

Pseudopotential many-body theory of the electronic structure of 10^3 - 10^6 atom quantum nanostructures

Alex Zunger

National Renewable Energy Laboratory, Golden, CO 80401

Semiconductor quantum-dots can now be made either by techniques of colloidal chemistry (yielding nearly spherical 20-50 D diameter, strain-free, surface-passivated dots of Si, CdS, CdSe, InP and InAs) or via epitaxial growth (yielding strained, lens-shape dots of 20-70 D height, embedded in a semiconductor matrix, e.g., InAs/GaAs). Such objects are now in the forefront of research as they were shown last year to be the world's smallest lasers and because they hold promise for quantum-computing and highly efficient solar cells. In the last few years these dots were studied experimentally by a new generation of breakthrough spectroscopies capable of addressing a single dot or molecule with unprecedented resolution of a small fraction of 1 meV. Such studies revealed novel features, unexpected by single-particle theory. Our theoretical work on dots started by shopping-around for available techniques (e.g., k^*p effective-mass), concluding that they are not appropriate for such small dots with strong inter-band and inter-valley coupling. We then developed our own techniques. They involve two steps: (1) the single-particle problem is solved via order-N plane-wave-basis approach to empirical pseudopotentials. This affords a pseudopotential solution of up to one million atom objects without encountering the LDA error. We include strain, surface passivation, arbitrary shapes and inter-band coupling; (2) In the second step, the many-body problem is addressed by calculating all inter-electronic (screened) Coulomb and exchange integrals from the pseudopotential wave functions and using them in a configuration interaction (multiple Slater determinant) expansion. The quantities calculated include the excitonic spectrum, multi-excitons, charged excitons as well as the effects of dot size and shape on their electronic structure. These methods were applied to Si, Ge, CdSe, InP and InAs/GaAs.

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