

Towards Reality in Nanoscale Materials XI

11 – 13 February 2025
Levi, Finland



Organizers

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Financial and media support

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Programme

	Tuesday		Wednesday		Thursday
	<i>chair: Ugeda</i>		<i>chair: Pawlak</i>		<i>chair: Fasel</i>
9:00	Khajetoorians	9:00	Fasel	9:00	Ugeda
9:40	Cahlik	9:40	Grill	9:40	Grutter
10:00	Carva	10:00	Weymouth	10:00	Gerstmann
10:20	<i>Coffee</i>	10:20	<i>Coffee</i>	10:20	<i>Coffee</i>
	<i>chair: Friedrich</i>		<i>chair: Batzill</i>		<i>chair: Ganguli</i>
11:00	Feng	11:00	Besley	11:00	Pawlak
11:40	Hofmann	11:40	Zhou	11:40	Schulz
12:00	Jolie	12:00	Achilli	12:00	Franzke
12:20	Ghorbani-Asl	12:20	Hinaut	12:20	Fischer
12:40	<i>Lunch</i>	12:40	<i>Lunch</i>	12:40	<i>Lunch</i>
	<i>chair: Besley</i>		<i>chair: Greplova</i>		<i>chair: Drost</i>
16:20	Ajayan	16:20	Valenti	16:20	Swart
17:00	Mustonen	17:00	Fumega	17:00	Cortés-del Río
17:20	Åhlgren	17:20	Aumayr	17:20	Reichmayr
17:40	<i>Coffee</i>	17:40	<i>Coffee and posters</i>	17:40	<i>Coffee</i>
	<i>chair: Valenti</i>				<i>chair: Ghorbani-Asl</i>
18:20	Greplova			18:20	Batzill
19:00	Friedrich			19:00	Kotakoski
19:20	Jing			19:20	
				21:00	<i>Workshop dinner</i>

Contents

Organizers	3
Support	3
Programme	4
Contents	5
Tuesday	10
Alex Khajetoorians A quantum simulator to study electronic structure in the Hofstadter limit	12
Ales Cahlik Observation of Multiferroicity in two-dimensional NiBr ₂	13
Karel Carva Magnetism and THz excitations in quasi-2D systems under external perturbations	14
Xinliang Feng Beyond Layers: Unveiling the Potential of Organic 2D Crystals in Emerging Material Science	15
Stephan Hofmann Fast Descriptors and Accelerated Process Development for 2D materials	16
Wouter Jolie Polarons in single-layer MoS ₂	17
Mahdi Ghorbani-Asl Encapsulated alkali metals between bilayer graphene: a computational study	18
Pulickel Ajayan Phases of Boron-Carbon-Nitrogen Compositions	19
Kimmo Mustonen Hexatic Phase in Covalent 2D Crystals	20
Harriet Åhlgren Synthesis of 2D monolayer gold on functionalised graphene	21
Eliska Greplova Autonomous Quantum Control in the Age of AI	22
Rico Friedrich Controlling Electronic and Magnetic Properties of 2D Non-van der Waals Materials by Data-driven Design	23
Liwei Jing Strain-induced two-dimensional topological crystalline insulator .	24

Wednesday	25
Roman Fasel Quantum magnetism in nanographene spin chains	26
Leonhard Grill Switching in 2D molecular layers – role of the atomic-scale surroundings	27
Alfred J. Weymouth Lateral force microscopy reveals the sides of molecules and probes individual chemical bonds	28
Elena Besley Electronic structure of two-dimensional bipartite lattices constructed from six-nodal monomers	29
Shengqiang Zhou Ion-irradiation induced magnetic phase transition in 2D semiconductor CrSBr	30
Simona Achilli Novel carbon 2D materials and functionalized graphene: theory and experiments	31
Antoine Hinaut Tuning thermal expansion of supramolecular networks	32
Roser Valenti Exploring Correlated phases and topology in van der Waals platforms	33
Adolfo Fumega Nature of the Unconventional Heavy-Fermion Kondo State in Monolayer CeSi11	34
Friedrich Aumayr Coulomb-driven nanopore formation in 2D materials by impact of slow highly charged ions	35
Thursday	37
Miguel Ugeda Superconductivity in octahedrally coordinated (1T) layered dichalcogenides	39
Peter Grutter Spatially resolved trap states and random telegraph noise in semiconductors	40
Uwe Gerstmann Screening and relaxation in weakly coupled 2D heterostructures: implications for tunable molecular spin-coupling	41
Rémy Pawlak Proximity-induced superconductivity in molecular quantum systems	42
Fabian Schulz On-surface synthesis of nanographenes with polyradical states	43
Katharina Lorena Franzke Magnetic fingerprint of single spins at 2D nanostructures	44
Jeison Fischer Spin polarization of an Anderson impurity in MoS2 mirror twin boundaries	45
Ingmar Swart Local noise measurements on a d-wave superconductor	46
Eva Cortés-del Río Observation of Yu-Shiba-Rusinov states in superconducting graphene	47
Fanny Reichmayr Raman Marker Bands for In-situ Quality Control During Synthesis Of 2D c-MOFs	48
Matthias Batzill Modification of 2D-Transition Metal Dichalcogenides by Excess Metal Incorporation	49

Jani Kotakoski Corrugation-dominated elastic modulus of defect-engineered graphene	50
Posters	51
Tiago Antão Electric Field Control Of Moiré Skyrmion Phases in Twisted Multiferroic NiI ₂ Bilayers	53
Büşra Gamze Arslan Observation of Edge States on NiI ₂ /NbSe ₂	54
Thuy An Bui 2D metal-iodides in bilayer graphene encapsulation	55
Nan Cao Heisenberg S=1/2 Antiferromagnetic Molecular Chain	56
Madhuri Chennur Electrical impact of defect evolution on multi-layer WSe ₂	57
Aaron Dunbrack Monolayer Moiré: Replacing Layers With Valleys In Graphene	58
Jana Dzibelova Atomic-resolution investigation of 2D hematene	59
Linda Feuerstein Nitrile Groups as Build-in Molecular Sensors for Interfacial Effects at Electrocatalytically Active Planar Carbon-Nitrogen Materials	60
Akash Gupta Single-electron charging on Au nanoparticles with Radio Frequency-excitation detected by pendulum Atomic Force Microscope	61
Shuyu Huang Moiré energy dissipation driven by nonlinear dynamics	62
Mitisha Jain Molecular Dynamics Simulations of He and Ar Ion Irradiation of 2D MoS ₂ on Au and SiO ₂ Substrates	63
Umair Javed Pore shape selection in hBN with electron beam induced chemical effects	64
Netta Karjalainen Hamiltonian parameter learning with artificial neural networks	65
Clara Kofler From four t(w)o three: How 4D-STEM measurements of 2D materials lead to 3D information	66
Pekka Koskinen The structure and stability trends in atomically thin metallenes	67
Daniil Krukliniskii Point and line defects in 2D and bulk CrSBr compounds from first-principles calculations	68
Artem Kuklin Stability Challenges in Predicted 2D Materials with Nonequivalent Sublattices: Topological Constraints and the Limits of PBC	69
Lauri Kurki Automated structure discovery for scanning tunnelling microscopy	70
Greta Lupi Atomic-scale quantum magnets have gained significant attention due to their potential	71
Manuel Längle Two-dimensional noble gas clusters in a graphene sandwich	72
Anshika Mishra Manipulation of Charge Defects in Monolayer NiBr ₂	73
Rabia Nawaz Low energy ion irradiation of few-layer MoS ₂	74
Marcel Niedermeier Quantum computing topological invariants of two-dimensional quantum matter	75
Lorena Niggli Dynamic heterogeneity in the self-induced spin glass in elemental neodymium	76
Anastasiia Nihei Heterostructures of 2D Materials from an Integrated Computational Approach	77

Hermann Osterhage Fock-Darwin states in artificial atomic structures	78
Ram Prakash Pandeya Molecular Order Induced Charge Transfer in a C60- Topological Insulator Moiré Heterostructure	79
Diana Propst Unraveling the highly defective regime of graphene and hexagonal boron nitride on the scale of atomic resolution: Automated image acquisition and analysis	80
Nandhini Ravindran Overcoming challenges in preparation of monolayer hBN and vdW heterostructure TEM samples	81
Prosun Santra Tailoring the electromechanical properties of two-dimensional materials via defect engineering	82
Yubing Wang Few layer plates & scrolls of the magnetic topological insulator MnBi ₂ Te ₄	83
Christoph Wilhelmer First-principles investigations of noise in ultra-scaled 2D field effect transistors	84
Nian Wu Precise Large-Scale Chemical Transformations on Surfaces: Deep Learning Meets Scanning Probe Microscopy with Interpretability . . .	85
Walter Zuccolin Single-layer boron phosphide on metallic surfaces: screening of promising substrates by first-principles	86
Hans Ågren Point and complex defects in single-layer transition metal dichalcogenides	87
Participants	88
Index	90

Tuesday 11.02.2025

- chair: Ugeda*
- 9:00 - 9:40 **Khajetoorians**
A quantum simulator to study electronic structure in the Hofstadter limit
- 9:40 - 10:00 Cahlik
Observation of Multiferroicity in two-dimensional NiBr₂
- 10:00 - 10:20 Carva
Magnetism and THz excitations in quasi-2D systems under external perturbations
- 10:20 - 11:00 *Coffee*
- chair: Friedrich*
- 11:00 - 11:40 **Feng**
Beyond Layers: Unveiling the Potential of Organic 2D Crystals in Emerging Material Science
- 11:40 - 12:00 Hofmann
Fast Descriptors and Accelerated Process Development for 2D materials
- 12:00 - 12:20 Jolie
Polarons in single-layer MoS₂
- 12:20 - 12:40 Ghorbani-Asl
Encapsulated alkali metals between bilayer graphene: a computational study
- 12:40 - 16:20 *Lunch*
- chair: Besley*
- 16:20 - 17:00 **Ajayan**
Phases of Boron-Carbon-Nitrogen Compositions
- 17:00 - 17:20 Mustonen
Hexatic Phase in Covalent 2D Crystals
- 17:20 - 17:40 Åhlgren
Synthesis of 2D monolayer gold on functionalised graphene
- 17:40 - 18:20 *Coffee*
- chair: Valenti*
- 18:20 - 19:00 **Greplova**
Autonomous Quantum Control in the Age of AI
- 19:00 - 19:20 Friedrich
Controlling Electronic and Magnetic Properties of 2D Non-van der Waals Materials by Data-driven Design
- 19:20 - 19:40 Jing
Strain-induced two-dimensional topological crystalline insulator

A quantum simulator to study electronic structure in the Hofstadter limit

Alex Khajetoorians¹

¹Radboud University

Quantum simulators are a pathway to study novel physical phenomena which are difficult to predict or observe in synthesized materials. To this end, the physical behavior of materials ranging from gasses to superconducting qubits has been used to emulate Hamiltonians. The most iconic of which is the Hubbard model, where previously unobserved phenomena were seen. To date, there is still a lack of viable platforms for quantum simulation to study confined electrons in strong magnetic fields, where the magnetic length is on the order of the periodicity of the lattice. For typical crystals, this corresponds to magnetic fields that are unattainable with common laboratory magnets. Moreover, it is still a grand challenging to control the orbital and lattice symmetries in such platforms, as well as the long-range nature of the coupling. In this talk I will discuss a new quantum simulator to study electronic structure in the Hofstadter limit, which is based on using Cs atoms on the semiconducting surface of InSb. We begin by patterning Cs atoms on the surface with scanning tunneling microscopy, sculpting confinement potentials on the 2DEG which act as artificial atoms (i.e. quantum dots). I will review how these artificial atoms contain multi-orbital symmetry, and can be used to emulate molecular orbitals and molecular structure. I will then focus on the magnetic field response. I will show the response of these artificial atoms to strong magnetic fields, where they exhibit Fock-Darwin states. I will link this to the Hofstadter picture and further comment on perspectives to use this platform to study the role of spin-orbit coupling, interactions, and topology.

Observation of Multiferroicity in two-dimensional NiBr₂

Ales Cahlik¹ Antti Karjasilta¹ Adolfo Fumega¹ Javaria Arshad¹ Mohammad Amini¹ Anshika Mishra¹ Büşra Gamze Arslan¹ Robert Drost¹ Jose Lado¹ Peter Liljeroth¹

¹Aalto University

Multiferroics are materials that exhibit more than one type of ferroic order, such as ferromagnetism and ferroelectricity. Recent advancements in two-dimensional (2D) materials have identified monolayer NiI² as the first known monolayer type-II multiferroic, featuring magnetic spin spirals coupled with ferroelectric polarization. In this study, we report the discovery of multiferroicity in monolayer NiBr², further expanding the family of verified single-layer multiferroic materials. Using scanning tunneling microscopy (STM), we demonstrate that NiBr², grown epitaxially on highly oriented pyrolytic graphite (HOPG), shows behavior remarkably similar to that of NiI², with multiferroic order driven by a magnetic spin spiral and spin-orbit coupling. STM imaging reveals periodic modulations in the electrostatic potential below 5 K, appearing as stripe-like variations in the local density of states (LDOS), indicating the multiferroic order. Furthermore, we demonstrate that the boundaries of multiferroic domains can be manipulated using external electric fields, underscoring the strong magnetoelectric coupling in NiBr². Unlike NiI², NiBr² can be synthesized from a single precursor source, which significantly simplifies the growth process, highlighting the practical advantages of NiBr² for further studies. These findings not only confirm the broader occurrence of multiferroicity in Ni-based halides but also establish monolayer NiBr² as a promising platform for the exploration of van der Waals 2D multiferroic materials.

Magnetism and THz excitations in quasi-2D systems under external perturbations

Karel Carva¹ Krishna K. Pokhrel¹ Dávid Hovančík¹ Jiří Pospíšil¹

¹Charles University Prague

Magnetic van der Waals materials with very weak exchange coupling between magnetically ordered layers represent an interesting intermediate stage between the more explored cases of isotropic bulk-like exchange and the recently intensively studied ideal 2D limit (monolayer) [1]. We perform a complex investigation of lattice and magnetic excitations induced by external perturbations employing the synergy of DFT calculations, infrared, terahertz, and Raman spectroscopies in several quasi-2D materials. One of them is trihalide VI₃, where the transition to the long-range ferromagnetic order is accompanied by the observed variations of phonon frequencies induced by the strong magnetoelastic coupling. The acoustic magnon mode acquires here unusually high energy reaching to THz range, but dramatically softens at temperatures where a second lattice distortion has been reported in the literature. First-principles calculations show the strong connection of magnetic ordering to the lattice. Furthermore, a ground state with exceptionally high orbital momentum is predicted and the electronic configuration is compared to recent measurements based on the x-ray magnetic circular dichroism [3]. These findings suggest the possibility of controlling magnetic anisotropy in this system by selective occupation of specific lattice modes. A highly interesting connection between formation of helimagnetism and breaking of both inversion and rotational symmetry of the lattice has been recently discovered in single-layer Ni dihalides [4]. Here we examine how magnetism and lattice symmetry are affected by pressure in NiBr₂. References:

[1] M. Gibertini, et al., *Nat. Nanotech.* 14, 408 (2019)

[2] D. Hovančík, et al., *J. Phys. Chem. Lett.* 13, 11095 (2022)

[3] D. Hovančík et al., *Nano Lett.* 23 (2023) 1175

[4] Song et al., *Nature* 602, 7898 (2022).

