

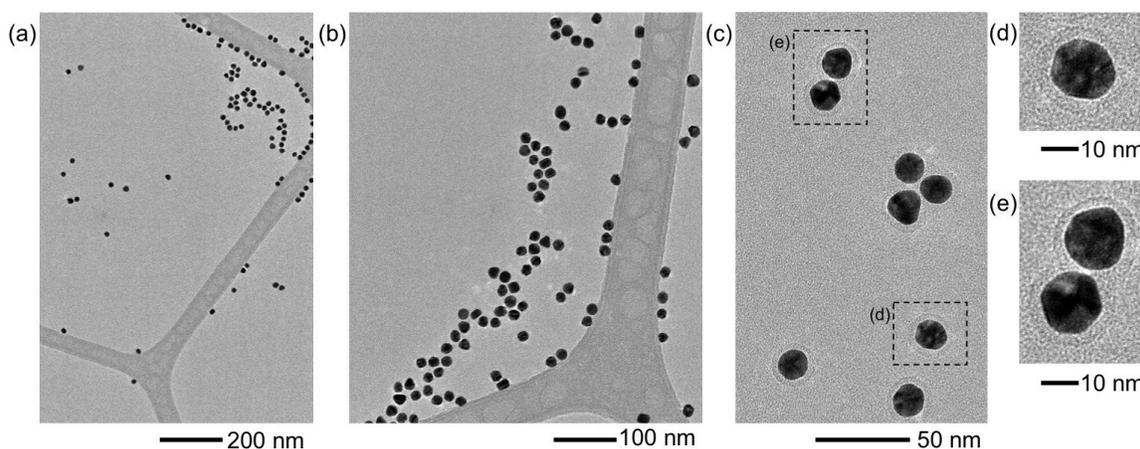
### Supporting Information

## In-Situ Electron Microscopy of the Self-Assembly of Single-Stranded DNA-Functionalized Au Nanoparticles in Aqueous Solution

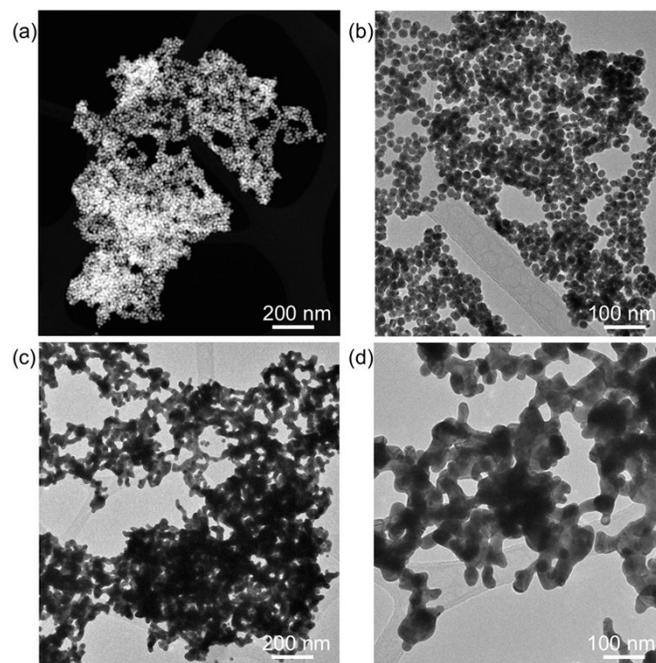
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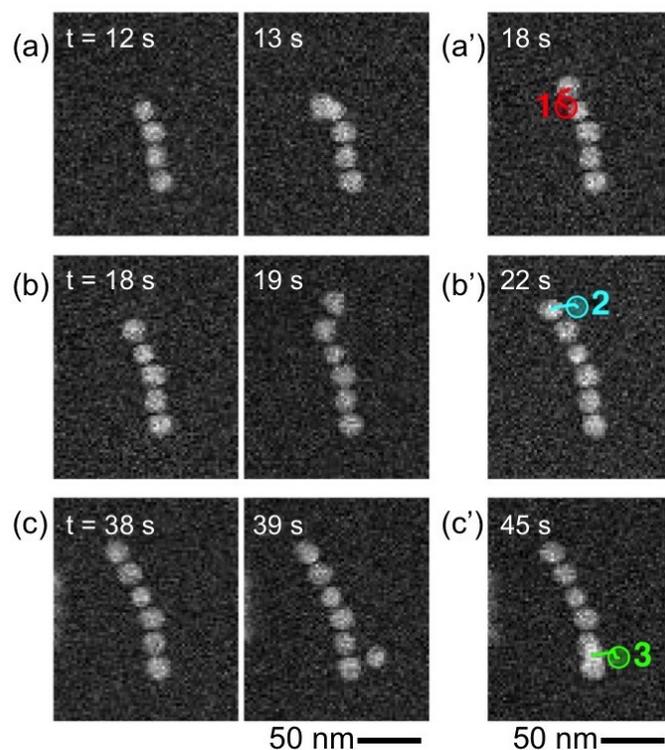
### Supporting Figures



