Electronic transport in complex molecules: structural and dynamical correlations

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The ultimate nanoscale conductor is reached perhaps at the single long-molecule level. For example, the prototypical biological molecule of DNA, has been proposed as a natural conductor with many ideal properties: It is naturally occurring as well as easily synthesized, it is prone to “self-assembly” and useful to create desired “scaffolding” at the nanometer scale, and it may allow electronic transport along its π-bond stack of neighboring bases. Similarly, other flexible complex molecules may exhibit interesting transport behavior (such as the “molecular metals” based on phthalocyanine). A host of different experimental results suggest a non-trivial role of molecular configurations, both from a static (composition) sense, as well as in the sense of the dynamical configurations.

In this talk we present the role of structural and dynamical correlations on the transport properties of flexible long molecules. It is known that chemical constraints built in the given molecular structure give rise to correlations in the local electronic energies and effective hopping amplitudes - even in apparently disordered chains (such as in typical DNA molecules). We study the role that these correlations may have on the ability of electrons to travel along the chain, and present results for the anticipated I-V characteristics. The role of “contacts” and details on the particular DNA sequence are also elucidated and found to be extremely important on the observed current amplitudes. Similarly, we look at the effects on Fermi broadening on the low temperature I-V and compare with ongoing experiments in Ohio and elsewhere.

On the other hand, flexible molecules have a relatively soft “twiston” mode that gives rise to an unusual and interesting electron-phonon interaction. As the twist is thought to affect directly the hopping between molecular units, this gives rise to a twiston-polaron as the lower energy quasiparticle. We consider the general characteristics of such objects and study their effective mass and effective bandwidth. We find that a Holstein polaron formulation of the twiston-electron quasiparticle which includes nonlinear coupling and nonlinear restoring forces results in power-law bandwidth suppression with coupling strength (unlike the exponential behavior of the usual vibrational modes). These results suggest that dynamical behavior of the electronic hopping is important for a full understanding of the transport in these unique one-dimensional-like nanoscale wires.