Size effects on the fluorescence efficiency of biocompatible CdTe quantum dots.

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In the last years, semiconductor nanocrystals, usually called Quantum Dots (QDs), have attracted intense attention due to their desirable properties, such as a wide absorption band, high emission yield, good chemical stability and possibility of multi-photon excitation [1]. As a consequence of all the outstanding properties, QDs have been already used in numerous applications such as biological labelling and imaging, and photovoltaic and optoelectronic devices [2].

Because of quantum size effects, their optical properties can be tuned just by changing their particle size. This fact has been already used to tailor the emitted colour. Of particular interest is the use of QDs for biomedical fluorescence imaging. These dots should be easily dispersed in biological fluids and so in water. As different QD sizes can be in principle used it is of primordial importance to investigate the size effects on the QDs fluorescence efficiency.

In this work we have investigated in detail how the fluorescence quantum efficiency of CdTe-QDs (which are commonly used for optical bioimaging) depends on the QD size by means of Thermal Lens (TL) spectroscopy. The non-monotonous variation of the fluorescence quantum efficiency with the dot size has been tentatively explained in terms of surface effects that enhance the non- radiative rate for small QDs.

The CdTe-QDs investigated in this work were provided by Plasmachem Inc. Six different CdTe-QDs were used with nominal emission peak wavelengths at 510, 550, 615, 660, 680 and 710 nm, corresponding to sizes of 1.2, 2.3, 3.6, 3.8, 4.3 and 4.7 nm, respectively. The QDs (0.3% in mass) were dispersed in distilled water with no evidence of precipitation. TL measurements were made by using the same experimental set up as in ref [3].

Figure 1, shows a typical TL transient signal for the CdTe-QDs/water solutions, in this case for those emitting at 660 nm (3.8 nm diameter). From the best fit to the theoretically expected TL transient decay (solid line) [1], the so called TL induced phase shift, θ, and the thermal diffusivity are straightforwardly obtained. Then the quantum efficiency, η, is calculated from the Equation 1. In this equation λ_{exc} is the excitation wavelength, $\langle\lambda_{em}\rangle$ is the average emission wavelength, C is a constant related to the host liquid (C=279 W^{-1} in our case), and $\Theta = \theta / P_{abs}$ (P_{abs} being the absorbed pump power).

$$\eta = \left[1 - \frac{\Theta}{C}\right]\frac{\langle\lambda_{em}\rangle}{\lambda_{exc}} \qquad (1)$$

For the particular case of the 3.8 nm, Θ =182 W^{-1}, and considering the excitation (488 nm) and emission (660 nm) wavelengths, a fluorescence quantum efficiency of 0.47 was determined. Then, by analyzing the TL transient signals of CdTe-QDs of the different sizes, we have determined their quantum efficiencies. Figure 2 shows a plot of the quantum efficiency as a function of dot diameter. An

inspection of this plot reveals two different trends: the quantum efficiency rises with decreasing QD size down up to 3.8 nm where a maximum in the quantum efficiency is obtained. Then this trend is reversed so that decreasing sizes produce a decrease in the quantum efficiency. Then, the obtained results indicate that dots of about 4 nm would be the most indicated to be used as luminescent probes for high resolution biomedical fluorescence imaging.

The observed quantum efficiency decrease for dots smaller than about 4 nm indicates that surface effects are playing an important role in the non radiative rate.

References

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Figures

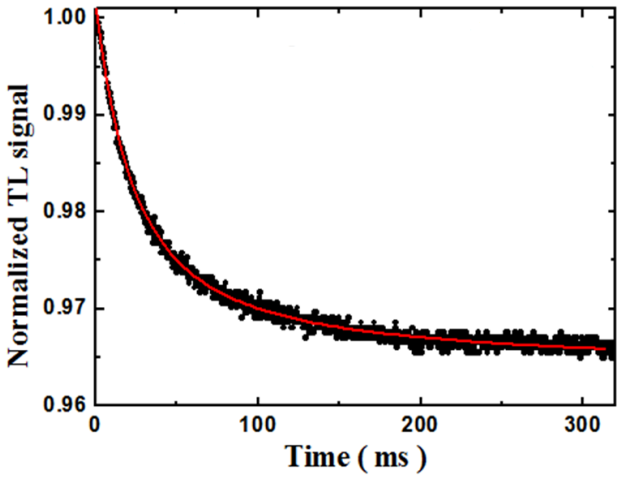

Figure 1. Normalized transient TL signal for CdTe-3.8 nm QDs at room temperature, excitation at 488 nm with a pump power of 0.42 mW. The solid line is the best fit using to the TL theoretical transient decay [1].

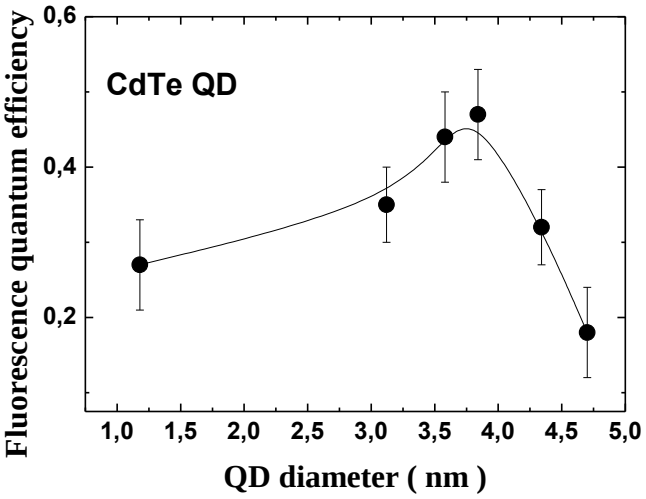

Figure 2. Fluorescence quantum efficiency versus size for the CdTe-QDs (room temperature and 488 nm excitation wavelength).