TT 58: Graphene I: Growth, Structure and Substrate Interaction (joint session O/TT)

Time: Thursday 10:30–12:00   Location: GER 37

TT 58.1 Thu 10:30 GER 37

Design principles for doping graphene for electrochemical CO2 reduction: Insights from Theory — ●SUDARSHAN VIJAY[1], JOSEPH GAUTHIER[2], HENDRIK HEENEN[1], VANESSA BUKAS[1], HENRIK KRISTOFFERSEN[1], and KAREN CHAN[1] — [1]CatTheory, Department of Physics, Technical University of Denmark — [2]SUNCAT Center for Interface Science and Catalysis, Department of Chemical Engineering

Graphene based 2D catalysts hold great promise for CO2 reduction to CO and CH4. Recent experimental investigations [1,2] show metal doped Iron-Nitrogen-Carbon (Fe-N-C) catalysts are able to reduce CO2 to CO at low overpotentials and with high selectivity. However, modelling these materials in an electrochemical environment poses several open challenges. In this work, we present a theoretical investigation on Fe-N-C catalysts which includes the effect of potential, interfacial pH, change in local spin states to properly elucidate the mechanism for CO2 reduction. We find that the electronic structure of Fe-N-C resembles graphene more than it does a metal, with significantly fewer states at the fermi level. Charge dependence of binding energies of key intermediates depend on the position of the highest energy d-orbital with respect to the fermi level. Using computed reaction energetics coupled with mean-field kinetic models, we are able to ascertain the mechanism for CO2 reduction and compare our results with experimental findings. We extend this analysis to other 2D material systems and propose rational design principles.

[1] Science 14 Jun 2019: Vol. 364, Issue 6445, pp. 1091-1094 [2] ACS Energy Lett. 2018, 3, 4, 812-817

TT 58.2 Thu 10:45 GER 37

Graphene/S/Ru(0001): a model system for studying intercalation — ●LARS BUSS[1,2], JENS FALTA[2,3], MORITZ EWERT[1], POLINA SHEVERDYAEVA[4], PAOLO MORAS[4], and JAN INGO FLEGE[1] — [1]Applied Physics and Semiconductor Spectroscopy, BTU Cottbus-Senftenberg, Cottbus, Germany — [2]Institute for Solid State Physics, University of Bremen, Bremen, Germany — [3]MAPEX Center for Materials and Processes, University of Bremen, Bremen, Germany — [4]Istituto di Struttura della Materia, Consiglio Nazionale delle Ricerche, Trieste, Italy

The epitaxial growth of single-layer graphene on transition-metal substrates enables the growth of micrometer-sized islands with excellent crystalline quality, but the possibly strong binding to the substrate has proven detrimental to its materials properties. However, intercalation of foreign atoms lifts the interlayer coupling, restoring its unique electronic structure. We have investigated the intercalation of sulfur underneath graphene on Ru(0001) with low-energy electron microscopy (LEEM) and micro-diffraction (μLEED). We find that sulfur deposited from a molecular precursor at elevated temperatures intercalates through the edge of the island, eventually leading to wrinkles in the graphene. Intriguingly, the overlaying graphene limits the number of possible S/Ru(0001) reconstructions below, preventing the formation of less dense reconstructions like the p(2×2)-S and ($\sqrt{3} \times \sqrt{3}$)-S surface phases. Intensity-voltage LEEM and angle-resolved photoemission (ARPES) prove the free-standing character of the sulfur intercalated graphene, which is found to be p-doped by 380 meV.

TT 58.3 Thu 11:00 GER 37

Au intercalation under epitaxial graphene on Ru(0001): the role of graphene edges — ●SEBASTIAN GÜNTHER[1], TEVFIK ONUR MENTES[2], ROBERT REICHELT[1], ELISA MINUSSI[2], BENITO SANTOS[2], ALESSANDRO BARALDI[2], and ANDREA LOCATELLI[2] — [1]TUM, Dept. Chemie, Lichtenbergstr.4, D-85748 Garching — [2]Sincrotrone Trieste, AREA Science Park I-34149 Trieste

Au intercalation at the graphene-Ru(0001) interface is investigated at elevated temperature by using low energy electron - and x-ray photo-electron emission microscopy (LEEM/XPEEM). Graphene (g) growth by ethylene decomposition at 1030 K on a Au pre-covered Ru surface pushes the Au adatoms towards the g-free surface area. When instead, evaporating Au on a partly g-covered surface, a modified Stranski-Krastanov growth on Ru with two atomic wetting layers followed by 3D islands is observed. At 970 K the growth follows a precise order: 1)The first wetting Au layer is grown exclusively on the g-free area of the Ru surface. 2)After completion of the Au/Ru layer, Au intercalation below g-flakes sets in. 3)Having completed the g/Au/Ru layer,

second wetting Au layer growth ouside the g-covered area takes place. 4)After completion of the Au/Au/Ru layer, intercalation of the second Au layer underneath g sets in. At 970 K, Au is shown not to stick to g-covered surface regions. This crucial finding singles out the g-edges as the only intercalation channel. Chemical maps at different stages during Au growth point to a pronounced kinetic barrier at the g-edges preventing intercalation before the Au monolayer is completed outside the g-covered regions.

