Collective electronic excitations in graphene-based systems: first-principle and model calculations

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Two-dimensional crystals are promising building blocks for the design of new materials. For example, graphene can be stacked up into multi-layer graphene, or wrapped into low-dimensional nanostructures like carbon nanotubes and fullerenes. In order to predict their properties one has to understand the influence of dimensionality and interactions between the building blocks. In this work, we concentrate on collective electronic excitations (plasmons) in graphene-based systems. Plasmons can be understood as oscillations of the electron density with a given wavelength (momentum) and frequency (energy). The plasmon dispersion, i.e., the relation between energy and momentum, can be probed by angular-resolved electron energy-loss spectroscopy (EELS) in an electron microscope. We use ab-initio and model calculations to theoretically investigate the energy-loss function (1–40eV) of different carbon nanostructures.

First, the dielectric response of isolated graphene is calculated from first-principles. Using a plane-wave pseudopotential code [1,2], we calculate the momentum- and energy-dependent electron energy-loss function for isolated graphene within the random-phase approximation (Fig. 1). These results are compared with tight-binding calculations [3]. In particular, we discuss the validity of the two-dimensional description of graphene which is often applied in model calculations by neglecting the extension of the orbitals in perpendicular direction [4].

Second, we investigate plasmons in multi-layer graphene (Fig. 2). Using a layered-electron gas model [5] we observe a splitting of the graphene plasmon into several plasmon bands according to the number of layers. In standard EELS experiments, the beam is perpendicular to the graphene layers and only the symmetric plasmon mode (Fig. 2a) can be observed [6]. In contrast, our calculations predict that the other modes (Fig. 2b+c) can be measured using angular-resolved EELS on tilted multi-layer graphene in the case of large perpendicular momentum transfers. We present first experimental results obtained on a low-voltage optimized Libra-based transmission electron microscope prototype (ZEISS) which is equipped with an electrostatic monochromator and a C_S-corrector [7,8].

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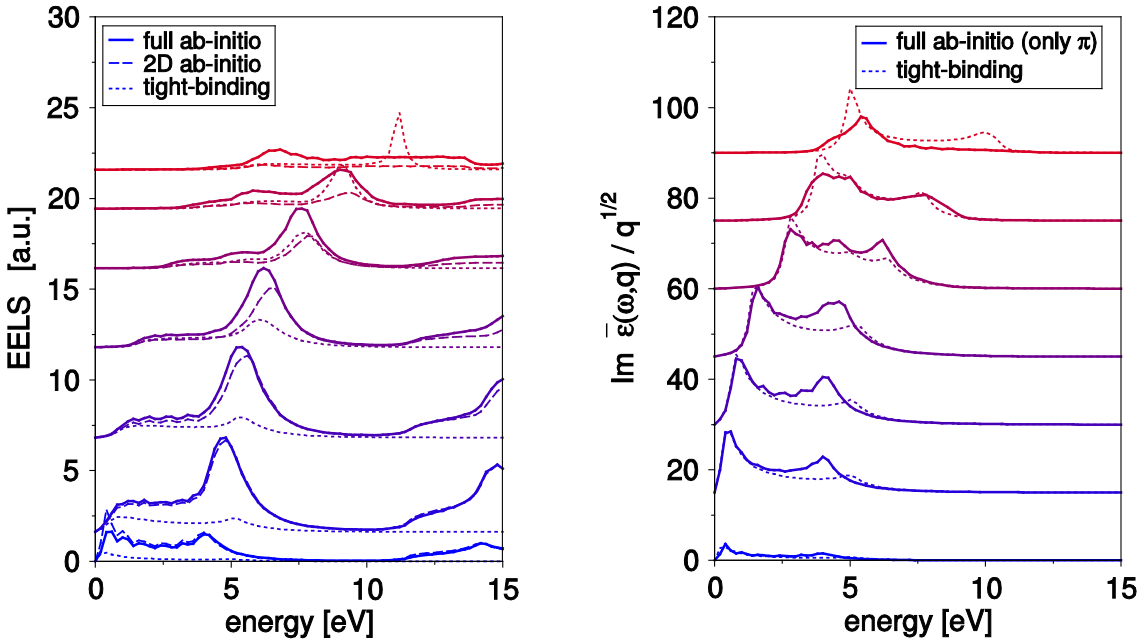

Figure 1. *Left: The calculated energy-loss function of isolated graphene for different in-plane momentum transfers q along the ΓM direction. We find important differences between the results of the full ab-initio calculation (solid line), an ab-initio calculation in 2D (dashed) where the extension of the orbitals in perpendicular direction is neglected, and tight-binding calculations (dotted). Right: In contrast, ab-initio and tight-binding calculations show very similar results for the polarisability of graphene. All spectra are scaled and shifted along the y-axis for better visibility.*

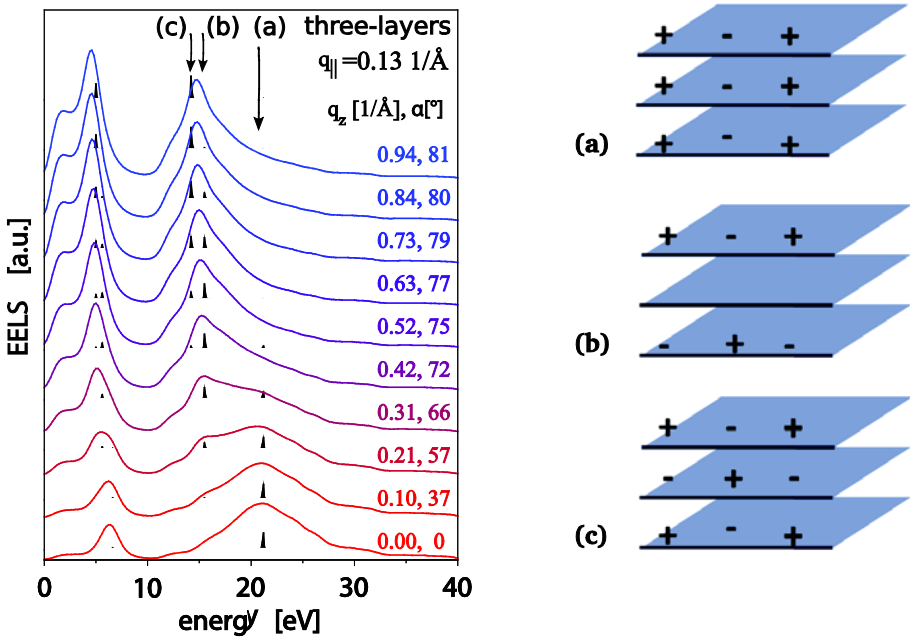

Figure 2. *Left: Angular-resolved energy-loss spectra of a three-layer graphene system for fixed in-plane momentum transfer $q_{||}$= 0.14 Å and increasing perpendicular momentum transfer q_z calculated from isolated graphene using the layered electron-gas model. Arrows indicate the energy and strength of the contributing plasmon bands. All spectra are scaled and shifted along the y-axis for better visibility. Right: Visualization of the charge oscillation for the different plasmon modes in three-layer graphene. For in-plane momentum transfer (q_z=0), only the symmetric mode (a) can be excited. For large q_z, also the anti-symmetric plasmon mode (c) contributes to the energy-loss spectrum.*