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Enhanced photoluminescence from embedded PbSe colloidal quantum dots in silicon-based random photonic crystal microcavities

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The experimental observation of enhanced photoluminescence from high-Q silicon-based random photonic crystal microcavities embedded with PbSe colloidal quantum dots is being reported. The emission is optically excited at room temperature by a continuous-wave Ti-sapphire laser and exhibits randomly distributed localized modes with a minimum spectral linewidth of 4 nm at 1.5 μm wavelength. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2954007]

There have been a host of attempts to extract light from silicon and to demonstrate lasing with radiative host materials embedded within or deposited on top of silicon. Nanostructured silicon emitters with various degrees of quantum confinement have also been investigated for light emission, with limited success.[1,2] The two essential requirements for coherent emission are a gain medium with a high quantum efficiency and a resonant cavity with a high quality factor. A promising approach is to use chemically synthesized nanocrystals, such as Pb(S, Se) and CdSe colloidal quantum dots (QDs) as gain media, embedded in a high-Q silicon-based microcavity. Enhanced luminescence has been demonstrated with Pb(S, Se) QDs embedded in Si photonic crystal (PC) cavities.[3,4] The colloidal QDs, which exhibit size-tunable luminescence with high efficiency ($>80\%$) in the near infrared (IR) range, represent a technologically interesting choice of gain medium for potential applications in silicon photonics.[5,6] In this letter, we report the experimental observation of enhanced photoluminescence (PL) from PbSe QDs embedded in silicon *random* PC microcavities.

PCs are periodic dielectric structures, usually two-dimensional (2D) arrays of air holes in high-refractive-index membranes, that selectively inhibit light propagation in certain bands of frequencies.[7] Destroying the periodicity of the lattice introduces small defects which act as optical cavities with high Qs wherein light can be localized by total internal and Bragg reflections. Q factors of the order of 10^6 have been measured in engineered microcavities in 2D PCs.[8] On the other hand, Topolancik *et al.* have recently investigated and reported a different approach to photon localization in PCs, which relies on random structural perturbations introduced uniformly throughout the crystal by deliberately changing the shapes and orientations of the lattice elements (air holes).[9] Such random disorder superimposed onto the crystal causes backscattering which impedes propagation of Bloch waves along line defects defined in the 2D lattice. Extended modes that propagate with a low group velocity at frequencies approaching the mode edge become spatially confined in sections of the disordered waveguide. This subtle interplay of order and disorder was predicted to give rise to Anderson localization in disordered lattices.[10] Incorporation of suitable gain media into these structures could enable self-optimized lasing from random nanocavities operating around the guided mode's cutoff, similar to what has been observed at the photonic band edge in *crescent*-deviation disordered PCs.[11] It is worth noting that disordered waveguide structures could support self-optimized nanocavity lasers with significantly smaller modal volumes and lower thresholds than the large-area, disordered PC band-edge lasers.[11]

The fabrication of the devices uses a simple scheme of incorporating colloidal PbSe QDs into the random PC microcavities. The disordered PCs were fabricated on silicon-on-insulator substrates using standard electron-beam lithography and reactive ion etching. A line-defect waveguide is formed by equally spaced circular holes defined in a hexagonal lattice of randomly rotated squares. The top image of the fabricated structure is shown in the scanning electron micrograph (SEM) in Fig. 1(a). The thickness of the silicon slab ($h=220$ nm), the radius of the defect holes ($r=105$ nm), and the lattice constant ($a=470$ nm) and the fill factor ($\sim 30\%$) of the bulk PC were chosen so that the cutoff of the guided mode aligns spectrally with the PL peak of colloidal PbSe QDs at 1510 nm. The dispersion of the waveguide in the underlying periodic crystal calculated by plane-wave expansion method and the room temperature PL spectrum of the dots are shown in Fig. 1(c). The superimposed random scatterers which trigger mode-edge localization can be viewed as the difference between circles in the underlying (ideal) crystal and randomly oriented squares in the disordered crystal.

