Photon-Assisted Tunneling through Molecular Conduction Junctions with Graphene Electrodes

Boris D. Fainberg

Faculty of Sciences, Holon Institute of Technology, 5810201, Holon, Israel
and School of Chemistry, Tel-Aviv University, 69978 Tel-Aviv, Israel
fainberg@hit.ac.il

The field of molecular-scale electronics has been rapidly advancing over the past two decades, both in terms of experimental and numerical technology and in terms of the discovery of new physical phenomena and realization of new applications. In particular, the optical response of nanoscale molecular junctions has been the topic of growing experimental and theoretical interest in recent years. A way of the control of the current through molecular conduction nanojunctions is the well-known photon-assisted tunneling (PAT) [1,2]. The main idea is that an external field periodic in time with frequency ω can induce inelastic tunneling events when the electrons exchange energy quanta ω with the external field. Here we propose and explore theoretically a new approach 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) [3]. Our approach is based on the excitation of dressed states of the doped graphene electrode with electric field that is parallel to its surface (Fig.1), having used unique properties of graphene, like strongly non-linear electromagnetic (EM) response [4], linear dependence of the density of states on energy, and the gapless spectrum that can change under the action of external EM field. 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 using non-equilibrium Green functions. Fig.2 show photon assisted current for a molecular junction with one graphene electrode and another one - a metal electrode for large momenta (far from the Dirac point). One can see the steps when the potential of the graphene electrode achieves the values corresponding to the photon energy, the doubled photon energy etc. The steps are found on the background that decreases linearly for a n-doped graphene electrode and increases linearly for a p-doped electrode when the potential of the graphene electrode increases. This is related to the linear dependence of DOS on energy. We have also shown that using graphene electrodes can essentially enhance currents evaluated at side-band energies $\sim n\hbar\omega$ 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 (Fig.3) that is accompanied with opening the gap induced by intraband excitations (for the interband transition contributions to the gap in an undoped graphene see [5] and references there). The quasienergy shows a quadratic dependence on momentum near the gap (solid line in Fig.3) that gives rise to slow falling down Fourier-harmonics with the harmonics index. Since the n-doped graphene is a well-defined Fermi liquid, we estimate the imaginary part of its the self-energy for the excitations of $n\hbar\omega$ above the Fermi energy. The estimates show that a rapid relaxation of the Fermi distribution to the “dressed” (Floquet) states seems a reasonable assumption. Recently Floquet states of surface Dirac fermions of a topological insulator have been observed experimentally [6].

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. The latter processes are accompanied by surface plasmon excitation in the graphene sheet that makes them more efficient. Our results illustrate the potential of graphene contacts in coherent control of photocurrent in molecular electronics, supporting the possibility of single-molecule devices.

References

[1] G. Platero and R. Aguado, Phys. Rep., **395** (2004) 1.
[2] S. Kohler, J. Lehmann, and P. Hanggi, ibid., **406** (2005) 379.
[3] B. D. Fainberg, Phys. Rev. B, **88** (2013) 245435.
[4] E. Hendry, P. J. Hale, J. Moger, A. K. Savchenko, and S. A. Mikhailov, Phys. Rev. Lett., **105** (2010) 097401.
[5] H. L. Calvo, H. M. Pastawski, S. Roche, and L. E. Torres, Appl. Phys. Lett., **98**, (2011) 232103.
[6] B. H.Wang, H. Steinberg, P. Jarillo-Herrero, and N. Gedik, Science, **342** (2013) 453.

Figures

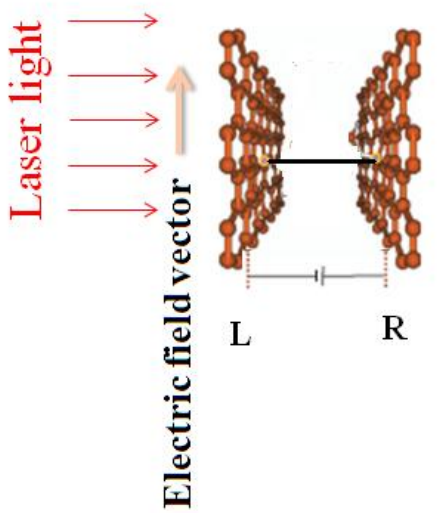

Fig.1. Molecular bridge (thick horizontal line) between left (L) and right (R) graphene electrodes with applied voltage bias.External electromagnetic field acts on the electrodes.

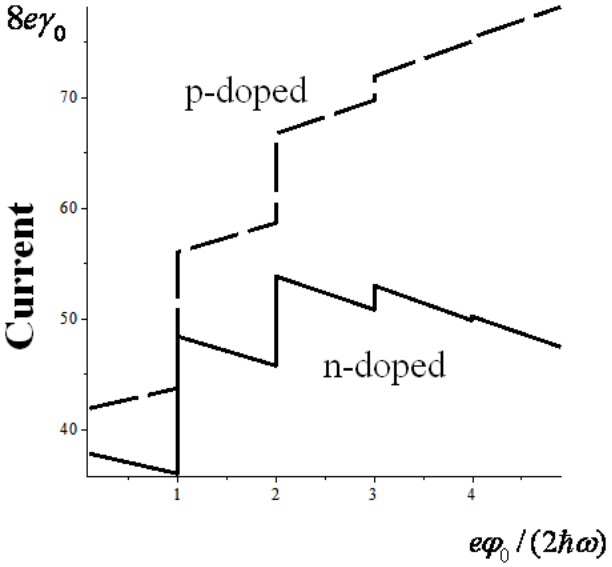

Fig.2. Current in the case of large momenta for n-doped (μ>0, solid) and p-doped (μ<0, dashed) graphene electrode as a function of applied voltage bias.

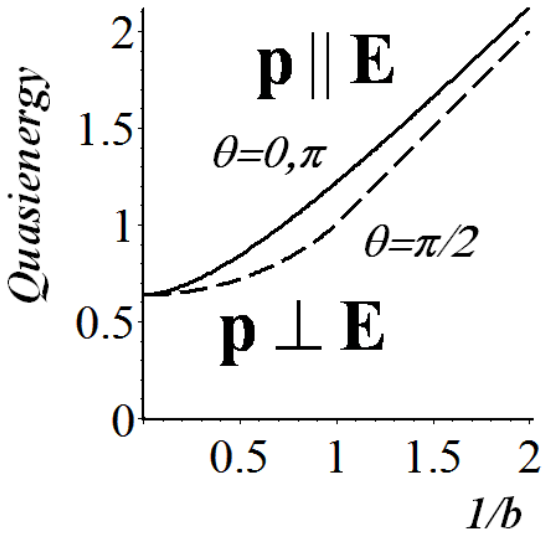

Fig.3. Quasienergies for momentum parallel (solid line) and perpendicular (dashed line) to electric field as functions of unperturbated energy vp weighted per the work done by the electric field during one fourth of period.