Substrate interactions of graphene directly grown on van-der Waals type insulators

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A few ways to grow graphene are known today, among them the decomposition of SiC surfaces [1] and graphene catalysis on metals [2] are the most prospective for future applications in microelectronics. The first is rather expensive due to the costs of the SiC and requires high thermal budgets; the second needs a layer transfer to Silicon. A possible third way would be graphene growth directly on insulating materials, which allow device fabrication without layer transfer.

In this contribution, we present the direct growth of few layer graphene down to bilayers on an insulating van-der Waals substrate containing Si. We will address for the first time the interaction of the grown graphene with the insulating substrate by means of spectroscopic techniques like Raman and synchrotron-radiation based XPS.

A freshly cleaved sample of Muscovite with a stoichiometry of $KAl_2(AlSi_3O_{10})(F,OH)_2$ and 3T (trigonal) structure was annealed in UHV to 800-1000°C and exposed to a direct beam from a solid carbon source (source-substrate distance: 0.3 m). Muscovite is considered as one of the flattest materials available, similar to graphene, and both are van-der Waals systems. In addition, Muscovite is a good insulator due to its band gap of 7.8 eV.

The samples were analyzed with µ-Raman spectroscopy (Renishaw InVia) with a wavelength of 633 nm and a spot size of ~0.5 µm. The laser power at the sample surface was ~0.07 mW. X-ray photoelectron spectroscopy (XPS) was performed at BESSY II (Berlin/Germany) at beamline U56/2 PGM 2 with a SPECS / PHOIBOS 100 hemispherical electron analyzer. The beam diameter was 800 x 100 µm.

Figure 1 shows results of the Raman map. The bright areas (points 3 & 4) in the optical micrograph (panel a) turn out to be few layer graphene with good quality, grown via direct carbon deposition on a van-der Waals alumino silicate. These points exhibit 2 and 4 layers of graphene, respectively. The surrounding is composed of “highly reduced graphene oxide” HRGO (point 2) and of “SiC-like” species (point 1) several 100 µm apart. The spatial distribution of the D-line intensity (the strong intensity of the HRGO D-line was extracted and excluded by a fitting routine) monitors the quality of the grown graphene. The observed quality (small D-line, panel b) is highest for higher number of layers (here 4) (with low 2D/G intensity, panel d). A small shift of the G-line with respect to intrinsic graphene samples by ~3 cm^{-1} indicates an unintentional background doping [3], most probably due to interaction with the insulating substrate.

Spatial resolved XPS is presented in Fig.2. Here, the C 1s emission of the sample was detected when the sample was stepwise scanned by the synchrotron-radiation beam. Three distinct regions can be observed. One exhibits a brought emission in the energy range of carbides at 283 eV (blue) and a weak satellite at 288 eV (carbon-oxygen bonds), for simplicity labeled “SiC-like”. The next spectrum (red) has one line at 284.7 eV (C-C bonds) [4]. The small width of this XPS emission and its asymmetric character are typical for highly conducting species like graphene / graphite. The black spectrum has a strong emission at 288 eV with contributions in the C-C bond and carbide region. In analogy to our Raman results we label this area HRGO; however the single XPS components do not fully reflect the characteristic spectrum of a graphene oxide.

In summary, we demonstrated the feasibility to grow graphene directly on an insulator. In contrast to the van-der Waals like nature of Muscovite and graphene we have clear evidence that an interaction stronger than van-der Waals forces appears which includes chemical bonds. This allows high quality graphene growth for higher numbers of layers but limits the quality of bilayers.

References

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Figure 1

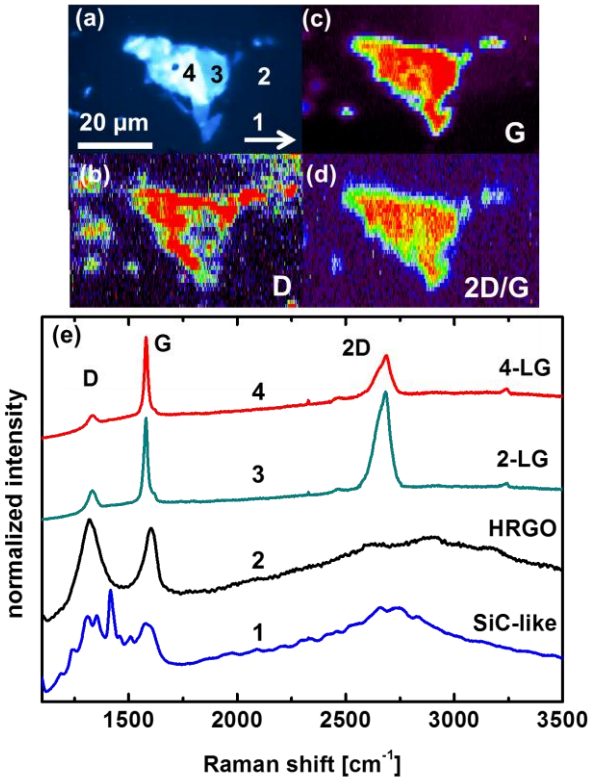

Fig.1. Raman spectroscopic investigation of few layer graphene grown on insulator. Optical micrograph (a), Raman intensity maps of the D and G line and the ratio 2D/G (b-d). The corresponding spectra recorded at the points 1-4 (e).

Figure 2

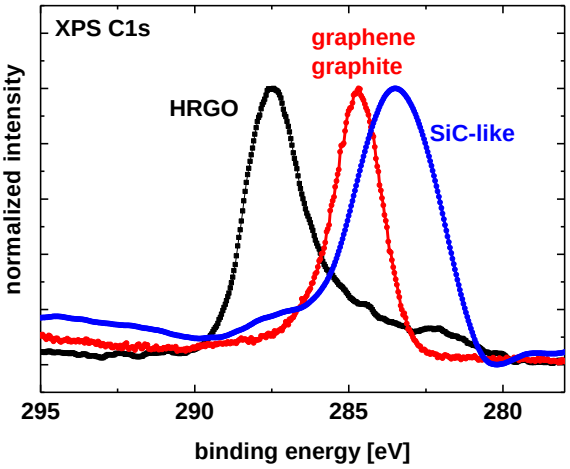

Fig.2. Synchrotron radiation based XPS of the C 1s line of few layer graphene grown on insulator. The spectra represent distinct regions on the substrate.