O 93: Graphene II: Electronic Structure and Growth

Time: Friday 10:30–12:30   Location: GER 37

O 93.1   Fri 10:30   GER 37

Energy dissipation on magic angle twisted bilayer graphene —
●Marcin Kisiel, Alexina Ollier, Urs Gysin, and Ernst Meyer — Department of Physics, University of Basel, Klingelbergstrasse 82, CH-4056 Basel, Switzerland

Understanding nanoscale energy dissipation is nowadays among few priorities particularly in quantum systems. While traditional Joule dissipation omnipresent in today's electronic devices is well understood, the energy loss of the strongly interacting electron systems remains largely unexplored. Twisted bilayer graphene (tBLG) is a host of interaction-driven correlated insulating phases, when the relative rotation is close to the magic angle (1.08deg). Here, we report on low temperature (5K) nanomechanical energy dissipation of tBLG measured by sharp tip of the pendulum atomic force microscope (pAFM). Ultrasensitive cantilever tip acting as an oscillating gate over the quantum device shows dissipation peaks attributed to different fractional filling of the flat energy bands. pAFM provides exquisite spatial resolution and thus allows to determine the twist angle distribution of tBLG. Strikingly it does it without literally touching the sample surface. Application of magnetic fields provoked strong oscillations of the dissipation signal at 3/4 band filling, which we identified as familiar to Aharonov-Bohm oscillations. The work demonstrates that nanomechanical energy dissipation provides a rich source of information on the dissipative nature of the correlated electronic system of tBLG, with implications for coupling a mechanical oscillator to the quantum devices.

O 93.2   Fri 10:45   GER 37

Raman features of graphene and weyl semimetals beyond the standard nonadiabatic theory — ●Nina Girotto and Dino Novko — Institute of Physics, Zagreb, Croatia

Although graphene has already been thoroughly studied with Raman spectroscopy, there still exists a disagreement about the broadening mechanisms of the E2g mode and its actual temperature dependence [Nano Lett. 10, 466 (2010)]. Along with the importance of the nonadiabatic effects in graphene, higher order electron-phonon scattering processes also significantly impact the phonon spectrum. Specifically, the electron-phonon-induced lifetime and energy renormalization of the electron-hole pair excitations bring additional temperature dependence in the electron-coupled phonon modes and their corresponding linewidths. The nonadiabatic theory relying on first principles calculations, developed in [Phys. Rev. B 98, 041112(R) (2018)], is here successfully applied to graphene in various doping regimes and to Weyl semimetals, which contain a 3-dimensional analog of Dirac points in the electron band structure and are, therefore, excellent candidates for achieving a nonadiabatic regime.

O 93.3   Fri 11:00   GER 37

Near-field optical investigation of tetralayer graphene reveals ABCB stacking — ●Konstantin G. Wirth[1], Jonas B. Hauck[2], Alexander Rothstein[3], Christoph Stampfer[3], Dante M. Kennes[2], Lutz Waldecker[3], and Thomas Taubner[1] — [1]I. Institute of Physics (IA), RWTH Aachen University — [2]Institute for Theory of Statistical Physics, RWTH Aachen University — [3]2nd Institute of Physics, RWTH Aachen University

The crystallographic structure of few-layer graphene (FLG) greatly influences its electronic and optical properties. Local probing of the stacking order is usually done by optical measurements, such as infrared absorption [1] or Raman spectroscopy, which work for noncontacted and encapsulated samples but are limited in lateral resolution by diffraction to a few micrometer. Here, we directly probe the electronic properties of tetralayer graphene (4LG) with amplitude and phase-resolved near-field spectroscopy with a broadly tunable laser source over the energy range from 0.28 to 0.56 eV. We differentiate between rhombohedral and Bernal stacking sequences and reveal domains of the previously elusive third stacking order of 4LG, namely ABCB[2], by addressing distinctive interband contributions in optical conductivity. These results establish near-field spectroscopy at the interband transitions as a semi-quantitative tool, enabling the recognition and characterization of FLG domains.

[1] Mak et al., PNAS, 2010 107, 14999-15004 [2] Wirth et al., ACS Nano 2022, 16, 16617-16623

O 93.4   Fri 11:15   GER 37

Image potential states as local probes for graphene magnetism — ●Maciej Bazarnik[1,2,3], Roland Wiesendanger[3], and Anika Schlenhoff[1] — [1]Institute of Physics, University of Münster, Germany — [2]Institute of Physics, Poznan University of Technology, Poland — [3]Department of Physics, University of Hamburg, Germany

Image-potential states (IPSs) evolving in front of polarizable surfaces are known to serve as quantum sensors for various properties, including surface magnetization and interfacial coupling at buried interfaces. Using a spin-polarized scanning tunneling microscopy (SP-STM) setup, the spin-polarized IPS can be probed locally. In hybrid systems such as iron (Fe) -intercalated graphene (Gr) on Ir(111), Fe is known to grow pseudomorphically on the Ir substrate in a monolayer thickness limit. In contrast, the Gr overlayer exhibits a substantial buckling within the moiré unit cell, resulting in a laterally varying hybridization to the Fe. As a result, the monolayer Gr exhibits a laterally varying spin-polarization within the moiré unit cell.

Here, we present resonant tunneling studies on Fe-intercalated Gr, showing a much stronger contrast between the fcc and hcp parts of the moiré unit cell compared to conventional STM images on this system. Moreover, our SP-STM studies show that the IPSs are sensitive to the interlayer coupling of Gr to the underlying ferromagnet. The resulting local IPS spin-polarization can be used to probe the atomic-scale magnetism within the moiré unit cell of the ferromagnet graphene hybrid system.

