1. Introduction

2D materials can display remarkable electronic properties, but with all the atoms at the surface these properties are easily obscured by scattering off substrate-induced random potentials or adsorbed species. It is therefore desirable to protect these layers by using flat, inert substrates or by removing the substrate altogether [1–10]. Hexagonal boron nitride (hBN) is considered an ideal substrate because of its atomic flatness and its ability to segregate hydrocarbon contaminants into bubbles. Outside these bubbles, hBN provides a pristine surface with significantly reduced charge fluctuations, thus giving access to the low energy electronic properties of graphene and other 2D layers placed on top. Development of methods to pick up and transfer 2D-materials using hBN has led to ultra-clean, high-quality, encapsulated devices such as FETs [11], photodetectors [12], and light emitters [13]. Graphene encapsulated in hBN [14] has revealed a wide range of intrinsic properties including micron-scale ballistic transport [15], electron optics [16, 17], magnetic focusing [18], and Moiré superlattices which exhibit interesting magneto-transport properties [19–21]. Adding a second graphene layer on top of the first further allows one to tune the band structure by controlling the relative angle between the graphene flakes [22, 23]. Devices made from these structures are quickly gaining attention in the physics research community after the observation of correlated electronic ground states at low temperatures [24, 25].

A common challenge in the fabrication of any 2D heterostructure is evaluating and maintaining the cleanliness of the planar interfaces. Protecting the electronic properties of a 2D sample by encapsulation is a delicate process. The quality of such encapsulated devices depends on the location and number of contaminants trapped within the device and on the reliability of the electrical contacts. Typical 2D heterostructures can be constructed in air or in an inert atmosphere glovebox (for reduced contamination) using the polymer stamp method (described in...
We observe that even with substrate components annealed and encapsulation performed in a glovebox, samples often exhibit collections of hydrocarbons in bubbles within the structure, electrostatic charging, adsorbates, strain build-up, and even formation of cracks and tears during encapsulation. Typically, optical microscopy and AFM topography are used to determine regions of the device which are free of bubbles and wrinkles before these relatively defect free regions are isolated by plasma etching (see figure 1(a)); to produce a high-quality device.

Previous works have determined the degree of electronic disorder by measuring the spatial variation of the Fermi level of graphene placed on top of hBN and contacted with an electrode by STS mapping [3, 4, 6, 8, 10]. Transport measurements probe the degree of low-energy electronic scattering by measuring the energy spread of the high resistive state at charge neutrality in encapsulated graphene layers after employing plasma etching and electrode deposition [26]. Here we utilize non-contact AFM-based electrostatic measurements of potential variations near the surface of encapsulated graphene heterostructures. Using simple analysis, we relate these measured quantities to the charge inhomogeneity in the buried graphene layer. Further, we employ high-resolution electrostatic imaging to locate graphene samples, as well as sub-micron sized structural defects such as tears and folds. Our measurements technique benefits from not requiring electrical contact to the graphene layer, making it possible to image and characterize other encapsulated samples non-invasively during different stages of device fabrication.

2. Methods

Devices are fabricated with the standard polymer stamp method using the strong adhesion between graphene and hBN (see supporting information) and placed onto p-doped silicon wafers with a capping layer of 300 nm SiO₂. Devices were constructed using remotely controlled micromanipulator stages inside an argon-atmosphere glovebox and oxygen and water levels maintained below 0.1 ppm.

Samples are measured using an NT-MDT SolverNEXTP system with gold coated AFM tips with typical cantilever stiffness of 3.5 N m⁻¹ (HA-NC probes from K-Tek nano). Relative humidity in the AFM measurement chamber is maintained below 5% by a slow flow of dry nitrogen gas in order to avoid effects of water accumulation on the sample surface.

Electrostatic measurements are performed in the two-pass scheme where in the first pass, the topography is measured using non-contact AFM; on the second pass, electrostatic forces are monitored with the tip retracted 10 nm from the measured sample surface. We note that it is important to ground the doped silicon backgate during measurement to allow quantitative analysis and to avoid charging/discharging of the tip or sample.

High-resolution electrostatic imaging is performed by measuring the electrostatic force gradient near the surface using electric force microscopy (EFM). The cantilever is excited using a piezo block (as in non-contact topography mode) and a DC bias is applied. The applied bias modifies the electrostatic force felt by the AFM tip and induces a phase shift between the driving signal and the cantilever oscillation which can be related to the effective tip-backgate capacitance and the surface potential by

\[
\Delta \Phi = -\arcsin \left( \frac{Q}{k} \right) \quad (1)
\]

where \( Q \) and \( k \) are the quality factor and spring constant of the AFM cantilever, respectively, and \( F_\omega (V_b) \) is the electrostatic force experienced by the tip due to the sample:

\[
F_\omega (V_b) = -\frac{\partial C}{\partial z} (V_b - V_S)^2. \quad (2)
\]

Here, \( C \) is the total capacitance between the tip and backgate, \( V_b \) is the applied tip bias, and \( V_S \) is the potential at the sample surface.

