# O 94: Focus Session: Proximity Effects in Epitaxial Graphene I

Proximity-induced correlation effects in low dimensional electron gases are essential for the design of new quantum materials with tailored electronic, magnetic and optical properties. The recent reports on superconductivity in twisted bilayer graphene is an intriguing example. Inherently, proximity coupling often comes along with further effects, e.g. hybridization, charge transfer, etc., so that intended quantum phenomena become easily quenched. Epitaxial graphene (EG) resembles a truly 2D electron gas system and is known for its ability of manifold and flexible functionalization schemes at its vacuum and interface site, e.g. by doping, encapsulation with high-Z materials as well as formation of superlattices. This Focus Session will provide an overview of the current state of research and possible perspectives.

Organizer: Christoph Tegenkamp (TU Chemnitz)

Time: Thursday 15:00–18:00 Location: MA 141

**Topical Talk** O 94.1 Thu 15:00 MA 141

**Tailoring the electronic structure of epitaxial graphene on SiC** — •Kathrin Küster — Max-Planck-Institut für Festkörperforschung, Stuttgart, Germany

Inducing correlation effects in low dimensional electron gases by proximity coupling is a powerful concept for the design of new quantum materials with tailored electronic, magnetic and optical properties. The DFG research unit FOR5242 investigates spin-orbit interaction and electronic correlations of 2D electron gas systems using epitaxial graphene grown on SiC substrates. After a brief introduction to the objectives of the research unit, I will present two different projects of our group. (i) By intercalation with lanthanides like Gd [1] and Yb [2,3], graphene can be driven to extreme doping levels where the system deviates from the low-energy limit of massless Dirac fermions. (ii) In contrast, intercalation of graphene with Pb leads to the formation of almost charge neutral graphene. In general intercalation with heavy elements is an interesting research area since those might induce spin-orbit coupling effects and exotic collective phenomena. Besides the influence of the intercalants on the electronic properties of the graphene, also the investigation of the electronic and structural properties of the intercalants is an exciting topic on its own since they often strongly deviate from their 3D counterparts.

[1] S. Link et al. PRB 100, 121407(R) (2019); [2] P. Rosenzweig et al. PRB 100, 035445 (2019), [3] P. Rosenzweig et al. PRL 125, 176403 (2020); [4] B. Matta et al. Phys. Rev. Research 4, 023250 (2022); [5] P. Schädlich et al. Adv. Mater. Interfaces 2300471 (2023).

O 94.2 Thu 15:30 MA 141

**Investigating intercalated Lead layers underneath epitaxial graphene** — •Philip Schädlich[1,2], Franziska Schölzel[1], Peter Richter[1,2], and Thomas Seyller[1,2] — [1]Technische Universität Chemnitz, Institut für Physik, 09126 Chemnitz — [2]Center for Materials, Architectures and Integration of Nanomembranes (MAIN), 09126 Chemnitz

Intercalation is a promising approach for tailoring the electronic structure of epitaxial graphene on SiC. Beyond that, it enables the formation of otherwise unstable two-dimensional (2D) phases of elements and opens a route to investigate the interplay between the two 2D materials and the substrate [1,2].

We have studied in detail the Pb intercalation process itself, as well as the structure and electronic properties of the 2D Pb layer by means of low-energy electron microscopy and photoelectron spectroscopy (PES). Probing the electronic structure using angle-resolved PES reveals a strict (1×1) periodicity of the intercalated Pb with respect to the substrate. The quasi-freestanding graphene turns out to be effectively screened from the doping influence of the substrate leading to charge-neutrality. In fact, the 2D Pb layer is compensating the spontaneous polarization of the substrate. Our results experimentally quantify the interaction between the 2D Pb layer, the substrate and the graphene layer and demonstrate a first step towards controlling the diversity of 2D Pb phases.

