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PhD Thesis Defense ALEXANDER BLOCK 'Quantifying Nanoscale Carrier Diffusion with Ultrafast Optical and Photocurrent Microscopy'

ALEXANDER BLOCK

September 06, 2019

Thesis Defense, September 6, 14:00. ICFO Auditorium

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Molecular Nanophotonics

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Heat transport in solids is one of the oldest problems in physics, dating back to the earliest formulations of thermodynamics. The classical laws of heat conduction are valid as long as the observed time and length scales are larger than the relaxation time and mean free path of the underlying microscopic heat carriers, such as electrons and phonons. With the advent of ultrafast lasers and nanoscale systems these regimes can now be surpassed and new refined models of heat transport are needed. In particular, the interaction of ultrashort light pulses with matter can excite electrons to high temperatures, leading to a local non-equilibrium of electrons and phonons. Under these conditions, also the transport properties of the carriers are altered.

So far, these effects have typically been studied in the time domain. The cooling of photo-excited hot electrons has been studied both in metals as well as novel 2D materials, such as graphene. However, due to a lack of spatio-temporal resolution, it has not been possible to distinguish the effects of hot-electron diffusion from other cooling mechanisms, such as electron-phonon coupling.

In this thesis, I directly track such ultrafast heat and carrier diffusion in space and time with ultrafast microscopy. By using the recently developed technique of probe-beam-scanning transient-absorption microscopy on thin gold films I directly resolve, for the first time, a transition from hot-electron diffusion to phonon-limited diffusion on the picosecond timescale. I support the understanding of these complex dynamics by theoretical modeling of the thermo-optical response based on a two-temperature model.

I apply the same technique to study hot carrier diffusion in atomically thin monolayer graphene. By comparing differently prepared samples, I study the strong influence of external parameters, such as production type, substrate, and environment on carrier diffusion.

Finally, I study hot carrier diffusion in exfoliated and encapsulated graphene devices with a novel technique of ultrafast spatio-temporal photocurrent microscopy based on the photothermoelectric effect. I extract diffusion dynamics for electrically characterized samples with the help of theoretical spatio-temporal modeling, thereby testing the fundamental relationship between electrical and thermal carrier transport.

The precise quantification of ultrafast and nanoscale carrier transport with these state-of-the-art techniques leads to a broader understanding of non-equilibrium dynamics and could ultimately help the design, optimization, and heat management of the next generation of ultra-compact (opto-) electronic devices, such as solar cells, photodetectors, or integrated circuits.

Friday, September 6, 14:00. ICFO Auditorium

Thesis Advisor: Prof Dr Niek van Hulst