Supporting Information for

DNA Base Pair-Stacking Crystallization of Gold Colloids

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1. Synthesis of gold nanospheres (Au NSs)

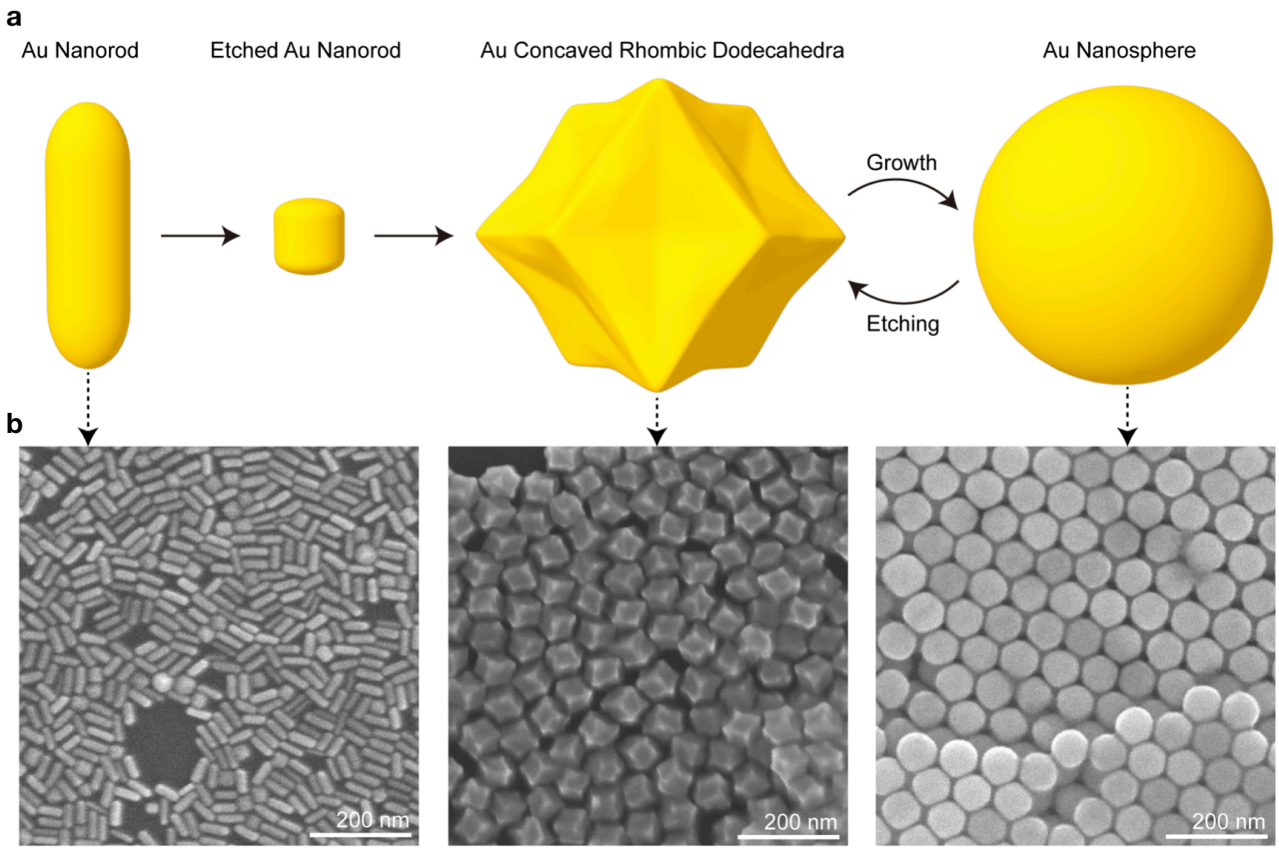

Figure S1. Synthetic route for gold nanospheres (Au NSs). (a) Schematics illustrating the iterative reductive growth and oxidative dissolution process to prepare the highly uniform Au NSs. Growth of Au concaved rhombic dodecahedra and subsequent etching into Au NSs were repeated in cyclic fashion to refine the quality of the Au NSs. (b) Scanning electron microscope (SEM) images of Au nanoparticles correspond to the synthetic route.

