



## Active Motion of a Janus Particle by Self-Thermophoresis in a Defocused Laser Beam

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We study self-propulsion of a half-metal coated colloidal particle under laser irradiation. The motion is caused by self-thermophoresis: i.e., absorption of a laser at the metal-coated side of the particle creates local temperature gradient which in turn drives the particle by thermophoresis. To clarify the mechanism, temperature distribution and a thermal slip flow field around a microscale Janus particle are measured for the first time. With measured temperature drop across the particle, the speed of self-propulsion is corroborated with the prediction based on accessible parameters. As an application for driving a micromachine, a microrotor is demonstrated.

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Self-propulsion arouses interest among scientists because of its interesting dynamics, associated theoretical challenges to nonequilibrium transport phenomena, and possible applications, including microswimmers, nanomachines, and drug delivery [1–4]. Biological self-propelled molecules or biomotors [5], such as ATPase and myosin, convert chemical energy to mechanical motion through chemomechanical coupling. Such thermodynamic coupling also causes self-propulsion in physical systems as exemplified by nanorods' motion in hydrogen peroxide solutions [6]. In contrast to self-propulsion, phoretic motions, such as electrophoresis, dielectrophoresis, and thermophoresis, generally described by the linear response theory, are the directional motions of material in given external fields. Making a bridge between a phoresis and a self-phoretic motion [7] would give more insight into the mechanisms for self-propulsion. In this Letter, we produced Janus particles (the particle containing two different surfaces on its two sides [8]), by evaporating gold coat on the hemisphere of silica or polystyrene spheres, and analyzed self-propulsive motion under the laser irradiation. The Janus particle moves along a local temperature gradient which is generated by absorption of laser at the metal-coated hemisphere. The local driving gradient and flow near a microscale Janus particle are experimentally observed for the first time.

Thermophoresis of colloids in solution has been intensively discussed recently [9,10] motivated by new experimental observations. To utilize thermophoresis as a mechanism for self-propulsion of a particle, it is essential to create a local temperature gradient by the particle itself. To prepare Janus particles, a monolayer of microscale silica or polystyrene particles is prepared by a drying process. Gold is deposited by thermal evaporation to create a 25 nm thick coating on the half-side of each particle [11]. An image of 1  $\mu\text{m}$  Au-silica Janus particles is shown in Figs. 1(a) and 1(b). A thin chamber containing a solution and particles is sandwiched by two cover glasses with a 20  $\mu\text{m}$  spacer. The cover glasses are treated by BSA to prevent particles from sticking to the surfaces. A laser

(Nd:YAG, 1064 nm) is fed from the bottom of the chamber through an oil immersion objective (100 $\times$ , 1.35 numerical aperture). The laser is defocused to irradiate a wider area of a diameter about 9  $\mu\text{m}$  [Fig. 1(c)], where 20% of maximum intensity is defined as its edge. Images of particles are collected through the same objective by a high speed camera at 300 frames per second.

1  $\mu\text{m}$  Au-silica Janus particles are placed in a chamber at a low concentration to insure only one particle is caught within the view under the microscope. After applying the laser, a particle is pushed to the upper surface of the chamber and moves around within the laser irradiated region. The two-dimensional trajectories of a particle are shown in Figs. 1(d)–1(f). The particle moves faster under

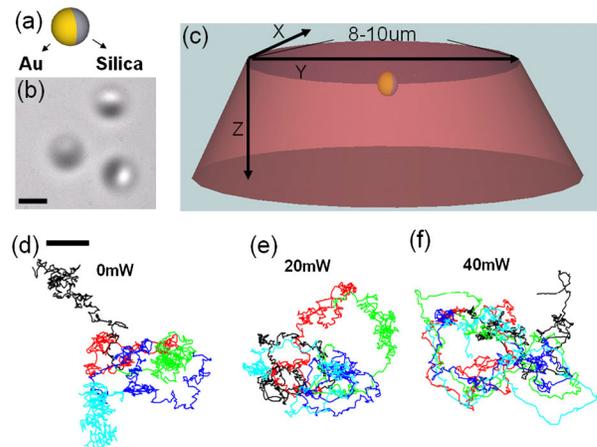


FIG. 1 (color online). (a),(b) Janus particle and its bright field image under microscope. The dark sides of the particles shown in (b) are the Au coatings (scale bar: 1  $\mu\text{m}$ ). (c) Schematic drawing of a Janus particle placed in a thin chamber (thickness: 20  $\mu\text{m}$ ). The laser, red (gray) cone, is fed in the Z direction from the bottom and the particles are visualized in the XY plane. (d) to (f) The trajectories of a Janus particle in the XY plane within 10 seconds. Each color corresponds to an interval of 2 seconds. (d) Without laser irradiation. (e) 20 mW, and (f) 40 mW. Bar: 1  $\mu\text{m}$ .

laser irradiation than the normal diffusion and the direction of motion follows the polarity of the Janus particle [see movie 1 in supplementary material (SM) [12]].

