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HUMBOLDT-UNIVERSITÄT ZU BERLIN

Collaborative Research Centre 951

HIOS

Hybrid Inorganic/Organic Systems for Opto-Electronics

Symposium 2022

06.10. — 07.10.2022

Campus Adlershof

Berlin, Germany

Book of Abstracts

TU berlin

Universität Potsdam

Freie Universität Berlin

MAX-PLANCK-GESELLSCHAFT
FRITZ-HABER-INSTITUT

HZB Helmholtz
Zentrum Berlin

Welcome!

It is our pleasure to welcome you to the fourth international symposium on Hybrid Inorganic/Organic Systems for Opto-Electronics (HIOS). Hosted by the Collaborative Research Centre (CRC) 951, the HIOS Symposium will provide an open and creative platform for the scientific exchange of the latest achievements in the exciting field of hybrid inorganic/organic systems, comprising inorganic semiconductors, conjugated organic materials, and metal nanostructures.

During two days, fourteen invited talks, presented by international experts and by members of the CRC 951, will cover HIOS structure and morphology, electronic structure and control of inorganic/organic interfaces, charge and energy transfer, hybrid optical excitations, plasmonic phenomena, and much more. The scientific program is complemented by a poster session offering networking opportunities for all participants.

We are looking forward to see you in Berlin-Adlershof and we hope that you will enjoy your time at the **HIOS SYMPOSIUM 2022**.

With best regards,

Oliver Benson

Norbert Koch
Spokesperson CRC 951

Oliver Benson
Vice-Spokesperson CRC 951

4th Symposium on

Hybrid Inorganic/Organic Systems for Opto-Electronics 2022

October 06th – 07th, 2022; Berlin-Adlershof

Thursday, October 06th

09:00 – 09:55	Registration
09:55 – 10:00	Welcome
10:00 – 10:30	**Ralph Ernstorfer**, FHI Berlin *Momentum-resolved view on singlet fission and exciton dynamics in molecular crystals and heterostructures*
10:30 – 11:00	**Seth Marder**, U Colorado Boulder *Interface Chemistry for Hybrid Organic Inorganic Electronics and Opto-electronics*
11:00 – 11:30	Coffee Break
11:30 – 12:00	**Andreas Knorr**, TU Berlin *Excitation Transfer in Functionalized Atomically Thin Materials*
12:00 – 12:30	**Goki Eda**, NUS *Quantum engineering of 2D semiconductors*
12:30 – 15:00	Lunch & Poster Session
15:00 – 15:30	**Claudia Draxl**, HU Berlin *Opto-electronic excitations in TMDC-based systems explored by many-body theory*
15:30 – 16:00	**Mariana Rossi**, MPSD Hamburg *Understanding Defects and Molecular Adsorbates on Monolayer TMDC*
16:00 – 16:30	**Katharina Franke**, FU Berlin *Anchoring molecular adsorbates to S defects on monolayers of MoS2 on Au(111)*
16:30 – 17:00	Coffee Break
17:00 – 17:30	**Kurt Busch**, HU Berlin *Modelling resonator-based active nano-photonic functional elements*
17:30 – 18:00	**Emil List-Kratochvil**, HU Berlin *Electronic and Photonic Neuromorphic Device Concepts Based on HIOS*

Friday, October 07th

10:00 – 10:30	**Cinzia Casiraghi**, U Manchester *Water based 2D material inks: from printed electronics to biomedical applications*
10:30 – 11:00	**Dieter Neher**, U Potsdam *Charge transfer across the hybrid TMDC/organic interface*
11:00 – 11:30	Coffee Break
11:30 – 12:00	**Kirill Bolotin**, FU Berlin *Generating and exploring ultrastrong electric field via molecular gating*
12:00 – 12:30	**Benjamin Schwartz**, UCLA *Understanding and controlling the mobility of carriers in chemically-doped semiconducting polymers*
12:30 – 13:00	**Oliver Benson**, HU Berlin *Chiral coupling in hybrid plasmonic nanostructures*
	Closing

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Invited talks

The oral presentations will be held in the
Erwin-Schrödinger-Zentrum at Campus Adlershof, Berlin:
Room 0'119, Rudower Chaussee 26, 12489 Berlin.