PbSe QDs were synthesized using a noncoordinating solvent technique.[5,6] The synthesis procedure starts with the preparation of a solution of PbO and oleic aid and the subsequent heating of the solution up to an elevated temperature of 160 °C. Rapid injection of selenium-trioctylphosphine reagents into the hot solution induces the nucleation of PbSe and subsequently cooling down the reaction temperature to 135 °C allows the nuclei to grow into highly crystalline nanoparticles. The size of PbSe QDs can be tailored by care-

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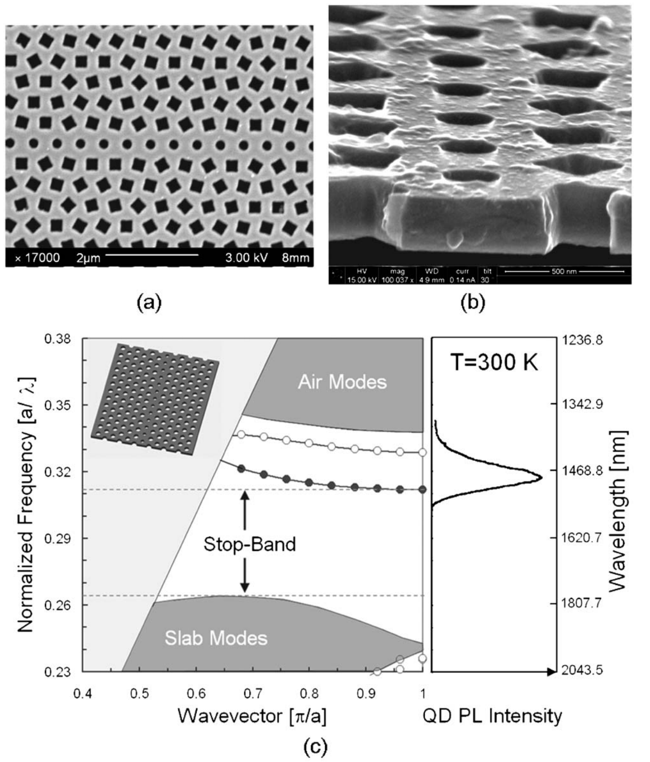

FIG. 1. (a) SEM of the fabricated Si-based 2D membrane disordered PC microcavity, (b) of a cross section of the PC showing PbSe QDs embedded into PC microcavities, and (c) calculated dispersion of the defect waveguide in ideal crystal shown in the inset (hollow circles denote odd modes and solid circles denote even modes).

fully controlling the growth conditions. The QD growth was monitored using visible/near IR absorption spectroscopy to achieve the desired wavelength emission wavelength around 1.55 μm.

The width of the localization band and the positions of random resonators before QD deposition were measured with a 1475–1580 nm broadly tunable laser source which was coupled laterally into the waveguide. The vertically scattered light emitted from random cavities was collected with a high-resolution objective lens and recorded with either a photodiode to obtain vertically scattered spectra from small sections of the waveguide or with an IR camera to obtain 2D spatially resolved spectra shown in Fig. 2(a). The plot shows an approximately 40 nm broad band filled with confined fields with various localization lengths. Note that these are random patterns, i.e., every device has a unique spectral signature and both Q factor and localization position may vary across the pattern and from pattern to pattern. Figure 2(b) shows a resolved projected spectrum collected from a 5 μm long section of the disordered PC waveguide. The spectrum exhibits a high Q (~55 000) resonance near 1512 nm. Such randomly distributed and localized high-Q resonances are typical for random cavities based on multiple scattering feedbacks,[12,13] which will be reflected in the following characterization of active devices as well.

