Thermodynamically Driven Approach Toward Engineering Nanomanufacture of Single-Sized Quantum Dot Molecular Nanocrystal Ensembles

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Colloidal photoluminescent (PL) semiconductor nanocrystals are fascinating materials; over the past decade, they have attracted significant attention for both fundamental science and practical applications. It has been acknowledged that quantum confinement becomes operative when the size of a nanocrystal becomes less than or comparable to that of a photogenerated exciton in its corresponding bulk materials. Particularlly, quantum dots (QDs) are spherical nanocrystals with their excitons confined in three spatial dimensions and bandgap size dependented. Moreover, their optical properties including bandgap absorption and emission can be tuned via size, structure, and composition. For example, the bandgap of CdSe QDs is tunable across the visible range (~400 nm to ~700 nm). In general, QDs are of particular interest in studying fundamental photophysics with great potential in various applications including energy, biological/medical, and sensing.

However, the size-dependent characteristics make regular quantum dots (RQDs) fascinating, but create intrinsic difficulties to study. Such difficulty can be easily understood, with the variation in size within one colloidal ensemble sample giving rise to inhomogeneous spectral broadening, in addition to homogeneous spectral broadening.

The synthesis of colloidal QDs typically requires source compounds, solvents as reaction media, and surfactants as surface ligands providing colloidal stability and surface passivation. Both hot-injection and non-injection-based approaches lead to colloidal nanocrystal ensembles (i.e., RQDs) with a certain degree of size distribution. At present, the formation mechanism of the nanocrystals is far from being understood. At the same time, it is challenging, and for sure a leading-edge area, to design and synthesize colloidal nanocrystal ensembles free of size distribution and thus inhomogeneous spectral broadening.

This presentation will address our latest advances in the design, synthesis, and engineering of single-sized QDs exhibiting bandgap emission with only homogeneous spectral broadening. We now name such novel QDs “molecular nanocrystals”; in the past, we named them magic-sized QDs (MSQDs). They represent the successful precise control of identical numbers of atoms and/or molecules in each nanocrystal. So far, multiple families of MSQDs have been effectively produced for various materials, including CdS, CdSe, and CdTe.

More importantly, this presentation will discuss about the formation mechanism of these MSQDs. Our fundamental understanding brings insights in the molecular compound nucleation (but no growth) of identical MSQDs as well as nucleation and growth leading to the formation of regular quantum dots (RQDs). Such new understanding together with the rational design of non-injection strategies towards large-scale productions with high synthetic reproducibility of various MSQDs lead to potential advancement of various applications including solar cells, solid-state lightings, and other optoelectronic devices. The opportunity in future chemical process engineering development of non-injection-based chemical nanomanufacturing processes, plus “molecular engineering" of MSQDs to create new families of molecular crystals with unique or improved properties, will be also discussed.

References

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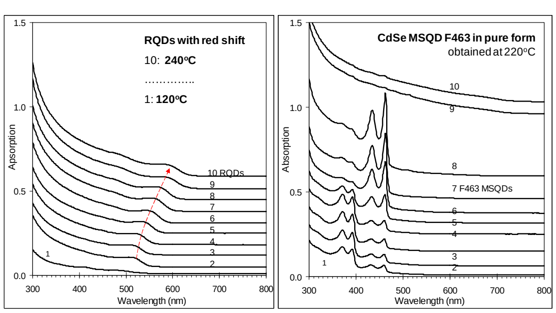

Figure 1. MSQD vs RQD ensembles: different formation mechanisms during our non-injection syntheses.

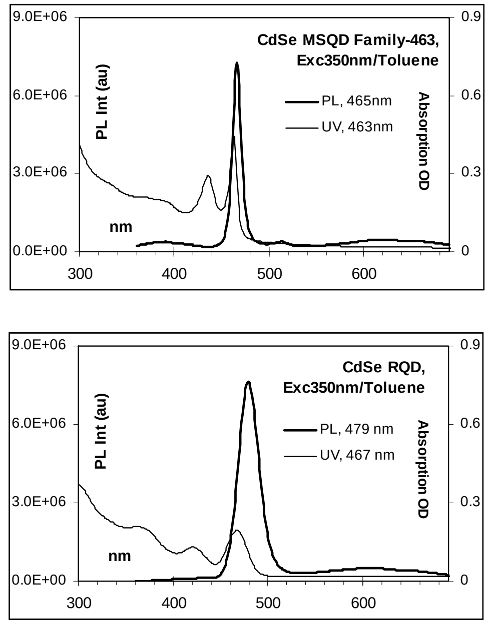

Figure 2. Different optical properties of CdSe MSQD vs RQD ensembles.

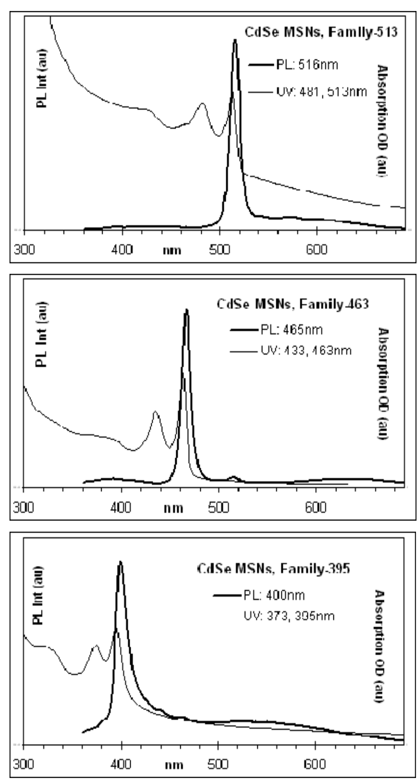

Figure 3. Multiple families of different pure species of CdSe MSQD produced.