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Pair Tunneling through Single Molecules

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Coupling to molecular vibrations induces a polaronic shift, and can lead to a negative charging energy, U. For negative U, the occupation of the ground state of the molecule is even. In this situation, virtual pair transitions between the molecule and the leads can dominate electron transport. At low temperature, T, these transitions give rise to the charge-Kondo effect [1]. We developed the electron transport theory through the negative-U molecule [2] at relatively high T, when the Kondo correlations are suppressed. Two physical ingredients distinguish our theory from the transport through a superconducting grain coupled to the normal leads [3]: (i) in parallel with sequential pair-tunneling processes, single-particle cotunneling processes take place; (ii) the electron pair on the molecule can be created (or annihilated) by two electrons tunneling in from (or out to) opposite leads. We found that, even within the rate-equation description, the behavior of differential conductance through the negative-U molecule as function of the gate voltage is quite peculiar: the height of the peak near the degeneracy point is independent of temperature, while its width is proportional to T. This is in contrast to the ordinary Coulomb-blockade conductance peak, whose integral strength is T-independent. At finite source-drain bias, V>>T, the width of the conductance peak is $\sim$V, whereas the conventional Coulomb-blockade peak at finite V splits into two sharp peaks at detunings V/2, and -V/2. Possible applications to the gate-controlled current rectification and switching will be discussed.

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