Experimental Report proposal HC- 899, ID03

"Ru silicide synthesis and oxidation below epitaxial graphene on Ru(0001)", by R. Larciprete, P. Lacovig and S. Lizzit

The possibility to intercalate Si below epitaxial graphene (GR) supported on Ru(0001) as well as on other transition metal substrates is a well assessed process. The aim of this experiment was to investigate the surface reactions which follow the adsorption of Si on the Ru substrate below graphene in order to reveal Si ad Ru intermixing, interface roughening due to island nucleation and, eventually, crystalline phase formation. A further objective was to study the structural properties of the ultrathin SiO_2 layer obtained by oxidizing the Si/Ru interface.

The Ru(0001) surface was prepared by using the standard sputtering and annealing procedure. Graphene was grown by CVD of ethylene at 1100 K. Si was evaporated at low flux on the sample held at 720 K. During the experiment we have prepared two samples which differed for the amount of evaporated Si. In the first case the evaporation was carried out in steps up to about 2.5 ML and each time we performed a complete sample characterization whereas the second sample was prepared by evaporating in once about 4ML of Si. Oxidation was carried out at 640 K by exposing the samples to O_2 at pressure of $1x10^{-3}$ mbar for one hour.

Graphene grown on Ru(0001) forms a highly perfect superstructure composed of regions of alternating weak and strong chemical interactions, in which, according to recent surface X-ray diffraction results, (25×25) unit cells of graphene sit on (23×23) unit cells of ruthenium [1]. Moreover it has been shown that both the graphene layer and the underlying substrate are corrugated, the Ru corrugation decaying slowly over several monolayers into the bulk [2]. Fig.1 shows the in-plane scan along the *h* direction in the neighborhood of the Ru(01) crystal truncation rod (CTR) at *l*=0.2 reciprocal lattice units (r.l.u.). In agreement with the results of Ref.1 two satellites appear on either side of the Ru (01) CTR due to the graphene superstructure. The Ru(1,0) (2,0) (1,1) (0,1) and (0,2) CTRs have also been measured on the Gr/Ru(0001) interface and their analysis will allow a direct comparison with the results reported in Ref. [2]. The Ru(0,1) CTR (*hkl* = 1,0,*l*) is shown in Fig.2.

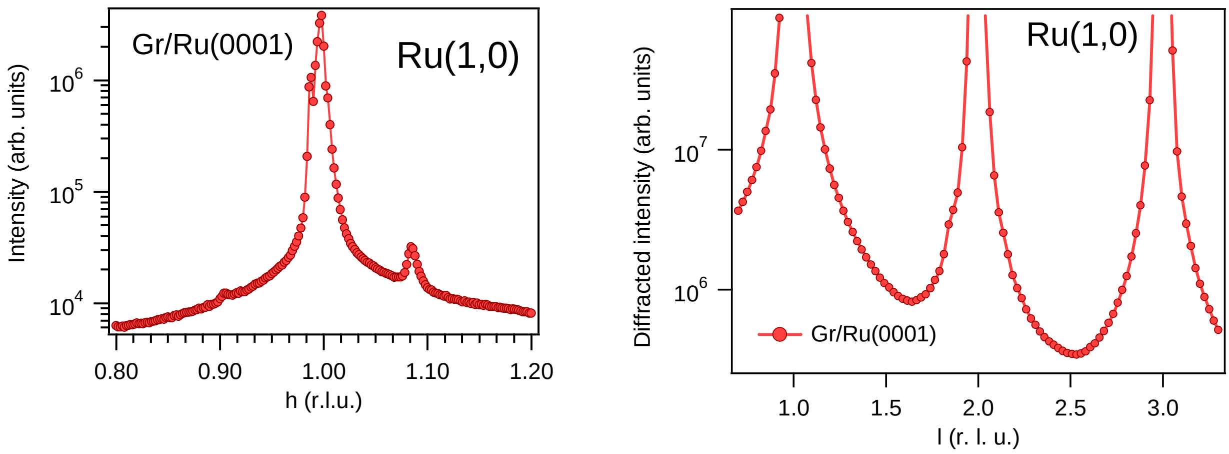
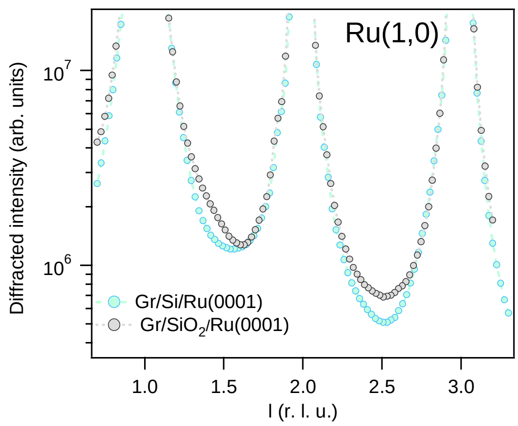