We measured the mean square displacement (MSD) of the moving particle. The MSDs for different laser irradiation powers are shown in Fig. 2(a). The power of the laser is varied from 0 to 40 mW before the objective. We observed that the irradiation of the laser increases the MSD several folds for a short time scale ( $\leq 1$  second) and the confinement of the particle for a longer time scale ( $\geq 3$  second). The latter can be explained by the two-dimensional trapping effect due to strong scattering from the metal part of the particle [13], although the laser is defocused. On the other hand, the behavior at the shorter time scale cannot be attributed by the trapping effect.

Based on these observations, we consider that the dynamics of a Janus particle is composed of three kinds of motions, Brownian motion, self-propelled motion induced by the laser, and optical trapping confinement. These three kinds of motion can be separated by two different time scales; the time constant  $\tau_k$  of the laser trapping effect and the rotational diffusion time constant  $\tau_r$  of self-propelled motion induced by the laser.

To discuss the time scale of the trapping effect, we employ the motion of a self-propelled particle in a harmonic potential without inertia as a first approximation. The MSD of the particle can be obtained as (see SM),

$$\text{MSD} = 4D\tau_k(1 - e^{-t/\tau_k}) - \frac{2V^2\tilde{\tau}}{\tau_k^{-1} + \tau_r^{-1}}e^{-t/\tau_r} + \frac{2V^2\tau_k}{\tau_k^{-1} + \tau_r^{-1}}\left(1 + \frac{\tilde{\tau}}{\tau_r}e^{-t/\tau_k}\right), \quad (1)$$

where  $\tilde{\tau} = 1/(-\tau_r^{-1} + \tau_k^{-1})$ ,  $D_r = 1/2\tau_r$  is the rotational diffusion constant,  $D$  the diffusion constant of the particle,  $V$  the self-propelling velocity,  $k$  the spring constant,  $\tau_k = k_B T/Dk$  the time constant due to the harmonic potential,  $T$  the temperature, and  $k_B$  is Boltzmann constant. When  $\tau_k \gg \tau_r$ , Eq. (1) tends to  $\text{MSD} \simeq (4D + 2V^2\tau_r)\tau_k(1 - e^{-t/\tau_k})$ , thus  $\tau_k$  can be obtained from MSD and Eq. (1) by an approximation. The values of  $\tau_k$  are from 0.4 to 3 s depending on the laser power.

The self-propelled motion along the particle's polarity  $\mathbf{n}(t)$  is characterized by the rotational diffusion of the polarity.

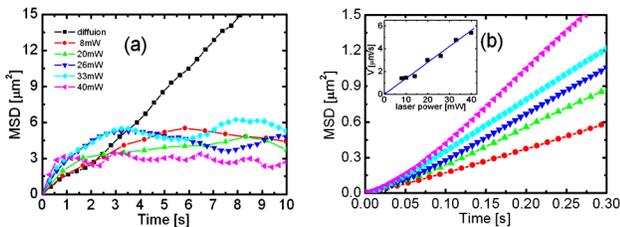


FIG. 2 (color online). (a) MSD of self-propulsion of the Janus particles under different laser power. (b) The MSD in small time scale. Inset: the relation between self-propelled velocity and laser power.