2. Glass microfluidic chamber for crystallization of DNA-conjugated Au NSs.

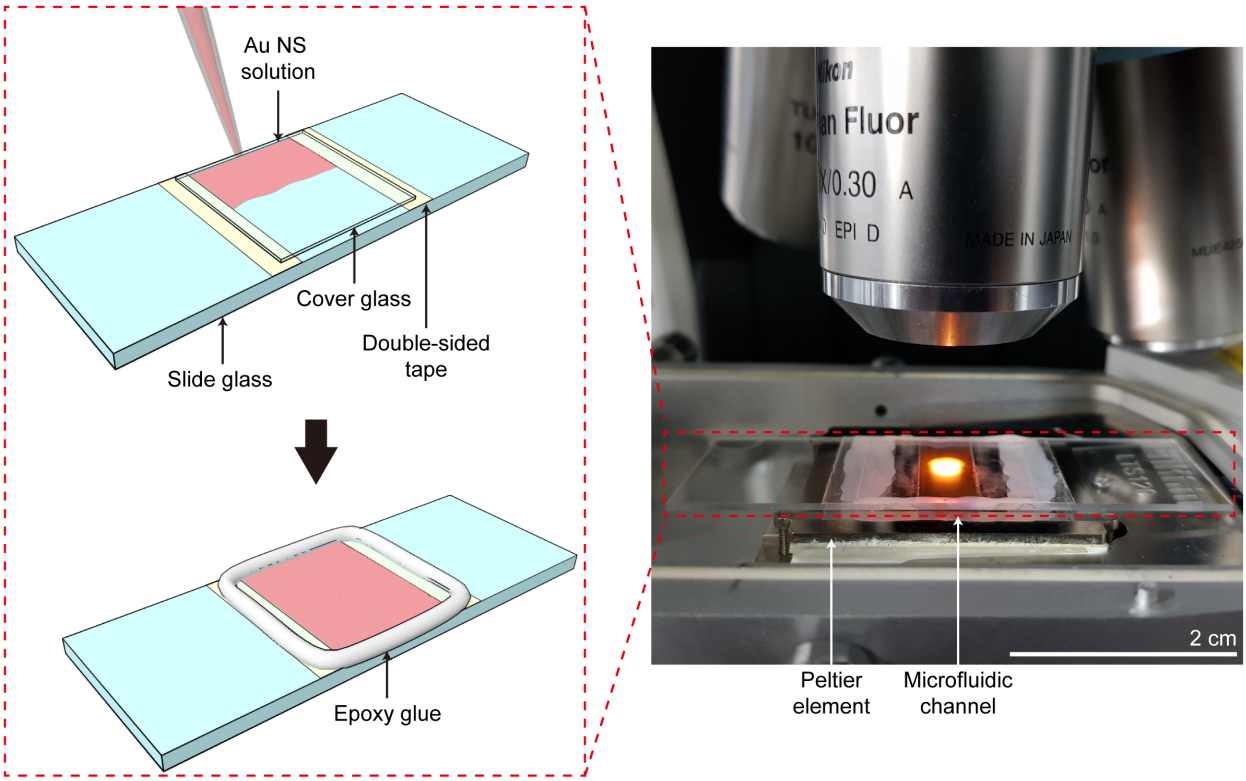

Figure S2. Preparation of glass microfluidic chamber and experiment set up for the assembly of DNA-conjugated Au NSs. Schematics in the left panel illustrate the detailed procedure for the preparation of microfluidic chamber containing DNA-conjugated Au NS suspensions. The chamber was assembled by placing cover glass on top of slide glass taped with double-sided adhesive tape. Then, the Au NSs suspensions were pipetted within the microfluidic chamber and sealed with epoxy glue. Once the epoxy glue is hardened, the chamber was placed on the Peltier element for thermal annealing (right panel).

