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On the derivative discontinuity in molecular junctions[1]

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Both the wave and particle aspects of the electron play essential roles in transport through single-molecule junctions. The wave character is implicit both in the Landauer formula used to understand nanoscale transport and in the very chemical bonds holding the junction together, while the particle aspect is manifested in phenomena such as Coulomb blockade and shot noise. The dominant computational paradigm for transport in single-molecule junctions involves local or semilocal approximations to density functional theory combined with nonequilibrium Green's functions. This approach does exceptionally well at describing the wave aspect of the electron, but fails to describe the particle aspect—due to the omission of the derivative discontinuity in the exchange-correlation potential that arises in the limit of vanishing lead-molecule coupling. To understand the role of the derivative discontinuity in molecular junctions, we investigated the transport and occupancy of a simple Anderson model of a molecular junction. We showed[2] that the exact single-particle Kohn-Sham potential of density functional theory reproduces the linear-response transport of the Anderson model exactly, despite the lack of a Kondo peak in its spectral function. Using Bethe ansatz techniques, we calculated this potential exactly for all coupling strengths, including the cross-over from mean-field behavior to charge quantization caused by the derivative discontinuity. The implications of our results for more complex molecular junctions will be discussed.

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[2]Justin P. Bergfield, Zhenfei Liu, Kieron Burke, Charles A. Stafford, arXiv:1106.3104v2