

Abstracts



Monday afternoon

Two-dimensional materials: from individual devices to complex systems

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The materials that have enabled the information technology revolution over the past decades will soon reach their physical limits. Novel nanomaterials and technologies have therefore become a major focus of current solid-state device research, with two-dimensional (2D) materials being one of the most promising candidates. While much progress has been made on an individual device level, only a few studies have looked into more complex systems. In this talk I will present realizations of electronic and photonic systems, comprising a large number of 2D material-based devices. In particular, I will discuss digital and analog electronics, photonic systems, as well as systems for machine learning. I will present investigations of 2D device variability, its physical origins, and how it affects system performance.

Two-dimensional monoelemental materials beyond graphene

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Monoelemental two-dimensional (2D) materials are at the forefront of current material research. Beyond graphene, which is intensively studied over more than one decade, the other related materials remain almost unexplored. The research activities in the field of other layered materials like phosphorene, black phosphorus monolayer, are rapidly growing in the last few years. Compare to graphene, all these materials are non-zero band-gap semiconductors. This property opens new application possibilities in electronic and optoelectronic devices. Also, the research in the field of energy storage and conversion, as well as sensors and other fields, is rapidly growing. The important properties are based on electronic structure and are strongly related to the material morphology, especially the number of layers. The band-gap energy increases for monolayer materials. It is very important since the rapid increase of band-gap energy allows to utilize the visible region of spectra. The properties of 2D materials can be further controlled by their functionalization. The non-covalent functionalizations are well known in phosphorene chemistry in order to modify its electronic properties and increase its chemical stability towards oxidation. The covalent functionalizations for monoelemental layered material are much less explored and currently exist only a few reports.[1]

Silicon and germanium can also form layered structures, which are not planar as graphene. Compared to carbon in the form of graphite, the 2D allotropes of Si and Ge are not formed from the bulk elements. Their layered forms are obtained either by “bottom-up” procedure by deposition on substrates with suitable symmetry or by “top-down” approach based on chemical exfoliation of precursors with suitable composition like Zintl phases with MX₂ composition where M is an alkaline earth metal (or lanthanide) and X is silicon or germanium forming hexagonal lattice separated by the metal atoms. Selective removal of such atoms can provide silicene or germanene derivatives – substituted silicenes or germanenes. The chemical exfoliation of Zintl phases provides a platform, which allows direct functionalisation of silicene or germanene. The reaction with acid typically provides hydrogenated silicene and germanene. The other reports in silicene and germanene chemistry are based on reaction with alkyl iodides with Zintl phases providing alkylated derivatives. The recent findings in silicene and germanene chemistry show many possibilities for their derivatisations, which open their future applications in optoelectronic as well as the other fields.

Also in the 15th group, so-called pnictogens, can be found elements exhibiting layered structures – phosphorus, arsenic, antimony and bismuth. Phosphorus and arsenic form true layered structures in their orthorhombic forms where between the layers are only weak van der Waals interactions. The rhombohedral allotropes adopted by As, Sb and Bi exhibiting interlayer interactions, which are increasing from arsenic to bismuth with a significant decrease of the anisotropy. The exfoliation of bulk pnictogens was reported for all fourth elements in this group.[2] However, especially the pnictogens with rhombohedral structure show a significant reduction of lateral size due to stronger interlayer interactions. The non-covalent functionalization was reported for phosphorene with various aliphatic and polyaromatic molecules and their derivatives. The functionalized molecules can effectively change the electronic structure of phosphorene and, in addition, significantly improve its stability towards oxidation. The reports on non-covalent functionalization of other pnictogens are much less reported. The covalent functionalization of pnictogens was first reported in 2016 and since that time still only few reports are in literature.[1] For the covalent functionalization, there were reported mainly radical reactions.

The current state-of-the-art in the research of monoelemental two-dimensional materials synthesis, exfoliation and functionalization will be presented together with possible future applications.

Acknowledgements: This work was supported by the Czech Science Foundation (GACR No. 19-26910X).

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[2] Pumera, M., Sofer, Z., *Adv. Mater.*, 2017, 29, 1605299

Monday afternoon

Anharmonic semiconductors - Lessons Learned from Halide perovskites

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In semiconductor physics, the dielectric response, charge carrier mobility and other electronic material properties at finite temperatures, are always treated within the framework of the harmonic approximation. This approach is very successful in capturing the properties of tetrahedrally bonded semiconductors such as silicon and GaAs.

In my talk, I will show that 2D and 3D halide perovskites are fundamentally different due to their strongly anharmonic lattice dynamics. Large amplitude, local polar fluctuations induced by lattice anharmonicity localize the electronic states and enhance the screening of electric charge within the material. In other words, in some aspects, halide perovskites behave more like a liquid than a crystalline solid. I will also discuss the implications of these findings on other families of semiconductors such as organic and rock-salt semiconductors.

Creating 2D Metals for Extreme non-Linear Optics and Quantum Applications

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Quantum materials (QMats) are prime candidates for next-generation energy-efficient technologies, such as topological quantum computing, quantum sensing, and neuromorphic computing. While van der Waals 2D materials exhibit a compellingly wide range of exotic and potentially useful properties such as charge density waves, topological insulator edges, and superconductivity, one can also realize these properties by *stabilizing new 2D allotropes of traditionally 3D superconductors and magnets*. In this talk I will discuss our twist on epitaxial graphene, dubbed confinement heteroepitaxy (CHet), that enables the creation of 2D forms of 3D materials (e.g. 2D-GaN, 2D-Ga, In, Ag)[1-4] and *decouples* the growth of the metals from other 2D layers, thereby enabling a new platform for creating artificial quantum lattices (AQLs) with atomically sharp interfaces and designed properties. As a specific example, we synthesize plasmonic layers that exhibit >2000x improvement in nonlinear optical properties, and 2D-superconductors combined with topological insulators as the building block of next generation “2D” topological superconductors. Confinement heteroepitaxy opens up avenues for enabling a virtual “legoland” of hybrid quantum materials.

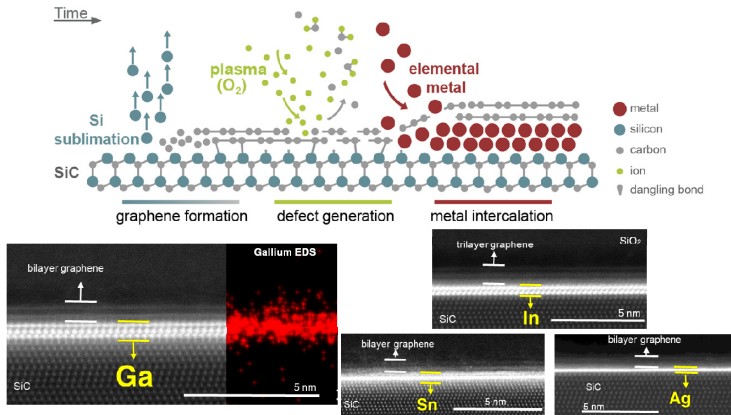


Figure 1: The process of confinement heteroepitaxy to form 2D metals such as 2D-Ga, In, Sn, and Ag.

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3. An Air-Stable and Atomically Thin Graphene/Gallium Superconducting Heterostructure. arXiv:1905.09938
4. Epitaxial Graphene Intercalation: A Route to Graphene Modulation and Unique 2D Materials. arXiv:1905.09261

Monday afternoon

Your paper in *Physical Review Letters*

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From its conception to publication and beyond, a paper in a reputable physics journal entails interactions among authors, editors, referees, conference chairs, journalists, department chairs, university deans, funding agencies, and others. The dissemination of physics is rapidly changing in the age of developments such as optimized search engines, online repositories, social media, and multimedia. I will address how, in particular, PRL (first published 60 years ago) and the *Physical Review* family of journals (established over 125 years ago) continue to play a central role in advancing and diffusing the knowledge of physics in this landscape.

Exfoliated 2D Transition Metal Dichalcogenides: preparation, modification and useVoiry, Damien¹¹ Institut Européen des Membranes (IEM, CNRS UMR5635, ENSCM, UM), University of Montpellier, France

Two-dimensional (2D) materials such as graphene and Transition Metal Dichalcogenides (TMDs) are fascinating objects impacting broad domains in chemistry and physics [1]. 2D materials have also emerged as electrocatalysts demonstrating excellent activity for key electrochemical processes including the hydrogen evolution reaction (HER), the oxygen evolution reaction (OER) and the CO₂ reduction reaction (CO₂RR) [2]. The properties of TMDs can be tuned by changing the elemental composition, the thickness and the atomic structure [1]. TMDs can indeed adopt two different coordinations for the metal atoms: either a trigonal prismatic coordination or an octahedral coordination that corresponds to 2H and 1T phases respectively [3]. Nanolaminate membranes made of exfoliated nanosheets such as graphene oxide (GO) are also promising candidates for molecular sieving *via* size-limited diffusion in the 2D capillaries of nanolaminate membranes [4]. Our research aims to investigate the behavior of 2D materials as building-blocks of nanolaminate membranes and for novel electrochemical reactions. We have notably explored the role of the structural phase and the defects on the HER performance from group-6 TMDs in order to optimize the thermodynamics and the kinetics of the catalytic reaction. Controlling the surface chemistry of the 2D nanosheets can provide additional levers for enhancing the sieving performance of the nanolaminate membranes.

Through several examples, I will present our recent strategies for engineering exfoliated nanosheets of TMDs. My presentation will highlight how these strategies can be used for fine tuning the properties of the 2D TMDs and other 2D materials for molecular sieving and electrocatalysis.

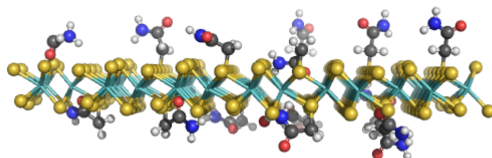


FIG. 1. Schematic showing the side view of a covalently functionalized TMD nanosheet.

[1] Wang, Q. H., Kalantar-Zadeh, K., Kis, A., Coleman, J. N. & Strano, M. S. Electronics and optoelectronics of two-dimensional transition metal dichalcogenides. *Nat. Nanotechnol.* **7**, 699–712 (2012).

[2] Voiry, D., Shin, H. S., Loh, K. P. & Chhowalla, M. Low-dimensional catalysts for hydrogen evolution and CO₂ reduction. *Nat. Rev. Chem.* **2**, 0105 (2018).

[3] Voiry, D., Mohite, A. & Chhowalla, M. Phase engineering of transition metal dichalcogenides. *Chem. Soc. Rev.* **44**, 2702–2712 (2015).

[4] Joshi, R. K. et al. Precise and Ultrafast Molecular Sieving Through Graphene Oxide Membranes. *Science* **343**, 752–754 (2014).

