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## Integrating top-down nanopatterning with bottom-up self-assembly to fabricate photonic cavities with atomic-scale dimensions

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The two main techniques for fabricating nanostructures and functional devices are top-down nanopatterning and bottom-up self-assembly. Top-down nanopatterning offers precise control of pattern placement and scalability but is limited in achieving the smallest feature sizes [1]. Bottom-up self-assembly involves building blocks organizing themselves into functional structures through non-covalent interactions, offering atomic-scale resolution but less geometric freedom and production yield [2]. Combining the best of both approaches would be of great value to science and technology, especially nanophotonics because, on the one hand, light confinement is limited by the size of the smallest feature that can be fabricated [3, 4, 5] and, on the other hand, the potential of photonic technologies rely on the reliable fabrication of large-scale circuits [6]. An important proposed challenge is to confine light to unprecedented levels in air bowties with aspect ratios > 100 [3].

In this work, we use a novel method to fabricate photonic cavities with atomic-scale dimensions that use surface forces such as van der Waals, Casimir, and capillary forces [7] to self-assemble structures patterned only through lithography and etching [8]. The geometry and normalized electric field of the fundamental optical mode of our cavity design is shown in Fig 1a and 1b. The fundamental cavity mode has a resonance wavelength of  $\lambda = 1524$  nm, a quality factor of  $Q = 5 \times 10^4$ , and a mode volume of  $V = 3.4 \times 10^{-4} \lambda^3$ . The extremely small mode volume stems from light confinement to a 2 nm air bowtie (Fig 1c and 1d).

We fabricate the designed air-bowtie nanobeam cavities in silicon-on-insulator wafers (Fig 2a) by selfassembling two halves initially separated by a gap  $g_f$  (Fig 2b) and suspended by two folded guided cantilevers. When the buried-oxide layer is etched, the two halves and springs are released, and the surface forces cause a deterministic collapse to form a 2 nm bowtie gap (Figs 2c and 2d). The resolution of the nanofabrication limits the absolute value of  $g_f$ . However, the relative distance between  $g_b$  and  $g_f$  is not limited by resolution, and, thus, the bowtie width,  $g_i$  in the final device is limited only by surface roughness, enabling the realization of bowties with atomic-scale features (see high-resolution transmission electron microscope image in Fig 2d). Figure 2e shows the scattered far-field spectrum of a self-assembled cavity obtained with cross-polarized optical microscopy and its resonance fitted with a Fano lineshape. Our approach showcases the potential of integrating top-down nanopatterning and bottom-up self-assembly to realize photonic nanocavities with atomic-scale dimensions for applications such as single-photon nonlinearities and single-photon sources.

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Figure 1. Design and geometry of a bowtie nanobeam cavity with atomic-scale dimensions. (a) The geometry of the bowtie nanobeam cavity. (b) The normalized electric field of the fundamental cavity mode,  $|\mathbf{E}|$ , with a logarithmic color map. (c) The single bowtie unit cell with the following parameters: lattice constant, *a*, bowtie angle,  $\varphi$ , bowtie width,  $W_b$ , nanobeam width, *H*, and device-layer thickness, *t*. The inset illustrates the bowtie width, *g*, of 2 nm at the center of the bowtie. (d) . The normalized electric field of the fundamental cavity mode at the central bowtie unit cell with a linear color map. The inset illustrates the electric field tightly confined around the 2 nm bowtie width.



**Figure 2.** Fabrication and optical characterization of the self-assembled atomic-scale photonic cavities. (a) Tilted (20°) scanning electron microscope (SEM) image of a self-assembled nanobeam cavity. (b) Topview SEM image of a single bowtie unit cell before self-assembly, with a fabricated gap,  $g_f$ , and a bowtie gap,  $g_b$ . (c) Tilted (20°) SEM image of a bowtie unit cell after self-assembly with an approximate bowtie width of 2 nm. (d) Top-view high-resolution transmission electron microscope image of the central region of the bowtie with an approximate 2 nm gap. The interplanar distance between the (022) silicon crystal planes is 0.19 nm. (e) Scattered far-field spectrum of a self-assembled nanobeam cavity measured using cross-polarized optical microscopy. The inset shows the resonance of the fundamental mode at the wavelength of 1521.5 nm, and a quality factor of  $(4.2 \pm 0.1) \times 10^4$ , extracted by fitting a Fano lineshape to the resonance.