[1] C. Ghosal, Phys. Rev. Lett. 129, 116802 (2022), [2] P. Rosenzweig, Phys. Rev. B 101, 201407(R) (2020)

O 94.3 Thu 15:45 MA 141

**Pb-intercalated epitaxial graphene on SiC: Charge transfer mechanism and spin-split interlayer bands** — •Bharti Matta[1], Philipp Rosenzweig[1], Kathrin Küster[1], Craig Polley[2], and Ulrich Starke[1] — [1]Max-Planck-Institut für Festkörperforschung, Stuttgart, Germany — [2]MAX IV Laboratory, Lund, Sweden

Intercalation of epitaxial graphene is a robust way of modifying its properties and stabilizing two-dimensional (2D) interlayers at the graphene/SiC interface. Pb being a heavy element superconductor has a great potential as an intercalant due to possible proximity effects in graphene. In this work, angle-resolved photoelectron spectroscopy (ARPES) shows that the near charge-neutrality of Pb-intercalated quasi-freestanding monolayer graphene on SiC (Pb-QFMLG) involves charge transfer from both interlayer-Pb and SiC. This is based on the observation that at 20 K, the $p$-doping of Pb-QFMLG increases by $\approx 9 \times 10^{10}$ cm$^{-2}$ compared to room temperature, which can be attributed to the freezing out of SiC bulk $n$-dopants. The bands of interlayer-Pb cross the Fermi level, confirming its metallic nature. ARPES in the repeated Brillouin zone confirms the (1×1) alignment of Pb relative to SiC. Constant initial state mapping as a function of photon energy reveals dispersionless Pb bands, corroborating their 2D nature. Potassium adsorption results in predominant charge transfer into graphene, inducing a strong $n$-doping of $\approx 10^{14}$ cm$^{-2}$. Further, spin-resolved ARPES uncovers a strong spin splitting of the interlayer bands. However, any proximity-induced spin splitting in graphene is still ambiguous. This ongoing work is supported by DFG through FOR 5242.

O 94.4 Thu 16:00 MA 141

**Electronic structure of intercalated epitaxial graphene: A first principles study** — •Andres Unigarro[1], Florian Günther[2], Franziska Schölzel[1], Niklas Witt[3], Thomas Seyller[1], Tim Wehling[3], and Sibylle Gemming[1] — [1]Institute of physics, TU Chemnitz, Chemnitz, Germany — [2]UNESP, Rio Claro, Brazil — [3]Universität Hamburg, Hamburg, Germany

Two-dimensional materials such as graphene are fascinating because they combine unique mechanical and electronic properties. Proximity effects introduced from the interaction with intercalates and intercalation layers change the electronic, optical and transport properties of epitaxial graphene (EG) while preserving the regular honeycomb structure. Elements of the IV group such as Pb can be used as intercalants below a graphene sheet, leading to partly well-defined hetero bilayer with different functionalities. Heavy atoms like Pb furthermore, promise to introduce additional effects such as spin-orbit coupling to the electron gas of graphene. Using a density functional based tight binding approach, we investigate the modifications in the electronic structure of EG due to proximity effects induced by intercalation.

O 94.5 Thu 16:15 MA 141

**Mott states under cover: Silicon intercalation of epitaxial graphene** — •Niclas Tilgner, Zamin Mamiyev, and Thomas Seyller — TU Chemnitz Institut für Physik

Mott-Hubbard bands are well known from different superstructures on SiC(0001). Typical examples are the siliconrich $(3 \times 3)$ and $(\sqrt{3} \times \sqrt{3})R30^\circ$ reconstruction, but also the unreconstructed $(1 \times 1)$ surface. In all cases the electrons occupying the dangling bonds show a non-negligible interaction among themselves, which gives rise to a Mott-Hubbard metal-insulator transition. The dispersion and the energetic splitting of the upper and lower Hubbard band was found to scale with the distance between the dangling bonds and the dielectric properties of the environment, respectively. It could be achieved to prepare an analogous system underneath graphene by intercalation of silicon. The talk focuses on the major results, which were acquired. Experiments using diffraction techniques (LEED, SPA-LEED) allow the evaluation of the silicon arrangement, that is found to consist of different patches with $(\sqrt{3} \times \sqrt{3})R30^\circ$, $(2 \times 2)$ and $(3 \times 3)$ ordering. Furthermore, investigations of the prepared samples with photoemission techniques (ARPES, XPS) reveal several surface states, which can partly be attributed to the dangling bonds of the silicon adatoms and therefore to one of the Hubbard bands. Unintuitively, no avoided crossing with the Dirac cone is observed. Possible reasons for this circumstance will be discussed.

