Supplementary Information:
Light-Activated Self-thermophoretic Janus Nanopropellers

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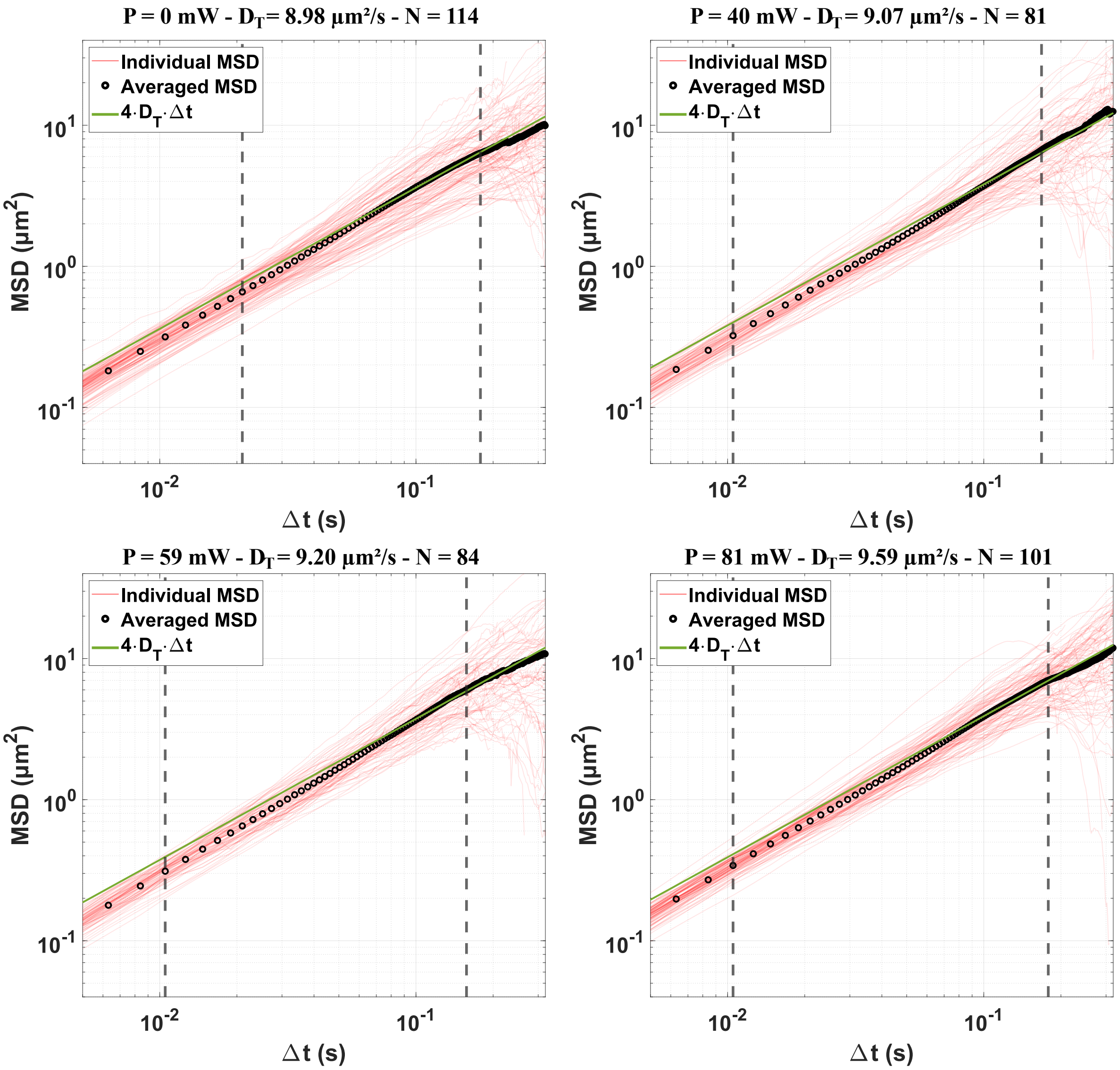

FIG. S1. Mean squared displacement (MSD) of gold nanoparticles under varying illumination conditions. Individual MSD traces are shown as light red lines, while open circles represent the ensemble-averaged MSD. The green lines correspond to a linear fit of the averaged MSD, from which the diffusion coefficients are extracted. Vertical gray dashed lines indicate the fitting range.

