

Growth and Electronic Structure of Epitaxial Graphene on Silicon Carbide

Aaron Bostwick¹, Eli Rotenberg¹, Jessica McChesney^{1,2}, Taisuke Ohta^{1,2},
Thomas Seyller³, and Karsten Horn²

¹Advanced Light Source, Lawrence Berkeley Laboratory, 1 Cyclotron Road, Berkeley California 94720

²Fritz Haber Institute of the Max Planck Society, Faradayweg 4 – 6, D-4195 Berlin Germany

³Lehrstuhl für Technische Physik, Institut für Physik der Kondensierten Materie, Universität Erlangen, Germany

1. Introduction

Graphene, the two-dimensional lattice of hexagonally coordinated carbon atoms, has many fascinating properties that have put it into the limelight of current solid state physics and materials science: it is the thinnest known material in the universe and the strongest one ever measured. Its charge carriers are unique in that they possess zero effective mass, exhibit a giant intrinsic mobility, and can travel for micrometers without scattering at room temperature. Graphene can sustain a current density six orders of magnitude higher than that of copper, and shows record thermal conductivity and stiffness, to name just a few of its intriguing properties. Electron transport in graphene is described by a Dirac-like equation, which allows the investigation of relativistic quantum phenomena in a benchtop experiment [1]. Moreover, it offers the prospect of novel electronic devices, such as field effect transistors in which the spin orientation of the electron is used to provide switching action, its use in sensing devices and much more. The talk will cover several basic science-related issues, as well as aspects of graphene research important for future applications. One most important aspect of research into graphene's properties is that as a true 2D material, being only one atom thick ("the thinnest object conceivable" [2]) it is accessible to the vast repertoire of surface-sensitive methods such as scanning probe techniques [3,4], photoelectron spectroscopy [5,6], various optical techniques [7] etc.

