O 82: Graphene I: Adsorption, Intercalation and Doping

Time: Thursday 15:00–17:15 Location: GER 37

O 82.1 Thu 15:00 GER 37

Graphene quantum dot nanoarray in a van der Waals heterostructure — •Keda Jin[1,2,3], Junting Zhao[1,2,3], Tobias Wichmann[1,3,4], F. Stefan Tautz[1,3,4], Jose Martinez-Castro[1,3], Markus Ternes[1,2,3], and Felix Lüpke[1,3] — [1]Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich, 52425 Jülich, Germany. — [2]Institut für Experimentalphysik II B, RWTH Aachen, 52074 Aachen, Germany. — [3]Jülich Aachen Research Alliance (JARA), Fundamentals of Future Information Technology, 52425 Jülich, Germany. — [4]Institut für Experimentalphysik IV A, RWTH Aachen, 52074 Aachen, Germany.

Graphene quantum dots (GQDs) are promising candidates for spin qubits because of their long spin coherence and short switching time. Although isolated GQDs have been created in van der Waals (vdW) heterostructures, the fabrication of GQD nanoarrays is still an ongoing challenge. Here, we realize a regular two-dimensional array of GQDs in a vdW heterostructure composed of graphene on a single layer of $1T$-$NbSe_2$ on top of $2H$-$NbSe_2$ ($Gr/1T/2H$-$NbSe_2$) by combining mechanically assembled vdW heterostructures and nanoscale phase engineering. Using scanning tunneling microscopy (STM) and non-contact atomic force microscopy (nc-AFM), we demonstrate that the $1T$-$NbSe_2$ phase periodically dopes the graphene layer, which in response exhibits quantized charge states that can be manipulated by the local electric field of the tip. Our findings show a possible route for the creation of a highly dense GQD nanoarray with a potential application in the quantum technologies.

O 82.2 Thu 15:15 GER 37

Sn-induced superstructures at the EG/SiC(0001) interface and their influence on EG — •Zamin Mamiyev, Chitran Ghosal, and Christoph Tegenkamp — Institut für Physik, Technische Universität Chemnitz

Intercalating chemical elements into the buffer layer/SiC(0001) interface is a promising approach to delaminate a single carbon layer forming extended epitaxial graphene (EG) layers with new properties. In this regard, Sn is an interesting candidate because the triangular monolayer structure on SiC(0001) was shown to exhibit a Mott state [1].

By means of low energy electron diffraction we studied recently in detail the intercalation of Sn below the buffer layer grown before on SiC(0001) and found 1×1 and $(\sqrt{3}\times\sqrt{3})$ Sn induced phases depending on details of the annealing procedure [2]. As deduced from transport but also local spectroscopy, the former is metallic while the latter reveals a band gap opening. In addition, we found a $(\sqrt{3}\times\sqrt{3})R30°$ interference pattern with respect to the EG lattice. This so-called broken Kekulé order forms due to the breaking of the bond order in EG and often found in the vicinity of buckled $Sn(1\times1)$ subsurface clusters opening the channel for K,K' scattering in EG. The electronic structure and, in particular, the formation of a Mott gap will be investigated by electron energy loss spectroscopy.

[1] S. Glass et al., PRL 114, 247602 (2015). [2] Z. Mamiyev and C. Tegenkamp, Surf. & Int. 34, 102304 (2022).

O 82.3 Thu 15:30 GER 37

Stabilizing high-overlap organic crystals by n-doped graphene — •Fabio Calcinelli and Oliver Hofmann — Graz University of Technology, Graz, Austria

Organic thin-films are known for their tuneable properties, which are strongly dependent on the polymorph the films assume. Traditional ab-initio studies of thin films geometries remain computationally prohibitive, due to the immense number of possible configurations, and this results in the need for structure-property relationships to help in the design of materials with certain target properties.

In this contribution, we examine the correlation between the energy of polymorphs and their interlayer LUMO-LUMO overlap for benzoquinone on n-doped graphene. Employing smart-data machine learning, we use a small number of DFT calculations to predict the energy and the LUMO-LUMO overlap for a large sample of polymorphs. We assess the impact of the n-doping of the substrate on this overlap/stability relationship by performing the energy and overlap evaluation for different intensities of n-doping.

We find that, for low dopant concentrations, there exists no relationship between interlayer overlap and stability, while for high dopant concentrations, it appears that polymorphs with high LUMO-LUMO overlap are predominant in the stability ranking. We explain this different behavior as the consequence of charge transfer from the substrate to the first molecular layer producing different LUMO occupations for different dopant concentrations, resulting in a stabilizing effect of high LUMO-LUMO overlap for high concentrations only.