By measuring polarity of the particle from the images, the rotational diffusion time constant  $\tau_r$  was obtained by calculating the correlation function  $\langle \mathbf{n}(t) \cdot \mathbf{n}(0) \rangle = \exp(-t/\tau_r)$  (data not shown). The values of  $\tau_r$  are typically fitted between 0.1 to 0.2 s depending on the laser power. The dependence may come from the jumps of the polarity through the direction normal to the observing plane, which are more probable under higher laser power. This leads to shorter  $\tau_r$  obtained by 2D projection of the polarity. In a time scale shorter than  $\tau_r$ ,  $t \ll \tau_r \ll \tau_k$ , the rotational diffusion and the trapping effect can be neglected, and the motion can be treated as Brownian motion with a fixed directed motion. The MSD of Eq. (1) in a short time scale becomes,

$$\text{MSD} = 4Dt + V^2t^2. \quad (2)$$

The contribution from Brownian motion is  $4Dt$  and that of directed motion is  $V^2t^2$ . Similar behavior has been discussed in hydrogen peroxide solutions but without potential [14] and in the motility of the cell [15]. Equation (2) is used to fit the MSD in Fig. 2(b) up to 0.1 s to obtain a set of different  $V$  values for different laser powers with a fixed  $D$ . The data can be well fitted by Eq. (2) and  $D$  is obtained as  $0.38 \mu\text{m}^2/\text{s}$ . Comparing with the diffusion coefficient of a free particle ( $0.47 \mu\text{m}^2/\text{s}$ ), smaller value might be caused by the additional friction over the wall. The relation between self-propelling velocity  $V$  and the laser irradiation power is shown in the inset of Fig. 2(b). The overall linear relation confirms that the self-propulsion of a particle is powered by the laser irradiation linearly.

We propose that the directed motion of the Janus particle is induced by the temperature gradient generated by itself which we call self-thermophoresis. The metal side absorbs the laser of the intensity  $I$  with an absorption efficiency  $\epsilon$ . The heat generation at the surface of the particles with the radius  $R$  is balanced with thermal diffusion and thus gives  $q(\theta) = \kappa \mathbf{e}_n \cdot \nabla T$  where  $\kappa$  is the heat conductivity of water and  $\mathbf{e}_n$  is the outward normal unit vector on the surface. The temperature distribution on the surface is obtained from the heat transfer equation as

$$T(R) = T_0 + \sum_{n=0}^{\infty} \frac{q_n R}{(n+1)\kappa_o + n\kappa_i} P_n(\cos\theta) \quad (3)$$

where  $T_0$  is the temperature at infinity and  $P_n(\cos\theta)$  is the Legendre polynomial of order  $n$ . The thermal conductivity inside and outside the particle is  $\kappa_i$  and  $\kappa_o$ , respectively. The heat flux  $q(\theta)$  is expanded using the polynomial with the coefficients  $q_n$ . The temperature is asymmetric around the particle due to half-side absorption. The gap of temperature across the two sides of Janus particles is  $\Delta T = 3\epsilon IR/2(2\kappa_o + \kappa_i)$ . The propulsive velocity  $\mathbf{V}$  for a self-thermophoretic particle is given by a surface integral of effective surface slip velocity  $\mathbf{v}_s = \mu(\theta)\nabla T$  driven by local temperature gradient [7].  $\mu(\theta)$  is mobility characterized by interactions between the particle and fluids. On the other hand, exactly the same gradient can be imposed externally in which the velocity is described by the Soret

coefficient,  $S_T = D_T/D$  as  $\mathbf{V} = -DS_T\nabla T$ . Here  $D_T$  is the thermodiffusion coefficient. This suggests that the self-thermophoretic velocity is described not only by the mobility but also by the Soret coefficient, which has been rather extensively studied (see SM). The velocity is

$$\mathbf{V} = -DS_T \frac{\Delta T}{3R}, \quad (4)$$

where  $S_T$  of a Janus particle is interpreted as the average of the two coefficients of silica beads and those covered uniformly by gold. In Eq. (4),  $\Delta T$  is proportional to the laser power and accordingly velocity  $\mathbf{V}$  is proportional to it, which agrees with the MSD measurements. To further verify the proposed self-phoretic mechanism, the flow and temperature around a Janus particle and the magnitude and sign of Soret coefficient are compared with observations in the following.

The flow around a spherical colloid in temperature gradient was reported recently [16]. We expect a similar flow caused by self-thermophoresis might exist around the Janus particle. We fixed a  $3 \mu\text{m}$  Au-polystyrene Janus particle on the glass surface and added  $40 \text{ nm}$  polystyrene fluorescent particles to the solution as tracers. The distribution of tracers and motion of a large tracer are shown in Fig. 3(a). The motion of a large tracer clearly shows the direction of the circulating flow near the heated Janus particle. We also found that the concentration of tracers is high in the noncoated side and low in the coated side. It can be explained by the balance between the drag by the thermal slip flow induced by the Janus particle and the thermophoresis of the tracer itself. Tracers near the particle surface are driven from a colder to hotter region by the thermal slip and reinjected to the colder side by the circulation. But the tracers are repelled from the hotter region due to positive Soret effect, resulting in a higher density of tracers at the colder side of the Janus particle.

