Lasing from InGaP quantum dots in a spin-coated flexible microcavity

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Abstract: We report the realization of a mechanically flexible microcavity laser emitting at 657 nm using spin coating. These optically pumped vertical cavity surface emitting lasers use InGaP colloidal quantum dots as the active medium and alternating polymer layers of different refractive indices as the Bragg mirrors. Results of photoluminescence measurements indicating enhancement in spontaneous emission are presented. We also demonstrate the possibility of peeling the device off the substrate yielding a flexible structure that can conform to any shape and whose emission spectra can be mechanically tuned. This new class of hybrid lasers combines advantages of organic and inorganic materials.

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OCIS codes: (140.3948) Microcavity Devices; (250.5590) Quantum Dot Devices; (140.7270) Vertical Emitting Laser.

References and links
spectral properties and wide range of potential applications in bio-imaging/sensing, displays, molecular beam epitaxy and metal有机 chemical vapor deposition. Fabricated using traditional and expensive semiconductor growth techniques such as wavelength range are important for realizing such systems. Most red emitting VCSELs are these POFs have a local attenuation minimum at 650 nm and hence VCSELs emitting in this (POF) are becoming the choice for low-cost short haul optical communications systems. Today, applications for short-range fiber optical communication such as Gigabit Ethernet have replaced edge emitting lasers by VCSELs as sources and plastic optical fibers and displays. Since their initial conception in 1979 [1], vertical cavity surface emitting lasers (VCSELs) have found numerous applications ranging from free space, plastic optical fiber, multi and single channel communication systems, scanning, printing, machine vision, optical switching and displays. Today, applications for short-range fiber optical communication such as Gigabit Ethernet have replaced edge emitting lasers by VCSELs as sources and plastic optical fibers (POF) are becoming the choice for low-cost short haul optical communications systems. These POFs have a local attenuation minimum at 650 nm and hence VCSELs emitting in this wavelength rage are important for realizing such systems. Most red emitting VCSELs are fabricated using traditional and expensive semiconductor growth techniques such as molecular beam epitaxy and metal organic chemical vapor deposition. Colloidal QDs have garnered much attention in the recent times due to their attractive spectral properties and wide range of potential applications in bio-imaging/sensing, displays,
telecommunications and quantum cryptography. They allow spin-coating based processing, the possibility of self-assembly, compatibility with silicon platform and tunability in a wide array of materials with specific absorption and emission properties. Most device demonstrations using such QDs have used either cadmium (Cd) based or lead (Pb) based systems and the toxicity of these materials are a concern. These semiconductor nanocrystals produce bio-hazardous wastes, specifically classified under Restriction of Hazardous Substances Directive (RoHS). InGaP (Indium gallium phosphide) QDs obtained from Evident Technologies used in the present work is an attractive low-toxic alternative due to the absence of heavy metal components.

Embedding photon emitters in microcavities alters their emission properties due to the ability of these structures to confine and enhance electromagnetic fields. Recently there have been several attempts to achieve this goal with colloidal QDs by embedding them in distributed feedback structures, poly (methymethacrylate) spheres, silica microspheres, one dimensional microcavities, two and three–dimensional photonic crystals and microdisk structures [2-16]. From the stand point of realizing a compact laser, such structures are important due to their smaller footprint and more importantly their ability to decrease the lasing threshold due to the smaller optical mode volumes. Indeed lasing has been observed from colloidal QDs by embedding them in a variety of microcavity structures such as microspheres, distributed feed back structures, cylindrical cavities, and microring structures [2-7].

The simplest class of such microcavities is the one dimensional microcavity consisting of a cavity layer sandwiched between two sets of Distributed Bragg Reflectors (DBRs) which comprise of alternating layers of materials with different refractive indices. Despite the simplicity of these structures, they have found wide range of applications in photonic devices such as VCSELs, microcavity LEDs, detectors and filters. Most DBRs for VCSEL structures are fabricated using techniques such as MBE, MOCVD, plasma enhanced chemical vapor deposition, or sputtering. Recently, techniques such as self assembly of block copolymers, co-extrusion of two polymers and spin coating have been used to realize DBR and distributed feedback (DFB) lasers in organic medium [17-24]. Spin coating has also been extensively used to realize organic lasers wherein the emissive organic material is spin coated on to Bragg mirrors, DFB structures, two dimensional photonic crystals, and photopatterned into microdisk structures [25-32]. Optically and electrically pumped VCSEL devices demonstrated to date in organic and inorganic material systems have used solid substrates with the exception of the recent demonstration of surface emitting lasers that utilized two dimensional photonic crystal based reflectors with a thick (~ 1μm) organic dye based gain medium to realize a flexible laser structure [33, 34]. Other demonstrations of flexible lasers in the past have used distributed feedback mechanism to realize lasing [19, 35]. In contrast, we present a one dimensional microcavity structure consisting of a half wavelength thick cavity sandwiched between two distributed Bragg reflectors to realize the VCSEL.

