Enhanced infrared emission from colloidal HgTe nanocrystal quantum dots on silicon-on-insulator photonic crystals

Chunxia Wang,[1,2] Jürgen Roither,[1] Raimund Kirschschlager,[1] Maksym V. Kovalenko,[1] Moritz Brehm,[1] Thomas Fromherz,[1] Qiang Kan,[2] Pingheng Tan,[3] Jian Liu,[3] Hongda Chen,[2] and Wolfgang Heiss[1,a)]
[1]*Institute of Semiconductor and Solid State Physics, Johannes Kepler University Linz, A-4040 Linz, Austria*
[2]*State Key Laboratory for Integrated Optoelectronics, Institute of Semiconductors, Chinese Academy of Sciences, Beijing 100083, People's Republic of China*
[3]*State Key Laboratory for Superlattices and Microstructures, Institute of Semiconductors, Chinese Academy of Sciences, Beijing 100083, People's Republic of China*

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A two dimensional silicon-on-insulator based photonic crystal structure is used to enhance the emission from colloidal HgTe nanocrystal quantum dots embedded in a thin polymer film. The enhancement is resonant to the leaky eigenmodes of the photonic crystals due to coherent scattering effects. Transmittance and photoluminescence experiments are presented to map the leaky mode dispersion and the angle dependence of the emission enhancement factor, which reaches values up to 80 (650) for vertical (oblique) emission in the telecommunication wavelength range. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3194151]

Silicon-on-insulator (SOI) has emerged as a promising platform for passive photonic functions due to its transparency at telecommunication wavelengths, its high refractive index contrast, and the well established Si technology. SOI is perfectly suited for devices based on two-dimensional (2D) photonic crystal (PhC) structures, such as high quality photonic cavities to confine light on nanoscales,[1] thermo-optical modulators to control the group velocity of light,[2] and PhC waveguides with small bend radii, which are integrated with PhC microcavities.[3] Even though optical gain has been demonstrated in periodic nanopatterned crystalline silicon[4] and Raman lasing in continuous mode has been achieved,[5] the development of efficient silicon based optical emitters is still a challenge due to the indirect nature of the band gap of silicon and, thus, its very low light emission efficiency. Most promising approaches to improve this situation are hybrid structures, where, e.g., III-V semiconductor heterostructures as optical active materials are combined with silicon-based waveguides.[6,7] While on this way even electrically pumped lasing has been demonstrated by wafer bonding of AlGaInAs on SOI substrates,[7] a convenient way to obtain light emitting hybrids is given, if the active material can be deposited from top onto patterned Si structures from solutions. Thus several attempts have been reported where infrared emitting colloidal PbS nanocrystal quantum dots (NC-QDs) are infiltrated into silicon-based PhC microcavities.[8–12] The coupling of the luminescence with the cavity modes led to narrow band emission lines and partly to an enhancement of the NC-QD emission intensity at the cavity modes wavelengths.[11]

An alternative attempt to enhance the emission from NC-QDs by a PhC structure was recently demonstrated by Ganesh *et al.*,[13] who demonstrated a doubly resonant scheme: optical excitation of the NC-QDs is performed resonantly via leaky modes of a photonic structure and light extraction is also enhanced by coupling to leaky modes. This doubly resonant approach leads to a NC-QD emission enhancement over a wider spectral region than obtained by the microcavity attempts. While Ganesh *et al.*[13] demonstrated the fluorescence extraction enhancement for colloidal NC-QDs operating in the visible spectral region, based on a TiO_2 PhC, here results are presented for the near infrared, making use of a silicon-based PhC slab. Instead of the commercially available PbS NC-QDs, HgTe NC-QDs are used, which have been previously suggested as a promising material for applications in the telecommunication wavelength region.[14,15] For the present system, significantly improved fluorescence extraction with an enhancement factor up to 650 is demonstrated for oblique emission (80 for vertical emission), obtained even under nonresonant pump excitation conditions. The extraction is highest close to the leaky eigenmodes above the light line in the PhC energy dispersion, which is confirmed by a comparison between photoluminescence (PL) spectra and angle dependent transmittance measurements.

