

**Supporting Information**

**Analysis of Metallic Nanoparticle-DNA Assembly Formation in Bulk Solution via  
Localized Surface Plasmon Resonance Shift**

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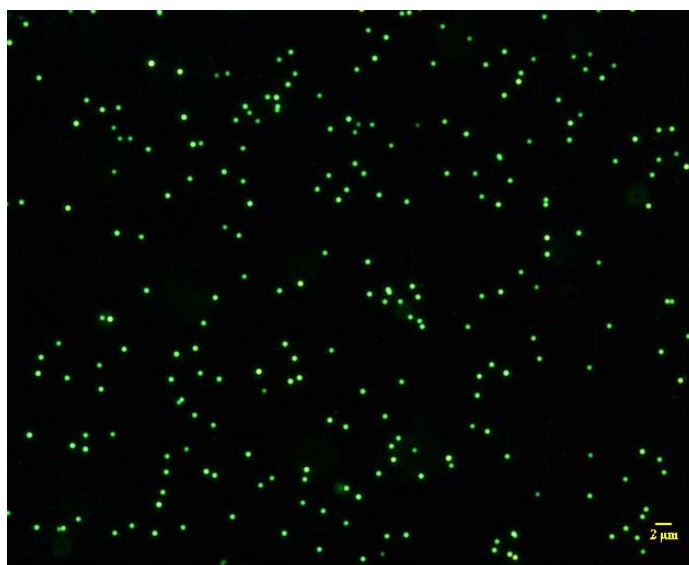


Figure S1: Dark field image of as-synthesized nAu with mean diameter  $51.5\text{nm} \pm 3.69\text{ nm}$ .