Beyond Layers: Unveiling the Potential of Organic 2D Crystals in Emerging Material Science

Xinliang Feng¹

¹Technische Universität Dresden

Organic 2D crystals with extended in-plane conjugation and strong interlayer coupling are a class of layered materials with distinctive electronic structures and exotic physicochemical phenomena, offering significant application potential. A common synthesis method is bottom-up synthesis, creating single-layer to few-layer 2D polymers/supramolecular polymers, 2D conjugated polymers or covalent-organic frameworks, and 2D conjugated metal-organic frameworks. A key challenge is achieving controlled 2D polymerization in two directions under thermodynamic/kinetic control in solution, solid state, or at the surface/interface. In my talk, I will present novel 2D polymerization methods and design strategies to achieve efficient 2D conjugation in specific 2D conjugated polymers, like 2D poly(arylenevinylene)s and 2D poly (benzimidazobenzophenanthroline) ladder-type structures. These polymers provide a platform for high intrinsic carrier mobilities, crucial for future organic opto-electronics and spintronics. Next, I will discuss our recent advancements in 2D conjugated metal-organic framework materials, emphasizing their applications in MOFtronics and beyond. Then I will discuss on-water surface chemistry as a potent synthetic platform for organic 2D crystals and their van der Waals heterostructures, utilizing water-surface confinement for enhanced chemical reactivity and selectivity. A key focus will be on the surfactant-monolayer-assisted interfacial synthesis method, known for its efficiency in arranging precursor monomers on the water surface and controlled 1D/2D polymerization. These distinct 2D crystal structures, with tailorable conjugated building blocks, tunable pore sizes and thicknesses, and remarkable electronic structures, are promising for applications in electronics, optoelectronics, and spintronics. Additionally, I will discuss other physicochemical phenomena and application potentials of organic 2D crystals, such as in membranes and emergent energy devices.

Fast Descriptors and Accelerated Process Development for 2D materials

Stephan Hofmann¹ Jinfeng Yan¹ Ryo Mizuta¹ Ye Fan¹ Vitaly Babenko¹

¹University of Cambridge

We systematically explore nucleation, crystal growth and process mechanisms for quantum materials at the atomic monolayer limit using CVD-based approaches coupled to new process vectors such as catalytic enhancement. This talk focuses on the development of a microreactor set-up for high-throughput operando scanning electron microscopy and a closed-loop platform for accelerated process discovery. We focus on WS₂ as a model material system and explore surface-based mechanisms linked to sodium tungstates, NaOH and NaCl on SiO₂ and sapphire substrates in conjunction with widely used gaseous (MO)CVD precursors. We report various hot salt substrate corrosion phenomena that lead to the substrate spreading of W intermediates and significantly influence the growth mechanisms. We can directly resolve the formation and wetting behaviour of liquid droplets and show that the growth scenario particularly for mono-layers is not following a vapour-liquid-solid (VLS) model. Holistic calibration of SE contrast allows us to reveal WS₂ nucleation and in-plane growth kinetics. We introduce a fast-screening descriptor approach for holistic data-driven optimization across underpinning large parameter spaces.

Polarons in single-layer MoS₂

Wouter Jolie¹ Camiel van Efferen¹ Arne Schobert² Tfyche Tounsi¹ Laura Pätzold² Michael Winter² Jeison Fischer¹ Thomas Michely¹ Roberto Mozara² Tim Wehling²

¹Universität zu Köln, Germany

²Universität Hamburg, Germany

Wouter Jolie¹, Camiel van Efferen¹, Arne Schobert², Tfyche Y. Tounsi¹, Laura Pätzold², Michael Winter², Mark Georger¹, Affan Safeer¹, Christian Krämer¹, Jeison Fischer¹, Jan Berges³, Thomas Michely¹, Roberto Mozara², Tim Wehling^{2,4,1} Universität zu Köln, Germany² Universität Hamburg, Germany³ Universität Bremen, Germany⁴ The Hamburg Centre for Ultrafast Imaging, Germany Polaron are quasiparticles that arise from the interaction of electrons or holes with lattice vibrations. Though polarons are well-studied across multiple disciplines, experimental observations of polarons in two-dimensional crystals are sparse. We use scanning tunneling microscopy and spectroscopy to measure the properties of polarons emerging from coupling of non-polar zone-boundary phonons to Bloch electrons in n-doped metallic single-layer MoS₂. The latter is kept chemically pristine via a protective graphene layer. Tunneling into the vibrationally coupled polaronic states leads to a series of evenly spaced peaks in the differential conductance on either side of the Fermi level. Using density functional theory calculations, we show that the energy spacing stems from a renormalized M-point phonon, responsible for the formation of stable polarons in metallic MoS₂.

Encapsulated alkali metals between bilayer graphene: a computational study

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The intercalation of various atomic and molecular species into layered materials has recently emerged as a new paradigm in materials science research, given its crucial role in advancing energy and ion storage technologies [1]. Notably, a dense arrangement of lithium between graphene sheets, described as closely packed lithium multilayers, was observed in an in-situ TEM study [2]. More recently, the intercalation of double layers of K, Rb, and Cs, with a $C^6M^2C^6$ composition, was also reported between bilayer graphene [3]. In this study, we systematically investigate the intercalation of alkali metals (AMs), specifically sodium, potassium, rubidium, and cesium, between graphene sheets using density functional theory (DFT) calculations. We examine the structures and energetics of both monolayer and bilayer intercalates with closely packed structures and compare these findings with those of AM atoms adsorbed on graphene. By evaluating the intercalation energetics across various AM concentrations, we demonstrate that while single-layer intercalation of AMs is energetically favorable, bilayer configurations can form at higher AM concentrations, particularly for Cs. Our DFT calculations reveal that the projected separations between AM atoms at the most energetically favorable concentrations are larger than those observed experimentally. Additionally, we investigate the effects of pressure-related to spatial confinement between graphene sheets—and charge transfer between AM atoms and graphene on the stability of intercalated layers. This study offers new insights into the atomic structure and physical properties of novel two-dimensional layered materials encapsulated within bilayer graphene.

References

- [1] A.V. Krasheninnikov, et al., Nano Lett. 24, 12733 (2024).
- [2] M. Kuhne, et al., Nature 564, 234 (2018).
- [3] Y.-C. Lin, et al., Nat. Commun. 15, 6445 (2024).

Phases of Boron-Carbon-Nitrogen Compositions

Pulickel Ajayan¹

¹Rice University

The ternary phase space consisting of boron, nitrogen and carbon shows a number of thermodynamically stable phases of binary and ternary nature. There has been tremendous amount of work in the past on optimizing conditions to grow these phases and understanding their stability. This talk will focus on some of these interesting materials that have compositions of C, BN, CN and BCN. The effort in synthesizing hexagonal as well as cubic phases of these compositions will be discussed with the goal of achieving 2D and 3D structures with specific applications in mind.

Hexatic Phase in Covalent 2D Crystals

Kimmo Mustonen¹ Thuy An Bui¹ David Lamprecht¹ Jacob Madsen¹ Peter Kotrusz² Clemens Mangler² Viera Skakalova² Jani Kotakoski¹

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Ice melting into water is an example of a first order phase transition characterized by a discontinuous change in one or more thermodynamic parameters such as entropy. According to Kosterlitz, Thouless, Nelson, Halperin, and Young [1-2], however, the melting process is fundamentally different in 2D materials where it unfolds continuously and is mediated by thermally excited topological defects. This rather exotic intermediate thermodynamic state that is neither purely crystalline nor liquid, is dubbed as the hexatic phase, and it is characterized by quasi-long-range orientational and short-range translational orders. Whereas different physical phenomena on various length scales have been successfully used to test this idea [3-4], due to the high melting point and low thermodynamic stability of covalent 2D crystals, detailed observations of solid-to-liquid transitions in these materials have thus far remained inaccessible. Here, in contrast, we overcome these limitations by combining crystals of 2D silver iodide in graphene sandwich in a van der Waals heterostructure setting [5] and observe the AgI crystals in an atomically resolved scanning transmission electron microscope (STEM) at temperatures reaching 1160 °C. By using a convolutional neural network to track atom positions in thousands of atomically resolved STEM images, we show that 20-30 °C below the AgI melting temperature, a highly dynamic state emerges, which is shown to be the first observation of the hexatic state in covalent materials.

[1] Kosterlitz, J. M., & Thouless, D. J. (1972) *Journal of Physics C: Solid State Physics*, 5(11), L124.

[2] Nelson, D. R., & Halperin, B. I. (1979) *Physical Review B*, 19(5), 2457.

[3] Zheng, X. H., & Grieve, R. (2006) *Physical Review B-Condensed Matter and Materials Physics*, 73(6), 064205.

[4] Huang, P. et al. (2020) *Nature Nanotechnology*, 15(9), 761-767.

[5] Mustonen et al. (2022) *Advanced Materials*, 34(9), 2106922.

Synthesis of 2D monolayer gold on functionalised graphene

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Kimmo Mustonen¹ Elena Besley² Jani Kotakoski¹ E. Harriet Åhlgren³

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The search for truly 2D metals has been ongoing for sometime. Gold nanostructures possess many interesting properties such as plasmonic and catalytic activity that make them especially interesting candidate for metallene synthesis. Already in early 2000 it was shown, that small gold clusters can form flat energetically favorable configurations through s-p-d hybridization of up to 12 atoms in size [1]. Here, we show a robust approach to synthesise monolayer gold on freestanding functionalised graphene membrane [2]. Using low energy ion irradiation, we prepare the graphene sheet with defects and introduce dangling bonds that will support the lateral growth of gold. We then add gold atoms by landing them at the ultra-low energy of 25 eV onto the membrane. The gold atoms are trapped in the open dangling bonds and start the goldene growth on top of the membrane. Atoms that land on a pristine area of graphene will migrate on the surface until they are trapped in a dangling bond or attach into a growing goldene structure. Increasing the amount of gold will result in increasing the lateral size as well as the thickness. We study the atomic structure of the goldene structures by employing high-resolution scanning transmission electron microscopy and the chemical composition by electron energy loss spectroscopy. Complementary ab initio -based atomistic simulations reveal the growth mechanism and elucidate the role of the defects in the graphene membrane and how they affect the energy landscape of the system.

[1] H. Häkkinen, Atomic and electronic structure of goldclusters: understanding flakes, cages and superatoms from simple concepts, *Chem. Soc. Rev.*, 2008, 37, 1847-1859.

[2] W. Joudi, S. Ghaderzadeh, A. Trentino, K. Mizohata, K. Mustonen, E. Besley, J. Kotakoski, E. H. Åhlgren, Two-dimensional one-atom-thick goldene grown on defect-engineered graphene, submitted.

Autonomous Quantum Control in the Age of AI

Sam Katirae-Far¹ Yuta Matsumoto¹ Joseph Rogers¹ Vinicius Hernandez¹
Thomas Spriggs¹ Valentina Gualtieri¹ Brennan Undseth¹ Lieven
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Abstract: Contemporary quantum devices keep increasing in size and complexity. While this enormous progress allows both academia and industry to explore quantum systems at an unprecedented scale, the newest generation of devices also comes with unparalleled control and calibration challenges. Enter artificial intelligence: At the heart of all control problems lies a data interpretation task. How much can the predictive and generalization power of AI help us tackle these challenges? In this talk I will explain how to apply modern computational methods, including AI, to control and learn physics of contemporary quantum devices and I will give a perspective on how AI can aid us as we scale up the newest generation of quantum technologies.

Controlling Electronic and Magnetic Properties of 2D Non-van der Waals Materials by Data-driven Design

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Two-dimensional (2D) materials and their heterostructures provide an extensive platform for realizing novel electronic and magnetic functionalities at the nanoscale. While individual 2D systems are traditionally obtained from bulk layered compounds bonded by weak van der Waals (vdW) forces, the recent surprising experimental realization of non-vdW 2D compounds derived from non-layered crystals [1,2] opens up a new direction. They exhibit qualitatively new features due to the cations at their active surfaces. As outlined by our recent data-driven investigations also employing autonomous first-principles calculations [3, 4], several dozens of new candidates showcase a wide range of appealing electronic, optical, and in particular magnetic properties owing to the (magnetic) cations at the surface of the sheets. We find that the oxidation state of the surface cations of the 2D systems as well as accounting for strong surface relaxations upon exfoliation are crucial factors determining their stabilization. At the same time, chemical tuning by surface passivation provides a valuable handle to further control the electronic and magnetic properties of these novel 2D compounds and eventually to even induce ferromagnetism as demonstrated by hydrogenation of 2D CdTiO₃ [5]. Moderate (negative) passivation energies suggest that the property design achieved by the passivation can be reversible. These features thus make non-vdW 2D materials an attractive platform for fundamental as well as applied nanoscience and, in particular, spintronics.

[1] A. Puthirath Balan et al., *Nat. Nanotechnol.* 13, 602 (2018).

[2] A. Puthirath Balan et al., *Mater. Today* 58, 164 (2022).

[3] R. Friedrich et al., *Nano Lett.* 22, 989 (2022).

[4] T. Barnowsky et al., *Adv. Electron. Mater.* 9, 2201112 (2023).

[5] T. Barnowsky et al., *Nano Lett.* 24, 3974 (2024).

Strain-induced two-dimensional topological crystalline insulator

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Topological crystalline insulators (TCIs) host topological phases of matter protected by crystal symmetries. Topological surface states in three-dimensional TCIs have been predicted and observed in IV-VI SnTe-class semiconductors. Despite the prediction of a two-dimensional (2D) TCI characterized by two pairs of edge states inside the bulk gap, materials challenges have thus far prevented its experimental realization. Here we report the growth and characterization of bilayer SnTe on the 2H-NbSe₂ substrate by molecular beam epitaxy and scanning tunneling microscopy. We experimentally observe two anticorrelated, periodically modulated pairs of conducting edge states along the perimeters of the sample with a large band gap exceeding 0.2 eV. We identify these states with a 2D TCI through first principles calculations. Finally, we probe the coupling of adjacent topological edge states and demonstrate the resulting energy shift driven by a combination of electrostatic interactions and tunneling coupling. Our work opens the door to investigations of tunable topological states in 2D TCIs, of potential impact for spintronics and nanoelectronics applications at room temperature.

