

DYNAMICS ON THE NANOSCALE: Time-domain ab initio studies of quantum dots and carbon nanotubes.

Device miniaturization requires an understanding of the dynamical response of materials on the nanometer scale. A great deal of experimental and theoretical work has been devoted to characterizing the excitation, charge, spin, and vibrational dynamics in a variety of novel materials, including carbon nanotubes, quantum dots, conducting polymers, inorganic semiconductors and molecular chromophores. We have developed state-of-the-art non-adiabatic molecular dynamics techniques and implemented them within time-dependent density functional theory in order to model the ultrafast photoinduced processes in these materials at the atomistic level, and in real time.

Quantum dots (QD) are quasi-zero dimensional structures with a unique combination of molecular and bulk properties. As a result, QDs exhibit new physical properties such as carrier multiplication, which has the potential to greatly increase the efficiency of solar cells. The electron-phonon and Auger relaxation in QDs compete with carrier multiplication. Our detailed studies of the competing processes in PbSe QDs rationalize why carrier multiplication was first observed in this material.

The electron-phonon interactions in carbon nanotubes (CNT) determine the response times of optical switches and logic gates, the extent of heating and energy loss in CNT wires and field-effect transistors, and even a superconductivity mechanism. Our ab initio studies of CNTs directly mimic the experimental data and reveal a number of unexpected features, including the fast intrinsic intraband relaxation and electron-hole recombination, the importance of defects, the dependence of the relaxation rate on the excitation energy and intensity, and an understanding of the role of active phonon modes.

Our real-time atomistic simulations create a detailed picture of these and related materials, allow us to compare and contrast their properties, and provide a unifying description of quantum dynamics on the nanometer scale.