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Electronic properties of quantum-dot molecules

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Abstract

We investigate the electronic structure of several double quantum dot systems: (i) hetero-dot artificial molecules built from two CdS chemically synthesized nanocrystals and (ii) two vertically stacked pyramidal self-organized InAs/GaAs quantum dots. The calculations are performed using the empirical tight-binding approach. The results of calculations show significant coupling between the nanocrystals that form a quantum-dot molecule. When two quantum dots are close enough, the strong coupling can split and reorder energy levels, change state symmetries and make substantial changes in optical spectra. Formation of double-dot states having bonding and antibonding character by analogy to diatomic molecules is shown.

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Keywords: Quantum dots; Artificial molecules

By analogy to natural molecules and solids, the complex systems made of ensembles of nanocrystals can be anticipated and experimentally achieved. Techniques of self-organized growth allow for fabrication of two-dimensional and three-dimensional arrays of quantum dots [1]. Formation of close packed quantum-dot solids can also be achieved by methods of chemical synthesis of nanocrystals [2–4]. Absorption and luminescence spectra of arrays of nanocrystals show significant differences from the spectra obtained for non-interacting quantum dots that suggest formation of delocalized electronic states [3,4].

The first step towards understanding the physics of quantum dot solids is to study artificial molecules, i.e., systems built of two coupled nanocrystals. We investigate two such systems, namely (i) hetero-dot artificial molecules built from two CdS chemically synthesized nanocrystals and (ii) two vertically stacked pyramidal self-organized InAs/GaAs quantum dots, with an empirical tight-binding model.

Let us first consider a double quantum dot built of two different CdS nanocrystals, of radii 2.91 nm (NC1) and 1.86 nm (NC2), respectively. Their centers are separated by 4.65 nm. The double-dot contains 5333 atoms, but only one atom is common to both nanocrystals joined by 16 chemical bonds. The larger nanocrystal has denser energy spectrum, with some of its energy levels close to the energy levels of the smaller nanocrystal. One such example is the ground electron state (s-type) of NC2, with the energy 0.298 eV and the first excited electron energy level (p-type, triply degenerate) of NC1, with energy 0.301 eV. Four new double-dot *molecular orbitals* are formed from these four single-nanocrystal states. As with diatomic molecules, two of the states are p-type,

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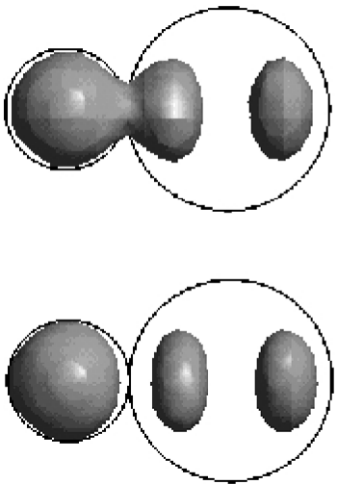

Fig. 1. Density isosurfaces (50%) of electron bonding-like (top) and antibonding-like (bottom) states ($s \pm p_x$) of a double-dot built from two different CdS QDs. Circles mark sizes of each nanocrystal.

localized in NC1 and polarized perpendicular to the double-dot axis. The other states are polarized along this axis and have densities distributed in the entire double-dot. The state with the lowest energy has bonding character, while the other one is an antibonding mixture of p_x(NC1) and s(NC2) states. Their charge densities are shown in Fig. 1.

Now, let us consider two vertically stacked InAs pyramids on wetting layers (WL) embedded in a large GaAs box. The pyramid height is 1.8 nm and the size of its square base is 3.6 nm. Both pyramids together with the WLs contain more than 4700 atoms. The largest GaAs box considered, contains more than 40,000 atoms. We find that the pyramids couple strongly, even if they are separated by several monolayers (ML) of the surrounding GaAs. When the separation distance decreases to 3 ML, the splitting of the electron and hole energy levels becomes comparable to the inter-level energy spacing of the single pyramid and the energy levels can cross. When the top of the lower quantum dot touches the bottom

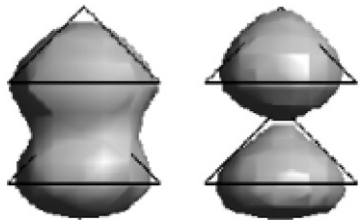

Fig. 2. Density isosurfaces (50%) of electron bonding-like (left) and antibonding-like (right) states of a double-dot built from two pyramidal vertically stacked InAs/GaAs QDs on WLs.

of the upper WL, the splitting of the ground electron level reaches 150 meV. The lowest electron state of such a double-dot has strong bonding character (symmetric-like combination of the wave functions of individual dots). The first excited state reveals clearly antibonding nature, with a nodal plane parallel to the WL. The corresponding charge densities are shown in Fig. 2.

In summary, we have studied coupling effects in two different double-dot systems: (i) artificial molecules built of two CdS chemically synthesized nanocrystals and (ii) two vertically stacked pyramidal self-organized InAs/GaAs quantum dots. Charge densities show the formation of bonding and antibonding-like states in various double-dot systems. The results help to explain differences observed between optical spectra of dense arrays of quantum dots and of non-interacting quantum dots.

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