The device structure consists of a SOI based 2D PhC slab, covered by a thin polymer film containing colloidal HgTe NC-QDs [a schema of the sample is shown in Fig. 1(a)]. The smart cut SOI wafer has a SiO_2 intermediate layer with a thickness of 1.0 μm and a 250 nm *p*-type Si layer on top. The pattern of the PhC is fabricated by deep ultraviolet lithography (248 nm illumination source) and inductively coupled plasma etching, performed at a pressure of 1.6 Pa and with an etch gas mixture containing SF_6 [flux 60 SCCM (SCCM denotes cubic centimeter per minute at STP)] and C_4F_8 (flux 65 SCCM). The sample has a triangular air-hole pattern with a lattice constant $a=700$ nm and a hole radius $r=180$ nm [Fig. 1(b)]. On top of the PhC a poly(methyl methacrylate) (PMMA) film is spin cast containing 1% in weight of colloidal HgTe NC-QDs. The ~160 nm thick PMMA protects the NC-QDs from oxidation and, thus preserves their luminescence efficiency.[11] Additionally, it improves the symmetry of the devices refractive index profile by pushing the center of the optical modes from the Si/SiO_2 interface toward the Si waveguide core by 7 nm [according to the refractive index of PMMA at 1.4 mm wavelength of 1.53 (Ref. 16)], which increases their interactions with the

a)Electronic mail: wolfgang.heiss@jku.at.

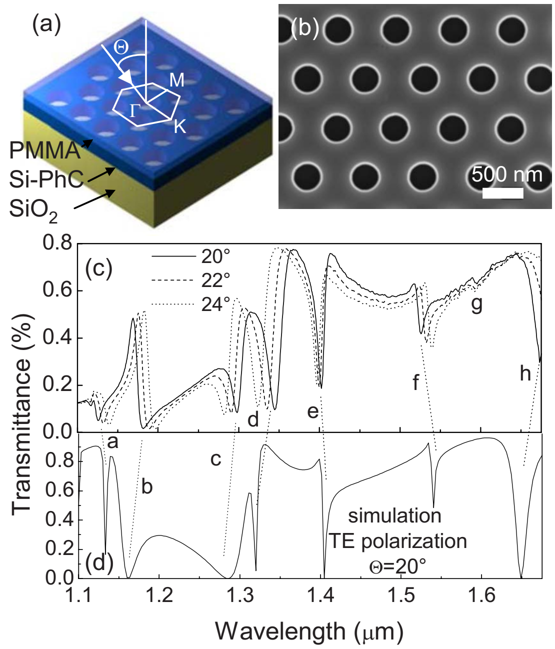

FIG. 1. (Color online) (a) Schematic structure of the 2D photonic crystal covered by a PMMA-film containing 1% HgTe NCs. Indicated also is Θ, the angle of incidence and the high symmetry points in the reduced Brillouin zone Γ, K, and M. (b) Scanning electron micrograph of the fabricated sample. (c) Transmittance spectra at various angles of incidence for *S*-polarization. The spectral anomalies are labeled a–h. (d) Simulated transmittance spectrum for TE polarization in qualitative agreement with the experimental data in (c).

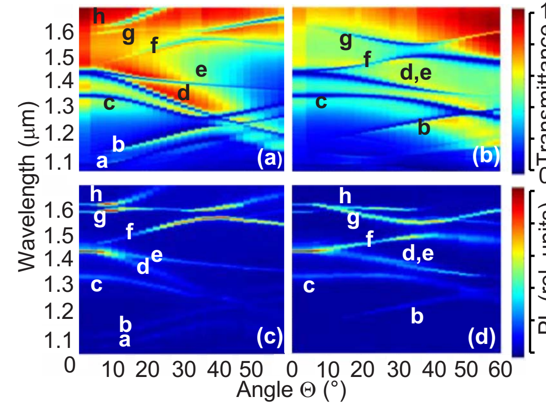

FIG. 2. (Color online) Angle-resolved optical characterization of the PhC covered by 160 nm PMMA, containing 1% HgTe NCs, along the Γ-K direction. (a) Contour plots of the transmittance spectra vs angle of incidence for (a) *S* and (b) *P* polarization. [(c) and (d)] Contour plots of the PL for *S* and *P* polarizations.

