Directed assembly of perovskite nanocrystals on topographically and chemically patterned surfaces

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Colloidal nanomaterials display a broad range of unique chemical and physical properties that make them prime candidates as nanoscale building blocks for the development of future technologies. Towards this goal, one of the main challenges resides in developing methods to manipulate these materials with a level of precision comparable with their small size. Achieving such high control typically requires a transition from solution phase to solid state; it is during this transition that the system is brought from the random and dynamic spatial distribution of a colloidal solution to an ordered, static assembly. To this end, one of the most effective approaches is to lithographically pattern the substrate with guiding features that act on the assembly process and yield the desired assembly configuration. The patterning can be topographical, chemical, or a combination of the two. In this work we study the effects of topography and surface chemistry on the assembly behavior of perovskite nanocrystals (PNCs), a novel type of nanocrystals with outstanding optical properties and great potential for applications in optoelectronics and photonics.[1]

The PNCs subjects of this study are cubic in shape with side length of about 10 nm. They are surrounded by organic ligands terminating with apolar groups; they are thus soluble in apolar solvents and have good affinity for hydrophobic surfaces. The PNCs solution is deposited by spin-coating on silicon substrates that are patterned with narrow trenches — by electron beam lithography and plasma etching — and which surface is chemically functionalized to change the degree of hydrophobicity. The effects of the pattern size and surface chemistry are carefully examined. Trenches width and depth are systematically varied between 20 and 50 nm and between 20 and 100 nm, respectively, so that to accommodate a limited number of nanocubes. As shown in Figure 1, shallow trenches do not produce a regular assembly while trenches with depth of 100 nm successfully confine PNCs into ordered lines as narrow as made of one or two PNCs and extending for hundreds of nanometers without gaps or defects. Arranging PNCs in such ordered, 1D-like features allows careful studies of the collective mechanisms of exciton diffusion and recombination in PNCs assemblies, which in turn determine the optoelectronic behavior of the system and offer fundamental guidance in engineering new optoelectronic devices made of PNCs.

PNCs are bright emitters but their emission can be further amplified if it is coupled with a resonant photonic crystal (PhC) mode. Our simulations, shown in Figure 2a, highlight the electromagnetic field magnitude inside a PhC.[2] By controlling PNCs assembly on the PhC, as shown in Figures 2c and 2e, we can study the coupling between the PhC mode and the PNCs emission as a function of the PNCs assembly geometry. Confining PNCs on the regions of maximum electric field maximizes photoluminescence amplification with minimal distortion of the PhC mode.

As shown in these two examples, controlling PNCs assembly opens new avenues for the development of novel nanostructured optoelectronic and photonic devices that take full advantage of PNCs nanoscale size, their solution processability, and their bright and narrow linewidth photoluminescence.

[1] L. Protesescu, et al. *Nano Letters* 2015, 15, 3692-3696.
[2] V. Mocella and S. Romano. *Physical Review B* 2015, 92