**Topical Talk** O 94.6 Thu 16:30 MA 141

**Proximity spin-orbit coupling and topological interfaces in graphene / alloyed transition metal dichalcogenide heterostructures** — •Stephen Power[1] and Zahra Khatibi[2] — [1]School of Physical Sciences, Dublin City University, Ireland — [2]School of Physics, Trinity College Dublin, Ireland

Spin-orbit coupling (SOC) in graphene can be enhanced by proximity effects in stacked graphene/transition metal dichalcogenide (TMDC) heterostructures. The TMDC layer composition determines the nature and strength of the resultant SOC induced in graphene.

Here, we demonstrate the evolution of the SOC in graphene with the composition of an alloyed TMDC layer ($W_xMo_{1-x}Se_2$). Density functional theory is used to simulate systems with different compositions and distributions, and allows local and global signatures of the metal-atom alloying to be clarified. The low-energy spin and electronic behavior follow a effective medium model which depends only on the composition ratio $x$.

While graphene/$MoSe_2$ has a simple band gap, valley-Zeeman-driven inverted bands are present in graphene/ $WSe_2$. The topological state of mixed systems can thus be tuned by varying $x$, with band gap closure occurring at a critical value. Furthermore, heterostructures with W- and Mo-domains can host boundary states similar to those between AB- and BA-stacked domains in bilayer graphene. Finally, we trace the electronic evolution of alloyed heterostructures from homogeneous effective medium to domain-localised bands as the domain size increases.

O 94.7 Thu 17:00 MA 141

**Non-equilibrium carrier dynamics and band structure of graphene on 2D tin** — •Maria-Elisabeth Federl[1], Niklas Hofmann[1], Leonard Weigl[1], Johannes Gradl[1], Niklas Witt[2], Tim Wehling[2], Biao Yang[3], Neeraj Mishra[4], Camilla Coletti[4], and Isabella Gierz[1] — [1]University of Regensburg — [2]University of Hamburg — [3]Technical University Munich — [4]Istituto Italiano di Tecnologia, Pisa

Van der Waals heterostructures are novel artificial materials with tailored electronic properties that might enable new technologies in the fields of optoelectronics, spintronics, and quantum computing. Wafer-scale heterostructures with clean interfaces are easily obtained by confinement heteroepitaxy on SiC(0001) [1] where various elements are intercalated below the covalently bound carbon buffer layer on SiC. Confinement heteroepitaxy has recently been used to stabilize a 2D layer of Sn with exotic electronic properties [2-4]. A possible interaction with the quasi-freestanding graphene layer on top remains unexplored. Using time-and angle-resolved photoemission spectroscopy, we find a surprisingly short-lived non-thermal carrier distribution inside the Dirac cone of graphene as well as a rigid down-shift of the whole $\pi$-band that we attribute to interlayer hybridization revealed by density functional theory.

[1] Nat. Mater. 19, 637 (2020)
[2] J. Phys. D: Appl. Phys. 49, 135307 (2016)
[3] Appl. Phys. Express 11, 015202 (2018)
[4] Phys. Rev. Lett. 122, 126403 (2019)

O 94.8 Thu 17:15 MA 141

**Spatially Resolved Sn Intercalation of Epitaxial Buffer Layer of Graphene on SiC** — •Benno Harling and Martin Wenderoth — IV. Physical Institute, Georg-August-University Göttingen, Germany