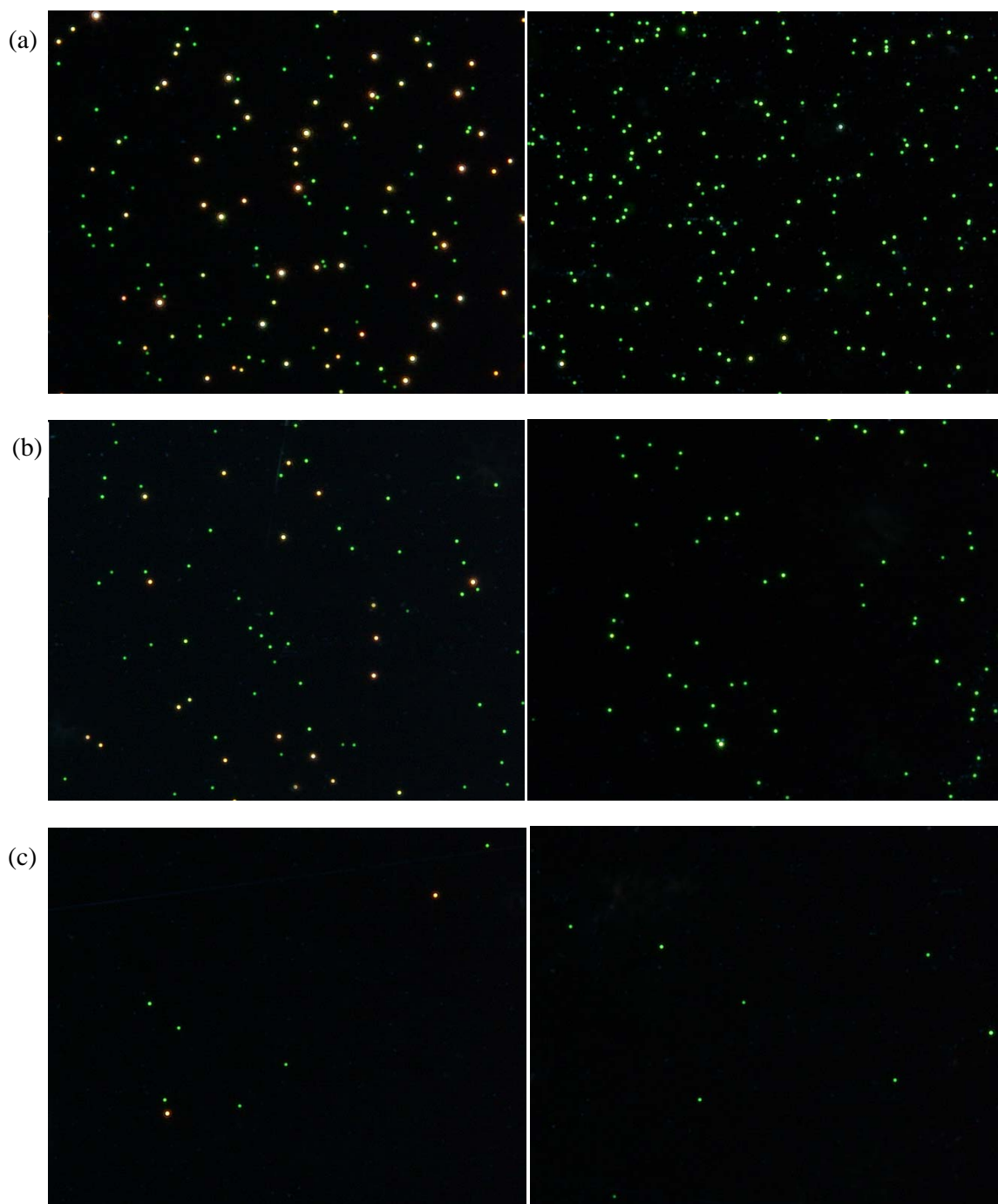


Figure S2. Probe Concentration & Hybridized Duplex Length Study (Scheme 1). Comparison of the dark field micrographs on hybridization percentage between complementary probes nAu-P/nAu-cP<sub>27</sub> at decreasing concentration (a) 400fM, (b) 100fM, and (c) 10fM, recorded after 1h-hybridization. For each set of concentration, left panel constitutes of probes nAu-P & nAu-cP<sub>27</sub> at constant ratio of 1/1; right panel is control comprises only single type of probe. The scattering color of all controls remains more than 95% green even after 24h. Images were taken from the bottom plane of liquid chamber, to which particles settled. Buffer: 140mM Tris, 140mM NaCl, 2mM MgCl<sub>2</sub>.

