Electronic Supporting Information.

Plasmonic staining of DNA molecules with photoinduced Ag nanoparticles monitored using dark-field microscopy

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(1) Sensitivity of the current plasmonic imaging examined by Mie theory

Ag nanoparticles (NPs) exhibit strong optical absorption and scattering extending from the near UV to near IR spectral range, which are assigned to plasmon resonance as a consequence of the collective oscillations of the conduction electrons coupled with incident light.^{1,2} The resonance frequency as well as the strength of the plasmon dipole moment strongly depends on their size and shape of NPs.^{1,2} In the current study, we measured plasmon resonance light scattering images (plasmonic images) of Ag NPs with average diameter ~ 30 nm and confirmed them by SEM in Fig. 3. We have examined here the sensitivity of plasmonic imaging as a function of Ag NP size.

We examined a minimum scattering cross-section of detectable Ag NPs in the current system by comparing calculated scattering cross-sections based on Mie theory and plasmonic images.^{1,2} Figure S1a shows plasmon resonance light scattering spectra calculated using single Ag NPs with diameter from 30 to 90 nm based on a reported dielectric function of silver metal.³ Figure S1b1 to b4 shows plasmonic images obtained from purchased Ag NPs (BBinternational, silver colloid, London) with average diameter from 20, 40, 60, and 80 nm. From Fig. 1 we can notice that the minimum diameter of detectable Ag NPs exists between 20 and 40 nm. We confirmed that Ag NPs with diameter of ~30 nm is detectable in the experiment. Thus, we can conclude that a minimum scattering cross-section of detectable Ag NPs is ~5 x 10^{-11} cm².



Figure S1. (a) Plasmon resonance light scattering spectra calculated using single Ag NPs with diameter from 10 to 90 nm. (b1 to b4) Plasmonic images of Ag NPs with average diameter from 80, 60, 40, and 20 nm. White scale bar is $10 \mu m$.

(2) Confirmation of existence of DNA molecules on glass surfaces using DNA-specific fluorescent dye

To confirm the existence of DNA molecules on the glass surfaces, we introduce standard staining method of DNA molecules with fluorescent dye molecules. A portion of 5 μ l aqueous mixture of T4 DNA (0.2 μ g/ μ l), DNA-intercalating fluorescent dye molecule (YOYO-1, 1 mM, Molecular Probes) and 2- mercaptoethanol (4 % (v/v)) sandwiched as mentioned in the present paper was irradiated by a green laser (488 nm, 25 mW) focused by a small lens placed besides a dark-field condenser as shown in Fig. S2a. The laser line is filtered by notch filter.

As shown in Fig. S2b, the fluorescence images exhibit DNA molecules bound at both extremities on the slide glass. The most DNA molecules form line having spatially homogeneous distributions. The DNA molecules size distribution is around 10 μ m, about 5-6 fold smaller than those in the original state (~57 μ m). In the current study, DNA molecules were uncombed by any techniques, e.g. Molecular combing,⁴ so that the DNA molecules may naturally exist coiled state. Thus, we consider that these lines are single DNA molecules on glass surfaces without forming conjugates in the absent of AgNO₃.



Figure S2. (a) Schematic representation of the experimental setup for fluorescence detection. (b) Fluorescence microscopic images of T4 DNA molecules stained with YOYO-1. Black scale bar is 5 μ m.

References for Supporting Information

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