

## O 41: Graphene: Growth, Substrate Interaction, Intercalation, and Doping

Time: Wednesday 10:30–12:30

Location: S052

O 41.1 Wed 10:30 S052

**Using polyaromatic hydrocarbons for graphene growth** — ●MATTHEW A. STOODLEY<sup>1,2</sup>, BENEDIKT P. KLEIN<sup>1,2</sup>, LUKE A. ROCHFORD<sup>2</sup>, MATTHEW EDMONDSON<sup>3</sup>, MARC WALKER<sup>2</sup>, TIEN-LIN LEE<sup>1</sup>, ALEXANDER SAYWELL<sup>3</sup>, REINHARD J. MAURER<sup>2</sup>, and DAVID A. DUNCAN<sup>1</sup> — <sup>1</sup>Diamond Light Source, Didcot, United Kingdom — <sup>2</sup>University of Warwick, Coventry, United Kingdom — <sup>3</sup>University of Nottingham, Nottingham, United Kingdom

Graphene, due to its widespread potential applications, has been one of the most studied materials this century. The most common approach to produce high quality graphene is through epitaxial growth utilising chemical vapour deposition (CVD) on copper surfaces. This method exploits the catalytic activity of copper to form highly crystalline, large area graphene. Traditionally, precursors used to grow graphene on Cu(111) require high pressures and elevated substrate temperatures, however, many cutting edge characterisation techniques cannot be used at such conditions. In this work, polyaromatic hydrocarbons (PAH) are used to grow high quality graphene on Cu(111) via CVD at relatively low surface temperatures. Furthermore, the low vapour pressure of the PAH permits us to synthesise graphene films in ultra-high vacuum conditions. We present a characterization of the grown graphene, by utilising a wide variety of techniques including Scanning Tunnelling Microscopy, X-ray photoelectron spectroscopy, normal incidence x-ray standing waves, and near edge X-ray adsorption fine structure.

O 41.2 Wed 10:45 S052

**Stone-Wales defect: molecular model system reveals increased interaction with Cu(111) surface** — ●BENEDIKT P. KLEIN<sup>1,2,3</sup>, ALEXANDER IHLE<sup>4</sup>, STEFAN R. KACHEL<sup>1</sup>, LUKAS RUPPENTHAL<sup>1</sup>, SAMUEL J. HALL<sup>2</sup>, DANIEL EBELING<sup>4</sup>, RALF TONNER-ZECH<sup>1</sup>, REINHARD J. MAURER<sup>2</sup>, ANDRE SCHIRMEISEN<sup>4</sup>, and J. MICHAEL GOTTFRIED<sup>1</sup> — <sup>1</sup>Philipps-Universität Marburg, Germany — <sup>2</sup>University of Warwick, Coventry, UK — <sup>3</sup>Diamond Light Source, Didcot, UK — <sup>4</sup>Justus-Liebig-Universität Gießen, Germany

The properties of the graphene/metal interface are crucially influenced by the enhanced interaction of defects in the graphene layer with the metal substrate. However, due to experimental and computational constraints, it is difficult to investigate this interaction directly. We combine calculations with experimental analysis of large organic molecules adsorbed on metal surfaces as molecular model systems. We chose two model molecules, azulene and pyrene, which have the same aromatic topology as the prototypical Stone-Wales defect and the ideal graphene lattice, respectively. When adsorbed on the Cu(111) surface, we could show using TPD that the model defect binds much stronger to the surface. nc-AFM and NIXSW results furthermore prove a reduction in adsorption height while XPS, UPS, and NEXAFS show an increased electronic hybridisation between molecule and surface. DFT results agree with these findings and show a localized interaction with the metal surface, both for the molecular model systems and the defect embedded into the graphene layer.

O 41.3 Wed 11:00 S052

**Determining the stability and catalytic formation of graphene on liquid Cu using machine-learning potentials** — ●HAO GAO<sup>1</sup>, VALENTINA BELOVA<sup>2</sup>, MACIEJ JANKOWSKI<sup>2</sup>, HENDRIK H. HEENEN<sup>1</sup>, GILLES RENAUD<sup>2</sup>, and KARSTEN REUTER<sup>1</sup> — <sup>1</sup>Fritz-Haber-Institut der MPG, Berlin, Germany — <sup>2</sup>ESRF, Grenoble, France

