Active Motion of Janus Particle by Self-thermophoresis in Defocused Laser Beam: Supplementary information

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I. MOVIES

Movie 1: Real-time motion of a 1 \( \mu m \) Au-silica Janus particle under laser irradiation. The movie is taken at the condition that the laser power is 40 mW, measured before the objective, and the diameter of the irradiated range is about 9 \( \mu m \).

Movie 2: Real-time motion of twin Janus particles (micro-rotor) which consist of two 1 \( \mu m \) Au-silica Janus particles with the one being tethered to the surface as shown in Fig. 4. The bright spot below the micro-rotor is the center of the laser. The on-off control of the laser is controlled by a shutter manually. The micro-rotor rotates only when the laser is on (laser power: 50 mW, measured before the objective).

II. DERIVATION OF EQ.(1)

Let us consider the probability distribution \( P(\mathbf{r}, \mathbf{n}) \) of position \( \mathbf{r} \) and orientation (polarity) \( \mathbf{n} \) of a Janus particle spatially confined in two dimensions (xy-plane) and rotating in three dimensions. In addition, the particle is trapped in the xy-plane with the harmonic potential \( U = \frac{1}{2}kr^2 \) with \( r = |\mathbf{r}| \). \( k \) is the spring constant of the potential. The probability distribution yields following Fokker-Planck equation [1]

\[
\frac{\partial P(\mathbf{r}, \mathbf{n})}{\partial t} = -\frac{\partial}{\partial \mathbf{r}} \cdot (\mathbf{v} P) - \mathcal{R} \cdot (\omega P)
\]  

(1)

where \( \mathcal{R} = \mathbf{n} \times \frac{\partial}{\partial \mathbf{n}} \) is the rotational operator, and the velocity is given by

\[
\mathbf{v} = -D \frac{\partial}{\partial \mathbf{r}} (k_B T \ln P + U) + V \mathbf{n},
\]

(2)

where \( V \) is interpreted as self-propulsive velocity of a Janus particle. This implies that the unit vector \( \mathbf{n} \) is chosen as the direction of self-propulsion. The angular velocity is given by

\[
\omega = \frac{1}{\zeta_r} \mathbf{N} = -\frac{1}{\zeta_r} \mathcal{R}(k_B T \ln P + U),
\]

(3)

where \( \zeta_r \) is rotational friction constant and \( \mathbf{N} \) is torque. The closed form of the equation is written as

\[
\frac{\partial P(\mathbf{r}, \mathbf{n}, t)}{\partial t} = \frac{\partial}{\partial \mathbf{r}} \cdot D \left( \frac{\partial P}{\partial \mathbf{r}} + \frac{P}{k_B T} \frac{\partial U}{\partial \mathbf{r}} \right) - V \frac{\partial}{\partial \mathbf{r}} \cdot (\mathbf{n}P) + D_r \mathcal{R} \cdot \mathcal{R} P.
\]

(4)

For a sphere, the translational and rotational diffusion coefficients are respectively given by \( D = k_B T/(6\pi \eta R) \) and \( D_r = k_B T/(8\pi \eta R^3) \) where \( \eta \) is shear viscosity in bulk and \( R \) is the radius of a sphere [2].

The correlation functions are calculated using the Green’s function; for example,

\[
\langle \mathbf{n}(t) \cdot \mathbf{n}(0) \rangle = \int d\mathbf{n}d\mathbf{n}' d\mathbf{r}d\mathbf{r}' [\mathbf{n} \cdot \mathbf{n}' \mathcal{G}(\mathbf{n}, \mathbf{n}', \mathbf{r}, \mathbf{r}, t)] P_{st}(\mathbf{r}', \mathbf{n}')
\]

(5)

where \( P_{st} \) is the probability at steady states. The Green function yields

\[
\frac{\partial \mathcal{G}(\mathbf{r}, \mathbf{r}', \mathbf{n}, \mathbf{n}', t)}{\partial t} = \frac{\partial}{\partial \mathbf{r}} \cdot D \left( \frac{\partial \mathcal{G}}{\partial \mathbf{r}} + \frac{k}{k_B T} \mathbf{r} \right) \mathcal{G} - V \frac{\partial}{\partial \mathbf{r}} \cdot (\mathbf{n} \mathcal{G}) + D_r \mathcal{R} \cdot (\mathcal{R} \mathcal{G})
\]

(6)

with the initial condition

\[
\mathcal{G}(\mathbf{r}, \mathbf{r}', \mathbf{n}, \mathbf{n}', 0) = \delta(\mathbf{r} - \mathbf{r}') \delta(\mathbf{n} - \mathbf{n}').
\]