HgTe NC-QDs placed in the holes of the PhC.

The HgTe NC-QDs with a mean size of to 4.5 nm were synthesized in aqueous solution via a room temperature reaction between $Hg(ClO_4)_2$ and H_2Te gas, with thioglycerol as stabilizer.[15] After the synthesis the hydrophilic ligands are exchanged to docanethiol, a hydrophobic thiol, to make the NC-QDs soluble in PMMA. The HgTe NCs show bright PL at room temperature with a quantum yield up to 40% (Ref. 15). The size of the colloidal HgTe NC-QDs is chosen to be about 4.5 nm to obtain their emission at wavelengths around 1.4 μm, within the transparent region of Si. The emission wavelength region of the chosen HgTe NC-QDs shows a good overlap with the leaky modes of the designed PhC, at normalized frequencies[17] (a/λ) above the light line and the optical band gap of the PhC.

The optical characterization of the PhC devices was performed with a combined setup for transmittance and PL experiments. The core of it is a goniometer on which the sample is fixed in front of a pin hole to allow rotation around the Γ-M axis of the PhC by $\Theta = \pm 60°$. $\Theta = 0°$ represents the direction normal to the samples surface [see Fig. 1(a)]. Since the sample was uniformly patterned over an area of 1 mm^2 no microscope setup is required. For transmittance experiments white light from a halogen lamp is collimated onto the sample and it is focused by a lens onto the entrance of a fiber spectrometer, equipped with an InGaAs detector line array. *S* and *P*-polarized responses (*S* and *P* denote the direction of the electromagnetic waves electric field vector perpendicular or parallel to the plane of incidence) are analyzed by a linear polarizer. To examine the 2D PhC-assisted extraction of the colloidal NC-QD fluorescence, far-field PL detection is carried out in the same setup, by focusing the 514 nm line of an Ar^+ ion laser onto the pattern, i.e., the same point as the halogen lamp is focused. Switching between PL and transmittance is performed by blocking one of the excitation beams.

To determine the optical band structure of the PhC covered by a NC-PMMA film, transmittance spectra are measured for various angles of incidence. The measurements probe solely the leaky modes, which lie within the light cone of free photons with in-plane vectors $k \leq \omega/c$ (Ref. 18). The external excitation of leaky modes has been shown to cause sharp resonant anomalies such as maxima, minima, and dispersive forms in reflectivity,[18] which are consequently found also in transmittance. Examples for these spectral features are shown by the spectra obtained for $\Theta = 20°$, 22°, and 24° (and *S* polarized light) in Fig. 1(c), where they are labeled by small letters (from a to h). All these spectral features (except a small dip in the experimental spectrum labeled g) are in qualitative agreement with the simulated transmittance spectrum shown in Fig. 1(d). The latter is based on a rigorous coupled wave analysis, performed by making use of a commercial software (DIFFRACTMOD v1.0, Rsoft Design Group, Inc.) and the refractive index dispersion of Si. Depending on the polarization, with changing Θ the energies of these features shift and their strengths and line shapes change. The angular dependence of the resonant features directly maps the energy dispersion of the leaky modes in the reduced Brillouin zone,[18,19] whereby the angle of incidence, Θ, corresponds to the in-plane photon wave vector by $k_{\parallel} = (\omega/c)\sin\Theta$. Thus the energy dispersion can be directly seen in the contour plots in Fig. 2, exhibiting the transmittance as function of wavelength and Θ for *S*-polarized (a) and *P*-polarized light (b). There the resonant spectral transmittance minima form dark lines, clearly exhibiting different angle dependencies for the two orthogonal polarization directions. In defect-free, lossless systems, the leaky modes should be associated with 100% reflection upon external excitation,[13] causing the observed minima in transmittance.