3. Absorption and scattering cross section of 30 nm and 75 nm Au NSs

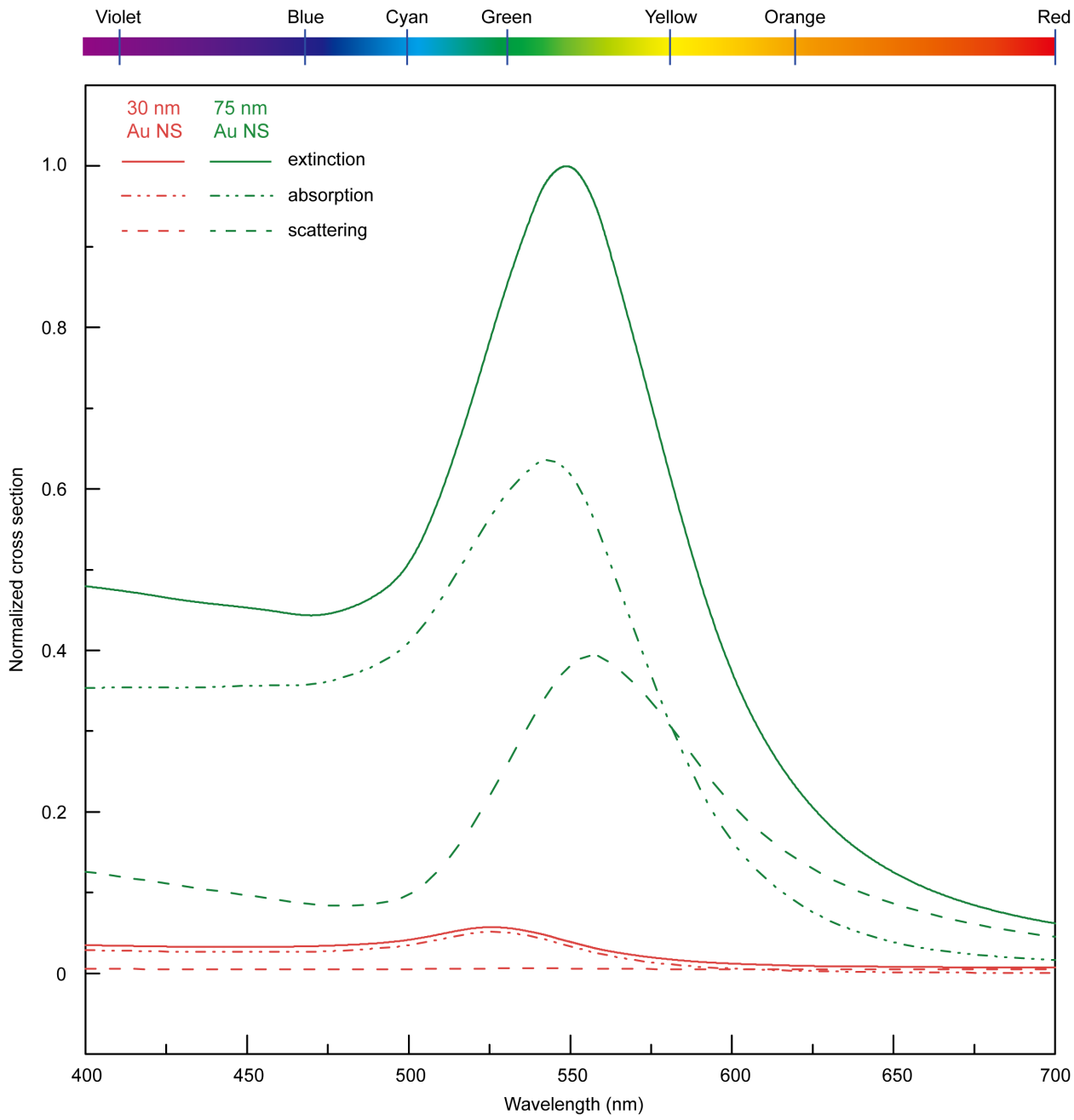

Figure S3. Numerical simulation (using FDTD method) of absorption and scattering spectra of 30 (red lines) and 75 nm (green lines) Au NSs. The solid, dash-dot-dot, and dashed lines correspond to extinction, absorption, and scattering cross section, respectively. All