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Giant absorption enhancement in colloidal quantum dot supercrystals through dipolar coupling

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Nanocrystals are thought to be viable candidates for the fabrication of solar cells, light emitting diodes, photodetectors, ... In such a device context, nanocrystals will always be deposited in thin layers. However, colloidal nanocrystal properties (e.g. absorption cross sections, exciton lifetime, ...) are typically evaluated in solution using effective medium approaches such as the Maxwell-Garnett (MG) model¹. These models assume that the nanocrystals behave as uncoupled dipoles embedded in a host medium with given permittivity. However, it is clear that this assumption no longer holds for a close packed film of nanocrystals where those dipoles tend to couple through electromagnetic multipolar interactions. Using properties measured in solution to evaluate the performance and physics of thin film devices is therefore incorrect, or at least a serious simplification.

The basic idea is that the internal field of a particle in close proximity to other dipoles will be a superposition of the influence of the external field and the induced dipolar fields of the neighbouring particles. We expect that this type of electromagnetic coupling will increase the absorption cross section relative to that of the particles in solution. We therefore define an 'enhancement factor 'E'', being the ratio between the absorption cross section in film to the cross section in solution.

To access the enhancement E experimentally, we measure the absorption spectrum of single and multiple close packed monolayers of nanocrystals on glass substrates using a standard UV-VIS-NIR spectrophotometer. The layers are deposited using Langmuir-Blodgett deposition² and are well defined (i.e. particle density and lattice symmetry are uniform over large areas). The enhancement predicted by our intuitive reasoning is substantial, leading to a size-dependent value of E up to 5 for PbS monolayers. The effect is smaller in CdSe because of the smaller dielectric screening in these materials. The latter probably decreases the influence of a dipole on its neighbours.

Starting from our intuitive coupling idea, we can calculate E from theory as a function of particle size. To achieve this, we use the coupled dipole model (CD model) which was developed to understand the localised plasmonic response of coupled arrays of metallic nanoparticles³. We are able to predict the size-dependence of the measured enhancement, both for CdSe and PbS using a limited number of free fitting parameters.

We use monolayers since they are 2D structures that are easy to model in a scalar approach. In a device context (e.g. a solar cell) nanocrystals will be stacked in multiple layers to achieve a certain thickness (e.g. to absorb sunlight). Our model can be extended to predict the behaviour in these technologically very relevant multilayers. It turns out the enhancement is also present in these stacked systems.

We also expect the coupling in these nanocrystal solids to affect the emission properties. Research is ongoing to investigate collective properties such as sub- and superradiance. This will provide us with a complete picture on the very exciting optical behaviour of quantum dot solids, extending their applicability to new fields as quantum optics. We believe our results have paved the way to a more general understanding of nanocrystal solids, the latter being very relevant for real-life applications of nanocrystals such as thin-film solar cells. We show that in close packed layers, properties such as absorption cross section are not intrinsic to the nanocrystal but are determined by their interactions with other particles. These interactions can be tuned through ligand length and particle size, both of which are easily accessible during the colloidal synthesis or through post-deposition processing (e.g. ligand exchange).

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Santa Fe, New Mexico, USA
May 13 - May 18, 2012