Here we demonstrate the fabrication of a solution-processed mechanically-flexible optically pumped VCSEL emitting at 657 nm. The entire VCSEL structure, including the DBR mirror, is fabricated via spin coating on a glass substrate. A schematic drawing of the one-dimensional microcavity is shown in Fig. 1(a). Alternating layers of polymers of two different refractive indices were stacked to form the DBR mirrors [20-24]. The high and low refractive index polymers chosen were poly-N(vinylcarbazole) (PVK), and cellulose acetate (CA), with refractive indices of 1.683 and 1.475 at 600 nm respectively. Solvents were chosen such that the solvent for one polymer does not dissolve the other. PVK is soluble in non-polar solvents such as chlorobenzene, whereas CA is soluble in polar solvents such as alcohol.

The microcavity structure was realized on a glass substrate. Alternating layers of poly-N(vinylcarbazole) (PVK), and cellulose acetate (CA) of quarter wavelength thickness were stacked to form the DBR mirrors. Here, PVK was dissolved in Chlorobenzene (28 mg/ml) and CA in diacetone alcohol (30 mg/ml). The PVK layer was spun at 3000 RPM for 40 seconds and then placed on a hotplate at 80°C for 15 minutes, resulting in thicknesses of ~ 90 nm. Following this the ~ 100 nm thick CA layer was spin coated at 4500 RPM for 40 seconds, and placed on a 120°C hotplate for 15 minutes to remove the solvent. This process is repeated...
to obtain the DBR stack mirror. Following the realization of the twenty period bottom DBR, a PVK cavity layer of $\lambda/2n_{PVK}$ thickness (~190 nm) embedded with InGaP/ZnS core/shell QDs is spin coated at 2000 rpm. Here, $n_{PVK}$ is the refractive index of PVK. The InGaP/ZnS core/shell QDs dispersed in toluene (2.0 mg/ml) having peak emission wavelength of 650 nm and nanocrystal diameter of ~ 6 nm was obtained from Evident Technologies. The concentration of QDs in PVK was optimized to obtain the maximum emission intensity, which was found to be 30% v/v of colloidal QDs with respect to the PVK solution (39 mg/ml) by dispersing 0.3 ml of the QDs in 1 ml of PVK solution. This corresponds to a fill factor of approximately 2% of QDs in the cavity layer. Following this, ten and half period DBR consisting of alternating PVK/CA layers was realized on top of the cavity layer by spin coating. The thicknesses of the layers were controlled through spin speed and concentration of the polymer. The layer thicknesses were calibrated separately using reflectivity measurements prior the fabrication of the entire structure.

Following the fabrication of the microcavity structure, it was peeled off the glass substrate to form a free standing microcavity. Photographs of the free standing microcavity as well as on a cylindrical surface are shown in Fig. 1(b). These photographs clearly demonstrate the ability of these flexible microcavities to conform to any shape.

The optical microcavity was characterized using reflectivity and photoluminescence (PL) measurements. All optical measurements reported here were carried out at room temperature. Fig. 1(c) shows the experimental reflectivity of the microcavity structure at normal incidence. Reflectivity measurements were carried out using a fiber coupled Tungsten-Halogen lamp as the white light excitation source. Light from the excitation source was focused to spot size of approximately 0.5 mm in diameter and the reflected light was collected by a fiber coupled spectrometer. The spectral position of the cavity mode was designed to overlap with the emission maximum of the InGaP/ZnS core/shell QDs. The quality factor of the microcavity was found to be ~ 70. The inset of Fig. 1(c) shows the normalized reflectivity spectrum of a stand alone DBR consisting of 20 periods at normal incidence at five different locations. Greater than 95% reflectivity and a 100 nm stop-band was obtained using twenty periods of the structure. The reflectivity spectrum also showed excellent uniformity across the sample. The maximum reflectivity observed from the entire structure in the vicinity of the region where lasing was observed was 99%.