data are normalized by the maximum value of extinction cross section of 75 nm Au NS.

4. Macroscopic view of Au NS crystallization during thermal annealing

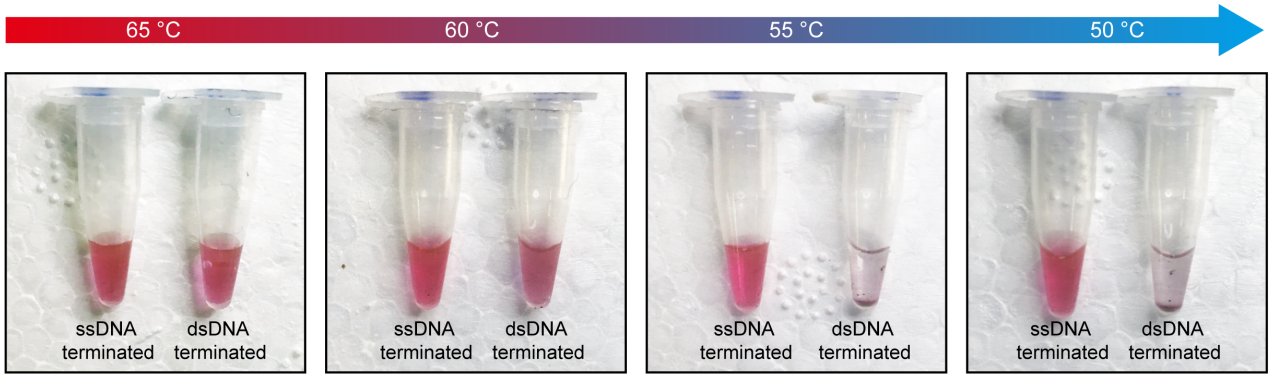

Figure S4. Macroscopic color change of DNA-conjugated Au NS suspensions upon thermal annealing processes (from 65 to 50 °C). The concentration of NaCl was set as 300 mM.

