Momentum dependent electron energy-loss in single and multi-layer graphene

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Graphene with its unique structural and electronic properties has caused a lot of excitement in recent years. At low energies charge carriers in graphene act as massless two-dimensional (2D) Dirac electrons, which give rise to interesting properties including extremely high charge mobility and fractional quantum Hall effect [1,2]. Recently the nature of 2D plasmons in graphene has also been in the focus of attention [3]. 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 behavior in graphene, but also for potential applications in nano-plasmonics [4]. Here we are concerned with the dispersion of high-energy plasmons (>3eV), known as $\pi-$ and $\pi-\sigma$ plasmon in free-standing single- and multi-layer graphene.

We have applied angle-resolved electron energy-loss spectroscopy (EELS) in a low-voltage transmission electron microscope (TEM) to measure the momentum-dependent energy-loss function (ELF) of suspended single- and multi-layer graphene for the two reciprocal symmetry direction $\Gamma-M$ and $\Gamma-K$. Samples were prepared using mechanical exfoliation. Experiments were done on a Libra-based TEM prototype (ZEISS) operated at 20kV and 40kV [5]. We determined the energy and momentum resolution to be 0.2eV and 0.1-0.2$\mathring{A}^{-1}$. Our achieved spatial resolution was around 100-200nm.

In Fig. 1 we have plotted EEL spectra of suspended single-layer graphene. In accordance with experiments of Eberlein et al [6], we find the two plasmon peaks at small q-values to be around 5eV and 15eV for both symmetry directions. At smaller q-values the spectra for $\Gamma-M$ and $\Gamma-K$ are similar. We find significant differences at q-values larger than 0.5$\mathring{A}^{-1}$. In $\Gamma-M$ direction and above a value of 0.8$\mathring{A}^{-1}$ the $\pi-$plasmon splits into two peaks with a shoulder at around 5eV similar to $\pi-$plasmons observed in carbon nanotubes (CNT) [7] and graphite [8]. We see a similar shoulder for the $\Gamma-K$ direction. Comparison to density functional theory calculations (RPA) shows, that in both cases the behaviour of the $\pi-$plasmon is well reproduced. In contrast RPA does not correctly describe the behaviour of the $\pi-\sigma-$plasmon. Furthermore, for single-layer, free-standing graphene we report a quasi-linear dispersion of the $\pi-$plasmon for both symmetry directions [9]. In addition we present measurements, illustrating the changes of the ELF with increasing number of graphene layers (up to 6). Here the behaviour can be understood in terms of a simple layered electron gas (LEG) model. Additionally we present the layer dependent plasmon dispersion behaviour (Fig. 2) and compare it to graphite, showing that at small q-values 6-layered graphene is still significantly different from graphite but appears graphite like at large q-values.

References

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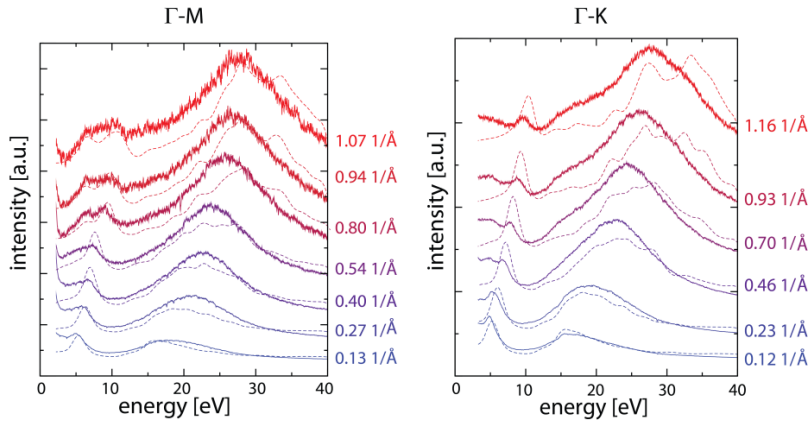[5]] U. Kaiser et al, Ultramicroscopy **111** (2011), p.1246-1239.
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Figure 1. *Experimental in-plane energy-loss of suspended, single layer graphene (solid) and ab initio (RPA) calculations (dotted). Shown are spectra for momentum transfers along Γ−M (left) and Γ−K (right). Incident electron energy was 20keV. Energy resolution is ΔE = 0.2eV and momentum resolution Δq =+/-0.1 1/Å. Spectra have been shifted vertically and scaled for better visualization.*
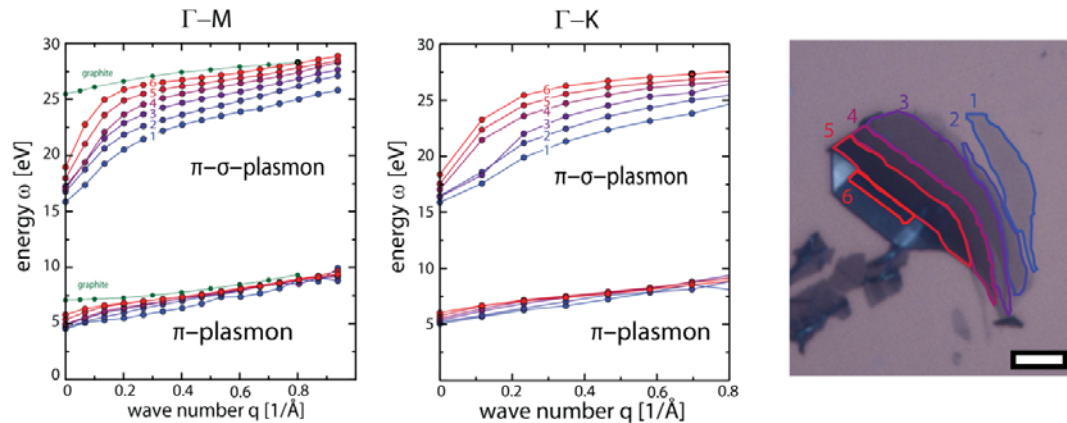
Figure 2. *Measured energy-momentum dependence of both the π− and π−σ−plasmon in suspended graphene. Shown are dispersion curves for in-plane momentum transfers along Γ-M (left) and Γ-K (middel). For Γ−M the plasmon dispersion of graphite was added as reference (green). Peak positions were extracted from spectra of up to six layers of AB-stacked graphene sheets. Different colors indicate different number of layers. Incident electron energy was 40keV. Energy resolution is ΔE=0.2eV and momentum resolution is Δq=+/-0.2 1/Å. (right) Light optical images showing the probed graphene sample. Colored numbers indicate the number of graphene layers . Scale bar is 2μm.*