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Supplementary Information for
Coupled Oscillation and Spinning of Photothermal Particles in Marangoni Optical Traps

Hyunki Kim[a,1], Subramanian Sundaram[b,c,1], Ji-Hwan Kang[a,d], Nabila Tanjeem[a,e], Todd Emrick[a*], Ryan C. Hayward[a,e*]

[a] Polymer Science and Engineering, University of Massachusetts, Amherst, MA, USA.

[b] Biological Design Center, Boston University, Boston, MA, USA.

[c] Wyss Institute for Biologically Inspired Engineering, Harvard University, MA, USA.

[d] Chemical Engineering, California State University, Long Beach, CA, USA.

[e] Chemical and Biological Engineering, University of Colorado, Boulder, CO, USA

[1] These authors equally contributed.

* Corresponding authors: Ryan C. Hayward, Todd Emrick

Email: ryan.hayward@colorado.edu, tsemrick@mail.pse.umass.edu

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Other supplementary materials for this manuscript include the following:

Movies S1 to S5

Table S1. Model parameters

C_p	Specific heat capacity (4180 J Kg^{-1} K^{-1})
μ	Dynamic viscosity of water (0.890×10^{-3} N s m^{-2})
γ_T	Temperature coefficient of surface tension, (-0.1379 mN m^{-1} K^{-1})
k_w	Thermal conductivity of water (0.6 W m^{-1} K^{-1})
k_{th}	Thermal conductivity of gel (0.6 W m^{-1} K^{-1})
ρ_w	Density of water (997 Kg m^{-3})
ρ_m	Density of gel (997 Kg m^{-3})
t_d	Thickness of disk (10 μm)
a	Width of disk (300 μm)
h	Convection heat transfer coefficient (W m^{-2} K^{-1}), from eq. (2)
R_{th}	Effective thermal resistance (K W^{-1}), from eq. (3)
C_{th}	Effective thermal capacitance (J K^{-1}), from eq. (4)
α	Effective optical absorption coefficient of disk
Λ_{drag}	Effective drag scaling factor
Λ_m	Effective mass scaling factor
Λ_{th}	Thermal coupling coefficient

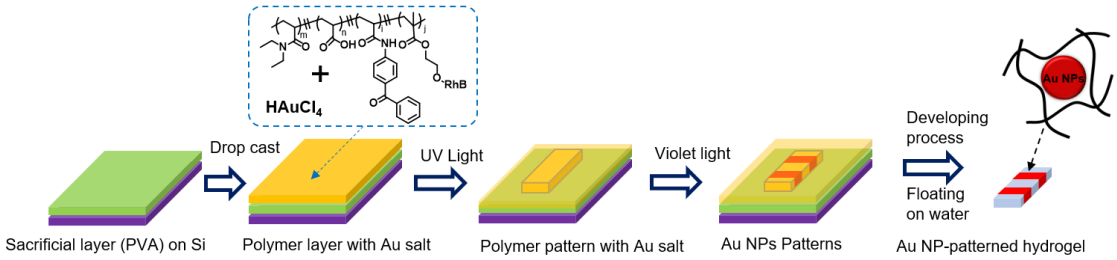

Fig. S1. Schematic diagram for the fabrication of HNDs by (1) drop-casting on a PVA-coated Si wafer, (2) drop-casting polymer-Au salt solution, (3) patterning hydrogels by UV exposure, (4) patterning Au NPs embedded in the hydrogel using high dose of violet light, (5) development and release of HNDs.

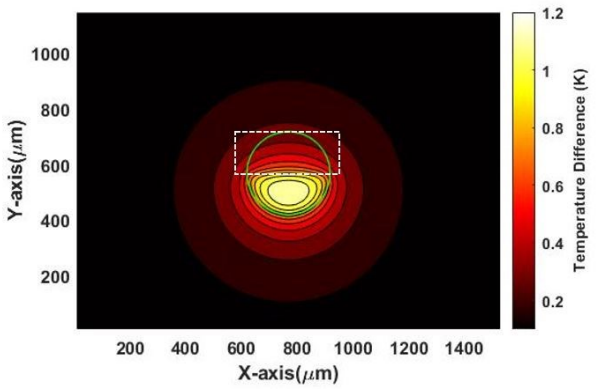

Fig. S2. Estimated temperature profile of a circular HND (green circle) located off-center in an optical trap (white dotted line), with light intensity 3 W/cm^2 using a Green's function approach.

