Reversible Formation and Hydrogenation of Deuterium-Intercalated Quasi-Free-Standing Graphene on 6H-SiC(0001)

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Using LEED and High Resolution Electron Energy Loss Spectroscopy (HREELS), we have studied two reversible hydrogenation processes of graphene/6H-SiC. We confirm that the high temperature hydrogenation around 700°C of the C-rich (6v3x6v3)R30° of 6H-SiC leads to a quasi free-standing monolayer graphene (QFMLG) through deuterium intercalation [1,2] under the carbonaceous buffer layer (SiC-BL). In contrast to the buffer layer, this QFMLG, stable under a moderate annealing, shows quasi-metallic properties. The decrease of the full-width at half-maximum of the elastic peak in HREELS shows that a subsequent room-temperature hydrogenation of the QFMLG restores an insulating phase with a bandgap dependent on the hydrogen coverage, while the carbon honeycomb structure remains intact.

The insulating character of the hydrogenated QFMLG allows to probe the vibrational properties of Si-H or Si-D species under the QFMLG. While no abstraction could be observed during hydrogenation (deuteration) of a D-intercalated (resp. H-intercalated) QFMLG at room temperature, the high temperature hydrogenation leads to a complete isotope substitution, i.e. SiC-D/QFMLG becomes SiC-H/QFMLG [3].

The evolution of the HREELS elastic peak FWHM of SiC-D/QFMLG-H upon thermal annealing show marked changes which reveal the successive desorption of hydrogen chemisorbed on grapheme followed by the desorption of the intercalated deuterium, ending in a SiC-BL, and which can be used as a simple fingerprint to identify the nature of the hydrogenated system. These reversible hydrogenation processes appear promising for the practical realization of useful graphene-based devices.

References

[1] C.Riedl et al., Phys.Rev.Lett.**103** (2009) 246804
[2] S.Watcharinyanon et al.,Surf.Sci. **605** (2011) 1662
[3] F.C. Bocquet, R. Bisson, J.-M. Themlin, J.-M. Layet, and T. Angot, submitted to Phys.Rev.B (2012)

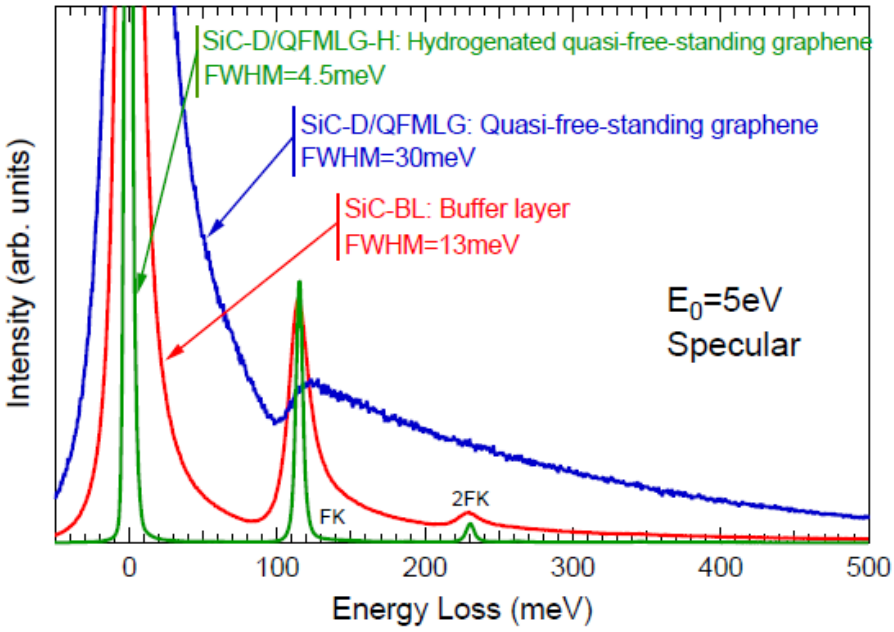

Figure 1 : HREEL spectra of SiC-BL (red), SiC-D/QFMLG (blue), and SiC-D/QFMLG:H (green) samples.

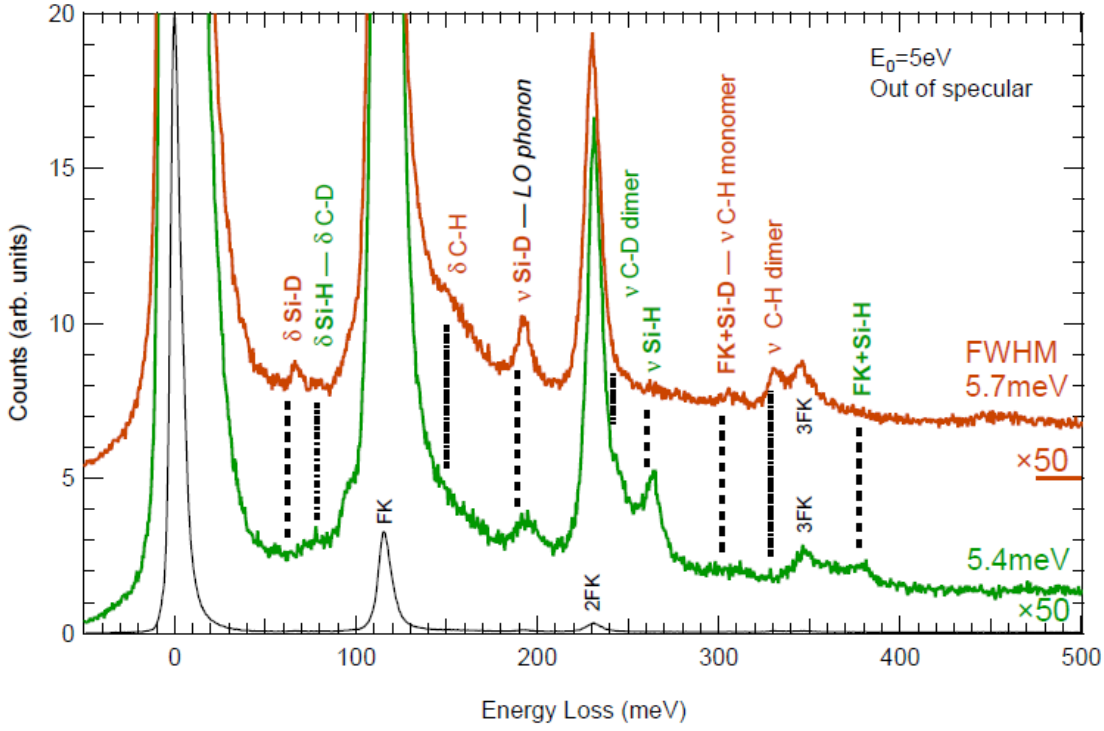

Figure 2 : HREEL spectra of deuterated H-intercalated SiC-H/QFMLG:D (black, and green – x50), and hydrogenated D-intercalated SiC-D/QFMLG:H (red) samples.