Wednesday 12.02.2025

- chair: Pawlak*
- 9:00 - 9:40 **Fasel**
Quantum magnetism in nanographene spin chains
- 9:40 - 10:00 Grill
Switching in 2D molecular layers - role of the atomic-scale surroundings
- 10:00 - 10:20 Weymouth
Lateral force microscopy reveals the sides of molecules and probes individual chemical bonds
- 10:20 - 11:00 *Coffee*
- chair: Batzill*
- 11:00 - 11:40 **Besley**
Electronic structure of two-dimensional bipartite lattices constructed from six-nodal monomers
- 11:40 - 12:00 Zhou
Ion-irradiation induced magnetic phase transition in 2D semiconductor CrSBr
- 12:00 - 12:20 Achilli
Novel carbon 2D materials and functionalized graphene: theory and experiments
- 12:20 - 12:40 Hinaut
Tuning thermal expansion of supramolecular networks
- 12:40 - 16:20 *Lunch*
- chair: Greplova*
- 16:20 - 17:00 **Valenti**
Exploring Correlated phases and topology in van der Waals platforms
- 17:00 - 17:20 Fumega
Nature of the Unconventional Heavy-Fermion Kondo State in Monolayer CeSiI
- 17:20 - 17:40 Aumayr
Coulomb-driven nanopore formation in 2D materials by impact of slow highly charged ions
- 17:40 - 19:40 *Coffee and posters*

Quantum magnetism in nanographene spin chains

Roman Fasel¹

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Recent advancements in on-surface synthesis have enabled the creation of unprotected open-shell nanographenes, opening pathways to realize quantum spin models. This presentation examines nanographene spin chains, focusing on three cases: (i) spin- $\frac{1}{2}$ Heisenberg chains from covalently linked olympicenes [1], (ii) spin-1 Haldane chains from triangulenes [2], and (iii) spin- $\frac{1}{2}$ alternating-exchange Heisenberg chains derived from Clar's goblet molecules [3,4]. Using on-surface synthesis, hydrogenation, and STM tip-induced dehydrogenation, we control spin chain length, parity, and termination. Spin excitations are characterized through inelastic electron tunneling spectroscopy and high-level theoretical calculations. In agreement with the corresponding spin models, we observe gapped excitations in integer-spin chains, gapless excitations in half-integer-spin chains, spin fractionalization in spin-1 Haldane chains, and spinon excitations in spin- $\frac{1}{2}$ Heisenberg chains. For spin- $\frac{1}{2}$ alternating-exchange chains, we identify three phases defined by ground state degeneracy and edge excitations. Additionally, Fourier analysis of tunneling data reveals the energy-momentum relation of triplons, bosonic spin-1 quasiparticles. Our results demonstrate the potential of bottom-up synthesis for creating and examining prominent spin models with large exchange interactions, raising hopes for future non-cryogenic quantum spin-based devices.

[1] C. Zhao et al., arXiv:2408.10045 (2024).

[2] S. Mishra et al., Nature 598, 287 (2021).

[3] C. Zhao et al., Nat. Nanotechnol. (2024). <https://doi.org/10.1038/s41565-024-01805-z>.

[4] S. Mishra et al., Nat. Nanotechnol. 15, 22 (2020).

[5] C. Zhao et al., Phys. Rev. Lett. 132, 046201 (2024).

Switching in 2D molecular layers - role of the atomic-scale surroundings

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Azobenzene molecules are prototypical switches, which change between trans and cis isomers upon an external stimulus. Their study in two-dimensional layers on surfaces gives insight into their properties on the single-molecule scale by scanning tunneling microscopy (STM) and allows to investigate how the molecular function is affected by its atomic-scale surroundings. Here, various examples of 2D layers of molecular switches will be presented, using voltage pulses from the STM tip and light as stimuli. The former is very local, allowing to 'write' atomic-scale structures into a 2D molecular layer [1], while the latter can remotely switch many molecules before their optical response is studied at the single-molecule level. The metallic surface underneath a 2D molecular layer is often seen only as a support, but can also affect the switching process [2]. Here, it will be discussed how the role of the metal surface can be reduced by controlled decoupling of the molecules from the surface. This was done by lifting the molecular switching unit, either by using a tripod 'stator' [3], and by involving bulky side groups attached to each molecule [1]. Additionally, molecular switches were also studied by non-contact atomic force microscopy (nc-AFM) on a bulk insulator surface, revealing efficient and reversible photo-switching that depends on the excitation wavelength [4]. Beside the surface, also the direct surrounding of each molecule can affect the switching. This leads to the presence or absence of isomerization processes, depending on the molecular assembly pattern [5], and to cooperative effects that determine which molecule in the neighborhood of a cis isomer is most likely to be switched next. [1] S. Jaekel et al., *Surf. Sci.* 744, 122468 (2024); [2] M. Alemani et al., *J. Phys. Chem. C* 112, 10509 (2008); [3] C. Nacci et al., *Angew. Chem. Int. Ed.* 57, 15034-15039 (2018); [4] S. Jaekel et al., *ACS*

Lateral force microscopy reveals the sides of molecules and probes individual chemical bonds

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Elisabeth Riegel¹ Oliver Gretz¹ Franz J. Giessibl¹

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Understanding 2D materials and heterostructures often requires atomic-scale insights. Scanning tunneling microscopy (STM) and atomic force microscopy (AFM) are the most widely used techniques for these investigations, with recent advances enabling characterization at the picometer scale. Lateral force microscopy (LFM), a derivative of frequency-modulation AFM (FM-AFM), involves oscillating the tip laterally along the surface. By implementing LFM with a qPlus sensor, we achieve lateral amplitudes smaller than atomic diameters, thus ensuring high spatial resolution. Our low-temperature experiments utilize a CO-functionalized tip, similar to conventional FM-AFM setups. A key advantage of our method is the ability to separately measure conservative and dissipative interactions, leveraging the strengths of the frequency-modulation technique. The conservative interaction channel enables probing the sides of adsorbates, which is challenging for normal-force FM-AFM but can be controllably achieved with LFM. Notably, this approach allows direct imaging of hydrogen atoms at the terminal sides of adsorbed molecules. The dissipation channel is more short-range than the tunneling current of STM. It yields information about the potential energy surface that is so sensitive that we can clearly observe differences between covalent bonds.

Electronic structure of two-dimensional bipartite lattices constructed from six-nodal monomers

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Abstract: This work is inspired by recent experimental advancements in sustainable synthesis of two-dimensional graphene-like frameworks from hexasubstituted benzene molecules through a reversible polymerization reaction. We generalise a concept of six-nodal monomers as building blocks for two-dimensional systems and present a class of bipartite periodic lattices, that are generated from the monomers of various structures using homology theory. We analyse the electronic structure of these lattices within the extended Hubbard model and make several general conclusions about topology of their Fermi surface and existence of the Van Hove singularities. We show that proposed lattices with equivalent sites exhibit semi-metal behaviour with conical intersections on the Fermi surface similar to the ones observed in graphene. Introducing heteroatoms to these systems allows to obtain semi-conductors with narrow band gaps ranging 1–2 eV, that can be controlled by varying structure of the monomer and nature of heteroatoms.

Ion-irradiation induced magnetic phase transition in 2D semiconductor CrSBr

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CrSBr is rapidly gaining attention as a prominent candidate within the family of van der Waals magnetic semiconductors [1-3]. Below the Néel temperature of 132 K, the material is supposed to exhibit prototypical A-type antiferromagnetic order. In this work, CrSBr single crystals were synthesized by chemical vapor transport, showing pure antiferromagnetic properties [4]. The pristine CrSBr crystals and flakes were then subjected to ion irradiation, which produces structural defects in the crystals in a controllable way. We observe a transition from antiferromagnetic to ferromagnetic behavior in CrSBr [5, 6]. Already at moderate fluences, ion irradiation induces a remanent magnetization with hysteresis adapting to the easy-axis anisotropy of the pristine magnetic order up to a critical temperature of 110 K. Density functional theory calculations suggest that the displacement of constituent atoms due to collisions with ions and the formation of interstitials favor ferromagnetic order between the layers. Increasing irradiation fluences gradually lowers the Curie temperature, reflecting the impact of crystalline degradation. This suggests that by finely tuning the irradiation parameters and employing precise lithography techniques, it is possible to selectively modulate induced ferromagnetism in CrSBr in terms of magnetization strength, critical temperature, and spatial distribution.

[1] N. P. Wilson, K. Lee, J. Cenker et al., *Nat. Mater.* 20, 1657 (2021).

[2] F. Dirnberger, J.M. Quan, R. Bushati et al., *Nature* 620, 533 (2023).

[3] K. Lin, X. Sun, F. Dirnberger et al., *ACS Nano* 18, 2898-2905 (2024).

[4] F. Long, K. Mosina, et al., *Appl. Phys. Lett.* 123, 222401 (2023).

[5] F. Long, M.Ghorbani-Asl, K. Mosina, et al., *Nano Lett.* 23, 8468-8473 (2023).

[6] F. Long, et al. *Adv. Phys. Res.*, 3, 2400053 (2024)

Novel carbon 2D materials and functionalized graphene: theory and experiments

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Tailoring the properties of 2D materials through functionalization and material design represents one of the goals of the research in the field. In view of applications, a deep understanding of the quantum properties at the nanoscale is mandatory, allowing a parallelism between properties and functionalities and enabling efficient realization protocols. We present two different examples of design and characterization of 2D materials, combining our theoretical analysis with experimental measurements. The first paradigm consists in the assembly of novel 2D carbon networks via temperature-assisted on-surface synthesis of molecular precursors on metals. We demonstrate the possibility to obtain π -conjugated, hybrid sp - sp^2 -2D-carbon structures. Moreover, we perform a multidisciplinary characterization of the steps in the synthesis via *ab initio* calculations, STM, Raman and ARPES experiments and we discuss the role of the metal substrate in the final properties of the 2D product. [1-6] As a second example of functionalization we briefly present the results obtained for the implantation of magnetic defects (Mn, V) in graphene/Cu via ultra-low energy implantation, with applications ranging from spintronics to single-atom catalysis and representing an ideal platform to study Kondo physics. We characterize via *ab initio* methods and different experimental techniques the local structure of the defects, their statistics and the induced magnetic properties. [7,8]

[1] Fratesi, G. et al., *Materials* 11, 2556 (2018).

[2] Rabia, A. et al., *Nanoscale* 11, 18191 (2019).

[3] Rabia, A. et al., *ACS Appl. Nano Mater.* 3, 12178 (2020).

[4] S. Achilli et al. *2D Materials* 8, 044014 (2021).

[5] S. Achilli et al. *Phys. Chem. Chem. Phys.* 24, 13616-13624 (2022).

[6] P. D'Agosta, S. Achilli et al. *arXiv:2404.06896*

[7] P.-C. Lin et al. *ACS Nano* 15, 3, 5449-5458 (2021)

[8] R. Villareal. et al. *ACS Nano* 18, 27, 17815-17825 (2024)

Tuning thermal expansion of supramolecular networks

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Thermal expansion is a fundamental property of solid systems that governs many of their mechanical applications. Due to various mechanisms involving intrinsic molecules flexibility, their propensity to conformational changes or their intermolecular interactions, molecular architecture allows large or negative thermal expansion coefficients. Here, we investigate the thermal response of 2D supramolecular networks on metal surfaces via molecular scale SPM, at 5 and 300K, of hexabenzocoronene molecules equipped with peripheral alkyl side chains [1]. We determine lattice dimension increase, resulting in a giant thermal expansion coefficients up to $980 \pm 10 \cdot 10^{-6} \text{ 1/K}$, twice larger than other molecular systems hitherto reported in literature. By modifying the side chains length, the expansion can be fine-tuned while maintaining its order of magnitude [2]. Molecular dynamics simulations of the SW molecule assemblies support the giant expansion coefficient and reveal a mechanism of expansion based on large thermal fluctuations of the alkyl chains as a result of entropic effects and large anharmonic vibrations. This results in temperature-dependent intermolecular interactions that promote the giant expansion of the supramolecular network with temperature [2,3].

[1] A. Hinaut et al., *Nanoscale* 10, 1337 (2018).

[2] S. Scherb et al., *Small* 2024, 2405472.

[3] S. Scherb et al., *Commun Mater* 1, 8 (2020).

Exploring Correlated phases and topology in van der Waals platforms

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In recent years a plethora of new correlated states have been observed by stacking, twisting and straining two dimensional van der Waals materials of different kind. Some prominent examples are twisted bilayer graphene, bilayer heterostructures of graphene with the spin-orbit assisted Mott insulator α -RuCl₃ –a candidate for Kitaev spin physics–, or bilayer heterostructures of the Mott insulator 1T-TaS₂ with the metal 1H-TaS₂. Unique to these bilayer structures is the possible emergence of phases not foreseeable from the single layers alone, such as heavy fermions, Kondo insulators, quantum spin liquids, correlated metals, or topological superconductors. In this talk I will discuss the microscopic modelling of such heterostructures by a combination of first-principles calculations, effective-model considerations, many-body techniques and statistical methods and will present exemplary cases on the emerging correlated electronic and magnetic properties [1-5] and will compare with experimental results.

[1] Rai et al. PRX, 14 (3), 031045 (2024)

[2] Crippa et al. Nature Communications 15 (1), 1357 (2024)

[3] Akram et al. Nano Letters 24, 890 (2024)

[4] Hu et al. PRL 131, 166501 (2023)

[5] Yang et al. Nature Materials 22, 50 (2023)

Nature of the Unconventional Heavy-Fermion Kondo State in Monolayer CeSiI

Adolfo O Fumega¹ Jose Lado¹

¹Aalto University

Heavy-fermion compounds are materials whose properties stem from the co-existence of a magnetic lattice coupled to a nearly free electron gas forming what is known as the Kondo lattice model. The phase diagram of these systems displays some of the most intriguing many-body states: unconventional superconductivity, critical phases, different sorts of magnetic orders, or heavy-fermion phases. Traditionally, Kondo systems have been studied in bulk materials. Remarkably, the recent isolation of CeSiI in the few-layer limit has established this compound as the first 2D van der Waals heavy-fermion material [Nature 625, 483-488 (2024)]. Interestingly, these experiments report an unconventional momentum-dependent Kondo-screened phase and a dominant magnetic ordered phase at low temperatures in this 2D heavy fermion system. In this presentation, we will introduce a DFT+pseudofermion methodology, which combines density functional theory calculations with a pseudofermion formalism to capture heavy fermion Kondo materials [Nano Letters 24 (14), 4272-4278 (2024)]. This methodology allows us to analyze the multiorbital heavy-fermion electronic structure of monolayer CeSiI. In particular, it allows us to determine the competition between the magnetic exchange interaction of the localized spins and the Kondo coupling with the gas of electrons. We show that magnetic exchange interactions promote an unconventional nodal heavy-fermion hybridization, thus explaining the origin of the unconventional heavy-fermion behavior observed in experiments. Our work introduces a novel methodology to describe heavy-fermion systems from an ab initio perspective in combination with a pseudofermion formalism, whose application can be extended to other heavy-fermion compounds and van der Waals heterostructures.

