Optical Binding Outside the Focal Spot Leads to Swarming of Gold Nanoparticles

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In the past few decades, optical manipulation of small particles has triggered a wide variety of studies from fundamental research to practical applications. However, these light-matter interactions were essentially limited inside the irradiated area. Previously, our group reported a large swarming assembly of 200 nm gold nanoparticles (Au NPs) formed by laser trapping at the glass/solution interface.[1] The assembly grows on a spatial dimension much larger than the focal spot and displays laser polarization dependence. The Au NPs in the swarms dynamically fluctuate while being spatially confined in a volume, indicating that the optical potential is expanded outside of the irradiated area of the incident laser.

In this work, we applied single particle tracking analysis to investigate the initial stage of the optical trapping and swarming of 400 nm Au NPs, tracing the origin of the optical potential expansion. As a result, correlated motion between particles is not only observed inside but also outside the laser focus. The external NPs show an expanding arc-shaped distribution with a characteristic half-wavelength inter-arc distance (Fig. 1a and 1b). The obtained results strongly support that optical binding can occur outside the irradiated area and extend its network through multiple scattering among more optically-bound NPs, leading to the dumbbell-shaped swarming assembly (Fig. 1c). The generality of this phenomenon is demonstrated by using the NPs of different sizes (200, 300 and 400 nm) and materials (Au, Ag and SiO_2-shelled Au NPs). The present finding provides new insights for optical trapping and optical binding, which will allow us to extend the current understanding of these fields.

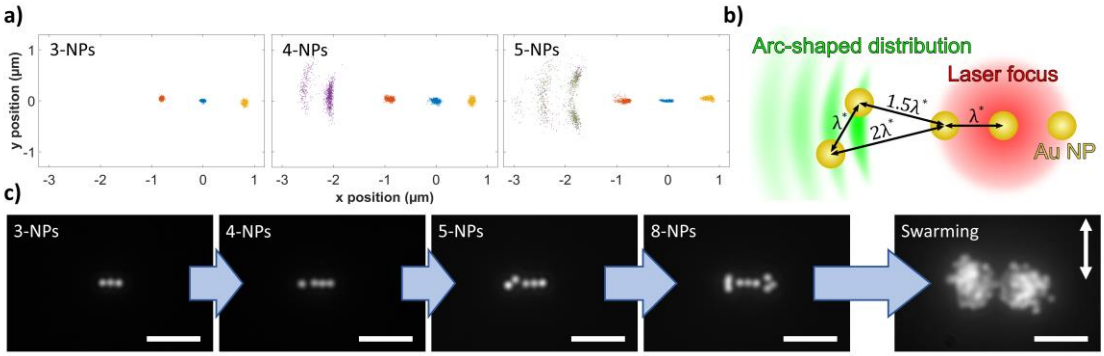

Figure 1. (a) Spatial distribution of 400 nm Au NPs in 3- to 5-NPs system. (b) Illustration showing the observed arc-shaped distribution outside the focus and wavelength-related interparticle distance. (c) Darkfield scattering images of 400 nm Au NPs showing the temporal evolution from 3-NPs system to swarming assembly. The white arrow indicates the direction of linear polarization. The scale bar is 5 μm.

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