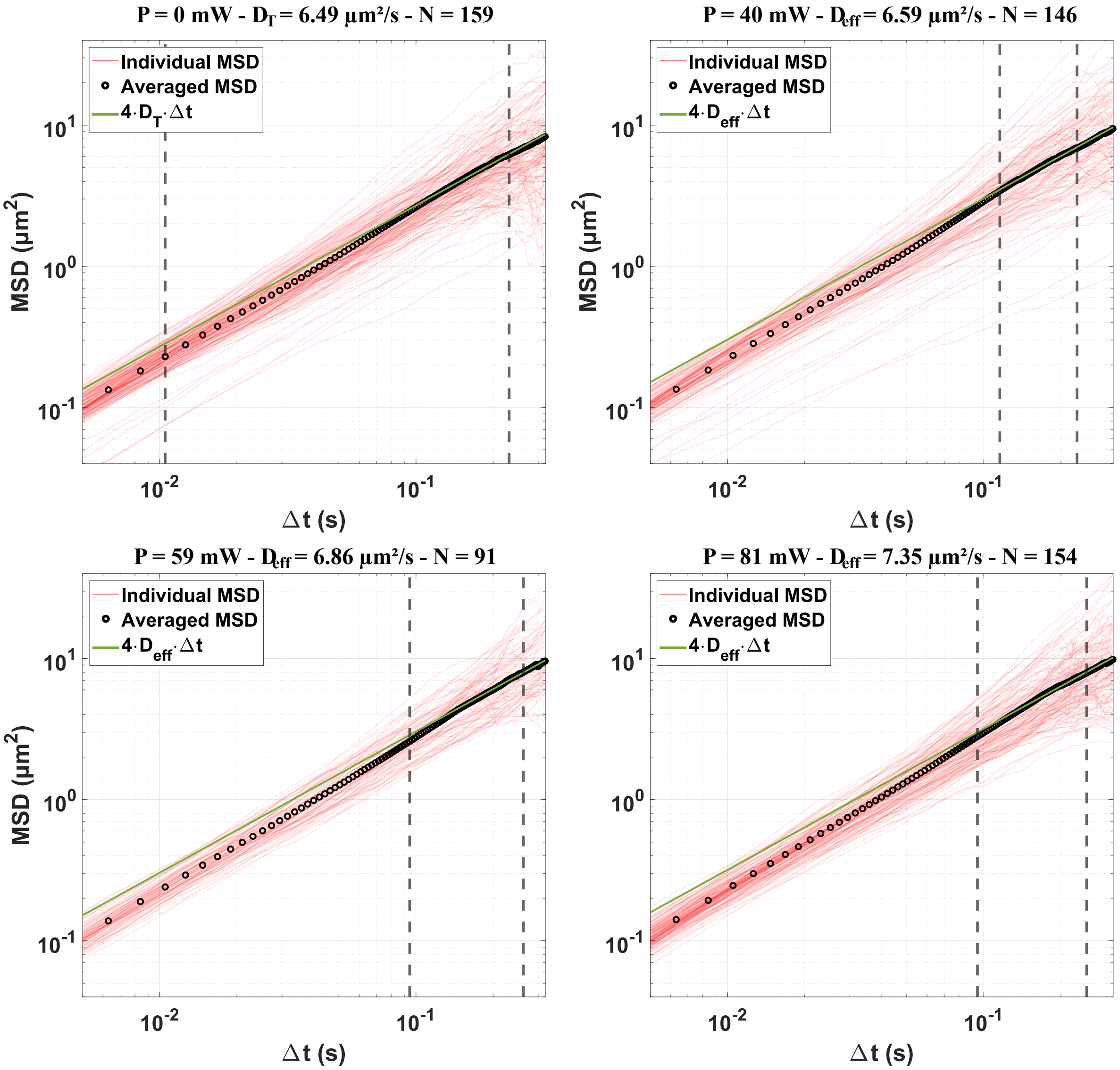

FIG. S2. Mean squared displacement (MSD) of Janus nanoparticles under varying illumination conditions. Individual MSD traces are shown as light red lines, while open circles indicate the ensemble-averaged MSD. The green lines represent a linear fit to the averaged MSD, used to extract the diffusion coefficients. Vertical gray dashed lines indicate the fitting range.

I. THEORY OF HOT BROWNIAN MOTION (HBM)

According to the Stokes-Einstein equation, the translational diffusion coefficient of a spherical colloid of radius R is:

$$\mathrm{D_T} = \frac{k_B T}{6\pi\eta R} \tag{S1}$$

where k_B is the Boltzmann constant, T the absolute temperature, and η the solvent viscosity. The temperature dependence of Eq. S1 arises from both the thermal energy, $k_\mathrm{B}T$, and the viscosity $\eta(T)$. For water, the temperature dependence of viscosity is well described by the Vogel–Fulcher law:

$$\eta(T) = \eta_\infty \exp[A/(T - T_{VF})], \tag{S2}$$

with $\eta_\infty = 0.02984$ mPa.s, $A = 496.9$ K, and $T_{VF} = 152.0$ K [1]. The resulting temperature dependence of the normalized diffusion coefficient is plotted in Fig. S3 (blue dashed line).