(7)
With integration by part, the equation for the correlation function is obtained as

$$
\frac{\partial}{\partial t} \langle \mathbf{n}(t) \cdot \mathbf{n}(0) \rangle = \int d\mathbf{n}' d\mathbf{r}' [\mathcal{L} \mathbf{n} \cdot \mathbf{n}' G(\mathbf{n}, \mathbf{n}', t) P_n(\mathbf{r}', \mathbf{n}')] \tag{8}
$$

with the operator $\mathcal{L} = \frac{\partial}{\partial t} D \cdot \frac{\partial}{\partial r} - \frac{Dk}{\kappa} \mathbf{r} \cdot \frac{\partial}{\partial r} + V \mathbf{n} \cdot \frac{\partial}{\partial r} + D_r \mathbf{R}^2$. Using $\mathcal{L} \mathbf{n} = -2D_r \mathbf{n}$, we obtain

$$
\langle \mathbf{n}(t) \cdot \mathbf{n}(0) \rangle = \exp(-2D_r t). \tag{9}
$$

Similar calculation is applied to the correlation function $\langle \mathbf{n}(t) \cdot \mathbf{r}(0) \rangle$ and $\langle (\mathbf{r}(t) - \mathbf{r}(0))^2 \rangle = 2\langle (\mathbf{r}(t))^2 \rangle - \langle \mathbf{r}(t) \cdot \mathbf{r}(0) \rangle$. With $\mathcal{L} \mathbf{r} = -Dk/(k_B T) \mathbf{r} + V \mathbf{n}$, the equation for the translational motion is explicitly given as

$$
\frac{\partial}{\partial t} \langle (\mathbf{r}(t))^2 \rangle = 4D - \frac{2Dk}{k_B T} \langle (\mathbf{r}(t))^2 \rangle + 2V \langle \mathbf{n}(t) \cdot \mathbf{r}(t) \rangle, \tag{10}
$$

and

$$
\frac{\partial}{\partial t} \langle \mathbf{r}(t) \cdot \mathbf{r}(0) \rangle = -\frac{Dk}{k_B T} \langle \mathbf{r}(t) \cdot \mathbf{r}(0) \rangle + V \langle \mathbf{n}(t) \cdot \mathbf{r}(0) \rangle, \tag{11}
$$

where $\langle \mathbf{n}(t) \cdot \mathbf{r}(t) \rangle = V/(\tau_k^{-1} + \tau_r^{-1})$. Solving the equation, we finally obtain

$$
\langle (\mathbf{r}(t) - \mathbf{r}(0))^2 \rangle = 4D \tau_k (1 - e^{-t/\tau_k}) - 2V \langle \mathbf{n}(t) \cdot \mathbf{r}(t) \rangle \tau_r e^{-t/\tau_r} + 2V \langle \mathbf{n}(t) \cdot \mathbf{r}(t) \rangle \tau_k \left( 1 + \frac{\tau_r}{\tau_k} e^{-t/\tau_k} \right), \tag{12}
$$

where $\tau = 1/(-\tau_r^{-1} + \tau_k^{-1})$, $\tau_r = 1/(2D_r)$ and $\tau_k = k_B T/(Dk)$. When $k = 0$, the results is equivalent to the results in [3].

### III. Derivation of Eq.(3)

Let us consider a particle with laser-absorbing surface moving with the velocity $V$. When the characteristic length $\lambda$ of interaction between the particle and fluid is much shorter than the radius $R$ of the particle, we may approximate surface of the colloid to be flat. A temperature gradient around the particle induces a slip flow in the direction of latitude angle $\theta$ with the axis of the direction of motion [4, 5]

$$
\mathbf{v}_s = v_s \mathbf{e}_\theta = \mu(\theta)(\nabla T) \mathbf{e}_\theta \tag{13}
$$

where $\mathbf{e}_\theta$ is the tangential unit vector on surface, $\mu(\theta) = -(k_B / \eta)|\Gamma|\lambda$ is a mobility coefficient of a particle by a temperature gradient, $\lambda$ is characteristic length, $\lambda = \Gamma^{-1} \int c_0 y(e^{-\beta U_0} - 1) dy$ and $\Gamma = \int c_0 (e^{-\beta U_0} - 1) dy$ with the potential $U_0$ of the interaction between the particle and fluid of density $c_0$.