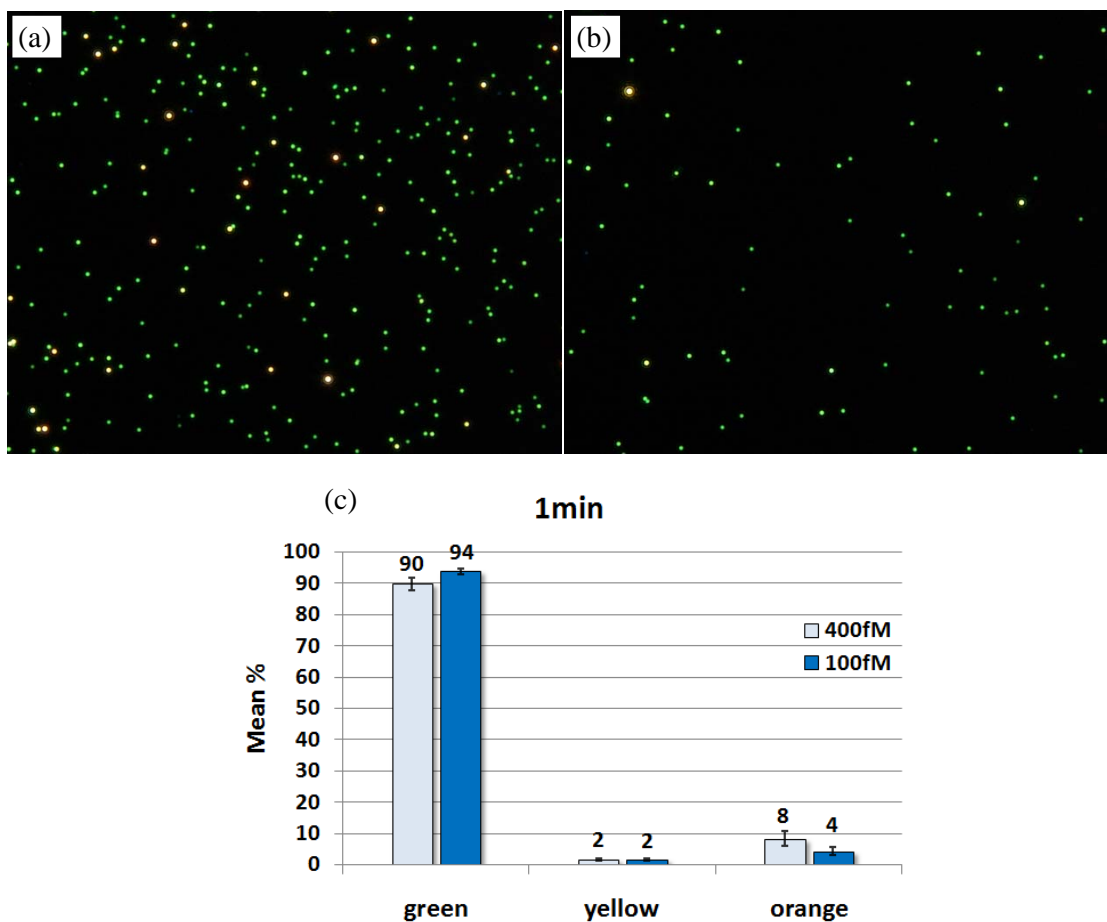


Figure S3. Nanoassemblies formed at 1 min for (a) 400fM (b) 100fM of complementary probes 50nm nAu-P/50nm nAu-cP<sub>27</sub>, both at constant ratio of 1/1, and (c) the corresponding hybridization percentages for 400fM (light blue), and 100fM (blue). Images were taken from the bottom plane of liquid chamber, to which particles settled. Buffer: 140mM Tris, 140mM NaCl, 2mM MgCl<sub>2</sub>.

