Synthesis of nanocrystalline graphene on Al_2O_3(0001) by molecular beam epitaxy

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The implementation of graphene in future nanoelectronics will depend, among many factors, on the capability of producing this material in large-scale and with high quality directly on substrates that are compatible with standard semiconductor technology. Additionally, growing mono- or few-layer graphene on semiconducting/insulating substrates is highly desirable since it enables its electronic characterization and device implementation without the need of a transfer process. In this context, molecular beam epitaxy (MBE) is particularly attractive since it may allow a precise layer-by-layer growth of graphene on such substrates that may result in high-quality material.

In the present work, we investigate the growth of graphene by MBE on 2-inch Al_2O_3(0001) wafers. The *c*-plane sapphire was chosen due to its hexagonal symmetry, which may facilitate graphene epitaxy since its in-plane lattice constant of 4.75 Å is about twice the graphene one (2.45 Å). Besides, the high thermal stability of sapphire allows growth experiments at high substrate temperatures. The carbon films were grown in a MBE system equipped with a solid source of carbon, which consists of a highly ordered pyrolytic graphite (HOPG) filament operating typically at temperatures of about 2400 ºC. Quadrupole mass spectrometry (QMS) analysis reveals that the source emits mostly atomic C, although CO-related species are also observed. The power applied to the source was kept constant in order to obtain similar carbon fluxes. The structural and electronic properties of films prepared with different growth times (between 15 and 480 min) and temperatures (from 800 to 1000 ºC) were studied. The thickness of the films, which cover the whole wafer surface, varied from around 1 to 14 nm depending on the growth time, as determined by atomic force microscopy after a lithography process.

The structural properties of the samples were investigated by Raman spectroscopy. The spectra show the characteristic peaks of a graphitic/graphene structure (see Fig. 1), namely the disorder/defect induced mode at ~1360 cm^{-1} (D peak) [1], the E_{2g} mode at ~1600 cm^{-1} (G peak) [2], and a peak induced by a double-resonance electron-phonon process at ~2700 cm^{-1} (2D peak) [3]. We observe that the sharpness of the D, G, and 2D peaks increase with the growth temperature, for samples prepared with the same growth time [Fig. 1 (a) and (b)]. Such variation is directly connected to an improvement in the degree of crystallinity in the film.

For films prepared with growth times between 15 and 120 min, the analysis of the Raman peaks parameters, such as the relation between the full width at half maximum (FWHM) of the G peak and the intensity ratio between the D and G peaks [I(D)/I(G)], reveals that graphene domains are formed in the carbon film, however with high degree of disorder. On the other hand, for longer growth times [e.g. 480 min, Fig 1(c)] nanocrystalline graphene films with higher quality could be grown, as evidenced by the sharper and much more intense 2D peak. In this case, the observed D peak is likely related to the borders of the graphene domains. It is also worth to notice that the single peak shape of the 2D line [Fig. 1(c)] shows a lack of graphite stacking order between adjacent layers [4]. Thus, despite its thickness (~14nm), the film may behave electronically as a monolayer due to an electronic decoupling between each layer, similarly to what is observed for graphene grown on the C-face of SiC [5]. Finally,

to evaluate the electrical transport quality of the films, we conducted magneto transport measurements using a van der Pauw geometry. A carrier mobility of 120 cm^2/Vs at room temperature and a strong *p*-type doping (~1x10^{13} cm^{-2}) were measured for the films deposited at 1000 ºC for 480 min. Further experiments aiming at the achievement of graphene layers with better structural quality (i.e. larger domains) and thus better transport properties are currently under progress.

References

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Figure

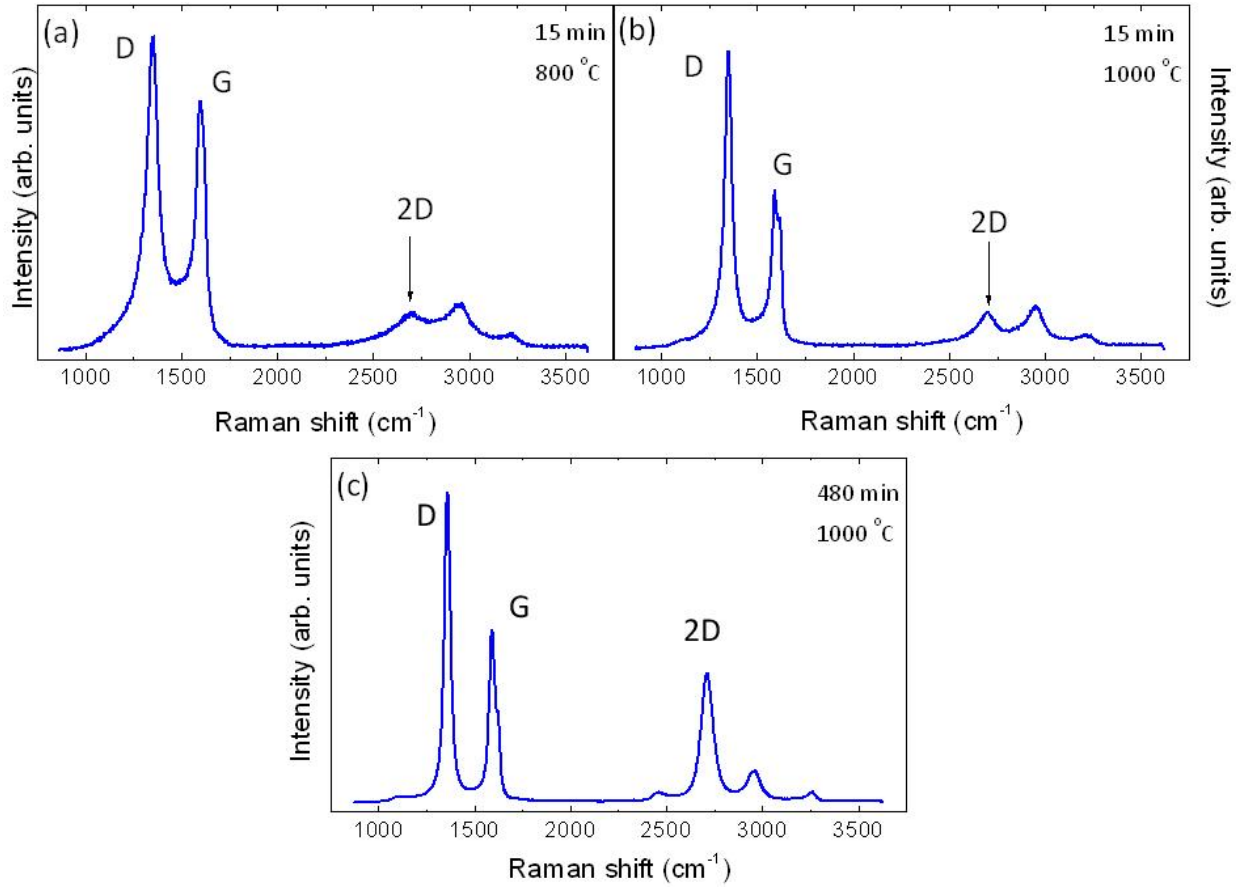

Figure 1 - Raman spectra of MBE grown nanocrystalline graphene films prepared at 800 ºC (a) and 1000 ºC (b, c) for growth times of 15 min (a, b), and 480 min (c). It can be noticed that for increasing time and temperature the 2D peak intensity increases and the FWHM of the peaks decrease, showing the improvement in the structural quality of the grown layers.