Secondary Hot-Carrier Generation in Graphene

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For many optoelectronic applications, such as photodetection and light harvesting, it is highly desirable to identify materials in which an absorbed photon is efficiently converted to electronic excitations. The unique properties of graphene, such as its gapless band structure, flat absorption spectrum and strong electron-electron interactions, make it a highly promising material for efficient broadband photon-electron conversion [1]. Indeed, recent theoretical work has anticipated that in graphene multiple electron-hole pairs can be created from a single absorbed photon during energy relaxation of the primary photoexcited e-h pair [2]. A photoexcited carrier relaxes initially trough two competing pathways: carrier-carrier scattering and phonon emission. In the former process the energy of photoexcited carriers remains in the electron system, being transferred to secondary electrons that gain energy (become hot), whereas in the phonon emission process the energy is lost to the lattice as heat. While recent experiments have shown that photoexcitation of graphene can generate hot carriers [3], it remains unknown how efficient this process is with respect to optical phonon emission.

In this talk we discuss the energy relaxation process of the primary photoexcited e-h pair in doped single-layer graphene [4]. In particular, we quantify the branching ratio between the two competing relaxation pathways. Given the challenging timescale with which these processes occur, we employ an ultrafast Optical pump – Terahertz probe measurement technique, where we exploit the variation of the photon energy of the pump light. Changing this photon energy is crucial as it allows us to prepare the system with photoexcited carriers having a prescribed initial energy determined by the photon energy, and follow the ensuing energy relaxation dynamics. We show experimentally, in combination with theoretical modeling, that carrier-carrier scattering is the dominant relaxation process. This process leads to the creation of secondary hot electrons that originate from the conduction band, a process we refer to as “hot-carrier multiplication”. Since hot electrons in graphene can drive currents, multiple hot carrier generation makes graphene a promising material for highly efficient broadband extraction of light energy into electronic degrees of freedom, enabling high-efficiency optoelectronic applications.

References

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