White Light Nanophosphors for Solid State Lighting

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Motivation—Development of highly efficient GaN-based solid-state illumination sources requires new types of phosphors that 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 while maintaining optical transparency in the visible, 450 to 700 nm regime.

Accomplishment—It has been established that semiconductor quantum dots (QDs), due to the confinement of the photogenerated electron-hole (e-h) pairs, have absorbance and emission energies determined by the size of the dot. We have discovered that, for sufficiently small CdS or CdSe dots, with diameters, d ~ 2 nm, the enormous interfacial area of the dots and lack of translational symmetry allows one to independently adjust the absorbance onset and the emission energy. The decoupling of these two features allows one to select a material type and size to match the desired excitation source while changing the interface morphology or chemistry to alter the emission color. Thus, we have demonstrated blue, green, and red emission from a single size QD sample without changing its absorbance onset. Such extremely small dots, like organic dyes, have negligible self-absorbance of their emitted light. For solid-state lighting this allowed us to optimize the QD phosphor optical properties to ensure that 90-100% of the ~ 400 nm excitation LED source is absorbed in the required short optical path. This materials development enabled the first demonstration of spectrally broad, white emission from a single size QD of cadmium sulfide (CdS) by simple changes in the interface passivation of the dot. The absorbance edge was selected to match the excitation energy of a Cree™ UV LED, while techniques for incorporating the QDs into a transparent epoxy dome without agglomeration of the dots were developed. The latter is key to maintaining high fluorescent quantum yields. Our white emitting device is shown in Fig. 1, while Fig. 2 compares the spectral energy distribution of our nanophosphor to that of the most commonly used GE "soft-white" conventional phosphor used in fluorescent lighting. (This GE phosphor has no absorbance above 300 nm and so cannot be used with an LED) It is possible to obtain subtle shifts in the tint of the white coloration of our CdS QD nanophosphor by further changes in the surfactant used to passivate the surface, shifting the broad energy distribution to either the blue or the red of that shown in Fig. 2, yielding a blue-white or orange-white nanophosphor. We measured a record quantum efficiency of 60% from the encapsulated blue-white QD phosphors in an actual device.

Significance—The use of near-UV LEDs in white light illumination sources requires novel phosphors which can efficiently re-emit all the absorbed ~ 400 nm excitation. Conventional white light phosphors fail to absorb this energy while larger QD phosphors suffer from self-absorbance, which makes white light production by mixing of blue, green, and red emitting QDs very difficult. We have shown that smaller dots, with over 70% of their atoms at surface sites, can have their emission energies tuned by changes in the interface passivation alone, while their absorbance is determined by their size and remains invariant. The basic scientific principle of separation of absorbance from emission in very small QDs is extendable to novel systems, such as Si or Ge QDs doped with emitting ions like Mn.

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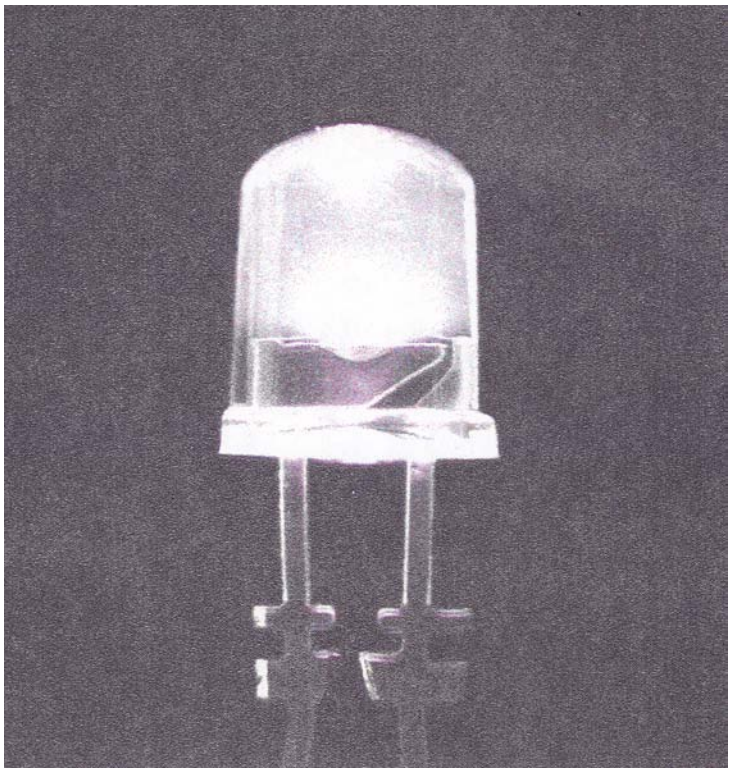

Figure 1. Photograph of a white light emitting LED using a commercial CREE 390 nm excitation LED and d = 2 nm CdS quantum dots incorporated into the epoxy dome.

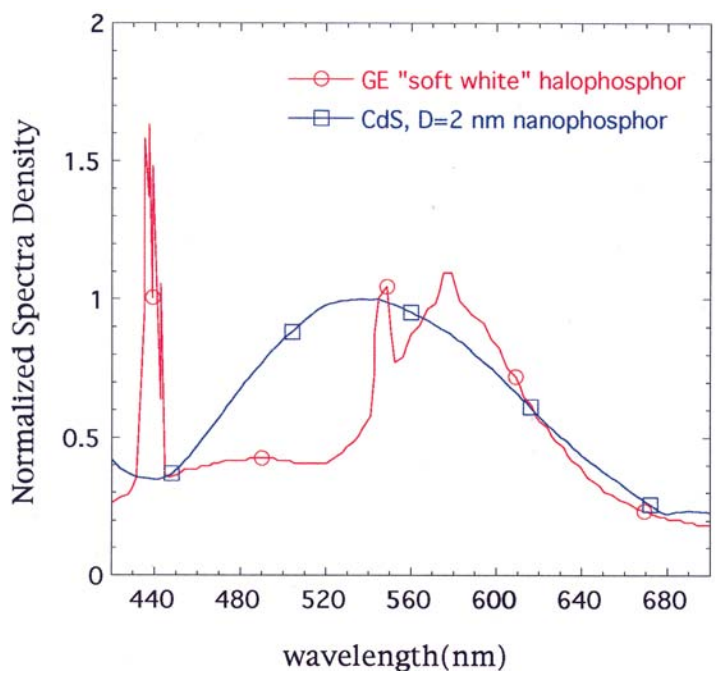

Figure 2. Spectra Energy Density of a white emitting CdS nanophosphor compared to that of a commercial GE "soft-white" phosphor used in fluorescent lamps. The latter must be excited below 300 nm, and thus cannot be used with a GaN-based LED.