

Temperature dependent interaction of graphene with thin TiO₂ and Al₂O₃ layers

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In many graphene device concepts a single or few layer graphene is neighboring device elements composed of metals, dielectrics or semiconductors forming interfaces [1]. The properties of these interfaces and their evolution during technological processing are likely to have a big impact on the performance of graphene devices. In this work, using Raman spectroscopy, we investigate the interaction of thin TiO₂ and Al₂O₃ layers with single layer graphene during annealing treatment to shed some light on the interface reactivity and thermal stability in these material systems.

Graphene flakes were exfoliated from natural graphite and deposited onto 300 nm SiO₂/Si substrates. Subsequently the samples were covered with thin (5-15 nm) layers of TiO₂ (Al₂O₃) by atomic layer deposition at 200 °C. Ti (IV) isopropoxide and TMA precursors were used for TiO₂ and Al₂O₃, respectively. For temperature Raman measurements, graphene flakes sandwiched between TiO₂ (Al₂O₃) and SiO₂ were heated up in flowing N₂ to the target temperature with a rate of 20 °C/min (see Fig.1). Before starting the measurement samples were held at the chosen temperature for 30 min to minimize the drift of the laser spot along the surface. The laser beam ($\lambda = 514\text{nm}$) was focused using a 50x long-working-distance objective lens with NA=0.5. The laser power on the sample surface was ~ 0.3 mW.

Fig.2 (a) shows the evolution of the Raman G band of a graphene sample at various experiment stages. For the as-deposited graphene flake the G peak is located at 1585 cm⁻¹ and is slightly up-shifted with respect to the intrinsic graphene samples (~1580 cm⁻¹ [2]) indicating a hole-doping ($n \sim 1 \times 10^{12}/\text{cm}^2$) possibly due to O₂ adsorbed on graphene [3]. TiO₂ deposition results in a further blue shift of the G peak to ~1591 cm⁻¹. This shift can be interpreted as either an increased hole-doping ($n \sim 4 \times 10^{12}/\text{cm}^2$) [4] or a compressive biaxial strain of ~0.08% in the graphene sheet caused by the TiO₂ layer [5]. Increasing temperature results in a red shift of the G peak. Assuming a linear change of the temperature-induced Raman shift in the chosen range, a G peak temperature coefficient is estimated to be -0.36 cm⁻¹/°C [Fig. 2(b)]. This value is significantly higher than that obtained recently for uncovered graphene on SiO₂ in vacuum (-0.019cm⁻¹/°C) [6] and air ambient (-0.016cm⁻¹/°C) [7]. The disorder-induced Raman D band (~1350cm⁻¹) was not detected during annealing implying a negligible change in the sp² hybridization. This suggests that the SiO₂/graphene/TiO₂ stack is thermally stable at least up to 600 °C and that the graphene sample is still of high quality after the thermal treatment [8].

Beside Raman signatures, we investigate the morphology and chemical composition of the oxide layers covering graphene before and after the thermal treatment and discuss the prospects of using them as barrier dielectrics in graphene high frequency tunneling devices.

