Paramagnetic and fluorescent manganese-doped PbS nanocrystals

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The controlled incorporation of dopant impurities in a single colloidal nanocrystal (Quantum dot, QD) is a challenging field of research with potential for numerous applications in nanotechnology. Of particular interest is the incorporation of 3d transition metal ions (Mn, Co, etc) whose d-shell electronic configurations imprint the nanocrystal with unique magnetic and optical properties [1]. Here we report the successful synthesis in aqueous solution of colloidal manganese-doped PbS nanocrystals. Our "one pot" synthesis approach produces nanoparticles that combine within one structure, paramagnetic properties of transition metals (see EPR spectra in Fig. 1a) with efficient/tuneable photoluminescence (PL) emission in the near-infrared (NIR) wavelength range (850-1200 nm) (Fig. 1b). Interestingly, the formation of alloyed (PbMn)S nanocrystals also leads to a thermal energy shift of the QD PL emission, which decreases with increasing Mn-content thus leading to a temperature independent emission for Mn-contents of about 10% (see inset in Fig. 1b).

These new functional nanoparticles are of fundamental and technological interest. The solubility of PbS:Mn nanoparticles in physiological solvents and their dual functionality, i.e. NIR luminescence and paramagnetism, open up exciting prospects for future exploitation of these nanocrystals as imaging labels for combined fluorescence and magnetic resonance imaging (MRI). Although chelated Mn-complexes can be employed as contrast agents in MRI, a Mn-doped nanocrystal will be less susceptible to chemical alterations (i.e. less toxic). Thus these nanocrystals will offer unique combined functionalities and imaging opportunities.

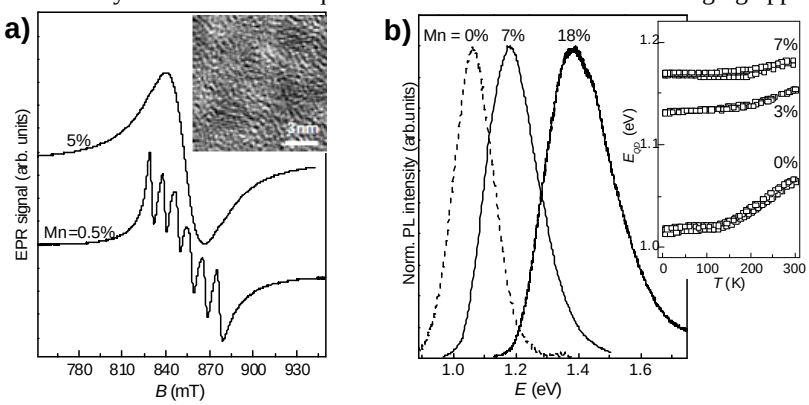

Figure 1 (a) Room temperature K-band EPR spectra for (PbMn)S QDs. Six resonances are observed for samples with Mn < 3% due to hyperfine interaction between the *d*-electrons and the nuclear Mn spins. Inset: HR TEM image of (PbMn)S with 10% Mn. (b) Room temperature photoluminescence (PL) spectra of PbS (dashed line) and (PbMn)S QDs and temperature dependence of the PL peak position (Inset).

[1] S. T. Ochsenbein and D. R. Gamelin, Nature Nanotechnology 6, 112 (2011).