An update on properties and synthesis of MoS₂ and WS₂ nanotubes

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The availability of WS₂ multiwall inorganic nanotubes (MWINT-WS₂) in large amounts led to the extensive investigation of their properties suggesting numerous applications such as strengthening of polymers, substrate for hydrogen storage, catalyst for hydrogen evolution and hydro-desulfurization, optoelectronics, etc.

Thus, bulk photovoltaic effect (BPVE) in intrinsic INT-WS₂ was observed due to their small band gap, broken inversion symmetry, low dimensionality and polar structure [1]. The generated photocurrent is orders of magnitude larger than in other BPVE materials.

Low temperature cathodoluminescence (CL) of single-wall WS₂ nanotubes (SWINT), produced by high-power plasma irradiation of ~100 nm MWINT-WS₂ nanotubes [2], demonstrates blue shift, confirming quantum confinement effect in a few nm diameter tube [3].

Solid-gas reaction of Mo/W oxides with H₂/H₂S gases applied for the synthesis of MWINT-WS₂ [4] and WS₂/MoS₂ inorganic fullerene-like (IF) nanoparticles [5,6] resulted in pure phases and scalable processes. However, due to the instability of the intermediate products, similar reaction to obtain nanotubes of MoS₂, was found to be highly challenging, resulting in miserable yield, difficult control and poor reproducibility. Current study succeeded to overcome these obstacles without the assistance of a catalyst and reports on sustainable high-yield synthesis of MoS₂ nanotubes 20-100 nm in diameter and up to 10 μm in length [7]. Individual INT-MoS₂ were characterized by TEM, EDS, selected area electron diffraction (SAED) and Raman.

INT of MoS₂ are both 40% lighter and 40% stronger compared to their WS₂ analogous and hence more beneficial for tribological and composite applications. Being semiconductors, both MoS₂ and WS₂ nanotubes are good candidates for photovoltaics and optoelectronics.

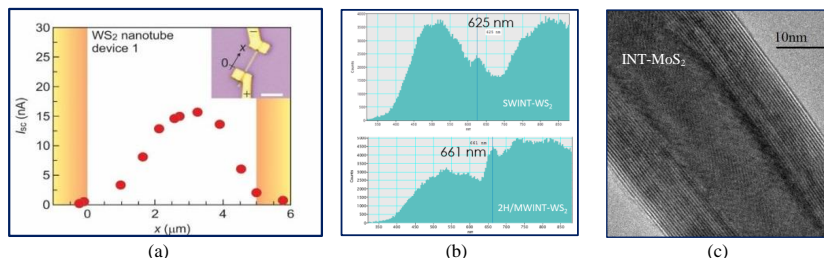
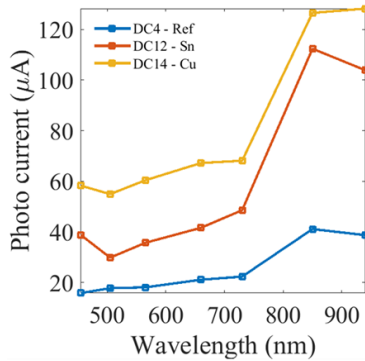


FIG. 1. (a) Bulk photovoltaic effect (BPVE) in intrinsic INT-WS₂ - due to the small band gap, broken inversion symmetry, low dimensionality and polar structure. The generated photocurrent is orders of magnitude larger than in other BPVE materials; (b) cathodoluminescence (CL) of SW- and MW-INT of WS₂. Blue shift in CL of SWINT confirms quantum confinement effect in a few nm diameter tube; (c) TEM micrograph of MoS₂ nanotube obtained via solid-gas reaction by the reproducible, scalable, catalyst-free and size-controlled synthesis, for the first time 24 years after their discovery in 1995 [8];

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- [2] V. Bruser, R. Popovitz-Biro, G. Seifert, R. Tenne, A. Zak, et al., Inorganics, 2, 177, (2014).
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Defects Activation by Metal Intercalation in MoS₂ Thin FilmsMoshe Kirshner,¹ Chen Stern,¹ Kristie Koski,² Ashwin Ramasubramaniam,³ and Doron Naveh¹¹ Faculty of Engineering and Institute of Nanotechnology and Advanced Materials, Bar-Ilan University, Ramat-Gan, Israel 52900² Department of Chemistry, University of California Davis, Davis California USA 95616³ Department of Mechanical and Industrial Engineering, University of Massachusetts Amherst, Amherst MA USA 01003

Intercalation chemistry of layered compounds and particularly zero-valent metals in semiconductor host was established over years of studies that established firm understanding of the structural properties and phases of these materials. Zero-valent metals can form self-organized structured monolayers in the Van der Waals gap of the host material, yet we show for the first time the result of such intercalation in volume-constricted MoS₂ and the resulting electronic properties of such layers for the cases of copper and tin intercalation in MoS₂. We study the effects of zero-valent metal intercalation on the electronic and optoelectronic properties of the host material. We find that metal intercalation activate traps by flooding those with free electrons. The intercalation of copper and tin enhance both the conductivity and photoresponse of Si-MoS₂ photodiodes. We estimate experimentally the band alignment and trap location and density towards a comprehensive understanding of the potential of such intercalations for optoelectronics and photocatalysis.

FIG. 1. Photocurrents of bare Si-MoS₂ diode (blue), Sn-intercalated MoS₂ and copper-intercalated MoS₂.

Hybridized indirect excitons in MoS₂/WS₂ heterobilayers

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Ensembles of indirect or interlayer excitons (IXs) are intriguing systems to explore classical and quantum phases of interacting bosonic ensembles. IXs feature enlarged lifetimes due to the reduced overlap of the electron-hole wave functions [1,2]. A field effect structure with few layer hexagonal boron nitride (hBN) as insulator and few-layer graphene as gate-electrodes facilitates an electric field control of the IXs in a MoS₂/WS₂ heterobilayer [see Fig. 1 (a)] [2]. A multiplet structure in the IX emission band can be observed even at room temperature. Stark shift measurements reveal the presence of a finite out-of plane dipole of the IXs. Due to a different strength of the dipole and a distinct temperature dependence, we identify the IXs to stem from optical interband transitions with electrons and holes in different valleys of the heterostructures [2]. We observe a field dependent level anti-crossing for the energetically lowest emission line forming hybridized indirect excitons at low temperatures (cf. Figs. 1b and 1c) [2,3]. We discuss this behaviour in terms of a finite coupling of the electronic states of the two TMDC monolayers (cf. Fig. 1d) making it possible to control the probability for the hole to reside in the MoS₂ or WS₂ layer of the artificial vdW solid by application of an external electric field. Such hybridized states have great potential for the realization of robust charge qubits at relaxed experimental conditions. Overall, our results demonstrate the design of novel nano-quantum materials prepared from artificial van der Waals solids with the possibility to in-situ control their physical properties via external stimuli such as electric fields.

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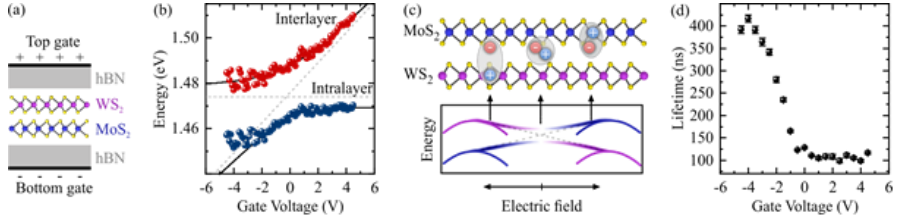


FIG. 1. (a) Schematic depiction of the MoS₂/WS₂ heterobilayer field effect structure. (b) Field dependent level anti-crossing of the energetically lowest interband transitions from the Σ -point to the K-point at a bath temperature of 10K. (c) Proposed model for the level repulsion. The hole at the K-point changes the hosting layer in dependence of the gate voltage due to a finite layer coupling. (d) Gate voltage dependent PL lifetime tuning. The PL lifetime shift from 400 ns to 100 ns supports the interpretation of a gradual change of the excitonic nature from interlayer to intralayer. (modified from [2])

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Ultra-thin van der Waals crystals as semiconductor quantum wells

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While two-dimensional (2D) materials research has mainly focused on the monolayer forms of van der Waals crystals, the broad range of properties of intermediate thickness few-layer systems has remained less well explored. Conceptually we may view few-layer 2D semiconductors as semiconducting quantum wells, with their characteristic subband spectra. Whereas the primary optical transitions of the 2D semiconductors generally fall in the visible and near infrared (IR) ranges, their intersubband transitions present an as yet largely unexploited opportunity to extend the optical activity further into the IR and THz ranges. Such transitions, depending on the material, its thickness and doping, will densely populate an energy range ~ 0.8 eV to 0.05 eV for films ~ 10 layers [S. J. Magorrian *et. al.*, Phys. Rev. B 97, 165304 (2018); D. A. Ruiz-Tijerina *et. al.*, Phys. Rev. B 98, 035411 (2018)].

The first experimental observation of intersubband transitions in a 2D semiconductor, WSe₂, was recently demonstrated using near-field THz spectroscopy [P. Schmidt *et. al.*, Nat. Nanotechnol. 13, 1035 (2018)]. While this permitted direct measurements of the transitions, it requires the use of demanding detection techniques. We will demonstrate how the subband structure of 2D semiconductors may be studied by combining the use of resonant tunnelling (Fig. 1) and photoluminescence excitation spectroscopy, exemplifying the technique for 4 – 7 layer films of InSe. We achieve a comprehensive understanding of both the conduction and valence subband spectra, including their evolution due to the quantum confined Stark effect, in agreement with theory and angle-resolved photoemission spectroscopy.

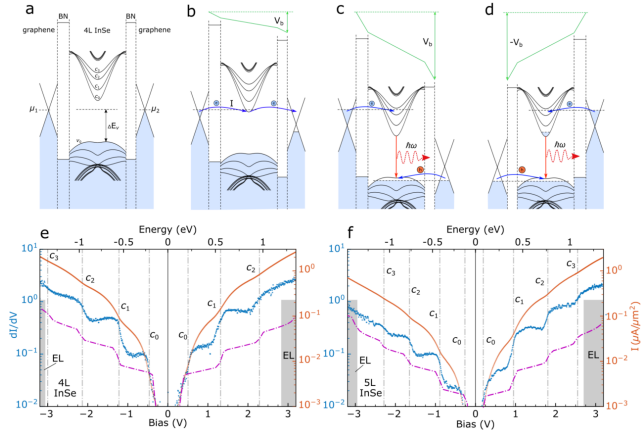


FIG. 1. Low-temperature resonant tunnelling spectroscopy of conduction band subbands in 4- and 5-layer InSe films: Band alignment in 4L InSe device, for unbiased (a), weakly biased (b) and strongly biased in forward (c) and reversed (d) configurations. Current density (orange), dI/dV (blue) and density of states from tight binding model (purple, arb. u.) for 4L (e) and 5L (f) InSe film, with the number of layers found using AFM topography. Energy/Bias relation found using onset of electroluminescence.

