Uniform Atomic Layer Deposition of Al_2O_3 on Graphene by Reversible Hydrogen Plasma Functionalization

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A novel method to form ultrathin, uniform Al_2O_3 layers on graphene using reversible hydrogen plasma functionalization followed by atomic layer deposition (ALD) is presented. ALD on pristine graphene is known to be a challenge due to the absence of dangling bonds, leading to non-uniform film coverage. We show that hydrogen plasma functionalization of graphene leads to uniform ALD of closed Al_2O_3 films down to 8-nm in thickness. Hall measurements (figure 1) and Raman spectroscopy reveal that the hydrogen plasma functionalization is reversible upon Al_2O_3 ALD and subsequent annealing at 400 °C, and in this way does not deteriorate the graphene's charge carrier mobility. This is in contrast with oxygen plasma functionalization, which can lead to a uniform 5-nm thick closed film, but which is not reversible and leads to a reduction of the charge carrier mobility.
Density functional theory (DFT) calculations attribute the uniform growth on both H_2 and O_2 plasma functionalized graphene to the enhanced adsorption of Trimethylaluminum (TMA) on these surfaces. A DFT analysis of the possible reaction pathways for TMA precursor adsorption on hydrogenated graphene predicts a binding mechanism that cleans off the hydrogen functionalities from the surface, which explains the observed reversibility of the hydrogen plasma functionalization upon Al_2O_3 ALD.

Figures

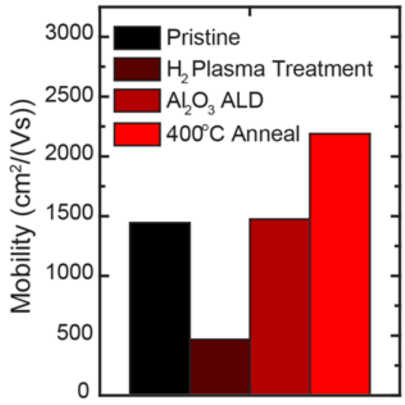

Figure 1: Charge carrier mobility of pristine graphene, H_2 plasma treated graphene, H_2 plasma treated graphene with 8 nm ALD AL_20_3 on top, and after 400°C anneal.

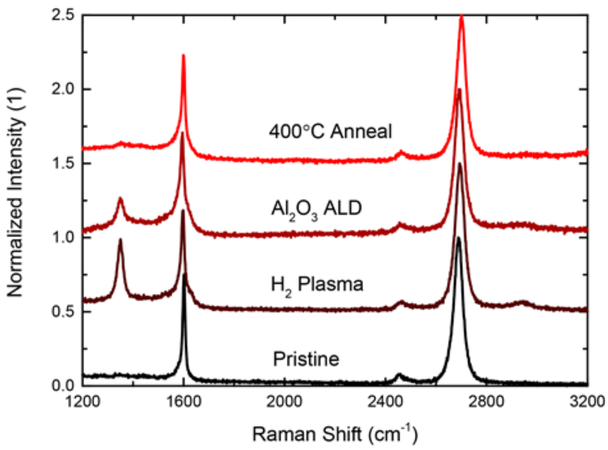

Figure 2: Raman spectrum of pristine graphene, H_2 plasma treated graphene, H_2 plasma treatedgraphene with 8 nm ALD AL_20_3 on top, and after 400°C anneal.