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Momentum-resolved electron energy-loss spectroscopy of free-standing single- and multi-layered graphene

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Graphene with its unique structural and electronic properties has caused a lot of excitement in last couple of years. Recently the nature of 2D plasmons in graphene has also been in the focus of attention [1,2]. These plasmons are collective excitations of electrons, which are confined on a plane and interact via Coulomb forces. They can be characterized based on their dispersion relation, i.e. the dependence of the plasmon energy on wavelength. Insights into the properties of these 2D plasmons in graphene are not only important for the fundamental understanding of the electronic structure and collective behaviour in graphene, but also for potential applications in nano-plasmonics [3].

Here, we are concerned with the dispersion of high-energy plasmons (>3eV), known as π- and $\pi+\sigma$-plasmon, in free-standing single- and multi-layer graphene. We have applied momentum-resolved electron energy-loss spectroscopy in a Libra200-based low-voltage transmission electron microscope (ZEISS) equipped with a monochromator and in-column Omega-filter to measure the momentum-dependent energy-loss function for two non-equivalent in-plane directions, i.e. ΓM and ΓK.

In figure 1 we have plotted experimental spectra of free-standing single-layer graphene covering momentum transfers along both symmetry direction over the entire 1st Brillouin zone. At small q-values, we find two plasmons peaks located at 4.9eV and 15.3eV and in good agreement to previous findings [4]. The measured loss function for both symmetry directions are identical up to an intermediate momentum transfer of about 0.5 1/Å, with significant differences appearing at larger q-values. In ΓM -direction, above a value of 0.8 1/Å, the π-plasmon splits into a double peak, with a shoulder at around 5eV, similar to observations in carbon nanotubes [1]. For ΓK, no splitting is observed. These experimental results are then compared to ab-inito calculations in the framework of time-dependent density functional theory (TDDFT).

In addition we will present measurements, illustrating the changes of the loss-function with increasing number of layers (up to 6) (figure 2) and a careful study of out-of-plane excitations in single- and multi-layered graphene. These observations are than discussed in terms of a simple layered electron gas model.

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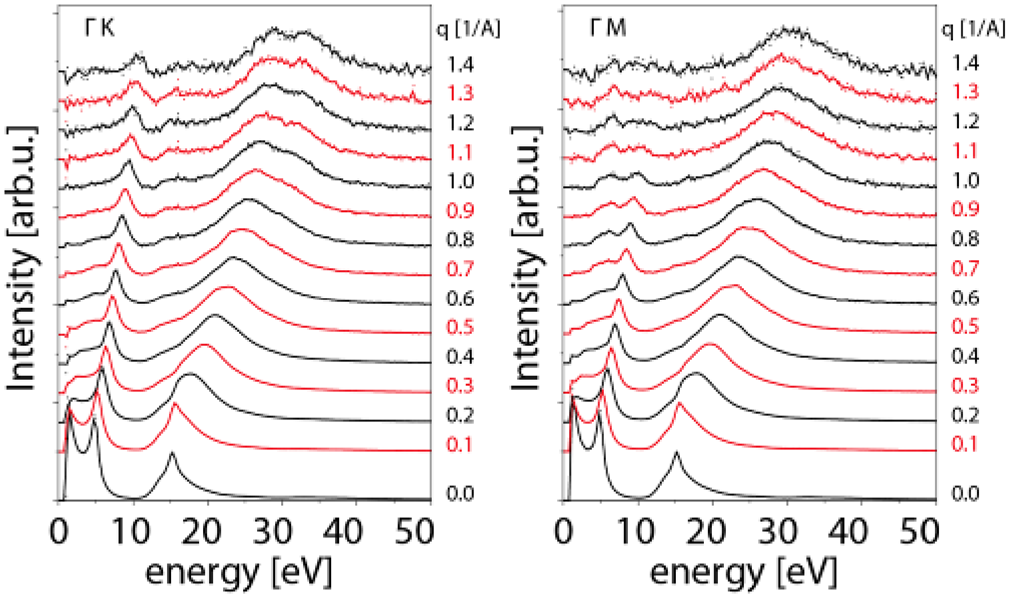

Figure 1. Experimental in-plane energy-loss of free-standing, single-layer. Shown are spectra for momentum transfers along ΓK (left) and ΓM (right). Energy resolution is ΔE = 0.2eV and momentum resolution Δq =+/-0.1 1/Å. Spectra have been shifted vertically and scaled for better visualization

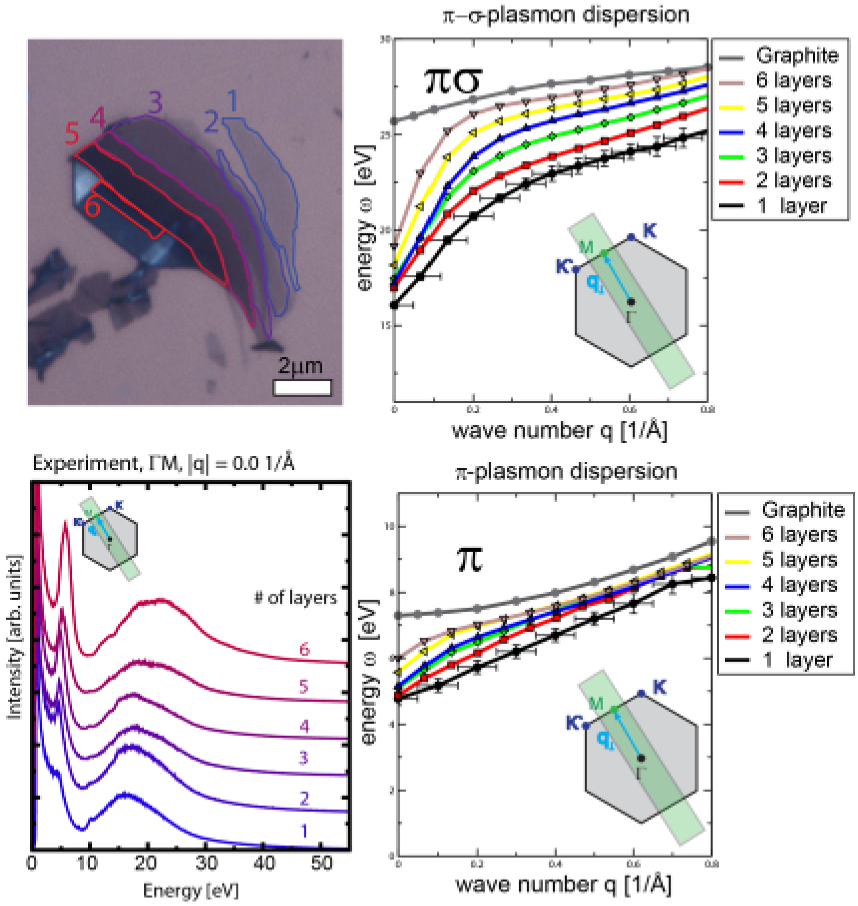

Figure 2. Measured energy-momentum dependence of both the π− and π+σ−plasmon in free-standing graphene. Shown are dispersion curves for in-plane momentum transfers along ΓM (left) and ΓK (middle). For ΓM the plasmon dispersion of graphite was added as reference (gray). Peak positions were extracted from spectra of up to six layers of AB-stacked graphene sheets. Different colours indicate different number of layers. Incident electron energy was 40keV. Energy resolution is ΔE=0.2eV and momentum resolution is Δq=+/-0.1 1/Å. (right) Light optical images showing the probed graphene sample.