Two and One Dimensional Light Propagation in Layer-by-Layer Deposited Colloidal Nanocrystal Waveguides

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Optical waveguides containing high concentrations of colloidal nanocrystals have been fabricated by layer-by-layer deposition on planar and patterned glass substrates. The two- and one dimensional waveguiding in these structures is demonstrated by propagation loss experiments. The experimental results obtained for various film thicknesses and widths of the waveguide stripes indicate that the losses are dominated by surface roughness. The deposition on the structured samples does not lead to any additional losses. This fact and the exceptionally high content of nanocrystals make these structures highly suitable for photonic applications like laser or optical amplifiers.

Introduction

Colloidal semiconductor nanocrystals (NCs) [1] – [3] give a widely size tunable room temperature luminescence [4], [5] with quantum efficiencies close to unity [6], and they are soluble in various organic and inorganic solvents [7]. All these characteristics provide a flexible platform for the development of integrated photonic devices like optical amplifiers and lasers operating at room temperature and over a broad spectral range. The main difficulty in achieving NC-based lasing is the very efficient nonradiative Auger recombination [8], so that laser operation is achieved so far only with pulsed excitation [9] – [11]. Strategies to improve the laser performance are (a), to increase the NC concentration in the active layer in order to increase the gain [12], (b), the use of high-finesse optical feedback structures [9] – [11], and (c), the optimization of the quality of the optical waveguides. Therefore, here we present a systematic investigation of the optical properties of two (2D) and one dimensional (1D) waveguides (WGs) optically activated by strongly luminescent colloidal CdTe NCs. To obtain structures with smooth surfaces and high NC volume fractions the layer-by-layer deposition technique [13] is applied. The waveguiding properties are investigated by propagation loss measurements. These measurements are done for waveguides with different parameters (width, thickness).

Sample Preparation

The NC WGs are fabricated by controlled deposition of CdTe NC/polymer films onto glass substrates by the layer-by-layer assembly method. This makes use of the alternating adsorption of (sub)monolayers of positively charged poly(diallyldimethylammonium chloride) (PDDA) molecules and negatively surface-charged CdTe NCs,

each provided in aqueous solutions. The total film thickness is controlled by the number of PDDA/CdTe NC bilayers and by the deposition time for each monolayer. For a film with 40 bilayers we obtain a typical thickness of 120 nm. Since the effective thickness of a PDDA molecule (approx. 1 nm) is several times smaller than that of the used CdTe NCs with a diameter of approximately 3.2 nm [14], the layer-by-layer technique allows to obtain exceptionally high NC concentrations (close to 60% in volume as determined from the optical film density).

For the fabrication of 1D WGs, the NC/polymer bilayer films were deposited on substrates which were patterned with grooves, with a width of 5, 10, 20 and 40 µm and a length of 2 cm. The grooves with a depth of 450 nm were wet chemically etched by a buffered HF solution whereby a 30 nm thick Cr layer was used as etch mask (see inset of Fig 1(g)).

Results and Discussion

The waveguiding properties of the NC/polymer films are studied by propagation loss measurements, making use of the NC luminescence. In particular, an Ar-ion laser was used to excite the NCs from a direction perpendicular to the sample surface while the photoluminescence (PL) emitted in lateral direction is collected from the sample edge by a microscope objective. The PL spectra are recorded as a function of the distance z, measured between the excitation spot, which is moved by a mirror, and the edge of the sample (see inset in Fig 1(c))

