Gold plated and thick shell quantum dots: two examples of colloidal quantum dots with much improved optical properties

M. Nasilowski, P. Spinicelli, B. Dubertret*
LPEM, ESPCI, 10 rue Vauquelin, 75005 Paris, France
*e-mail : benoit.dubertret@espci.fr

The quest for the perfect quantum dot (QD) is a drive for both chemists and physicists. Since the landmark synthesis of colloidal semiconductor nanocrystals[1], many studies have tried to understand and limit QD emission blinking in time[2].

The most widely accepted explanation for the emission intensity flickering is the presence of an excess charge, in or in close proximity to the nanocrystal[3] that can recombine non radiatively with the exciton through Auger processes.. This results in heat, lower quantum yield, and fluctuation of the QD emission in time when observed at the single particle level.

Recently, two routes have been proposed to decrease the efficiency of Auger recombination: composition gradient between the core and the shell[4] and thick shell QDs[5]. While this two routes have led to QDs with improved fluorescent properties, they both have their limitations: limited quantum yields for gradient QDs and strongly temperature dependent quantum yield for thick shell QDs.

We present here two new generations of quantum dots with unmatched optical properties. The first one, a CdSe/CdS QD with a thick CdS shell and a gradient interface between the core and the shell, exhibit a quantum yield of 100% at room temperature, with a perfectly stable, non-blinking, fluorescence emission over long periods of time (hours). We measured a similar quantum yield for the monoexciton and for the biexciton, which shows that Auger recombinations are completely suppressed in these QDs. At high excitation powers, these QDs show multiexcitonic emission even at the single dot level, so that single QDs have excitation dependent light emission, another proof of the complete suppression of Auger recombination.

The second one consists of a single quantum dot encapsulated in a silica shell coated with a continuous gold nanoshell.[6] It provides a system with a stable and Poissonian emission at room temperature that is preserved regardless of drastic changes in the local environment. This novel hybrid quantum dot/silica/gold structure behaves as a plasmonic resonator with a strong Purcell factor. The gold nanoshell also acts as a shield that protects the quantum dot fluorescence and enhances its resistance to high-power photoexcitation or high-energy electron beams.

These two types of new QDs bring a rupture in the family of colloidal QDs in the sense that they are the first examples of 100% quantum yield QD at room temperature even at high excitation power, regardless of their charge state and with robust optical properties.

[1] C. B. Murray, D. J. Norris, and M. G. Bawendi, *J. Am. Chem. Soc.*, vol. **115**, no. 19, pp. 8706–8715, Sep. 1993.
[2] M. Nirmal, B. O. Dabbousi, M. G. Bawendi, J. J. Macklin, J. K. Trautman, T. D. Harris, and L. E. Brus, *Nature*, vol. **383**, no. 6603, pp. 802–804, Oct. 1996.
[3] M. Kuno, D. P. Fromm, H. F. Hamann, A. Gallagher, and D. J. Nesbitt, *J. Chem. Phys.*, vol. **115**, no. 2, p. 1028, 2001.
[4] G. E. Cragg and A. L. Efros, *Nano Lett.*, vol. 10, no. 1, pp. 313–7, Jan. 2010.
[5] C. Javaux, B. Mahler, B. Dubertret, A. Shabaev, A. V Rodina, A. L. Efros, D. R. Yakovlev, F. Liu, M. Bayer, G. Camps, L. Biadala, S. Buil, X. Quelin, and J.-P. Hermier, *Nat. Nanotechnol.*, vol. **8**, no. 3, pp. 206–12, Mar. 2013.
[6] B. Ji, E. Giovanelli, B. Habert, P. Spinicelli, M. Nasilowski, X. Xu, N. Lequeux, J.-P. Hugonin, F. Marquier, J.-J. Greffet, and B. Dubertret, *Nat Nano*, vol. **10**, no. 2, pp. 170–175, Feb. 2015.