TT 58.4 Thu 11:15 GER 37

Covalent functionalization of epitaxial graphene on cubic-SiC(001) — ●DMITRII POTOROCHIN[1,2,3], OLGA MOLODTSOVA[1,2], VICTOR ARISTOV[1,4], ALEXANDER CHAIKA[4], MAXIM RABCHINSKII[5], MARINA BAIDAKOVA[2,5], NIKOLAI ULIN[5], PAVEL BRUNKOV[2,5], DMITRY MARCHENKO[6], and SERGUEI MOLODTSOV[2,3,7] — [1]DESY, Hamburg, Germany — [2]ITMO University, Saint Petersburg, Russian Federation — [3]TU Bergakademie Freiberg, Freiberg, Germany — [4]ISSP RAS, Chernogolovka, Russian Federation — [5]Ioffe Institute RAS, Saint Petersburg, Russian Federation — [6]Helmholtz-Zentrum Berlin, Berlin,Germany — [7]European XFEL, Schenefeld, Germany

Some properties of graphene, although being unique, can serve as a deterrent factor to its use in some fields of technology. For instance, the bandgap absence complicates the fabrication of graphene-based logic elements of electronics. Furthermore, the transparency in the visible spectral region imposes a restriction on the use of graphene as an active material for elements of photonics (photodetectors, photovoltaics, etc.). Covalent functionalization of graphene is a promising approach to overcome such limitations. In the current report, we present an investigation of the electronic structure and morphology of epitaxial graphene on cubic-SiC(001) covalently modified by organic dyes. Results of high-resolution X-ray photoelectron spectroscopy (HR-XPS), photoemission electron microscopy (PEEM), and scanning tunneling microscopy (STM) studies are given. This work was supported by RAS, RFBR (Grant Nos. 17-02-01139, 17-02-01291), and Minobrnauki of Russia (Project 3.3161.2017/4.6).

TT 58.5 Thu 11:30 GER 37

Controlled formation of nanobubbles in graphene — ●PIN-CHENG LIN[1], RENAN VILLARREAL[1], HARSH BANA[1], KEN VERGUTS[2,3], STEVEN BREMS[3], STEPHEN DE GENDT[2,3], MANUEL AUGE[4], FELIX JUNGE[4], HANS HOFSÄSS[4], HOSSEIN GHORBANFEKR[5], M FALLH[5], FRANÇOIS PEETERS[5], MEHDI NEEK-AMAL[5], CHRIS VAN HAESENDONCK[1], and LINO DA COSTA PEREIRA[1] — [1]Quantum Solid State Physics, KU Leuven, 3001 Leuven, Belgium — [2]Departement Chemie, KU Leuven, 3001 Leuven, Belgium — [3]imec, 3001 Leuven, Belgium — [4]II. Institute of Physics, University of Göttingen, Göttingen 37077, Germany — [5]Department of Physics, University of Antwerp, 2020 Antwerp, Belgium

Strained nanobubbles have been used to engineer the electronic structure of graphene through the creation of pseudomagnetic fields (e.g. via strain imposed by a selected substrate or mechanical actuators), however, they provide limited controllability. Here we report on the controlled formation of noble gas (He, Ne, Ar) nanobubbles in graphene (on various substrates) using ultra-low energy (ULE) ion implantation. ULE ion implantation allows us to precisely tune the number of implanted ions and their kinetic energy, which in turn controls the bubble formation efficiency and bubble density. Our experimental approach is based on scanning tunneling microscopy/spectroscopy (STM/STS), synchrotron X-ray photoelectron spectroscopy (XPS), Raman spectroscopy, among others, complemented by density functional theory (DFT) and molecular dynamics calculations (MD), which give insight into the bubble formation and stability mechanisms.

TT 58.6 Thu 11:45 GER 37

Simulating the scattering of a hydrogen atom from graphene using a high-dimensional neural network potential. — ●SEBASTIAN WILLE[1,2], MARVIN KAMMLER[2], MARTÍN L. PALEICO[3], JÖRG BEHLER[3], ALEC M. WODTKE[1,2], and ALEXANDER KANDRATSENKA[2] — [1]Institute for Physical Chemistry, Georg-August University Göttingen, Germany — [2]Department of Dynamics at Surfaces, Max Planck Institute for Biophysical Chemistry, Göttingen, Germany — [3]Theoretical Chemistry, Georg-August University Göttingen, Germany

Understanding the formation of covalent bonds due to atomic-scale motions and energy dissipation pathways involved is an ongoing challenge in the field of chemistry. Recent measurements of the translational energy loss distribution of hydrogen atoms scattered from graphene showed a bimodal pattern. The second generation reactive empirical bond order potential was fitted to ab initio electronic structure data obtained from embedded mean-field theory to generate a potential energy surface (PES). First-principles dynamics simulations using the provided PES were able to reproduce the bimodal feature of the energy loss spectrum and were in qualitative agreement with experimental results. But these investigations could not fully provide a detailed description of the scattering and sticking mechanisms. Therefore, we developed a full-dimensional neural network PES by fitting to the density functional theory data in order to further reduce the remaining errors by the fitting procedure of the PES underlying molecular dynamics simulations performed.