To characterize active devices, colloidal PbSe QDs were embedded in the nanoscale air holes comprising the line defects in disordered PCs. To maximize the density of QDs coupling with the microcavities, the samples were soaked in the PbSe QD solution for several hours. The SEM image in Fig. 1(b) shows a cross section of the PCs embedded with PbSe QDs. The devices were optically excited at room temperature with a continuous wave (cw) Ti:sapphire laser operating at 810 nm. Emission from the QDs in the microcavities was focused by a high-resolution 100× objective lens with an effective focus length $f=2$ mm and a numerical aperture of 0.7. The diffraction-limited size of the focus spot of the pump beam ($\lambda=810$ nm and ~3.5 mm aperture or beam size) is ~1 μm. The localized modes have the minimum localization lengths of few lattice constants. The small focal spot allows us to efficiently pump these highly localized modes. It should also be mentioned that the disordered structure supports multiple spatially overlapping modes of various localization lengths (modal volumes) at most probing frequencies (wavelengths). This can be seen clearly in 2D contour map in Fig. 2(a) which shows randomly distributed (hot spots) revealing positions of the localization regions. It means that, in the active structure, multiple modes are likely to be excited. The output spectrum was analyzed with a 0.75 m high-resolution spectrometer and detected with an InGaAs photomultiplier tube using phase lock-in amplification. The pump light is blocked by a bandpass filter placed in front of the spectrometer. Unlike emission from the conven-

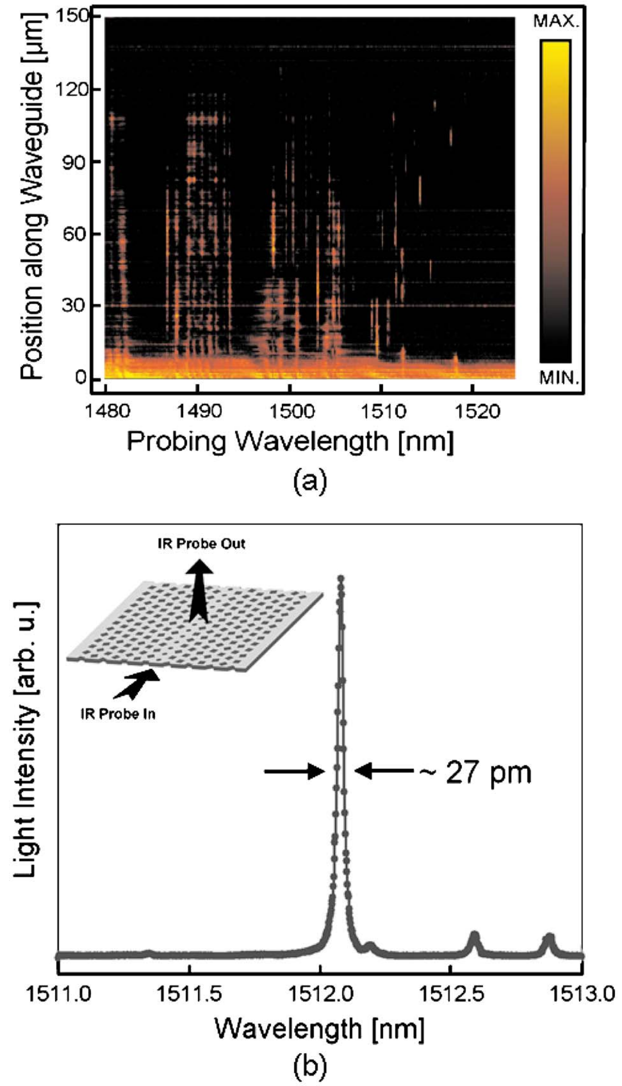

FIG. 2. (Color online) (a) Contour plot of the spatially resolved spectra of a 150 μm long disordered waveguide. (b) Example of a well-localized, high-Q resonance in the passive random PC microcavities. The probing and collection directions are indicated in the inset.