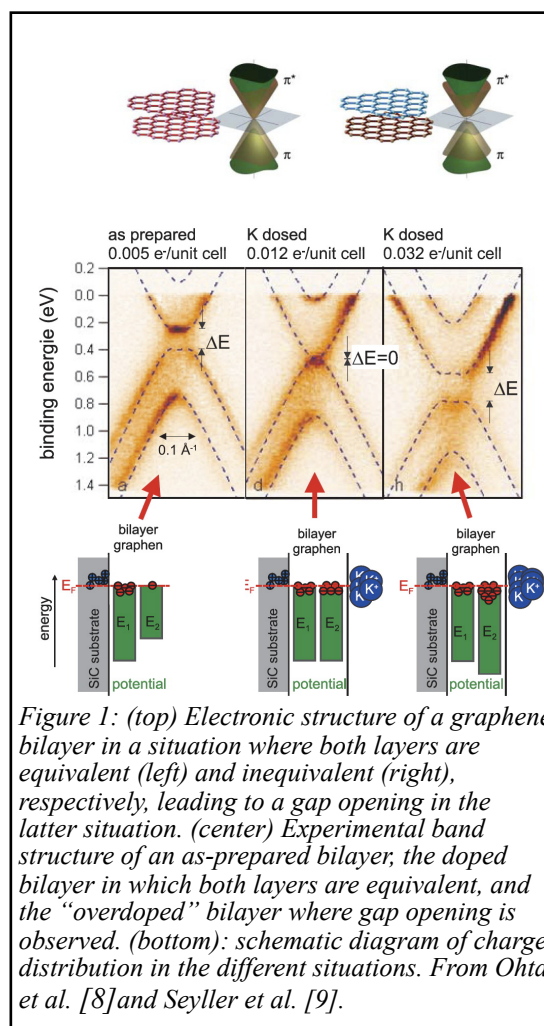
2. Electronic Structure

The band structure of graphene is unique in that the Fermi surface consists of a point at which the valence and conduction bands meet, such that its density of states is zero at the Fermi level – graphene is thus a semimetal. Its band structure can be modeled in a simple tight binding picture, and since it is possible to prepare single as well as multilayers of graphene ("few layer graphene"), the evolution of the electronic structure from a purely 2D material towards the formation of a 3D solid, i.e. the onset of interaction between the layers, can be studied in unprecedented detail. The bilayer is a special case since it marks the transition from the massless charge carriers mentioned above, to "massive" ones. Photoemission has played an important role in elucidating the emergence of a band gap at the so-called Dirac point; this can emerge in the bilayer if the layers are rendered inequivalent through the action of the substrate, through doping or an electric field; this situation is schematically depicted at the top of Figure 1. The center panel shows the experimental band structure as observed using angle-resolved

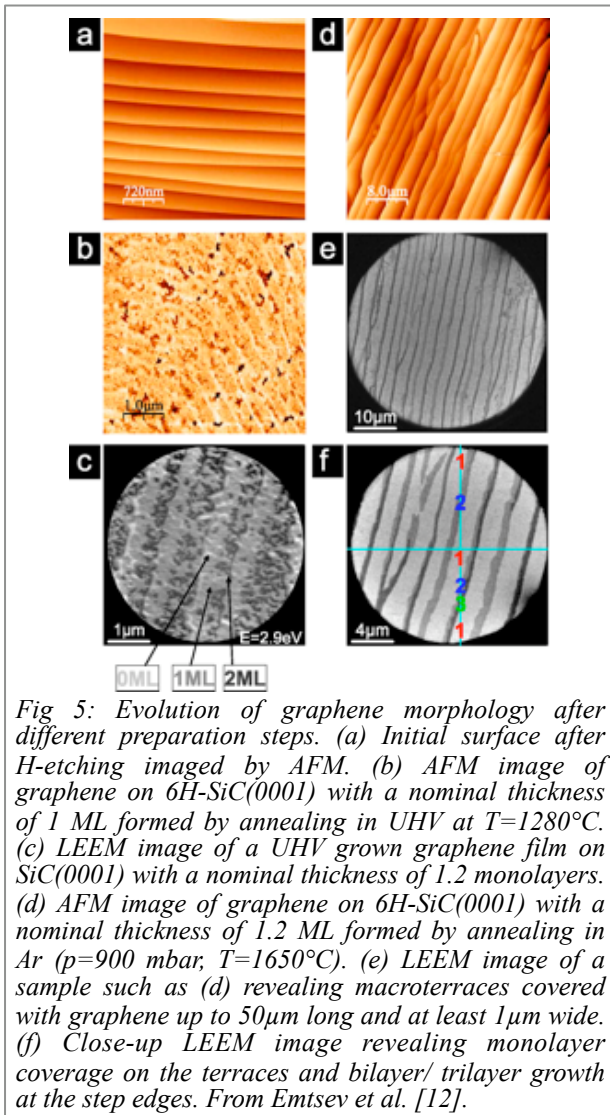
photoemission [8]. The as-prepared bilayer is already doped through charge transfer from the substrate, and by additional doping through the adsorption of potassium atoms which transfer their charge to the bilayer, an situation where the two layers are equivalent is achieved (center panel). Overdoping of the outside layer causes gap opening again; the schematic charge distribution is shown in the bottom of Figure 1 [9]. Bilayers are of particular interest since the emergence of a gap is crucial for using graphene in switching devices. The single layer offers the chance to study the influence of many-body interactions, such as electron-phonon and electron-electron processes which leave a particularly clear signature in the photoelectron or scanning tunneling spectra.