Magneto-spectroscopy of interlayer excitons in hBN-encapsulated TMDC heterostructures

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Transition-metal dichalcogenide (TMDC) monolayers are direct-gap semiconductors with peculiar spin-valley coupling. They are characterized by inequivalent valleys at the corners of the Brillouin zone (K points), which can be selectively excited via circularly polarized light. Combining two different TMDCs can lead to a type-II band alignment, so that optically excited electron-hole pairs form interlayer excitons (IEX). Encapsulating such a heterobilayer in hexagonal boron nitride (hBN) suppresses inhomogeneous broadening and allows spectroscopic observation of subtle effects.

In crystallographically aligned heterobilayers consisting of the TMDCs MoSe₂ and WSe₂, which have a very similar lattice constant, a moiré-like periodic modulation of the potential landscape occurs, providing a trapping potential for IEX [1]. For crystallographic alignment of the adjacent layers, there are two possibilities giving rise to optically bright IEX recombination. In the aligned structure, interlayer transitions between the MoSe₂ conduction band and the WSe₂ valence band K valleys with the same valley index are direct in reciprocal space. In the anti-aligned structure, interlayer transitions between valleys with different indices are *k*-space direct. This strongly modifies the energetic splitting of IEX photoluminescence (PL) emission in a magnetic field [2].

Here, we report on PL spectroscopy of IEX in several hBN-encapsulated MoSe₂-WSe₂ heterobilayers (Figure 1a) with different crystallographic alignment. In some structures, we are able to resolve multiple, spectrally separated IEX emission peaks, in agreement with recent observations [1], see Figure 1b,c. Tracking the energetic shifts of the different IEX features in large magnetic fields, we find a complex behavior that strongly depends on the sample. However, we are able to identify systematic differences between crystallographically aligned and anti-aligned structures. We compare our experimental findings to density-functional-theory-based calculations of magnetic field effects on the electronic band structure of the heterobilayers.

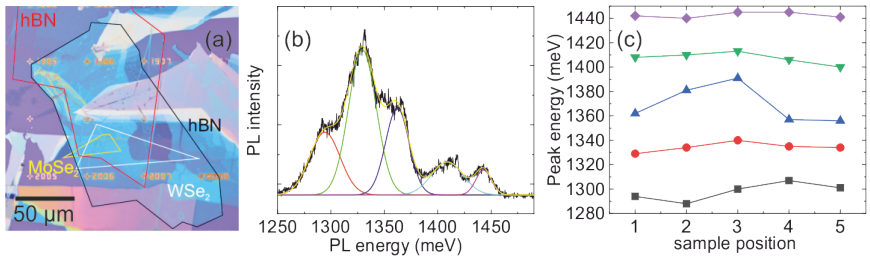


FIG. 1. (a) Optical microscope image of hBN-encapsulated heterobilayer. (b) PL spectrum of heterobilayer shown in (a), measured at 4.5 K. We observe multiple, spectrally separated peaks in the interlayer exciton (IEX) emission energy range. (c) Dependence of IEX peak positions on sample position.

Tuesday afternoon

Dielectric disorder and exciton transport in 2D semiconductors

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Semiconducting two-dimensional materials are found to exhibit a number of highly intriguing phenomena, including spin-valley locking, efficient light-matter coupling, and strong Coulomb interaction. The ongoing research is further driven by an increasing variety of recently available heterostructures to tune and extend the properties of van der Waals systems through artificially assembled stacks. These avenues strongly motivate increased attention to the consequences of environmental influences and the role of proximity effects for the key properties of local electronic structure. Furthermore, considering that the fundamental optical excitations in these systems are dominated by strongly bound exciton states, a key necessity arises to study their spatial properties, that remained barely explored so far. Fundamental understanding of both energy landscapes in the 2D plane and the movement of excitonic and electronic quasiparticles across them is thus highly relevant with respect to basic physics and in view of the potential use of 2D materials in future optoelectronic technology.

Here, I will address the topics of environmental energy fluctuations and exciton propagation in van der Waals semiconductors. In the first part, I will present an alternative, fundamental source of disorder in 2D materials based entirely on the local changes of the Coulomb interaction due to fluctuations of the external dielectric environment. The consequences for optical and transport properties of excitons and free charge carriers will be discussed. The importance of dielectric homogeneity in heterostructures will be further highlighted, demonstrated by spatial mapping of the fluctuations and by identifying the elimination of the dielectric disorder as a main consequence of material's encapsulation. In the second part, direct measurements of room temperature exciton propagation will be presented and discussed, focusing on the influence of disorder and non-trivial optical transport phenomena beyond the linear regime.

Tuesday afternoon

Electron and exciton-phonon interaction in atom-thin multivalley semiconductors

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In two-dimensional crystals the enhanced density of states of lattice vibrations results in significant interaction between the acoustic phonons and electronic quasiparticles: electrons and excitons. Here we address theoretically two situations where such interaction gives rise to novel and unexpected effects.

First, we study theoretically intervalley coupling in transition-metal dichalcogenide monolayers due to electron interaction with short-wavelength phonons [1]. This intervalley polaron coupling results in (i) a renormalization of the conduction band spin splitting and (ii) an increase of the electron effective masses. We also calculate the renormalization of the cyclotron energy and the Landau level splitting in the presence of an external magnetic field. We uncover an inter-valley magneto-phonon resonance where the separation between the Landau levels equals to the intervalley phonon energy. Similar, but much weaker effects are also expected for the valence band holes. These results might also help to resolve the discrepancy between *ab initio* values of the electron effective masses and the ones deduced from magneto-transport measurements.

Second, we address excitonic transport in the presence of highly non-equilibrium acoustic phonons [2]. The phonon “hot-spot” acts as a source of the phonon wind which drags excitons resulting in the non-diffusive exciton transport. We demonstrate that a halo-like spatial distribution of excitons akin observed in [3] can be formed as a result of the exciton drag by non-equilibrium phonons.

This work was supported in part by Russian Science Foundation (project # 19-12-00051).

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[2] M. M. Glazov, arXiv:1905.01621 (2019).

[3] Marvin Kulig, Jonas Zipfel, Philipp Nagler, Sofia Blanter, Christian Schüller, Tobias Korn, Nicola Paradiso, Mikhail M. Glazov, and Alexey Chernikov, *Phys. Rev. Lett.* **120**, 207401 (2018).

Tuesday afternoon

Excitons in Two-Dimensional Transition-Metal Dichalcogenides: Why does the Effective Hydrogen Model Fail?

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Ever since the first experimental measurements of the exciton spectra in two-dimensional (2D) transition-metal dichalcogenides (TMDC), it was realised that the commonly used effective hydrogen model is not applicable because it overestimates the binding energies of the excitons in these materials. First attempts to understand theoretically the exciton spectra were based on a modified interaction potential – indeed, due to the layered structure of 2D TMDC, the interaction is screened in a non-local manner, and one should use the Keldysh potential rather than the usual $1/r$ Coulomb interaction for the interaction between the electron and the hole constituting the exciton. While this approach yields better numerical results for the spectra in view of experimental findings, it does not account for another major effect: in 2D TMDC, the valence and conduction bands are strongly coupled in spite of the rather substantial band gap. In modern language, this band coupling, which is a signature of Dirac excitons [1], is accounted for by the so-called Berry curvature, which yields pertinent corrective terms to the hydrogenic model. These corrective terms can also be viewed as an effective pseudospin-orbit coupling where the pseudospin represents the atomic orbits responsible for the bands at the direct gap.

In this presentation, we show how the Berry-curvature terms arise in the effective model for excitons in 2D TMDC and discuss their effect on the exciton spectra [2], both in the case of neutral and charged excitons (trions) [3]. We show that these terms conspire with the Keldysh potential and that their inclusion provides a better estimate of the screening length in comparison with *ab initio* calculations.

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Spatiotemporal exciton dynamics in atomically thin materials

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Monolayer transition metal dichalcogenides (TMDs) exhibit a remarkable excitonic landscape including bright exciton states, but also a variety of dark excitons that cannot be accessed by light due to the required momentum-transfer or spin-flip [1]. Solving 2D material Bloch equations for excitons, phonons and photons, we obtain a microscopic access to the time-, momentum-, and space-resolved non-equilibrium dynamics. In particular, we shed light on the interplay of optics, ultrafast dynamics and diffusion processes of excitons in this technologically promising class of atomically thin materials. In joint theory-experiment studies, we demonstrate:

- (i) Formation, thermalization, and photoemission of bright and dark intra- and interlayer excitons in TMD monolayers and related van der Waals heterostructures (Fig. 1a) [2-4].
- (ii) Pronounced low-temperature signatures of momentum-dark excitons in photoluminescence as well a clear transition of oscillator strength to the dark states in infrared spectroscopy probing intra-excitonic transitions [5,6].
- (iii) Exciton propagation and formation of spatial exciton rings (halos) due to the emission of hot phonons acting as a heating mechanism and giving rise to strong spatial temperature gradients and thermal drifts (Fig. 1b) [7].

The gained microscopic insights into the spatiotemporal dynamics of excitons are crucial for understanding, predicting, and controlling exciton optics, dynamics and transport, which is of central importance for the design of future high-performance optoelectronics devices.

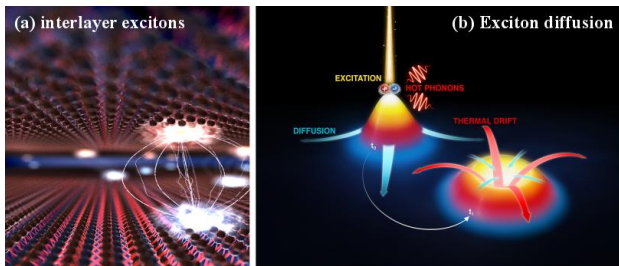


FIG. 1. (a) Formation of interlayer excitons in a van der Waals heterostructure, where the Coulomb-bound electron and hole are localized in different layers. (b) Exciton diffusion and halo formation in a transition metal dichalcogenide. Exciton relaxation by hot-phonon emission creates a long-lived spatial gradient in excitonic temperature. This gives rise to a thermal drift dragging excitons out of the central region and resulting in the formation of spatial rings (halos).