Coulomb-driven nanopore formation in 2D materials by impact of slow highly charged ions

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The formation of nanopores in 2D materials through the impacts of slow highly charged ions is mainly influenced by the electron mobility of the target and the charge state of the projectile, i.e., the potential energy carried into the collision [1], [2]. Experiments show that in materials with high electron mobility, like single-layer graphene (SLG), rapid electron replenishment prevents long-term damage. However, materials with lower mobility, such as MoS₂, may disintegrate when excited by a highly charged ion due to the formation of a long-lasting positive charge patch [3], initiating a Coulomb repulsion of the positively charged atoms the target layer. A minimum charge state (threshold charge) of the projectile is, however, necessary to form nanopores. We present experimental results for different 2D materials including vdW-heterostructures of 2D materials and model the interaction between HCIs and 2D materials, focusing on electron transfer, charge transport, and atomic dynamics under Coulomb forces [4]. The model successfully predicts the relationship between pore size and the projectile's charge state, showing that materials with higher charge mobility require a higher threshold charge for pore formation. For SLG, no pore formation occurs regardless of the incoming ion charge, since the high electron mobility in SLG enables fast re-neutralization of the electron-depleted impact area thus ensuring long-time stability of the target. The stability of these materials under HCI irradiation can be summarized in a phase diagram, highlighting the relationship between charge mobility and nanopore creation, consistent with experimental findings.

[1] J. Schwestka, et al., ACS Nano 14, 10536 (2020).

[2] R. Kozubek, et al., J. Phys. Chem. Lett. 10, 904 (2019).

[3] A. Niggas, et al., Phys.Rev.Lett 129, 086802 (2022).

[4] A. Sagar Grosse, et al., Nano Lett. 22, 9676 (2022).

Thursday 13.02.2025

- 9:00 - 9:40 *chair: Fasel*
Ugeda
Superconductivity in octahedrally coordinated (1T) layered dichalcogenides
- 9:40 - 10:00 Grutter
Spatially resolved trap states and random telegraph noise in semiconductors
- 10:00 - 10:20 Gerstmann
Screening and relaxation in weakly coupled 2D heterostructures: implications for tunable molecular spin-coupling
- 10:20 - 11:00 *Coffee*
- 11:00 - 11:40 *chair: Ganguli*
Pawlak
Proximity-induced superconductivity in molecular quantum systems
- 11:40 - 12:00 Schulz
On-surface synthesis of nanographenes with polyradical states
- 12:00 - 12:20 Franzke
Magnetic fingerprint of single spins at 2D nanostructures
- 12:20 - 12:40 Fischer
Spin polarization of an Anderson impurity in MoS₂ mirror twin boundaries
- 12:40 - 16:20 *Lunch*
- 16:20 - 17:00 *chair: Drost*
Swart
Local noise measurements on a d-wave superconductor
- 17:00 - 17:20 Cortés-del Río
Observation of Yu-Shiba-Rusinov states in superconducting graphene
- 17:20 - 17:40 Reichmayr
Raman Marker Bands for In-situ Quality Control During Synthesis Of 2D c-MOFs
- 17:40 - 18:20 *Coffee*
- 18:20 - 19:00 *chair: Ghorbani-Asl*
Batzill
Modification of 2D-Transition Metal Dichalcogenides by Excess Metal Incorporation
- 19:00 - 19:20 Kotakoski
Corrugation-dominated elastic modulus of defect-engineered graphene
- 19:20 - 19:40
- 21:00 - 23:00 *Workshop dinner*

Superconductivity in octahedrally coordinated (1T) layered dichalcogenides

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Materials exhibiting multiple superconducting phases are exceptionally rare in nature. The few known examples of multiphase superconductors feature complex phase diagrams, where distinct phases can be independently induced by external stimuli such as pressure or magnetic fields. In this talk, I will present our recent experimental results on the van der Waals 4Hb-TaSSe polytype, which exhibits the coexistence of two superconducting condensates with different spatial localizations within the material. The layered structure of 4Hb-TaSSe, consisting of alternating T-type and H-type polymorphs, facilitates the development of two effectively decoupled superconducting phases with distinctly different microscopic properties. Using high-resolution quasiparticle tunneling and Andreev reflection spectroscopy on the two polymorph layers, we identify two superconducting gaps of different sizes in each layer. These gaps exhibit characteristics consistent with weakly coupled condensates, potentially involving different pairing symmetries. The coexistence of these condensates is further supported by our measurements of critical temperatures and upper critical magnetic fields, which differ significantly between the polymorph layers. To explore the possible superconducting ground states, we employ a minimal model based on ab-initio calculations, which captures many of the experimental observations. Our findings challenge the current understanding of superconductivity in low-dimensional systems and open new possibilities for customizable superconducting devices capable of independently operating multiple superconducting states.

Spatially resolved trap states and random telegraph noise in semiconductors

Peter Grutter¹

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In frequency-modulated atomic force microscopy the measured frequency shift is quadratic in applied bias for metallic samples and probes. However, for semiconducting samples, band bending effects must be considered, resulting in non-parabolic bias curves. We have developed a framework to quantitatively describe a metal-insulator semiconductor (MIS) device formed out of a metallic AFM tip, vacuum gap, and semiconducting sample. We show how this framework allows us to measure dopant concentration, bandgap and band bending timescales of different types of defects on semiconductors with nm scale resolution on Si, 2D MoSe₂ and pentacene monolayers [1,2]. We also measure temporal two-state fluctuations of individual defects at the Si/SiO₂ interface with nanometer spatial resolution using frequency-modulated atomic force microscopy with single electron sensitivity. We demonstrate that two-state fluctuations are localized at interfacial traps, with bias-dependent rates and amplitudes. When measured as an ensemble, the observed defects have a 1/f power spectral trend at low frequencies [3]. Low-frequency noise due to two level fluctuations inhibits the reliability and performance of nanoscale semiconductor devices, and challenges the scaling of emerging spin based quantum sensors and computers. The presented method and insights provide a more detailed understanding of the origins of 1/f noise in silicon-based classical and quantum devices, and could be used to develop processing techniques to reduce two-state fluctuations associated with defects.

[1] M. Cowie, et al., Phys. Rev. Lett. (accepted); arxiv.org/abs/2306.13648

[2] M. Cowie, et al., Phys. Rev. Materials 6, 104002 (2022); arxiv.org/abs/2109.15275

[3] M. Cowie, et al., Proc. Natl. Acad. Sci. USA 121 (44) e2404456121 (2024); arxiv.org/abs/2403.07251

Screening and relaxation in weakly coupled 2D heterostructures: implications for tunable molecular spin-coupling

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Phthalocyanines (Pc) at surfaces are prototype molecules which can host magnetic ions in a well-defined surface environment. The precise external control of the individual spins via electric fields, however, require a detailed knowledge of the influence of the substrate. In a combined experimental and theoretical study, we have investigated the structural and electronic properties of Pb and Mn-phthalocyanine adsorbed on three different graphene-related substrates. Formation of almost identical densely packed PbPc molecular layers with strongly tilted molecules were found on both n-type and p-type doped 2D templates. On HOPG, in contrast, the dispersing molecular states of the less deformed molecular adsorbate stress the importance of substrate mediated interaction and proximity coupling [1,2]. Substituting Pb by Mn we show that this effect is able to trigger the magnetic coupling within the molecular monolayer. The adsorption on either epitaxial or pyrolytic graphene leads to interesting differences in the adsorption behavior and the resulting electronic and magnetic properties. Our analysis demonstrates that the different deformation ability of the vdW coupled systems, like their actual thickness and buckling, triggers the molecular morphology, exhibits a proximity-coupled band structure, and determines the magnetic properties of the adsorbed molecules. It thus provides important implications for future 2D design concepts of molecules at surfaces for sensing as well as information storage devices.

[1] T.T. Nhung Nguyen, T. Sollfrank, C. Tegenkamp, E. Rauls, U. Gerstmann, Phys. Rev. B 103, L201408 (2021).

[2] D. Slawig, M. Gruschwitz, U. Gerstmann, E. Rauls, C. Tegenkamp, J. Phys. Chem. 125, 20087 (2021).

Proximity-induced superconductivity in molecular quantum systems

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Richard Hess¹ Jelena Klinovaja¹ Daniel Loss¹ Ernst Meyer¹

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Topological superconductivity emerges in chains or two-dimensional arrays of magnetic atoms coupled to a superconducting substrate. Another solution for building such atomically precise magnetic structures could be based on the self-assembly or on-surface synthesis of magnetic molecular systems. This would also make possible to control the charge state (spin) of these molecular systems using an external gate voltage as demonstrated for single molecules, an important prerequisite for their future implementation as topological qubits. I will first compare the synthesis of iron-based metal-organic chains on Pb(111) and Ag(111). Using low-temperature scanning probe microscopy, the structures and spectral signatures by tunneling spectroscopy of coordinated Fe atoms with high $S = 2$ spin-state will be shown. I then will discuss our recent works on the supramolecular assembly of radical molecules directly adsorbed on Pb(111), whose charge state can be tuned from anionic to neutral state by the electric field tip of a scanning tunneling microscope (STM). An anionic molecule obtained by an electron donated from the substrate carries a spin-1/2 state [2], as confirmed by the observation of Yu-Shiba-Rusinov (YSR) states in tunneling spectra at millikelvin temperature. At the border of supramolecular islands, low-energy sub-gap states appear localized at edges with an exponential decay towards the island interior. Tight-binding calculations deduce that both the localization and the spectral signature of these states are consistent with Majorana modes protected by mirror symmetry in an antiferromagnetic spin lattice [3]. Overall, our results open up a vast playground for the synthesis of topological organic-superconductor hybrids.

[1] J.C. Liu et al. *Adv. Sci.* 2024, in review

[2] C. Li et al. *Nat. Comm.* 2023 14, 5956

[3] R. Pawlak et al. *PNAS*. 2024, in review

[4] C. Li et al. *ACS Nano* 2024, in review

On-surface synthesis of nanographenes with polyradical states

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π -magnetism in carbon-based nanostructures offers a potential route towards molecular spintronics [PRL 102, 157 (2009)]. However, its realization relies on the precise control over the charge and spin state of the molecular building blocks. One promising candidate to achieve such level of control are triangulene nanographenes and their derivatives. Triangulene molecules are open-shell polycyclic aromatic hydrocarbons with a spin polarization arising from a sublattice imbalance in the finite-sized graphene flakes [PRL 62, 1201 (1989)]. Their electronic and magnetic properties can be engineered through, e.g., variation of their size or heteroatom doping. Here, we show how a combination of solution and on-surface synthesis allows us to fabricate with atomic precision different spin-carrying nanographenes based on nitrogen-doped triangulenes. We characterize their structure as well as electronic and magnetic properties by a combination of low-temperature scanning tunneling microscopy, noncontact atomic force microscopy and Kelvin probe force microscopy, and rationalize our findings by extensive electronic structure calculations. Our results demonstrate how to modify the magnetic ground state, excitation energies and polyradical character of triangulene-based nanographenes through precise control over their atomic structure. *The presented work is carried out in collaboration with the groups of Thomas Frederiksen at Donostia International Physics Center, Diego Peña at Universidade de Santiago de Compostela, and Pavel Jelinek of the Institute of Physics of the Czech Academy of Sciences.

Magnetic fingerprint of single spins at 2D nanostructures

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The recent combination of electron spin resonance and scanning tunneling spectroscopy (ESR–STM) provide a new platform to access single spins of atoms and molecules on surfaces [1, 2]. Characteristic hyperfine (hf) splittings due to the interaction between the electronic spin and the magnetic moments of the nuclei can be measured and compared with theoretical data from density functional theory (DFT). In comparison with defects in bulk material, however, the calculated data deviates considerably from the experimental values. Limited accuracy of the xc functionals or the direct influence of the electric field of the STM-tip have been discussed as possible reasons. In this work, we show that large parts of the observed discrepancies are due to a basically relativistic effect, the suppression of orbital quenching at the surface. In semiconductors, the deformation of atomic orbitals by the crystal field leads to the orbital moment being effectively averaged out. We have developed a fully relativistic method that allows the calculation of this orbital contribution for complex structures [3]. We show that this effect scales with spin-orbit coupling in particular when the unpaired electron is localized in quasi-atomic p-like orbitals. In this case, the orbital part of the hf splitting is far from negligible. It even becomes dominant, surpassing the dipolar contribution by a factor of five. For Pb ions as well as Pb phthalocyanine (PbPc) molecules on the MgO/Ag(111) substrate, this leads to additional hf splittings in the GHz range. It is thus by no means negligible, but required to achieve overall accuracy in predicting the hf interactions of single spins in 2D nanostructures.

[1] S. Baumann, et al., *Science* 350, 417 (2015)

[2] L. Farinacci et al., *Nano Letters* 22, 8470 (2022).

[3] K.L. Franzke, et al. *J. Phys.: Conference Series*. Vol. 2701. No. 1. IOP Publishing, 2024.

Spin polarization of an Anderson impurity in MoS2 mirror twin boundaries

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Mirror twin boundaries (MTB) are line defects in monolayer MoS2 islands grown via molecular beam epitaxy on graphene on Ir(111). MTBs form a well-defined 1D system of finite length, hosting confined states. When the highest occupied confined state is at the Fermi level, it splits into a singly occupied and doubly occupied states, defining an Anderson impurity. When the magnetic moment of this singly occupied quantum confined state of the MTB is screened by the conduction electrons from the bath, a Kondo resonance is established [1]. Here, we report spin-polarized scanning tunneling spectroscopy measurements on a MTB in presence of an out-of-plane magnetic field at different temperatures. We detect changes in peak intensity of the quantum confined impurity states with varying magnetic field, mirroring the changes seen in the split Kondo resonance. These intensity changes of the confined states are the result of their spin-polarized nature under external magnetic field exceeding the thermal energy. Quantitative comparison with numerical renormalization group (NRG) calculations provides evidence of the notable spin polarization of the spin-resolved impurity spectral function under the influence of a magnetic field. Moreover, we extract the field and temperature dependence of the impurity magnetization from the differential conductance measurements and demonstrate that this exhibits the universality and asymptotic freedom of the $S = 1/2$ Kondo effect. Our work shows that mirror twin boundaries can be used as a testing ground for theoretical predictions on quantum impurity models. [2]References: [1] C. van Efferen, J. Fischer, T. A. Costi, A. Rosch, T. Michely, W. Jolie, Nat. Phys. 20, 82-87 (2024). 10.1038/s41567-023-02250-w [2] M. Bagchi, T. Y. Tounsi, A. Safeer, C. van Efferen, A. Rosch, T. Michely, W. Jolie, T. A. Costi, J. Fischer, Accepted in Phys. Rev. Lett. (2024). arXiv:2407.14667

Local noise measurements on a d-wave superconductor

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The pseudogap stands out in the phase diagram of the cuprate high-temperature superconductors because its origin and relationship to superconductivity remain elusive. Two competing hypotheses have emerged: one associating the pseudogap to preformed electron pairs and another attributing it to local order, such as charge density waves. Here, we present unambiguous evidence supporting the pairing scenario, using local shot-noise spectroscopy measurements in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$. Our data demonstrates that the pseudogap energy Δ_{PG} is associated with electron pairing, with pairing energies reaching up to 70 meV. This finding excludes the possibility of the pseudogap arising solely from local orders, and instead indicates a clear relation between the pseudogap phase and Cooper pair formation. Furthermore, we observe that the pairing energy is spatially heterogeneous. These results suggest that the limiting factor for higher T_c in cuprates is not the pairing strength, but rather the phase coherence. Our work resolves a long-standing controversy in the field of high-temperature superconductivity and may reignite the hope to increase T_c in high-temperature superconductors by increasing phase coherence.

