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High temperature MBE of graphene on sapphire and hexagonal boron nitride flakes on sapphire

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The discovery of graphene and its remarkable electronic properties has provided scientists with a revolutionary material system for electronics and optoelectronics. Here, the authors investigate molecular beam epitaxy (MBE) as a growth method for graphene layers. The standard dual chamber GENxplor has been specially modified by Veeco to achieve growth temperatures of up to 1850 °C in ultrahigh vacuum conditions and is capable of growth on substrates of up to 3 in. in diameter. To calibrate the growth temperatures, the authors have formed graphene on the Si-face of SiC by heating wafers to temperatures up to 1400 °C and above. To demonstrate the scalability, the authors have formed graphene on SiC substrates with sizes ranging from 10 × 10 mm² up to 3-in. in diameter. The authors have used a carbon sublimation source to grow graphene on sapphire at substrate temperatures between 1000 and 1650 °C (thermocouple temperatures). The quality of the graphene layers is significantly improved by growing on hexagonal boron nitride (h-BN) substrates. The authors observed a significant difference in the sticking coefficient of carbon on the surfaces of sapphire and h-BN flakes. Our atomic force microscopy measurements reveal the formation of an extended hexagonal moiré pattern when our MBE layers of graphene on h-BN flakes are grown under optimum conditions. The authors attribute this moiré pattern to the commensurate growth of crystalline graphene on h-BN. © 2016 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution 3.0 Unported License.

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I. INTRODUCTION

The discovery of graphene and its remarkable electronic properties by Geim and colleagues in 2004 (Ref. 1) has provided scientists and engineers with a material system for revolutionizing electronics and optoelectronics. The initial breakthrough discovery focused on the properties of a planar field effect transistor made by mounting a sheet of exfoliated graphene on the oxidized surface of a doped silicon wafer. Pioneering experiments on these devices have elucidated graphene's remarkable electronic properties—the high

carrier mobility and velocity with zero effective mass and the very low density of states around the Dirac neutrality point. However, the planar graphene transistor suffers from a fundamental property of massless, “relativistic” fermions. Whereas a field effect transistor based on Si or GaAs can be switched off by closing the conducting channel with a gate voltage, an electron current-carrying state in graphene changes into a hole state by “tunneling” through the potential barrier, so current continues to flow. This makes it difficult to switch a planar graphene transistor into a nonconducting off state. The Manchester group overcame this limitation by realizing a new type of graphene transistor architecture, namely, a field-effect transistor based on quantum tunneling

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from one graphene electrode through a thin insulating barrier.² Later a variant of this device: a barrier transistor or “barristor” fabricated from a layer of graphene had been grown by chemical vapor deposition (CVD).³ Multilayer graphene-based devices are evolving rapidly; for example, the Manchester group have now observed and modeled voltage-tuneable negative differential conductivity (NDC) up to room temperature in graphene tunnel transistors.⁴ This opens up the exciting possibility of new fast logic devices, which exploit the multivalued $I(V)$ curve and of high-frequency mixers and fast oscillators and mixers, which exploit the nonlinearity and the gain that can be achieved in the NDC region. This type of device structure also has potential for photonic applications.

The Manchester group and other groups internationally have used small (tens of micrometers) exfoliated layers of graphene and of other layered crystals such as hexagonal boron nitride (h-BN) to construct their heterostructures layer-by-layer. A viable commercial technology based on this type of devices will require large area wafers (>2 in. diameter) with a precise control over the thickness and composition of the layers and, in addition, a reliable processing method for low resistance contacts and interconnects.

The growth of graphene monolayers by CVD on single crystalline transition metals such as Co, Pt, Ir, Ru, and Ni has been studied extensively in recent years.^{5–7} In the CVD process, the metal film is exposed to a hydrocarbon flow under ambient or low pressure at temperatures of ~600–1000 °C. After the process, the graphene films are transferred to the required substrate. It has been demonstrated that the CVD-produced graphene is not only suitable for displays, solar cells, and other applications requiring large areas of graphene, but also is of sufficient quality to make functional electronic devices. However, the small grain size (~1 μm) places severe limits on orientational order in multilayer structures. The presence of impurities arising from atmosphere and the release of the graphene from the metal substrate combined with subsequent handling indicate that, for a fully developed technology, the vertical devices envisaged here cannot be easily realized by sequential stacking of exfoliated of the CVD-grown layers.