Momentum-resolved view on singlet fission and exciton dynamics in molecular crystals and heterostructures

Ralph Ernstorfer

Fritz Haber Institute of the Max Planck Society, Berlin (Germany)

Singlet fission may boost photovoltaic efficiency by transforming a singlet exciton into two triplet excitons and thereby doubling the number of excited charge carriers. The primary step of singlet fission is the ultrafast creation of the correlated triplet pair. While several mechanisms have been proposed to explain this step, none has emerged as a consensus. The challenge lies in tracking the transient excitonic states. We use time- and angle-resolved photoemission spectroscopy [1] to observe the primary step of singlet fission in crystalline pentacene and gain intimate knowledge about the localization and the orbital character of the exciton wavefunctions recorded in momentum maps [2]. This allowed us to directly compare the localization of singlet and bitriplet excitons and decompose energetically overlapping states based on their orbital character. We find direct evidence of the charge-transfer character of the singlet state and show that the fission process occurs in a charge-transfer-mediated mechanism. We apply this experimental approach also to study exciton dynamics [3] in the van der Waals heterostructures monolayer-WSe2-graphene and reveal the competition between charge and energy/exciton transfer process as well as a previously not observed intraband Meitner-Auger energy transfer process [4].

[1] M. Puppin et al., Rev. Sci. Inst. 90, 23104 (2019).
[2] A. Neef et al., arXiv:2204.06824
[3] S. Dong et al., Natural Sciences 1, e10010 (2021).
[4] S. Dong et al., arXiv:2108.06803

Interface Chemistry for Hybrid Organic Inorganic Electronics and Opto-electronics

Seth R. Marder

Renewable and Sustainable Energy Institute, Department of Chemical and Biological Engineering, Department of Chemistry, and Materials Science Program, University of Colorado Boulder, Boulder (USA)

Organic semiconductors have attracted interest for electronic applications due to their potential for use in low-cost, large-area, flexible electronic devices. Here we will report on recent developments pertaining to surface modifiers and dopants that could impact the charge injection/collection processes in organic light emitting diodes, organic field effect transistors, and organic photovoltaic and HOIS devices. In particular, we will examine how phosphonic acids assemble on ITO substrates and can be used to enhance device performance in a variety of ways. I will also briefly outline some studies on the development of 1D and 2D HOIS perovskite materials time permitting.

Excitation Transfer in Functionalized Atomically Thin Materials

Andreas Knorr

Nichtlineare Optik und Quantenelektronik von Halbleitern, Institut für Theoretische Physik,
Technische Universität Berlin, Berlin (Germany)

The decoration of atomically thin materials with nanostructures allows to study a variety of excitation phenomena at two dimensional interfaces. This includes different charge-, Dexter- or Förster-type transfer processes in heterostructures of atomically thin semiconductors functionalized with molecules, metal nanoparticles and graphene. In this talk, we discuss an approach to the theoretical description of different excitation transfer and strong coupling processes after optical excitation and the formation of hybrid excitations, their propagation and decay. Throughout the talk, a comparison with recent experiments (absorption, luminescence and angle resolved photoemission spectroscopy) will be provided.

Quantum engineering of 2D semiconductors

Goki Eda

Department of Physics, National University of Singapore, Singapore
Department of Chemistry, National University of Singapore, Singapore
Centre for Advanced 2D Materials, National University of Singapore, Singapore

Point defects in solids are often sources of optically accessible quantum states. Color centers in diamond and SiC are well-known examples with great potential for quantum technologies. In the past few years, defects in 2D semiconductors received considerable interest following the initial reports on the observation of single photon emission [1]. The identity of defects responsible for the quantum emission, however, remains unknown. In this talk, I will discuss our recent efforts to generating optically active dilute atomic defects in 2D semiconductors with the aim to accessing their quantum nature [2]. One example is the impurity doping of monolayer WS_2, which is found to result in bright sub-gap emission even at ppm concentrations [3]. We show that such emission arises from ionized-acceptor-bound excitons, a three-body charge complex analogous to a negative trion. Interestingly, these bound exciton complexes exhibit sizeable valley polarizability, reflecting their partial free exciton character. We further discuss a scanning probe approach to rapidly quantifying impurities in the ultra-dilute limit ($< 10^{10}$ cm-2) in ambient conditions [4].

[1] Perebeinos, Nature Nanotech. 10, 485 (2015).
[2] Loh et al. Nano Lett. 21, 5293 (2021).
[3] Loh et al. Manuscript in preparation.
[4] Nam et al. Manuscript in preparation.

