Wavelength photoexcitation dependence of ultrafast phonon and plasmon dynamics in Graphite

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The richness of the electron energy momentum distribution of a material suggests various interesting behaviors when it is driven out of its equilibrium state. Depending on the level of the excitation, we can expect different dynamics. The most straightforward way to experimentally make these observations is to directly measure the k-space of the material. The transition from the ground state to various excited states can be achieved using photoexcitation with a laser pulse.

We are interested in graphite which shows a strong electron-phonon coupling due to the presence of a Dirac cone at the Γ point. The zero-band gap allows electronic transition at a broad energy scale from deep IR to UV. Depending on the level of excitation energy, the excited electrons take different in the energy-momentum landscape. We aim to measure the scattering behaviors of these hot electrons after two different photoexcitation wavelengths: 400nm (3.1 eV) and 800 nm (1.55 eV).

To make these observations, we used two different techniques. The time and momentum electron energy loss spectroscopy (tr-q-EELS) to investigate the plasmon dynamics and the shot-to-shot acquisition ultrafast electron diffraction (UED) to investigate the phonon dynamics through the diffuse scattering. Both spectra show significant differences when photoexcited with different wavelengths.

These observations are supported by theoretical works that suggest that the discrepancies in the spectra are due to different temperatures of the strongly coupled optical phonons (E_{2g} and A_1' SCOP modes) and the overall lattice temperature. We suggest that the excitation at higher photon energy may lead to a larger excited electron packet with enlarged momentum driving the generation of SCOP with different electron-phonon coupling.

In summary, we combined tr-q-EELS and UED to fully inspect the reciprocal space dynamics of Graphite after different photoexcitation wavelengths. Depending on the excited energy level, the hot electrons decay differently to SCOP suggesting distinct electron-phonon coupling.