# Nanocrystal Based Microcavity Light Emitting Devices

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Highly luminescent, colloidally synthesized semiconductor nanocrystals are used to fabricate microcavity light emitting devices operating around 650 and 1540 nm. They consist of a Bragg interference mirror from standard optical materials, an active layer consisting of either CdSe nanocrystals for operation in the visible or HgTe nanocrystals with a strong emission in the telecommunication wavelength range, a spacer layer to achieve the desired cavity length, and a metallic top mirror. Both types of devices give highly directional, narrow mode emission with a beam divergence below 2° clearly evidencing that the nanocrystal emission couples to the microcavity resonances. Furthermore, operation up to 75°C is demonstrated without degradation of the nanocrystals, which is promising for potential applications.

## Introduction

Semiconductor nanocrystals (NCs) synthesized by wet-chemical methods have been investigated because of their strongly size-dependent electrical and optical properties. The high flexibility of the synthesis processes allows one to obtain NCs with different shapes and with energy band gaps ranging from the mid-infrared to the UV depending on both the elemental composition and the size of the particles. Moreover, the very high photoluminescence (PL) quantum efficiencies close to unity achieved at room temperature make NCs ideal candidates to improve the performance of light emitting devices. Therefore, NCs have been used to increase the efficiency of plastic light emitting diodes and to obtain single photon sources operating at room temperature. In addition, the operation of NC-based laser devices such as distributed feedback- and microring lasers has been demonstrated. Recently, NCs have not only been used to realize spherical resonators with tremendously high quality factors but also to achieve ultra-low threshold continuous wave lasing at room temperature. While the emission of single NCs, films of NCs and spherical resonators is evenly distributed over all directions in space, in the present work we investigated microcavity light emitting devices, giving strongly directional emission from NCs in continuous wave operation mode. The microcavity devices contain as active material either CdSe NCs for operation in the visible at wavelengths around the damping minimum in plastic optical fibers or HgTe NCs with a strong emission in the telecommunication wavelength range.

## Design and Fabrication

In the microcavity structures either CdSe or HgTe NCs are placed between a Bragg interference mirror and a metallic mirror. The devices were designed by transfer matrix calculations using the optical constants of the individual layer materials, determined by transmission spectroscopy of single reference layers.

Primarily, for the HgTe based microcavity devices (MC-HT), dielectric Bragg interference mirrors consisting of five pairs of $TiO_2$ and $SiO_2$ layers with a thickness of a quarter of the design wavelength of 1540 nm were deposited on a glass substrate by ion assisted electron beam evaporation. This results in mirror reflectivities of 95.5% in the center of the stop band. On top of the reflector ending up with a $SiO_2$ Bragg layer, a densely packed film of HgTe NCs was deposited by the layer-by-layer assembly method. A sequence of 20 bilayers of poly(diallyldimethylammonium chloride) and HgTe NCs results in a smooth and closely packed film with a thickness of 50 nm. The NC-film was covered by a spacer layer of $SiO_2$ to achieve (a) the desired cavity length for a single resonance right at the center of the Bragg mirror stop band as well as (b) a small refractive index at the interface to the metallic mirror on top of the structures to obtain a high overall reflectivity. The thickness of the spacer layer is varied to tune the wavelength of the cavity resonance. On top of the structures a 200 nm thick silver mirror was deposited by DC magnetron sputtering.

For the CdSe based devices (MC-CS), Bragg mirrors also consisting of five pairs of $TiO_2$ and $SiO_2$ layers but designed for a wavelength of 650 nm were deposited on a 22 microns thick, freshly cleaved sheet of pristine mica, whose surfaces are parallel to each other. On the other side of the mica substrate a film of CdSe NCs acting as the optical active material in our devices was deposited and covered by PMMA to protect them during deposition of the metallic top mirror.

## Results

All devices were characterized by reflectivity measurements clearly showing a broad Bragg mirror stop band with a maximum reflectivity at the stop band center of 95.5% in perfect agreement with the expected value from our design simulations.

For the MC-HT devices a $SiO_2$ spacer layer thickness of 170 nm results in single resonator mode right at the center of the Bragg mirror stop band at 1540 nm with a full width at half maximum (FWHM) of only 19.5 meV. By increasing the spacer layer thickness and thus the cavity length, the resonance can be deliberately detuned to lower photon energies [1].

