Placing quantum dots in 3D photonic crystals and finding them back

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It is a major outstanding goal in Nanophotonics to precisely place quantum emitters inside a three-dimensional (3D) metamaterial. It is well-known that such control offers exquisite control over cavity QED, spontaneous and stimulated emission, and even non-linear optics [1]. Theory predicts that the emission of an emitter, *e.g.* a quantum dot, varies spatially on 100s nm scale [2]. Thus, the challenge is to place emitters with a precision better than $\Delta x < 100$ nm. We present our newly developed chemical toolbox to fix the positions of quantum dots with a polymer brush layer with thicknesses in the 10s nm range in silicon nanostructures [3].

Once 3D positioning is successful, a second challenge arises, namely *how* to non-destructively find *where* the emitter sits in the 3D structure. Since nanophotonic materials are necessarily opaque, optical microscopy has insufficient penetration depth, apart from its limited resolution. While scanning electron microscopy (SEM) offers sufficient spatial resolution, it has a small penetration depth hence only the sample surface is viewed (see Fig. 1(A)) but not the bulk. X-ray techniques are promising tools, in view of excellent penetration depth, non-destructive character, and nm spatial resolution. Therefore, we study a 3D Si photonic band gap crystal with infiltrated PbS nanocrystal quantum dots by X-ray fluorescence tomography [4].

Our photonic crystals have the inverse woodpile structure (Fig. 1(A)) that exhibits a broad full and complete 3D band gap [5]. The crystal are made by CMOS-compatible methods using deep reactive ion-etching through tailored masks. Fluorescence tomography was performed at the ESRF (beamline ID-16NI). X-rays (17 keV photon energy) are focussed in the sample. We collect data at 17 different angles while rotating the crystal from 0 to 180°. Projection maps are obtained at every angle, followed by standard tomographic reconstruction to obtain the 3D atom density distribution with 50 nm spatial 3D resolution for each chemical element.

Fig. 1(B) shows a projection map of the number of lead atoms - from the quantum dots - in one crystal. The volume is a cube that contains the 3D photonic crystal structure that is surrounded by bulk silicon above and by the 2D array of deep pores that are etched first. Close inspection of the 3D volume after reconstruction indeed reveals two sets of pores running in the Z and the X-directions, matching the design (Fig. 1(A). The structure is periodic with lattice parameters that also match the design very well. It appears that the quantum dots are located throughout the whole crystal volume. Their position correlates well with elements characteristic of the polymer brush layer. Finally, we find that after the X-ray experiment the quantum dots remain optically active.

We conclude that 3D X-ray fluorescence tomography has great potential to solve many future questions on 3D optical metamaterials for nanophotonic research and applications, including cavity arrays, physically unclonable functions, and precise localization of light emitters as qubits and for enhanced lighting efficiency.

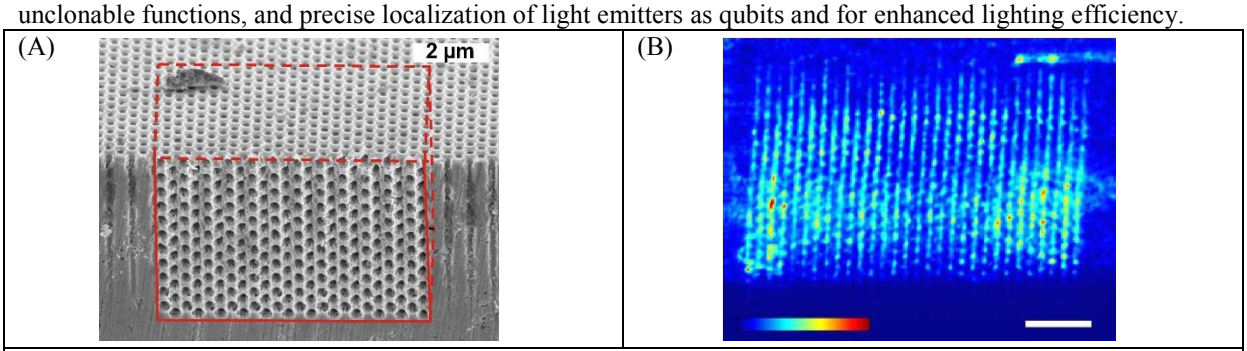

Fig. 1. (A) SEM image of the surface of a 3D Si inverse woodpile photonic crystal. The estimated extent is shown by dashed lines. The 3D crystal is surrounded by a large 2D array of pores that is first etched in the Z-direction, from [5]. **(B)** Projection map of the number of Pb atoms per pixel (area 50 x 50 nm) and integrated along the propagation direction of the beam, for sample orientation at θ=-47°, as in the SEM on the left. The color scale is the areal atomic densitiy (in units atoms/pixel area) ranging from 0 to 420000 Pb atoms per pixel. The scale bar represents 2 µm as in Fig. 1(A).

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