O 82.4 Thu 15:45 GER 37

Theoretical studies of nitrogen-doped corrugated graphene on metal supports — •Ivan Abilio[1] and Krisztián Palotás[1,2] — [1]Wigner Research Center for Physics, Budapest, Hungary — [2]ELKH-SZTE Reaction Kinetics and Surface Chemistry Research Group, Szeged, Hungary

For maximizing the usability of N-doped graphene materials, the microscopic understanding of their structures (graphitic, pyridinic, pyrrolic, etc) and electronic properties at the atomic scale is crucial. Motivated by this, density functional theory calculations of N-doping configurations in corrugated graphene single layers on Ir(111) and Ru(0001) substrates were performed. Taking systematically selected N-doped graphene defect configurations, their energetic preference is found to be correlated with their atomic structure. Scanning tunneling microscopy simulations with different tip models [1] were performed, and a great variety of STM image contrasts showed a good agreement with experimental STM images. The negatively charged N atoms embedded into the corrugated graphene layer suggest single atom catalytic potential for these structures.

[1] G. Mándi and K. Palotás, Phys. Rev. B 91, 165406 (2015).

O 82.5 Thu 16:00 GER 37

Formation model of AlF_3 intercalated aggregates on HOPG surfaces for rechargeable battery applications — •Sindy Rodríguez Sotelo[1,2], Adriana Candia[1], Igor Stanković[3], Mario Passeggi Jr.[1,2], and Gustavo Ruano[4] — [1]IFIS, CONICET-UNL, Santa Fe, Argentina — [2]FIQ, UNL, Santa Fe, Argentina — [3]Scientific Computing Laboratory, Center for the Study of Complex Systems, IPB, Belgrade, Serbia — [4]CAB, CNEA, Bariloche, Argentina

Rechargeable batteries based on ion intercalation are currently of great interest, with lithium being the most studied and with the best performance. However, the limited reserves of this material and its scarce distribution in the world have led to the research for new alternatives. In our recent work, we studied the intercalation of AlF_3 in HOPG as a new possibility to use this molecule in rechargeable batteries and to understand its role when used as a solvent component [1,2]. Based on our recent reports, in this work we theoretically and experimentally characterize the dynamics of the AlF_3 interlayers, to discuss the possibility of decoupling the first graphene layers from the HOPG under ultra-high vacuum conditions. We show experimental measurements performed by STM, XPS and AES, also proposing a model to glimpse the growth and behavior of AlF_3 in the surface layers of the HOPG substrate, to provide clarity regarding the formation of the AlF_3/HOPG interface. We complement the analysis with calculations in the formalism of DFT and molecular dynamics (MD). **References** [1] A.E. Candia et al. Carbon, 186 (2022) 724. [2] Rodríguez et al. Phys. Chem. Chem. Phys, 23 (2021), 19579.

O 82.6 Thu 16:15 GER 37

Atomic and electronic structure of intercalated Pb layers in proximity to epitaxial graphene — •Chitran Ghosal, Markus Gruschwitz, Julian Koch, and Christoph Tegenkamp — Institut für Physik, TU Chemnitz, Chemnitz, Germany

Selective intercalation into the buffer layer/SiC(0001) interface can realize atomically sharp interface layers near epitaxial graphene that resemble novel 2D heterosystems. High-Z elements are interesting candidates, not only because they are expected to host Dirac fermions, but also because spin effects are expected in graphene.

Intercalation of Pb leads to the formation of different phases,

which we have studied in detail using scanning tunneling microscopy. As recently shown, intercalated Pb bilayers form nanostripes under graphene and show fingerprints of plumbene. These Pb layers are rotated with respect to graphene, which breaks the sublattice symmetry and is associated with an electronic gap in graphene [1]. Hexagonally arranged bubble-like structures with an average size of 2.3 nm were also found. Scanning tunneling spectroscopy (STS) again revealed the formation of mini-gaps in the 2D heterosystem. Spatially resolved mapping of the electronic structure also clearly showed the existence of a network of edge states around the edges of the bubbles.

