CVD growth and characterization of Graphene on Ge(100) substrates

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Abstract

The direct growth of graphene on CMOS compatible substrates represents a game-changing breakthrough [1]. Germanium is an ideal candidate: it is a semi-metal, it does not form a stable carbide and large-area single-crystal Ge surfaces on Si wafers are becoming routinely available.

Here we present a systematic study of the CVD growth of graphene on Ge(100) substrates. The samples were grown in a Aixtron Black Magic CVD system using H₂ and CH₄ as precursor gases and Ar as a carrier gas. The growth process was investigated as a function of temperature T, H₂/CH₄ flux ratio R, growth time, background pressure. We found a very narrow window of temperatures suitable for the deposition of good quality graphene ranging between 925°C and the melting temperature of Ge. A proper multi-step ramp in temperature was adopted in order to avoid surface damages such as corrugation and pits. The resulting surface is suitable for a large area deposition of graphene and allows for graphene growth at T=935°C, only a few degree Celsius below the Ge melting point.

Raman spectroscopy and XPS were used to study the graphene quality, domain size and coverage of the grown structures. SEM and AFM microscopy were used for morphological investigations of both graphene films and germanium substrates. Conductive-AFM measurements were carried out in order to investigate the correlation between morphological features and sample local electric conductance. The joint use of these techniques allowed for a full characterization of the as grown graphene samples without the need of a transfer process on alternative suitable substrates. This characterization route enables the follow: the Ge surface nano-faceting induced by the presence of CH₄ at high temperature and the development of a graphene layer on the Ge nano-faceted surface as a function of the growth parameters.

A standard Raman analysis of the spectra reported in Figure 1 proves that we were able to deposit a uniform monolayer graphene having size domains larger than 500 nm by using a flux ratio R=100 and T=935°C. For lower values of R we found a multi-layer structure with isolated domains of few-layer graphene deposited on a conformal monolayer film.

The XPS intensity ratio between C 1s and Ge 3d core levels of the same sample demonstrates that these monolayer domains uniformly cover the whole Ge surface, pointing to a self limiting growth condition. The comparison between Raman, XPS and AFM/SEM data indicates that the self limiting growth process is lost at lower values of R for the reduced H₂ etching effect. As a matter of fact the presence of bilayer graphene domains having average sizes of 250 nm and a covering percentage of about 20% is revealed for R=40.

References

Figure 1 Raman spectra of samples grown for 60 min. at different temperatures and H₂/CH₄ flux ratio $R$ ($R=40$, blue line and $R=100$, red line). For comparison, the Raman spectrum of a sample grown at lower temperature ($T=910^\circ$C, green line) is also shown, highlighting the poor quality of the resulting graphitic structures.