PLASMON EXCITATIONS IN GRAPHENE USING ELECTRON ENERGY LOSS SPECTROSCOPY

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Plasmon excitations in graphene have been studied recently by electron energy loss spectroscopy (EELS), both at high incident energies (∼100 keV) with trajectories that traverse graphene layers in scanning transmission electron microscope (STEM) [1] and at low incident energies (∼10 eV) under the near-specular reflection conditions in a high-resolution reflection EELS (HREELS) [2]. I present recent results on theoretical modeling of the experimental spectra obtained in Refs. [1] and [2].

In the case of high-energy plasmon excitations in free-standing, multi-layer graphene (MLG) consisting of N graphene sheets, the experimental EEL spectra exhibit both π and $\sigma + \pi$ plasmon peak structures that are obtained as integrated over a significant range of wavenumbers q [1]. By treating the MLG as layered electron gas with in-plane polarizability modeled by a two-dimensional, two-fluid hydrodynamic model [3], we obtain the EEL spectra that show very good agreement with the experimental results of Eberlein **et al.**[1] for $N < 10$, as shown in Fig.1. In particular, one notices that the low-energy π plasmon peak undergoes relatively small changes with increasing number of graphene layers, whereas the high-energy $\sigma + \pi$ plasmon structure evolves from a broad peak at about 15 eV for $N = 1 - 2$ to a peak at about 25 eV at $N > 10$. This effect points to possible analogy with the interplay between surface and volume plasmon modes in graphite, which should be further explored.

On the other hand, by using the low-energy electron scattering from epitaxial single-layer graphene in HREELS, one obtains wavenumber-resolved spectra that reveal the dispersion of graphene's low-energy π plasmon [2]. In particular, at long wavelengths one finds the evidence for strong coupling of graphene's π plasmon with a Fuchs-Kliever surface phonon in the SiC substrate [2,4]. The results of our modeling [5] of the loss function for epitaxial graphene are compared with the experimental HREEL spectra of Liu **et al.**[2] in Fig.2. We used Mermin's procedure to account for finite damping rate γ in the RPA dielectric function for graphene assuming linear π electron bands [6,7], whereas the substrate effects were modeled by a local dielectric function for non-dispersing optical phonon mode in SiC [8]. One sees in Fig.2 that a relatively large value of $\gamma \approx 0.4$ eV is needed to reproduce the HREEL spectra in regions around the π plasmon peaks, whereas inclusion of the substrate phonon only qualitatively reproduces spectral features at low frequencies, and hence warrants further refinements of the HREELS model for graphene.

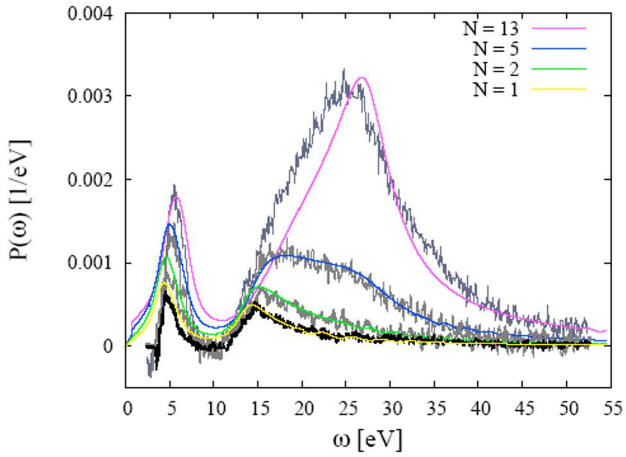

Figure 1: Probability density $P(\omega)$ for energy loss ω for free-standing graphene with N= 1, 2, 5 and 13 layers (smooth curves in colors), along with the corresponding experimental EEL spectra [1] (noisy curves in gray).

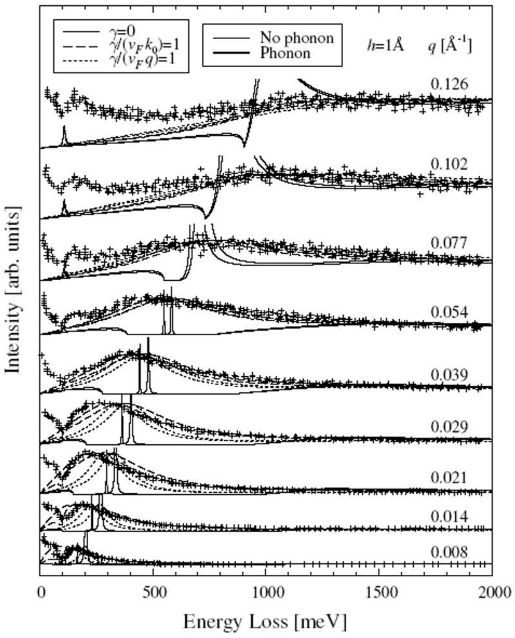

Figure 2: Loss function based on Mermin's treatment of graphene's RPA dielectric function with damping rate γ versus electron energy loss for graphene with charge-carrier density $n = 1.9 \times n_0$, where $n_0 = 10^{13} cm^{-2}$, on a SiC substrate with the gap of $h = 1\mathring{A}$ for $\gamma = 0$ (solid curves), constant damping rate $\gamma = v_F k_0$, where $k_0 = \sqrt{\pi n_0}$ (dashed curves), and linearly dispersing damping $\gamma = v_F q$ (dotted curves). The thick and thin lines show model results with and without the inclusion of the substrate's optical phonon, respectively, while symbols show the experimental HREELS data [2].

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