Influence of Electron Correlations on Quasiparticle Energies and Lifetimes in an Atomic Nanowire Coupled to Electrodes

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Abstract

As electronics devices scale to sub 10 nanometer lengths, the distinction between “device” and “electrodes” becomes blurred. A common approach to the modeling of molecular-scale electronics has relied on a combination of a density functional treatment for electronic structure and the non-equilibrium Green’s function formalism for transport, optionally with use of the GW approximation to correct the single-particle energy levels to account more accurately for screening effects. Recently, the effects of allowing lead excitations couple to molecular device states [1] has been explored and the inclusion of electron-electron interactions on electrodes coupled to a ‘device region’ has been formulated for the NEGF approach to transport [2].

In this work, the interaction between device and leads is studied in a simple model of a molecular tunnel junction. Using a complex absorbing potential [3], we are able to reproduce the single-particle energies of a device region including a description of the effects of the “semi-infinite” electrodes. With this approach, we are able to model the effect of coupling of a quantum device to electrodes while systematically studying the effect of many-electron interactions between the device and lead regions. Varying the device-lead coupling strength, the effect of electron correlation on energy shifts and lifetimes of electronic states on the device region is studied by permitting the electron correlation or “many-electron interactions” to be more accurately treated through the inclusion of an increasing number of many-particle states in a configuration interaction expansion [4]. We find that the prediction of the electronic states of a device region is sensitive to both the amount of device-lead coupling and to the amount of electron correlation that is included in a calculation. The two effects mix in a complicated way, implying that detailed treatments of the electronic structure of nanoscale devices are required to predict electronic behaviour such as charge transport and photoexcitations in a molecular junction.

References

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Figures