

QUANTUM DOT MOLECULES AND CHAINS

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Abstract A review of results from theoretical investigations of several systems composed of two or more coupled quantum dots (known as *artificial molecules* or *quantum dot solids*) is presented. All the calculations are performed within an empirical tight-binding theory. It is shown that coupling between nanocrystals can split and reorder energy levels and change state symmetries. The results help to understand and explain differences observed in optical spectra of arrays of quantum dots in comparison to the spectra obtained for non-interacting nanocrystals. We show also how an external electric field influences the properties of coupled quantum dots.

1. Introduction

By analogy to diatomic molecules and crystalline solids, *artificial* and *quantum dot solids* can be built from coupled semiconductor nanocrystals. Coupled dots created by etching techniques or electrical confinement from quantum-well systems were studied first due to their possible applications as single-electron resonant tunneling devices [1, 2, 3, 4, 5, 6, 7, 8]. Systems of vertically stacked self-assembled quantum dots are intensively investigated now to determine how the coupling between nanocrystals in a dense array of such dots influences properties and performance of quantum dot lasers [9, 10, 11, 12]. Linear chains of electrically confined quantum dots have been proposed recently for a realization of multi-qubit gates [13].

The densest ensembles of quantum dots are obtained for chemically synthesized nanocrystals [14, 15, 16, 17, 18]. Kagan et al [14] first found luminescence from close-packed CdSe nanocrystals totally different than

the luminescence of non-interacting dots in a dilute solution. Several other experiments [16, 17] have shown that optical properties of dense arrays of chemically synthesized nanocrystals are significantly different from properties of individual dots.

We present here a review of the results of our investigations on the formation of delocalized states in coupled quantum dots and linear chains of nanocrystals. We work within the empirical tight-binding theory which, as a microscopic approach, is well suited for precise investigation of the coupling on the atomic scale. The presented results help to explain why the optical properties of coupled nanocrystals are so different from the properties of individual quantum dots. We present also some preliminary results showing how an external electric field influences delocalized states in coupled systems.

2. Theory

In the empirical tight-binding approach (TB) the one-particle wavefunction is represented in an orthogonal basis set of atomic orbitals $\phi_\alpha(\mathbf{r} - \mathbf{R}_J)$ [19],

$$\Psi(\mathbf{r}) = \sum_J \sum_\alpha c_{\alpha,J} \phi_\alpha(\mathbf{r} - \mathbf{R}_J), \quad (1)$$

where α denotes an orbital of a given symmetry and \mathbf{R}_J is an atomic site. In our model, each atom is described by 5 orbitals (s, p_x, p_y, p_z , and s^*). We assume that atoms occupy the sites of a zinc-blend structure. Only atoms embedded in a volume of a given system are taken into account. The interaction in the Hamiltonian is restricted to on-site and nearest neighbors only. The Hamiltonian matrix elements

$$t_{\alpha'\alpha R'_J R_J} = \langle \phi_{\alpha'}(\mathbf{r} - \mathbf{R}'_J) | H | \phi_\alpha(\mathbf{r} - \mathbf{R}_J) \rangle \quad (2)$$

are treated as empirical parameters obtained by fitting bulk-band structure to experimentally known band gaps and effective masses. There are 13 different TB parameters for each material. The surface dangling bonds are passivated by shifting their energies high above the conduction band edges. The one particle quantum dot states are found by diagonalizing the TB Hamiltonian matrix with the use of an iterative eigenvalue solver.

To study linear infinite chains of nanocrystals we first define the supercell of a given chain. The TB wavefunction components corresponding to atomic sites at opposite supercell boundaries differ in phase by e^{iqD} , where D is the chain period. Therefore, the TB elements for \mathbf{R}_J and \mathbf{R}'_J at adjacent supercells are $t_{\alpha'\alpha R'_J R_J} = e^{iqD} t_{\alpha'\alpha R'_J R_{J+D}}$. A static external electric field is described by adding $e\vec{F}\vec{r}$, where F is the electric