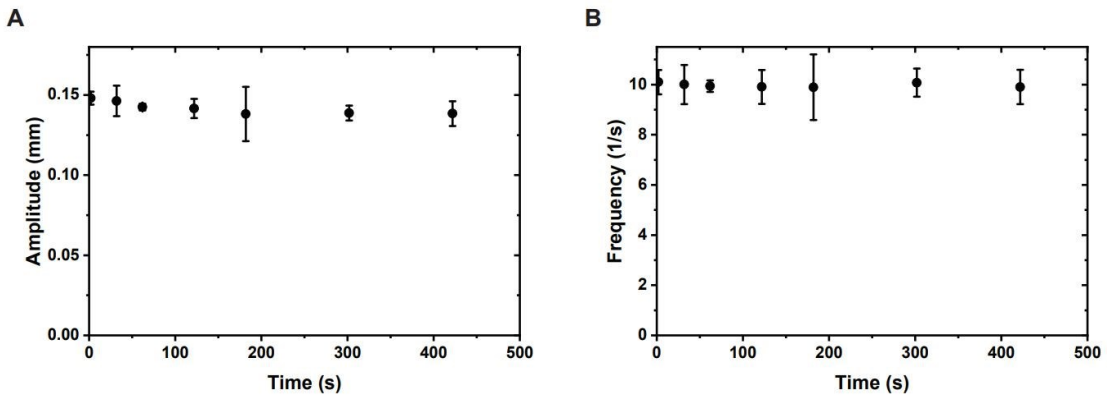

Fig. S3. Amplitude (A) and frequency (B) of a single HND oscillator over a period of 400 s (trap height : 140 µm, light intensity: 2.5 W/cm^2).

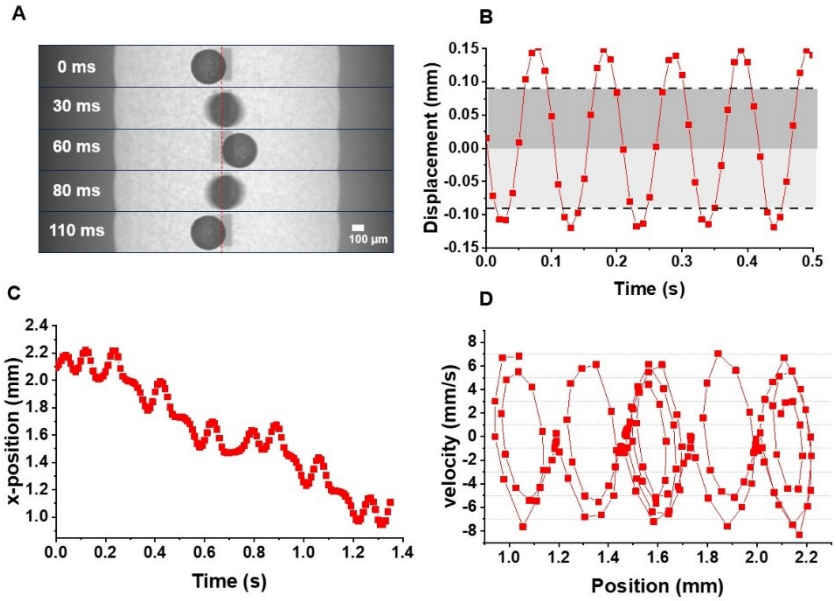

Fig. S4. (A) Time-lapse images of oscillating HNDs under asymmetric illumination. (B) Oscillatory motion of HNDs under asymmetric trap patterns, providing off-centered oscillatory motion. (C) Position vs. time plot of trap-hopping HNDs under multiple asymmetric traps as shown in Fig. 1C. (D) Velocity vs. position plot of trap-hopping HNDs from Fig. 1C.

