

## Supporting Information

# Probing Spatiotemporal Stability of Optical Matter by Polarization Modulation

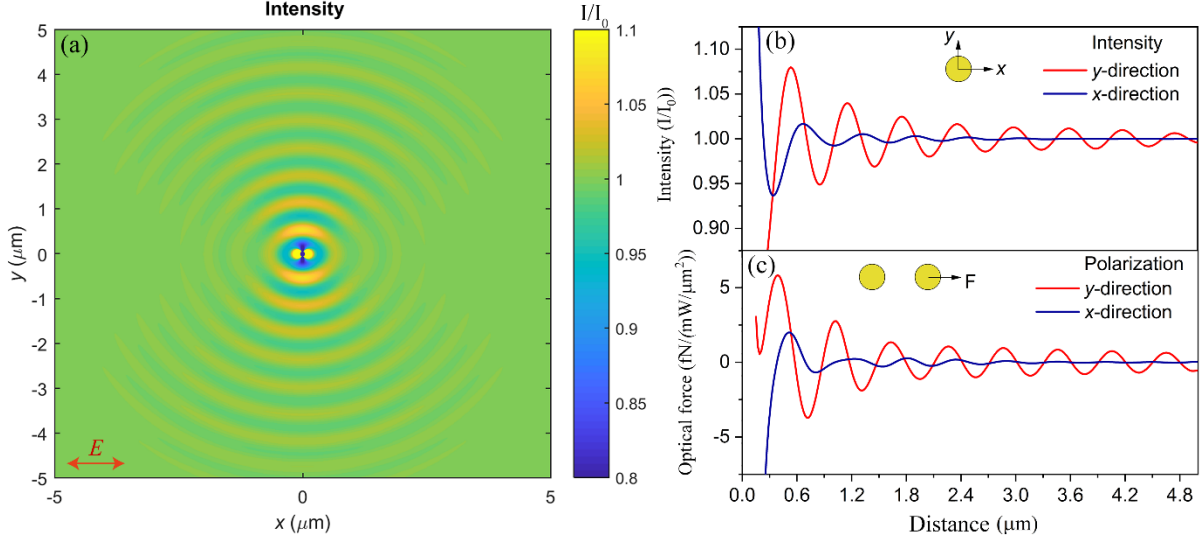
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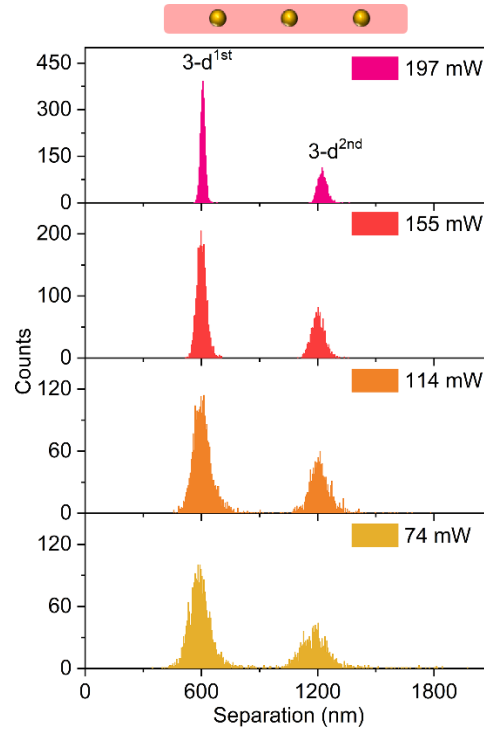
### Videos

**Video S1.** A montage of eight independent clips showing single to eight gold nanoparticles trapped by an optical line with 0.5 Hz polarization modulation.

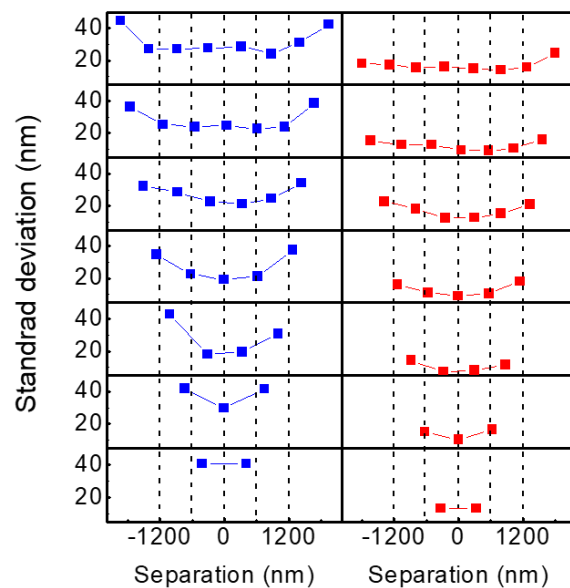
**Video S2.** A montage of seven independent clips showing optical binding of five Au nanoparticles confined by an optical line with different polarization modulation frequencies.



