Nanofabrication via Force Engineering for Extreme Nanodevices

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Next generation nanodevices require scalable, yet deterministic and pristine integration of rapidly growing library of nanomaterials into miniaturized device designs. Amongst these are colloidal nanoparticles made through bottom-up chemical synthesis with diverse properties, unique and superior to their bulk counterparts. For these properties to be leveraged in new device paradigms, the nanoparticles need to be integrated into deterministic designs with single particle resolution whose properties can be finely engineered. Resolution requirements and chemical incompatibilities, render conventional top-down fabrication strategies insufficient for their processing. To this end, diverse bottom-up assembly strategies have been explored, yet, commonly lack the desired versatility, scalability, and spatial structural control.

Here, we introduce a platform in which nanoscale forces, including van der Waals and capillary, can be engineered through chemical and structural considerations to guide scalable and deterministic integration of nanoparticles into functional nanodevices. In particular, we introduce two strategies (Figure 1). In one approach summarized in Figure 1a, forces are used to guide dry contact printing of spatially arranged colloidally-synthesized nanoparticles onto arbitrary surfaces and with diverse designs.[1] In the second approach shown in Figure 1b, rather than spatial arrangement and printing of pre-synthesized particles, the nanocrystals are directly synthesized on-site enabled by guided confinement of precursor solutions through the nanoscale forces. Using these techniques, we achieve arrays of patterned nanoparticles with sub-50 nm positional accuracy and dimensions down to 30 nm. We use these features to demonstrate reconfigurable metal-molecule-metal actuators with sub-nm sensitivity, emitter-coupled plasmonic cavity arrays based on a nanoparticle-on-mirror design, and on-chip halide perovskite nanoscale light-emitting diodes with single nanocrystal pixels. These examples highlight the new opportunities that engineering fundamental forces offers in integrating nanoparticles and translating their unique properties into functional devices.

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[1] W. Zhu *et al.*, Nanoparticle contact printing with interfacial engineering for deterministic integration into functional structures, *Sci. Adv.* **8**, 43 (2022).

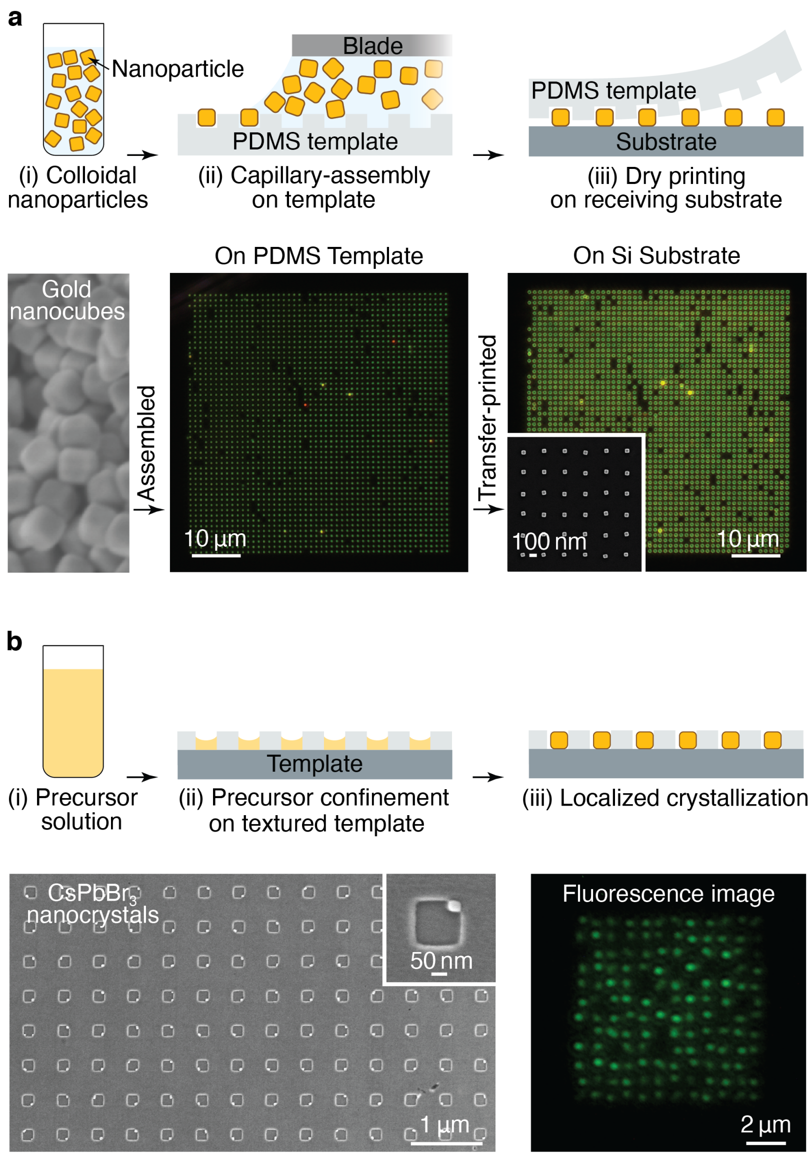

Figure 1: Nanofabrication via Force Engineering: (a) Colloidal nanoparticles are assembled into a topographical template with a deterministic spatial arrangement leveraging capillary forces. Then, the patterned particles are dry transfer-printed onto arbitrary surfaces through optimizing the van der Waals interactions. An example based on colloidal gold nanocubes is shown where dark-field scattering images of an array of particles formed on the template and printed on a silicon substrate are included with the inset showing a magnified scanning electron microscope (SEM) image of the array. (b) A topographical template with chemically-textured surface is used to control nanoscale forces to allow precursor solution confinement for localized nanoparticle growth on-site. An example forming arrays of lead halide perovskite nanocrystals ($CsPbBr_3$) is shown. Left image shows the SEM image of an array and the right is the fluorescence image of the $CsPbBr_3$ array with emission at ~ 510 nm.