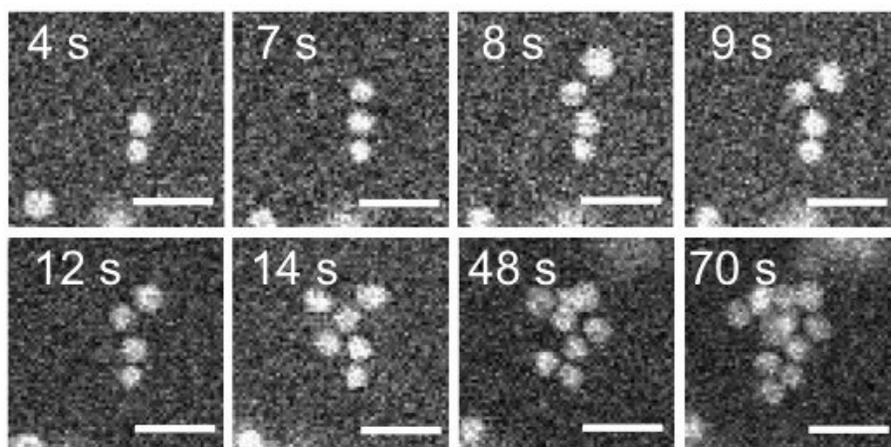
**Figure S1. Assemblies of ssDNA-Au nanoparticle conjugates formed at pH 3.** (a) Small assemblies generated in acidic solution, drop-cast onto a TEM grid and imaged using ex-situ TEM in vacuum. (b) Close-up TEM image of some of the small assemblies. (c) Higher-magnification TEM image, showing the extent of the (dry) DNA shell around the Au nanoparticles. (d), (e) Detail views of the areas marked in (c). The DNA shells are clearly visible as bands of ~4 nm thickness around the Au nanoparticles.



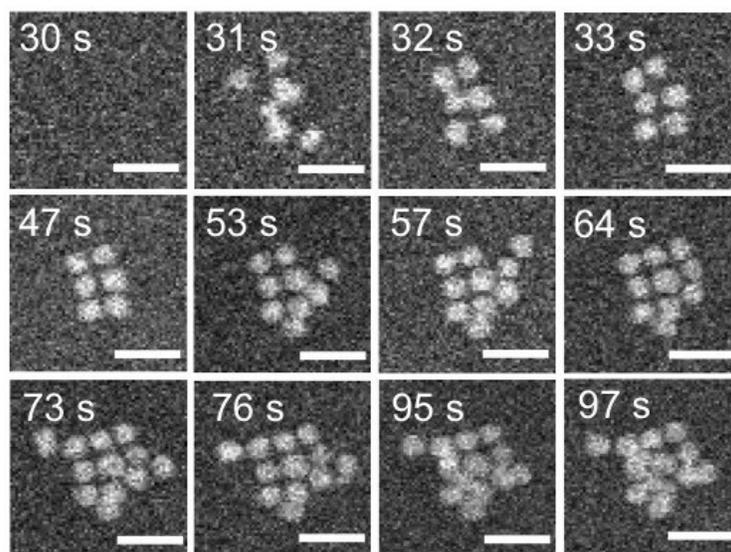
**Figure S2.** TEM images of thiolated single strand (ss) DNA functionalized Au nanoparticles and citrate terminated Au nanoparticles at pH 1. **(a), (b)** ssDNA conjugated Au nanoparticles remain intact and aggregate/self-assemble at pH 1. **(c), (d)** Citrate-capped Au nanoparticles coalesce upon lowering of the pH.



