

## Exchange coupled donor dimers in nanocrystal quantum dots

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Doping of semiconductor nanocrystals (NCs) is a promising means of tailoring the electronic, optical, and magnetic behavior of NCs for application in for example solar cells, thin film transistors, and (opto)electronics and thus major research efforts have been directed toward this topic in recent years [1]. Incorporation of heterovalent donor and acceptor impurities into NCs has already been reported for NCs of ZnO [2], Si [3], SiGe [4], CdSe [5], and InP [6]. Despite these significant advances, the electronic structure of doped NCs is still largely unresolved. A central problem concerns the coupling between dopants when multiple impurities are incorporated in a single NC. In this context, understanding the yet unknown electronic properties of donor dimers in NCs is a crucial first step toward the general comprehension of NC doping. For a donor dimer in a single NC, the two dopants are separated at most by a few nanometers and thus interact very strongly by exchange. This interaction yields discrete electronic levels in a single NC and the impurity subband model applied for bulk semiconductors with equivalent doping concentrations becomes unrealistic. Moreover, in most applications of doped NCs, a macroscopic amount of NCs (NC ensemble) will be used in a single device. Due to the random site location of dopants, dimers in different NCs will exhibit distinct electronic properties. Hence, the net effect of dopants on the electronic properties of NC ensembles will be largely governed by statistics.

In this work, using electron paramagnetic resonance we experimentally probe the exchange of donor dimers in NCs via a deviation of their triplet-state magnetic resonance from Curie paramagnetism. We show that the exchange coupling of the closely-spaced donors can be well described by effective mass theory, which allows the consideration of statistical effects crucial in NC ensembles. Whilst a dimer induces discrete states in a NC, their energy splitting differs by up to three orders of magnitude for randomly placed dimers in a NC ensemble, due to an enormous dependence of the exchange energy on the dimer configuration.

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