The recently discovered rapid, high-quality synthesis of graphene (Gr) on liquid Cu catalysts is microscopically still poorly understood. This is due to the difficult characterization of the Cu liquid surface. Especially in atomistic simulations, the large length and time scales necessary to reliably emulate the temporal evolution of the liquid are a major challenge. Corresponding molecular dynamics simulations require large simulation cells and need to span well into the nanosecond regime – an endeavor presently intractable via first-principles methods. In this work we use computationally efficient machine-learning potentials (MLPs) trained to density-functional theory (DFT) data in order to extrapolate the first-principles predictive power to the required scales. Detailed benchmarking confirms that our MLP captures the involved physics well, accurately reproducing the experimentally determined Gr adsorption height. We apply the MLP to further study the catalytic mechanism of Gr synthesis in order to rationalize distinct

reaction kinetics found experimentally. Our work draws a path for the use of reliably trained MLPs as a multiscale modeling technique to explore previously uncharted computational problems. In that we provide new insight into the domain of liquid metal catalysts which generally lack atomic-scale understanding.

O 41.4 Wed 11:15 S052

**Properties of epitaxial graphene on various SiC terminations and polytypes investigated by low-energy electron microscopy** — ●PHILIP SCHÄDLICH<sup>1,2</sup>, DAVOOD MOMENI PAKDEHI<sup>3</sup>, FLORIAN SPECK<sup>1,2</sup>, KLAUS PIERZ<sup>3</sup>, and THOMAS SEYLLER<sup>1,2</sup> — <sup>1</sup>Technische Universität Chemnitz, Chemnitz, Germany — <sup>2</sup>Center for Materials, Architectures and Integration of Nanomembranes (MAIN), Chemnitz, Germany — <sup>3</sup>Physikalisch-Technische Bundesanstalt, Braunschweig, Germany

The epitaxial growth of graphene on SiC has been improved from UHV growth to ambient pressure synthesis [1], and recently to the polymer-assisted sublimation growth (PASG) [2]. It results in an enhanced nucleation of the buffer layer and suppressed step bunching, which usually occurs upon the graphene formation by sublimation. Thus, PASG leads to homogeneous monolayer graphene with minimum step size equivalent to one or two SiC-bilayers. As a result of the latter, graphene on various surface terminations of the SiC substrate can now be systematically studied.

We utilize low-energy electron microscopy (LEEM) to identify the underlying substrate structure and to elucidate its delicate impact on the characteristic I(V) curves for different SiC polytypes. In addition, we investigate how the decoupling of the buffer layer from the substrate by hydrogen intercalation [3] influences the aforementioned effects.

[1] K. V. Emtsev et al., Nat. Mat. 8, 203 (2009). [2] M. Kruskopf et al., 2D Mater. 3 (4), 041002 (2016). [3] C. Riedl et al., PRL 103, 246804 (2009).

O 41.5 Wed 11:30 S052

**Quasiparticle Interference Effects Revealed in Potassium-Intercalated Epitaxial Monolayer Graphene** — TOBIAS HUEMPFNER, ●FELIX OTTO, ROMAN FORCKER, and TORSTEN FRITZ — Institute of Solid State Physics, Friedrich Schiller University Jena, Helmholtzweg 5, 07743 Jena, Germany

Epitaxial graphene is known for its fascinating physical properties, such as a linear dispersion at the K point, an anomalous quantum Hall effect and even superconductivity. These effects are based on the unique electronic structure of graphene, which can be tuned, e.g., by intercalation of metals. In this study, we scrutinize samples of epitaxial monolayer graphene (EMLG) on SiC(0001) intercalated with potassium by means of local and area-averaging experimental methods at low temperatures. For the highest accessible K concentration we find a highly ordered (2 × 2) superstructure that the K atoms form below the uppermost graphene layer. Further, we observe that the K atoms also reside below the buffer layer of the EMLG on SiC(0001) sample that is located between the quasi-freestanding graphene sheet and the SiC substrate. This causes an effective decoupling of the buffer layer and a transformation to K-intercalated epitaxial bilayer graphene (EBLG). This configuration promotes strong *n*-doping of the graphene sheets, where besides a rigid shift of the Dirac bands to higher binding energies also filling of two parabolic interlayer bands is observed.