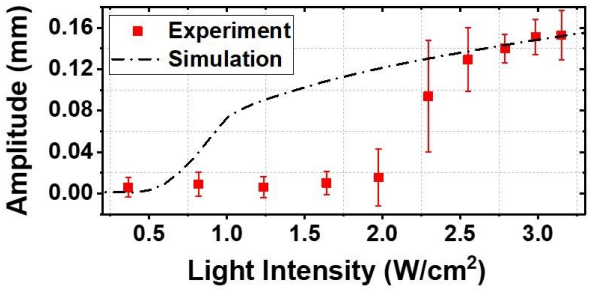

Fig. S5. Comparison of HND oscillation amplitudes seen in simulation and experiment under varying light intensity, with a trap height of 140 μm.

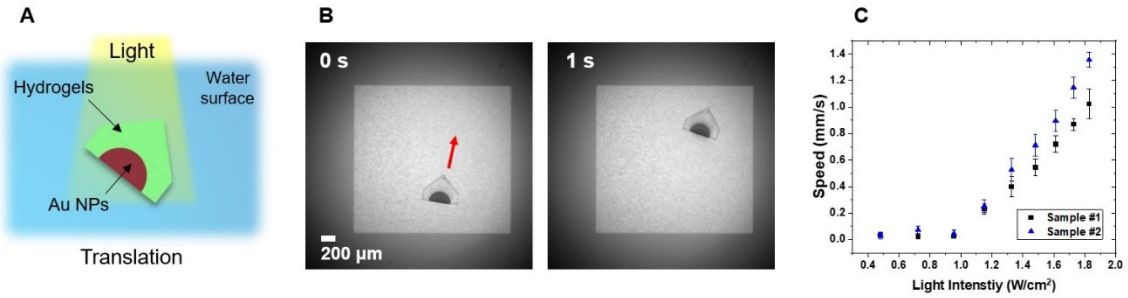

Fig. S6. (**A**) Schematic diagram of HNDs exposed to light, showing unidirectional translation. (**B**) Time-lapse images of HNDs under light. (**C**) Speed of the translational motion vs. light intensity of the HNDs with bottom edge length of 540 μm.