Observation of Yu-Shiba-Rusinov states in superconducting graphene

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Nowadays, the properties of pristine graphene are well known. This means that we are now in a position of going one step further and introduce new properties that are not naturally found on it. In this work, we report the observation of the exotic Yu-Shiba-Rusinov states by combining the antagonistic nature of magnetism and superconductivity on graphene. We induce superconductivity in graphene by the proximity effect brought by adsorbing nanometer scale superconducting Pb islands. Using an ultra-high-vacuum (UHV) and low temperature (4K) scanning tunnelling microscope (STM), we detected the emergence of Yu-Shiba-Rusinov states on naturally existing graphene grain boundaries, which unequivocally demonstrate the existence of magnetic moments in this chemically pure line defect. The rise of Yu-Shiba-Rusinov states in graphene make this system a suitable platform to promote Majorana states, key building blocks for topological quantum computing.

Raman Marker Bands for In-situ Quality Control During Synthesis Of 2D c-MOFs

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Two-dimensional conjugated metal-organic frameworks (2D c-MOFs) are a subclass of MOFs that are of particular interest for electrocatalysis due to their good intrinsic conductivity. The electrochemical properties of such 2D frameworks are strongly related to their structure, which in turn is influenced by the synthesis conditions. However, even under formally identical conditions, MOF crystals with different structural properties are obtained, and to date there is no easy-to-apply method to predict the quality of MOF crystals already during synthesis. In the present work, we monitored the formation of phthalocyanine-based 2D MOFs at the air-water interface using in-situ Raman spectroscopy and identified Raman marker bands that characterise the degree of linker aggregation, the reaction progress, and the yield of MOFs formed during the reaction. Using transmission electron microscopy (TEM) measurements on the MOF crystals after synthesis, a correlation between the Raman marker bands and the resulting crystalline domain size distribution of the MOF could be derived. Thus, a method for a non-invasive, fast and simple in-situ quality assessment of the synthesized MOFs was established. These results are an important step towards the automation of MOF synthesis.

Modification of 2D-Transition Metal Dichalcogenides by Excess Metal Incorporation

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The reduced structural constraints in 2D (van der Waals-) materials enable the incorporation of excess atoms into the 2D-lattice. These excess metals may result in point-, extended- defects, or cause a transformation into new stoichiometric phases. In this talk we discuss different mechanisms to induce compositional variations by reacting transition metal dichalcogenides (TMDs) with metal atoms or by removing chalcogen atoms. Not surprisingly, given the broad nature of the TMDs, different transition metal groups respond differently to metal rich conditions. The early transition metal dichalcogenides (group 5 transition metals) have a preference to insert excess metals in between TMD layers [1,2,3]. Group 6 TMDs may incorporate elements at interstitial sites which may cause restructuring into metal-rich mirror twin grain boundary networks [4,5] that can order into novel phases. In contrast, group 10 PtTe₂ can transform into a different stoichiometric phase, i.e., 2D Pt-mototelluride [6,7]. Understanding the processes by which TMDs react with excess metals shine light on synthesis processes and point towards methods for modifying or synthesizing novel 2D materials with new desirable functionalities. References:

- [1] M. Bonilla, et al. *Adv. Mater. Interf.* 7, 2000497 (2020)
- [2] K. Lasek, et al. *ACS Nano* 14, 8473 (2020)
- [3] V. Pathirage, et al. *Nano Letters* 23, 9579 (2023)
- [4] P.M. Coehlo, et al. *ACS Nano* 12, 3975 (2018)
- [5] V. Pathirage, et al. *Mater. Today Nano* 22, 100314 (2023)
- [6] K. Lasek, et al. *ACS Nano* 16, 9908 (2022)
- [7] K. Lasek, et al. *Nano Letters* 22, 9571 (2022)

Corrugation-dominated elastic modulus of defect-engineered graphene

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Pristine graphene is famously the stiffest material ever measured, but, according to simulations, it becomes softer when defects are introduced. The only report with clearly opposite results is from 2010 [1], where graphene indentation was modeled directly. However, the result only holds when the system is similar in size to the indentation tip [2]. Therefore, it was surprising when graphene was reported [3] to stiffen in experiments when irradiated with low-energy Ar. Interestingly, experiments with oxygen plasma gave the opposite result [4]. Here, we create atomically clean graphene and expose it to low-energy Ar before characterization via autonomous atomic-resolution scanning transmission electron microscopy and image analysis [5] and nanoindentation. The samples remain in vacuum through all steps [6]. We find a drastic decrease in the elastic modulus of the defect-engineered graphene in agreement with Ref. [4], but in stark contrast with Ref. [3]. Surprisingly, the decrease is also much stronger than what is predicted by most theoretical studies in the literature. We solve these discrepancies by carrying out experiments without removing surface contamination and by developing a model to account for both the softening of graphene due to removal of strong sp^2 -bonds and its corrugation due to local strain at defects, and comparison to atomistic simulations. In summary, we show that the softening is almost entirely caused by corrugations due to defects larger than single-vacancies and that single-vacancies can be neglected.

[1] Kvashnin, Sorokin and Kvashnin, Fullerenes, Nanotubes and Carbon Nanostructures 18, 497 (2010)

[2] Kvashnin and Sorokin, J. Phys. Chem. Lett. 6, 2384 (2015)

[3] López-Polín et al., Nat. Phys. 11, 26 (2015)

[4] Zandiatashbar et al., Nat. Commun. 5, 4186 (2014)

[5] Trentino et al., Nano Lett. 21, 5179 (2021)

[6] Mangler et al., Microsc. Microanal. 28 (S1), 2940 (2022)

Posters

- Tiago Antão
Electric Field Control Of Moiré Skyrmion Phases in Twisted Multiferroic NiI₂ Bilayers
- Büşra Gamze Arslan
Observation of Edge States on NiI₂/NbSe₂
- Thuy An Bui
2D metal-iodides in bilayer graphene encapsulation
- Nan Cao
Heisenberg S=1/2 Antiferromagnetic Molecular Chain
- Madhuri Chennur
Electrical impact of defect evolution on multi-layer WSe₂
- Aaron Dunbrack
Monolayer Moiré: Replacing Layers With Valleys In Graphene
- Jana Dzibelova
Atomic-resolution investigation of 2D hematene
- Linda Feuerstein
Nitrile Groups as Build-in Molecular Sensors for Interfacial Effects at Electrocatalytically Active Planar Carbon-Nitrogen Materials
- Akash Gupta
Single-electron charging on Au nanoparticles with Radio Frequency-excitation detected by pendulum Atomic Force Microscope
- Shuyu Huang
Moiré energy dissipation driven by nonlinear dynamics
- Mitisha Jain
Molecular Dynamics Simulations of He and Ar Ion Irradiation of 2D MoS₂ on Au and SiO₂ Substrates
- Umair Javed
Pore shape selection in hBN with electron beam induced chemical effects
- Netta Karjalainen
Hamiltonian parameter learning with artificial neural networks
- Clara Kofler
From four t(w)o three: How 4D-STEM measurements of 2D materials lead to 3D information
- Pekka Koskinen
The structure and stability trends in atomically thin metallenes
- Daniil Krukliniskii
Point and line defects in 2D and bulk CrSBr compounds from first-principles calculations
- Artem Kuklin
Stability Challenges in Predicted 2D Materials with Nonequivalent Sublattices: Topological Constraints and the Limits of PBC
- Lauri Kurki
Automated structure discovery for scanning tunnelling microscopy
- Greta Lupi
Hamiltonian learning quantum magnets with non-local impurity tomography
- Manuel Längle
Two-dimensional noble gas clusters in a graphene sandwich

- Anshika Mishra
Manipulation of Charge Defects in Monolayer NiBr₂
- Rabia Nawaz
Low energy ion irradiation of few-layer MoS₂
- Marcel Niedermeier
Quantum computing topological invariants of two-dimensional quantum matter
- Lorena Niggli
Dynamic heterogeneity in the self-induced spin glass in elemental neodymium
- Anastasiia Nihei
Heterostructures of 2D Materials from an Integrated Computational Approach
- Hermann Osterhage
Fock-Darwin states in artificial atomic structures
- Ram Prakash Pandeya
Molecular Order Induced Charge Transfer in a C60-Topological Insulator Moiré Heterostructure
- Diana Propst
Unraveling the highly defective regime of graphene and hexagonal boron nitride on the scale of atomic resolution: Automated image acquisition and analysis
- Nandhini Ravindran
Overcoming challenges in preparation of monolayer hBN and vdW heterostructure TEM samples
- Prosun Santra
Tailoring the electromechanical properties of two-dimensional materials via defect engineering
- Yubing Wang
Few layer plates & scrolls of the magnetic topological insulator MnBi₂Te₄
- Christoph Wilhelmer
First-principles investigations of noise in ultra-scaled 2D field effect transistors
- Nian Wu
Precise Large-Scale Chemical Transformations on Surfaces: Deep Learning Meets Scanning Probe Microscopy with Interpretability
- Walter Zuccolin
Single-layer boron phosphide on metallic surfaces: screening of promising substrates by first-principles
- Hans Ågren
Point and complex defects in single-layer transition metal dichalcogenides

Electric Field Control Of Moiré Skyrmion Phases in Twisted Multiferroic NiI2 Bilayers

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Twisted magnetic van der Waals materials provide a flexible platform to engineer unconventional magnetism. Here we demonstrate the emergence of electrically tunable topological moiré magnetism in twisted bilayers of the spin-spiral multiferroic NiI₂. We establish a rich phase diagram featuring uniform spiral phases, a variety of $k\pi$ -skyrmion lattices, and nematic spin textures ordered at the moiré scale. The emergence of these phases is driven by the local stacking and the resulting moiré modulated frustration. Notably, when the spin-spiral wavelength is commensurate with the moiré length scale by an integer k , multi-walled skyrmions become pinned to the moiré pattern. We show that the strong magnetoelectric coupling displayed by the moiré multiferroic allows the electric control of the $k\pi$ -skyrmion lattices by an out-of-plane electric field. Our results establish a highly tunable platform for skyrmionics based on twisted van der Waals multiferroics, potentially enabling a new generation of ultrathin topologically-protected spintronic devices.

Observation of Edge States on NiI₂/NbSe₂

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Topological superconductivity is an interesting phenomenon that may result from the interactions of superconductors with magnetic materials. It represents a quantum phase in the superconducting state that hosts stable edge modes, Majorana zero modes, against local disturbances. This robustness has led to the idea of using topological superconductors for fault-tolerant quantum computation. There are only a few examples of topological superconductors to date, and the need to explore material platforms to engineer topological superconductors with desirable properties remains. As topological superconductivity arises from the interaction of superconductors and magnets, stacked van der Waals materials are a highly promising platform to engineer topological phases in superconductors. Recently, multiferroicity of nickel diiodide (NiI₂) was shown down to monolayer limit. With its helical magnetic order and intrinsic spin-orbit coupling, NiI₂ is an excellent candidate to achieve topological superconductivity. In this work, our study on monolayer NiI₂ grown on superconducting bulk NbSe₂ is presented. This system is characterized using scanning tunneling microscopy STM and spectroscopy STS. The effect of doping due to the growth of NiI₂ on different substrates is investigated. Our observations revealed that NiI₂ does not show ferroelectricity, likely due to the charge transfer from the NbSe₂ substrate. A particular focus of this study was at the edges of the NiI₂ islands. We found clear signatures of edge modes in the NiI₂/NbSe₂ system. Whether these arise from topological effects remains to be seen in future studies. Our results show that the combination of 2D materials is a promising way to create designer materials with scientifically and technologically relevant properties.

2D metal-iodides in bilayer graphene encapsulation

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The first man-made 2D structures were those produced a hundred years ago by intercalating alkali and earth metals into graphite. The isolation of graphene sparked renewed interest to intercalation compounds that back then, for the first time, could be grown between only two layers of graphene in monolayer form. For electron microscopy images meant that an unobstructed atomic-level view to graphite intercalation compounds became a reality, turning a new page in studies of 2D structures and their self-assembly [1]. We have on our own right developed a graphene oxide intercalation technique wherein 2D metal-iodides of various kinds are synthesized in encapsulation. In our recent study it was demonstrated that for instance a high-temperature van der Waals layered phase of CuI can be stabilized as a monolayer under room temperature conditions by encapsulation [2]. In the present work we use the same approach and demonstrate the synthesis of an array of further 2D metal-iodides that include e.g. AgI, NiI₂, AuI₂, CoI₂, FeI₂ and MnI₂. These materials are visualized and characterized by atomically resolved scanning transmission electron microscopy (STEM) and electron energy loss spectroscopy (EELS).

[1] Krasheninnikov et al., *Nano Lett.* 2024, 24, 41, 12733-12740

[2] Mustonen et al., *Adv. Mater.* 34 (2022) 2106922

Heisenberg $S=1/2$ Antiferromagnetic Molecular Chain

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Magnetic exchange interactions between localized spins in π -electron magnetism of carbon-based nanostructures have attracted tremendous interest due to their promise for future quantum technologies. In particular, carbon-based spin systems offer an ideal platform for exploring many-body quantum phenomena, such as gapped excitations, strong spin entanglement, and fractionalized excitations. However, Heisenberg $S=1/2$ molecular chains with a given antiferromagnetic coupling remain unexplored, despite the flexibility they would offer in future spintronic devices. In this work, we report the realization of entangled antiferromagnetic quantum spin- $1/2$ Heisenberg model with diazahexabenzocoronene oligomers, obtained by surface-assisted homo-coupling on Au(111). Extensive low-temperature scanning tunneling microscopy/spectroscopy measurements on finite-size antiferromagnetic chains show that even-numbered spin chains exhibit a collective state with gapped excitations, while odd-numbered chains feature a Kondo excitation. Density functional theory and many-body calculations confirm the presence of an antiferromagnetic coupling J between nearest neighbours in the entangled quantum states, which is responsible for the observed quantum phenomena and demonstrates a strong dependence on the parities of the chain. These results provide a feasible way for the realization of the entangled quantum Heisenberg $S=1/2$ model. Moreover, this work presents opportunities for further studies on the quantum behaviour of many-body systems (e.g., topological spin lattices with fractional excitations).

