Supporting Information for

Plasmon Switching of Gold Nanoparticles through Thermo-Responsive Terminal Breathing of Surface-Grafted DNA in Hydrated Ionic Liquid

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**g. S1** (a) TEM image of the AuNPs adopted in this study. (b) Extinction spectra of AuNPs before and after DNA functionalization.
**Fig. S2** Typical TEM images of the assembled dsDNA-AuNPs obtained in aqueous solution.
Fig. S3 Time-dependent extinction profile of the DNA-AuNPs in hydrated TBAB at various volume ratios upon addition of com-DNA. The corresponding volume ratio of TBAB is shown with the collected extinction spectra measured at different times. The final AuNP concentration of 4 nM was used in all the experiments.
Fig. S4 (a) Photographs of the DNA-AuNPs in hydrated TBAB at various volume ratios. (b) Time-dependent extinction profile of the DNA-AuNPs in hydrated TBAB. The corresponding volume ratio of TBAB is shown with the collected extinction spectra measured at different times. The final AuNP concentration of 4 nM was used in all the experiments.
Fig. S5 (a) Photographs of the DNA-AuNPs (4 nM) in hydrated TBAB at various volume ratios upon addition of mis-DNA. (b) Time-dependent extinction profile of the DNA-AuNPs in hydrated TBAB upon addition of mis-DNA. The corresponding volume ratio of TBAB is shown with the collected extinction spectra measured at different times. The final AuNP concentration of 4 nM was used in all the experiments.
Fig. S6 Typical TEM images of the dispersion of the dsDNA-AuNPs obtained in 20% TBAB.
**Fig. S7** Effect of IL and salt on the colloidal stability of AuNPs. (a,b) Photographs for bare AuNPs in various hydrated TBAB without (a) and with (b) 550 mM NaCl. (c,d) Photographs for bare AuNPs in various hydrated EMIM-DCA without (c) and with (d) 550 mM NaCl.
Fig. S8 Time-dependent extinction profile of the DNA-AuNPs in hydrated EMIM-DCA at various volume ratios upon introduction of com-DNA. The corresponding volume ratio of EMIM-DCA is shown with the collected extinction spectra measured at different times. The final AuNP concentration of 4 nM was used in all the experiments.
Fig. S9 (a) Photographs of the DNA-AuNPs (4 nM) in hydrated EMIM-DCA at various volume ratios. (b) Time-dependent extinction profile of the DNA-AuNPs in hydrated EMIM-DCA. The corresponding volume ratio of EMIM-DCA is shown with the collected extinction spectra measured at different times. The final concentration of the AuNPs was 4 nM.
Fig. S10 (a) Photographs of the DNA-AuNPs (4 nM) in hydrated EMIM-DCA at various volume ratios upon addition of mis-DNA. (b) Time-dependent extinction profile of the DNA-AuNPs in hydrated EMIM-DCA upon introduction of mis-DNA. The corresponding volume ratio of EMIM-DCA is shown with the collected extinction spectra measured at different times. The final concentration of the AuNPs was 4 nM.
Fig. S11 Ionic strength-dependent optical properties of the DNA-AuNPs. (a,b) Extinction spectra of the DNA-AuNPs in the presence of NaCl of different concentrations (a) and the corresponding plots of their spectral intensities (b). (c,d) Extinction spectra of the DNA-AuNPs in the presence of MgCl₂ of different concentrations (c) and the corresponding plots of their spectral intensities (d).
Fig. S12 Temperature-dependent extinction of hydrated TBAB (1%). The spectral profiles indicate a decrease in its extinction with increasing temperature in the range of 25–40°C.
Fig. S13  (a) Temperature-dependent extinction profiles of the dsDNA-AuNPs in aqueous solution.  (b) Plots of the plasmonic peak positions of the dsDNA-AuNPs during the repeated heating to 40°C and cooling to 25°C.