Effect of substrate polishing on the growth of graphene on 3C-SiC(111)/Si(111) by high temperature annealing

B.Gupta^a, I. Di Bernardo^b, P. Mondelli^b, A. Della Pia^b, M.G. Betti^b, F. Iacopi^c, C. Mariani^b, N. Motta^{*a}

^a School of Chemistry, Physics and Mechanical Engineering and Institute for Future Environments, Queensland University of Technology, 2 George Street, Brisbane 4001, QLD, Australia

^b Dipartimento di Fisica, Università di Roma La Sapienza, Piazzale Aldo Moro 5, I-00185 Roma, Italy

^c Queensland Micro and Nanotechnology Centre, Griffith University, Nathan Campus 4111, QLD, Australia

Abstract

We analyse the effects of substrate polishing and of the epilayer thickness on the quality of graphene layers grown by high temperature annealing on 3C-SiC(111)/Si(111) by Scanning Tunneling Microscopy, X-ray Photoelectron Spectroscopy, Raman spectroscopy, Low Energy Electron Diffraction and High Resolution Angle Resolved Photoemission Spectroscopy. The results provide a comprehensive set of data confirming the superior quality of the graphene layers obtained on polished substrates, and the limitations of the growth obtained on unpolished surfaces.

*Corresponding author: email: n.motta@qut.edu.au

1. Introduction

The rush towards the realization of graphene based devices over the last decade is motivated by its wide range of exceptional physical and mechanical properties [1, 2]. Though the very

existence of a material as a free standing membrane fascinated the scientific community, its potential relies on its intrinsic structure: graphene is a network of sp^2 hybridized carbon atoms arranged in a hexagonal lattice with a double basis, and its specific chemical bonds make it impermeable and resistant to the introduction of atomic impurities, besides providing a great breaking strength. Moreover, its band structure exhibits a linear dispersion around the $\bar{K}$ point of the surface Brillouin Zone, which is responsible for its extremely high electron mobility ($2.5 \times 10^5 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ [3]) and makes it a potential platform for advanced electronics.

Since its first isolation in 2004 via micromechanical cleavage of Highly Oriented Pyrolytic Graphite (HOPG) [4], several methods have been proposed to synthesise graphene in sizeable quantities as its effective employment in industry depends on the capability to produce large area, high quality carbon layers on a technologically relevant substrate. Despite being a very simple and economic procedure, micromechanical cleavage does not offer exact control over the size and number of graphene patches, and requires transfer to the desired substrate [4]. Chemical Vapour Deposition (CVD) and Temperature Programmed Growth (TPG) can provide high quality and large area samples [5, 6], but do not overcome the problem of transferring graphene and purification processes via chemicals to remove catalysts and precursors might be necessary. Another proposed route is the unzipping of Carbon NanoTubes (CNTs) to form Graphene nanoribbons [7], but the usage of chemicals which could affect graphene properties is unavoidable in this case as well. Thermal desorption from SiC crystals is presently considered the most reliable technique to scale up the graphene growth to commercially available single-crystal wafers, allowing the development of an industrial scale process for graphene electronics [8].

Commercial SiC crystals are very expensive and limited in size, posing a limit to graphene/SiC industrial usage. This recently led to the realization of 3C-SiC thin films on

bulk Si(111) crystals [9-13]. The heteroepitaxy involves a series of technological challenges in terms of film homogeneity and integrity, nevertheless these composite substrates are much cheaper than bulk SiC and are also compatible with the current Si-based technology. It has been shown that graphitization on 3C-SiC(111)/Si(111) occurs in a similar fashion to graphene growth on SiC(0001), as these two substrates exhibit the same structure down to the fourth atomic layer, and the number of graphene layers can be easily tuned by changing the annealing temperature[14].

Despite the intense scientific activity around the epitaxial growth of graphene on 3C-SiC, a full analysis of the effects of substrate roughness and of epilayer thickness of 3C-SiC(111)/Si(111) on the quality of graphene layers is still missing. The advantage of polishing has been previously suggested [15, 16], but not evidenced as mandatory to achieve defectless, large area graphene layers. In this paper we analyse the effects of polishing and substrate roughness on 3C-SiC(111) epilayers on Si (111) before and after graphene growth, providing a clear indication of the ideal surface characteristics to obtain high quality graphene layers.