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Wednesday morning

Defect induced and layer-modulated magnetism in ultrathin metallic PtSe₂

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Intrinsic, two-dimensional (2D) magnetic materials are attracting increasing attention for their unique properties including layer-dependent magnetism and electric field modulation. Yet, experimentally inducing magnetism into otherwise non-magnetic 2D materials remains a challenge. Here, we investigate magneto-transport properties of ultrathin PtSe₂ crystals and demonstrate unexpected magnetism. Our electrical measurements show the existence of either ferromagnetic or anti-ferromagnetic ground state orderings depending on the number of layers in this ultra-thin material. First-principles calculations suggest that surface magnetism induced by the presence of Pt vacancies and the RKKY exchange couplings across ultra-thin films of PtSe₂ are responsible for the observed layer-dependent magnetism. Considering the existence of such unavoidable growth-related vacancies in 2D materials, these findings can expand the range of 2D ferromagnets into materials that would otherwise be overlooked.

Flatlands beyond graphene: Magnetic Ordering in Ultrathin Crystals

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Two-dimensional (2D) materials exhibit intriguing physical properties and many of these materials can be manually assembled into atomic-scale multilayer devices under ambient conditions, owing to their exceptional chemical stability. Efforts have been made to add a magnetic degree of freedom to these 2D materials via defects, but only local magnetism has been achieved. Only with the recent discoveries of 2D materials supporting intrinsic ferromagnetism (such as CrI₃, Cr₂Ge₂Te₆ etc.) have stacked spintronic devices become realistic. Assembling 2D multilayer devices with these ferromagnets under ambient conditions remains challenging due to their sensitivity to environment degradation, and magnetic order at room temperature is rare in van der Waals materials. Here, we report the growth of air-stable ultrathin epsilon-phase iron oxide crystals that exhibit magnetic order at room temperature [1]. These crystals require no passivation and can be prepared in large quantity by cost-effective chemical vapor deposition (CVD) technique. We find that the epsilon phase, which is energetically unfavorable and does not form in bulk, can be easily made in 2D down to a seven unit-cell thickness. Magneto-optical Kerr effect (MOKE) magnetometry of individual crystals shows that even at this ultrathin limit the epsilon phase exhibits robust magnetism with coercive fields of hundreds of mT (Figure 1).

Another material is V₅S₈, a metal with an antiferromagnetic ground state below the Néel temperature $T_N \sim 32$ K and a prominent spin-flop signature in the magnetoresistance (MR) when $H||c \sim 4.2$ T [2]. We study ultrathin crystals of V₅S₈, focusing on temperatures close to T_N and the evolution of material properties in response to systematic reduction in crystal thickness. Transport measurements just below T_N reveal magnetic hysteresis that we ascribe to a metamagnetic transition, the first-order magnetic field-driven breakdown of the ordered state. The reduction of crystal thickness to ~ 10 nm coincides with systematic changes in the magnetic response: T_N falls, implying that antiferromagnetism is suppressed; and while the spin-flop signature remains, the hysteresis disappears, implying that the metamagnetic transition becomes second order as the thickness approaches the 2d limit. These results highlight the advantages of ultrathin crystals grown by CVD on Van der Waals substrates as promising candidates towards air-stable 2D magnetism and integration into 2D spintronic devices.

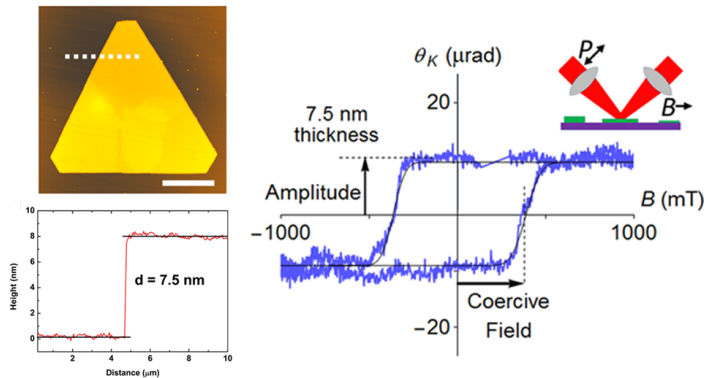


FIG. 1. **Left:** AFM image of a representative ultra-thin epsilon-Fe₂O₃ crystal with triangular shapes grown on a mica substrate and corresponding thickness measurement; **Right:** longitudinal MOKE measurement at room temperature (295K) of the same crystal, clearly showing room-temperature magnetic order in the ultra-thin epsilon-Fe₂O₃ with well-defined transitions, and coercive fields of hundreds of mT.

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Data-driven discovery of novel 2D materials and heterostructures

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Atomically thin two-dimensional (2D) materials represent a rapidly moving frontier of condensed matter physics. The reduced phase space and low dielectric screening in these extremely thin materials lead to enhanced many-body effects and unique physics, which can be further tuned by external probes or by stacking the 2D materials into van der Waals heterostructures.

It will be shown how the electronic and optical properties of 2D materials and their heterostructures can be accurately predicted by combining classical electrostatic models with many-body quantum mechanics. I will discuss novel types of excitonic states in 2D semiconductor heterostructures and illustrate the concept of dielectric band structure engineering, which exploits the dependence of the quasiparticle energy on the local dielectric environment[1]. Finally, I will present our recent efforts to establish a comprehensive computational database of 2D materials using an automatic high-throughput framework based on the Atomic Simulation Environment (ASE) (<http://c2db.fysik.dtu.dk>)[2] and demonstrate how it can be used to identify 2D materials with interesting physical properties such as ferromagnetism and non-trivial band structure topologies[3].

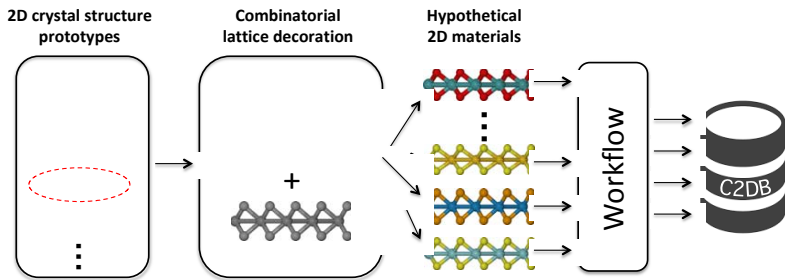


Figure. The materials in the C2DB are initially generated by decorating an experimentally known crystal structure prototype with atoms chosen from a (chemically reasonable) subset of the periodic table.

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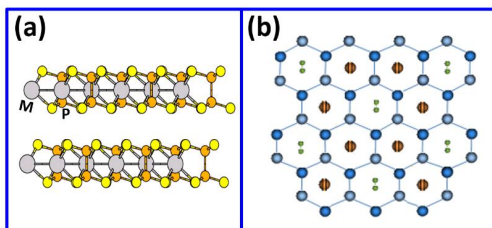
Two dimensional compounds beyond graphene and beyond transition metal di-chalcogenides

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Inorganic lamellar compounds are characterized by strong intra-layer chemical bonding and only weak van der Waals interaction between adjacent layers. Single layers are composed of metal ions sandwiched between chalcogenide or halide atoms, to form three- or four-atom thick slabs. Each metal is positioned at the center of a symmetric or distorted octahedron, where adjacent octahedra share edges for the formation of a single layer. Although the bulk forms of lamellar materials have been studied extensively in the previous century, individual layers of the most common materials—MoS₂ and WS₂ transition metal di-chalcogenides, have received much attention in the past decade due to their decent conductivity, electronic band gap, in-plane stiffness, and above all, exhibiting a honeycomb Brillouin zone with distinguished K and K' symmetry points of opposing helicities, enabling potential applications in spin-based devices. The current document proposes the investigation of metal phosphor tri-chalcogenides with the general chemical formula MPX₃ (M=metal, X=chalcogenide), closely resembling the metal di-chalcogenides, but one-third of the metal ions are replaced by a phosphor pair (P-P) (Scheme 1a). MPX₃ offers a large range of chemical compositions tunable by the M and X elements, and consequently forming semiconductors with band gap energies covering the range from UV to the near infrared. A top view of a single layer reveals a honeycomb

arrangement of the metal ions with P-P at the center of a hexagon (Scheme 1b). Furthermore, the metal ions within a layer produce a ferromagnetic or anti-ferromagnetic arrangement, endowing those materials with unique magnetic and magneto-optical properties. The valley degree of freedom is correlated with the antiferromagnetic ordering. The electronic band structure is dominated by the [P₂X₆]⁴⁻ units, contributing s- and p-atomic orbitals, while the metal d-orbitals hybridize with X(p) orbitals. Most common MPX₃ compounds have monoclinic or orthorhombic crystallographic structures with a space group C₂ or P2₁/c symmetry, respectively.

The talk at the Flatlands meeting will include description of the optical and magneto-optical properties of a vast number of MPX₃ single slabs, Moiré patterning and multiple layers. The description will focus on the influence of the internal magnetism on the optical properties of the materials, e.g., degree of circular polarization in the magneto-reflectance or magneto-photoluminescence spectra. Generally, the MPX₃ family show additional physical properties beyond graphene and beyond the well-known Mo(W)X₂ compounds, with tunable electronic and magnetic properties.



Scheme 1: (a) A side view of MPX₃ structure; (b) Top view of a single layer (brown, blue and green dots denotes the metal, chalcogenide and phosphor atoms).

Liquid phase exfoliation of organic sheet stacks- A case study

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Liquid phase exfoliation (LPE) has become an important production technique giving access to single and few-layered nanosheets in colloidal dispersion. It has been shown to be applicable to a whole host of inorganic crystals.^[1] In addition to layered inorganic crystals, organic sheet stacks such as 2D polymers or covalent and metal organic frameworks have received increasing attention in recent years. Such organic sheet stacks can be made by a number of techniques, e.g. using reversible solution phase chemistry, polymerisation after assembly on the liquid-air or liquid-liquid interface or polymerisation after precisely aligning monomers in single crystals. With the latter approach, perfectly single crystalline 2D polymer sheet stacks such as structure **1** can be synthesised.^[2]

We now used this 2D polymer to test the feasibility of state of the art liquid phase exfoliation in combination with established size selection techniques.^[3] We found that crystalline nanosheets can be obtained after exfoliation in solvents, however at very low yield. In aqueous surfactant solutions, where monolayer populations are typically higher,^[1] the nanosheets of **1** undergo a structural distortion likely due to Coulombic interactions of the positively charged sheets and the negatively charged surfactant. This has prompted us to develop a post-polymerisation modification of **1** to the neutral pyridine-based polymer **2** by reaction of the single crystals with gaseous ammonia. This reaction converts approximately 1.4×10^6 pyrylium ions per sheet μm^2 into the corresponding pyridines and is amongst the most complex post-polymerizations with molecular definition.

