Direct View of Hot Carrier Dynamics in Graphene by Time and Angular Resolved Photoelectron Spectroscopy

J. C. Johannsen1, S. Ulstrup2, F. Cilento3, A. Crepaldi1, M. Zacchigna4, C. Cacho5, J. C. E. Turcu5, E. Springate5, F. Fromm6, C. Raidel7, T. Seyller8, F. Parmigiani1,2, M. Grioni1, P. Hofmann2.

1. Institute of Condensed Matter Physics, École Polytechnique Fédérale de Lausanne (EPFL), 1015 Lausanne, Switzerland.
2. Department of Physics and Astronomy, Interdisciplinary Nanoscience Center (NANO), Aarhus University, 8000 Aarhus C, Denmark.
4. KOM-CNR Laboratory TASC, Area Science Park, 34012 Trieste, Italy.
5. Central Laser Facility, STFC Rutherford Appleton Laboratory, Didcot OX11 0QX, United Kingdom.
6. Institute for Physics, Technische Universität Chemnitz, 09126 Chemnitz, Germany.
7. Department of Physics, University of Trieste, 34127 Trieste, Italy.


For correspondence and requests: federico.cilento@eletra.eu

Introduction, Theory, and Motivation

Graphene is one of the crystalline forms of carbon, together with graphite, carbon nanotubes, fullerenes, and diamond. In this most common form, carbon atoms are arranged in a regular hexagonal pattern, forming a perfect two-dimensional crystal. Graphene is an excellent conductor of both heat and electricity, and is among the strongest known materials. Its strength, like that of diamond, is derived from the strong carbon-carbon covalent bonds. The material is electrically neutral, having zero electrical conductivity. However, when a carbon atom is electrically empty, the entire graphene sheet is electrically neutral. These properties make graphene a unique material for a wide range of applications, including electronic, medical, and chemical ones. Its perfect bidimensionality leads to a wealth of unique properties.

All these properties arise from the peculiar electronic structure, characterized by a linear dispersion relation for electrons and holes, described by the Dirac equation. This relation holds at low energies near the six corners of the two-dimensional Brillouin Zone (BZ), leading to zero effective mass for electrons and holes near the Fermi level. Because the linear dispersion relation is maintained at the Fermi surface, the material behaves like relativistic particles described by the Dirac equation. Hence, the electrons and holes are called Dirac fermions and the six corners of the BZ are called the Dirac points (DPs).

To this end, we chose an on-the-equilibrium approach, under the form of the Time and Angular Resolved Photoelectron Spectroscopy (TR-PARPES) technique. In order to characterize how the extra energy is rapidly injected in the solid system through the pump pulse, we used TR-PARPES directly to verify how the electronic distribution is modified after extra energy is absorbed by the material.

The large 2D of graphene requires high energy photons (hv=1.1 eV) in order to probe the electronic band structure in the vicinity of the DPs. Hence, we performed time-resolved experiments at the ARTESI facility (Didcot, STFC – 44z, G1).

Non-Equilibrium Approach & TR-ARPES @ ARTEMIS

Out-of-equilibrium spectroscopies with temporal resolution of ~100 fs allow to discriminate the intrinsic features of single solid materials thanks to their spectral features and dynamics in the non-equilibrium signal.

TR-ARPES provides the direct view on the evolution of the electronic structure and the electronic distribution, after the system has been brought out of equilibrium by the absorption of a pump pulse. At ARTEMIS, the probe beam is a tunable 100 fs pulse train at the next harmonics (16-32 eV).

In graphene, the peculiar electronic structure at the Fermi level allows for the ultrafast availability of extra energy absorbed by electrons through different excitation mechanisms. We also pointed out that supercollisions, i.e., the scattering of electrons with photons, mediated by impurities, constitutes a fundamental channel in the relaxation mechanism. The two mechanisms, determining the strength of the electron-photon coupling, result in a net result on the evolution of the system.

In this paper, we analyze the electronic temperature of the system as a function of the pump-probe delay time. In this way, we reduce the electronic temperature of the system to a constant value, and study the dynamics of the electronic system.

Experimental Data

Data Analysis

Relaxation Dynamics

Electron-Boson Coupling

Supercollisions

Lifetime Evolution

Conclusions

Non-equilibrium spectrosopies revealed effective in order to solve a long-standing issue in graphene, namely, the nature and strength of the electron-photon coupling, responsible in determining the relaxation timescale of the excess energy absorbed by electrons through different excitation mechanisms. We also pointed out that supercollisions, i.e., the scattering of electrons with photons, mediated by impurities, constitutes a fundamental channel in the relaxation mechanism. The two mechanisms, determining the strength of the electron-photon coupling, result in a net result on the evolution of the system.

In this work, we analyze the electronic temperature of the system as a function of the pump-probe delay time. In this way, we reduce the electronic temperature of the system to a constant value, and study the dynamics of the electronic system.

Non-equilibrium spectrosopies revealed effective in order to solve a long-standing issue in graphene, namely, the nature and strength of the electron-photon coupling, responsible in determining the relaxation timescale of the excess energy absorbed by electrons through different excitation mechanisms. We also pointed out that supercollisions, i.e., the scattering of electrons with photons, mediated by impurities, constitutes a fundamental channel in the relaxation mechanism. The two mechanisms, determining the strength of the electron-photon coupling, result in a net result on the evolution of the system.