Steady state PL measurements were carried out using the 488 nm line of an Argon-Ion laser as the excitation source. The emission from the microcavity sample showed cavity linewidth limited narrower emission with the integrated spectral intensity being ~ 2 times that of QDs in free space (Fig. 2(a)). Results of angle resolved PL measurements are shown in Fig.
2(b). The emission spectrum follows the cavity mode till 40°. Beyond this point, the emission maximum of the QDs lies outside the stop band of the DBR and hence does not show any angle dependence. The above measurements were carried out with the microcavity laid out on a flat substrate. Following this, the microcavity was wrapped around glass cylinders with different radii. The PL emission collected at 20° was compared for the three cases and is shown in Fig. 2(c). Clearly, the emission wavelength blue shifts and intensity of emission decreases upon bending the microcavity to smaller radii. As expected, this effect was observed only for non-normal collection angles since light being emitted normal originates from the cavity region that is not curved by the bending.

![Figure 2](image)

**Fig. 2.** (a) Steady state photoluminescence spectrum of the InGaP quantum dots in the microcavity (red). Luminescence maxima corresponding to the cavity mode and the band edge modes are indicated. The luminescence spectrum of the bare quantum dots in toluene is also shown for comparison (black). (b) Angle dependent photoluminescence spectrum of the microcavity device on a flat substrate. The emission spectrum follows the cavity mode till 40°. Beyond this point, the emission maximum of the quantum dots lie outside the stop band and hence does not show any angle dependence. (c) Photoluminescence observed at 20° collection angle from the microcavity wrapped around cylinders of different radii. The emission wavelength is found to blue shift and intensity of emission decreases upon bending the microcavity to smaller radii.

Time resolved PL measurements carried out on the InGaP QDs in toluene show bi-exponential decay with lifetimes of 16 ns and 85 ns. These lifetimes were found to decrease to 10.8 ns and 78 ns when the QDs were embedded in the PVK host. This is attributed to increased nonradiative processes that occur due to surface degradation when the QDs are dispersed in the PVK host. These lifetime measurements were carried out on spin coated samples with identical QD concentrations on glass substrate. More detailed study on the effect of the host matrix on luminescence properties of these QDs is currently underway.

To characterize the lasing properties of the flexible microcavity structure, the device was optically pumped using the second harmonic of an NdYAG laser (532 nm) with 5ns pulses operating at 10 Hz. The emission from the device was collected using a fiber coupled spectrometer. The input pump power was varied using a variable attenuator. Figure 3 shows the average output power as a function of average pump power. As can be seen from Fig. 3, the lasing threshold is at 27 mW and the slope efficiency (top emission) is ~12%. At pump power greater than 50 mW, the laser output was found to saturate primarily due to heating effects. The variation of the full width at half maximum (FWHM) at the lasing wavelength as a function of pump power is also shown in the inset of Fig. 3. The FWHM shows a threshold behavior consistent with the light output versus input measurements. Also shown in the inset is the emission spectra obtained above the threshold. The observation of threshold in light output as well as significant linewidth narrowing above threshold provides clear evidence of lasing in the flexible microcavity discussed here [36].
This demonstration of a new class of hybrid optically pumped VCSEL realized through a simple spin coating technique that combines the advantages of organic and inorganic systems represents a significant step towards low-cost, mechanically flexible lasers that have efficient luminescence properties, can cover a wide spectral range, and can conform to any shape. Through design of appropriate charge injection scheme, an electrically pumped version should be realizable. In addition to lasing, we have also demonstrated the possibility of tuning the emission wavelength of the flexible microcavity through mechanical bending. Such flexible microcavities are expected to play a key role in reconfigurable photonic switches and conformal optical apertures.

Acknowledgments

This work was supported by the US Army Research Office (Grant no. W911 NF-07-1-0397). M. L. acknowledges support from the DOD Center for Nanoscale Photonics at the City College of CUNY. We thank M. Gerhold, L. Deych, A. Genack, and A. Lisyansky for useful discussions. We also acknowledge Evident Technologies for providing the InGaP quantum dots used in the present work.