The advantages of imaging with EFM are two-fold. Firstly, the measurement signal is nearly quadratic in the applied bias. This allows one to controllably tune the contrast between regions with different effective capacitances. Second, the phase shift measured is proportional to the second derivative of the capacitance which is highly localized to the tip apex; providing superior resolution to measurement modes which probe the force rather than the force gradient [27–30].

Surface potential measurements are performed with amplitude modulated Kelvin force probe microscopy (AM-KPFM) where DC and AC signal is applied to the tip at frequency \( \omega \), inducing frequency dependent components to the electrostatic force. The force component proportional to \( \cos \omega t \) is given by

\[
F_\omega (z) = -\frac{\partial C}{\partial z} (V_{DC} - V_S) \cdot V_{AC} \cos \omega t, \quad (3)
\]

where \( V_{DC} \) and \( V_{AC} \) are the applied DC and AC components of the bias applied to the tip. During the KPFM measurement, this component is monitored and the DC bias is controlled by feedback to nullify \( F_\omega (z) \), i.e. find \( V_{DC} \) such that \( V_{DC} = V_S \) determining the surface potential of the sample.

On Si/SiO₂, the sample surface potential can be related to work function difference between tip and backgate modified by charges within the structure:

\[
V_S = \frac{\Delta \Phi}{e} - \frac{q}{C_b}, \quad (4)
\]

where \( \Delta \Phi \) is the workfunction difference between the gold-coated tip and the silicon backgate, \( e \) is the charge of an electron, \( q \) is the total charge trapped within the structure beneath the tip, and \( C_b \) is the capacitance between the localized charge and the grounded backgate [31].
Now, assuming graphene forms parallel plate capacitor with the backgate and that the charge in graphene is uniformly distributed (i.e. tip-graphene image interactions are negligible), we find that the difference in surface potential measured between regions with encapsulated graphene present and regions without graphene can be given simply by

$$\Delta V_S = \frac{\varepsilon_{hBN} t_{bot} + \varepsilon_{ox} t_{bot}}{\varepsilon_{0} t_{bot} + \varepsilon_{hBN}} \varepsilon_{2D},$$

where \(t_{bot}\) is the thickness of the silicon oxide (300 nm), \(\varepsilon_{ox}\) is the dielectric constant of silicon oxide (about 3.9), \(\varepsilon_{hBN}\) is the out-of-plane dielectric constant of hBN (about 3.8), \(\varepsilon_{0}\) is the permittivity of free space, \(t_{bot}\) is the thickness of the hBN flake beneath the graphene, and \(\varepsilon_{2D}\) is the 2D charge density of the buried graphene layer. Thus, after measuring the thickness of the bottom hBN flake by AFM topography we are able to extract the 2D carrier concentration of the encapsulated graphene layer by simply comparing its local surface potential to that of the surrounding regions.

3. Results and discussion

3.1. Electrostatic force microscopy (EFM)

We first image the samples by EFM phase shift mapping. The optical micrograph of an encapsulated double layer graphene sample is shown in figure 1(a). It consists of two stacked graphene monolayers (~zero-degree twist angle) and is left electrically isolated during this measurement. The first pass of the measurement records the sample topography and is displayed in figures 1(c) and (d). The encapsulated graphene flake is buried between a 71 nm hBN flake underneath and a 50 nm hBN flake on top. By applying a bias between the AFM tip and the silicon backgate, we are able to selectively image the buried graphene layer. Figure 1(e), we measure the EFM phase shift map along the dashed line in (g) shows the sharp edge resolution obtainable with this technique.
at a DC bias $V_b = -6 \text{ V}$ in the same region shown in figure 1(c) and observe a large phase shift difference when the tip passes above the region of encapsulated graphene.

Note that we are also able to observe the local gold backgate which runs beneath the sample in the phase shift measurement, showing that this method may be employed for other thin conducting materials.

We measure the phase shift versus bias in different regions of the device (figure 1(f)): a region of bare hBN (no graphene), an encapsulated monolayer graphene flake, and one region where two graphene layers overlap (BLG). In each region, the signal minimum is related to the surface potential while the curvature relates to the second derivative of the local capacitance. The phase shift minima nearly coincide in all regions of the device; however, there is a large enhancement of $\frac{\partial^2 C}{\partial z^2}$ above the graphene layer compared to the surrounding regions both due to the screening ability of the graphene layer and due to the close proximity of the graphene to the tip. Thus, the contrast between the regions of encapsulated graphene and the surrounding hBN stack grows with increasing magnitude of $V_b$.