When performing LPE and subsequent size selection on the uncharged polymer **2**, crystallinity of the nanosheets was preserved. By optimising the size selection, mg fractions with up to 29% of monolayers of this novel 2D polymer with an average length of ~ 130 nm were obtained. By the application of secondary centrifugation cascades, this content was increased further to 33% with a length of ~ 140 nm. This is the first time that the amount of monolayers of an exfoliated organic 2D material has been quantified. Overall, LPE of **2** is equally efficient as graphite exfoliation. The solution processability allows to cast thin films enabling the characterisation by vibrational spectroscopy. We expect that recent advances in exfoliation of graphite or inorganic crystals (e.g. scale-up, printing etc.) can be directly applied not only to this 2D polymer, but also to other organic sheet stacks such as covalent organic frameworks. This will result in a whole host of novel 2D nanosheets with precisely tailored structure, e.g. pore size, holding great promise for applications as separation membranes.

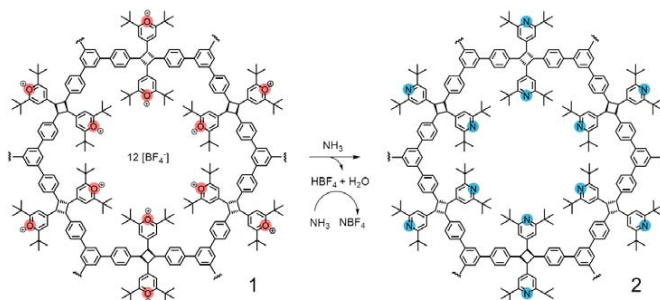


FIG. 1. Structure of the two organic polymers under study. The pyridine-based polymer **1** can be transformed into **2** by post-polymerization modification in the single crystal.

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Phase selective synthesis of two-dimensional crystals

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The direct synthesis of two-dimensional (2D) 1T or 1T' MoS₂ with high phase purity remains as a challenge due to the thermodynamically metastable nature arose from their rather higher formation energy, which dramatically hinders their fundamental studies on superconductivity, magnetism, ferroelectricity and potential applications in electronics, catalysis and batteries. Combined with theoretical calculations, we designed a potassium-assisted chemical vapor deposition (CVD) method and succeeded to the phase-selective growth of 1T' MoS₂ monolayers and the 1T'/2H hetero-phase bilayers. We also characterized the in-plane spectroscopic and electrical anisotropy of 2D 1T' phase MoS₂. Our work opens up a way for the direct synthesis of 2D metastable TMDCs and provides new insights into the construction of novel 2D heterostructures, and thus will further extend their functionalities and applications. ^[1]

In addition to the phase selective synthesis of 2D MoS₂, we also achieved the phase selective synthesis of 2D MoTe₂ by tellurizing MoO_x thin films with varied stoichiometry. Based on the patterned synthesis of hetero-phase structures of MoTe₂, we constructed field effect transistors (FETs), arrays of logic devices and radio frequency devices using 2H and 1T' MoTe₂ as channels and contacts, respectively. The obtained devices can work immediately after the synthesis process without further processing, and thus avoided materials degradation induced by the conventional lithographic procedures. This one-step and one-piece construction strategy combined with the unique planar structure of 2D atomic crystals not only bring extra freedom to the device design and optimization but also reduce the contact barriers and interfacial impurities to the largest extent and therefore, opens a new avenue for building high performance devices using 2D electronic components. ^[2]

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Thursday morning

Optical properties of 2D materials

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The physical properties of two-dimensional semiconductors, such as transition-metal dichalcogenides (TMDCs), can be modified through the presence of adjacent materials, as in van-der-Waals heterostructures or in covalently functionalized layers. We present recent results on the interlayer excitons in MoSe₂-WSe₂ and MoS₂-WSe₂ heterostructures. Furthermore, we discuss Raman spectra of chemically functionalized MoS₂ flakes. If the functionalization is based on chemically exfoliated flakes, we observe oxidation processes, which depend on the temperature and excitation power. This is in contrast to mechanically exfoliated, on-substrate functionalized MoS₂ layers. The defect-activated LA (longitudinal acoustic) phonon from the K/M points in the Brillouin zone indicates successful functionalization.

Two-dimensional Topological Polymers

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The discovery of graphene initiated an immense research effort in the field of two-dimensional (2D) crystals. Graphene shows extraordinary high stability, intriguing electronic, including topological, properties, and chemical inertness. Soon it was clear that 2D crystals can be formed from virtually all layered materials by top-down (exfoliation...), but also bottom-up (chemical vapor deposition...) approaches. The family of 2D materials contains a lot of remarkable phenomena, for example 2D semiconductors that get metallic when the number of layers is increased, 2D topological insulators, and 2D metals.

It is somewhat less known that graphene represents also the prototype 2D polymer. 2D polymers (also 2D covalent-organic frameworks – 2D COFs) are a rather new family of synthetic 2D crystals where molecular units are stitched together with strong bonds. This offers a regular crystal lattice and thus materials comprising all collective phenomena that are known from solid state physics, however, with a much richer diversity due to the essentially infinite number of molecules that can be considered. A recent breakthrough was the discovery of chemical coupling reactions that achieve full conjugation between the constituting molecules, which is the precondition for the formation of 2D semiconductors with ballistic transport properties.

For the 2D polymers I will to focus on structural diversity: while mathematically, 11 tilings are possible in two dimensions (the so-called Kepler nets), nature offers much less structural diversity in crystalline 2D materials. By picking suitable molecular building units we can form lattices with structural topologies that impose, in turn, electronic topologies. One of the examples that I will highlight is the kagome structure, which produces both Dirac points and flat bands (Figure 1) [1].

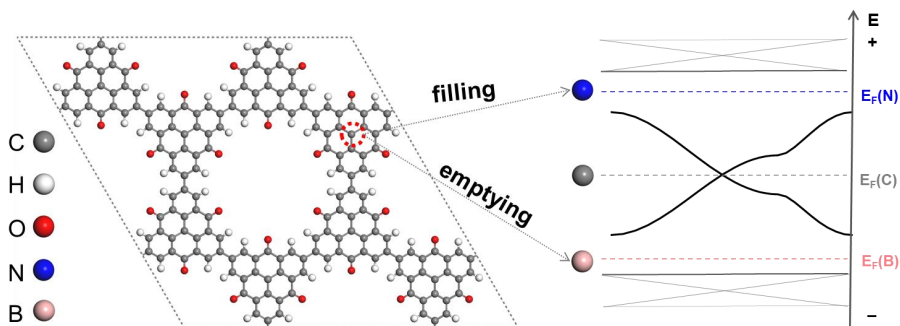


FIG. 1. Unit cell of a heterotriangulene-based kagome lattice and corresponding band structure. If the centre atoms are chosen as C, Dirac points are located at the Fermi level. If all centre atoms are substituted by N (B), one electron per monomer is added (removed) from the structure, and the Fermi level shifts above (below) the Dirac band, making the conduction (valence) band to be a flat band.

Optical spectroscopy of 2D perovskites in high magnetic fields

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Organic-inorganic halide perovskites have become the next big thing in emerging semiconductor materials, with their unprecedented rapid development and successful application in high-performance photovoltaics. Yet, their inherent instabilities over moisture, remain a crucial challenge for these materials. This directed the interest of the scientific community to perovskites derivatives such as 2D perovskites. These materials are significantly more stable and possesses higher tunability of their properties which expand the field of their application from energy harvesting through LED to single materials white light emitters.

This natural quantum wells can be directly synthesized [1] or obtained by the ligand-assisted exfoliation from the 3D perovskites [2] forming so called nanoplates. 2D perovskites are often regarded as perfect quantum wells, because they are not plagued by inter-facial roughness or intermixing characteristic of epitaxially grown quantum wells. The well layers consists of planes of lead-halide octahedra, separated by organic spacers. The research of this natural perovskite quantum wells is in infancy stage and their structural, dielectric, optical, and excitonic properties remain to be explored especially influence of organic spacers. Interestingly, despite bandedge states are composed of lead and halide orbitals organic ligands affects the band structure and electron-phonon coupling via distortion of the octahedral cages. The optical properties of these materials are dominated by strong electron-hole attraction resulting from dielectric confinement. While the large variety of ways to tune the optical properties of 2D perovskites is undoubtedly their huge advantage, the interaction between different parameters hinders our fundamental understanding of their properties. To date their structural, dielectric, optical, and excitonic properties remain largely unexplored.

In this work we show how magnetic field can be applied understand the basic band-structure properties of 2D perovskites. Very often this materials exhibits complex absorption and emission spectrum with many side-bands which origin can be attributed to phonon replicas, bound excitonic states or mixture of different thickness nanoplatelets in case of colloidal systems. Here we address this issue for $(C_nH_{2n+1}NH_3)2PbI_4$ (with $n=4, 6, 8, 10, 12$) and nanoplatelets exfoliated from $MAPbI_3$ by means of optical spectroscopy in magnetic field up to 67T. In case of $(C_nH_{2n+1}NH_3)2PbI_4$ we show that complex absorption features shift parallel in magnetic field indicating phonons related nature of the observed side bands. Moreover our studies supported by modeling, unambiguously demonstrate that the reduced mass of exciton change about 30% upon phase transition in this materials. This is reflected by 3 time decrease of diamagnetic shifts coefficient for low temperature crystal phase. Since the phase transition occurs close to room temperature our founding can provide additional way for 2D perovskite properties engineering via moderate cooling achievable by Peltier coolers. Moreover the application of magnetic field to colloidal nanoplates allows us to identify transitions nature in their reach absorption spectrum. The different diamagnetic coefficients we observed point to an exciton size that can be tuned by quantum confinement effect via the variation of the thickness of nanoplatelets. Moreover the application of magnetic fields allows to easily distinguish between ground and excited excitonic states transitions.

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Critical role of interfaces in halide perovskite-based devices

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Interfaces are known to play a crucial role in semiconductor devices. This is also true for halide perovskite-based devices. Here, we take interest in two specific heterostructures through joint experimental and computational investigations. Firstly, we consider the case of the interface between MAPbI₃ and the hole transporting material NiO. We show that the great performances obtained arise from the quality of the perovskite film grown on NiO and near-ideal energy band alignment. We demonstrate that vacancies and dopants play a crucial role in that level matching [1,2].

