Coherent laser control of current via molecular nanojunctions with semiconductor and graphene contacts

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Abstract — We propose new approaches to coherent control of transport via molecular junctions. The first method is based on the application of intrinsic semiconductor contacts and optical frequencies below the semiconductor bandgap. Our analytical theory predicts a new phenomenon, referred to as coherent destruction of induced tunnelling, which extends the phenomenon of coherent destruction of tunnelling frequently discussed in the previous literature. We also propose to use graphene electrodes as a platform for effective photon assisted tunneling through molecular conduction nanojunctions. Our results illustrate the potential of semiconductor and graphene contacts in coherent control of photocurrent.

Keywords — Molecular conduction nanojunctions; dressed states; semiconducting and graphene electrodes

I. Introduction

The optical response of nanoscale molecular junctions has been the topic of growing experimental and theoretical interest in recent years, fueled by both the rapid advance of the experimental technology and the premise for long range applications. The ultimate goal of controlling electric transport with coherent light, however, has proven challenging to realize in the laboratory. One difficulty that has been noted in the previous literature is substrate mediated processes. Light shine on a molecular system in contact with a metal substrate is adsorbed by the substrate, rather than by a molecular bond or the molecule-surface bond in the vast majority of cases, leading to the excitation of hot carriers. The latter may interact with the molecule, and lead to the coherence loss. Other competing processes include heating of the electrodes (one of which often consists of an STM tip) and undesired energy transfer events.

II. Molecular Nanojunctions with Semiconductor Contacts

In the first part of this work we propose a new approach [1] to coherent control of transport via molecular junctions, which bypasses several of the hurdles to experimental realization of optically manipulated nanoelectronics noted above. The method is based on the application of intrinsic semiconductor (SC) contacts and optical frequencies below the SC bandgap. We consider a molecular junction consisting of a molecular moiety that possess a permanent dipole moment \( \mathbf{D} \) and is in contact with two intrinsic SC electrodes. The interaction of a nonresonant electromagnetic (EM) field with such systems leads to modulation of their energetic spectrum by the field frequency \( \omega \). The modulation under discussion alters the arrangement of molecular electronic states and may substantially change the electron and hole transfer rates between the molecular bridge and the SC contact, due to the strong dependence of these rates on the position of a molecular level relative to the conduction band and valence band. In other words, we are referring to optically induced tunneling. We also predict the effect of coherent destruction of induced tunneling (CDIT) that extends the certain effect of CDT.

III. Molecular Nanojunctions with Graphene Contacts

In the second part of the work we propose and explore theoretically a new approach [2] to coherent control of electric transport via molecular junctions, using either both graphene electrodes or one graphene and another one - a metal electrode (that may be an STM tip). Our approach is based on the excitation of dressed states of the doped graphene electrode with electric field that is parallel to its surface, having used unique properties of graphene. We have calculated a semiclassical wave function of a doped graphene under the action of EM excitation and the current through a molecular junction with graphene electrodes. We have shown that using graphene electrodes can essentially enhance currents evaluated at side-band energies in molecular nanojunctions that is related to the modification of the graphene gapless spectrum under the action of external EM field. We have calculated the corresponding quasienergy spectrum that is accompanied with opening the gap induced by intraband excitations. A side benefit of using doped graphene electrodes is the polarization control of photocurrent related to the processes occurring either in the graphene electrodes or in the molecular bridge.

References