A zoomed-in phase shift map is taken at the corner of the device where the two encapsulated graphene monolayers overlap (figure 1(g)) and a line cut through the AFM topography scan along the teal line in (b). The graphene sits on top of a hBN flake 37 nm thick with three different thicknesses of hBN on top: 11.6 nm, 21.3 nm, and 30.7 nm. (e) The cantilever phase shift recorded at $V_b = -6 \text{ V}$ for three different thicknesses of encapsulating hBN marked in (c) shows a monotonic decrease of the force gradient as thickness increases. (f) The cantilever phase shift versus lift height taken at $V_b = -3 \text{ V}$ above another encapsulated graphene sample (not shown) displaying the sharp decrease in contrast between the encapsulated graphene and surrounding hBN as the tip-sample separation grows large.

![Figure 2](image-url)
3.2. Kelvin probe force microscopy (KPFM)

Next we employ AM-KPFM to map the surface potential of the sample shown in figure 1. The results are displayed in figure 3. The region where graphene is encapsulated shows a lower surface potential with respect to the tip than the surrounding boron nitride indicating a difference in local charge density given by equation (5). By determining the thickness of the bottom hBN flake (from figure 1(d)) and evaluating using equation (5), we can determine the 2D charge density within the graphene layer to be \( n_g = 0.92 \times 10^{10} \text{ cm}^{-2} \). This level of doping is typical for encapsulated graphene devices fabricated in a dry atmosphere. Despite fabrication in a controlled environment using fresh, clean materials, we always observe some small amount of charge bound to the graphene layer. This charge density is large enough to provide the contrast observed in the surface potential map.

Due to the charge trapped in the embedded graphene, we observe a surface potential contrast between the encapsulated monolayer and bilayer graphene. The difference in doping dependence of monolayer and bilayer graphene stems from the difference in low energy band structures. To maintain the same Fermi level, the bilayer graphene requires a larger charge density, as seen in figure 3(b). With the surrounding boron nitride stack as background, we find a charge carrier concentration in the bilayer region of \( n_{BLG} = 1.16 \times 10^{10} \text{ cm}^{-2} \). Note that in AFM topography (figure 1(c)), neither the graphene edge nor the monolayer/bilayer boundary are visible. By contrast, in KPFM these features are well resolved making it possible to select specific sample regions such as monolayer, bilayer, or boundary for fabricating well characterized devices.

Upon fabricating and attaching electrodes to the graphene layer, the trapped charge will change due to the work function differences between the graphene layer and the contacting metal. Thus, the absolute charge density is not a good metric for device quality. Instead, we may look at the surface potential fluctuations within the encapsulated graphene region. The potential within the graphene layer is expected to be uniform and thus, any surface potential fluctuations must originate from charge traps outside the graphene layer. Surprisingly, despite the presence of bubbles observed in sample topography (figure 3(a)), the
measured surface potential is not affected. However, we do observe fluctuations in the surface potential across the device surface reflected in the RMS spread of the surface potential distribution (figure 3(d)). Additionally, in some regions of the device, we observe local potential fluctuations (up to 70 mV) which are not associated with topographical features (figures 3(e) and (f)). These features provide strong electronic scattering centers which might accidentally be included in a processed device selected by using AFM topography alone. It is well-known that charge traps, which are typically found within the insulating SiO2 substrate, and at the various interfaces, provide a random potential and scattering centers which reduce the device’s transport properties. We measure the RMS variation in surface potential in several regions of the device. For the monolayer (g) and bilayer (BLG) regions, we find RMS variations of $\delta V_p = 26.09$ mV and $\delta V_{BLG} = 18.73$ mV corresponding to charge variations of $\delta n = 1.51 \times 10^6$ 1 cm$^{-2}$ and $\delta n_{BLG} = 1.08 \times 10^6$ 1 cm$^{-2}$, respectively. The values obtained are comparable to those measured by transport in similar devices [26]. Thus, we can use this metric to identify pristine regions of the sample that are suitable for further processing so as to maximize device quality.

We provide, in the supporting information, a description of the encapsulation procedure, measurements of dirty and defective samples identified by EFM, and preliminary results regarding the effects of annealing on sample uniformity.

4. Conclusions

In conclusion, we have demonstrated that conductive scanning probe techniques are well suited for achieving high-contrast and high-resolution imaging of 2D conductive layers encapsulated in hBN. We show that the small interaction volume between the tip and sample enables the user to visualize edges of the embedded flake at the sub-micron scale. Employing surface potential mapping with KPFM, we identify the positions of charged contaminants within and on top of the heterostructure through fluctuations of the measured surface potential identifying electronically defective and pristine regions of the buried graphene. The techniques used are extensions of atomic force microscopy which are commercially available or can be implemented through simple upgrades to existing AFM systems. We expect that these methods will be valuable tools for fabricating high quality transport devices by avoiding contaminated and defective regions of the encapsulated flake(s). Finally, we hope that these techniques will lead to the production of pristine heterostructures, open new avenues for 2D device characterization methods, and provide opportunities for novel research.

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