By PL also the energy dispersion is mapped, whereby the leaky modes result in spectral maxima. These maxima, shown in the contour plots in Figs. 2(c) and 2(d) for *S* and *P*-polarized light (bright lines), exhibit angular dependencies exactly matching those found by the transmittance experiments. The PL maxima arise because the eigenmodes above light line (leaky modes) have high field strength within the

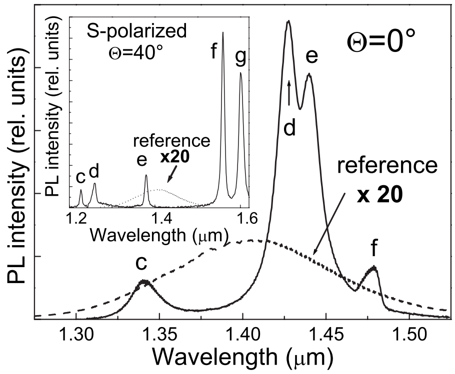

FIG. 3. PL spectra from the HgTe NCs on top of the 2D PhC and on a flat Si reference for vertical emission. The inset shows the PL spectrum for *S* polarization and $\Theta = 40°$ in comparison to the reference obtained for 0°.

PhC slab whereas they are of propagating nature in free space. Thus the emission of the NC-QDs, which are located in the air holes can easily feed these modes with a large portion of their radiating power. This coupling of the isotropically radiated emission of the NC-QDs into the leaky modes results also in a spatial redistribution of the radiation[20] and in a substantial enhancement of its apparent emission intensity.

To quantify the enhancement effect, in Fig. 3 the emission spectrum from an unpatterned area of the device is compared with one obtained from the PhC structure for $\Theta = 0°$. While the reference spectrum from the flat Si surface basically resembles the emission from the HgTe NC-QDs, exhibiting a single emission line with a peak around 1400 nm and a width of about 150 nm, the spectrum from the PhC structure shows four maxima, corresponding to the leaky modes c–f. Most important is that the emission is strongly enhanced at all wavelengths, even though the radiation is emitted perpendicular to the periodic structure of the 2D PhC. This vertical emission is enabled due to slow Bloch modes close to the Γ-point, the center of the first Brillouin zone.[21] For the resonance mode d at 1430 nm the emission intensity is 80 times higher as from the reference. With increasing angle the emission of unpatterned 2D slabs usually decreases, following a cosine-shaped dependence. For the PhC a more complicated angle dependence is observed and some modes even become more intense with increasing Θ. The modes g and h exhibit their highest intensity, e.g., at $\Theta \sim 10°$ and the mode f at 40°, where it exhibits an anticrossing with the g resonance [Fig. 2(c)–*S* polarization]. Most pronounced is the enhancement of the f resonance at the rotation angle of 40°, where the resonance is found at 1560 nm and its intensity is six times higher as at 0°. The intensity of the reference for 0° at this wavelength is, however, quite small so that for the enhancement factor a huge value of 650 is obtained (see inset in Fig. 3). This actual value is substantially higher than what is reported for PhC enhanced fluorescence performed in the visible spectral region.[13] The large difference in extraction efficiency between the patterned and the smooth surface can be assigned to the fact, that in the latter case the NC-QDs excite mostly guided modes in the waveguide slab with a large portion of their emission power through evanescent fields. While the light is trapped by the guided modes, the leaky modes in the PhC modes provide an effective escape way into the free space.[22]

In conclusion, we have investigated the infrared emission from colloidal HgTe NC-QDs embedded in a polymer thin film, deposited on top of a silicon-based 2D PhC structure. By angle-resolved transmittance and PL the device leaky mode dispersion above light line is determined. Resonant to the leaky modes the luminescence is enhanced by two orders of magnitude due to strong coherent scattering effects. Since the extraction enhancement is obtained over a broad spectral range, the combination of infrared emitting NC-QDs with 2D PhCs might be applied, e.g., to reduce the amount of light trapped in light emitting diodes[20] resulting in future Si-based high-brightness light sources.

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