When excited under light illumination, Brownian particles can be considered as steady-state heat sources for which Rings *et al.* have developed a theoretical description of this non-equilibrium process [1, 2]. Their framework generalizes the Stokes-Einstein relation (Eq. S1) by introducing an effective temperature,

$$T_\mathrm{HBM} \simeq T_0(1 + \frac{5}{12}\Delta T), \tag{S3}$$

where ΔT is the temperature difference between the particle surface and the bulk solvent at temperature T_0 and viscosity η_0, and an effective viscosity η_HBM, given by Eq. S4:

$$\frac{\eta_0}{\eta_\mathrm{HBM}} = 1 + \frac{193}{486}\left[\ln\frac{\eta_0}{\eta_\infty}\right]\frac{\Delta T}{T_0 - T_{VF}} - \left[\frac{56}{243}\ln\frac{\eta_0}{\eta_\infty} - \frac{12563}{118098}\ln^2\frac{\eta_0}{\eta_\infty}\right]\left(\frac{\Delta T}{T_0 - T_{VF}}\right)^2 + O(\theta^3) \tag{S4}$$

By combining Eqs. S3 and S4 with Eq. S1, the normalized translational diffusion coefficient for a particle undergoing hot Brownian motion can then be expressed as:

$$\begin{aligned}\frac{\mathrm{D_{HBM}} - \mathrm{D_0}}{\mathrm{D_0}} \approx &\left[1 + \frac{193T_0}{243A}\ln^2\frac{\eta_0}{\eta_\infty}\right]\frac{\Delta T}{2T_0} \\ &- \left[1 - \ln\frac{\eta_0}{\eta_\infty} - \frac{386T_0}{81A}\ln^2\frac{\eta_0}{\eta_\infty} + \frac{448T_0^2}{81A^2}\ln^3\frac{\eta_0}{\eta_\infty}\left(1 - \frac{12563}{27216}\ln\frac{\eta_0}{\eta_\infty}\right)\right]\frac{\Delta T^2}{24T_0^2}\end{aligned} \tag{S5}$$

Its temperature dependence is shown in Fig. S3 (orange solid line).

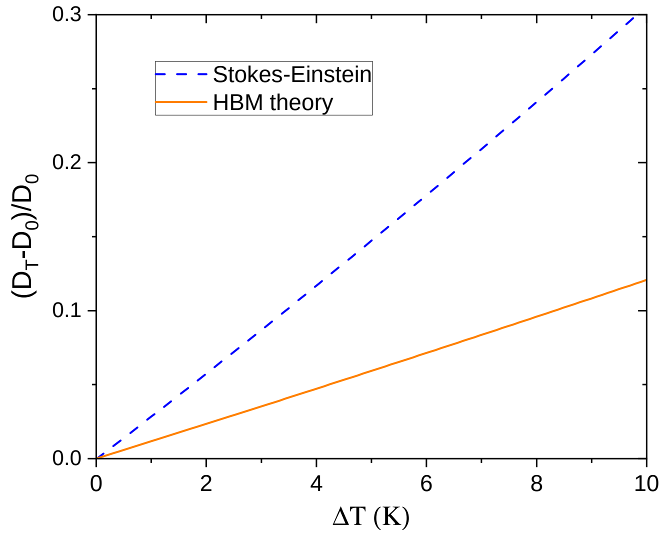

FIG. S3. Temperature dependence of normalized translational diffusion coefficient calculated from hot Brownian motion (HBM) theory using Eq. S5 (orange solid line) compared with the prediction of the Stokes-Einstein equation (Eq. S1, blue dashed line).

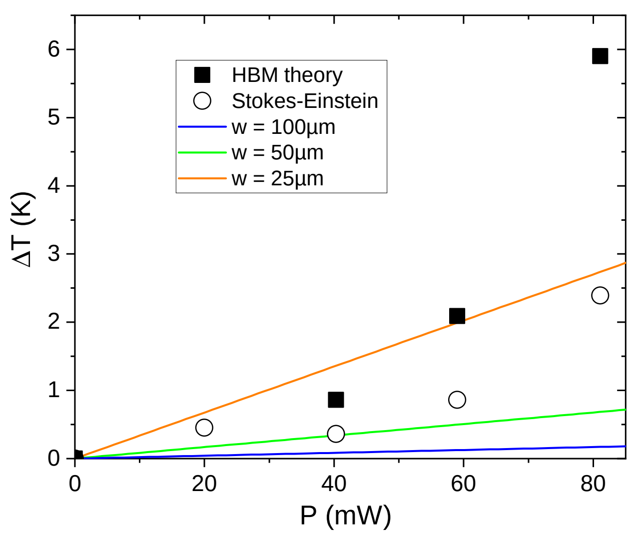

FIG. S4. Temperature increase as a function of laser intensity calculated from the experimental rescaled diffusion coefficient (Fig. 9(b)) using HBM theory (Eq. S5 and Fig. S3) consistently exceeds the prediction of Eq. 4 (main text) for different beam sizes, w. This indicates that the nanoparticles are not thermally independent.

II. SUPPLEMENTARY MOVIE

The Supplementary Movie S1 shows Janus nanoparticles observed by dark-field microscopy in the absence of laser illumination.

[1] Daniel Rings, Markus Selmke, Frank Cichos, and Klaus Kroy. Theory of Hot Brownian Motion. *Soft Matter*, 7(7):3441–3452, March 2011.

[2] Daniel Rings, Romy Schachoff, Markus Selmke, Frank Cichos, and Klaus Kroy. Hot brownian motion. *Phys. Rev. Lett.*, 105:090604, Aug 2010.