Opto-electronic excitations in TMDC-based systems explored by many-body theory

Claudia Draxl

Institut für Physik & IRIS Adlershof, Humboldt-Universität zu Berlin, Berlin (Germany)

Organic-inorganic hybrid systems are fascinating materials from a theory point of view as various interactions take place on the same energy scale. Here, we investigate the interplay between electron-electron interaction, electron-hole correlations, and spin-orbit coupling (SOC). With the examples of pyrene and pyridine on MoS2, we discuss the level alignment at the interface. In both cases, semi-local density-functional theory finds type-II alignment. In contrast, owing to the mutual dynamical screening of the interface constituents, considering electron-electron interactions on the GW level of many-body perturbation theory, it is changed to type I. The correct band structure, in turn, is the basis for exploring many-body interactions and exciton physics in these low-dimensional structures. Solving the Bethe-Salpeter equation, we find besides intra-layer excitons on the MoS2 side, also hybrid and charge-transfer excitons at the interface, both in the visible range of the optical spectrum. Moreover, we study how SOC and a gold substrate impact the excitations in MoS2. All calculations are performed with the full-potential all-electron computer package exciting that employs linearized augmented planewave + local-orbital (LAPW+lo) basis sets.

Understanding Defects and Molecular Adsorbates on Monolayer TMDC

Mariana Rossi

Max-Planck-Institut für Struktur und Dynamik der Materie, Hamburg (Germany)

Anchoring molecular adsorbates to S defects on monolayers of MoS2 on Au(111)

Katharina J. Franke[1], J. Rika Simon[1], Christian Lotze[1], Paul Wiechers[1], Juan Pablo Guerrero Felipe[1], Dmitrii Maksimov[2], Ana M. Valencia[3,4], Caterina Cocchi[3,4], Björn Kobin[4], Stefan Hecht[4], Mariana Rossi[2]

[1] Freie Universität Berlin, Berlin (Germany)
[2] MPI for the Structure and Dynamics of Matter, Hamburg (Germany)
[3] Carl von Ossietzky Universität Oldenburg, Oldenburg (Germany)
[4] Humboldt-Universität zu Berlin, Berlin (Germany)

Monolayers of transition-metal dichalcogenides are highly promising candidates for optoelectronic devices. Special attention has to be devoted to defects in these two-dimensional materials as already a minute amount dramatically influences the electronic properties, most notably by the introduction of subgap states.

Here, we grow monolayers of MoS2 on a Au(111) surface and characterize their atomic-scale properties with scanning tunneling microscopy and spectroscopy at low temperature under ultra-high vacuum conditions. Hybridization of the S states with the metal substrate leads to a reduced semiconducting band gap as compared to a free-standing layer. Physisorbed molecules on a monolayer MoS2 are very well decoupled electronically from the Au substrate due this band gap. The molecules thus preserve their gas-phase properties.

To explore the influence of chemisorbed molecules on the properties of MoS2, we introduce S vacancies into the top layer. These exhibit a Kondo resonance at the Fermi level reflecting the presence of an unpaired electron spin. We then deposit the thiol-based molecule CF3-3P-SH (tri-fluoro-methyl-p-terphenyl-thiol) on the defect-rich surface. Many of these molecules attach to the S vacancies. We find two categories of anchored molecules, those with and without a Kondo resonance. Density-functional theory calculations reveal that singly-occupied defects states persist when a dehydrogenated molecule is attached, while the hydrogen-terminated ones lead to doubly occupied defect states. These results explain the presence/absence of a Kondo resonance in experiment.

Modelling resonator-based active nano-photonic functional elements

Kurt Busch

AG Theoretische Optik & Photonik, Humboldt-Universität zu Berlin, Berlin (Germany)
Max Born Institute, Berlin (Germany)

Micro-resonators represent fundamental ingredients for the realization of active nano-photonic functional elements. Ordinarily, they come in two forms, the standing- and the travelling-wave type. In this talk, several interesting enhancements of traditional resonator-based systems will be discussed. Specifically, planar Bragg-resonators can be enhanced by introducing by metallic gratings, thereby introducing distributed-feedback and defect-engineering concepts which, in turn, modify the lasing characteristics in such systems [1]. Similarly, judicious designs of symmetric coupled micro-ring resonators with uniform (linear) gain can reverse the optical energy distribution inside the structure, thereby providing a novel tuning knob [2]. Moreover, an intermediate link between the aforementioned resonator types exists. This novel class of photonic resonators supports hybrid optical modes, where, at one location along the resonator, the electromagnetic fields associated with such a mode feature a purely standing-wave pattern, while at a different location, the fields of the same mode represent a pure traveling wave character [3].