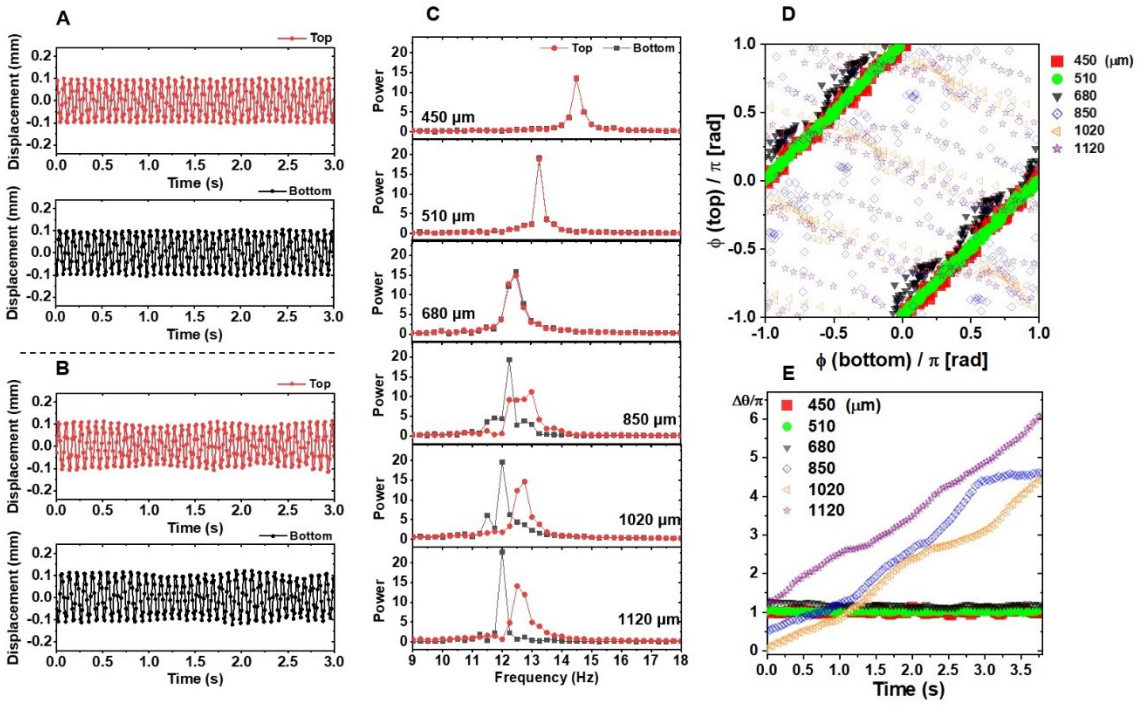

Fig. S7. Displacements of 2 coupled HNDs over time with center-to-center trap separation distance of 510 μm (**A**) and 850 μm (**B**). (**C**) Fast-Fourier transforms of the HND oscillations with respect to trap separation distances. (**D**) Phase plots for different trap separation distances. (**E**) Cumulative phase shift between the 2 HNDs over time, for various separation distances.

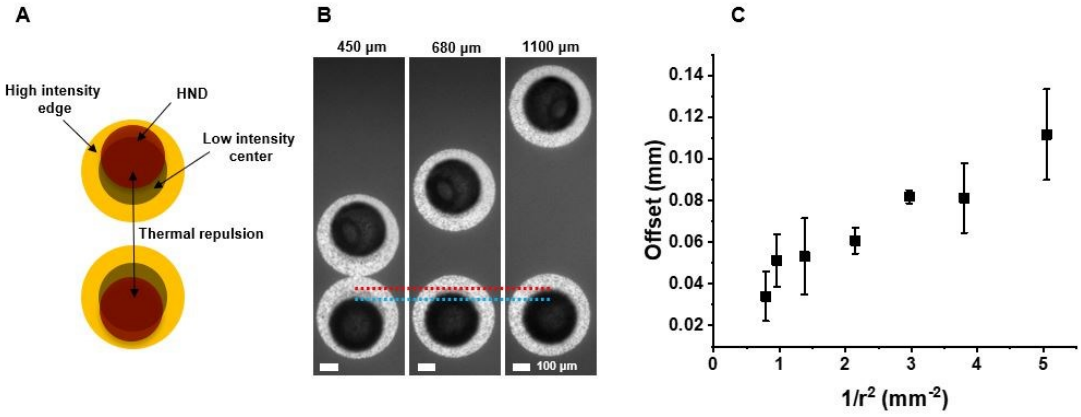

Fig. S8. (A) Schematic of the experiment to measure Marangoni repulsion between HNDs in non-oscillatory traps. (**B**) Images of trapped HNDs with center-to-center trap spacings indicated. (**C**) Measured offsets of HNDs from the trap centers plotted vs. $1/r^2$, where r is trap separation distance; for the proposed Marangoni repulsion and a trapping force that is linear in offset, the data should follow a straight line.

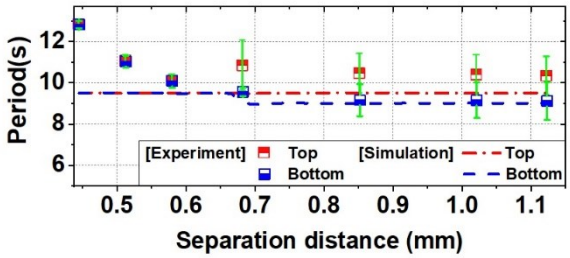

Fig. S9. Comparison between simulation and experiment for frequencies of the HNDs under varying separation distances.