**Figure S3. Particle incorporation during linear chain formation.** (a), (a') Attachment of a single-stranded DNA terminated Au particle to a four-particle linear chain ( $t = 12$  s) by a jump into contact ( $t = 13$  s) followed by edge diffusion and final incorporation (trajectory (1),  $t = 18$  s). (b), (b') Resulting five-particle chain ( $t = 18$  s), jump to vicinity of a sixth particle ( $t = 19$  s), and final attachment of this additional DNA-Au nanoparticle conjugate at the end of the chain (trajectory (2),  $t = 22$  s). (c), (c') Out-of-plane incorporation of a seventh particle by jump into the vicinity of the six-particle chain ( $t = 39$  s), followed by edge diffusion and hollow-site incorporation in the second layer (trajectory (3),  $t = 45$  s).



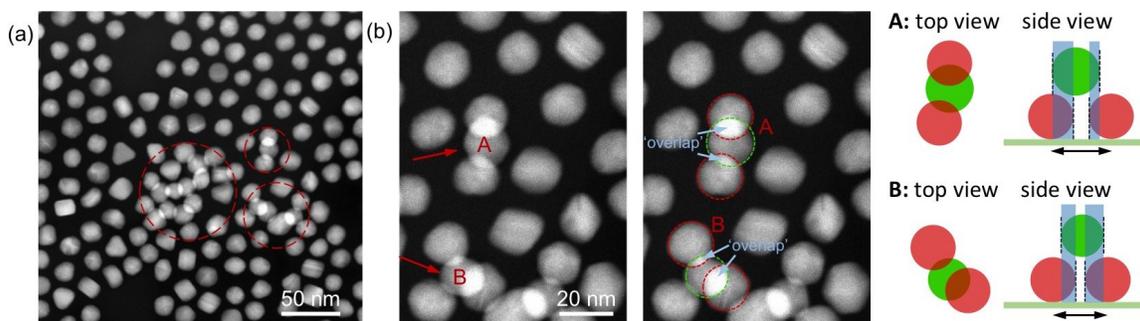
**Figure S4.** Interaction of ssDNA-terminated Au nanoparticles and transition from an initial linear to a final densely packed assembly. Scale bar: 50 nm.



**Figure S5.** Arrangement of ssDNA-conjugated Au nanoparticles on a square lattice ( $t = 33 \text{ s} - 47 \text{ s}$ ). Incorporation of additional particles appears to disrupt the planar square arrangement ( $t = 53 \text{ s}$  onward). Scale bar: 50 nm.

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**Figure S6.** (a)-(b) HAADF-STEM images of ssDNA functionalized Au nanoparticles. Dashed circles in (a) indicate areas with particles arranged vertically in two distinct layers. (b) magnified detail from (a) in which individual particles (A and B) in the second layer are marked with arrows. In HAADF-STEM (Z-contrast) imaging, the contrast is proportional to the thickness, i.e. thicker areas ('overlap') appear brighter. Thus, the bright contrast of particle A shows the areas of overlap of this particle with the two adjacent particles. This contrast indicates that particle A is not in the same layer as the two adjacent particles, i.e., it is centered over the spacing between the adjacent particles as shown schematically in side and top views of the arrangements around particles A and B. The interparticle spacings are measured between particles in the same layer (double arrow).

### Supporting Movies

**Movie S1.** Liquid cell electron microscopy observations of the self-assembly of single-strand DNA functionalized Au nanoparticles in aqueous solution. Scale bar: 200 nm.

**Movie S2.** Liquid cell electron microscopy observations of the self-assembly of citrate-terminated Au nanoparticles in aqueous solution. FOV: 1090 nm × 1090 nm.

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