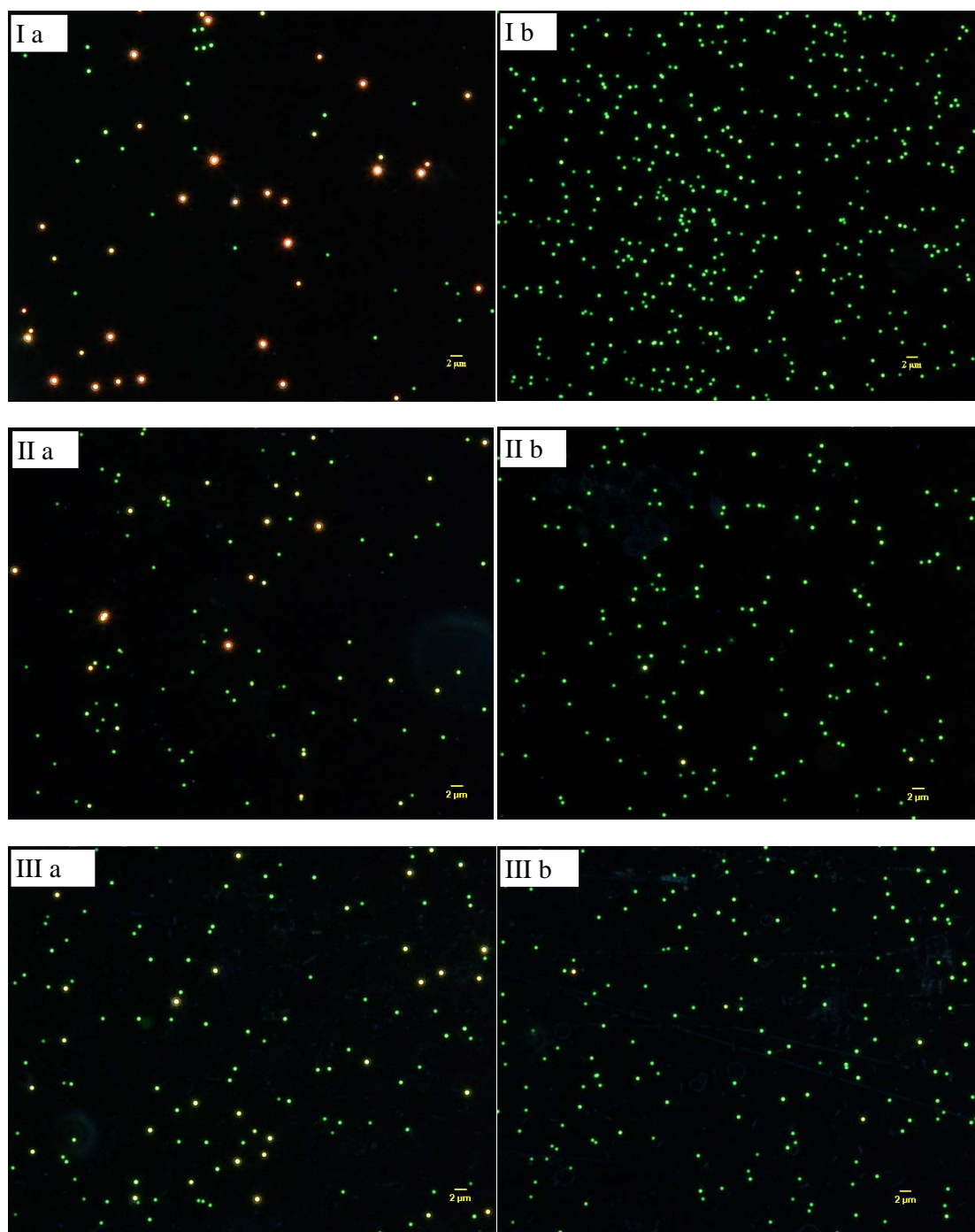


Figure S4: Size Effect Study (Scheme 1 vs. Scheme 2). Comparison of the dark field micrographs on hybridization percentage between complementary probes nAu-P/nAu-cP<sub>27</sub> for I(a) homo-size system with same probe size 50nm/50nm, and hetero-size systems with different probe sizes II(a) 50nm/20nm, III(a) 50nm/10nm, all at constant ratio of 1/1 (800fM each), recorded after 6h-hybridization. The corresponding controls are shown for I(b) homo-size system which comprises single type of 50nm probe at 1.6pM, and hetero-size systems which comprise 800fM 50nm probe & II(b) 800fM 20nm probe, III(b) 800fM 10nm probe, all carrying the same type of ssDNA sequences. Images were taken from the bottom plane of liquid chamber, to which particles settled. Buffer: 140mM Tris, 140mM NaCl, 2mM MgCl<sub>2</sub>.