In contrast to that, more than 30 narrow modes are found within the stop band around 650 nm in the MC-CS devices due to the mica sheet thickness of 22 microns [2]. Each resonator mode consists of a double peak structure with a width of 8.5 meV and a spacing of 5.8 meV whose energy splitting is found to scale with the thickness of the mica sheets [2], [3], indicating that it is solely caused by the birefringent properties of the used mica sheets.

For PL experiments, the samples were optically excited by commercially available laser diodes in cw operation mode at wavelengths of 980 and 532 nm at room temperature. For the MC-HT devices a single peak is found, with a FWHM close to 20 meV, which is about eight times smaller than the emission of a NC reference sample (Fig. 1 (a) (solid curves)). The peak energy coincides with the resonance energy deduced from the reflectivity experiments, clearly evidencing that the NC emission couples to the microcavity mode. By increasing the cavity length from 170 nm to 220, 270 and 320 nm $SiO_2$ spacer layer thickness, the emission of the microcavity can be spectrally tuned over a large range of nearly 90 meV towards lower energies, as shown in Fig. 1 (a).

Decreasing the cavity length results in the same tuning range but towards higher energies (not shown here). Thus, the microcavity emission can be tuned over a total range of 177 meV (1400-1750 nm), only limited by the width of the NC PL (Fig. 1 (a) (dashed curve)) which is comparable to that of the Bragg mirror stop band. Furthermore, temperature dependent PL experiments reveal only a moderate decrease of the device emission, so that at 75 °C still one third of the intensity at 25 °C is observed [1]. This decrease is solely caused by an increase of nonradiative recombination and not by

decomposition of the NCs, because after cycling the temperature several times up and down the same PL intensities are obtained.

The PL spectrum of a MC-CS device is presented in Fig. 1 (b) (solid curve) together with that of a NC reference layer (dashed curve). Obviously, the envelope of the device emission matches well to that of the reference layer, both exhibiting its maximum around 1.9 eV (650 nm).

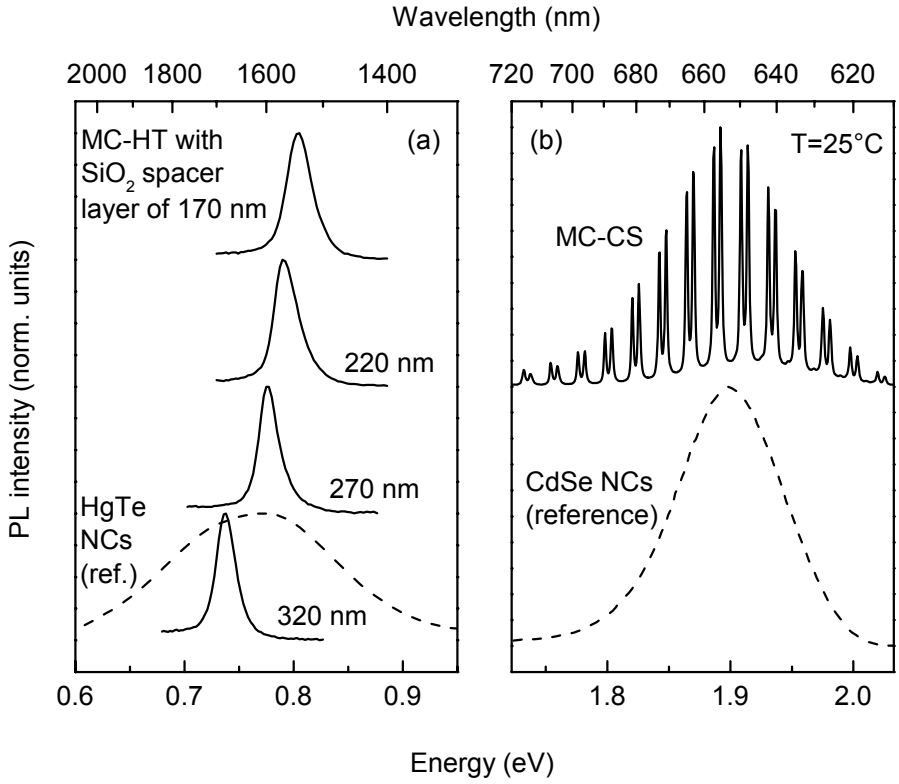


Fig. 1: Room temperature PL spectra of (a) four HgTe based devices (MC-HT) with different cavity lengths ($SiO_2$ spacer layer thicknesses of 170, 220, 270 and 320 nm, respectively) and (b) a CdSe based device (MC-CS) with a cavity length of approximately 22 microns. For clarity the spectra are shifted in vertical direction with respect to each other. In (a) as well as in (b) the PL spectrum of a NC reference layer is shown as a dashed curve.