The temperature profile was directly measured for an Au-polystyrene Janus particle with the diameter of  $3 \mu\text{m}$  fixed on the cover glass and subjected to laser heating. We used a temperature sensitive fluorescent dye, bis(2-carboxyethyl)5, 6 carboxyfluorescein (BCECF) [17]. The temperature profile is shown in Fig. 3(b), where the laser power is  $100 \text{ mW}$  and the diameter of the irradiated region is  $25 \mu\text{m}$ . Here the larger irradiated region is adopted to scale up the length scale together with the particle size (from  $1 \mu\text{m}$  to  $3 \mu\text{m}$ ). The average laser intensity per unit area  $I$  is equal to the condition where the laser power is  $13 \text{ mW}$  in the  $9 \mu\text{m}$ -diameter region. The temperature in the coated side is about  $2 \text{ K}$  higher than the noncoated side [Fig. 3(b)]. The temperature profile qualitatively agrees with the analytic result as shown in Fig. 3(b). For a  $1 \mu\text{m}$  bead, the temperature profile is hard to obtain experimentally; the temperature gap  $\Delta T$  across the particle is estimated as about  $0.7 \text{ K}$  based on  $3 \mu\text{m}$  data. By using the velocity data in the Fig. 2(b) inset, we can estimate  $S_T$  from Eq. (4) as  $10 \text{ K}^{-1}$ . This result is consistent with the Soret coefficient ( $26 \text{ K}^{-1}$ ) of the Au-silica Janus particle independently measured by single particle tracing [18] in an

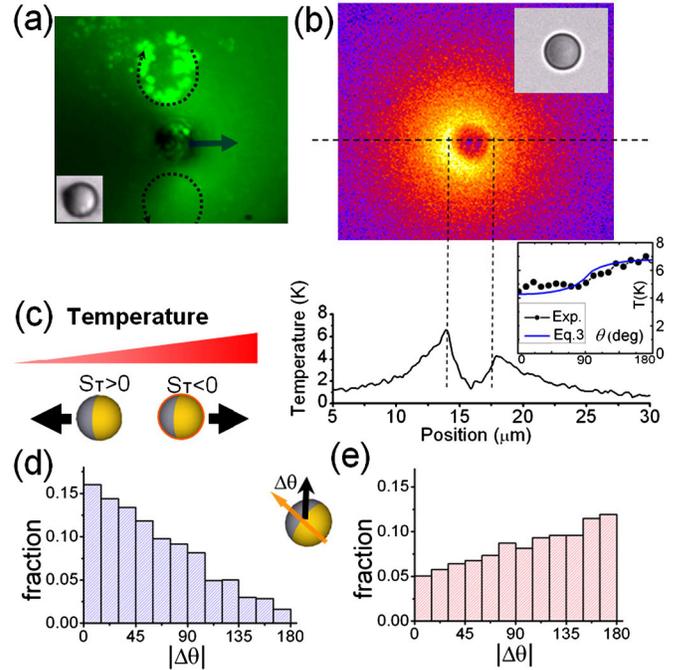


FIG. 3 (color online). (a) Distribution of  $40 \text{ nm}$  fluorescent tracers around a tethered  $3 \mu\text{m}$  Au-PS Janus particle under laser irradiation. The image is accumulated for  $5 \text{ s}$  and the bright spots are the trace of a larger particle. Dashed line arrows indicate the flows induced by temperature gradient and the solid arrow indicates the direction of motion in the nontethered condition. (b) The temperature distribution for tethered  $3 \mu\text{m}$  Au-PS Janus particle under laser irradiation. Inset: Surface temperature [solid line is from Eq. (3)]. (c) The direction of motion for a Janus particle in water and in a Triton X-100 solution. (d) Directional angle distribution of motion with respect to the polarity in water. Inset, black arrow: direction of motion; orange (grey) arrow: polarity. (e) Same kind of distribution as (d) in the Triton X-100 solution.

externally applied temperature gradient using an optically heated chamber [19,20].