**Figure S1.** Polarization-dependent optical binding interactions. (a) Calculated intensity distribution of electric field around a Au nanoparticle (dia. 100 nm, located at the origin) immersed in water and illuminated by a plane wave of  $\lambda = 800$  nm. The beam is incident along the  $z$ -direction with light polarization along the  $x$ -direction. The spatial distribution of the scattering field resembles the electric field emitted by an oscillating dipole, where in the near field the nanoparticle emits strongly along the polarization direction (i.e.,  $x$ -direction), but in the far field the emission (scattering) is stronger perpendicular to the polarization direction (i.e.,  $y$ -direction). Interference between the incident and scattered light can be clearly seen along the  $y$ -direction; it also occurs along the  $x$ -direction, but the field decays more quickly. (b) Corresponding intensity distribution along the  $x$ - and  $y$ -directions originated from the nanoparticle center. (c) Optical binding forces between two Au nanoparticles when the polarization is parallel or perpendicular to the axial direction. The similarity of curves in (b) and (c) clearly reveals the correlation between the polarization-dependent scattering and optical binding interactions, and shows that the induced dipole radiation plays an important role in the different optical binding interactions for polarization along and perpendicular to the chain direction.



**Figure S2.** Power-dependent optical binding of a chain with three gold nanospheres. The polarization is perpendicular to the chain direction. As the laser power increases, the distribution of particle separation becomes narrower, indicating stronger optical binding interactions.



**Figure S3.** Standard deviations of two to eight particles under different optical polarization. The blue data points came from polarization direction parallel to the chain orientation and the red data points came from perpendicular polarization direction. However, when the particle number is more than five, the optical stability decreases. A possible reason is that compressive forces from phase gradients become significant in longer chains,<sup>1</sup> so particles are shifted away from their equilibrium optical binding positions, making the chains less stable.

### Thermal energy for laser illuminated Au nanoparticles

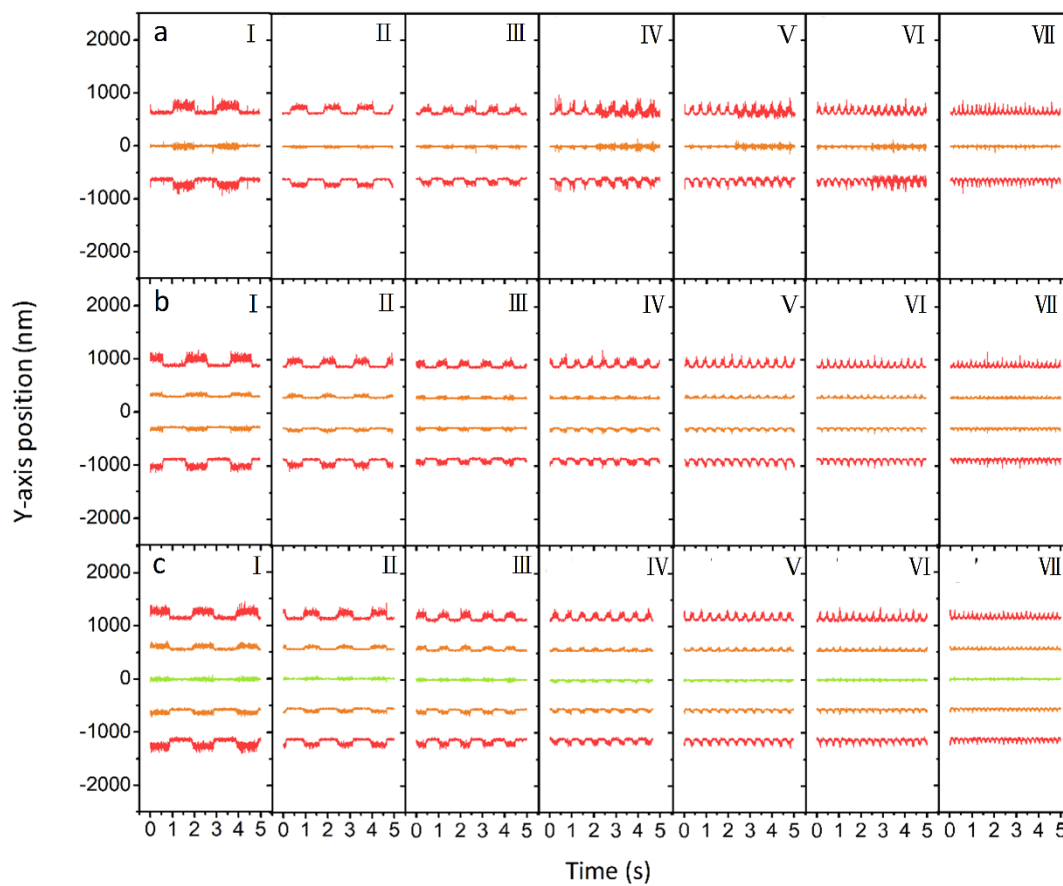
Here we estimate the average thermal energy for the nanoparticles in a one-dimensional chain. The optical line trap can be considered as an elliptical Gaussian beam with an average intensity<sup>2</sup>

$$I = P/(\pi r_x r_y), \quad (1)$$

where  $P$  is the laser power (190 mW),  $r_x$  (0.25  $\mu\text{m}$ ) and  $r_y$  (3  $\mu\text{m}$ ) are the radii along the short and long axes at which the field intensities drop to  $1/e^2$  of the maximum intensity at the trap center. The temperature increase at the surface of a nanosphere follows the relation<sup>3</sup>

