Applying the extended molecule approach to correlated electron transport: important insight from model calculations

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Abstract

Theoretical approaches of electric transport in correlated molecules usually consider an extended molecule, which includes, in addition to the molecule itself, parts of electrodes. In the case where electron correlations remain confined within the molecule, and the extended molecule is sufficiently large, the current can be expressed by means of Laudauer-type formulae. Electron correlations are embodied into the retarded Green's function of a sufficiently large but isolated extended molecule, which represents the key quantity that can be accurately determined by means of ab initio quantum chemical calculations. To exemplify these ideas, we present and analyze numerical results obtained within full CI calculations for an extended molecule described by the interacting resonant level model. Based on them, we argue that for narrower band (organic) electrodes the transport properties can be reliably computed, because the extended molecule can be chosen sufficiently small to be tackled within accurate ab initio methods. For wider band (metallic) electrodes, larger extended molecules have to be considered in general, but a (semi-)quantitative description of the transport should still be possible in the typical cases where electron transport proceeds by off-resonant tunneling. Our numerical results also demonstrate that, contrary to the usual claim, the ratio between the characteristic Coulomb strength and the level width due to molecule-electrode coupling is not the only quantity needed to assess whether electron correlation effects are strong or weak.

Keywords: molecular electronics, correlated molecular transport, molecular orbital gating, electrode-molecule contacts, nonequilibrium Green's functions, Keldysh formalism, interacting resonant level model

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