We assume the temperature distribution yields the heat equation

$$
\nabla^2 T_{i,o} = 0, \tag{14}
$$

where the subscripts $i$ and $o$ denote temperature fields inside and outside of a particle. The boundary conditions are continuity at the surface

$$
T_o(R) = T_i(R), \tag{15}
$$

and continuity of flux including absorption of laser $q(\theta)$

$$
-\kappa_o \mathbf{n} \cdot \nabla T_o + \kappa_i \mathbf{n} \cdot \nabla T_i = q(\theta). \tag{16}
$$

The absorption of heat $q(\theta) > 0$ is expanded using the Legendre Polynomials as $q(\theta) = \sum_{n=0}^{\infty} q_n P_n(\cos \theta)$. Solving the equation, we obtain the temperature distribution at the surface as

$$
T(R) = T_\infty + \sum_{n=1}^{\infty} \frac{q_n R}{(n+1) \kappa_o + n \kappa_i} P_n(\cos \theta), \tag{17}
$$
and accordingly the effective slip velocity as

$$v_s = \mu(\theta) \sum_{n=1}^{\infty} \frac{q_n}{(n+1)\kappa_0 + n\kappa_1} \frac{dP_n(\cos \theta)}{d\theta}. \quad (18)$$

When the velocity of the particle is slow enough, the low-Reynolds hydrodynamics may be assumed. The velocity of a particle is obtained either by solving the Stokes equation $\eta \nabla^2 \mathbf{v} = \nabla p$ with $\eta$ being shear viscosity of the fluid and $p$ being pressure, or using Lorentz Reciprocal theorem [2, 6]. Both methods lead to the same result

$$V = -\frac{1}{2} \int_0^\pi v_s \sin^2 \theta d\theta. \quad (19)$$

The velocity of a particle obtained above contains mobility $\mu(\theta)$, which is not the quantity measured easily, while the velocity of particles under linear temperature gradients is characterized by the material-dependent coefficient, Soret coefficient $S_T$. The coefficient for various materials have been extensively investigated and well known. It is therefore informative to characterize motion of Janus particles with those well-known parameters. Here we consider a particle under a linear temperature gradient imposed externally as

$$T = T_\infty + T_1 R \left[ \frac{r}{R} + \frac{1}{2} \left( \frac{R}{r} \right)^2 \cos \theta \right]. \quad (20)$$

The temperature gradient at infinity is $T_1$. The surface of the particle is uniform, and thus $\mu(\theta) = \mu_0$ is constant. The temperature on surface is then

$$T(R) = T_\infty + \frac{3}{2} T_1 R \cos \theta \quad (21)$$

which gives the tangential slip velocity on surface of particles with uniform coefficients as

$$v_s = -\frac{3\mu_0 T_1}{2} \sin \theta, \quad (22)$$

and this leads to the velocity of a particle

$$V = -\mu_0 T_1. \quad (23)$$

Now we introduce the Soret coefficient $S_T$ as

$$V = -DS_T T_1. \quad (24)$$

It is clear that the Soret coefficient is related with the mobility coefficient as

$$DS_T = \mu_0. \quad (25)$$

This results suggest that Soret coefficient can be used to characterize for Janus particle instead of the interaction potential, which is typically unknown and hard to measure.

We return to the velocity of Janus particles in which two sides have different interaction potentials and absorption rates of laser. This requires the expansion $\mu(\theta) = \sum_{n=0}^{\infty} \mu_n P_n(\cos \theta)$, and $\mu_0$ used above corresponds to the mean value of $\mu(\theta)$. The coefficients of the Legendre polynomial for Janus particle are explicitly described as

$$\begin{cases} q_{2l} = 0 \\ q_{2l+1} = \frac{4l+3}{2} \frac{(-1)^l (2l)!}{(l+1)! l!} \end{cases} \quad (26)$$

for $l = 0, 1, 2, \ldots$. From (18), the velocity of the particle is obtained as

$$V \sim \mu_n q_m. \quad (27)$$

with $n + m$ being odd integer for $n \geq 0$ and $m \geq 1$. However from (26), the even modes disappear except $n = 0$. Therefore, the only contribution to the velocity appears at $n = 0$ and $m = 1$, which suggests only the average of
two interaction potential is relevant. From the discussion above, the averaged interaction is described by two Soret coefficients:

\[ \mu_0 = \frac{D}{2} (S^0_T + S^G_T), \]  

(28)

where \( S^0_T \) is the Soret coefficient of a particle (silica for instance) and \( S^G_T \) is that of gold. With the efficiency of laser absorption \( \epsilon \) and its intensity \( I \), we obtain \( q_1 = \frac{3}{4} \epsilon I \), and finally the velocity of the Janus particle is described by the measurable variables as

\[ u = -\frac{1}{4} D (S^0_T + S^G_T) \frac{\epsilon I}{2\kappa_0 + \kappa_1}. \]  

(29)

Using \( \Delta T \) obtained from (17), we finally obtain Eq.(4) in the main text.


