fourth-order method, which has a high accuracy. In order to utilize the Runge-Kutta method to solve Eq. (8), the period *T* should be firstly divided into *m* equal time intervals. Define the span of each interval as *h*, and then we deduce *T*=*mh*. The recursive algorithm for the classical fourth-order Runge-Kutta equations can be written as

$$
\begin{aligned}
F_{v,1} &= a(r_i, v_i) \\
F_{r,1} &= v_i \\
F_{v,2} &= a(r_i + \frac{h}{2}F_{r,1}, v_i + \frac{h}{2}F_{v,1}) \\
F_{r,2} &= v_i + \frac{h}{2}F_{v,1} \\
F_{v,3} &= a(r_i + \frac{h}{2}F_{r,2}, v_i + \frac{h}{2}F_{v,2}) \\
F_{r,3} &= v_i + \frac{h}{2}F_{v,2} \\
F_{v,4} &= a(r_i + hF_{r,3}, v_i + hF_{v,3}) \\
F_{r,4} &= v_i + hF_{v,3}
\end{aligned}
\tag{9}
$$

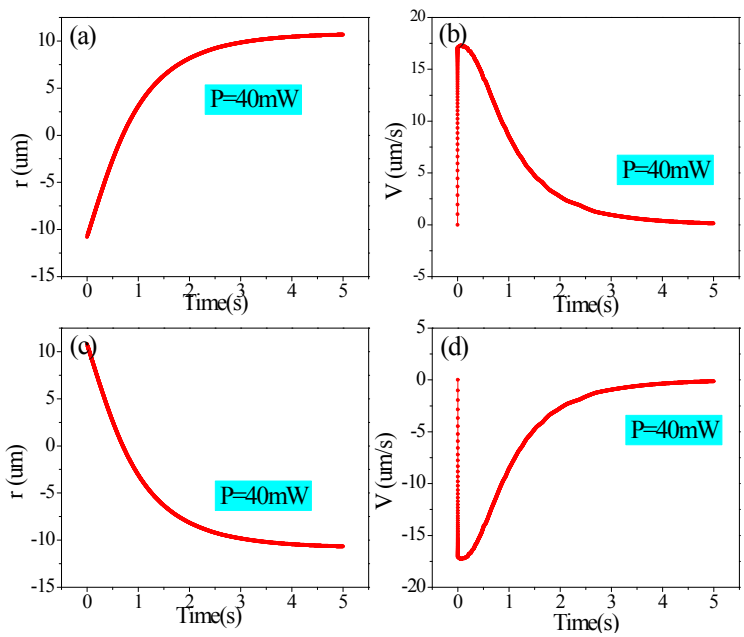
The velocity and position of a particle in the next time interval are obtained by

$$
\begin{aligned}
v_{i+1} &= v_i + \frac{h}{6}(F_{v,1} + 2F_{v,2} + 2F_{v,3} + F_{v,4}) \\
r_{i+1} &= r_i + \frac{h}{6}(F_{r,1} + 2F_{r,2} + 2F_{r,3} + F_{r,4})
\end{aligned}
\tag{10}
$$

Via iterative methods, we can get the velocity and position of the particles with high precision. The parameters we used in the calculation are as follows. The period T is 5 seconds and the time interval h is $1\times10^{-7}s$. The initial velocity of a Janus particle is set to be 0. We take the initial position r_0, the initial orientation γ of the Janus particle and the laser power P into account. The calculated velocity and position of the Janus particle in the LOT as a function of time under the condition of $r_0 = -10.0\mu\text{m},\ \gamma = 210°,\ P = 40\text{mW}$ are displayed in Figures S7(a)-(b). It is obvious that the particle moves continuously from the turning point A to B, under the propelling force of the LOT, and this corresponds to the T_{AB} process. For comparison, the calculated velocity and position of a Janus particle in a LOT over time under the condition of $r_0 = 10.0\mu\text{m},\ \gamma = 30°,\ P = 40\text{mW}$, are also displayed in Figures S7(c)-(d). It is obvious that the particle moves continuously from the turning point B to A, under the propelling force of the

LOT, and this corresponds to the T_{BA} process. Figures S7(e)-(h) are the same with Figures S7(a)-(d), respectively, except that the laser power is $P = 60\text{mW}$. The mechanical behaviors at the two laser power are similar, albeit the detailed value such as time duration of a single cycle motion as well as the maximum velocity are different.

The data in Figure S7 are for a single cycle of the round-trip motion of the Janus particle. They are used to construct the whole cyclic round-trip motion of the particle corresponding to the observation in experiments, which are illustrated in Figure 5 of the main body of this paper.



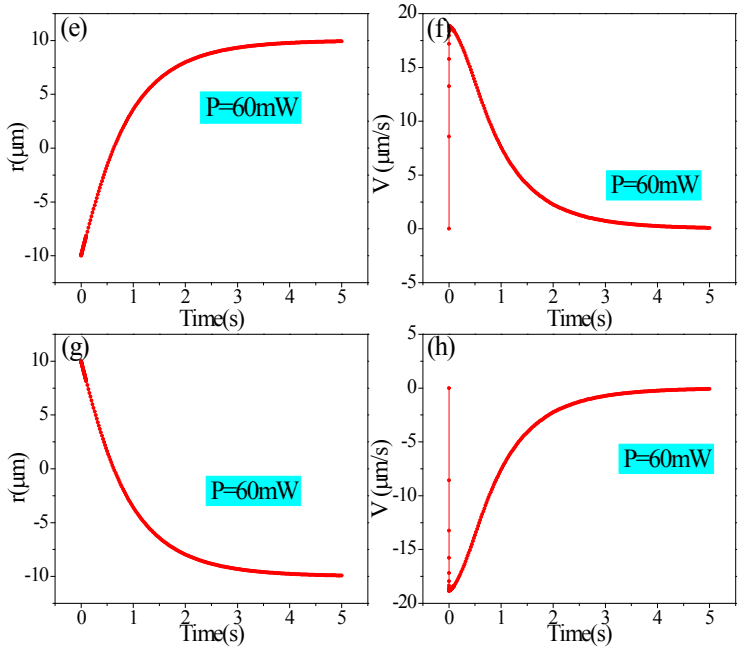


Figure S7. (a) Plot of the position over time and (b) the velocity over time under the condition of $r_0 = -10.0\mu\text{m},\ \gamma = 210°,\ P = 40\text{mW}$. (c) Plot of the position over time and (d) the velocity over time under the condition of. $r_0 = 10.0\mu\text{m},\ \gamma = 30°,\ P = 40\text{mW}$. (e) Plot of the position over time and (f) the velocity over time under the condition of $r_0 = -10.0\mu\text{m},\ \gamma = 210°,\ P = 60\text{mW}$. (g) Plot of the position over time and (h) the velocity over time under the condition of $r_0 = 10.0\mu\text{m},\ \gamma = 30°,\ P = 60\text{mW}$.

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