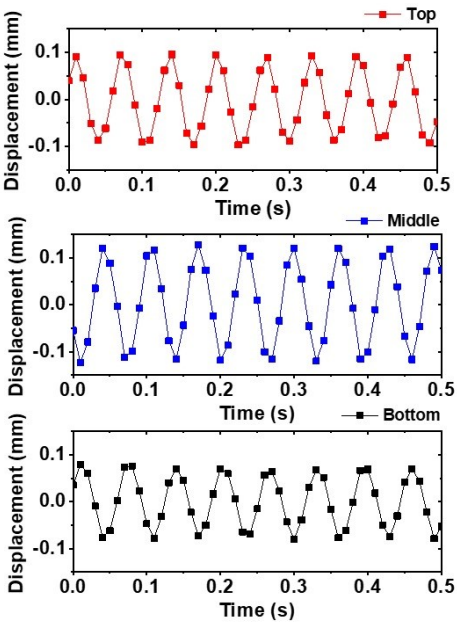

Fig. S10. Tracked displacements vs time for the three HNDs under the light pattern described in Fig. 3A.

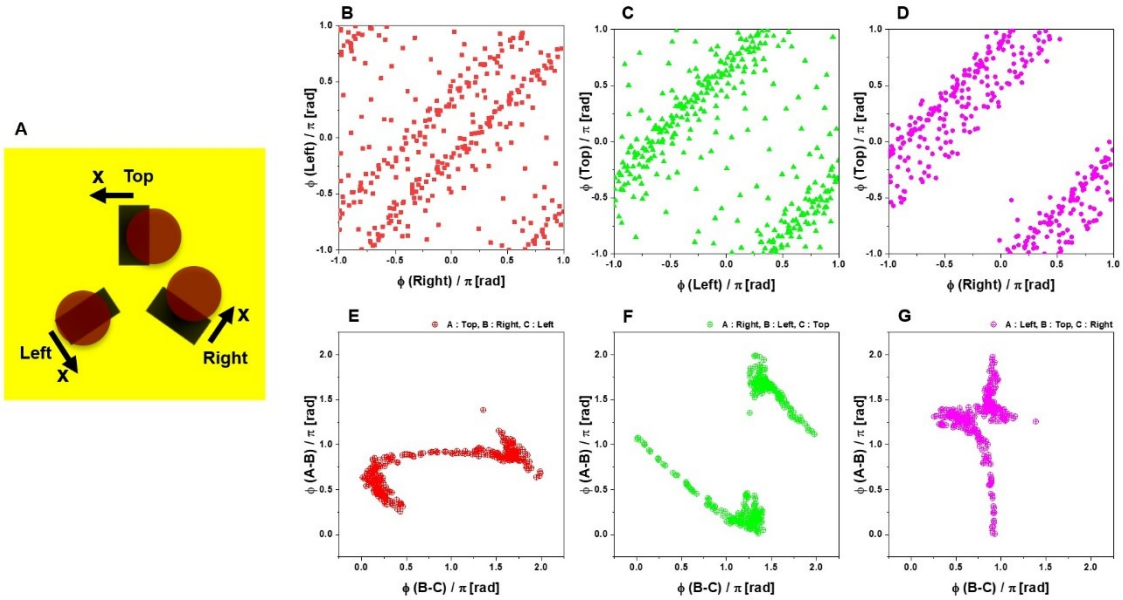

Fig. S11. (**A**) Spatial coordinates for the 3-HND ring oscillator pattern. (**B-D**) Phase plots for the HND pairs described in Fig. 3C. (**E-G**) Phase difference maps for HND pairs in the 3-HND ring network.

