10th International Conference on Molecular Electronics
November 29 - December 2, 2021

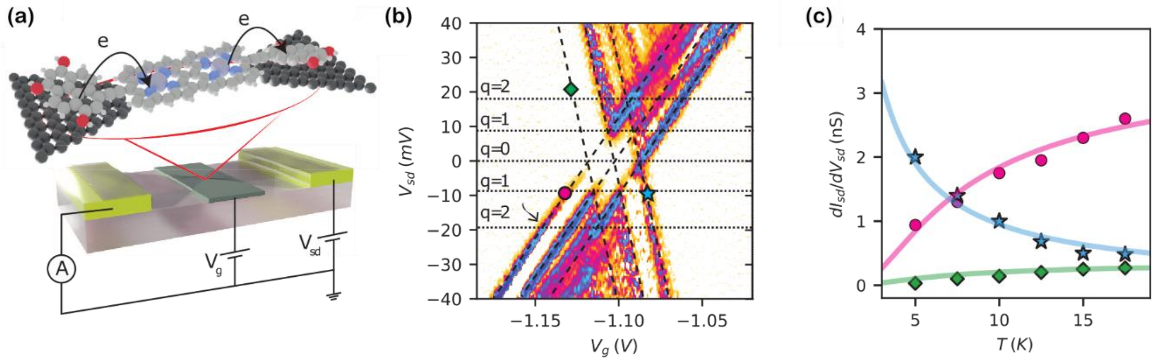
Electron-phonon interactions in weakly coupled single-molecule junctions

Xinya Bian, [a] Zhixin Chen, [a] Jakub Sowa, [b] Bart Limburg, [c] Andrew Briggs, [a] Harry Anderson, [c] Jan Mol, [d] and James Thomas [a]

[a] Department of Materials, University of Oxford, Parks Road, Oxford, UK. Email: james.thomas@materials.ox.ac.uk
[b] Department of Chemistry, Rice University, Houston, TX, USA.
[c] Department of Chemistry, University of Oxford, Chemistry Research Laboratory, Oxford, UK.
[d] School of Physics and Astronomy, Queen Mary University of London, London, UK.

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-blocked 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:

[1] Limburg B. et al. Anchor groups for Graphene-Porphyrin Single-Molecule Transistors, *Adv. Funct. Mater.,* 1803629 (2018)

[2] Thomas J. et al. Understanding resonant charge transport through weakly coupled single-molecule junctions *Nat. Commun.,* **10**, 4628 (2019),

[3] Luffe et al. Theory of vibrational absorption sidebands in the Coulomb-blockade regime of single-molecule transistors *Phys. Rev. B.* **77** 125306 (2008)

[4] Thomas et al. in preparation (2021)