Fluorescence Characterization of Strongly Packed Assembly of Silica Nanoparticles Formed under Femtosecond Laser Trapping

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We have first demonstrated a novel laser trapping phenomenon based on femtosecond (fs) laser which induces asymmetrically alternatively directional ejection of polystyrene (PS) nanoparticles (NPs).[1] This trapping and directional ejection has never been observed for cw laser trapping. In our systemic studies, we have proposed that particle assembly formation under fs laser trapping eventually results in the directional ejection. To elucidate our idea, we take advantage of fluorescence spectral measurement to characterize the formation of particle assembly during fs laser trapping.

We prepared silica NPs which surfaces are modified by perylene dyes (PDI) in order to study fluorescence spectroscopy. In their fs laser trapping, the directional ejection is observed similar to PS NPs, as shown in Fig(a). We found that the spectral shape is not much changed, showing a spectrum with two clear peaks λ= 550 nm and 590 nm, as shown in Fig(b). Notably, a red emission at λ= 650 nm appeared irregularly during the trapping, where the long wavelength emission of PDI indicates its π-stacked form. The temporal intensity of red emission is unsynchronized with that at the other two wavelengths, as shown in Fig(c). This suggests that a strong intermolecular interaction of PDIs belonging to different NPs are induced upon trapping. Besides, fluorescence spectra under CW laser trapping has also been measured. The directional ejection is never observed and the spectra shape is almost constant without appearing of red emission, which suggests that the trapping force of CW laser is unable to form an assembly during laser trapping.

We consider that, due to extremely high peak power of fs laser, NPs are strongly pressed with each other and packed together forming an assembly. This assembly is growing and receiving repetitive intense optical pressure leading to the directional ejection, which will be discussed.

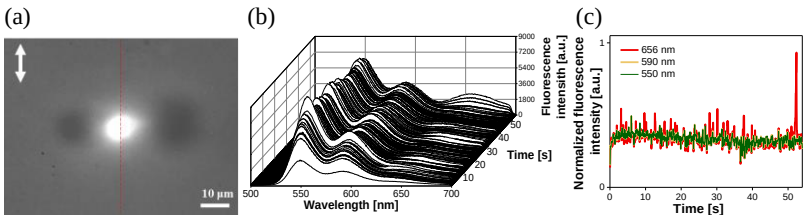

Figure (a) Incorporation of two transmission images showing fs laser trapping and directional ejections. Ejection direction is perpendicular to laser polarization. Two dark areas are ejected NPs. Center bright is multiphoton fluorescence. (b) Temporal multiphoton fluorescence spectra under fs laser trapping. (c) Normalized temporal fluorescence intensity at the peaks of 656 nm, 590 nm and 550 nm. A video frame and a temporal spectrum are accumulated by 33 ms and 0.1 s, respectively.

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1. Usman, A., Chiang, W.-Y. & Masuhara, H. *J. Photochem. Photobiol. A Chem.* **2012**, *234*, 83–90