O 41.6 Wed 11:45 S052

**Structural and electronic properties of a van der Waals heterostructure of two-dimensional Pb and epitaxial graphene** — ●BHARTI MATTA, PHILIPP ROSENZWEIG, OLAF BOLKENBAAS, KATHRIN KÜSTER, and ULRICH STARKE — Max-Planck-Institut für Festkörperforschung, Heisenbergstraße 1, 70569 Stuttgart, Germany

Intercalation is an established technique for manipulating the electronic properties of epitaxial graphene. Moreover, it is a way to confine otherwise unstable two-dimensional (2D) layers of elements, leading to unique physical properties compared to their bulk counterparts due to quantum confinement. In this work, we show uniformly Pb-intercalated quasi-freestanding monolayer graphene on SiC, which turns out to be essentially charge neutral. Additional bands – some crossing the Fermi level – can be clearly discerned, demonstrating the metallic character of 2D Pb sandwiched within the graphene/SiC het-

erointerface. The band velocity of the graphene Dirac cone changes at binding energies of around 0.6 eV and 1.1 eV, which points towards hybridization with these Pb bands. Low-energy electron diffraction reveals a  $10\times 10$  Moiré periodicity with respect to graphene. This is consistent with a triangular lattice of intercalated Pb that is strained by less than 2% relative to bulk-truncated Pb(111) and of which a  $(\sqrt{3}\times\sqrt{3})R30^\circ$  supercell matches  $2\times 2$ -SiC. However, finding direct signatures of the corresponding periodicities in the available band structure data is challenging. Our experimental results will provide a solid ground for further theoretical assessment of this system and better understanding of its properties.

O 41.7 Wed 12:00 S052

**Surface Transport Properties of Pb- and Sn-intercalated Graphene** — •M. GRUSCHWITZ, C. GHOSAL, Z. MAMIYEV, J. KOCH, S. WOLFF, T. SEYLLER, and C. TEGENKAMP — Institut für Physik, Technische Universität Chemnitz, Chemnitz, Germany

Intercalation experiments on epitaxial graphene are attracting a lot of attention at present as a tool to boost further the electronic properties of 2D graphene. In this work we studied the intercalation of Pb using buffer layers on 6H-SiC(0001). Recent DFT calculations suggest Pb atoms to act as electron donors thus allowing the doping level of the quasi-freestanding graphene layer to be tuned by the amount of intercalated material [1].

In this work we present results about the large-area intercalation of Pb and Sn, investigated by means of electron diffraction, scanning tunneling microscopy, photoelectron spectroscopy and in-situ surface transport. The intercalation of Pb and Sn results in formation of almost charge neutral graphene. In case of Pb, the intercalated layer consists of 2 ML and shows a strong structural corrugation. The epitaxial heterostructure provides an extremely high conductivity of  $\sigma = 100$  mS/ $\square$ . However, below 70 K we found a metal-insulator transition which we assign to the formation of minigaps in epitaxial

graphene, possibly induced by a static distortion of graphene following the corrugation of the interface layer [2]. Sn intercalation does not cause such a transition while yielding a conductivity of  $\sigma = 2.5$  mS/ $\square$ .

[1] J. Wang et al., PRB 103, 085403 (2021); [2] M. Gruschwitz et al., Materials 14, 7706 (2021)

O 41.8 Wed 12:15 S052

**Electron correlation effects in highly-doped single-layer graphene** — •VIVIEN ENENKEL<sup>1</sup>, PHILIPP ROSENZWEIG<sup>2</sup>, HRAG KARAKACHIAN<sup>2</sup>, FABIAN PASCHKE<sup>1</sup>, ULRICH STARKE<sup>2</sup>, and MIKHAIL FONIN<sup>1</sup> — <sup>1</sup>Fachbereich Physik, Universität Konstanz, 78457 Konstanz, Germany — <sup>2</sup>Max-Planck-Institut für Festkörperforschung, Heisenbergstraße 1, 70569 Stuttgart, Germany

Electronic correlations in graphene are expected to be strongly enhanced when there is a very high density of states at the Fermi level, giving rise to many-body states such as superconductivity or charge density waves (CDW) [1]. While the latter have been reported for example in Ca-intercalated bilayer graphene [2,3], in case of epitaxial monolayer graphene no direct evidence of correlated electronic ground states has yet been reported. We investigate heavily n-doped monolayer graphene on SiC(0001), obtained by Yb intercalation underneath zero-layer graphene. Here, a van Hove singularity (VHS) is pushed to the Fermi level, giving rise to an extended VHS, effectively pinning an almost non-dispersive flat band at EF [4]. Low-temperature STM reveals several distinct structures of Yb-intercalated graphene, which we attribute to differing arrangements of the Yb at the interface.  $dI/dU$  spectra show a pronounced gap feature centered at EF, whose response to field and temperature variations allows the interpretation of the feature as a CDW state. [1] M. L. Kiesel et al., Phys. Rev. B 86, 020507 (2012); [2] R. Shimizu et al., Phys. Rev. Lett., 114, 146103 (2015); [3] S. Ichinokura et al., ACS Nano 10, 2, 2761 (2016); [4] P. Rosenzweig et al., Phys. Rev. B 100, 035445 (2019).