[1] C. Ghosal et al., PRL 129, 116802 (2022)

O 82.7   Thu 16:30   GER 37

Tailoring permanent carrier density in epitaxial graphene by F4-TCNQ molecular doping for quantum Hall resistance standards — ●Yefei Yin, Atasi Chatterjee, Davood Momeni, Mattias Kruskopf, Martin Götz, Stefan Wundrack, Frank Hohls, Klaus Pierz, and Hans Werner Schumacher — Physikalisch-Technische Bundesanstalt, Bundesallee 100, 38116 Braunschweig, Germany

A prerequisite for the development of graphene electronics is the reliable control of its carrier density. Since epi-graphene on SiC already exhibits a high electron density of up 10^{13} cm^{-2}, the goal is a controlled reduction towards the charge-neutrality point. This was achieved by molecular doping with the acceptor F4-TCNQ doping stacks deposited on graphene. By precise adjustment of the dopant concentration and controlling the initial carrier density of the undoped graphene, we can tune the carrier density in a wide range from intrinsic n- to the p-type. Precision measurements of the quantum Hall resistance show a quantization accuracy of 10^{-9} which underlines the high quality and suitability of this doping method for electronic device application in metrology. [1] Our data show a correlation between the electron density and the onset of the i = 2 quantum Hall plateau which gives a valuable criterion for evaluating graphene-based resistance standards since the *classical* relation (n = ieB/h) to determine the centre of the quantum Hall plateau is not valid in epi-graphene. The results are discussed in the charge transfer model. [1] Y.Yin et al., Adv. Physics Res. 2022, DOI: 10.1002/apxr.202200015.

O 82.8   Thu 16:45   GER 37

Transport properties of intercalated epitaxial graphene — ●Markus Gruschwitz, Zamin Mamiyev, Chitran Ghosal, Susanne Wolff, Thomas Seyller, and Christoph Tegenkamp — Institut für Physik, TU Chemnitz, Chemnitz, Germany

Large-scale growth and electronic manipulation of graphene are of great interest for nanoelectronics applications. The former is already realized by epitaxial growth and intercalation of buffer layers on SiC(0001). Electronic properties such as doping level and gap opening of the decoupled graphene layer strongly depend on the element and the structure of the interface. In this study we investigated Sn, Bi and In intercalated graphene by means of nanoscopic transport, supplemented by STM, LEED and PES. Depending on the film thickness and temperature, (1x1) phases can be formed with all three elements, accompanied by the formation of free-standing graphene. The fully intercalated phases show metallic conductivity behavior (σ_{Sn}=0.63 mS/□, σ_{Bi}=0.74 mS/□). The slightly lower conductivity of 10% compared to clean MLG is most likely due to interface scattering rather than different doping levels of the EG. Further heating allows the formation of $\sqrt{3}$-structures for all intercalants. Thereby, the conductivities drop by 2-3 orders of magnitude, accompanied by an increase of the anisotropy with respect to SiC step structure. This is indicative of inhomogeneous desorption especially around the step edges. The detailed analysis with the variable range hopping model yield localization lengths that correlate with the size of intercalated areas.

O 82.9   Thu 17:00   GER 37

Magnetotransport and structural properties of Bi(110) islands on epitaxial graphene — ●Julian Koch[1], Sergii Sologub[1,2], Chitran Ghosal[1], and Christoph Tegenkamp[1] — [1]Institut für Physik, TU Chemnitz, Reichenhainerstr. 70, 09126 Chemnitz — [2]Institute of Physics, NAS of Ukraine, Nauki avenue 46, 03028 Kyiv

Magnetotransport measurements at 10 K using a 4 T magnet were performed on Bi islands on monolayer graphene/SiC with average thickness up to 3.6 bilayers (BL). They are supported by structural investigations using SPA-LEED and STM, which reveal that Bi predominately grows as needle-like islands with a (110) termination.

The analysis of the magneto and Hall conductivity gives the electron concentration and the mobility. The electron concentration slightly decreases with $4 \cdot 10^{11}$ cm^{-2}BL^{-1} from an initial value of $1.4 \cdot 10^{13}$ cm^{-2} for the clean surface. The mobility decreases from 2100 cm^{2}V^{-1}s^{-1} to a minimal value of 1630 cm^{2}V^{-1}s^{-1} at 2.4 BL. Moreover, the electron electron interaction contribution to the magneto conductivity, which is almost negligible for the clean surface, strongly increases linearly with approximately 0.44 mS/BL. At 3.6 BL it is equal to approximately half the Drude conductivity at zero field.

The weak localization (WL) of the clean surface gradually transitions to weak anti-localization (WAL) with increasing Bi coverage. This is associated with a decrease of the dephasing length of the scattered electrons, which is correlated with the average distance of the islands obtained from STM measurements.