An alternative method of growing graphene is to heat SiC to very high temperatures, in the range of 1250–1450 °C.^{8,9} This leads to the formation of an ultrathin epitaxial graphene film on the Si-terminated (0001) face of single-crystal 6H-SiC by thermal desorption of Si. Although this method can produce relatively large area graphene films, it is not an epitaxial technique suitable for engineering graphene-based heterostructures.

The ultrahigh vacuum environment offered by molecular beam epitaxy (MBE) together with the high purity of sources provides crucial advantages for the formation of heterostructures over large areas. Several years ago, we attempted to grow graphene in our existing MBE system at Nottingham on different substrates including SiC, GaAs, GaN, BN, and sapphire at growth temperatures of up to ~700 °C. We obtained amorphous carbon layers and our negative results were not published. High growth temperatures are essential for the growth of high quality graphene rather than amorphous carbon films. Recently,

graphene has been grown by MBE using both gaseous and solid sources for carbon.^{10–16} These studies indicate that high growth temperatures (above ~1000 °C) are required to provide the necessary mobility of carbon on the growth surface.

In this work, we investigated growth by MBE of graphene layers at extremely high substrate temperatures.

II. EXPERIMENTAL SETUP AND METHODOLOGY

We have studied the growth of graphene using a custom-designed dual chamber MBE system. The standard dual GENxplore MBE system has been specially modified by Veeco to achieve growth temperatures of up to 1850 °C (thermocouple temperatures) in ultrahigh vacuum conditions and is capable of growth on substrates of up to 3 in. in diameter. The growth chambers have a vertical configuration with the substrate heater mounted above the sources.

The engineering of future scalable and integrated devices based on high-quality graphene/boron nitride heterostructures will require substrate diameters of 2 in. or more. Ideally, the substrate should be stable at high temperatures ~1500 °C, easily available, and of relatively low cost. The best candidates at the current time are sapphire, templates of AlN on sapphire, boron nitride, silicon carbide (SiC), or bulk AlN substrates (if they become available at reasonable cost). In this study, we used SiC and sapphire substrates.

In MBE, the substrate temperature is normally measured using an optical pyrometer. Because we used transparent SiC and sapphire substrates, the pyrometer measures the temperature of the substrate heater, not the substrate surface. Therefore, our estimate of the growth temperature is based on a thermocouple reading.

We have used 10 × 10 mm² double side polished semi-insulating (0001) 4H-SiC from Norstel and 3-in. diameter double side polished n-doped 4H-SiC wafers from SiCrystal AG.

We use a SUKO-63 carbon sublimation source from Dr. Eberl MBE-Komponenten GmbH to grow graphene on sapphire at substrate temperatures between 1000 and 1650 °C (thermocouple temperatures). The double and single polished sapphire (0001) substrates from SurfaceNet GmbH ranged in size from 10 × 10 mm² up to 3-in. in diameter.

To grow graphene on h-BN surfaces, we have used h-BN flakes from high pressure and high temperature grown bulk BN crystals.¹⁷ The h-BN flakes were exfoliated using low tack acrylic adhesive tape (Loadpoint, Ltd.). The flakes were then deposited on the (0001) sapphire wafer by contact and slow removal of the adhesive tape. The substrates were then placed in toluene (99.9% CHROMASOLV for HPLC, Sigma Aldrich) overnight to remove the majority of the adhesive tape residue. The final removal of tape residue was completed by annealing in an H₂:Ar (5%:95%, respectively) gas flow at 0.15 slpm at 400 °C for 8 h in a quartz tube.