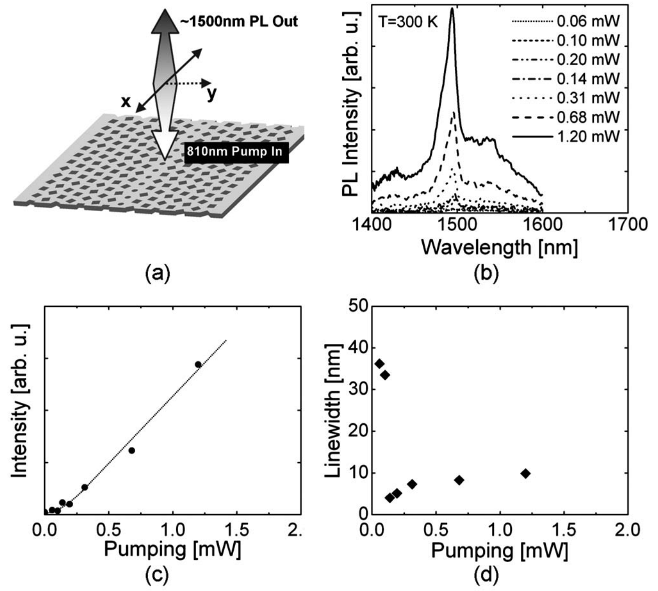

FIG. 3. (a) Schematic of the active cavity characterization scheme, (b) emission spectra of a silicon random PC microcavity with PbSe QDs measured at 300 K at different pump powers, (c) *L*-*L* characteristics, and (d) emission peak linewidth vs pump powers.

tional, engineered PC microcavities, the exact position of which is known, the output spectral characteristics for the random microcavities are sensitive to the location of the excitation. The disordered waveguides were probed systematically by scanning the focused pump beam along the waveguide axis (x), as shown schematically in Fig. 3(a). A strong dependence of excited modes' spectral characteristics on the excitation position was observed. Figure 3(b) shows a typical emission spectrum collected from a single excitation spot for varying excitation intensities. At lower pump intensity, the spectrum exhibits a broad spontaneous emission. Once the pump intensity exceeds a certain threshold, a much narrower emission peak emerges ($\sim$4 nm linewidth). It is possible that multiple random resonances are being excited according to the non-Lorentzian line shape of emission peak. There is a visible shoulder to the peak, and hence the linewidth of the emission peak is estimated by fitting the main peak without the shoulder. The plot of the peak emission intensity versus the pump intensity $(L\text{-}L)$, shown in Fig. 3(c), exhibits a soft threshold at $\sim$100 μW. Figure 3(d) depicts the measured narrowing of the emission linewidth above the pump threshold. The data shown in Fig. 3 do not indicate lasing, but suggest the onset of enhanced spontaneous emission coupled into localized modes as a result of strong feedback from random PC microcavities. Such feedback enables photon intensity around the resonance peak to quickly build up over that of the background luminescence.

The observation of lasing could also be prevented by the low fill factor of the QDs in the microcavity and the resulting low modal gain in our experiment. Techniques to enhance the QD density are currently being investigated. Another important issue is the luminescence efficiency of the colloidal PbSe dots. It is observed that the efficiency is reduced, possibly due to surface contamination and oxidation, when the QDs are dried on the silicon PC microcavities. The luminescence efficiency is the highest in a sol-gel form or in a polymer matrix solution. It has also been recently demonstrated that PbS/PbSe core-shell nanocrystals are immune to degradation during the drying process.[6] The use of such dots will significantly enhance the radiative efficiency and the output intensity of the microcavity light sources. These aspects are also being undertaken.

In conclusion, we demonstrate a silicon-based light emitter based on high-Q random cavities in disordered PC waveguides with embedded colloidal PbSe QDs. Emission with a minimum linewidth of 4 nm is observed. Such nanoscale light sources on silicon, with potential compatibility with complementary metal oxide semiconductor chips, could be of interest as optical interconnects in silicon photonics.

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