

## Correlated Dynamic Arrangement of Gold Nanoparticles in Laser Trapping at Glass/Solution Interface

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We are extending a systematic study on “Optically Evolved Assembly Formation in Laser Trapping” where optical potential expands from the focus to the outside of a few ten micrometer through scattering and propagation of the trapping laser. For gold nanoparticles (NPs) at glass/solution interface, our group demonstrated for the first time that the trapping evolves dynamically showing two groups of fluctuating NPs. We named the behavior as laser trapping and swarming<sup>[1]</sup>.

Here we report single-particle level laser trapping experiment to understand how gold NPs are arranged step-by-step outside the laser focus by controlling the precise number of particles. Single particle tracking analysis enables us to monitor the growing process of the assembly and to consider interparticle interaction. Interestingly, the fluctuation of gold NPs is correlated with each other especially in the direction perpendicular to laser polarization, forming an arranged structure at the focal spot. With the increase in the NP number from one to a few, they align outside the focus with a specific interparticle distance corresponding to the wavelength of the trapping laser. Although the NPs outside the focus are not irradiated directly, they keep some correlation with the particles inside the focal spot. The correlation coefficient decreases significantly when the particles go farther away from the focus.

The present result indicates that the expansion of optical potential originates from the interference of scattering light of the regularly aligned gold NPs in the focal spot. After the assembly grows large, multi-scattering of the NPs gives rise to the swarming where photothermal effect should be coupled.

[1] Tetsuhiro Kudo, Shang-Jan Yang, Hiroshi Masuhara, **Nano Lett.**, 2018, 18 (9), pp 5846–5853

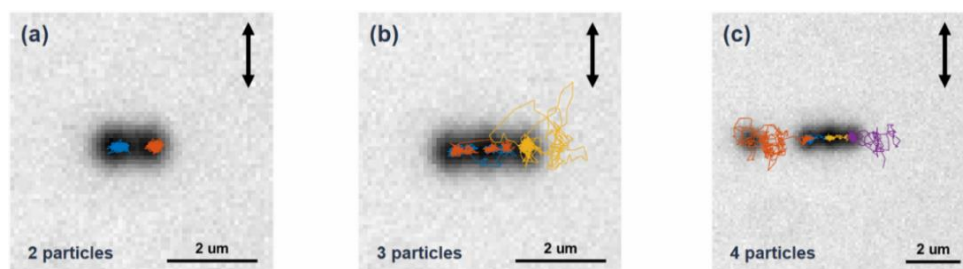


Figure 1. Single particle tracking trace of 400 nm gold NPs with 1064 nm laser trapping. Direction of laser polarization is illustrated as the arrow.