A wide range of techniques were used to characterize graphene layers: reflection high energy electron diffraction, Raman spectroscopy, x-ray photoelectron spectroscopy, atomic force microscopy (AFM), scanning tunneling microscopy, ellipsometry, and Hall effect measurements.

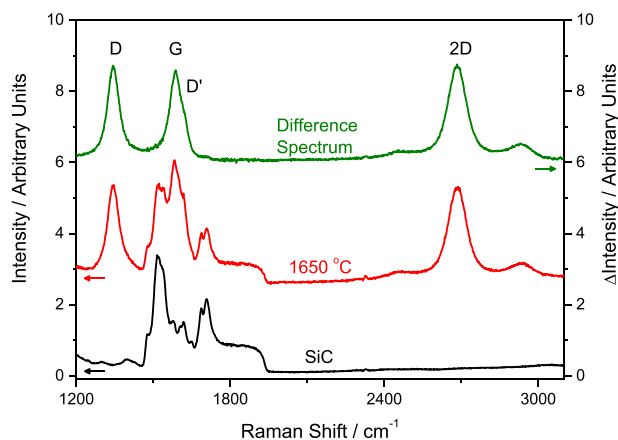


FIG. 1. (Color online) Raman spectra of SiC, after thermally annealing of SiC at 1650 °C and the difference spectrum showing the new peaks.

AFM measurements were performed in amplitude-modulated tapping (AC) mode in an Asylum Research Cypher-S AFM system using MultiAI-75 AFM probes (Budget Sensors) at set points between 400 and 600 mV. The STM measurements were performed in ambient conditions in a Molecular Imaging picospm STM using Pt:Ir (80%:20%) wire tips. Imaging was performed in constant tunneling current mode at a set point of 50 pA and a tip bias of +1 V. Images were processed using the GWYDDION software package.

Spectroscopic variable angle ellipsometry of the films was carried out using a J. A. Woollam M-2000 ellipsometer.

Raman spectra were recorded on a Horiba–Jobin–Yvon LabRAM Raman microscope, with an excitation laser wavelength of 532 nm operating at low power (~ 4 mW) and a 600 lines/mm grating. The detector was a Synapse CCD detector. The Raman shift was calibrated using an Si(100) reference sample. Raman spectroscopy provides a powerful tool for the rapid nondestructive analysis of graphene. Defect free graphene exhibits two key features in the Raman spectrum: the G band at ~ 1580 cm^{-1} arises from the degenerate iLO and iTO phonons at the Brillouin zone center, while the 2D band at ~ 2700 cm^{-1} is due to an intervalley

double resonance scattering phonon of the iTO phonon near the k point.¹⁸ This band has been shown to assist in the discrimination of multilayer graphene, splitting into two or more features for Bernal stacked graphene. However, care must be exercised in assigning a single 2D band to monolayer graphene; both multilayer Bernal stacked graphene grown by the thermal decomposition of SiC (Ref. 19) and turbostratic graphene²⁰ have been shown to exhibit one 2D band. The ratio of I_{2D}/I_G , which has also been used to discriminate layers in ordered graphene, is also unreliable in these instances. In addition to these features, the presence of defects in the graphene structure gives rise to the D and D' bands at ~ 1350 and 1620 cm^{-1} , respectively, which can be used to quantify the proportion and type of defects.^{18,21}

III. RESULTS

It is well established that graphene can be formed by heating of SiC to very high temperatures, 1250–1450 °C.^{8,9} This leads to the formation of an ultrathin epitaxial graphene film on the Si-terminated (0001) face of single-crystal SiC by thermal desorption of Si.