Electrical impact of defect evolution on multi-layer WSe₂

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Nanoelectronics lays the groundwork for developing innovative, cost-effective, miniaturized, and versatile materials. Within this field, 2D materials present numerous opportunities for tailoring nanoscale functionalities. Notably, structural defects in 2D materials can significantly influence their properties by scattering charge carriers and reducing mobility. However, these imperfections do not necessarily degrade device performance; instead, by analyzing defect types, densities, and distributions, we can uncover insights and even utilize these imperfections to our advantage such as doping, tuning band gaps, or enhancing catalytic activity (1). In this work, we will investigate effects of defects on multi-layer WSe₂ induced by a single broad-beam irradiation dose of Helium ion at 7.5 keV. Due to the role of substrate in the defect formation(2), Si/SiO₂ and hBN are employed as a charge-trapping bottom layer. Mechanical exfoliation technique is utilized as it ensures the quality of the material under investigation. All measurements in this study are conducted under ambient conditions, therefore changes of the defect states after irradiation are studied. Defect evolution is monitored one week and two weeks post-irradiation. Electrical contacts were fabricated by electron beam lithography (EBL) and different metals were implemented to compare the electrical compatibility with the 2D material. The devices exhibited intriguing behavior, showing detrimental characteristics after one week, but after two weeks, the current increased beyond the levels observed prior to irradiation. Further, Raman measurements were performed before/after ion irradiation to gain a deeper understanding of their fundamental behavior. References: 1) Wang, Dan, et al. *Nano Today* 16 (2017): 30-45.2) Fekri, *Advanced Electronic Materials*: 2400037.3) Telkhozhayeva, Madina, and Olga Girshevitz. *Advanced Functional Materials* (2024): 2404615

Monolayer Moiré: Replacing Layers With Valleys In Graphene

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In the past decade, moire materials have served as a new platform for strongly-interacting physics, particularly twisted bilayer graphene (TBG). One interesting limit of TBG which renders analytic methods tractable is the "chiral limit," wherein one neglects same-sublattice interlayer hopping and thereby finds exactly flat bands at the noninteracting level. In PRB 108, 075126, we consider another model with very similar behavior: on a single layer of graphene, add a potential with period larger than graphene by a factor of $(1+\epsilon)\sqrt{3}$ and rotated 90 degrees. At $\epsilon=0$, this results in a commensurate matching that folds the Dirac cones at K and K' on top of each other. For small but nonzero ϵ , this instead moire-couples the two valleys, replicating the chiral model of TBG but with valley substituted for layer. In this talk I will present this model, describe its similarities and differences from chiral TBG, characterize the interacting physics at integer filling, and discuss realistic implementations.

Atomic-resolution investigation of 2D hematene

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2D materials have attracted scientific research for decades, since they possess distinct physico-chemical properties compared to their 3D counterparts due to quantum confinement. These atomically thin structures such as graphene, hBN, etc. are typically based on so-called van der Waals systems. These 3D counterparts are defined by strong bonds oriented in-plane and solely weak bonding interaction between the layers. However, 2D materials can also be created from a parent material with strong bonding interactions in all three directions. For these, it is significantly more complicated to cleave them into individual layers. After cleavage into atomically thin layer, the non-van der Waals 2D materials could offer a playground to explore the changes in properties induced by dimensionality restriction. Recently, a new member of these non-van der Waals 2D materials has been introduced: hematene, the 2D form of α -Fe₂O₃ [1, 2]. The dimensional confinement of hematene structure is expected to introduce changes to the lattice [3]. This distortion of interatomic positions affects electronic configuration and therefore the magnetic, electronic and optical behavior of the system [4]. Here, we employ transmission electron microscopy and selected area electron diffraction to investigate the atomic structure and distortions of the lattice in thin hematene sheets with the aim to quantify the lattice distortions to facilitate the physical description of the material. References

[1] Puthirath Balan, A., et al., *Nature Nanotech.*, 13, 602, (2018), <https://doi-org/10.1038/s41565-018-0134-y>

[2] Dzibelová, J., et. al. *Appl. Mater. Today*, 34, 101881, (2023), <https://doi.org/10.1016/j.apmt.2023.101881>

[3] Friedrich, R. et al., *Nano Lett.*, 22, 989, (2022), <https://doi-org.uaccess.univie.ac.at/10>

[4] S. Maiti, K. et al., *Energy Environ. Sci.* 14, 3717, (2021) <https://doi.org/10.1039/D1E>

Nitrile Groups as Build-in Molecular Sensors for Interfacial Effects at Electrocatalytically Active Planar Carbon-Nitrogen Materials

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Planar carbon-based materials provide large surface areas and are composed solely of abundant elements, making them suitable to be used as electrocatalysts in future energy conversion technologies. Interfacial effects at the materials' surface determine the mechanistic cycle and include electric field effects, inductive effects and non-covalent interaction with the electrolyte, all of which significantly influence the efficiency of the overall electrocatalytic process. These effects can be characterized based on the vibrational Stark effect, which arises from the interaction between a local electric field and a vibrational mode of a surface-bound molecular group and can be measured by spectro-electrochemical methods such as in-situ Raman spectroscopy. In this work, we investigate a planar carbon material of C₂N stoichiometry that shows activity for the hydrogen evolution reaction (HER). Using Raman spectroscopy and first principle simulation, it is demonstrated that build-in nitrile groups display a defect character and act as Stark reporter groups. Specifically, the $\nu(\text{C}\equiv\text{N})$ vibrational mode at 2235 cm⁻¹ exhibits a linear shift in response to the applied electric voltage, according to the vibrational Stark effect under non-catalytic conditions in a KCl electrolyte. Under HER conditions in HCl electrolyte, deviations from the linear shift indicate rearrangement processes and a change of the local electric field within the electrochemical double layer, implying a semi-permanent conditioning of the electrode's surface due to the hydrogen evolution process.

Single-electron charging on Au nanoparticles with Radio Frequency-excitation detected by pendulum Atomic Force Microscope

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Single-electron charging has been keenly observed with the Atomic Force Microscope, wherein the oscillating tip is capacitively coupled to quantum dot-like nanostructures. This facilitates the gating of a single charge onto these nanostructures from a conducting back electrode reservoir [1,2]. This phenomenon can be observed as a huge increase of the cantilever energy dissipation. We studied Au nano-clusters grown onto Cu(100) surface, separated by thin layers of NaCl. Measurements are conducted at low temperatures (77K/5K) and ultra high vacuum, using a pendulum Atomic Force Microscope (pAFM) [3]. The pAFM uses an extremely soft cantilever with a high-quality factor, enabling it to be highly sensitive to dissipation signal. We imaged nanostructures with the Kelvin Probe Force Microscope (KPFM). The single electron charging of Au clusters was spatially resolved as Coulomb blockade rings through constant-height dissipation maps. Furthermore, an external radio frequency field of few GHz has been used in order to modify the charge state of the quantum dot-like nanostructures. The application of RF field introduces a novel element to our experimental approach, offering a nuanced control mechanism over the charge dynamics within the nanostructure systems. References:

[1] Stomp, Romain, et al. "Detection of single-electron charging in an individual InAs quantum dot by noncontact atomic-force microscopy." *Physical Review Letters* 94.5, 056802 (2005).

[2] Tekiel, Antoni, et al. "Room-temperature single-electron charging detected by electrostatic force microscopy." *ACS Nano*, 7.5, 4683 (2013).

[3] Gysin, U., et al. "Low temperature ultrahigh vacuum noncontact atomic force microscope in the pendulum geometry." *Review of Scientific Instruments*, 82.2 (2011).

Moiré energy dissipation driven by nonlinear dynamics

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The moiré superlattice in twisted van der Waals heterostructures is of central importance for the modulation of the electronic and optical properties of the system, yet the mechanical dissipation of such moiré systems remains largely unexplored. Here, we report the experimental observations of energy dissipation across both vertical and lateral directions along the moiré superstructures, revealing a significant increase in dissipation at moiré ridges compared to flat domains. Comparison of the measurements with a theoretical phononic dissipation model suggests that the local increase in energy dissipation originates from nonlinear instability dynamics of the moiré superstructure. Criteria for such moiré energy dissipation are established, which are expected to be broadly applicable to other van der Waals heterostructures. Our results extend the understanding of mechanical energy loss in moiré systems and pave the way for the rational design of new slidtronic and twisttronic devices and nanoelectromechanical systems in general.

Molecular Dynamics Simulations of He and Ar Ion Irradiation of 2D MoS₂ on Au and SiO₂ Substrates

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Using first-principles and analytical potential molecular dynamics, we study the response of 2D MoS₂ to electron and ion irradiation. We demonstrated how adsorbed carbon adatoms on MoS₂ can give rise to the formation of sulfur vacancies under electron beam with electron energies as low as 20 keV[1]. We further compared the S displacement cross-section, calculated with the inclusion of C adatoms, to the experimental data on S vacancies collected after electron irradiation. By incorporating a parameter to represent C adatom concentration into our model, we found a good agreement between the theoretical predictions and experimental results. With regard to ion irradiation, this study extends previous work on the formation of defect in 2D MoS₂ under electron beam by examining the effects of different substrates, specifically SiO₂ and Au, on defect production. We have found that depending on ion type and energy, the substrate can have a very strong or negligible effect on defect creation. In particular, SiO₂ substrate has minimal effect when He ions with 20eV-40 keV energies impact the system. Similarly, the presence of Au substrate has little influence on the sputtering yield of S/Mo atoms upon He irradiation. At the same time, we found an increased number of single S vacancies when the Au substrate is present for He ions with energies in the 100eV-40 keV range. For Ar ions (<6 keV), SiO₂ substrate effectively suppresses the defect production in MoS₂, while for the Au substrate, the reduction in sputtering yield from mono-layer was found for Ar ion with energies <800 eV. These results reveal that SiO₂ provides a stabilizing effect on MoS₂ during ion irradiation, with less defect formation compared to the Au substrate. These insights can help in the optimization of irradiation parameters for nano-patterning of 2D materials and tuning of their electronic properties, providing a foundation for precise defect engineering of 2D materials.

[1] M. Jain et al., PRM 8, 054004(2024)

Pore shape selection in hBN with electron beam induced chemical effects

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It is well-established that electron irradiation of hexagonal boron nitride (hBN) during transmission electron microscopy even at energies lower than 80 keV leads to the formation of triangular pores with nitrogen-terminated edges[1-4]. The cause of ejection of boron is suggested to be the combination of knock-on damage[2] radiolysis[3] and chemical etching due to the residual gases[4]. However, neither of these mechanisms has been conclusively shown to be the main cause. We resolve this by carrying out experiments with different acceleration voltages at different low-pressure atmospheres. At 1e-10 mbar, both 60 and 80 kV irradiation leads to round pores with no preference to either boron or nitrogen edges. From this data we estimate the displacement cross section at the pore edge to be roughly two orders of magnitude higher than in pristine material [5]. Experiments in nitrogen atmosphere show no difference. However, at oxygen partial pressure of ca. 1e-9 mbar, growth accelerates by ca. a factor of three, the shapes remain circular. But, at ca. 1e-8 mbar, the pores turn into triangles and the growth further spurs. This indicates, at higher pressures, the pore growth is dominated by a beam-assisted chemical process rather than direct electron-beam-damage. To ascertain this, we carry out experiment at 1e-9 mbar with a smaller field-of-view in an area already containing pores. The pores inside the imaged area remain circular, whereas those outside turn into triangles. Based on our ab initio simulations, oxygen atoms that are created from O₂ by the electron beam attach preferentially on B at pore edges. From this configuration, it is easier to remove the O and B together rather than just O. This explains the preference for nitrogen edges, and therefore, the triangles, under oxygen atmosphere.

[1] Jin et al, Phys. Rev. Lett. 102(2009)

[2] Kotakoski et al, Phys. Rev. B 82(2010)

[3] Cretu et al, Micron 72(2015)

[4] Gilbert et al, Sci Rep 7(2017)

[5] Bui et al, Small, 230192 (2023)

Hamiltonian parameter learning with artificial neural networks

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One-dimensional antisymmetric spin chains offer a versatile platform for studying realistic spin systems and have possible applications in spintronics and signal processing. Engineering spin models at the atomic scale using scanning tunneling microscopy, combined with local imaging of excitations via electrically driven spin resonance, offers a powerful method to visualize spin excitations. However, extracting the parameters of the many-body spin Hamiltonian is experimentally challenging, as this process must be performed indirectly. Usually, these indirect measurements either lack accuracy or become infeasible as the system size increases. Therefore, here we use supervised learning to infer the underlying Hamiltonian parameters via dynamical spin excitations. Here, we focus on $S=1/2$ spin chains, realized by hydrogenated Ti atoms in MgO films. We demonstrate a machine-learning algorithm to predict the Heisenberg exchange interactions J_1, J_2, J_3 and anisotropic exchange J_z , as well as antisymmetric Dzyaloshinskii-Moriya interaction JDMI for a one-dimensional many-body spin chain on the surface. We trained an artificial neural network with DMRG-simulated spin spectral functions and corresponding exchange parameters. We demonstrate that the algorithm can extract the different parameters of the Hamiltonian by providing the spatially dependent and frequency-dependent local excitations. These excitations can be directly measured via electrically driven spin resonance scanning tunneling microscopy. The algorithm is robust to noise in the dynamical spin excitations, offering an experimentally realistic strategy for Hamiltonian learning. Once trained, it can instantly extract the parameters from the measured spectral function. This methodology proposes confined excitations in quantum magnets as a powerful strategy for understanding the nature of quantum spin many-body models.

From four t(w)o three: How 4D-STEM measurements of 2D materials lead to 3D information

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While scanning transmission electron microscopy (STEM) can routinely resolve structures down to individual atoms, it is typically limited to 2D projections. However, measuring the 3D structure is often necessary to fully understand material properties, even for 2D materials such as graphene [1]. This becomes still more important when multiple 2D layers are combined. In this contribution, we present promising methods for measuring the 3D structure of moiré materials such as twisted bilayer graphene (tBLG). These exhibit a twist-angle dependent superstructure that influences several properties, including the interlayer distance [2]. All presented methods are based on four-dimensional STEM where the full diffraction pattern is available at each scan position. For direct measurement of the interlayer distance defocused Bragg interferometry shows promise. Here, the intensity distribution in overlapping regions of diffraction disks originating from separate material layers is studied to draw conclusions about the structure. The interference fringes in these regions are sensitive to out-of-plane modulations and both their phase [3] and rotation [4] have been used to determine average interlayer spacings. Another approach exploits the sensitivity of center of mass measurements to the local stacking order to verify 3D models by comparing experimental data with simulations [5]. By understanding, adapting, and further developing these methods and combining them with simulations we aim to create a robust workflow that can not only provide average values but also map the interlayer distance of tBLG as a function of the position in the moiré superstructure.

