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Beyond the Lorentzian Model in Quantum Transport: Energy-Dependent Resonance Broadening in Molecular Junctions[1] ZHENFEI LIU, Lawrence Berkeley Natl Lab, JEFFREY B. NEATON, UC Berkeley and Lawrence Berkeley Natl Lab — In quantum transport calculations, transmission functions of molecular junctions, as well as spectral functions of metal-organic interfaces, often feature peaks originating from molecular resonances. These resonance peaks are often assumed to be Lorentzian, with an energy-independent broadening function Γ. However, in the general case, the wide-band-limit breaks down, and the Lorentzian approximation is no longer valid. Here, we develop a new energy-dependent broadening function $\Gamma(E)$, based on diagonalization of non-Hermitian matrices within a non-equilbrium Green's function (NEGF) formalism. As defined, $\Gamma(E)$ can describe resonances of non-Lorentzian nature and can be decomposed into components associated with the left and right leads, respectively; and it is particularly useful in understanding transport properties in terms of molecular orbitals in asymmetric junctions. We compute this quantity via an ab initio NEGF approach based on density functional theory and illustrate its utility with several junctions of experimental relevance, including recent work on rectification in Au-graphite junctions.

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