To calibrate the growth temperatures in our GENxplor MBE system and to test the capabilities of the system, we have formed graphene on the Si-face of SiC by heating 10×10 mm² double polished semi-insulating (0001) 4H-SiC wafers to temperatures up to 1400 °C and above. Figure 1 shows the Raman spectrum of SiC, along with the spectrum obtained after annealing a 1 cm² sample of SiC at 1650 °C for 2 h. The formation of graphene is confirmed by the appearance of the D and 2D bands at ~ 1342 , and ~ 2683 cm^{-1} , respectively. Due to the presence of features from the SiC bulk, the G band was obscured; therefore, a different spectrum was obtained by subtracting the reference SiC spectrum from the one obtained for the annealed sample (Fig. 1). In order to ensure efficient subtraction of the SiC bands, a multiplier was applied to the reference SiC spectrum prior to subtraction. The G band is then clearly observed at ~ 1588 cm^{-1} .

Figure 2 shows STM images of graphene formed on (0001) 4H-SiC. Our images are in good agreement with previous studies, which identify regions of monolayer graphene

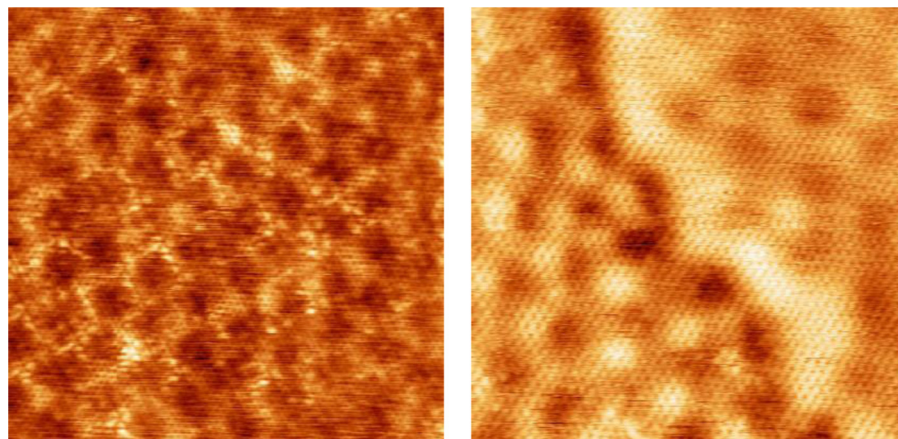


FIG. 2. (Color online) STM images of graphene on SiC: left (15 × 15 nm) monolayer graphene on the reconstructed SiC surface; right (11 × 11 nm) graphene lattice showing regions of monolayer coverage, bottom left and bilayer coverage upper right.

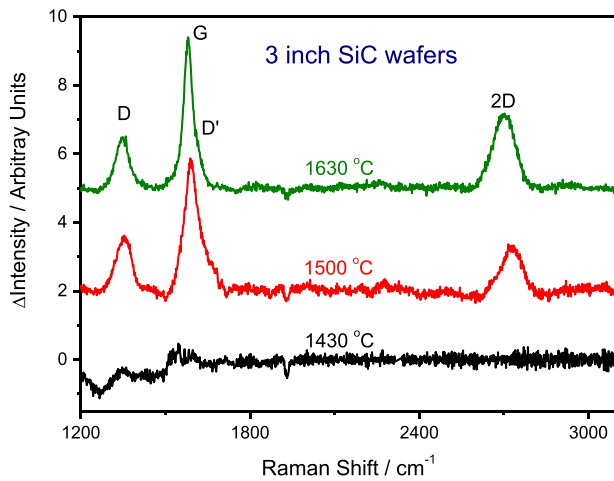


Fig. 3. (Color online) Raman difference spectra showing the additional peaks present after the thermal annealing of SiC at various temperatures on 3 in. wafers.

on the $6\sqrt{3} \times 6\sqrt{3}$ reconstructed SiC surface.²² A variation in the apparent contrast of the underlying reconstruction is observed in regions where bilayer graphene is formed. This is observed in the upper right corner of the image in Fig. 2 (right); note that the graphene's atomic lattice is observed across the complete surface in both the images in Fig. 2.

To demonstrate the scalability of the process, we have formed graphene on SiC substrates with sizes ranging from $10 \times 10 \text{ mm}^2$ up to 3-in. in diameter. Figure 3 shows the spectra of 3-in. 4H-SiC wafers annealed at various temperatures. At the lowest annealing temperature (1430°C), no evidence for the formation of graphene was observed, whereas for the two samples annealed above 1500°C , the presence of peaks assigned to graphene was clearly observed.