2. Experimental details:

Three different kinds of 3C-SiC(111) epilayers grown on Si(111) substrates were used to produce epitaxial graphene by annealing in UHV: 250nm thick unpolished (A), 1μm thick polished (B) and 1μm thick unpolished (C). Polishing was performed by a patented combination of chemical and mechanical steps (StepSiC®) at NOVASIC (France); the process removed 0.3 μm from the top layer with an error of ±10%. For this reason the 250nm sample could not be polished. Substrates are all P doped with resistivity 1-10 Ω-cm. Sample cleaning procedures and experimental details are described elsewhere [9]. The final annealing

temperature for epitaxial graphene growth on all the different substrates was $1250 \pm 15^\circ\text{C}$. The temperature on the 3C-SiC(111)/Si(111) substrate was measured by an optical pyrometer (IRCON Ultimex UX-20P). The 3C-SiC(111) surface was analyzed before and after annealing in situ by Scanning Tunnelling Microscopy (STM), X-ray Photoelectron Spectroscopy (XPS) and ex situ by Raman Spectroscopy [9].

Samples were taken out of the vacuum and subsequently analyzed at LoTUS laboratory, at the Physics Department – Università di Roma La Sapienza. Low-Energy Electron-Diffraction (LEED) data were taken after annealing at 550°C for two hours the samples introduced in UHV, obtaining good diffraction patterns, confirming the optimal recovering of graphene after air-transport by mild annealing [17]. Then High-Resolution Angle Resolved Photoelectron Spectroscopy (HR-ARPES) measurements of the samples have been performed by recording spectra at 77K, at a photon energy $h\nu=40.814\text{ eV}$ ($\text{HeII}\alpha$), with a high-resolution Scienta SES-200 hemispherical analyzer, by using a two-dimensional multichannel plate detector.

3. Characterisation:

3.2 Microscopic characterisation

(5×5) and $(1\times 1)\text{ }\mu\text{m}^2$ STM images of unpolished and polished 3C-SiC(111)/Si(111) substrates (defined as 3C-SiC(111) in the following) with different SiC epilayer thickness, after 10' annealing at 1250°C , are shown in Figure 1. Subtle changes cannot be appreciated with STM images at this scale, so we present only the images acquired after annealing. These images evidence the typical triangular features of the 3C-SiC(111) dislocation network, which is of the order of 200 nm for 250 nm thick samples (Figure 1a), and of 500 nm for $1\mu\text{m}$ thick samples (Figure 1b), indicating a release of the stress with the increase of the epilayer

thickness. Polishing clearly decreases the roughness and all these features look much smoother in Figure 1c, acquired on a polished $1\mu m$ thick epilayer.

Figure 1: Surface morphology obtained by STM after epitaxial graphene growth on unpolished (a,b,d,e) and polished samples (c,f). Top images: $(5\times 5)\mu m^2$; Bottom images: $(1\times 1)\mu m^2$. All images have been acquired at $V_b=2V$. (a,d) Sample A: unpolished $250nm$ thick 3C-SiC(111); $I=1nA$. (b,e) Sample B: unpolished $1\mu m$ thick 3C-SiC(111); $I=0.6nA$. (c,f) Sample C: polished $1\mu m$ thick 3C-SiC(111); (c): $I=0.6nA$ and (f): $I=730pA$.

Images 1d, 1e and 1f, $(1\times 1)\mu m^2$, provide further details of the surface morphology, evidencing more clearly the difference in the dislocation network between $250nm$ and $1\mu m$ samples (1d and 1e), and the atomic steps on the smooth surface obtained by polishing (1e). Some defects of 1 to $1.5nm$ height were spotted on the $250nm$ thick unpolished samples (Figure 1a), both before and after graphene growth. These defects were not found on the $1\mu m$ thick samples.

3.2.1 Roughness analysis

Table 1 and Fig 2 show the comparison of surface roughness of the samples before and after graphene growth on two unpolished (UP) and one polished (P) 3C-SiC(111) substrates. The roughness was calculated by analysing large scale $(5 \times 5) \mu m^2$ area STM images through the software Gwyddion [18].

Table 1: Roughness (nm) of $(5 \times 5) \mu m^2$ area of before and after graphene growth on 3C-SiC(111) samples measured by STM. The error is ± 0.5 nm

Sample Condition	Sample Type		
	(A) 3C SiC (111) unpolished 250nm thick	(B) 3C SiC (111) unpolished 1 μ m thick	(C) 3C SiC (111) Polished 1 μ m thick
As received	2.3 nm	5.1 nm	0.7 nm
After graphene growth	2.7 nm	5.1 nm	1.9 nm

The initial surface roughness of the 250 nm unpolished substrate is 2.3 ± 0.5 nm, while for the 1 μ m thick unpolished and polished substrates the initial surface roughness is 5.1 ± 0.5 and 0.7 ± 0.5 nm respectively (Figure 2). After graphene growth, the surface roughness changed to 2.7 ± 0.5 nm, 5.1 ± 0.5 nm, and 1.9 ± 0.5 nm respectively. It can be noticed that polishing reduces significantly the surface roughness.