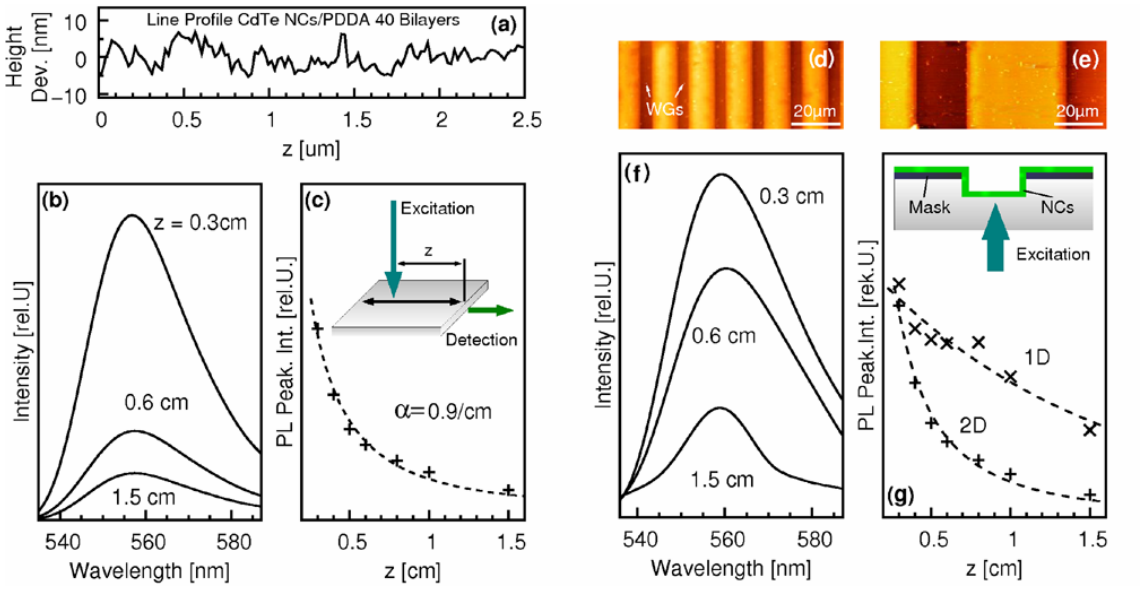

Fig. 1: (a) Surface scan of 2D 40 BL sample, (b) spectra recorded at different excitation distances (2D), (c) PL Intensity over several distances (2D), (d) and (e) AFM micrographs of 1d samples, (f) spectra recorded at different excitation distances (1D), (g) PL Intensity over several distances (1D)

Figure 1(b) shows the PL spectra for various distances z between excitation point and sample edge, revealing that increasing z results in a decrease of the PL intensity, regardless of the emission wavelength. Therefore, it is sufficient to examine the dependence of the peak intensity, as is given in Fig. 1(c). There, the experimental data are fitted by a function which is proportional to $(1/z)e^{-\alpha z}$ (solid line). Here the factor 1/z accounts for the intensity drop of a radial wave, originating from a point source, within a

2D slab. The exponentially decaying part is ascribed to the intensity drop caused by losses in the WG.

The luminescence spectra observed for the NCs in the grooves (Fig. 1(f)) exhibit a very similar shape than those for the planar films. The decay of the PL intensity with increasing propagation distance z, however, is by far smaller as in the 2D case. This is shown in detail in Fig 1(g), where the dependence of the peak PL intensities on z for the 1D and the 2D case are compared. For the 1D in particular, the decay can be fitted by a single exponential one, without the 1/z prefactor, which we quote as an indication for the truly 1D waveguiding of the light in the NC/PMMA film deposited on the patterned substrates. The resulting loss coefficients are the same for 1D and 2D samples, evidencing that purely 1D waveguiding occurs in the patterned samples.

To show that the WG losses are dominated by the surface roughness, we investigated 1D WGs with various layer thicknesses and widths. Reducing the thickness from 40 to 10 NC/PDDA bilayers results in an increase of the loss coefficient by a factor of 2. This is almost independent on the width of the substrate grooves, varied between 5 and 40 µm.

Conclusion

In summary the high potential of layer-by-layer deposited NC/PDDA films for applications in optical devices is demonstrated. In these films, deposited on planar as well as on patterned glass substrates two and one dimensional waveguiding is observed with penetration length of several centimeters.

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