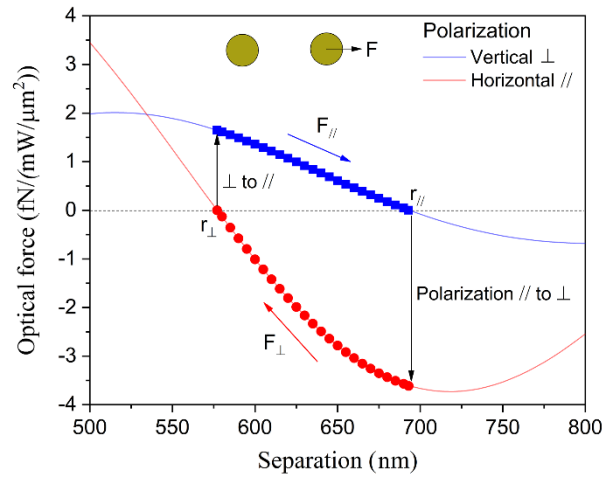
$$\Delta T = \sigma_{\text{abs}} I / (4\pi R \kappa_{\text{water}}), \quad (2)$$

where  $\sigma_{\text{abs}}$  is the absorption cross-section illuminated by  $\lambda_{\text{vacuum}} = 800$  nm laser in water (583  $\text{nm}^2$ ),  $I$  is the average laser intensity ( $8.1 \times 10^{10}$   $\text{W/m}^2$ ),  $R$  is the particle radius (50 nm), and  $\kappa_{\text{water}}$  is the thermal conductivity of water (0.6  $\text{W/(mK)}$ ). Consequently, the increased temperature at the surface of the gold nanoparticles is  $\sim 125$  K, and the average thermal energy for each Au nanoparticle in a one-dimensional chain is  $\sim 0.7 k_B T_0$ , where  $T_0 = 300$  K.



**Figure S4.** Trajectories of (a) three, (b) four and (c) five particles in line traps with different polarization modulation frequency. Panels I-VII correspond to modulation frequency of 0.5, 0.67, 1, 1.67, 2.5, 3.3 and 5 Hz, respectively.

## Timescales of optically driven motion and Brownian motion



**Figure S5.** Driven forces generated by the polarization modulation. The curves are calculated optical binding forces between two Au nanospheres under two polarization directions.  $F_{\perp}$  and  $F_{//}$  (marked by square and circle symbols) are the optical forces driving a nanosphere to the corresponding equilibrium separations at  $r_{\perp}$  and  $r_{//}$  upon polarization modulation.

The nanoparticle motion during the periodic modulation of polarization is mainly determined by the interparticle optical force and thermal fluctuation, where the former dominates during the polarization switching and the latter dominates when the nanoparticles locate at their equilibrium positions. Here we estimate the timescales of nanoparticle motion during the two processes. Figure S5 shows the driven forces on a Au nanoparticle when the polarization is switched between vertical and horizontal directions for two Au nanoparticles oriented horizontally. Considering a laser intensity of  $81 \text{ mW}/\mu\text{m}^2$ , the average driven force from the equilibrium separation  $r_{\perp}$  (577 nm) to  $r_{//}$  (693 nm) is  $F_{//} \approx 68 \text{ fN}$ , while that from  $r_{//}$  to  $r_{\perp}$  is  $F_{\perp} \approx -145 \text{ fN}$ . Assuming a uniform motion of a Au nanoparticle in a Stokes flow, the viscous drag force  $F_d = 6\pi\mu Rv$  balances the optical force from the fluctuation–dissipation relation,<sup>4</sup> so the velocity is given by

$$v = F_d / (6\pi\eta R) = F_{opt} / (6\pi\eta R), \quad (3)$$

where  $\eta$  is the dynamic viscosity of water and  $R$  is the radius of the nanoparticle. This formula gives an average velocity of  $81 \mu\text{m/s}$  under  $F_{//}$  and  $-173 \mu\text{m/s}$  under  $F_{\perp}$ , resulting in a transition time of 0.7 ms and 0.3 ms, respectively, for a distance of 58 nm (half the distance between the

two optical binding positions since the two nanoparticles move together).

When the nanoparticles locate at the optical binding positions, they can still diffuse inside the optical binding potential wells due to thermal fluctuations. The diffusion coefficient  $D$  of a spherical particle in a liquid is given by the Stokes-Einstein formula<sup>5</sup>

$$D = k_B T / (6\pi\eta R). \quad (4)$$

Under the thermal energy of  $0.7k_B T_0$ , we can get a diffusion coefficient of  $3.4 \times 10^{-12} \text{ m}^2/\text{s}$ . The time  $t$  for a Brownian particle diffusing a distance  $x$  can be estimated by<sup>6</sup>

$$t = x^2 / (2D), \quad (5)$$

which gives a time of 0.4 ms for a typical fluctuation distance of 50 nm (from Figure 5 in the main text).

#### References:

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