The device spectrum, however, shows 30 pronounced resonator modes with a spacing of only 19 meV and a width of each double peak of 8.5 meV so that almost 50 % of the NC PL spectrum is covered by the modes. Therefore, a large fraction of the NC PL is extracted from the device.

As a final point we discuss the angular dependence of the spontaneous emission which was determined in the same way as described in Ref. [2]. As depicted in Fig. 2, the experimental angular dependence can be well fitted by a Gaussian function with a half width at half maximum of 0.65° for the MC-CS device (MC-HT devices show similar results). For emission angles of only 2° off from the surface normal (see sketch in the inset), the emission intensities essentially drop to zero. Therefore, the narrow forward directed emission in combination with the symmetric beam profile and the narrow spectral width, resulting from the geometry of our cavities, is very promising for future applications of such devices.

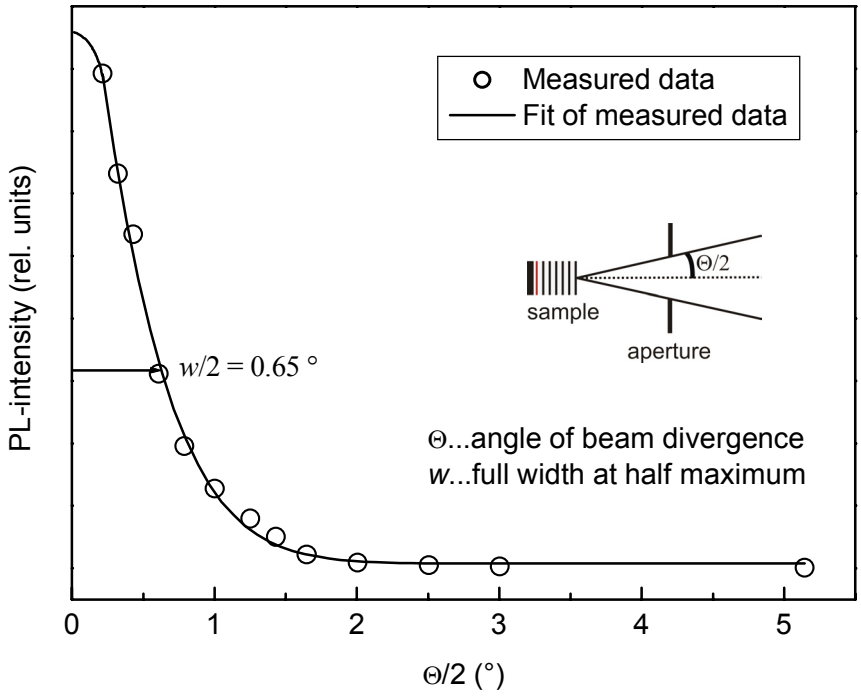


Fig. 2: PL intensity in dependence of half-angle of divergence. Measured data is represented by circles and the fit of measured data by a solid curve. This Gauss distribution results in a total beam divergence of 1.3°.

## Conclusion

In conclusion, we have demonstrated microcavity light emitting devices containing either CdSe NCs for operation in the visible at wavelengths around the damping minimum in plastic optical fibers or HgTe NCs with a strong emission in the telecommunication wavelength range. The CdSe based devices show emission from several resonances around 650 nm, with a small mode spacing, solely caused by the large cavity length of 22 microns. This allows one to extract a large fraction of the nanocrystal luminescence from the cavity. In contrast, the optical distance between the mirrors of the HgTe based devices is reduced to obtain a single resonance at the Bragg mirror stop band center. By varying the length of the microcavity, the resonator mode can be well tuned within the Bragg mirror stop band. Since the width of the NC PL is almost as large as the width of the stop band, also the microcavity emission can be tuned between 1.4 and 1.75 microns. This easily enables the adaptation of our devices to applications demanding a certain emission wavelength. Furthermore, the microcavity emission is strongly forward directed and a beam divergence of only 1.3° is demonstrated. Due to the simple and cost effective fabrication of the microcavity devices combined with their favorable optical emission properties with excellent stability also above room temperature, such devices might be attractive alternatives to currently available commercial laser diodes operating in this spectral range. For this electrical excitation is envisaged in the near future.

## Acknowledgements

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## References

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