Ultrafast electron probing of plasmon thermal dynamics

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Thermal engineering of plasmons and other forms of polaritons in nanomaterials offers an appealing way of controlling light−matter interactions down to nanometer and femtosecond spatiotemporal scales [1]. The study of ultrafast thermal dynamics traditionally relies on optical pump-probe experiments which are, however, limited in spatial resolution due to light diffraction. Electron energy-loss spectroscopy (EELS) performed on scanning transmission electron microscopes overcomes the optical diffraction limit by using electrons rather than light to map the material response with sub-Ångstrom spatial precision. In this work [2], we propose to study the ultrafast thermal dynamics of nanomaterials using an optical-pump/electron-probe (OPEP) technique with sub-Ångstrom and femtosecond spatiotemporal resolution.

Two-dimensional (2D) materials offer a splendid testbed for OPEP because they generally undergo substantial changes in their electronic structure under optical pumping [1]. We start by applying this technique to highly doped self-standing graphene, which hosts electrically tunable plasmons. The graphene sample is optically pumped with an ultrafast laser (Fig. 1a), which creates an elevated electronic temperature in the material that decays over a picosecond time scale. When probed with a delayed electron pulse, the graphene plasmon dispersion can be mapped out from the energy- and angle-resolved inelastically scattered electron distribution (Fig. 1b,c). The dynamics of rapid heating and cooling of graphene electrons is traced through the delay-dependent variations observed in the distribution of scattered electrons.

We further show that the OPEP technique can be applied to doped graphene and undoped graphite ribbon samples. Interestingly, ribbons break translational invariance and produce lateral plasmon confinement, resulting in a discretization in electron deflection. Finally, we also show that the described excitations and their temporal dynamics can be revealed by integrating the inelastic electron signal over a broad energy range, thus avoiding the need for highly monochromatic electron beams and precise spectrometers.

This previously unexplored approach to study the ultrafast dynamics of optical excitations can be of interest to understand and manipulate polaritons in 2D semiconductors and other materials exhibiting a strong thermo-optical response.

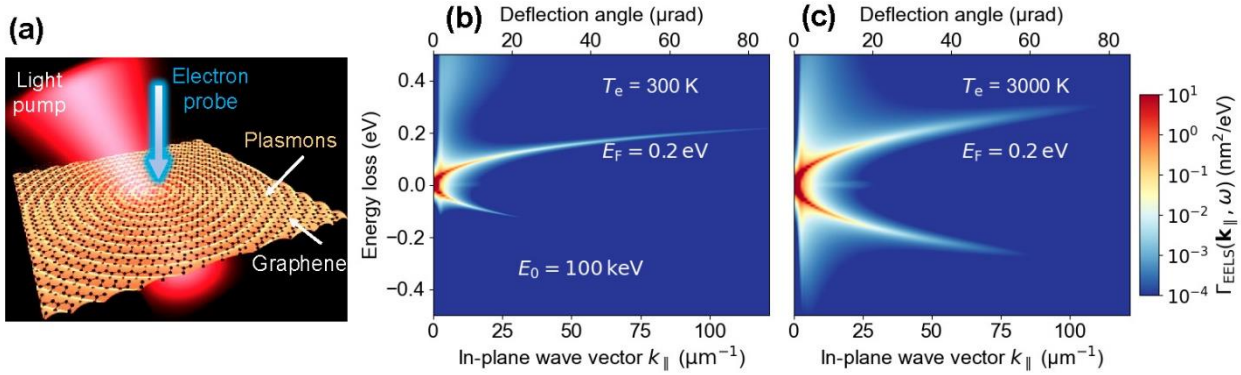

Fig. 1 OPEP characterization of plasmons in highly doped extended graphene. (a) Schematic representation of a graphene layer and OPEP configuration, with electrons impinging normal to the plane of the sample. (b, c) Momentum- and energy-resolved loss probability Γ_{EELS} ($k_\parallel$,ω) for 100 keV transmitted electrons, revealing features associated with plasmon excitation in the sample for (b) low (300 K) and (c) high (3000 K) electronic temperature regimes. The graphene Fermi energy is E_F=0.2 eV and the intrinsic damping is $\hbar/\tau$=4 meV.

References

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