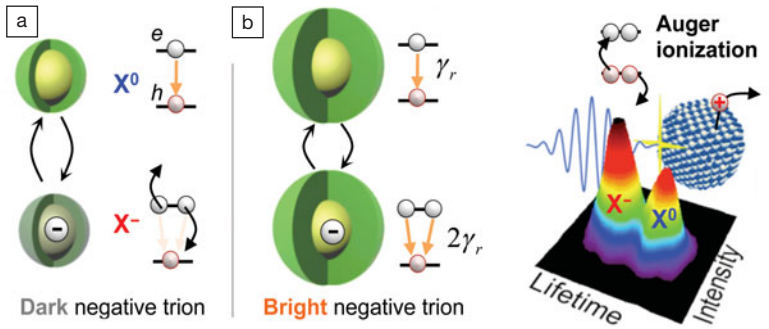
Nano Focus

Lifetime variation in giant nonblinking QDs due to switching between neutral and negatively charged states

Quantum dots (QDs) are attractive as nanoscale light sources, but the fluctuations in emission intensity from individual dots, which is known as "blinking," can limit their application. It has recently been shown that CdS/CdSe core–shell nanoparticles with thick shells do not exhibit blinking. However, as reported by a Los Alamos team, Christophe Galland (now at the University of Delaware), Yagnaseni Ghosh, Andrea Steinbrück, Jennifer A. Hollingsworth, Han Htoon, and Victor I. Klimov in the June 19 issue of *Nature Communications* (DOI: 10.1038/ncomms1916), giant nonblinking QDs (g-QDs) exhibit a pronounced variation in their emission lifetimes due to a switching between negatively charged and neutral states.

In their study, the researchers investigated the optical behavior of individual CdSe/CdS g-QDs with a ≥15 monolayer thick shell. Individual g-QDs produced stable photoluminescence intensities with variations fitting a single-peak Poisson distribution. Such behavior suggests emission occurred from a single state. However, lifetime measurements revealed the presence of two equally weighted lifetimes (19 ns and 39 ns), thereby demonstrating that two distinct states contribute to the emission.



In thin-shell QDs (a) charging is one of the mechanisms causing luminescence intensity fluctuations known as blinking. This is because light emission from the negatively charged exciton (X^-) is quenched by very fast Auger decay (negative trion pathway, curved arrows). In contrast, in g-QDs (b), nonblinking emission intensity is observed because Auger recombination of the negative trion is suppressed. The only signature of charging is a doubling of the radiative decay rate compared to the neutral exciton ($\gamma_r \rightarrow 2\gamma_r$; double orange arrow). This is indicated by the plot of time-resolved photon-counting data on the lifetime-intensity distribution map (lower right). Charge fluctuations are caused by Auger ionization of the g-QD, that is, by the decay of a biexciton through the fast positive trion pathway with ejection of the hole. This pathway is favored in g-QDs because of a pronounced asymmetry in spatial distributions of electron and hole wave functions in these nanostructures.

This behavior was linked to the process of nonradiative Auger recombination and its different effect on negatively charged excitons (negative trions) versus positively charged excitons (positive trions). Neutral QDs correspond to the bright optical state. In thin-shell QDs, charged excitons are essentially nonemissive because Auger decay is fast. However, in g-QDs, Auger processes are largely suppressed for negative trions. By adding a single electron to the QD, the number of radiative recombination pathways is doubled, as is the radiative decay rate. Charging is thus accompanied by a decrease in the photoluminescence lifetime without affecting the intensity ("lifetime blinking").

The investigators confirmed this scenario by studying the effect of controlled electrochemical charge injection on the QD photoluminescence and relaxation rates. At 0 V, the equal distribution between the two lifetimes was again observed. When either −0.5 V or −0.8 V was applied, electron injection was more favorable, and the shorter lifetime (19 ns) predominated without any change in photoluminescence intensity. The research team concluded that observed lifetime fluctuations are connected to random charging of the g-QDs with excess electrons. Moreover, they found that charging occurs by Auger ionization through ejection of a hole. This Auger decay pathway is favored in g-QDs because of a greater degree of confinement for holes than for electrons. These results show that nonblinking behavior is not incompatible with random charge fluctuations in the QD.

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Bio Focus

Wireless PV retinal prosthesis shows promise for restoration of sight

Retinal degenerative diseases lead to blindness due to the loss of photoreceptors even though the inner retinal neurons remain largely intact. Visual percepts, also called "phosphenes," can be produced by electrical activation of the inner retinal neurons. This alternate route to visual information has the potential for restoring sight to the blind. Current retinal prosthesis designs, with electrode arrays implanted in the retina facing either the ganglion cells or the inner nuclear layer, rely on serial telemetry to deliver stimulation signals to the electrodes, requiring bulky receiving and processing electronics and a trans-scleral cable. Surgery is complex and the design is difficult to scale up to attain higher visual acuity. In addition, patients cannot use natural eye movements to scan the visual scene because retinal stimulation patterns are transmitted from an external camera to the retinal implant, independent of eye orientation. These limitations can be overcome by devices that use

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