The effect of growth temperature was compared by growing samples at various temperatures from 1080 to 1740°C on $10 \times 10 \text{ mm}^2$ sapphire substrates, as demonstrated in the Raman spectra shown in Fig. 4. At the lowest growth

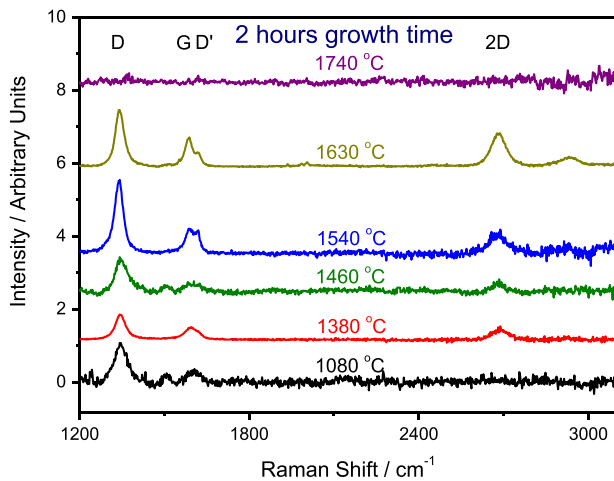


Fig. 4. (Color online) Raman spectra showing the effect of the substrate temperature on the growth of carbon films on sapphire. D and G bands were observed for all of the growth temperatures apart from the highest (1740°C). The 2D band is observed for the temperature range of 1380 – 1630°C .

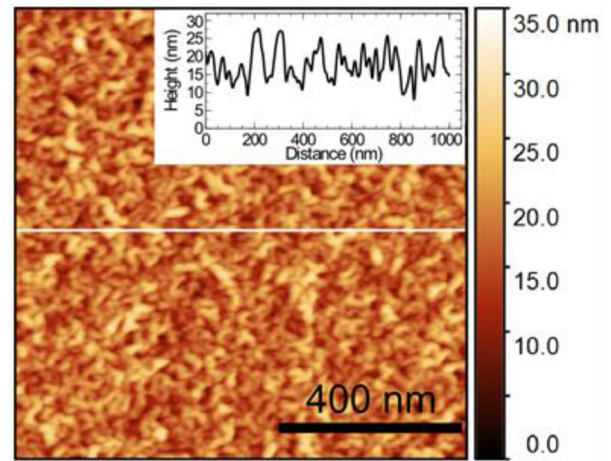


Fig. 5. (Color online) AFM image of the surface of a carbon film grown on sapphire, (inset) profile across the region indicated by the white line in the center of the image.

temperature, no 2D peak was observed with only the D and G peaks present, indicative of the formation of amorphous carbon. For temperatures between 1380 and 1630°C , the formation of graphene was observed as is evident from the presence of the D, G, and 2D bands. The 2D band shape is consistent with the formation of turbostratic graphene. At 1740°C , no bands were observed, which could be due to evaporation of carbon atoms from the hot substrate.

Carbon forms a continuous layer on the surface of the sapphire wafers, as shown in Fig. 5. However, the carbon layer is relatively rough as can be seen from the AFM profile in the inset of Fig. 5, which has a roughness of $R_q \approx 3 \text{ nm}$.