O 93.5   Fri 11:30   GER 37

Quantitative measurement and first principle prediction of the adsorption structure of graphene on on Cu(111) — ●Matthew A. Stoodley[1,2], Benedikt P. Klein[1,2], Luke A. Rochford[2], Tien-Lin Lee[2], David A. Duncan[2], and Reinhard J. Maurer[1] — [1]University of Warwick, Coventry, United Kingdom — [2]Diamond Light Source, Didcot, United Kingdom

Copper is the dominant substrate for the industrial scale growth of graphene, and graphene-functionalised copper foils are regularly employed as electrodes and heterogeneous catalysts. Yet, little is known about the structure of the graphene-copper interface. We report a quantitative measurement of the adsorption height of graphene on Cu(111) using the x-ray standing waves technique. Using state-of-the-art dispersion-corrected density functional theory, we tested different structural models of graphene on copper. Quantitative agreement with experiment could only be achieved by accounting for the moiré superstructure of the graphene layer on the copper surface. We further investigate how lattice strain affects long-range dispersion interactions and therefore the adsorption height of graphene on Cu(111) as well as graphene bilayer interlayer distances. We use these results to assess the predictive capabilities of common dispersion-corrected density functional approximations. Exploration of the adsorption energy of aromatic molecules of varying size, from benzene to graphene enables us to explore size effects between the substrate and organic overlayer.

O 93.6   Fri 11:45   GER 37

Determining the stability and catalytic formation of graphene on liquid Cu using machine-learning potentials — ●Hao Gao[1], Valentina Belova[2], Maciej Jankowski[2], Hendrik H. Heenen[1], Gilles Renaud[2], and Karsten Reuter[1] — [1]Fritz-Haber-Institut der MPG, Berlin, Germany — [2]ESRF, Grenoble, France

The 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 obtain free energy barriers of possible rate-limiting steps which can be compared to the

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 unchartered computational problems. In that we provide new insight into the domain of liquid metal catalysts which generally lack atomic-scale understanding.

O 93.7   Fri 12:00   GER 37

In-situ photoemission electron microscopy investigation of mono- and bilayer graphene growth on Ru(10$\bar{1}$0) — •Lukas Schewe, Cathy Sulaiman, Lars Buss, Moritz Ewert, and Jan Ingo Flege — Applied Physics and Semiconductor Spectroscopy, Brandenburg University of Technology Cottbus-Senftenberg

Epitaxial graphene growth has often been studied on close-packed transition metal substrates, e. g., the Ru(0001) surface, which is a well-studied model system for strong graphene-support interaction. Here, we focus on a Ru surface with different symmetry, i. e., the Ru(10$\bar{1}$0) surface, to investigate the influence of the presumably modified graphene-substrate interaction on the growth of epitaxial monolayer and bilayer graphene (MLG, BLG) islands. The structural and chemical differences of the graphene on the two different surfaces are investigated by photoemission electron microscopy (PEEM), delivering information on both morphology and electronic structure. In-situ PEEM observation of graphene growth on the Ru(10$\bar{1}$0) substrate by ethylene decomposition reveals the growth characteristics of MLG and BLG, the latter showing second layer nucleation via surface segregation of carbon. Furthermore, depending on growth temperature and relative orientation of the growing islands and surface steps, different growth characteristics are observed, in contrast to previous studies of the graphene/Ru(0001) system whereas similar electronic properties seem to prevail. Yet, when the MLG is decoupled from the Ru(10$\bar{1}$0) substrate via intercalation of oxygen a distinct shift in work function is identified, slightly different from the resulting shift on Ru(0001).

O 93.8   Fri 12:15   GER 37

Defective graphene by topological design — Benedikt P Klein[1,2], Matthew A Stoodley[1,2], Joel Deyerling[3], Michael Clarke[4], Luke A Rochford[1], Marc Walker[2], Raymundo Marcial Hernandez[5], Christian B Nielsen[5], Lars Sattler[6], Sebastian M Weber[6], Gerhard Hilt[6], Alexander Generalov[7], Alexei Preobrajenski[7], Leon BS Williams[1,8], Tien-Lin Lee[1], Alex Saywell[4], Willi Auwärter[3], Reinhard J Maurer[2], and •David A Duncan[1] — [1]Diamond Light Source, UK — [2]University of Warwick, UK — [3]Technische Universität München, Germany — [4]University of Nottingham, UK — [5]Queen Mary University of London, UK — [6]Carl von Ossietzky Universität Oldenburg, Germany — [7]MAX IV, Lund, Sweden — [8]University of Glasgow, UK

Defective graphene is a promising material for both applications in electronic devices and as substrates for modern catalysts. However, generating deliberate imperfection in graphene often relies on brute force methods, like ion sputtering. Herein, we demonstrate one-step bottom-up synthesis of defective graphene with incorporated 5- and 7-membered ring defects by using a polyaromatic feedstock that contains the same topology as the desired defect. Furthermore, the ratio of ideal to defective graphene could be controlled by the temperature used to grow the film. By a combination of X-ray standing waves, non-contact atomic force microscopy, X-ray photoelectron spectroscopy, near-edge X-ray absorption fine structure and density functional theory, we thoroughly characterise these networks and propose this method as a novel method of functionalising graphene.