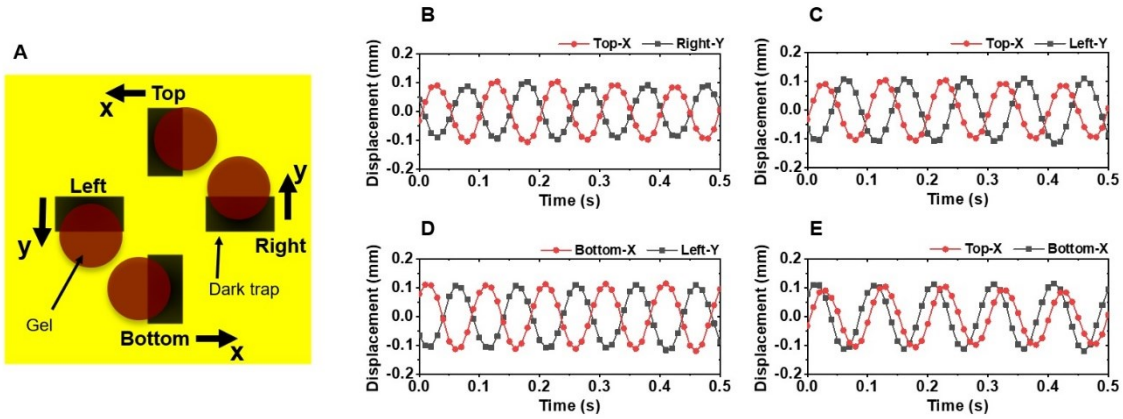

Fig. S12. (**A**) Spatial coordinates for the 4-HND ring pattern. (**B-E**) Temporal displacements of the four HNDs under the light pattern described in Fig. 3D.