After establishing the optimum growth temperature, graphene samples were prepared during various growth times (5 – 120 min) at a fixed growth temperature of 1630°C . The Raman spectra are summarized in Fig. 6. The ratio of the I_{2D}/I_G increases with time, similar to results shown earlier by the other MBE groups.¹³ The I_D/I_G ratio, after initially increasing between 5 and 20 min , decreased for subsequent

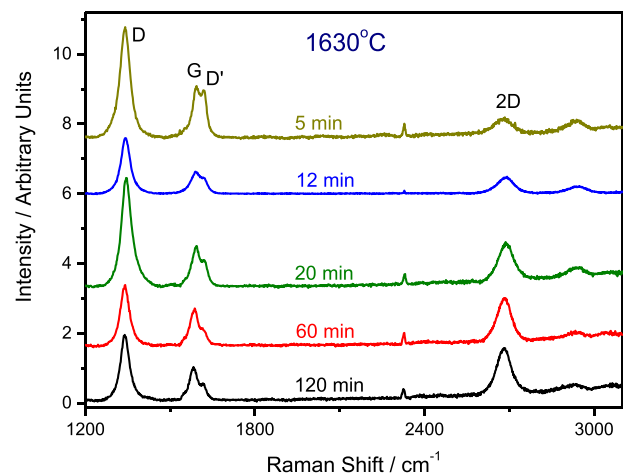


Fig. 6. (Color online) Raman spectra showing the effect of the growth time on the formation of carbon films on sapphire. The I_{2D}/I_G ratio increases with growth time, while the I_D/I_G ratio increases between 5 and 20 min and then subsequently decreases.

growth time. This could be due to an initial increase in the defect concentration at the short growth times; alternatively, the I_D/I_G ratio has been observed to decrease for very small domain sizes (<3 nm);²³ therefore, this trend could also be explained by an initial formation of very small domains, which then increase in size with growth time.

The carbon layers deposited onto the sapphire substrate are very thin, typically on the scale of a few tens of monolayers thick. As a result, AFM is used to measure the thickness of this material following growth. The carbon layer is weakly attached to the sapphire substrate and can be easily removed by a simple scrape with clean plastic tweezers, which exposes the underlying sapphire substrate. The thickness is then measured by scanning the edge of the region where the material has been removed, as shown in Fig. 7. In this example, the thickness of the carbon layer is ~ 17 nm. This method allows calibration of the growth rate. For example, using a carbon source filament current of 95 A, the layer growth rate is ~ 2 nm/h at a substrate temperature of 1500°C .

Our MBE films grown on sapphire are conducting with a resistance per square between ~ 10 and 200 k Ω depending on the layer thicknesses and growth conditions. Hall effect measurements on van der Pauw samples give room temperature p -type mobilities of up to 7 ± 2 cm²/V s and hole sheet densities of about $1\text{--}2 \times 10^{13}$ cm⁻² for layer thickness of 5 nm. The Hall voltage increases linearly with magnetic fields up to 16 T. This mobility is almost 1 order of magnitude higher than those reported for MBE growth of nanocrystalline graphene on sapphire substrates at lower MBE growth temperatures.²⁴

Spectroscopic variable angle ellipsometry of the films was carried out and the ellipsometric response was fitted using three Lorentz oscillators.²⁵ The three oscillators describe the exciton-dominated absorption peak, the Drude-like conductivity in the near-IR, and the broad response in the region of constant optical conductivity. The thickness of the film in Fig. 8 is found to be 13 ± 2 nm in agreement with AFM measurements. Using the Drude-like response, a resistance per square

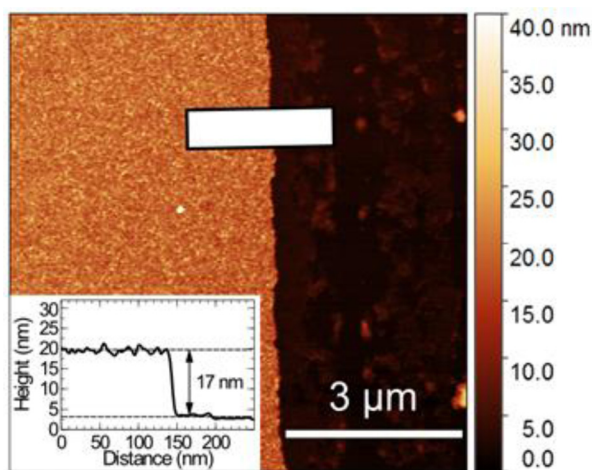


Fig. 7. (Color online) AFM image of the surface with the step between sapphire substrate and carbon layer (inset) line profile averaged across the region indicated by the white box showing the thickness of the carbon film.