5. Dark-field optical microscope (DFOM) image analysis for profiling melting temperature

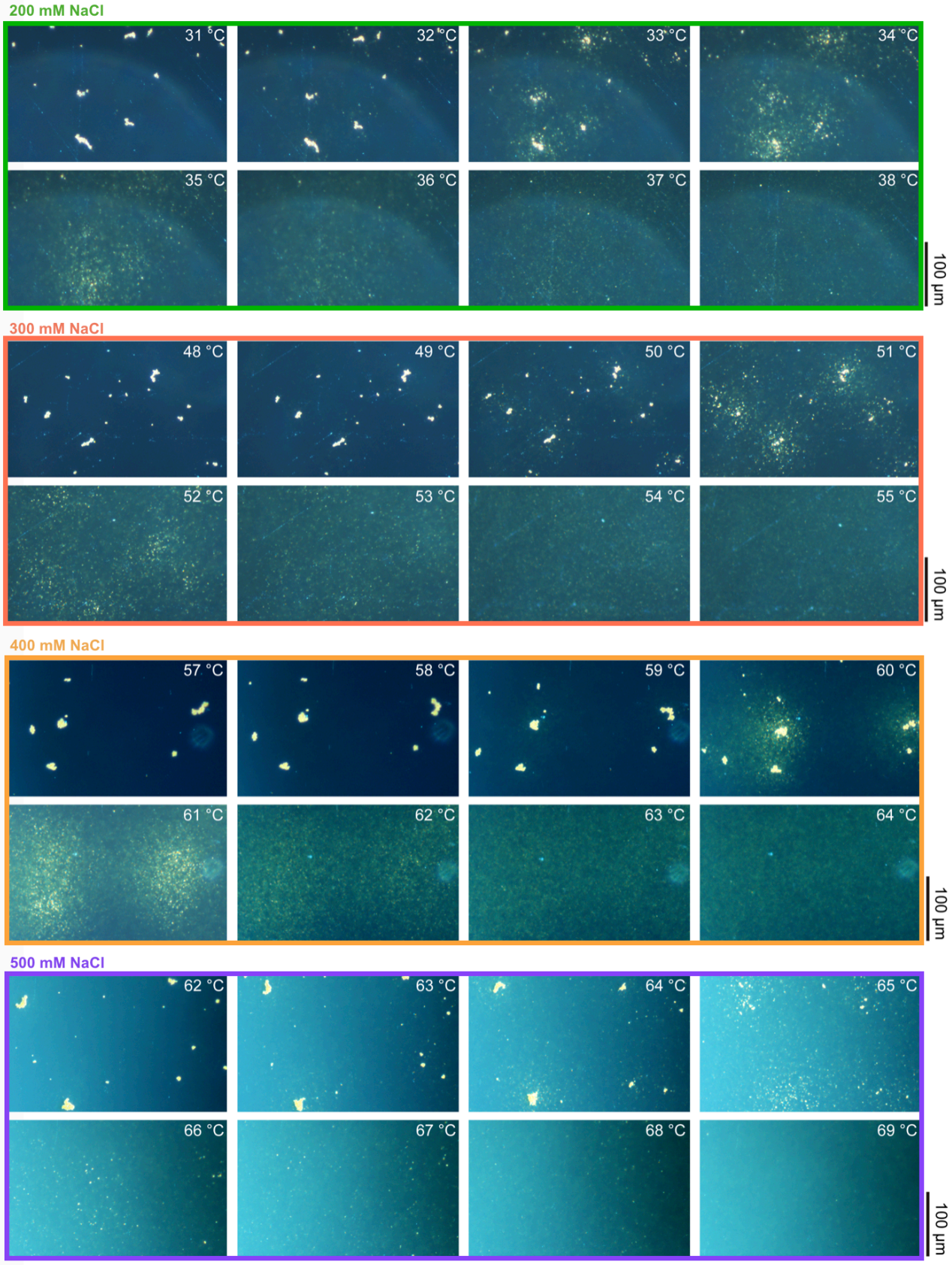

Figure S5. Dark-field optical microscope (DFOM) images of double stranded DNA- (dsDNA-) terminated Au NS suspensions at different annealing step which correspond to the individual dots in Figure 3c of the main manuscript. The concentration of NaCl was varied from 200 to 500 mM.