[1] Meyer et al., *Nature* 446, 60 (2007)

[2] Uchida et al., *Phys. Rev. B* 90, 155451 (2014)

[3] Latychevskaia et al., *Ultramicroscopy* 219, 113020 (2020)

[4] Zachman et al., *Small* 17, 2100388 (2021)

[5] Argentero et al., *Nano Lett.* 17, 1409 (2017)

The structure and stability trends in atomically thin metallenes

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Metallenes are atomically thin two-dimensional (2D) materials lacking a layered structure in 3D bulk. They can be stabilized by nanoscale constrictions like pores in 2D covalent templates, but the isotropic metallic bonding makes their stabilization difficult. A few metallenes have been stabilized experimentally, but their research is still largely based on computations. Here, we use density-functional theory to investigate the structure and stability of 45 different metallenes in six lattices and varying atomic densities. In this presentation, we discuss metallenes' basic properties in terms of energetics, dynamic stabilities, edge energies, edge stresses, physisorption, and basic electronic structure properties. The trends extracted from such systematic computations help identify the most promising metallenes for 2D stability and support the development of new strategies to synthesize larger and more stable metallene samples for plasmonic, optical, and catalytic applications.

Point and line defects in 2D and bulk CrSBr compounds from first-principles calculations

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D. Kruklynskii, M. Ghorbani-Asl, and A.V. Krasheninnikov Institute of Ion Beam Physics and Materials Research, Helmholtz-Zentrum Dresden-Rossendorf, 01328 Dresden, Germany The 2D CrSBr compound and its van der Waals layered counterpart have attracted a considerable amount of attention due to their peculiar electronic and magnetic properties [1,2]. However, the effects of defects on their characteristics have not yet been fully understood. At the same time, ion and electron irradiation has been experimentally demonstrated to be an effective tool to tune the properties of this material [3]. We use first-principles calculations to get insights into the structure and properties of defective CrSBr. We also study the production of defects in this system under electron irradiation by evaluating the displacement thresholds of atoms and calculating the displacement cross sections as functions of electron energy within the framework of the McKinley-Feshbach formalism. We further investigate the electronic properties of finite-width ribbons carved from CrSBr sheets. Our results should facilitate the defect-mediated engineering of the properties of CrSBr systems. References [1] N. Wilson, et al., Nat. Mater. 20 (2021) 1657. [2] E. Telford, et al., Nat. Mater. 21 (2022) 754. [3] F. Long, et al., Nano Lett. 23 (2023) 8468.

Stability Challenges in Predicted 2D Materials with Nonequivalent Sublattices: Topological Constraints and the Limits of PBC

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Two-dimensional materials are in the limelight of current nano research. They attract a lot of attention due to unique properties that could influence future technologies. Computational discovery of new two-dimensional materials has become a significant part of modern materials research in order to accelerate this area. The stability of these predicted materials becomes a new issue due to drawbacks of the periodic boundary condition (PBC) approximation that allows the passing of commonly used criteria of stability (like phonon and MD simulations). I will present our recent study devoted to the stability of a number of recently predicted 2D materials with nonequivalent sublattices. I will demonstrate that nonequivalent sublattices can lead to topological instability due to uncompensated structural stresses which can be artificially stabilized by PBC. Topology conservation theorem (TCT) will be introduced and it will be demonstrated that the lack of perfect filling of planar 2D crystalline space by structural units may cause the formation of structure waves, nanotubes, or rolls, saddle structures, aperiodic ensembles of irregular asymmetric atomic clusters. It will be proven that phonon dispersion and molecular dynamic calculations cannot be used as solid and final proof of either stability or metastability of predicted 2D materials with nonequivalent sublattices. It will be also shown that finite-size structural fragments can be stabilized and synthesized, while aromatic resonance and surface effect plays also an important role in the stabilization of 2D materials.

Automated structure discovery for scanning tunnelling microscopy

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Scanning tunnelling microscopy (STM) with a CO molecule on the probe apex captures sub-molecular level detail of the electronic and physical structures of a sample [1]. The resolution of the images is very high but the produced images are often difficult to interpret to reveal the atomic structure. To accelerate the analysis, we propose automated machine learning image interpretation tools to extract sample properties directly from bond-resolved STM images [2]. While our direct application is designed for characterising individual molecules on the surface, the methods presented can be applied for 2D material characterization. In recent years, there has been rapid development in image analysis methods using machine learning with particular impact in medical imaging. These concepts have been proven effective also in SPM in general and especially for extracting sample properties from AFM images [3,4,5]. We build upon these models and show that we can extract atomic positions directly from STM images. In particular, we employ rotation equivariant machine learning to improve robustness and data efficiency of the models. Finally, we test the chemical sensitivity of the method by predicting chemical compositions of some organic molecules.

[1] Cai, S., Kurki, L., Xu, C., Foster, A. S., Liljeroth, P., *J. Am. Chem. Soc.* 144, 20227–20231 (2022).

[2] Kurki, L., Oinonen, N., Foster, A.S., *ACS Nano*, 18, 17, 11130–11138 (2024).

[3] Alldritt, B., Hapala, P., Oinonen, N., Urtev, F., Krejci, O., Canova, F. F., Kannala, J., Schulz, F., Liljeroth, P., Foster, A. S., *Sci. Adv.* 6, eaay6913 (2020).

[4] Carracedo-Cosme, J., Perez, R., *npj Comput. Mater.* 10, 19 (2024).

[5] Oinonen, N., Kurki, L., Ilin, A., Foster, A.S., *MRS Bulletin* 47, 895–905 (2022).

Atomic-scale quantum magnets have gained significant attention due to their potential

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applications in quantum information processing and spintronics. Exploiting advancements in scanning tunneling microscopy (STM), it is now possible to measure and manipulate these systems with atomic precision. This project explores the application of supervised machine learning techniques to infer Hamiltonian parameters from STM data using non-local impurity tomography. We focus on two prototypical systems: a fermionic chain and a spin- $\frac{1}{2}$ chain, using the fermionic data as a benchmark to validate our approach for the spin chain. By training the NNs with theoretical datasets and subsequently fine-tuning them with noisy data, we aim to develop robust models capable of accurately predicting Hamiltonian parameters from experimental measurements. Our results demonstrate the efficacy of this approach in providing insights into the underlying interactions of quantum systems.

Two-dimensional noble gas clusters in a graphene sandwich

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The van der Waals atomic solids of noble gases on metals at cryogenic temperatures were the first experimental examples of two-dimensional systems [1]. Recently, such structures have also been created on surfaces under encapsulation by graphene, allowing studies at elevated temperatures through scanning tunneling microscopy [2,3]. However, for this technique, the encapsulation layer often obscures the arrangement of the noble gas atoms. Here we create Kr and Xe clusters between two suspended graphene layers by irradiation with singly charged low-energy ions. We uncovered their atomic structure through transmission electron microscopy [4]. We show that small crystals ($N < 13$) mostly arrange based on the simple non-directional van der Waals interaction, however, four- and five-atom clusters also show directional square-like bonding. Larger crystals also show deviations, possibly enabled by deformations in the encapsulating graphene lattice. We further discuss the dynamics of the clusters within the graphene sandwich and show that although all the Xe clusters with up to $N \approx 100$ remain solid, Kr clusters with already $N \approx 16$ turn occasionally fluid under our experimental conditions (under a pressure of 0.3 GPa). This study opens a way for the so-far unexplored frontier of encapsulated two-dimensional van der Waals solids with exciting possibilities for fundamental condensed-matter physics research.

[1] Jortner, J. et al., *J. Chem. Phys.* 42, 4250-4253 (1965).

[2] Herbig, C. et al., *2D Mater.* 3, 025032 (2016).

[3] Valerius, P. et al., *Phys. Rev. B* 96, 235410 (2017).

[4] Längle, M. et al., *Nat. Mater.* 23, 762-767 (2024).

Manipulation of Charge Defects in Monolayer NiBr₂

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Polarons are composite quasiparticles formed when an excess charge carrier becomes trapped by a local lattice distortion. Recent studies have revealed the presence of polarons in the transition metal dihalide CoCl₂, where they can be created, manipulated, and erased. This discovery opens up new avenues for studying polarons in van der Waals (vdW) materials, such as transition metal dihalides. We have identified similar polaron-like defects in monolayer NiBr₂, grown on a Highly Oriented Pyrolytic Graphite (HOPG) substrate via Molecular Beam Epitaxy (MBE). Using Scanning Tunneling Microscopy (STM) and Scanning Tunneling Spectroscopy (STS), we visualize the defects and characterize their electronic structure. Similar to observations in CoCl₂, we demonstrate their creation, erasure, and movement in NiBr₂. Additionally, STS measurements reveal a shift in the conduction band of NiBr₂ due to defects formed during the growth process. This is particularly intriguing because NiBr₂ exhibits multiferroic properties similar to those of NiI₂, and it would be interesting to study the interplay between polarons and multiferroicity. This work paves the way for future investigations into the manipulation of polarons in 2D materials, opening new possibilities for novel material designs and functionalities.

Low energy ion irradiation of few-layer MoS₂

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The development of functional 2D nanomaterials is critical for advancing the next generation of electronic and optoelectronic devices. This study focuses on modifying freestanding MoS₂ layers through ultra-low-energy argon and xenon ion irradiation. We investigate the structural changes induced by the ions targeting the van der Waals gap between the individual MoS₂ sheets as well as changes in the individual layers. Using the KIIA ion accelerator at the University of Helsinki, we irradiate few-layer MoS₂ structures with the ions, applying the various doses around 1×10^{14} – 1×10^{15} /cm² at room temperature. The changes in the structure vary from defects in the top layer to trapping of the ions in the interlayer gap, to pores and are controlled by the ion energy. We conducted a high-resolution scanning transmission electron microscopy (STEM) analysis using a Nion UltraSTEM 100 at the University of Vienna to investigate the effects further. This revealed interesting changes in the stacking order, as the initial AB stacking transformed into complex structures due to changes in the top layer like some areas seemed to have transformed into AA and AC stacking, with domain sizes sub-nanometer. Additionally, we use molecular dynamics (MD) to explore the effects of low-energy Ar on the MoS₂ bilayered system at the atomic level. Preliminary results show shallow damage at 20 and 30 eV ion energies, inducing point defects and small lattice distortions on the first layer. This effect is more noticeable at 40–50 eV, where Ar atoms penetrate the first of the two layers of MoS₂, get trapped in the gap, and create larger defects, e.g., pores. The stacking transformations may open interesting possibilities for strain engineering and band structure manipulation arising from MoS₂'s semiconducting nature, as well as changes in the surface's catalytic activity due to the formation of grain boundaries.

Quantum computing topological invariants of two-dimensional quantum matter

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Quantum algorithms provide a potential strategy for solving computational problems that are intractable by classical means. Computing the topological invariants of topological matter is one central problem in research on quantum materials, and a variety of numerical approaches for this purpose have been developed. However, the complexity of quantum many-body Hamiltonians makes calculations of topological invariants challenging for interacting systems. Here, we present two quantum circuits for calculating Chern numbers of two-dimensional quantum matter on quantum computers. Both circuits combine a gate-based adiabatic time-evolution over the discretized Brillouin zone with particular phase estimation techniques. The first algorithm uses many qubits, and we analyze it using a tensor-network simulator of quantum circuits. The second circuit uses fewer qubits, and we implement it experimentally on a quantum computer based on superconducting qubits. Our results establish a method for computing topological invariants with quantum circuits, taking a step towards characterizing interacting topological quantum matter using quantum computers.

Dynamic heterogeneity in the self-induced spin glass in elemental neodymium

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Spin glasses are a puzzling form of magnetic matter possessing seemingly random spins that continuously evolve, i.e. called aging dynamics [1]. Yet, it has been shown that spin glasses simultaneously exhibit memory and rejuvenation initiating a debate about the role of local length scales [2]. Much of what is known experimentally about spin glasses is based on ensemble thermodynamic measurements, hence there is a demand for experimental methods, which can study magnetization on the nanoscale. In contrast, the presence of spatially heterogeneous dynamics has been established in structural glasses ranging from polymers to colloids. This so-called dynamic heterogeneity refers to a spatial coexistence of frozen and active regions, where relaxation times among regions can vary by orders of magnitude. Recently, we discovered that the low temperature magnetic phase of elemental neodymium behaves like a proposed self-induced spin glass relying solely on spin frustration in the absence of disorder [3, 4]. Nd(0001) is characterized by the coexistence of locally ordered magnetic patterns in real space. Here, we explore its aging dynamics using spin-polarized scanning tunneling microscopy as a function of magnetic field and temperature. We observe that zero-field cooling freezes a specific set of metastable states into the spin glass and find a structured evolution towards a distinct set of states during aging dynamics. Spatially resolved magnetization dynamics reveal a coexistence of slow and fast dynamics providing evidence for the first experimental observation of dynamic heterogeneity in a spin glass system, thereby providing a natural bridge between the description of spin glasses and its non-magnetic counterparts.

[1] Binder et al., *Rev. Mod. Phys.* 58, 801-976 (1986)

[2] Lefloch et al., *EPL* 18, 647 (1992)

[3] Kamber et al., *Science* 368, 6494 (2020)

[4] Verlhac et al., *Nature Physics* 18, 905-911 (2022)

Heterostructures of 2D Materials from an Integrated Computational Approach

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Heterostructure interfaces of two-dimensional (2D) materials enable the realization of advanced electronic functionalities at the nanoscale. The efficient computational ab initio modelling of these systems is, however, a challenge as it requires proper lattice matching of the component 2D sheets with minimal strain. This often results in large structures with hundreds to thousands of atoms. Here, we utilize the newly developed Hetbuilder tool to automate the heterostructure cell construction based on coincidence lattice theory [1,2]. It is integrated with the AFLOW materials database and software [3,4] allowing for an efficient workflow for the structure generation from the bulk parent systems. We benchmark the approach by reproducing a wide range of favorable theoretically and experimentally studied 2D heterostructures predicted by Koda et al. [1]. We also perform ab initio calculations to study magnetic and electronic properties of heterostructures in a strong interaction regime derived from novel 2D materials. The ionic and covalent bonds at the interface give rise to a significant chemical coupling that activates substantial magnetic interactions and modulated electronic properties for specific materials [5].

[1] D. S. Koda et al., *J. Phys. Chem. C* 120, 10895 (2016).

[2] <https://zenodo.org/record/4721346>.

[3] M. Esters et al., *Comput. Mater. Sci.* 216, 111808 (2023).

[4] C. Oses et al., *Comput. Mater. Sci.* 217, 111889 (2023).

[5] A. Nihei et al., manuscript in preparation (2024).