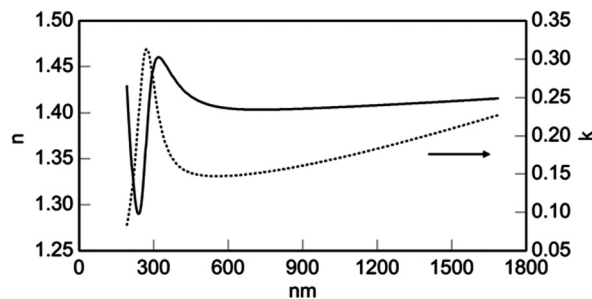


Fig. 8. Optical constants n and k of a graphene layer grown by MBE on a sapphire substrate.

of around 15 k Ω per square is found, in reasonable agreement with low frequency magneto-resistance measurements.

The quality of the graphene layers is significantly improved by growing on h-BN substrates. We have grown graphene on h-BN flakes mounted on sapphire substrates over a substrate temperature range from 700 to 1650°C (thermocouple temperatures). The amount of the carbon deposited on sapphire was significantly higher than that on the surface of h-BN flakes. This can be explained by the significantly higher mobility¹² and lower sticking coefficient¹⁶ of carbon on the BN surface. However, it may also partly be the result of a different temperature on the growth surface of h-BN flake mounted on sapphire due to different heat coupling.

Our AFM measurements reveal the formation of an extended hexagonal moiré pattern of period ~ 14 nm when our MBE layers of graphene on h-BN flakes are grown under optimum conditions. We attribute this moiré pattern to the commensurate growth of crystalline graphene on h-BN.^{26,27} We have performed extensive studies on the dependence of the period of the moiré pattern on the parameters of the MBE growth, and these results will be published elsewhere.²⁸

The quantity of defects within the material grown on h-BN is lower than on sapphire, as confirmed by a strong decrease in I_D/I_G ratio in the Raman spectra (Fig. 9). The appearance of a shoulder on the D peak is attributed to the Raman peak of h-BN.²⁹

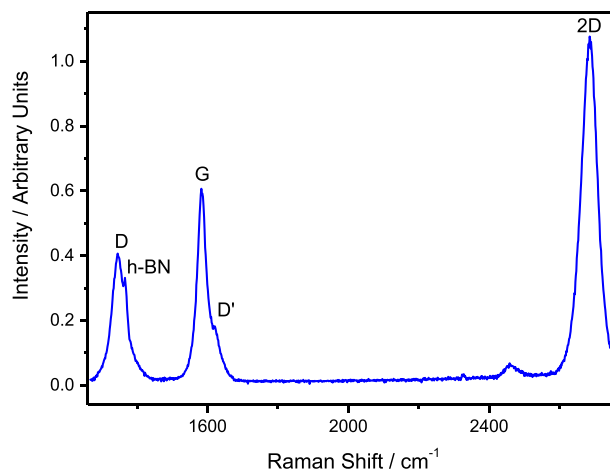


Fig. 9. (Color online) Raman spectrum of the material grown on an h-BN flake.

IV. SUMMARY AND CONCLUSIONS

In this work, we investigated growth by MBE of graphene layers at extremely high substrate temperatures. The standard dual chamber GENxplor was specially modified by Veeco to achieve growth temperatures of up to 1850 °C in ultrahigh vacuum conditions and is capable of growth on substrates of up to 3 in. in diameter. To calibrate the growth temperatures, we have formed graphene on the Si-face of SiC by heating wafers to temperatures above 1400 °C. We have used a carbon sublimation source to grow graphene on sapphire at substrate temperatures between 1000 and 1650 °C. The quality of the graphene layers is significantly improved by growing on h-BN substrates. Our AFM measurements reveal the formation of an extended hexagonal moiré pattern when our MBE layers of graphene on h-BN flakes are grown under optimum conditions.

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