Tunable Broad White Light Emission from Single Size Semiconductor Quantum Dots

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Motivation—The development of highly efficient GaN-based solid-state illumination sources requires new types of phosphors, which absorb near UV energy strongly while re-emitting visible, white light. Phosphors based on semiconductor nanoparticles offer advantages compared to conventional powder phosphors including negligible optical backscattering due to the small diameter of the dots. More importantly, we discovered that direct white emission from a single size dot is possible, obviating the need to blend two or more dots of differing size. Use of a single size dot eliminates self-absorbance of the emitted light at the high optical densities required for full absorbance of the excitation energy while maintaining optical transparency in the visible, 450 to 700 nm regime.

Accomplishment—We have demonstrated tunable broad white light emission from single size CdS semiconductor quantum dots (QDs) (see Fig. 2b). This accomplishment is made possible by our discovery that for sufficiently small CdS and CdSe quantum dots (QDs) of diameters ≤ 2 nm (or ~ 90 formula units) the onset of absorption (determined by quantum confinement, or dot size) and the emission energy, or color (determined by interfacial chemistry) can be independently controlled. The decoupling of these two features has allowed us to widely separate the absorption and emission to eliminate self-absorption of the emitted light and to tune the emission throughout the visible range from a population of single-size dots. In essence, these QDs behave much like organic laser dyes with significant adjustable Stokes shifts resulting in negligible self-absorption.

Key to this discovery is the ability to tailor the interface states of the QDs. This is accomplished in two ways during our inverse micellar synthesis and subsequent processing: (i) by the use of suitable surfactants that bind to selected sites on the QD surface causing changes in the energies and lifetimes of the interface states which determine the emission; (ii) the addition of surface active agents such as suitable electron or hole traps (e.g., by the addition of Zn^{2+} or excess S^{2-} ions, respectively) to a monodisperse population of CdS or CdSe QDs, which also tailor the emission.

Significance—Though quantum confinement of carriers by the finite size of a semiconductor QD can be used to adjust the color emitted, the overlap between absorbance and emission bands in the case of larger QDs leads to strong self-absorption of the emitted light (see Fig. 1). By synthesizing dots so small that a majority of their atoms are at the QD interface, the chemistry of this interface alone determines emission color, while the absorption is fixed by dot size. All the excitation light can then be converted to tunable, visible emission. Compared to conventional white light phosphors, our QDs can be selected by material type and size to have absorbance in the near-UV (360-420 nm) region, while conventional white light phosphors must be excited below 300 nm to emit in the visible (see Figs. 2a & b). This causes an inherently larger energy loss than excitation near 400 nm. The principles determining the optical properties of these new phosphors should extend to other nanosize semiconductors like Si, and, thus, these new nanomaterials have great potential as white phosphors for both solid-state lighting and fluorescence illumination devices.

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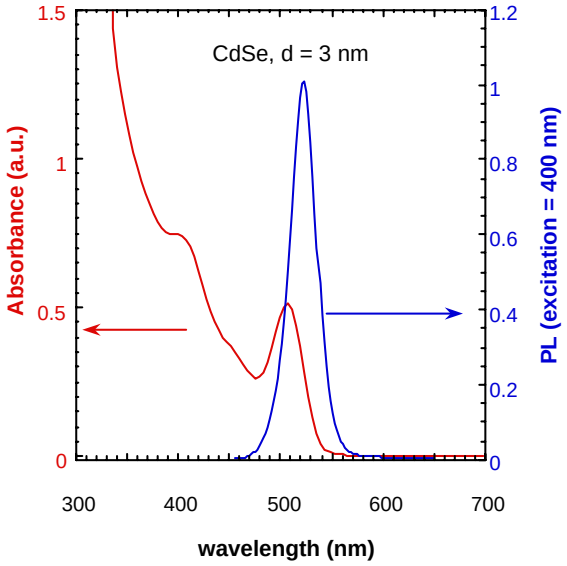

Figure 1. Strong overlap between **absorption** and **emission** leads to self-absorption in large Q dots.

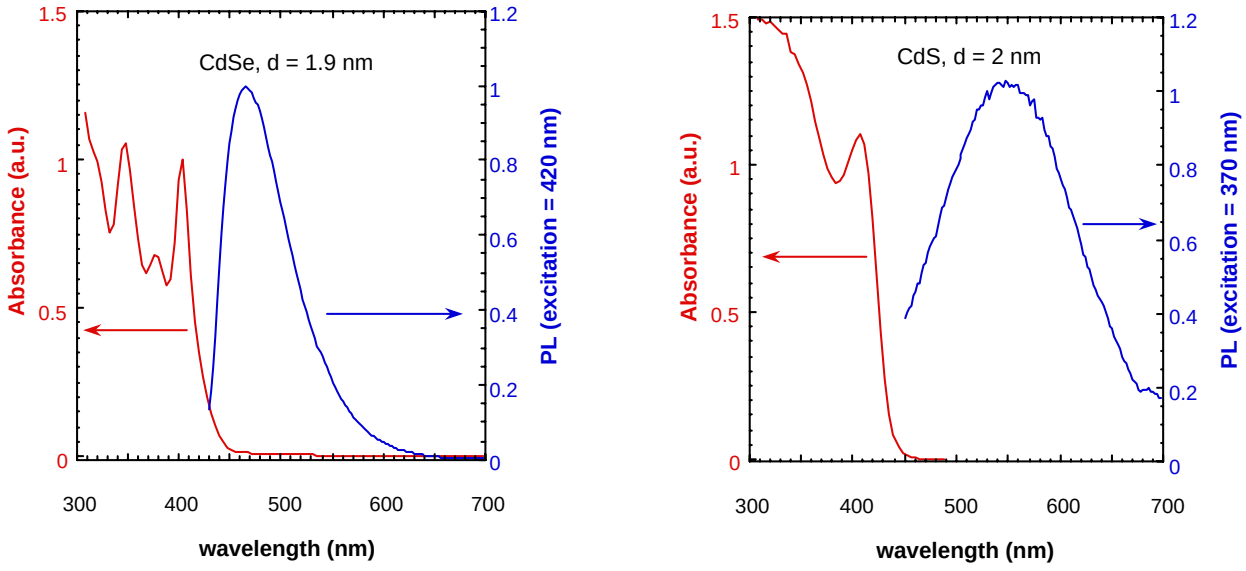

Figure 2. Significant Stokes shift in smaller Q dots results from decoupling of the **absorption** and **emission** and eliminates self-absorption. (a) CdSe, (b) CdS.