Intercalation, the insertion of atoms into a layered material such as graphene, has been shown to be a versatile means of altering electronic properties through proximity. While intercalation is typically discussed as a spatially homogeneous process, our study focusses on the inhomogeneities on the mesoscopic scale. An investigation of a partially intercalated epitaxial graphene buffer layer with tin was conducted using Kelvin Probe Force Microscope (KPFM). $\sqrt{3} \times \sqrt{3}$ and $1 \times 1$ phases of the intercalated tin can be discriminated as doping on the graphene layer corresponding to a specific shift in the work function. This allows to locate and quantify the different intercalation phases as well as the transition to the pristine buffer layer on a sub-micrometer scale. This opens insight into the diffusion of the intercalated atoms, i.e., the intercalation dynamics on this scale. Gradients from fully intercalated areas to the pristine buffer layer are observed and correlated with the topographic information. Our results show the strong impact of substrate steps as a significant boundary for the diffusion process, more specifically diffusion of Sn mainly happens on a terrace or via pin hole like channels across steps.

O 94.9 Thu 17:30 MA 141

**Low energy plasmons in Sn-intercalated quasi-free-monolayer-graphene** — •Zamin Mamiyev and Christoph Tegenkamp — Institut für Physik, Technische Universität Chemnitz

Graphene plasmons, particularly confined within structured and sandwiched graphene, represent a fascinating area of research. They hold the potential to facilitate the manipulation of light at sub-wavelength scales while providing a platform for investigating the many-body electronic interactions. Our study focuses on low-energy plasmonic excitations in epitaxial quasi-free monolayer graphene, created through the intercalation of Sn beneath the buffer layer on 4H-SiC(0001). [1]

The quantitative examination of the sheet plasmon dispersion reveals that the Sn-induced (1×1) interface exhibits metallic properties, leading to the formation of charge-neutral graphene. The dispersion of the sheet plasmon (activated by mild doping with K) is slightly redshifted with w.r.t. epitaxial graphene (EG). On the other hand, the Sn-diluted ($\sqrt{3} \times \sqrt{3}$) reconstruction forms intrinsically n-type doped graphene with a dispersion similar to the epitaxial monolayer graphene on the carbon buffer layer. Moreover, it appears that the dipolar coupling of longitudinal charge density fluctuations in EG to the interface layer triggers the formation of a plasmonic multipole component at higher frequencies. Our study shows that angle-resolved plasmon spectroscopy is a promising technique to investigate proximity effects of excitations in electronically weakly coupled 2D heterostructures.

[1] Z. Mamiyev and C. Tegenkamp, Surf. & Int. 34, 102304 (2022)

O 94.10 Thu 17:45 MA 141

**Evidence of Sn-induced Mott states coupled to Dirac electrons in epitaxial graphene** — •Chitran Ghosal[1], Zamin Mamiyev[1], Siheon Ryee[2], Niklas Witt[2,3], Tim Wehling[2,3], and Christoph Tegenkamp[1] — [1]Institute of Physics, Technische Universität Chemnitz, Reichenhainer Str. 70, Germany — [2]I. Institute of Theoretical Physics, University of Hamburg, Notkestraße 9-11, 22607 Hamburg, Germany — [3]The Hamburg Centre for Ultrafast Imaging, Luruper Chaussee 149, 22607 Hamburg, Germany

The adsorption of 1/3 ML of Sn on SiC(0001) was shown to reveal a robust 2D Mott state. In this work we studied the electronic structure of intercalated Sn below the buffer layer on SiC(0001) coming along with the formation of n-type doped graphene. SPALEED clearly revealed new $\sqrt{3}$-reconstruction spots after the intercalation process. By means of EELS and STS we analyzed in detail the electronic structure of this heterostructure. We found strong evidence of a hybridization between Sn-induced Mott states and the graphene $\pi$-bands. This leads to a gap opening of around 200 meV at the Dirac point. Moreover, a new state at around 1.2 eV emerged which we assign to the upper Hubbard band. The formation of narrow bands is supported by EELS measurements, showing besides the sheet plasmon also a pronounced interband transition at 1.5 eV. Combined DFT and dynamical mean field theory calculations support the emergence of strong Mott Hubbard correlations in this system. The emerging correlation effects show a pronounced stacking and charge transfer dependence and lead to spectral functions in good agreement with the experiments.