Electron – Phonon Couplings and Fano Resonances in Epitaxial Graphene Bilayer

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Keywords: graphene bilayer, Raman active mode, optical active mode, Fano shape line,

A vast majority of the optical studies of graphene have been performed by Raman spectroscopy, whereas infrared techniques that can provide complementary information, due to different selection rules, remain largely unexplored. Fourier transformed infrared attenuated total reflection spectroscopy (FTIR-ATR) and infrared reflectivity together with Raman measurements of epitaxial hydrogenated graphene bilayers grown on 4H-SiC(0001) by CVD are presented and discussed. The intercalation of hydrogen under a buffer and a single graphene layer was performed at high temperatures in the range 1000^0C – 1200^0C.

The FTIR-ATR measurements using polarized light revealed the Si – H stretch mode at 2128cm^{-1}, which proves that the SiC(0001) surface is saturated with hydrogen. The obtained bilayers are hole-doped (p = 1.3 – 1.6x10^{13} cm^{-2}), with the carrier mobility of 3000cm^2/Vs – 3500cm^2/Vs at 300K. In addition to the Si – H stretch line, a strong absorption is observed in both FTIR-ATR and reflectivity in the vicinity of the well-known Raman G band (close to1590cm^{-2}). However, the absorption line (detected in the reflection from the SiC substrate through the graphene bilayer) is shifted to a lower energy with respect to the Raman G line by about 10cm^{-1}, as shown in Fig. 1. This is the manifestation of an effect that in a plane optical phonon in the center of the Brillouin zone (G band) of bilayer graphene consist of an in-phase-mode (even parity) - being the Raman active one (RM), and an out-of-phase-mode (odd parity) - being the infrared active one (OM) [1]. Due to a large hole concentration, the RM and OM are no longer degenerate and the OM is shifted to a lower energy with respect to the RM. Such splitting between the Raman and optical modes of the G line was earlier reported only for a freestanding gated graphene bilayer in the electric fields [1].

In addition to finding the split between the energy of the Raman and infrared active modes we observed that the OM is of a Fano shape. The Fano shape of the infrared active G line was found in reflectivity and in FTIR-ATR, as well. Such Fano line shape of the optical mode of the G line was also earlier reported in the freestanding gated graphene bilayer [2]. The Fano shape absorption line, particulary strong in FTIR-ATR (Fig. 2) is observed for the first time for the graphene bilayer without influence of the electric field. The presence of Fano resonance indicates that infrared active G phonon mode is interacting with a continuum of hole excitations within the graphene bilayer valence band.

[1] Jan Yan et al., Phys. Rev. B 80 241417 (2009)
[2] A.B. Kuzmenko et al., Phys. Rev. Lett. 103, 116804 (2009)

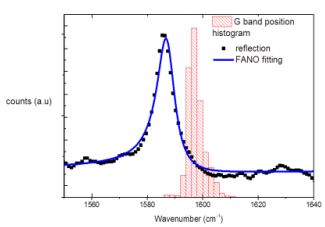

Fig. 1. The optical mode (OM) observed in reflectivity and the histogram of the Raman (RM) mode versus infrared wavenumber.

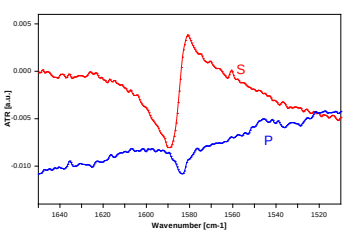

Fig. 2. The Fano resonance of the OM mode observed in s and p polarization in FTIR-ATR measurement.