Electron-phonon interactions in weakly coupled single-molecule junctions

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Abstract:
Off-resonant charge transport through molecular junctions has been extensively studied since the advent of single-molecule electronics and is now well understood within the framework of the non-interacting Landauer approach. Conversely, gaining a qualitative and quantitative understanding of the resonant transport regime has proven more elusive. Here, using a three-terminal device architecture,[1] resonant charge transport through weakly coupled graphene-based single-molecule junctions will be described.[2] The inadequacies of non-interacting Landauer theory will be demonstrated, and through a combination of temperature- and gate-voltage-dependent measurements of conductance the interplay between Marcus Theory and quantum descriptions of electron-phonon coupling will be unravelled. Furthermore, the observance of vibrational sidebands[3] in the Coulomb-blockade regions of charge transport provides experimental evidence of non-equilibrium vibrational dynamics in single-molecule junctions and allows vibrational relaxation times to be extracted that are orders of magnitude slower than expected from solution-phase electronic spectroscopy.[4]

(a) Architecture of three-terminal graphene–based single-molecule transistors. (b) Conductance map of resonant transport, displaying vibrational side-bands in the Coulomb-blocked regions. (c) Bosonic temperature dependence of vibrational signatures.

References: