Multiplication of photo-excited carriers in graphene

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We present microscopic calculations of the relaxation dynamics of photo-excited carriers in graphene. Our approach includes the major relaxation channels, i.e. carrier-carrier as well as carrier-phonon scattering, and considers the coupled ultrafast dynamics of carrier population, coherence, and phonon population resolved in time and momentum. In agreement with several differential transmission experiments, we observe a redistribution of the excited carriers to energetically lower states resulting in a hot Fermi-Dirac distribution on a hundred femtosecond timescale followed by phonon-induced cooling in the range of a few picoseconds [1-4].

Due to its zero-bandgap and the linear bandstructure of graphene, Auger-type relaxation channels become efficient, which are negligible in conventional semiconductors. In contrast to any other Coulomb-induced channels, these scattering processes modify the total carrier density by bridging the valence and conduction band. Two competing processes are important: Auger recombination (AR) annihilates an electron and a hole, whereas impact ionization (II) creates both (cp. fig. a). Here, we evaluate both Auger-type processes and find a significant influence on the relaxation dynamics in the form of ultrafast carrier multiplication (CM), a process where multiple charge carriers are generated from the absorption of a single photon [5].

The temporal evolution of the total carrier density during and after a 10 fs pulse (800 nm) is shown in figure b. To study the influence of Auger-processes, we compare two situations: First, only the excitation is simulated to obtain the number of optically excited carriers (blue line). In a second calculation we include the full Coulomb scattering resulting in a significant increase of the carrier density after the pulse (red line). This nonequilibrium effect is caused by an asymmetry between AR and II during the equilibration in favor of II. We note that the CM depends on the strength of the pulse, for higher excitations the CM is reduced. Including the carrier-(optical)phonon scattering we also estimate the efficiency of the CM under realistic conditions. As expected, we observe a phonon induced carrier recombination and temporal decay of the carrier density on a picosecond timescale. Thus, the carrier multiplication remains efficient up to a few picoseconds confirming the potential of graphene as a new material for photodevices.

References

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Figure: (a) Scattering scheme of Auger recombination and impact ionization. Both processes fulfill momentum and energy conservation. (b) Temporal evolution of the charge carrier density considering the exciting pulse (blue) and carrier-carrier scattering (red). The preference of II results in a significant increase of the carrier density during the equilibration.