Advances and challenges in single-molecule electron transport

Sumit Tewari[1,4], Richard Korytár[2], Ferdinand Evers[3], and Jan van Ruitenbeek[4]

[1]*Department of Materials, University of Oxford, OX1 3PH, Oxford, United Kingdom*
[2]*Department of Condensed Matter Physics, Faculty of Mathematics and Physics, Charles University, Ke Karlovu 5, 121 16 Praha 2, Czech Republic*
[3]*Institut für Theoretische Physik, Universität Regensburg, D-93053 Regensburg, Germany*
[4]*Huygens-Kamerlingh Onnes Laboratory, Leiden University, Niels Bohrweg 2, 2333CA Leiden, Netherlands*

Electronic transport properties for single-molecule junctions have been widely measured by several techniques, including mechanically controllable break junctions, electromigration break junctions or by means of scanning tunneling microscopes. In parallel, many theoretical tools have been developed and refined for describing such transport properties and for obtaining numerical predictions. Most prominent among these theoretical tools are those based upon density functional theory. In this review, theory and experiment are critically compared and this confrontation leads to several important conclusions. The theoretically predicted trends nowadays reproduce the experimental findings quite well for series of molecules with a single well-defined control parameter, such as the length of the molecules. The quantitative agreement between theory and experiment usually is less convincing, however. Many reasons for quantitative discrepancies can be identified, from which one may decide that qualitative agreement is the best one may expect with present modeling tools. For further progress, benchmark systems are required that are sufficiently well-defined by experiment to allow quantitative testing of the approximation schemes underlying the theoretical modeling. Several key experiments can be identified suggesting that the present description may even be qualitatively incomplete in some cases. Such key experimental observations and their current models are also discussed here, leading to several suggestions for extensions of the models towards including dynamic image charges, electron correlations, and polaron formation.