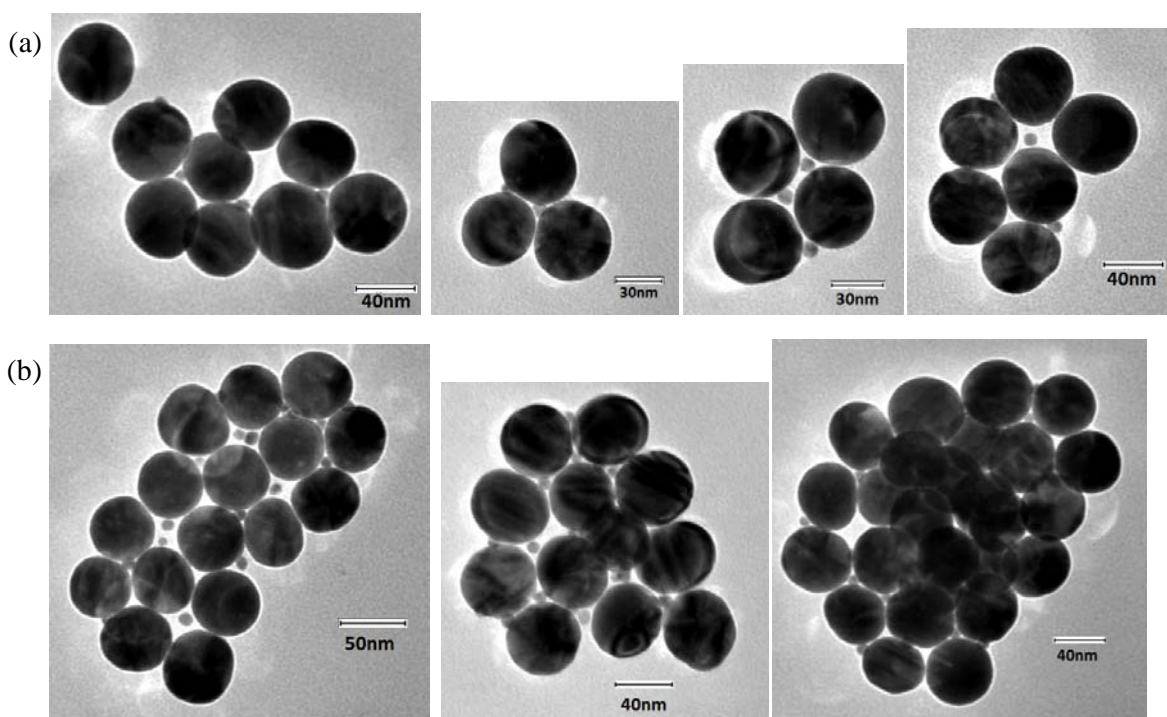


Figure S5: Size Effect Study (Scheme 1 vs. Scheme 2). Comparison of the TEM micrographs on hybridization efficiency between complementary probes nAu-P/nAu-cP<sub>27</sub> for hetero-size system with different probe sizes (50nm/10nm) at molar ratio (a) 1/1 (b) 1/3, recorded after 6h-hybridization in buffer of 140mM Tris, 140mM NaCl, 2mM MgCl<sub>2</sub>. The concentration of 50nm nAu-P is 800fM.

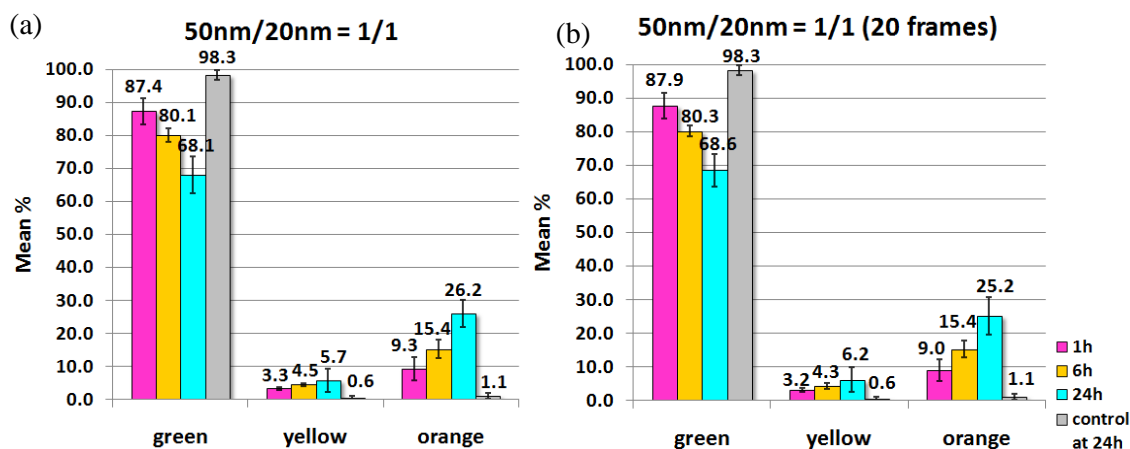


Figure S6: Comparison of the statistical distributions based on (a) 10, and (b) 30 dark field images. The data show the hybridization percentages between nAu-P/nAu-cP<sub>27</sub> for hetero-size system with different probe sizes (50nm/20nm) in Size Effect Study. Ratio of complementary probes: 1/1; concentrations of all species: 800fM. Each bar reported consists of four repeats, two with 50nm nAu-P/20nm nAu-cP<sub>27</sub>, & the other two with 20nm nAu-P/50nm nAu-cP<sub>27</sub>. Images are quantified by counting the hybridized-clusters ('yellow' & 'orange') vs. the individual probes ('green'). Buffer: 140mM Tris, 140mM NaCl, 2mM MgCl<sub>2</sub>.