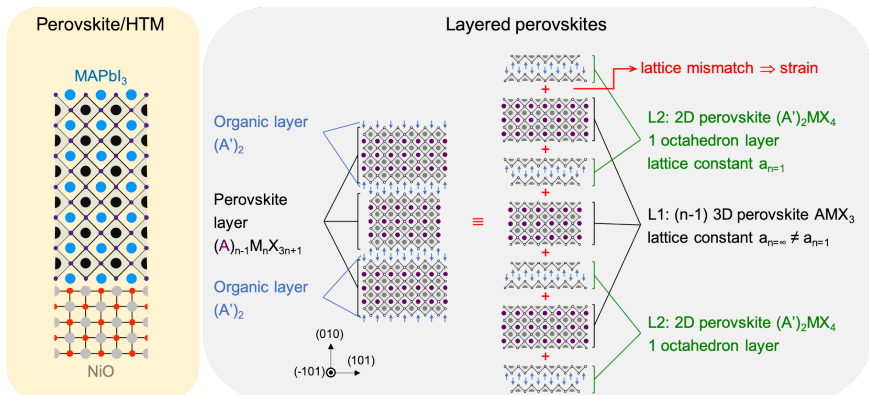


FIG. 1. Two crucial interfaces in halide perovskite devices. The MAPbI₃/NiO interface leads to very efficient solar cells. In layered perovskites, the mismatch between organic and perovskite moieties are responsible for key aspects of the device performances.

Next, we consider layered perovskites as heterostructures formed by organic layers and perovskite layers [3]. Based on experimental observations and first-principle calculations, we propose a flexoelastic model that permits rationalization of the surprising performances of layered-perovskite-based solar cells, despite intrinsic large exciton binding energies [4,5].

[1] W. Nie, M.K. *et al.*, Adv. Mater. **30**, 1703879 (2018)

[2] B. Traore, M.K. *et al.*, submitted manuscript

[3] B. Traore, M.K. *et al.*, ACS Nano **12**, 3321 (2018)

[4] J.-C. Blancon, M.K. *et al.*, Nat. Commun. **9**, 2254 (2018)

[5] M. Kepenekian *et al.*, Nano Lett. **18**, 5603 (2018)

Thursday morning

Charge carrier dynamics in thickness-controlled halide perovskite nanoplatelets

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Halide perovskite nanocrystals demonstrate intriguing optical properties such as near unity quantum yields, fast radiative recombination and wide emission tunability. Importantly, not only their size plays a role, but also the dimensionality, as previously shown for 2D nanoplatelets (NPLs), 1D nanowires and 0D quantum Dots (QDs).^{1, 2} Particularly the NPLs provide an excellent platform for understanding the fundamental properties of perovskites as their thickness can be tuned with monolayer-controlled precision.^{3, 4} Due to a reduced screening effect, not only do they exhibit extremely high exciton binding energies but also strongly enhanced carrier cooling rates.⁵ Using transient absorption and photoluminescence spectroscopy, we investigate the thickness-dependent properties of these nanoplatelets. In this presentation we present our insights into carrier relaxation, exciton-exciton annihilation, enhancing emission in the blue spectral range as well as energy transfer between NPLs of varying thickness.

- [1] J.A. Sichert et al. *Nano Lett.* 2015, 15, (10), 6521-6527.
- [2] Y. Tong et al. *Angew. Chem. Int. Ed.* 2017, 56, (44), 13887-13892.
- [3] V.A. Hintermayr et al. *Adv. Mater.* 2016, 28, (43), 9478-9485.
- [4] B.J. Bohn et al. *Nano Lett.* 2018, 18, (8), 5231-5238.
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Dimensional reduction of halide double perovskites: Insights from first principles calculations

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Dimensional reduction of the inorganic sublattice of organic-inorganic (hybrid) halide perovskites ABX_3 ($X=I^-$, Br^- , Cl^-) can be used to achieve electronic confinement in bulk solids, without the need for nanostructuring. Quasi-2D perovskites feature metal-halide sheets with n layers of an ordered perovskite lattice, separated by organic cations such as butylammonium ($BA=CH_3(CH_2)_3NH_3^+$). Lead-based quasi-2D halide perovskites [1] exhibit larger band gaps, less disperse bands and higher exciton binding energies than their 3D counterparts [2]. Double perovskites, a generalization of ABX_3 with two different B site cations, exhibit an even wider array of compositions and electronic structures, with band gaps between about 1 eV and 3.5 eV depending on composition [3,4]. Quasi-2D derivatives of the double perovskite $Cs_2AgBiBr_6$, corresponding to structures with one and two layers of alternating $AgBr_6$ and $BiBr_6$ octahedra, have recently been reported [5].

Here we present first principles calculations of quasi-2D double perovskite derivatives with $n=1-4$, using density functional theory (DFT) and Greens function-based many-body perturbation theory. For AgBi-based systems, our calculations reveal a conversion from an indirect to a direct band gap, as the inorganic lattice is thinned to a monolayer (Figure 1). We separate the effects of reduced dimensionality from those of structural distortions and strong spin-orbit coupling. Our results suggest optoelectronic properties distinctly different from typical quasi-2D lead-halide perovskites.

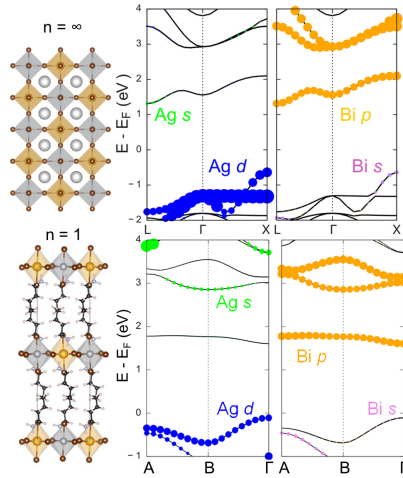


FIG. 1. DFT-PBE+SOC band structures of $Cs_2AgBiBr_6$ and the quasi-2D Ruddlesden-Popper perovskite $(BA)_4AgBiBr_6$ ($BA=CH_3(CH_2)_3NH_3^+$). Orbital contributions are shown in color. The indirect band gap of $Cs_2AgBiBr_6$ becomes direct, when the inorganic lattice is reduced to monolayer thickness.

- [1] B. Saparov *et al.*, Chem. Rev., **116**, 4558 (2016).
- [2] X. Hong *et al.*, Phys. Rev. B, **45**, 6961 (1992).
- [3] A. H. Slavney, L. Leppert *et al.*, Angew. Chem. Int. Ed. **57**, 12765 (2018).
- [4] L. Leppert *et al.*, arXiv:1903.11376 (2019).
- [5] B. A. Connor, L. Leppert *et al.*, J. Am. Chem. Soc., **140**, 5235 (2018).

Strain engineering in 2D materials: towards strain tunable optoelectronic devices

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Strain engineering is an interesting strategy to tune a material's electronic properties by subjecting its lattice to a mechanical deformation. Conventional straining approaches, used for 3D materials (including epitaxial growth on a substrate with a lattice parameter mis-match, the use of a dielectric capping layer or heavy ions implantation) are typically limited to strains lower than 2% in most cases due to the low maximum strains sustained by brittle bulk semiconducting materials. Bulk silicon, for example, can be strained only up to 1.5% before breaking. Moreover, these straining approaches induce static deformations of the semiconductor materials and therefore they are not suitable for tunable functional devices.

2D materials can be literally stretched, folded, bent or even pierced.[1] This outstanding stretchability (and the possibility of using dynamically varying strain) of 2D materials promises to revolutionize the field of strain engineering and could lead to "straintronic" devices – devices with electronic and optical properties that are engineered through the introduction of mechanical deformations.

In this talk I will discuss our recent efforts to study strain engineering in 2D materials and to exploit it to fabricate strain tunable functional optoelectronic devices.[2-6].

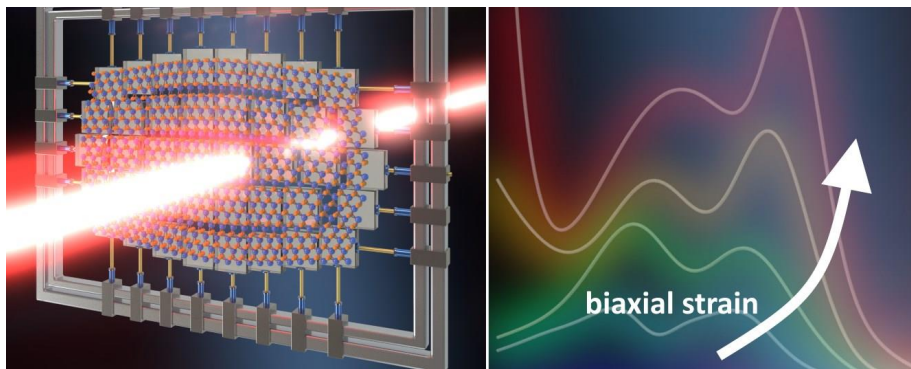


FIG. 1. (left) artistic representation of a single-layer MoS₂ photodetector subjected to biaxial strain. (right) photocurrent spectra acquired at increasingly high biaxial strain.

[1] R Roldán et al. *Journal of Physics: Condensed Matter* (2015) 27 (31), 313201

[2] A Castellanos-Gomez, et al. *Nano letters* (2013) 13 (11), 5361-5366

[3] J Querada, et al. *Nano letters* (2016) 16 (5), 2931-2937

[4] JO Island, et al. *Nanoscale* (2016) 8 (5), 2589-2593

[5] R Schmidt et al. *2D Materials* (2016) 3 (2), 021011

[6] R Frisenda, et al. *npj 2D Materials and Applications* (2017) 1 (1), 10

Evidence for Moiré Excitons in Van der Waals Heterostructures

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In vdW heterostructures formed by stacking two semiconductor monolayers, lattice mismatch or rotational misalignment introduces an in-plane moiré superlattice. While it is widely recognized that a moiré superlattice can modulate the electronic band structure and lead to novel transport properties including unconventional superconductivity and insulating behavior driven by correlations, its influence on optical properties has rarely been investigated experimentally [1-5].

We investigated the optical properties of a hexagonal boron nitride (hBN) encapsulated MoSe₂/WSe₂ heterobilayer (hBL) [2]. Due to the type-II band alignment between MoSe₂ and WSe₂, optically created electrons (holes) quickly transfer to the MoSe₂ (WSe₂) layer, leading to the formation of both intra- and interlayer excitons with very different properties (Fig. 1a). When the layers are aligned with a small twist angle and the inhomogeneous spectral linewidths are reduced with the capping layers, we observed several nearly equally spaced interlayer exciton resonances in the photoluminescence (PL) spectrum at low temperature⁴. Upon excitation with σ_1 circularly polarized light, the interlayer resonances exhibit alternating co- and cross-circularly polarized PL (Fig. 1b-c). We suggest that the alternating polarized emission originates from the atomic-scale spatial variations of the optical selection rules within a moiré supercell (Fig. 1d-e). The energy spacing and twist-angle dependence of the resonances and helicity of the emitted light are consistent with calculations of ground- and excited interlayer states confined within a moiré potential with ~ 150 meV lateral confinement, in agreement with first-principles calculations.

