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Colloidal ZnO quantum dots in ultraviolet pillar microcavities

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Abstract: Three dimensional light confinement and distinct pillar microcavity modes in the ultraviolet have been observed in pillar resonators with embedded colloidal ZnO quantum dots fabricated by focused ion beam milling. Results from a waveguide model for the mode patterns and their spectral positions are in excellent agreement with the experimental data.

OCIS codes: (350.4238) Nanophotonics and photonic crystals; (260.7190) Ultraviolet; (140.3948) Microcavity devices; (230.5590) Quantum-well, -wire and-dot devices.

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Semiconductor quantum dots are promising candidates for compact and robust single photon sources in quantum communications and quantum information processing systems [1]. Efficient single photon sources in the infrared have been demonstrated by embedding self-assembled semiconductor quantum dots into optical microcavities, such as microdisks [2], micropillars [3, 4], and photonic crystals [5]. In contrast, the ultraviolet (UV) region of the electromagnetic spectrum remains a challenge for building single-photon nanophotonic devices. Only recently, a single photon source based on GaN/AlN quantum dots has been demonstrated [6]. To optimize the light-matter coupling these quantum dots should be embedded in a microcavity, in analogy to sources operating in the infrared. However, there is great difficulty in combining all-epitaxial GaN quantum dot and cavity mirror growth processes [7, 8]. An efficient single photon source based on wide-bandgap self-assembled quantum dots inside an all-epitaxial microcavity has not yet been demonstrated. As an alternative, chemically synthesized colloidal CdSe/ZnS quantum dots also offer single photon emission capability at room temperature [9, 10]. Using gentle processing techniques, such colloidal quantum dots have been incorporated into all-dielectric pillar microcavities operating in the visible [11]. This approach circumvents many of the challenges of all-epitaxial growth.

In this work we present the first pillar microcavity with embedded semiconductor quantum dots operating in the UV. For this objective, colloidal ZnO quantum dots were chosen. The semiconductor ZnO possesses excellent optical properties. Both the large bandgap of ZnO of 3.4 eV and its exciton binding energy of 60 meV will be strongly beneficial to room-temperature operation of future single-photon devices. ZnO has a low refractive index compared to other semiconductors, which facilitates light extraction. This compound is also a promising candidate for spintronics applications [12]. ZnO quantum dots have shown long electron spin dephasing times even at room temperature [13] and have been successfully doped with magnetic ions [14].

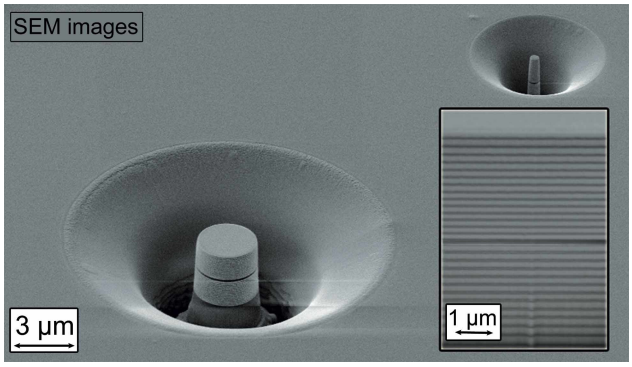

Fig. 1. SEM image of two ultraviolet micropillars with diameters of 3.2 µm and 600 nm, respectively, fabricated by focused ion beam milling and containing embedded colloidal ZnO quantum dots. Inset: Cross-sectional SEM image of the planar microcavity.

The UV pillar microcavities investigated here involve high-quality colloidal ZnO quantum dots embedded into an all-dielectric microresonator formed by HfO_2/SiO_2 Bragg mirrors. First, the bottom Bragg mirror was fabricated via RF-magnetron sputtering. It consisted of amorphous HfO_2 and SiO_2 alternating layers of $\lambda/4$ thickness (45 nm for HfO_2 and 62 nm for SiO_2) to reflect the UV photoluminescence of the colloidal ZnO quantum dots (Fig. 2(d)). Since the mirror materials have very low absorption in the near ultraviolet a reflectivity better than 99 % has been achieved with 15 layer pairs per mirror. After deposition

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of a SiO_2 spacer layer, 7.0 nm diameter ZnO quantum dots were suspended in liquid polysilazane [11] and spin-coated onto the mirror. The ZnO quantum dots were capped with dodecylamine to passivate surface defects and enhance their excitonic UV emission [15]. The spin-coated polysilazane layer solidified to a 30 nm thick SiO_2 layer in which the ZnO dots were embedded. Finally, SiO_2 was sputter-deposited to form a $\lambda/2$ cavity. This procedure ensures that the quantum dots are located in the electric field maximum of the vertical resonator. We estimate a submonolayer surface density for the ZnO dots in the central SiO_2 cavity layer. The planar cavity structure was completed by depositing another 15 pairs of HfO_2 and SiO_2 layers (see inset of Fig. 1).

In the planar resonator, only one-dimensional light confinement is present between the two Bragg mirrors because light can still leak out in lateral directions. In contrast, pillar microcavities provide three-dimensional light confinement due to total internal reflection at the resonator sidewalls. Therefore, pillar microcavity resonators were milled out of the planar cavity with a focused ion beam (FIB). Figure 1 shows scanning electron microscope (SEM) images of two such microresonators with diameters of 3.2 µm and 600 nm, respectively.