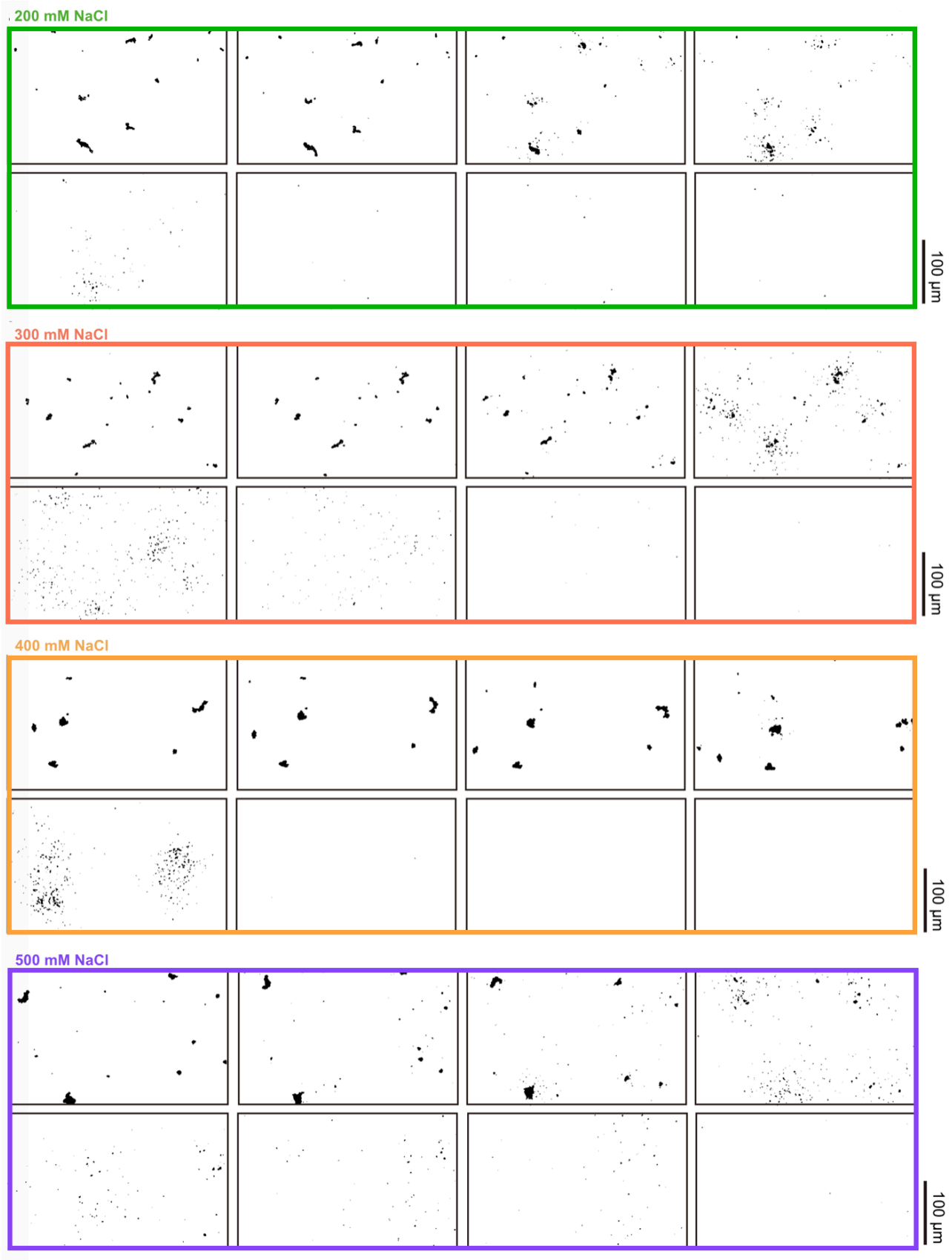

Figure S6. Contrast enhanced monochromatic images converted from DFOM images in Figure S5. Thresholds for the monochromatizating were adjusted based on hue and intensity of the assembled Au NSs crystals so that the dissociated Au NSs are rejected from reconstructed monochromatic images. From the above monochromatic images, the singlet fraction presented in Figure 3d of the main manuscript was quantified.