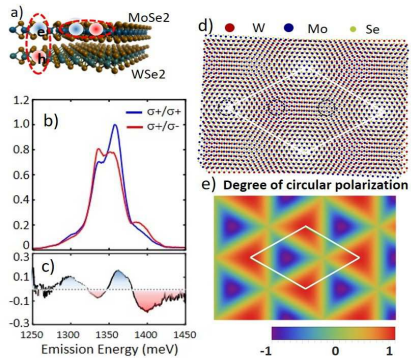


Fig. 1: (a) illustration of intra- and interlayer excitons in vdW heterobilayers; (b) multiple quantized interlayer exciton states; and (c) alternating circular emission; (d) illustration of the moiré superlattice and (e) spatially varying optical selection rules.

References:

- [1] Nano Lett. **18**, 7651–7657 (2018)
- [2] Nature **567**, 71-75 (2019);
- [5] Nature **567**, 66-70 (2019);
- [3] Nature **567**, 76-80 (2019);
- [4] Nature **567**, 81-86 (2019)

Quantum Hall Effect and Tunneling in Transition Metal Dichalcogenide Heterostructures

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Transition metal dichalcogenides (TMD) have attracted significant interest recently owing to their two-dimensional crystal structure, large band-gap, and strong spin-orbit interaction. Recent advances in sample fabrication have allowed the experimental study of low temperature magneto-transport of high mobility holes in WSe₂, and electrons in MoSe₂ and MoS₂. We review here the main results of these studies [1-4], which reveal clear quantum Hall states, and allow the extraction of fundamental electronic properties, such as carrier effective mass at the *K* and *Γ*-point band extrema, Landau level degeneracy, and spin-orbit coupling induced band splitting. The carrier density dependence on gate bias reveals negative electronic compressibility, and the quantum Hall state sequence dependence on carrier density reveals an interaction-enhanced Landau level Zeeman splitting consistent with the large carrier effective mass measured in these materials.

We also discuss the realization of rotationally controlled TMD heterostructures, consisting of two WSe₂ monolayers separated by hexagonal boron-nitride tunnel barriers. In samples where the two WSe₂ monolayers are rotationally aligned, we observe energy- and momentum-conserving (resonant) tunneling, manifested by a large conductance and negative differential resistance. In heterostructures where the two WSe₂ monolayers have a 180° relative twist, such that the Brillouin zone of one layer is aligned with the time-reversed Brillouin zone of the opposite layer, the resonant tunneling between the layers is suppressed. These findings provide evidence that, in addition to momentum, the spin-valley degree of freedom is conserved in vertical transport.

[1] B. Fallahazad *et al.*, *Phys. Rev. Lett.* **116**, 086601 (2016).

[2] H. C. P. Movva *et al.*, *Phys. Rev. Lett.* **118**, 247701 (2017).

[3] H. C. P. Movva *et al.*, *Phys. Rev. Lett.* **120**, 107703 (2018).

[4] S. Larentis *et al.*, *Phys. Rev. B* **97**, 201407(R) (2018).

[5] K. Kim *et al.*, *Nano Lett.* **18**, 5967 (2018).

Thursday afternoon

Field-Induced Superconductivity in Transition Metal Dichalcogenides

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Field effect was recently proved to be an effective way to dope various materials electrostatically. By introducing an electric double layer transistor, the electrochemical version of a conventional field effect transistor (FET), highly efficient FETs can be made with the capability of accumulating dense carriers ($\sim 10^{14}$ cm⁻²) mediated by the movement of organic ions at transistor channel surface. In this talk, I will focus on the experimental efforts using ionic gating as an effective tool to induce and control 2D superconductivity in semiconducting transition metal dichalcogenides down to one monolayer. The gate-induced carriers experiences both Rashba and Zeeman type spin-orbit interaction forming so-called Ising pairing. The spin configuration is strongly protected by the Zeeman type effective magnetic field, making this pairing state highly resilient against the in-plane magnetic field.

[1] J.M. Lu, O. Zheliuk, Q.H. Chen, I. Leermakers, N.E. Hussey, U. Zeitler, and J. T. Ye, *PNAS* 115, 3551(2018).

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Intrinsic valley Hall transport in atomically thin semiconducting transition-metal dichalcogenides

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Electrons hopping in two-dimensional honeycomb lattices possess a valley degree of freedom in addition to charge and spin. In the absence of inversion symmetry, these systems were predicted to exhibit opposite Hall effects for electrons from different valleys. Such valley Hall effects have been achieved by extrinsic means, such as substrate coupling, dual gating, and light illuminating. We realized intrinsic valley Hall transport without any extrinsic symmetry breaking in the non-centrosymmetric monolayer and trilayer MoS₂ [1]. We demonstrate experimentally that the nonlocal resistance measured from MoS₂ shows cubic scales with local resistance (see FIG. 1). Such a hallmark survives even at room temperature with a valley diffusion length at micron scale. By contrast, no valley Hall signal is observed in the centrosymmetric bilayer MoS₂. However, a DC electric field induced by dual gating can effectively break the inversion symmetry in bilayer MoS₂, resulting in valley Hall transport phenomena as evidenced by non-local transport measurement. The pronounced nonlocal signals are observed in our MoS₂ samples with length up to 16 μm and at temperature up to 300 K. Our work elucidates the topological origin of valley Hall effects and marks a significant step towards the purely electrical control of valley degree of freedom in topological valleytronics. Our observed intrinsic VHEs and their long valley diffusion lengths are promising for realizing room-temperature low-dissipation valleytronics.

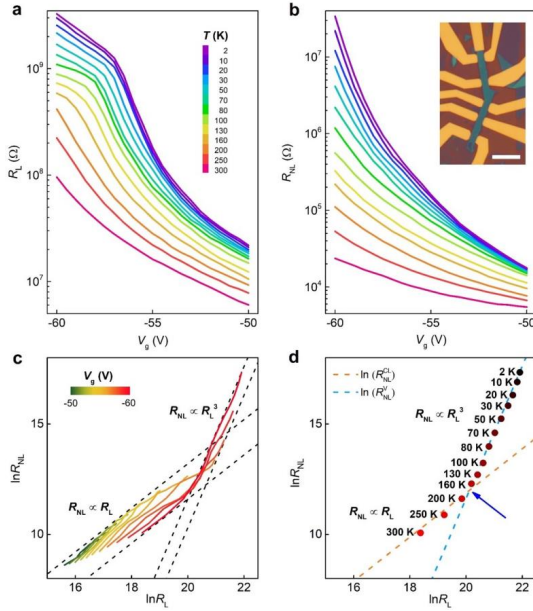


FIG. 1. Local and nonlocal resistances of monolayer MoS₂. a, b Semilog plots of RL and RNL as a function of V_g measured at varied temperatures. Inset of b: optical micrograph of our typical h-BN/MoS₂/h-BN device with multi-terminal Hall Bar configurations. Scale bar: 5 μm. c Scaling relations between ln R_L and ln R_{NL} at V_g ranging from -50 V to -60 V. When the electron density is relatively high, i.e., R_L and R_{NL} are small, R_{NL} is linearly proportional to R_L. When the electron density is relatively low, a crossover from linear to cubic scaling is observed. The critical density n_c = 4 × 10¹¹ cm⁻², with the gate voltage V_g = -57 V. d Crossover phenomenon by considering classical diffusion (R_{NL} ∝ R_L) and valley Hall transport (R_{NL} ∝ R_L³). The experimental data (solid circles, V_g = -60 V) clearly show two different regimes which are fitted by two linear curves (orange dashed line with slope 1 and blue dashed line with slope 3). The critical temperature is around 160 K~200 K, as marked by the blue arrow.

[1] Z. F. Wu, et al., “Intrinsic valley Hall transport in atomically thin MoS₂”, *Nature Comm.* 10 (2019)611. This research was support from the Research Grants Council of Hong Kong (16300717, C7036-17W).

Ultrafast Photophysics of Transition Metal Dichalcogenide Structures

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²*Center for Nanoscience and Technology, Italian Institute of Technology, Milan, Italy*

A material's functional properties are determined by its response to stimuli that drive it out of equilibrium. Some of the most important processes at the origin of such response occur on the femto- and picosecond time scale. Hence, their investigation requires appropriate time-resolved techniques such as femtosecond optical pump-probe spectroscopy. Two-dimensional semiconductors such as transition metal dichalcogenides (TMDs) bear exceptionally strong light-matter interaction and show great perspective for photonic, optoelectronic and light harvesting applications. Their optical response goes far beyond the single-particle picture, involving multi-particle excitations including excitons, trions, and biexcitons, as well as many-body effects such as band gap renormalization. Here, we disentangle these contributions to TMDs' optical response and extract the underlying electronic processes in selected examples including mono- and multilayer MoS₂ and WS₂, WS₂ nanotubes, and MoSe₂/WSe₂ heterostructures (HSs).

In mono- and multilayer flakes we find that during the first few ps the optical response is dominated by two antagonistic many-body effects, band gap renormalization and a screening of the Coulomb interaction that determines the exciton binding energy. As the hot electron-hole pairs relax towards the band minima (excitons at the K point in the monolayer, weakly bound electron-hole pairs at different k values in the multilayer case), Pauli blocking increasingly governs the transient optical spectra.

In WS₂ nanotubes, the cylindrical shape provides an intrinsic optical cavity whose resonant modes couple strongly to the electronic resonances. We develop a phenomenological coupled oscillator model with time-dependent parameters to describe the transient extinction spectra, allowing us to extract the underlying non-equilibrium electron dynamics. We find that, although resulting in very different spectral response due to the strong coupling, the electronic processes and their characteristic times in the nanotubes are very similar to those of layered multilayer TMDs.

Type II HSs, where the valence band maximum and the conduction band minimum are in separate layers, allow efficient charge separation, which is particularly beneficial for photovoltaics based on atomically thin materials. We study the charge transfer dynamics in a type II WSe₂/MoSe₂ HS. The pump beam, which is tuned to the smaller optical gap of the two monolayers (that of MoSe₂), injects excitons into the K/K' valleys of MoSe₂. We observe a clear delayed formation of the WSe₂ bleaching signal, which we attribute to a fast interlayer hole transfer occurring in less than 1 ps. The hole transfer yields an interlayer exciton (i.e. a bound state of an electron and a hole residing in two separate layers). Moreover, time resolved circular dichroism shows that the valley polarization is transferred from the excited layer to the adjacent one. The relaxation dynamics of the interlayer exciton is considerably slower (i.e. ~1 ns) compared to the excitons in isolated monolayers.