Fock-Darwin states in artificial atomic structures

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Quantum simulators are a pathway to study novel physical phenomena which are difficult to predict or observe in synthesized materials. To this end, the physical behavior of materials has been used to emulate model Hamiltonians [1,2]. The most iconic of which is the Hubbard model, where previously unobserved phenomena were seen [3]. To date, there is still a lack of viable platforms for quantum simulation to study confined electrons in strong magnetic fields, including controlling orbital and lattice symmetries as well as the long-range nature of the coupling. With such a platform, it would be possible to study the so-called Hofstadter limit, which requires magnetic fields that induce electron orbits on the same length scale as the periodicity of the lattice in question. For typical crystals, this corresponds to field strengths that are unattainable in a laboratory. Our approach to quantum simulation is based on patterning Cs atoms on the semiconducting surface of InSb [4]. We begin by arranging Cs atoms on the surface with scanning tunneling microscopy, sculpting confinement potentials on the 2DEG which act as artificial atoms (i.e. quantum dots). I will show the response of these artificial atoms to strong magnetic fields, where they exhibit Fock-Darwin states [5]. Furthermore, I will show how these artificial atoms can be coupled into molecular structures and the response of the resultant electronic structure to these magnetic fields. I will link this to the Hofstadter picture and further comment on perspectives to use this platform to study the role of spin-orbit coupling and topology.

[1] Bloch et al., Nat. Phys. 8, 267-276 (2012)

[2] Houck et al., Nat. Phys. 8, 292-299 (2012)

[3] Dehollain et al, Nature, 579, 528 (2020)

[4] Sierda et al, Science 380, 1048 (2023)

[5] Kouwenhoven et al., Rep. Prog. Phys. 64, 6 (2001)

Molecular Order Induced Charge Transfer in a C60-Topological Insulator Moiré Heterostructure

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Buckminsterfullerene (C60) has extensively been studied due to its various exotic electronic and magnetic properties which range from semiconductors in the pristine phase to metals or Mott insulators and even superconductors when C60 is doped by alkali atoms. Here, we study the electronic structure of a highly ordered ultrathin fullerene film (1ML C60) deposited on the topological insulator Bi4Te3 using ARPES, Raman, PL, and DFT methods. LEED analysis of the deposited film confirms (4X4) reconstruction of the C60 on a (9X9) supercell of the Bi4Te3 surface. The Fermi-surface mapping of the topologically protected surface state confirms a strong hexagonal warping deviating from the typical linearly dispersive Dirac bands. While we observe a hole doping to the TI with C60 deposition at room temperature as rigid shifting of the Dirac point, no charge transfer at low temperature is observed. The estimated hole doping at room temperature is 0.03 holes per C60 molecule. Photon polarization dependence reveals a complex band-dispersion of the HOMO and HOMO-1 molecular orbitals of C60. Temperature-dependent resonance Raman spectroscopy of the C60 pentagon pinch reveals the molecular ordering of the C60 thin film below 250K which is reminiscent of the structural transition in bulk C60. A significant change in the PL response of 1ML C60 film at low temperatures further confirms the molecular ordering in the ground state. Ab initio calculations of the reconstructed heterostructure suggest electron-doping of the C60 molecules due to tri-vacancy of the Te-terminated surface. Simulations performed for different molecular ordering reveals low electron affinity in the ground state affecting the charge transfer at low temperatures. Our work highlights that TI surfaces are excellent substrates for the growth of highly ordered layers of fullerenes. The work shown here also paves the way for further experiments using magnetic fullerenes and superconducting C60 films grown on TI.

Unraveling the highly defective regime of graphene and hexagonal boron nitride on the scale of atomic resolution: Automated image acquisition and analysis

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Defect-engineered 2D materials have become a highly active topic of study in the research community due to their unique and promising properties in comparison to the pristine material and many of the relevant defects are found on the atomic scale. The characterization of such defects requires stringent conditions, where it becomes necessary to probe large areas of the material with atomic resolution. A full realization of this goal is presented in this work, demonstrated on defect engineered graphene and hexagonal boron nitride (h-BN), where the induced defectiveness ranges from non-defective to the highly defective regime. Atomic resolution images are acquired using scanning transmission electron microscopy (STEM) annular dark field (ADF) imaging. The imaging process is automated, to reduce the time cost of the acquisition of datasets sufficiently large to represent the underlying statistics accurately, but also to reduce the observer bias when selecting images. This yields a large number of images, hundreds to thousands, which require their own automatic analysis in turn. Therefore, we use a convolutional neural network approach to automatically extract defect structures and their characterization from STEM ADF images, testing the scope of applicability of this approach on image sets of varying induced defectiveness. With this approach, the highest defect regime of graphene and h-BN have been characterized, illuminating their markedly different behaviors: Highly defective graphene becomes amorphous, while hBN does not, only leading to increased pore sizes and pore density, but otherwise showing crystalline regions. This approach is discussed in detail, showing its capabilities as a versatile characterization tool and illustrating some of the remaining shortcomings.

Overcoming challenges in preparation of monolayer hBN and vdW heterostructure TEM samples

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Understanding ion interaction with free-standing 2D materials such as hexagonal boron nitride and its combinations in van der Waals heterostructures opens a path to atomic-scale structural modification of materials with precision and high efficiency [1]. However, the preparation and transfer of this material onto transmission electron microscopy sample grids (TEM) is challenging due to poor coverage and heavy contamination, and unlike for graphene, no vendor offers grids with hBN. We have developed a method of sample preparation that is helpful for minimising sample contamination. We transfer commercial monolayer hBN grown on Cu foil onto gold Quantifoil grids using a chemical etching method [2]. Optimization of transfer parameters was required for uniform adhesion of single-layer hBN onto the grid, followed by the etching of Cu foil using FeCl₃ solution. To minimize contamination due to sample transfer, we optimized the durations of etching in the chemical bath followed by washing and drying the sample in distilled water and isopropyl alcohol. Additional contamination during transfer to the microscope or the ion-beam facility is avoided by use of our ultra-high vacuum (UHV) system and portable UHV suitcase [3]. Samples are further annealed at a heating stage connected to the UHV system to remove remaining surface contamination [4] before imaging using aberration-corrected scanning transmission electron microscopy (Nion UltraSTEM100). 1. Schwestka, J. et al., ACS Nano, 2020. 14(8): p. 10536-10543. 2. Bui, T.A. et al., Small, 2023. 19(39): p. 2301926. 3. Mangler, C. et al., Microscopy and Microanalysis, 2022. 28(S1): p. 2940-2942. 4. Tripathi, M. et al., physica status solidi (RRL), 2017. 11(8): p. 1700124.

Tailoring the electromechanical properties of two-dimensional materials via defect engineering

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We have investigated the response of four archetypal 2D materials—h-BN, graphene, MoSe₂, and phosphorene—to external strain, specifically focusing on systems containing substitutional impurities, using first-principles calculations. Our results indicate that the formation energy of defects can either increase or decrease under bi-axial tensile strain, depending on whether the atomic radius of the impurity atom is larger or smaller than that of the host atom [1]. Analysis of the strain maps shows that this behavior is linked to the compressive or tensile local strains introduced by the impurities, which interact with the applied external strain. Furthermore, we examined the impact of point defects, including substitutional impurities and vacancies, on the mechanical properties of h-BN and graphene. Notably, we found that at low concentrations (a few atomic percent), specific defects can counterintuitively enhance the Young's modulus of these materials [2]. This enhancement arises from defect-mediated strain fields around the impurities; when tensile strain near defects dominates, it increases the elastic energy under applied tensile strain, thereby strengthening the material. Conversely, predominantly compressive local strain induced by defects has the opposite effect. Our findings present new possibilities for combined strain and defect engineering to precisely tune the electromechanical properties of 2D materials. References:

[1] P. Santra, S. Ghaderzadeh, M. Ghorbani-Asl, H-P. Komsa, E. Besley, A.V. Krasheninnikov, *npj 2D Mater. Appl.* 8, 33 (2024).

[2] P. Santra, M. Ghorbani-Asl, S. Ghaderzadeh, E. Besley, A.V. Krasheninnikov, submitted (2024).

Few layer plates & scrolls of the magnetic topological insulator MnBi₂Te₄

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Yubing Wang, Daniel Wolf, Nicolas Perez Rodriguez, Thomas Brumme, Oliver Dreimann, Axel Lubk, Michael Ruck Magnetic topological insulators are intensively investigated for their potential application in quantum computing and spintronics. The currently most famous material of this kind is MnBi₂Te₄, a layered van der Waals compound that consists of magnetic septuple layers [Te-Bi-Te-Mn-Te-Bi-Te]. The surface of these materials is of special interest, since the topological and also other physical effects manifest there or originate from it. Moreover, the antiferromagnetic coupling of the layers makes a difference between few-layer 2D crystals of MnBi₂Te₄ with odd or even number of layers. Up to now, all investigations were performed on mechanically exfoliated material. We now developed an up-scalable electrochemical method for liquid-phase exfoliation. We succeeded in preparing thin flakes with thickness down to few nanometers, while lateral dimensions reach up to a hundred micrometers. Unexpectedly, we also obtained nanoscrolls of MnBi₂Te₄, which is an unprecedented morphology for (magnetic) topological insulators. All observed nanoscrolls consists of two septuple layers. The radius and therefore the number of turns proved to be a function of applied electrical voltage. Although rolling up is a continuous transformation that in principle should not affect the topology, the breaking of all symmetry, the incommensurable layers at different radii, and the magnetism are some of the aspects that immediately come to mind as fundamentally new.

First-principles investigations of noise in ultra-scaled 2D field effect transistors

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The implementation of 2D semiconductors in field effect transistors (FET) is a promising approach to decrease the spatial dimensions. However, such devices still lack the reliability to be produced on an industrial scale. This limitation arises from the high defect densities present in 2D materials, which degrade the performance of electronic devices. In particular, defects can lead to random telegraph noise (RTN) in FETs due to capture and emission of charge carriers at defect sites. While RTN is typically an undesired phenomenon in conventional electronics, it is possible to exploit such stochastic processes to simulate e.g. afferent neurons, which transform external signals into stochastic electrical spikes. Following this approach, a stochastic encoder for noise-afflicted medical-MNIST images can be constructed using 2D FETs, improving the inference accuracy compared to deterministic encoders [Nat. Comm., accepted (Nov. 2024)]. Measurements and subsequent TCAD simulations suggest that the RTN traces in the ultra-scaled devices originate from hole trapping at fast defects in the 2D substrate. The goal of this work is to determine the microscopic origin of the RTN signals through first-principles calculations. Based on STEM imaging, the types of defects occurring in the monolayer WSe₂ film were identified and subsequently investigated with hybrid functional DFT. The non-radiative charge transitions at various defect types in 1L-WSe₂ are analyzed using a multi-phonon model. In combination with the TCAD simulations, it is found that both the tungsten vacancy (V_W) and the selenium antisite (Se_W) exhibit hole charge transition levels and relaxation energies within the energy range to explain the detected RTN signals. Similar calculations for monolayer WS₂ and MoS₂ are carried out using the same computational setup, allowing for the comparison between different monolayer transition metal dichalcogenides.

Precise Large-Scale Chemical Transformations on Surfaces: Deep Learning Meets Scanning Probe Microscopy with Interpretability

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Scanning Probe Microscopy (SPM) techniques have shown great potential in fabricating nanoscale structures endowed with exotic quantum properties achieved through various manipulations of atoms and molecules. However, precise control requires extensive domain knowledge, which is not necessarily transferable to new systems and cannot be readily extended to large-scale operations. Therefore, efficient and autonomous SPM techniques are needed to learn optimal strategies for new systems, in particular for the challenge of controlling chemical reactions and hence offering a route to precise atomic and molecular construction. In this paper, we developed a software infrastructure named AutoOSS (Autonomous On-Surface Synthesis) to automate bromine removal from hundreds of Zn(II)-5,15-bis(4-bromo-2,6-dimethylphenyl)porphyrin (ZnBr2Me4DPP) on Au(111), using neural network models to interpret STM outputs and deep reinforcement learning models to optimize manipulation parameters. This is further supported by Bayesian Optimization Structure Search (BOSS) and Density Functional Theory (DFT) computations to explore 3D structures and reaction mechanisms based on STM images.

Single-layer boron phosphide on metallic surfaces: screening of promising substrates by first-principles

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Single-layer hexagonal boron phosphide (h-BP) is a predicted graphene-structured semiconductor exhibiting interesting properties for photovoltaic and photocatalytic applications [1] and is the most stable [2] between its proposed allotropes [3]. We investigate by ab-initio calculations the ground-state properties of h-BP on (111) surfaces of different d-shell metals of groups X, XI and XII, with the aim of identifying suitable substrates for its synthesis. Due to the lattice parameter mismatch with the substrates, the h-BP overlayer generates Moiré patterns that we describe using minimal simulation cells with a tolerance of about 3% for tensile or compressive strain applied to the overlayer. Adhesion energy, separation, and charge transfer between substrate and overlayer, projected density of states indicate that the least interacting substrate is Ag, closely followed by Au. The epitaxial h-BP monolayer assumes a buckled structure, with a buckling which is minimum on Ag and maximum on Au. We complete the investigation of h-BP on Ag(111) by considering also the formation of flakes of finite, small size, that are typically more strongly anchored to the substrate through their edges, zig-zag or Klein type. References.

[1] Suzuki T., Theoretical proposal of a revolutionary water-splitting photocatalyst: The monolayer of boron phosphide, *Applied Surface Science* 598 (2022) 153844.

[2] H. Şahin et al., Monolayer honeycomb structures of group-IV elements and III-V binary compounds: First-principles calculations, *Phys. Rev. B* 80 (2009) 155453.

[3] Zhili Zhu et al., Computational prediction of the diversity of monolayer boron phosphide allotropes, *Appl. Phys. Lett.* 109 (2016) 153107. Acknowledgements. We acknowledge support from the European Union- NextGenerationEU, through the ICSC-Centro Nazionale di Ricerca in High Performance Computing, Big Data and Quantum Computing.

Point and complex defects in single-layer transition metal dichalcogenides

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Layered transition metal dichalcogenides (TMDs) constitute an emerging class of materials that provide researchers a platform to realize fundamental studies and to design promising optoelectronic devices. While defects are an almost unavoidable part of TMDs, they bring additional interesting properties absent in defect-free layers. Moreover, the controlled introduction of defects in TMDs makes it possible to tailor the electromagnetic properties of the materials. Here we report defect-induced properties of single-layer PdSe₂ and demonstrate the emergence of magnetism at the nanoscale. Our first-principle calculations indicate that Se vacancies create in-gap defect states, which can be responsible for trapping of carriers. The complex square VPd+4Se vacancy induces spin-polarized states with a total local magnetic moment of 2 μ B per defect making it possible to introduce magnetization into PdSe₂ and therefore expand the family of 2D magnets. The defect formation energies are much lower compared to many other TMD materials, something that can explain the presence of a large number of Se defects after mechanical exfoliation of PdSe₂ layers, while the central location of the Pd atoms preserves them from exfoliation-induced defect formation. The negatively charged vacancies are prone to form and demonstrate spin-polarized states. The small diffusion barrier of VSe in 2D PdSe₂ leads to an easy room-temperature migration, while VPd demonstrates a high diffusion barrier preventing its spontaneous migration. As a guide for experimentalists, we simulate and characterize STM images in valence and conduction states and estimate the electron beam energy for a controllable production of various defect patterns. These intriguing findings make PdSe₂ an ideal platform to study defect-induced phenomena.

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