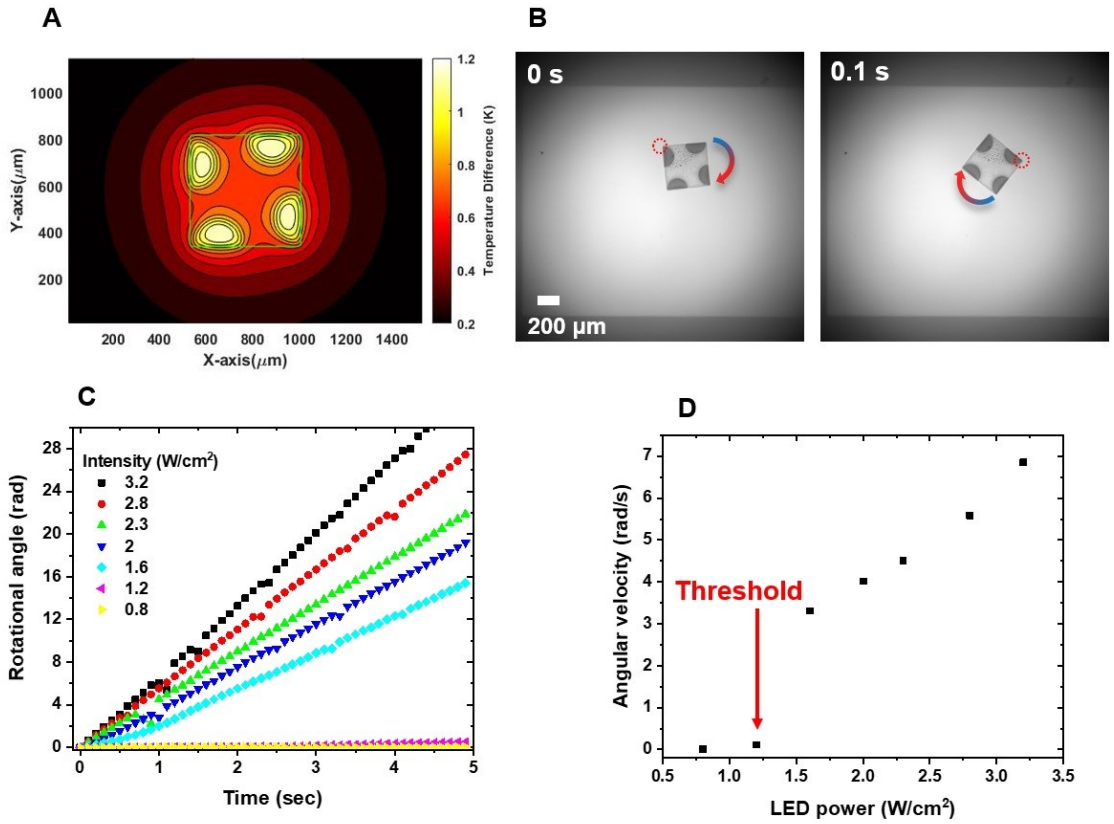

Fig. S13. (**A**) Estimated temperature profile around an HND spinner, subjected to uniform light intensity of 3 W/cm^2, using a Green's function approach. Solid green square line indicates the boundary of the gel. (**B**) Time-lapse images showing rotational motion under uniform illumination. (**C**) Variation of the rotational angle over time as a function of light intensity for HND spinners with edge length of 530 µm. (**D**) Angular velocity vs. light intensity for HND spinners. A threshold intensity to achieve spinning is observed, consistent with observations in the case of HND oscillation (Fig. S5) and propulsion (Fig. S6).

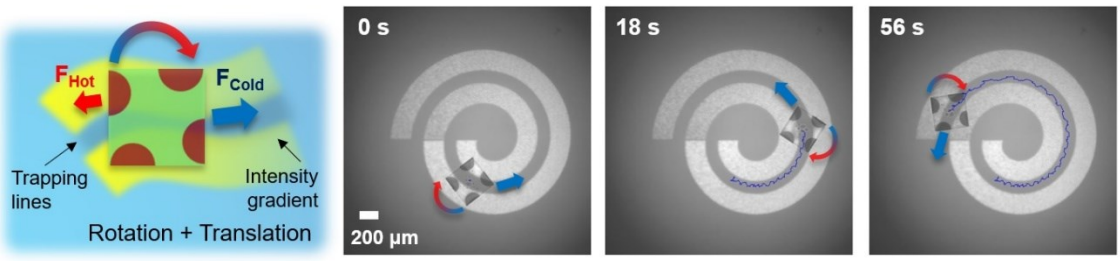

Fig. S14. An HND spinner following a path from high to low light intensity.

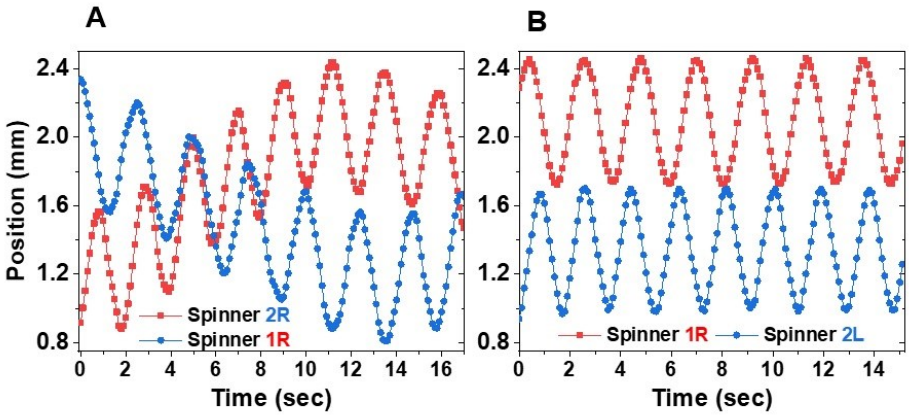

Fig. S15. Positions (y-axis) of the corners of square HNDs with respect to time for the two spinners in the Figure. 4G left (**A**) and right (**B**).

Movie S1 (separate file). Oscillatory motion of single HND under optical traps.

Movie S2 (separate file). 1D coupled oscillatory motion of HNDs.

Movie S3 (separate file). 2D coupled oscillatory motion of HNDs.

Movie S4 (separate file). Spinning and translational motions of gold nanoparticle patterned HNDs.

Movie S5 (separate file). Coupling of HND spinners under optical confinement.