Hot carrier optoelectronic devices based on van der Waals heterostructures

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Strong many-body interactions in 2D semiconductors result in efficient exciton–exciton annihilation (EEA), a four-body interaction involving the energy and momentum transfer between two holes and two electrons [1]. The resulting high-energy charge carriers are effectively at the surface and therefore can be transferred to the adjacent van der Waals layer at an ultrafast rate [2]. While the EEA effect has been identified experimentally in ultrafast transient absorption measurements its manifestation in the photocurrent has not been investigated. In this talk, I will discuss unconventional photoresponse in van der Waals heterostructure devices due to hot carriers derived from EEA in monolayer transition metal dichalcogenide (TMDs) [3]. We show that the photocurrent results from ultrafast transfer of Auger-induced high energy holes from monolayer TMD to the valence band of hexagonal boron nitride (hBN). Simultaneous measurements of photoluminescence quantum yield and photocurrent external quantum efficiency reveal that EEA play an important role above the generation rate of $10^{19} \text{ cm}^{-2}\text{s}^{-1}$.

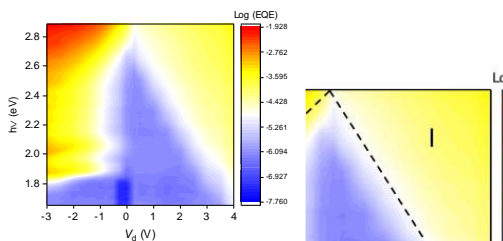


FIG. 1. External quantum efficiency (EQE) of photocurrent in metal-insulator-semiconductor (MIS) heterostructure (left), and illustration of photocurrent generation mechanism associated with EEA (right).

[1] Mouri et al. “Nonlinear photoluminescence in atomically thin layered WSe₂ arising from diffusion-assisted exciton-exciton annihilation” *Phys. Rev. B* 90, 155449 (2014)

[2] Hong et al. “Ultrafast charge transfer in atomically thin MoS₂/WS₂ heterostructures” *Nature Nanotech.* 9, 682 (2014).

[3] Linardy et al. Manuscript in preparation.

Control of the composition and the surface chemistry of II-VI semiconductor nanoplatelets

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2D colloidal II-VI semiconductor nanoplatelets (NPLs) are a unique class of nanoparticles with optical properties comparable to quantum well.¹ Their thicknesses, the unique confined direction, are defined at the atomic scale all along the nanoparticles without roughness which provide them optical properties without inhomogeneous broadening. In a synthesis all the NPLs present exactly the same number of monolayers. An effort has been dedicated to the growth of cadmium chalcogenides NPLs and their heterostructures. Indeed due to their shapes, these NPL can either be grown parallel to the confined direction leading to core/shell or perpendicular to the confined direction leading to core/crown NPLs.² In order to extend the optical properties of the NPLs, cation exchange has been proposed as a strategy to synthesis mercury chalcogenides nanoplatelets. Compare to spherical quantum dots emitting in the same range of wavelength, the HgTe NPLs present a similar PL efficiency (10%) but with a far narrower emission (60meV for an emission at 880nm) and shortest decay rate.³

In addition of the control of their compositions and quantum confinement, a control of the surface chemistry is another way to control the optical properties. Indeed NPLs can be stabilized by halides that act as X-L-type ligands, costabilized by amines which provide colloidal stability in nonpolar solvents.⁴ The hydrogen from the amine can participate in a hydrogen bond with the lone pair electrons of surface halides. In the case of CdSe, the improved surface passivation leads to an increase in the fluorescence quantum efficiency of up to 70% in the case of bromide. This ligand exchange is associated with a decrease in stress that leads to unfolding of the NPLs, which is particularly noticeable for iodide-capped NPLs.

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2. Nasilowski, M., Mahler, B., Lhuillier, E., Ithurria, S. & Dubertret, B. Two-Dimensional Colloidal Nanocrystals. *Chem. Rev.* **116**, 10934–10982 (2016).
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4. Dufour, M. *et al.* Halide Ligands to Release Strain in Cadmium Chalcogenide Nanoplatelets and Achieve High Brightness Laboratoire de Physique et d' Etude des Matériaux , ESPCI - Paris , PSL Research University , Sorbonne Université , CNRS , Institut des NanoSciences de Pari. *ACS Nano* **13**, 5326–5334 (2019).

Screening at the Surface of Black Phosphorus: Anisotropy, Doping, and Magnetism

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The layered semiconductor, black phosphorus (BP), has recently gained substantial momentum within the fields of physics, electronics and optoelectronics because of its layer-dependent band gap spanning the visible spectrum, high carrier mobility, strongly bound particle-hole excitations, and potentially anisotropic quantum Hall phases. Yet despite the fundamental role of screening in each of these material properties, the atomic scale dielectric response of BP remains largely unexplored.

In this talk, I will discuss our recent studies of electrostatic screening at the atomic scale on the surface of black phosphorus [1]. Using scanning tunneling microscopy to characterize the distributions of charged alkali adatoms, we quantify the electrostatic potential landscape arising from screened Coulomb interactions and Friedel-mediated charge carrier interactions. In conjunction with high-resolution angle resolved photoemission studies of the electronic band structure and ab initio calculations of the various energy scales, we are able to fully characterize the strongly anisotropic screening behavior at the black phosphorus surface. The extremely long-ranged, anisotropic screening has significant implications for charged impurity scattering in transport and excitonic quasiparticles in black phosphorus.

I will then provide an outlook to utilize the screening in black phosphorus to manipulate and stabilize multiple orbital configurations ($3d^8$ and $3d^9$) in a single Co atom adsorbed on the BP surface [2]; thus enabling a proof-of-concept single atom magnetic memory.

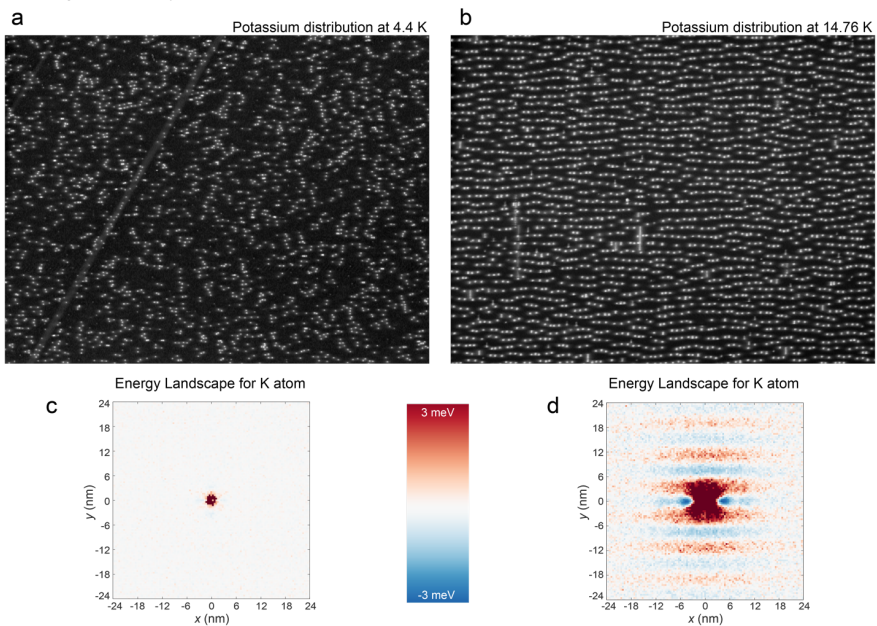


FIG. 1. Distribution of potassium atoms on black phosphorus (a) and their energy landscape (c) upon deposition at $T = 4.4\text{K}$. Distribution of potassium atoms (b) and their energy landscape (d) after annealing to $T = 14.76\text{K}$.

[1] Kiraly et. al. *arXiv cond-mat.str-el*, 02627 (2019).

[2] Kiraly et. al. *Nature Commun.* 9, 3904 (2018).

Friday morning

Additive-free MXene inks and direct printing of micro-supercapacitors

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Direct printing of functional inks is critical for applications in diverse areas including electrochemical energy storage, smart electronics and healthcare. However, the available printable ink formulations are far from ideal.

Either surfactants/additives are typically involved or the ink concentration is low, which add complexity to the manufacturing and compromises the printing resolution.

Here, we demonstrate two types of two-dimensional titanium carbide (Ti₃C₂T_x) MXene inks, aqueous and organic in the absence of any additive or binary-solvent systems, for extrusion printing and inkjet printing, respectively.

We show examples of all-MXene-printed structures, such as micro-supercapacitors, conductive tracks and ohmic resistors on untreated plastic and paper substrates, with high printing resolution and spatial uniformity.

The volumetric capacitance and energy density of the all-MXene-printed micro-supercapacitors are orders of magnitude greater than existing inkjet/extrusion-printed active materials.

The versatile direct-ink-printing technique highlights the promise of additive-free MXene inks for scalable fabrication of easy-to-integrate components of printable electronics.

Program of the 7th International Conference Flatlands beyond Graphene

2 – 6 September 2019, Toulouse, France

Hotel Mercure Atria, Compans Cafferelli, 31000 Toulouse

	Monday	Tuesday	Wednesday	Thursday	Friday
9h00		Damien Voiry	Andras Kis	J. Maultzsch	Ning Wang
9h30		Alla Zak	Jun Lou	Thomas Heine	C. Gadermaier
10h00		Doron Naveh	K.S. Thygesen	M. Baranowski	Goki Eda
10h30		Coffee	Coffee	Coffee	Coffee
11h00		U. Wurstbauer	Efrat Liftshitz	M. Kepenekian	S. Ithurria
11h30		S. Magorrian	Claudia Backes	A. Urban	Brian Kiraly
12h00		Tobias Korn	Liyang Jiao	Lin Leppert	V. Nicolosi
12h30		Buffet Lunch	Buffet Lunch	Buffet Lunch	Closing
14h00		A. Chernikov	Excursion to Albi	A.C. Gomez	
14h30		Misha Glazov		Xiaoqin Li	
15h00	Registration	Mark Goerbig		E. Tutuc	
15h30	Coffee	Ermin Malic		Jianting Ye	
16h00	Opening	Coffee		Coffee	
16h30	T. Mueller	Shotgun 1		Aperitif + Banquet in Albi	Shotgun 2
17h00	Zdenek Sofer	+	+		
17h30	Omer Yaffe	Poster Session	Poster Session		
18h00	J. Robinson	+	+		
18h30	Sami Mitra	Cocktail dinatoire	Cocktail dinatoire		
19h00	Welcome reception NOVOTEL				
21h00			Bus to		
22h30			Hotel Mercure		

Invited talks - 25 minutes + 5 minutes for questions/discussion

Shotgun – strictly 3 minutes/ 3 slides to advertise poster