

Solution processed microcavity structures with embedded quantum dots

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Abstract

The authors report the fabrication of a one-dimensional microcavity structure embedded with colloidal CdSe/ZnS core/shell quantum dots using solution processing. The microcavity structures were fabricated by spin coating alternating layers of polymers of different refractive indices (poly-vinylcarbazole—PVK, and poly-acrylic acid—PAA) to form the distributed Bragg reflectors (DBRs). Greater than 90% reflectivity was obtained using ten periods of the structure. The one-dimensional microcavity was formed by sandwiching a λ/n thick defect layer between two such DBRs. The emission of the quantum dots from the microcavity structure demonstrated directionality following the cavity mode dispersion and spectral narrowing. Room temperature time-resolved photoluminescence measurements carried out on this structure showed significant reduction in the photoluminescence decay time which is attributed primarily to nonradiative mechanism originating in the presence of the PVK host matrix. The photoluminescence decay time of the quantum dots was found to be ~ 1000 ps while for the quantum dots embedded in the polymer host and the microcavity were 400 and 150 ps, respectively.

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Colloidal quantum dots (QDs) have garnered much attention in the recent times due to their attractive spectral properties leading to wide range of potential applications in bio-imaging/sensing, display, telecommunication and quantum cryptography [1–5]. In comparison to self-assembled QDs fabricated using techniques such as molecular beam epitaxy, the colloidal chemistry technique offers more versatility and lower production cost. The colloidal QDs allow

spin-coating based processing, possibility of self-assembly, compatibility with silicon platform and tunability for a wide array of materials with specific absorption and emission spectra. By providing a wide range of functional wavelengths without having to change material systems, the colloidal chemistry route gives the photonic device designer a new and very valuable degree of freedom. In addition, through proper functionalization of their surfaces, the colloidal QDs allow ordered lateral self-assembly which is very difficult to achieve in the case of QDs grown using ultra high vacuum techniques such as molecular beam epitaxy (MBE).

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Embedding photon emitters in a periodic dielectric environment alters their emission properties [6,7]. This is due to the ability of the periodic structures to confine and enhance electromagnetic fields by several orders of magnitude. The simplest class of such periodic structures is the one-dimensional photonic crystals, more popularly known as Distributed Bragg Reflectors (DBRs) which comprise of alternating layers of materials with different refractive indices. Despite their simplicity, they have found wide range of applications in photonic devices such as vertical cavity surface emitting lasers (VCSELs), microcavity LEDs, detectors and filters. Most DBRs for inorganic materials are fabricated using techniques such as MBE, 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 periodic structures in organic medium [8–11].

While colloidal QDs have become one of the most attractive nanoscale fluorescent emitters, they have still not found widespread application in practical photonic devices. This is in part due to the difficulty in incorporating these QDs into photonic structures. Recently there have been several attempts to achieve this goal by embedding QDs in poly (methylmethacrylate) spheres, silica microspheres, one-dimensional microcavities, two and three-dimensional photonic crystals and microdisk structures [12–17]. Enhancement of spontaneous emission has also been observed from colloidal QDs embedded in the one-dimensional structures [15]. Most of the one-dimensional microcavity structures reported to date have used sputtered or thermally evaporated DBR mirrors. While they do give good reflectivity, often they do have undesirable effects on the optical properties of the QDs through surface defects. In addition, these techniques also require multiple deposition systems – one for colloidal quantum dots and the other for the mirrors. Hence, it is highly attractive to develop a technique for the fabrication of the microcavities which is compatible with the standard solution processing techniques of the colloidal quantum dots. Furthermore, solution processing allows the fabrication of the microcavity on wide variety of substrates including flexible ones.

Here we demonstrate the fabrication of a solution-processed one-dimensional polymer microcavity structure embedded with colloidal QDs. The entire structure, including the DBR mirrors were fabricated using spin coating. A schematic drawing of the microcavity structure is shown in Fig. 1. The microcavity structure is formed by introducing a PVK layer of λ/n thickness

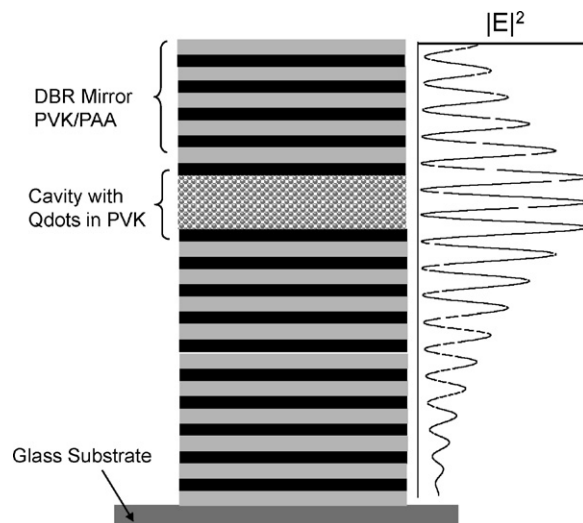


Fig. 1. Schematic drawing of the all-polymer microcavity structure along with the simulated intensity of electromagnetic field in the structure.

with embedded QDs as the defect layer between two DBRs. Transfer matrix simulations were carried out to design the microcavity structure and estimate the reflectivity and quality factor. The simulated electromagnetic field profile in the microcavity structure is also shown in Fig. 1.

The microcavity structure was realized on a glass substrate. Alternating layers of polymers of two different refractive indices were stacked to form the DBR mirrors [18]. To achieve high reflectivity and large stopband, the polymers for the DBR structure were chosen such that they have relatively high refractive index ratio. The high and low refractive index polymers chosen were poly-*N*(vinylcarbazole) (PVK), and poly acrylic acid (PAA), with refractive indices of 1.683 and 1.428 at 600 nm respectively. Solvents were chosen such that the solvent for one polymer does not dissolve the other polymer. PVK is soluble in non-polar solvents such as toluene or chlorobenzene but not in polar solvents such as water or alcohol, whereas PAA is soluble in alcohol but not in chlorobenzene. The polymers PVK and PAA with concentration of 0.28×10^{-4} and 3.12×10^{-2} M were spin coated at 4000 and 6000 rpm, respectively. Thin films of quarter wavelength thickness of the two polymers were alternately spin coated on a glass substrate to make the DBR structure. The bottom mirror consisted of ten periods of the DBR. A PVK cavity layer of λ/n_{PVK} thickness embedded with CdSe/ZnS core/shell QDs is sandwiched between two such DBRs. The cavity of λ/n thickness was obtained by spin coating the CdSe/ZnS QDs dispersed in the PVK solution at 3000 rpm. The CdSe/ZnS core/shell QDs dispersed in

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