[1] M. Kliem et al., Defect-State Lasing in Photonic Lattices of Metal–Organic Microcavities,
Adv. Photonics Res. 2, 2000116 (2021).
[2] A. Hashemi et al., Uniform optical gain as a non-Hermitian control knob, submitted (2022).
[3] Q. Zhong et al., The missing link between standing- and traveling-wave resonators, Nanophotonics 11, 4427 (2022).

Electronic and Photonic Neuromorphic Device Concepts Based on HIOS

Emil J. W. List-Kratochvil

Institut für Physik, Institut für Chemie & IRIS Adlershof, Humboldt-Universität zu Berlin, Berlin (Germany)

Artificial neural networks (ANN), inspired by biological nervous systems, enable signal processing beyond the capabilities of von Neumann computer architectures. Through dynamically adapting the connectivity (synaptic weights) in individual devices and by applying learning algorithms ANNs can offer in memory and tensor computing capabilities. Yet, to fully unleash the potential of hardware ANNs there is still a need for neuromorphic device concepts, which properly emulate all necessary synaptic functions adequately and allow for an easy integration into large scale hardware ANNs. In this contribution we will demonstrate electronic as well as plasmonic/photonic neuromorphic device concepts using different types of hybrid material systems. We will show that all presented devices concepts can be used to mimic fundamental functions of a synapse, such as long-term plasticity showing potentiation and depression, spike-time or spike-rate dependent plasticity or may be trained to realize AND- or OR logic gate operations.

This work was supported by the Deutsche Forschungsgemeinschaft through the CRC 951.

Water based 2D material inks: From printed electronics to biomedical applications

Cinzia Casiraghi

Department of Chemistry, University of Manchester, Manchester (United Kingdom)

Solution processing of 2D materials [1] allows simple and low-cost techniques, such as ink-jet printing, to be used for fabrication of heterostructure-based devices of arbitrary complexity. However, the success of this technology is determined by the nature and quality of the inks used. Our group has developed highly concentrated, defect-free, printable and water-based 2D crystal formulations, designed to provide optimal film formation for multi-stack fabrication [2]. I will give examples of all-inkjet printed heterostructures, such as large area arrays of photosensors on plastic [2], programmable logic memory devices [2], capacitors [3] and transistors on paper [3,4]. Furthermore, inkjet printing can be easily combined with materials produced by chemical vapor deposition, allowing simple and quick fabrication of complex circuits on paper, such as high-gain inverters, logic gates, and current mirrors [5]. Our formulation approach also allows to easily tune the charge of graphene, which is a key parameter in biomedical applications. [6-9].

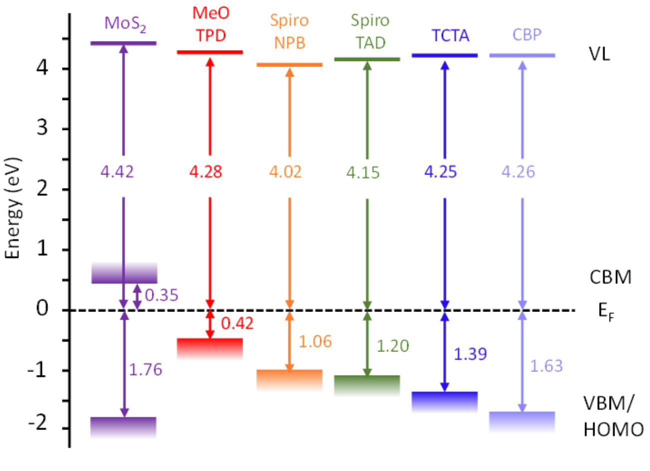
[1] Coleman et al, Science 331, 568 (2011).
[2] McManus et al, Nature Nano, 12, 343 (2017).
[3] Worsley et al, ACS Nano, 2018, DOI: 10.1021/acsnano.8b06464.
[4] Lu et al, ACS Nano, ACS Nano, 13, 11263 (2019).
[5] Conti et al, Nature comms, 11 (1), 1-9 (2020).
[6] Shin et al, Mol. Syst. Des. Eng., 2019, DOI:10.1039/C9ME00024K
[7] Shin et al, Faraday Discussion, https://doi.org/10.1039/C9FD00114J
[8] Shin et al, Nanoscale, 2020, DOI: 10.1039/D0NR02689A
[9] Tringides et al, Nature Nano, 2021 doi.org/10.1038/s41565-021-00926-z