Recently, it has been shown that the thermophoretic property of particles can be changed by adding surfactant, which would be absorbed on the surface of the particles [21]. The sign of the Soret coefficient of the Janus particle is also determined by single particle tracing. It shows that the Soret coefficient of the Janus particles is positive in water (moving to a colder region). Next, we add Triton X-100 to the solution ( $0.05 \text{ wt } \%$ ). We find that the Soret coefficient of the Janus particles becomes negative in Triton X-100 solution (particles move to warmer region). Figure 3(c) is the schematic figure of the directions of thermophoretic motion of a Janus particle in a temperature gradient.

We measured the angle  $\Delta\theta$  between the direction of the motion and the polarity of Janus particles in water and Triton X-100 solution as shown in Fig. 3(d) and 3(e). The motion of a Janus particle in pure water is toward the silica side, in other words, toward the colder side in the local temperature gradient created by local heating. Thus the direction of self-propelled motion agrees with that of thermophoresis in water [see Figs. 3(c) and 3(d)]. When the

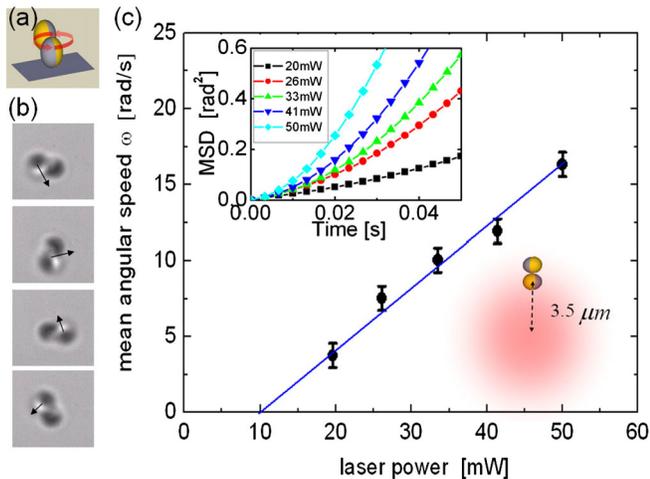


FIG. 4 (color online). (a) Schematic figure and its bright field image (b) of a microrotor during rotation. The time interval in (b) is 0.2 s for successive images. The arrow indicates the direction of rotation. (c) Rotational speed versus laser power. Inset: the MSD (left) and schematic top view of the microrotor and laser irradiation (right).

Soret coefficient becomes negative in Triton X-100 solution, Janus particles move toward the metal-coated (warmer) side [22] which confirms that the laser-induced self-thermophoresis motion does depend on the Soret coefficient. Such a reversal property of self-propelled motion is consistent with the reversal of phoretic motion and further confirms the proposed mechanism.

Finally we demonstrate that this method can be used to drive a micromachine. We carefully selected twin Janus particles which consist of two 1  $\mu\text{m}$  Au-silica Janus particles with the one being tethered to the surface [Fig. 4(a)]. We apply a focused laser at 3.5  $\mu\text{m}$  distance apart from the tethered particle [the inset of Fig. 4(c)]. As we increased the laser power, we observed that the twin particles start to rotate. The rotational direction depends on the polarity of the “vane” Janus particle. The particle moves in the direction indicated by the arrow (also see movie 2 in SM). Stable rotations of particles are observed in the range of laser power from 20 mW to 50 mW. Above a certain threshold in the laser power, the rotation speed increases linearly with the laser power [Fig. 4(c)]. The threshold for rotation may be due to an interaction potential hill existing between the bead and the glass surface. The linear velocity of the vane particle of motor can be calculated from the rotational velocity. The velocity varies linearly between 2 to 10  $\mu\text{m/s}$  as a function of the laser power. The velocity-power relation evaluated from the slope of Fig. 4(b) is 0.26  $\mu\text{m}/(\text{s mW})$ . The rotational motion has a stochastic nature at a low laser power. The MSDs of the rotor for a short time scale for different laser power are shown in the inset of Fig. 4(c). From the data, we obtain the

value of  $D_r$  for the rotor as (1.4  $\text{rad}^2/\text{s}$ ) by using a relation  $\text{MSD} = 2Dt + V^2t^2$  in one dimension.

We report the self-propulsion of Janus particles under a wide laser irradiation. The mechanism of active motion is explained by self-thermophoresis, which is related to photophoresis [23] by photothermal effect. The direction of motion is determined by the polarity of a Janus particle in contrast to the phoretic motions guided by the external fields. The external field induced self-propulsion gives us a new way to power and control micromachines. The basic principle is similar to the Crookes radiometer [24], but the scale is much more reduced.

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