Spontaneous emission from semiconductor nanocrystals in coupled spherical microcavities

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Spherical particles of a few micrometers in diameter can act as three-dimensional optical resonators providing the feedback required for enhanced linear and nonlinear optical processes such as photoluminescence (PL) and Raman scattering [1,2]. Polymer latex microspheres containing semiconductor quantum dots (or nanocrystals (NCs)) are promising candidates for the development of advanced sources, which can allow the manipulation of photons on the optical wavelength scale. In this work, we present a detailed experimental study of whispering gallery mode (WGM) structure in a single spherical microcavity containing CdTe nanocrystals, and of the evanescent coupled optical microcavity modes in a photonic molecule (PM) formed from two interacting microspheres. In our experiments we used melamine-formaldehyde (MF) latex microspheres of uniform size, 2.04±0.03 μm. A layer-by-layer deposition technique provides controllable coating of the microspheres with a shell of close-packed CdTe NCs of approximately 5 nm in diameter. Using low intensity excitation of a single MF/NCs microsphere by an Ar^+ laser (488 nm), a periodic (ripple) structure with narrow regular peaks was observed in the PL spectra and attributed to the strong field enhancement at the microcavity resonances. The shape, position and separation of the WGM peaks were theoretically analyzed over a wide spectral region using Mie theory.

To investigate the propagation modes in interacting spherical microcavities (a PM) we have measured PL spectra scanning a sample along the longitudinal axis of the PM. It was found that two major phenomena contribute to the observed lineshape of the mode structure. Firstly, superposition of uncoupled modes results in a pronounced double structure with the intensity distribution dependant on the excitation position. This observation demonstrates the possibility of optical interconnection using an array of microcavity waveguides. Secondly, distinct triplet structure was observed in the spectra, which is a result of splitting of a monosphere resonance. This structure, we believe, can be explained as a result of strong coupling between the photonic states of the two spherical microcavities forming the PM.

Time-resolved measurements showed slight modification of the spontaneous emission rate for NCs in a PM when compared to the spontaneous emission rate for NCs coating a single microsphere. We have also observed more than a 10-fold increase in the PL lifetime of the PM with increasing detection wavelength. This growth in lifetime is much higher than that observed for a planar film of close-packed NCs. Optical feedback via the WGMs of spherical microcavities can provide an increased probability of energy transfer to the emitting species, and in our case, the observed lifetime increase can be attributed to enhanced energy transfer between NCs of different sizes in a photonic molecule.

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2. Yu.P. Rakovich, J.F. Donegan, N. Gaponik, A.L. Rogach, Appl. Phys. Lett. **83** (2003) 2539.