6. Dissociation temperature of dsDNA used for the blunt-end-interaction

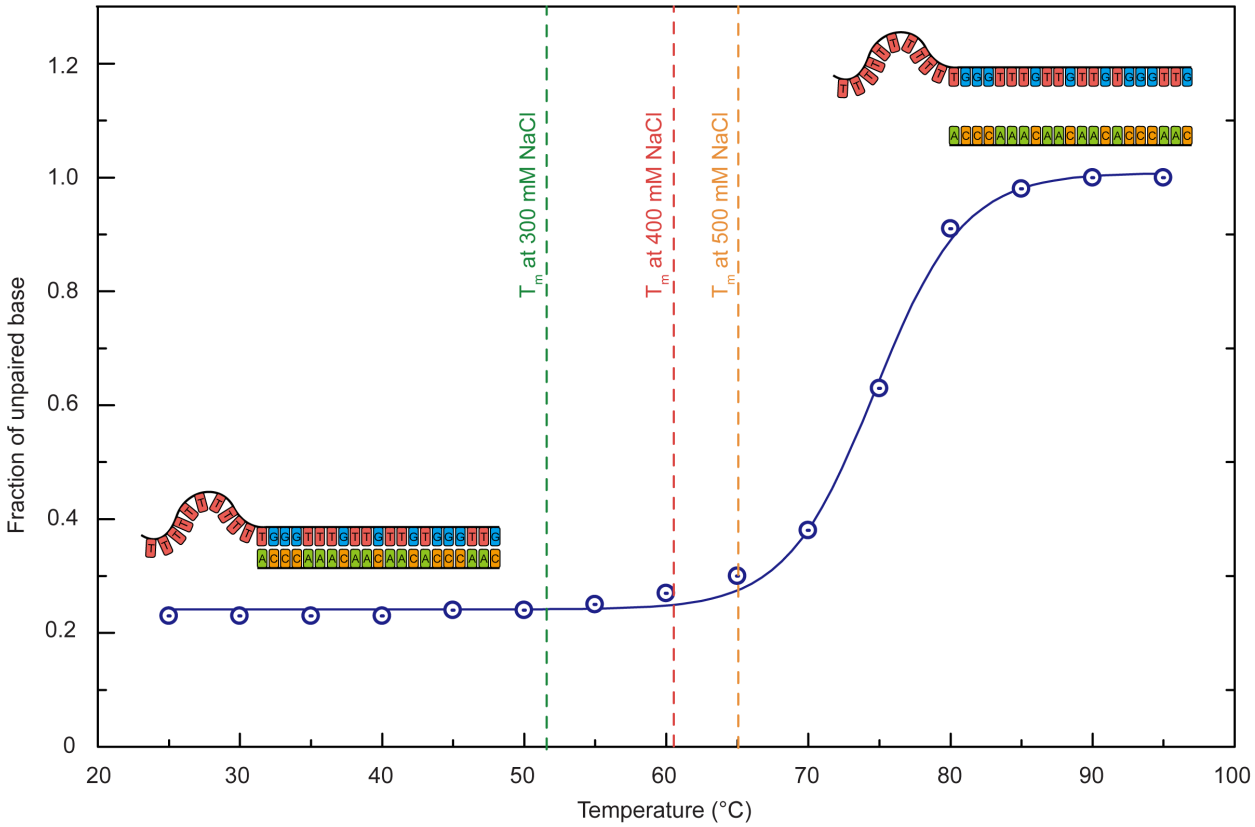

Figure S7. Theoretically predicted (using NUPACK)[1,2] melting profile of dsDNA used in blunt-end stacking crystallization of Au NSs. Blue dots correspond to the fraction of unpaired base on the dsDNA. Melting temperatures (T_m) of dsDNA-conjugated Au NS crystals at different NaCl concentration are also presented for comparison; green, red, and orange dotted lines correspond to T_m at 300, 400, and 500 mM NaCl concentration.