References

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Figure1

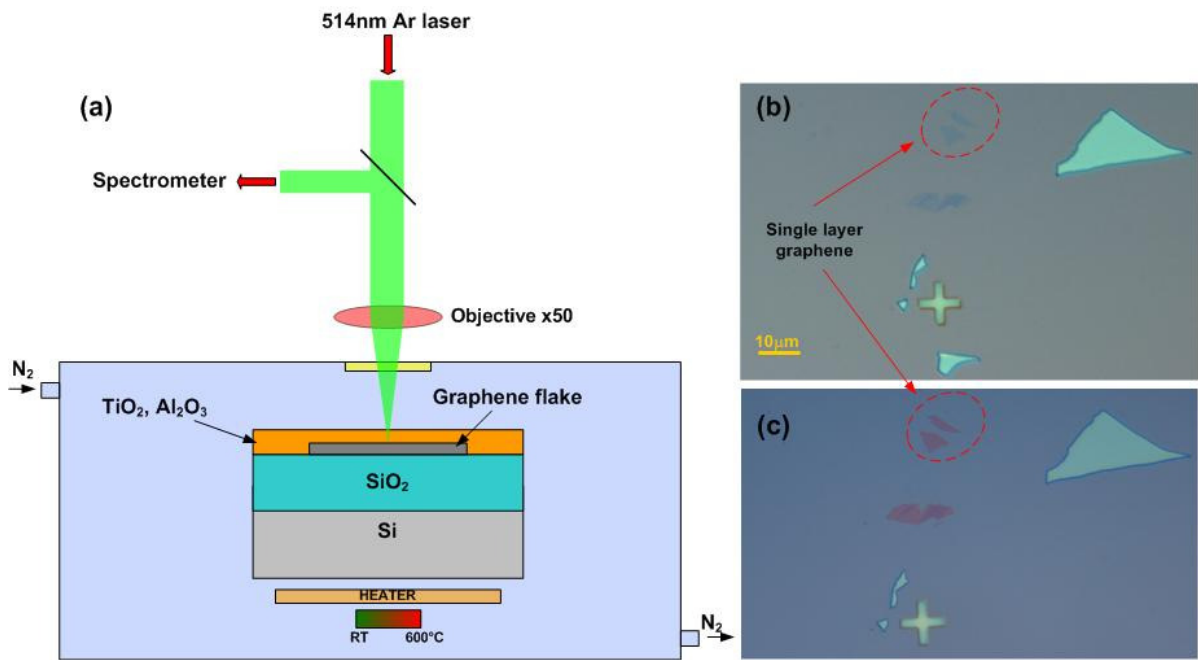


Fig.1. Schematic diagram of the experimental setup (a), optical image of single-layer graphene flakes before (b) and after deposition of ~ 5nm of TiO₂ (c).

Figure2

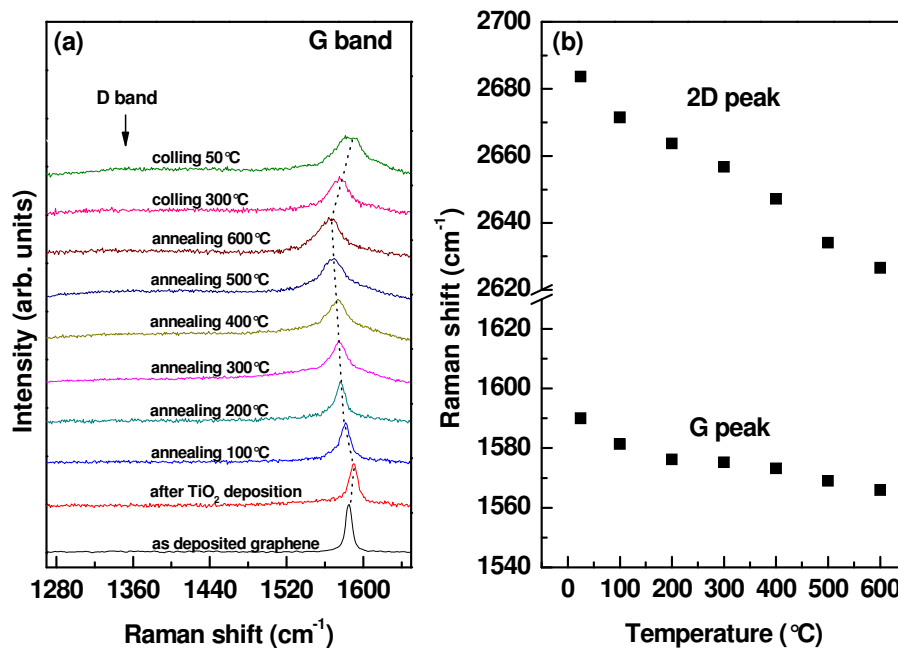


Fig.2. Raman spectra of graphene acquired after subsequent preparation steps (a). The dashed line is a guide to the eye. Variation of the G and 2D peak position as a function of the annealing temperature (b).