

Intrinsic Adsorption Dynamics of CVD Graphene Investigated by a Contactless Microwave Method

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Using a non-contact microwave dielectric resonator perturbation technique, the conductivity of large area CVD graphene can be determined by measuring the change in resonant peak shape of the cavity when coupled to graphene (Figure 1). [1, 2] Upon adsorption with Nitrogen dioxide (NO_2), a p-doping gas, the graphene sheet increases in hole conductivity which is determined through measurement of the linewidth at resonance.[3] Unlike traditional graphene conductivity measurements, which require metallic contacts, the use of microwaves ensures the only interaction contributing to the change in sheet conductivity is due to the graphene-gas adsorbate interaction.

Using this contactless gas sensing platform, we demonstrate NO_2 gas sensing at concentrations spanning three orders of magnitude. Interestingly, all the sensor response curves can be described using a surface coverage dependent Langmuir adsorption model (Figure 2). We find that the rate of adsorption is dependent upon overcoming an adsorption activation barrier which arises due to competitive adsorption processes with ambient gas species. More importantly, the height of this barrier increases along with the NO_2 surface coverage making sequential NO_2 adsorption exponentially more difficult. This surface coverage dependence presents a fundamental kinetic limitation intrinsic to all 2D material gas sensors. By understanding this kinetic limitation, an effective operational strategy for graphene gas sensors can be pursued.

Figures

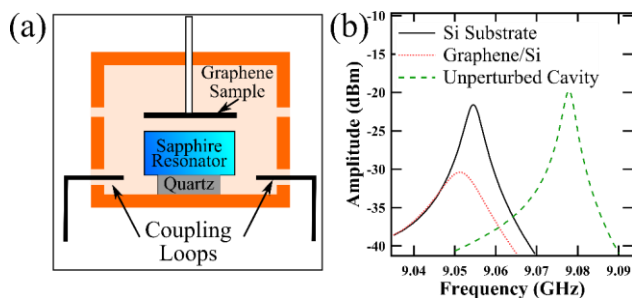


Figure 1: (a) Schematic of the graphene – dielectric resonator coupled system. By measuring the change in frequency and linewidth of the resonant mode (b) in the presence of graphene and graphene substrate the sheet conductivity can be measured. [1]

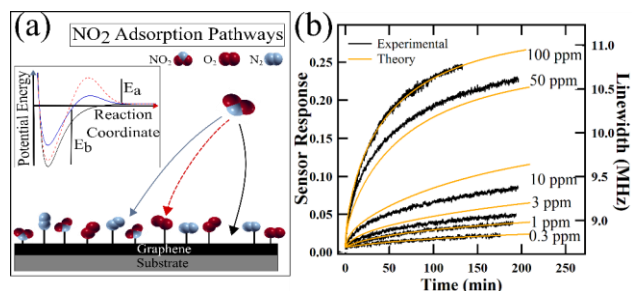


Figure 2: Schematic representation of the possible NO_2 adsorption pathways, emphasizing a graphene surface with a range of different adsorption activation energy barriers to be overcome for successful molecular adsorption. (b) Comparison between the experimental and theoretical sensor responses at concentrations spanning three orders of magnitude.

References

- [1] Hao et al. IET Circuits, Devices and Systems, **9**, 6 (2015) 397-402
- [2] Hao et al. Applied Physics Letters, **103**, 12 (2013) 2011- 123103
- [3] Black et al. Nanotechnology, **28** (2017) 395501