7. Numerical simulation for metallodielectric stopband of Au NS crystal with different space

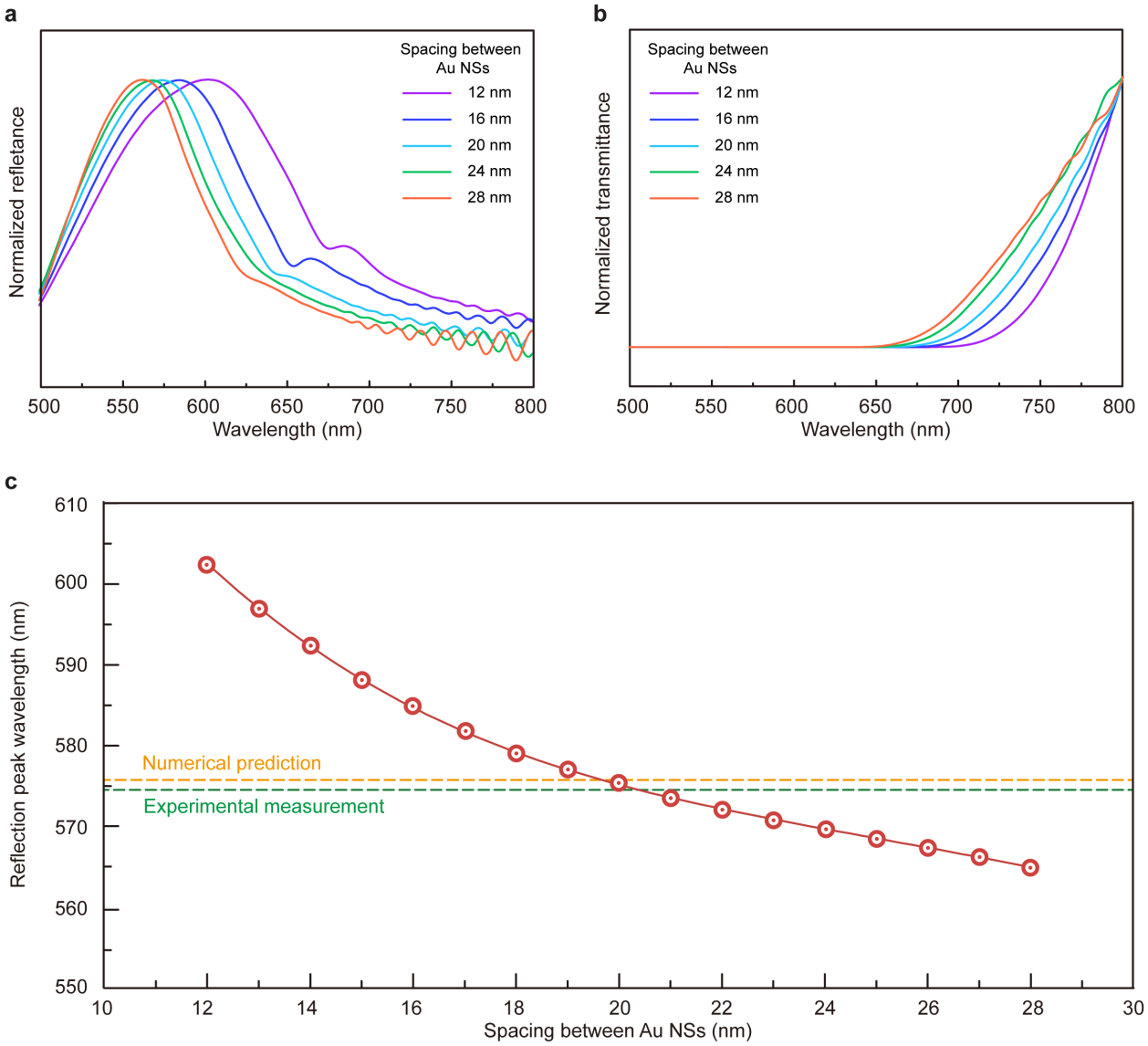

Figure S8. Numerical simulations for reflections and transmissions of Au NS crystal as a function of space between Au NSs: (a) reflection spectra, (b) transmission spectra, and (c) reflection peak wavelength (correspond to transmission minima) of Au NS crystal with different interparticle spacing. Numerically predicted (yellow dotted line) and experimentally measured (green dotted line) reflection peak wavelength, which are obtained from the spectra presented in Figure 5b-c of the main manuscript, are also included for guidance. Herein, 75 nm Au NSs were assumed to be assembled into face-centered-cubic (FCC) crystals as presented in Figure 5a of the main manuscript. The spacing between Au NSs was varied from 12 nm to 28 nm.

References

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