Figure 2: Roughness analysis of three samples taken before and after annealing: A: 250 μm unpolished; B: 1 μm unpolished; C: 1 μm polished.

3.2.2 Atomic resolution imaging

Figure 3(a) shows the epitaxial graphene step formation on 250 nm UP substrate (Sample A).

Figure 3: $(30 \times 30) \text{ nm}^2$ area STM images of epitaxial graphene step formation after annealing at 1250 $^{\circ}\text{C}$. (a) Sample A - 250 nm unpolished (V_b : 1.8V; I : 600pA); (b) sample B - 1 μm unpolished (V_b : 1.65V; I : 600pA) and (c) sample C - 1 μm polished (V_b : 1.5V; I : 100pA).

Several discontinuous terraces along with a remarkable amount of clusters are present on the surface. The terrace width varies between 3 and 7 nm . Fig 3(b), acquired on 1 μm thick

unpolished 3C-SiC(111) (Sample B) shows a notable roughness increase. Figure 3(c) shows that the terraces obtained on the polished substrate (Sample C) are larger, with an average width of 10–40 nm. It shows a continuous system of terraces all over the surface with very little or no clusters on the top. Surface reconstruction can be visible by zooming on one of the terraces.

Fig. 4 STM high resolution images of the polished sample (Sample C). Images (a,b,c) are acquired after annealing at 1175 °C: (a) the $(6\sqrt{3}\times 6\sqrt{3}) R 30^\circ$ surface reconstruction is visible (Vb: 0.82V; I: 440pA); (b) further zoom of image (a): (Vb:100mV and I:320pA); (c) schematic superimposed to image (b) showing the $(6\sqrt{3}\times 6\sqrt{3}) R 30^\circ$ (red line) and the (6×6) (yellow line) cells and the arrangement of the surface atoms. Images (d,e,f) are acquired after annealing at 1250 °C: (d) graphene showing the characteristic Bernal stacking (bias: 0.53V; I: 470pA); (e) graphene (left part of the image) with Bernal stacking coexisting with monolayer graphene (right, see text) (bias: 0.53V; I: 470pA); (f) honeycomb structure of graphene (bias: 0.49V; I: 400pA) with periodicity of 0.246nm. The crystal structure is superimposed in blue.

In Fig 4 STM high-resolution images are presented for the polished sample annealed at 1175 °C (panels a,b,c) and at 1250 °C (panels d,e,f). Fig 4a reveals an atomic arrangement compatible with the $(6\sqrt{3}\times 6\sqrt{3}) R 30^\circ$ reconstruction in the lower part of the image. A precursor area where the superstructure is not yet developed can be observed in the top left corner of the image. A zoom of the bottom area (Figure 4b and 4c) reveals clearly the $(6\sqrt{3}\times 6\sqrt{3}) R 30^\circ$ reconstruction, containing 108 Si and 108 C atoms per SiC bilayer and

338 atoms in a graphene layer. The lattice parameter of the superstructure is $32 \text{ \AA}$. This large reconstruction is caused by the different lattice parameters of graphene ($2.46 \text{ \AA}$) and SiC substrate ($3.08 \text{ \AA}$) [19]. STM images of the $(6\sqrt{3} \times 6\sqrt{3})R 30^\circ$ phase can include a corrugation with a (6×6) periodicity [20, 21], but low bias STM images often do not allow identification of a periodic arrangement of the surface atoms for this superstructure. Under certain tip conditions, the true structure can indeed be not clear [20], however we remark that the full atomic arrangement of the structure is still unresolved, due to the fact that it is a transition phase and several atomic arrangements, depending on the number and species of adatoms present, are possible. This surface reconstruction is attributed to a C-rich phase but does not have any graphitic properties, as the adlayer has a strong interaction with the substrate, and so it is considered a buffer layer (B) or an interface layer (I) [20].

The typical atomic arrangement of the A-B stacked graphene (Bernal stacking) is visible after annealing at 1250°C (Fig 4d-e); we notice that the honeycomb graphene structure is visible to the right of Fig 4e, suggesting a detaching of the first layer from the second [22]. Full atomic resolution of graphene (Fig 4f) showing a clear honeycomb structure with a periodicity of 0.246 nm was obtained also in several other areas of the sample [9, 23].