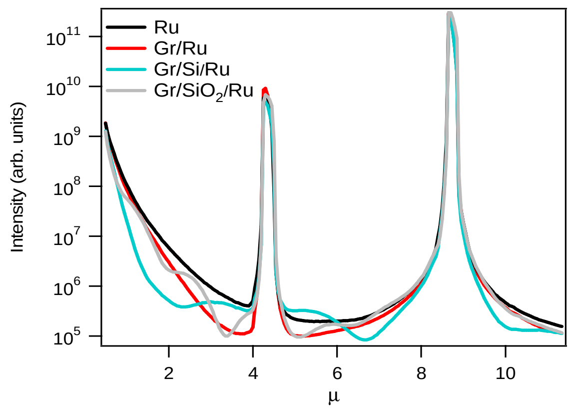
Fig.1 GR/Ru(0001) : left) in-plane scan measured along the *h* direction in the neighborhood of the (01) CTR of Ru at *l* =0.2 r.l.u.; right) Ru(1,0) CTR (*lhkl* = 1,0,*l*)

Silicon intercalation was followed by measuring the *l* scans at several Ru CTRs as well as the *h,k* and *l* scans at the graphene satellites of the Ru(01) and Ru(1,0) CTRs. Moreover we measured the X-ray reflectivity curves as a function of the Si coverage. Figs.2a and 2b show the Ru(0,1) CTRs (*h,k,l* = 1,0,*l*) and the X-ray reflectivity curves measured on the Gr/Ru(0001) surface before and after the intercalation of ~2.5 ML of Si. The analysis of the experimental data is currently in progress and is aimed at deriving a model for the Si gradient along the sample depth and the Ru-Si interface roughness.

Exposure of the Gr/Si/Ru(0001) sample to molecular oxygen in proper temperature and pressure conditions allows the intercalation of O_2 below graphene with the formation of SiO_2. The presence of the satellite peaks in the Ru CTRs after oxidation confirms that neither the O_2 intercalation nor the oxidation reaction induce evident damaging in the graphene layer. The synthesis of the SiO_2 layer coincides with the increase of the intensity between the Bragg peaks in the Ru(01) CTR (see Fi.g2a) and reflects in the modification of the X-ray reflectivity curves (see Fig.2b). The reciprocal lattice map measured on the Gr/SiO_2/Ru(0001) surface and reported in Fig.2c only shows the 1×1 diffraction spots of the substrate crystal indicating the amorphous structure of the oxide layer. Information on the Si adsorption, Ru lattice perturbation, substrate and adatom displacement as a function of Si coverage, as well as evaluation of the Si-Ru alloy and SiO_2 layer thickness, will be obtained through a detailed data analysis which is in progress.



a)



b)

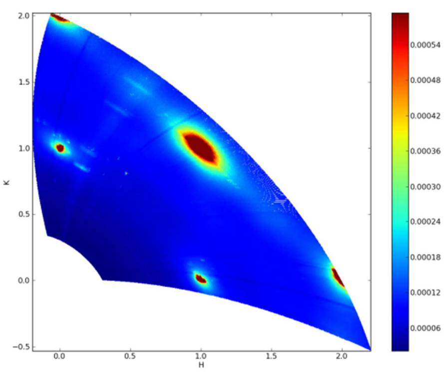


Fig.2 a) Ru(1,0) CTR (*hkl* = 1,0,*l*) measured on the Gr/Si/Ru(0001) surface, at Si coverage of ~ 2.5 ML before and after the interface oxidation at 640 K;
b) x-ray reflectivity curves measured at all reaction steps;
c) *h,k,l* map measured on the Gr/SiO_2/Ru(0001) surface.

c)

[1] Martoccia et al Phys. Rev. Lett. 101, 126102 (2008)

[2] Martoccia et al. New J. Phys. 12, 043028 (2010)

[3] S. Lizzit et al., Nano Lett. **12**, 4503 (2012)