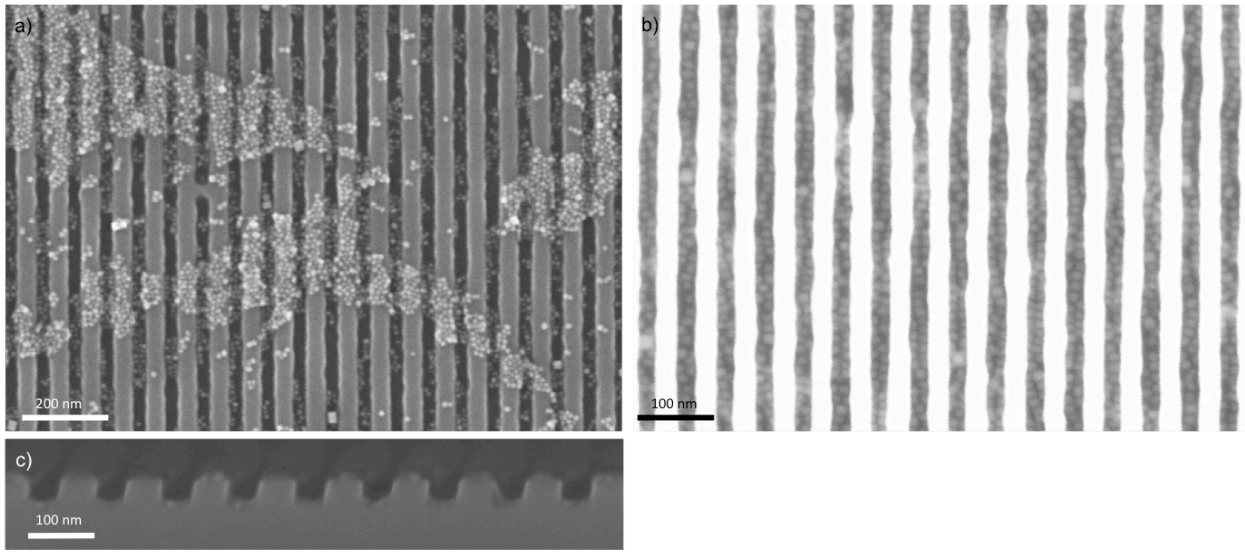

Figure 1. SEM images of PNCs spin-coated on a silicon substrate patterned with trenches of depth a) 30 nm, and b) 100 nm. c) Cross section SEM image of the sample in a) showing the trenches profile (30 nm depth). PNCs are visible on the top and bottom levels confirming the lack of confinement in the case of shallow trenches. Only the 100 nm deep trenches produce an ordered assembly, confining the PNCs on the bottom of the trenches and aligning them with the trench edges.

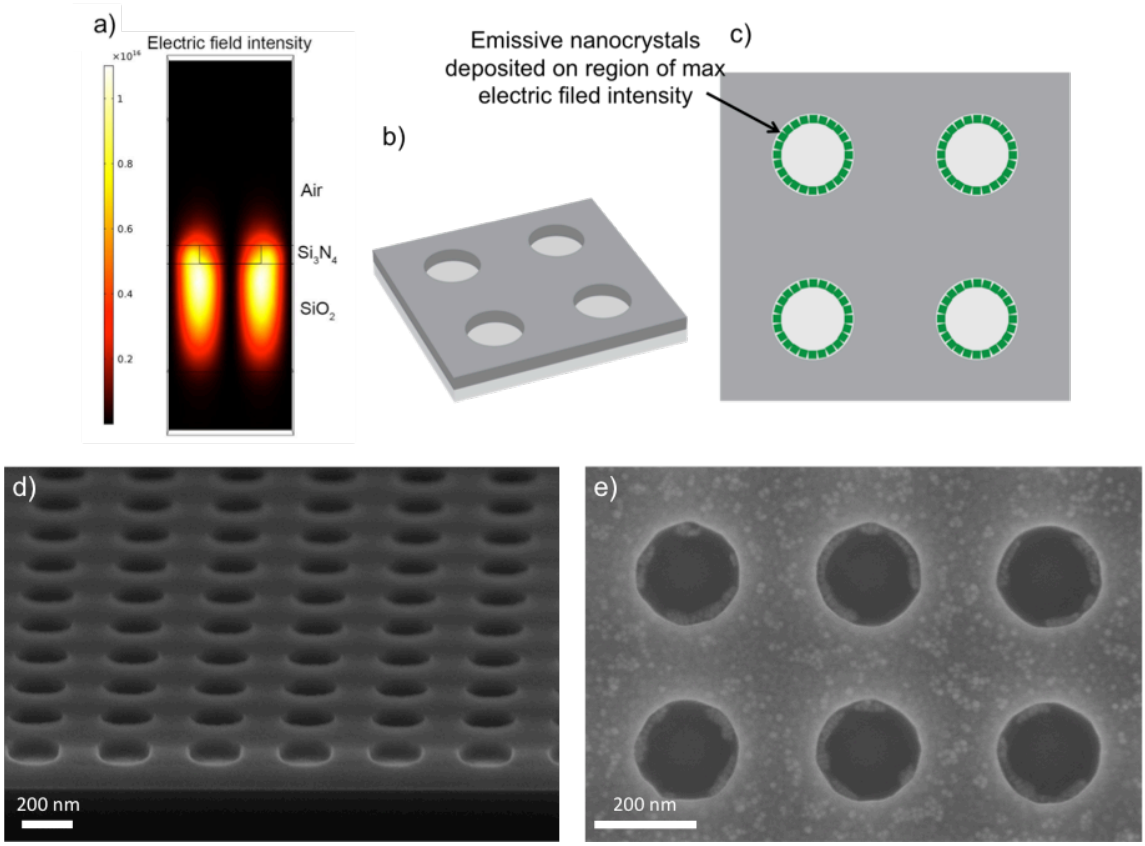

Figure 2. a) Simulated electric field intensity distribution in the longitudinal plane of the photonic crystal. The maximum field intensity on the surface is located at the inner edges of the etched circles. Schematic of b) the photonic crystal structure and of c) the target PNCs assembly in the region of maximum electric field. d) SEM image of the patterned photonic crystal. e) SEM image of PNCs assembled along the inner edge of the etched circles.