3.3 LEED

In order to ascertain the long-range crystallinity of our graphene we analyzed sample C ($1 \mu\text{m}$ polished 3C-SiC(111)) by LEED. In Figure 5a we can distinguish two hexagonal patterns with different lattice constants, rotated by 30° one respect to the other. As reciprocal lattice constant are inversely proportional to direct space ones, we attribute the innermost hexagon to SiC (1×1) reconstruction and the bigger one to graphene (Fig. 5a, bottom panel). This attribution is supported by the ratio of the hexagon sides: in real space,

$$\frac{a_{3C-SiC(111)}}{a_G} = \frac{3.08\text{\AA}}{2.46\text{\AA}} = 1.25$$

matching nicely within 2% the experimental result:

$$\frac{b_G}{b_{3C-SiC(111)}} = 1.23$$

Figure 5: LEED patterns of graphene grown on the polished sample annealed at 1225°C, collected at different primary beam energies. As a guidance for the eye, the geometrical patterns are superimposed to the data in the bottom figures: blue lines indicate the diffraction pattern of graphene, red dashed lines the pattern of the SiC and red solid lines join the diffraction spots related to the $(6\sqrt{3}\times 6\sqrt{3})R\ 30^\circ$ reconstruction.

In Fig. 5b, obtained at lower primary beam energy, the most intense spots can be ascribed to SiC (1x1) reconstruction, each of them surrounded by six fainter spots. These smaller hexagons, together with the hexagonally-arranged triangular groups of spots, are the result of double diffraction due to the interfacial layer. Diffraction spots are quite broad, and since thermal noise was avoided by keeping the sample at liquid nitrogen temperature, broadening can be attributed to limited domain size. There is no evidence of different rotational domains, but a minor component of defected sites or slightly tilted terraces cannot be excluded.

3.2 XPS and Raman Studies:

Figure 6 shows the high-resolution *C1s* XPS (6 a, b, c) and Raman spectra (Figure 6 d, e, f) of samples A (250nm unpolished), B (1 μ m unpolished) and C (1 μ m polished) after annealing at 1250 $^{\circ}$ C. The *C1s* XPS peak consists of three different carbon peaks: C in SiC (SiC~283eV), graphitic carbon (G~284.6eV) and C in the interface layer (I~285.8eV). Only a slight variation is found on the XPS intensity of the three carbon components of the three samples. The average number of graphene layers has been determined by using the intensities ratio of SiC and graphene peak as described elsewhere [9, 24] and it is equal to 4.5 $\pm$ 0.5 for the unpolished samples and to 4.2 $\pm$ 0.5 in the polished sample.

The Raman spectrum acquired on the three samples A, B and C after graphene growth shows three signature bands which can be attributed to graphene: D (1362 cm^{-1}), G (1607 cm^{-1}) and 2D (2719 cm^{-1}). In addition, a D+G band is also visible for all the samples. Weak peaks around the main G peak (at about 1510 and 1720 cm^{-1}) are visible [25, 26] on sample B, and to a lesser extent on Sample C, but not on sample A (Figure 6d). They are attributed to SiC [26, 27], and are visible only for the thick epilayer samples (B and C) where enough signal is generated at 1520 cm^{-1} from the overtone of the TO (X) phonon and at 1713 cm^{-1} from the combination of optical phonons near the M point [26]. We observe that these peaks are more

evident in sample B due to the high surface roughness, which may cause inhomogeneity in the graphene coverage. The data (Table 2) show that the position of D, G and 2D bands is a function of the type of substrate we used to grow graphene. The G band position for unpolished samples is 1607 and 1613 cm^{-1} respectively, while for polished samples it is 1592 cm^{-1} , with a peak shift towards lower wavenumbers for smoother surfaces. This shift has been attributed to local electron and/or hole doping, however in our case this is unlikely, as doping is the same for all our samples [26] and it could be attributed instead to the compressive strain [28] connected to roughness, as it has the same behaviour found in Fig.2.

Figure 6: (a, b, c) XPS spectra of epitaxial graphene grown on unpolished and polished substrates after graphene growth at 1250°C. The peaks related to SiC, graphitic carbon (G) and Interface layers (I) are represented by purple, green, and brown, respectively. (d, e, f) Raman Spectrum of unpolished and polished substrates annealed at 1250°C.