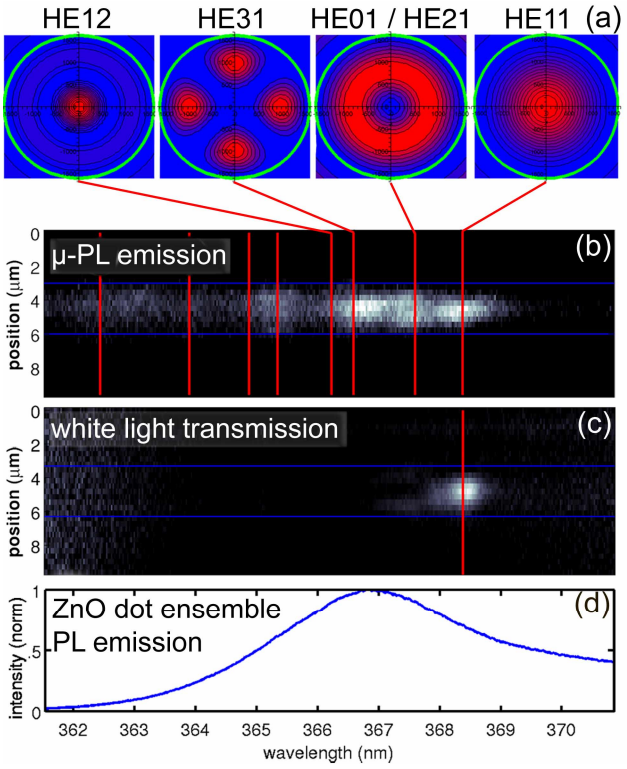

Fig. 2. (a) Simulated mode patterns of a 3.2 µm diameter ultraviolet pillar microcavity. (b) Top: Spatio-spectrally resolved photoluminescence from colloidal ZnO quantum dots in a 3.2 µm diameter micropillar resonator. Vertical red lines indicate calculated cavity mode positions. (c) Spatio-spectrally resolved white light transmission through the same micropillar. (d) Ensemble photoluminescence of the colloidal ZnO quantum dots measured outside the microcavity.

To demonstrate three-dimensional light confinement inside these UV microcavities, we performed micro-photoluminescence spectroscopy experiments. The ZnO quantum dots inside the cavity were excited with a continuous wave HeCd laser at a wavelength of 325 nm. The excitation light was focused on the pillar cavity with a UV objective with a numerical aperture of 0.5. The same objective was used to collect the photoluminescence, which was then spectrally dispersed in a monochromator (f = 0.5 m). The signal was recorded with a peltier-cooled CCD camera. The sample was kept at a temperature of 10 K in a helium flow cryostat.

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Figure 2(b) shows the photoluminescence spectrum from the 3.2 μm diameter pillar resonator of Figure 1. In our setup, the slit at the monochromator entrance provides spatial resolution in the vertical direction (Fig. 2(b) and c vertical axes) and spectral information in the horizontal direction (Fig. 2(b) and c horizontal axes). The fundamental cavity mode and several higher cavity modes are clearly visible. A broadband emission background emanating from the glass substrate has been subtracted from the spectrum. For the fundamental mode of the pillar microcavity a Q-factor of 250 has been determined. The spatial mode pattern of the first excited mode clearly differs from the fundamental mode.

To understand the measured photon emission spectrum we calculated the mode positions and patterns with an effective refractive index model in a wave-guiding structure [11]. The red lines in Fig. 2(b) mark the positions of the calculated cavity modes. The simulated mode positions are in excellent agreement with the experimental results. Since the recorded photoluminescence emission provides us with one-dimensional spatial resolution along the entrance slit of the spectrometer, the CCD image of Fig. 2(b) can be directly compared to a one-dimensional vertical cut in the center of the calculated two-dimensional mode patterns of Fig. 2(a). For example, the HE11 mode shows one intensity maximum, while the first excited mode HE01 consists of two vertically displaced components. To understand the appearance of higher cavity modes, the photoluminescence spectrum of an ensemble of ZnO quantum dots outside the cavity has been recorded, also at 10 K (Fig. 2(d)). The dot emission maximum is shifted slightly to longer wavelengths compared to the fundamental pillar mode position and it has a finite spectral width. Hence, there is still enough emission intensity at the higher cavity mode positions for these resonances to be observed in the micro-photoluminescence spectra of the pillar microcavity.

Figure 2(c) shows a white light transmission measurement of the same pillar microcavity. A UV xenon lamp has been used as a broadband light source. In contrast to the photoluminescence, the transmission spectrum is dominated by the fundamental mode of the pillar microcavity. Only a faint first excited mode is also visible. Due to the use of incoherent light in this experiment and a numerical aperture that is not matched to the pillar cavity, coupling to the totally symmetric fundamental resonance is dominant. In the photoluminescence experiment described before, the internal light source provided by the ZnO quantum dots emits light in all directions. Hence, in photoluminescence all pillar modes are observed.

In summary, our results demonstrate the first pillar microcavity with embedded semiconductor quantum dots operating in the ultraviolet. Pillar resonators with colloidal ZnO quantum dots have been fabricated via a combination of sputtering, solution processing of colloidal quantum dots, and focused ion beam milling. This approach circumvents some of the challenges that have previously confronted the formation of UV pillar microcavities. Micro-photoluminescence spectroscopy of the pillar resonators has shown several distinct microcavity modes in the ultraviolet. In the future, the ZnO quantum dot ensemble used in this study may be replaced by single ZnO quantum emitters, magnetically doped ZnO nanocrystals, or other UV emitters. With such approaches integrating solution processing and nanofabrication, robust single-photon sources operating at room temperature are within reach, and other nanophotonic devices based on colloidal quantum dots in dielectric or metallic [16] surroundings are on the horizon.

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