3. Graphene preparation

A crucial issue for any large scale usage of graphene has been to find a viable method to prepare wafer-size sam-



ples which can be used with (maybe adapted) procedures and methods in the semiconductor industry; the micro-mechanical cleaving (also known as adhesive tape) method, the basis of much of the studies into the intriguing physics of graphene, will most likely not be a route to mass production [1]. Graphitic layers on metal and semiconductor surfaces had been observed and investigated decades ago, and the growth of graphene on silicon carbide was found to result in useful samples [10,11], and has been used in a number of studies. In the latter system, two types of surfaces may be used, the silicon-terminated SiC(0001) as well as the carbon-terminated SiC(000-1) face. The former gives single and multilayer graphene arranged in a fashion similar to that in graphite,



while using the latter results in multilayers which, on account of the fact that consecutive layers are rotated against each other behave like ensembles of single layers. Graphene prepared on the silicon-terminated (0001) surface of silicon carbide offers a means for preparing the single- and few layer samples mentioned above. The preparation involves the preparation of a carbon buffer layer with a $6\sqrt{3} \times 6\sqrt{3}$ surface periodicity which saturates the SiC dangling bonds, followed by the formation of one or more graphene layers; this proceeds through high temperature annealing which leads to depletion of

silicon in the top layers. As shown in Figure 2, this procedure may lead to a considerable surface roughness (Figure 1b) with small patches of graphene on the order of tens of nanometers, and a distribution of thicknesses accompanied by deep holes in the surface. Clearly such surfaces are not very useful for transport studies and large scale applications; the problem is that the surface is not close to equilibrium in the annealing process carried out in ultrahigh vacuum. As shown in panels d–f, an alternative method offers vastly superior surface quality and homogeneity [12]. This involves annealing a silicon carbide surface under atmospheric pressures of argon, which creates a situation closer to equilibrium such that smooth surface morphologies can be achieved.

4. Outlook

Although an immense research effort is currently under way, with about 1500 papers on graphene published in 2008, many of the properties important for a basic science understanding and future applications need to be better understood, and roads towards large scale growth and patterning need to be explored. However, rapid progress has been made and one can reasonably expect that many of the issues will be addressed and resolved in the near future. Graphene, on account of its unique electronic structure, is a rich area of research in a novel field of solid state physics, and we can expect to identify more exciting aspects of this material, e.g. through functionalization. The talk will address the potential of photoelectron spectroscopy and other surface-related techniques to help understand the properties of this fascinating material.

References

1. A. K. Geim, Science **324**, 1530-4 (2009).
2. J. C. Meyer, A. K. Geim, M. I. Katsnelson, K. S. Novoselov, T. J. Booth and S. Roth, Nature **446**, 60-3 (2007).
3. G. M. Rutter, J. N. Crain, N. P. Guisinger, T. Li, P. N. First and J. A. Stroscio, Science **317**, 219-22 (2007).
4. T. Filleter, J. L. McChesney, A. Bostwick, E. Rotenberg, K. V. Emtsev, T. Seyller, K. Horn and R. Bennewitz, Phys. Rev. Lett **102**, (2009).
5. A. Bostwick, T. Ohta, J. L. McChesney, T. Seyller, K. Horn and E. Rotenberg, Eur.Phys. J. Special Top. **148**, 5-13 (2007).
6. W. A. de Heer et al., Sol. Stat. Commun. **143**, 92-100 (2007).
7. P. Blake, E. W. Hill, A. H. C. Neto, K. S. Novoselov, D. Jiang, R. Yang, T. J. Booth and A. K. Geim, Appl. Phys. Lett. **91**, (2007).
8. T. Ohta, A. Bostwick, T. Seyller, K. Horn and E. Rotenberg, Science **313**, 951-4 (2006).
9. Seyller, Th, Bostwick, A, Emtsev, K V, Horn, K, Ley, L, McChesney, J.L., Riley, J D, Rotenberg, E and Speck, F, phys.stat. sol. **245**, 1436 (2008).
10. I. Forbeaux, J. M. Themlin and J. M. Debever, Phys. Rev. B **58**, 16396-406 (1998).
11. C. Berger, Z. Song, T. Li, X. Li, A. Y. Ogbazghi and R. Feng, J.Phys.Chem. B **108**, 19912 (2004).
12. K.V. Emtsev, et al., Nat. Mater. **8**, 203 (2009).