The D band is visible in all samples, both polished and unpolished, confirming the presence of defects in our graphene, as evidenced also by STM analysis. The ratio of the intensity of D and G bands $I(G)/I(D)$ indicates the size of the graphene crystals. It has been demonstrated [29] that $I(G)/I(D)$ is proportional to the crystal size L_a : $I(G)/I(D) = L_a / C(\lambda)$, where the coefficient $C(514 \text{ nm}) \sim 4.4 \text{ nm}$. The lowest $I(G)/I(D)$ ratio was observed for sample B, 1µm

UP, confirming that the roughness found in STM limits the graphene crystal size (Table 2). The largest crystal size ($\sim 17\text{nm}$) is obtained as expected for sample C. The line shape and the full width at half maximum (FWHM) of 2D peak also are key factors in determining the number of graphene layers. For all of our samples only a single Lorentzian 2D peak shape has been observed, with a little difference in wavenumber and FWHM [25]. The FWHM of 2D peak for graphene grown on polished substrates (80 cm^{-1}) would indicate a good quality of the sample suggesting the presence of 4-5 monolayers of graphene, as found by Lee et al. [28] on 6H and 4H SiC (0001) substrates, matching our XPS peak analysis. On the other hand, the FWHM detected on unpolished substrates, 102 and 129 cm^{-1} , for 250 nm and $1\mu\text{m}$ SiC respectively, is well beyond the scale of ref [28], suggesting an effect connected to the roughness of the substrate.

In summary, from the shape and intensity of the 2D peak (see table 2) we deduce that the graphene flake average size is much smaller for the unpolished samples (8 nm) than for the polished ones (17 nm). As the XPS indicates a very similar average thickness, this implies a larger uncovered SiC surface in unpolished sample because of the inhomogeneous surface revealed by STM, resulting in the more intense substrate peaks found at 1520 cm^{-1} and 1713 cm^{-1} . We observe as well that the wavenumber of the 2D peak is lower for sample C compared to the other samples [28, 30], as well as the intensity ratio I_G/I_{2D} [31], which might indicate a slightly lower number of graphene layers in the polished sample as suggested by the XPS.

Table 2: Raman data of graphene grown on unpolished and polished substrate

Sample type	D band			G band			2D band			I_G/I_D	I_a (nm) $\approx 2.4 \times 10^{-10} (\lambda_i)^4 (I_G/I_D)$	I_G/I_{2D}
	Position	Area	FWHM	Position	Area	FWHM	Position	Area	FWHM			
A Unpolished (250nm) 1250°C	1361.7	$0.38 \cdot 10^6$	60.4	1606.7	$0.21 \cdot 10^6$	70.1	2719.4	$0.05 \cdot 10^6$	102.2	0.54	10.3	3.9
B Unpolished (1μm) 1250°C	1364.3	$7.17 \cdot 10^6$	73.3	1612.2	$2.94 \cdot 10^6$	56.2	2718.3	$0.56 \cdot 10^6$	129	0.41	7.8	5.2
C Polished (1μm) 1250°C	1358.8	$1.21 \cdot 10^6$	53.1	1592.0	$1.08 \cdot 10^6$	66.6	2704.8	$0.93 \cdot 10^6$	80.9	0.89	17.1	1.2

3.3 Electronic valence band structure of graphene/SiC/Si

High-resolution Angle-Resolved Photoelectron Spectroscopy measurements reported in Figure 7, were performed at 77K on graphene grown at 1225 °C, on a substrate equal to that of sample C (XPS of $CI1s$ reported in SI shows a thickness between two and three graphene monolayers). The data are recorded slightly off the $\bar{\Gamma} - \bar{K} - \bar{M}$ direction due to constraints of the sample holder. The band structure shows an overall agreement over the whole Brillouin Zone with the expected band structure of graphene [32], and in particular the π band presents the linear dispersion around the $\bar{K}$ point which is typical of massless Dirac fermions. The bottom of the π band at $\bar{\Gamma}$ is located at 7.99 eV below the Fermi level, and a faint σ band is visible as well.

Figure 7: Electronic valence band structure of graphene grown on polished 3C-SiC(111) acquired by ARPES at 77 K (Sample C).

