Inverse opals of lead sulfide nanocrystals for optical limiting applications

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Development of new materials with advanced optical properties is critical to the implementation of ultrafast networks and processors through optical modulators, limiters, switches, and routers.[1] Using purely optical means, all-optical limiters would allow transmission near unity at normal light intensities, while instantaneously decreasing it to minimal levels when the material is illuminated with a high intensity pulse. All-optical limiters can be produced from photonic crystals in which one of the component materials possesses optical nonlinearity.[1] Upon illumination, this photorefractive component changes its refractive index as function of light intensity. The stopband, defined as the wavelength range in which light is forbidden from propagating the photonic crystal, shifts its wavelength range as the photorefractive component changes its refractive index. Therefore, the all-optical limiter acts to limit light transmission at that wavelength range.

The incorporation of semiconductor nanoparticles with strong nonlinear optical properties in the photonic crystal offers a promising means to obtain all-optical limiters. Lead sulfide nanocrystal quantum dots (PbS NC) possess large nonlinear refractive index changes with a fast response time (picosecond) which is tunable in the telecommunication window.[2] This presentation reports on the synthesis of polymer colloid particles, the fabrication of photonic crystal, the infiltration of PbS NC into the photonic crystal and characterization of the resulting nanocomposite structure.

In a first stage, we synthesized polymer colloid particles with submicrometer sizes, which were used as the building blocks for the fabrication of photonic crystals. Since the formation of high quality crystals is of utmost importance, we have synthesized colloid particles with a monodispersity below 5% standard deviation. Using convective self-assembly, colloid particles assembled under optimized conditions, such as colloid crystallization temperature and particle concentration in the dispersion.[3] Crystals with sharp-edged and low transmission stopbands (~50% transmission and ~80 nm FWHM) were selected for the infiltration of PbS nanoparticles.

In the second stage, we synthesized PbS NC and characterized their structural and optical properties using TEM, spectrophotometry and ellipsometry. Next, we explored ways in which to infiltrate PbS NC into the photonic crystal. We considered using capillary forces to infiltrate PbS NC into interstitial spaces, infiltration under reduced pressure, and spin coating of PbS NC solution into photonic crystals. The approach to the infiltration of PbS NCs into photonic crystal determined the uniformity and fraction of PbS NC in the interstitial spaces. This seemingly simple step posed serious challenges in preserving the quality of the crystal and in obtaining a high fraction of PbS into the photonic crystal with uniformity.

To date, we found that the infiltration method utilizing capillary forces preserves the quality of the photonic crystal. Figure 1 shows the transmission spectrum of the colloid crystal before and after infiltration of PbS NC using capillary forces. Before infiltration, the crystal shows a sharp-edge and low transmission stopband (curve 1 centered at 1120 nm). Upon filtration of the crystal with PbS nanoparticles, the refractive index contrast between the colloids and the interstitial space decreases. As result the diffraction strength decreases and the stopband assumes a decreased attenuation (curve 2 centered at 1200 nm). We estimated the fraction of PbS NCs in photonic crystal by comparing the spectral shift in the stopband before and after infiltration. Using the multiple scattering method[4], we calculated the effective refractive index of the voids in the infiltrated photonic crystal (simulated curve 4 of Figure 1). The value of estimated refractive index of 1.33 is smaller than the refractive index of the pure PbS suggesting a 50% infiltration in the interstitial space. The SEM image of Figure 2 depicts the infiltration using capillary forces. The image demonstrates a gradient in PbS NC concentration along the height of the photonic crystal.

As another approach to increasing the PbS NC fraction and enhancing the uniformity in the photonic crystal, we explored the codeposition of water-soluble PbS NCs with polymer colloids. In this approach, PbS NCs were mixed with a colloid dispersion and self-assembled using convective assembly. Figure 3 is an SEM image depicting the uniformity and high PbS NC fraction obtain via codeposition. Other approaches, which are currently being investigated, include the growth of colloid crystal in confinement followed by introduction of PbS nanoparticles into the crystal by microfluidics means.

To summarize, on the basis that PbS NCs can be infiltrated into photonic crystals while preserving the optical properties of photonic crystals, we showed that inverse opals of lead sulfide nanocrystals show promise as all-optical limiters. We continue to optimize the fabrication of inverse opals of PbS NC in parallel with the measurement of the nonlinear optical response of our devices.

References:

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Figures:

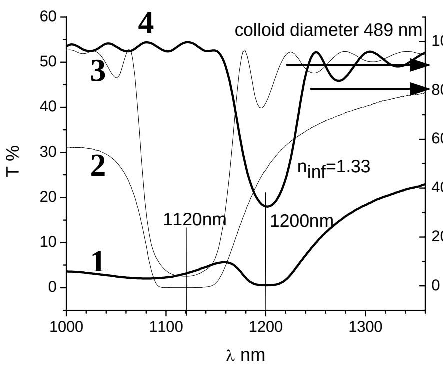

Figure 1: Optical transmission of photonic crystals without PbS NC (1), the corresponding simulation based on multiple scattering methods (3) with PbS infiltrated by capillary forces (2) and the corresponding simulation based on multiple scattering method (4).[4]

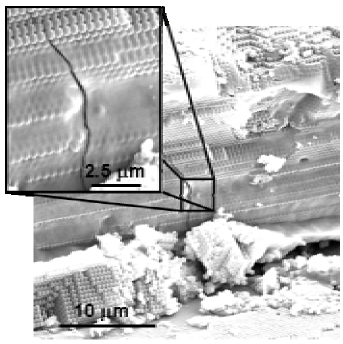

Figure 2: SEM image of a photonic crystal infiltrated with PbS NC using capillary forces.

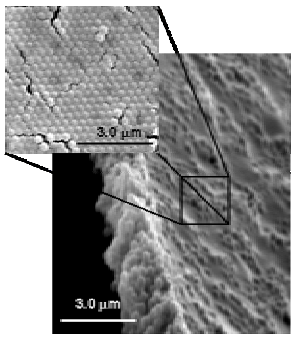

Figure 3: SEM image of a photonic crystal codeposited with water soluble PbS NC.