The Energy Distribution Curves (EDCs) relative to the Dirac cone apex are reported in Figure 8a. The π -band peak of each curve was fitted with a Gaussian function, and the centre of each

Gaussian is indicated by a red star in Figure 8a. The curve connecting the position of the Gaussian maxima as a function of the parallel momentum was in turn fitted with an hyperbola, which is the result of the intersection of a cone with a plane non containing a diameter of the cone, and therefore it is the curve we expect to see in the place of the Dirac cone. By fixing the known tilt angle with respect to the $\bar{\Gamma} - \bar{K}$ direction (1.7° , obtained by LEED measurements) it was possible to extrapolate the exact position of the vertex of the Dirac cone E_D . We find it to be located 0.29 eV below the Fermi level. It is worth noting that the shape and width of the π bands are very sensitive to the number of graphene layers, as clearly reported in recent ARPES [33] and spatial-resolved nano-ARPES [34] of multilayer graphene on bulk SiC. Thus the linearity and the width of the Dirac cone of the present ARPES data suggest the coexistence of grains of few layers graphene with different thickness and without rotational disorder. ARPES measurements have also been taken on graphene grown on a thinner unpolished 3C-SiC(111) layer (250 nm) (not shown here). Despite the very low signal intensity due to the roughness of the unpolished surface, the main features of the π band are in general agreement with the present data.

Figure 8: a) Energy Distribution Curves of the valence band around the apex of the Dirac cone. The red star on each curve indicates the position of the center of the Gaussian peak used to fit the peak of the p band. b) Sketch of the graphene Brillouin zone. Lines of different colors correspond to cuts along different azimuthal directions. c) Close up on the Dirac cone showing a small shift due to the rotation with respect to the $\bar{\Gamma} - \bar{K}$ direction of our sample. The yellow line is a guide for the eye to locate the Dirac cone.

4. Discussion:

An accurate analysis of the results on unpolished and polished 3C-SiC(111) substrates leads to the choice of the best condition for epitaxial graphene growth in UHV. Thicker polished substrates produce better results in terms of terrace widths and reduced roughness leading to an improved graphene quality. The roughness of samples as received drops substantially with polishing, while after graphene growth a small increase of the surface roughness was found for all substrates. Results obtained from XPS analysis on all the three samples would indicate that the number of epitaxial graphene layer is substantially unaffected by the roughness of the substrate, with a very small decrease in the polished sample compared to the unpolished ones.

The Raman data show clearly that the roughness of the substrate affects the size of graphene grains, with larger grains found in the polished sample (Table 2). ARPES, acquired on samples grown at a slightly lower T (1225 °C), suggests an average number of layers between 2 and 3, in line with previous results, and in perfect agreement with our XPS considering the temperature difference [9, 24, 35].

Microscopy measurements such as SEM and AFM (reported in SI), performed on the polished sample, reveal that the surface of the silicon carbide interlayer is crossed by a webcomb of valleys homogeneously arranged throughout the sample. It was previously claimed [21] that cracks may be induced in the SiC film because of the strain between Si and

SiC during the cooling process, but our AFM measurements report on shallow, wide valleys, allowing us to rule out any possibility of cracks reaching the Si substrate.

A study of the diffraction pattern points out the existence of a single domain over the whole sample, as we always observe a single series of spots related to SiC and graphene as well, while the crystallinity of the sample is granted by the presence of a well defined band structure. The electronic valence band state dispersion reveals a long-range ordered sample, even though some defects are present. In spite of the slight band broadening around the $\bar{\Gamma}$ point, the shape and the linearity of the Dirac cone suggest the presence of graphene grains with a defined symmetry and periodicity. We find the doping to be consistent with previous literature data of three layer graphene grown either on SiC thin film [21] and on bulk SiC crystals [22, 23]. This shift of the charge neutrality point is attributed to the charge transfer at the graphene-SiC interface.

4. Conclusion:

In conclusion, we found that polishing SiC substrate is the best choice to improve the quality and size of the graphene layers grown by annealing in Ultra High Vacuum. After polishing 1 μm 3C-SiC(111)/Si(111) substrates, surface roughness before growth drops from $\sim 5\text{nm}$ to 0.7nm . This helps in creating large terraces, where graphene formation is significantly improved with larger crystals and negligible amount of cluster formation on the surface as evidenced by STM.

Raman measurements and XPS results confirm that polished substrates produce bigger graphene crystals, providing an evaluation of the thickness and quality of graphene over the entire surface. HR-ARPES measurements, showing the typical electronic band state dispersion of graphene, confirm that large graphene grains belonging to a single domain grow

on polished substrates. In particular, we determine a Fermi velocity at the $\bar{K}$ point of the Brillouin zone close to the value observed for quasi-free-standing graphene on Ir [36], and from the Dirac cone we find only a slight n-type doping, much less than for graphene grown on bulk SiC.

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