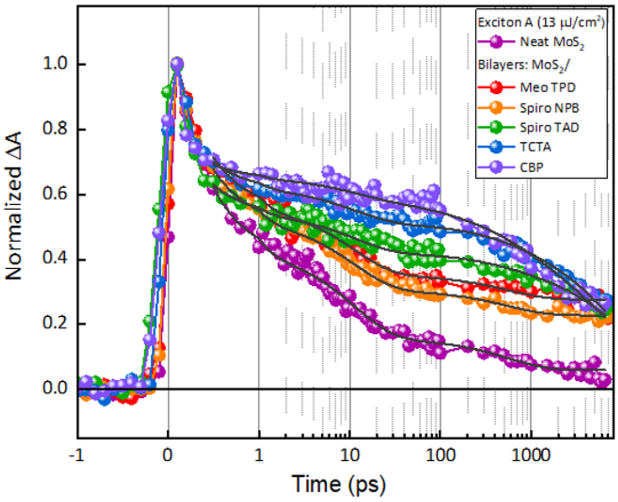
Charge transfer across the hybrid TMDC/organic interface

Dieter Neher[1], Meysam Raoufi[1], Sreelakshmi Chandrabose[1], Steffen Rühl[2], Sylke Blumstengel[2], Emil List-Kratochvil[2], Rongbin Wang[2], Norbert Koch[2]

[1] Institute of Physics and Astronomy, University of Potsdam, Potsdam (Germany)
[2] Institut für Physik & IRIS Adlershof, Humboldt-Universität zu Berlin, Berlin (Germany)

As direct bandgap semiconductors with high carrier mobility and absorption cross section, monolayer transition metal dichalcogenides (ML-TMDCs) are promising materials for optoelectronic applications. Here, we study the photoinduced charge transfer in hybrid systems combining ML-MoS_2 and ML-WSe_2 with selected organic semiconductors. The dynamics and fate of photogenerated species is monitored through transient absorption, photoluminescence and photocurrent spectroscopy. For a hybrid system with systematically tuned interfacial energy offset, we find that the efficiency of charge transfer is consistent with Marcus-theory. Hereby, a larger offset goes along with a lower efficiency of photoinduced transfer but also a longer lifetime of the charge separated state. These data rule out direct electron-hole recombination as the main decay pathway for photogenerated carriers in the considered system. For the combination of a novel non-fullerene acceptor with WSe_2, rapid triplet formation comes instead into play as an additional loss channel, driven by interfacial non-geminate recombination. Interestingly, this loss channel can be fully suppressed upon fluorination of the organic compound. Finally, intensity modulated photocurrent spectroscopy highlights the role of traps in determining the strength and kinetics of persistent photoconductivity. Frequency and intensity-dependent data are fully consistent with photodoping via trapping of photogenerated holes. Compared to neat MoS_2, the bilayer of ML-MoS_2 with H_2Pc exhibits an improved photoresponse speed and responsivity, which we assign to efficient interfacial charge transfer and trap filling.





Generating and exploring the effects of ultrastrong electric field in HIOS via molecular gating

Kirill Bolotin

Institut für Physik, Freie Universität Berlin, Berlin (Germany)

We consider a new type of HIOS: a 2D material with donor molecules on one side and acceptor molecules on the other. The charge transfer between the molecules generates electrical field inside the 2D material. We experimentally measure the field produced by this "molecular gating" and show that it reaches >4V/m, the highest field measured in a solid-state device. We then explore the effect of this intense field. First, we demonstrate that the field suffices to modify and eventually completely close the bandgap in bilayer 2D semiconductors driving a semiconductor to metal transition. Second, we show that the field lifts degeneracies between excitons in semiconductor 2D bilayer and brings different excitons into resonance. We demonstrate new types of excitonic states that arise because of that.

Understanding and controlling the mobility of carriers in chemically-doped semiconducting polymers

Benjamin Schwartz

Department of Chemistry & Biochemistry, University of California, Los Angeles (USA)

Chemical doping of conjugated polymers is a viable method for tuning the properties of organic electronic devices such photovoltaics and thermoelectrics. A lot work has gone into understanding the nature of the carriers produced upon doping, and the fact that the carrier delocalization (degree of polaron coherence) depends sensitively on the degree of order in the polymer and the coulomb interactions between the polymer and the dopant counterion. One of the most common molecular dopants for conjugated polymers is 2,3,5,6-tetrafluoro-7,7,8,8-tetracyanoquinodimethane (F4TCNQ), which is a powerful oxidizing agent that can easily create polarons on conjugated polymers such as poly(3-hexylthiophene-2,5-diyl) (P3HT). The nitrile vibrational modes of the F4TCNQ anion are observed to occur across a range of different frequencies in different samples of doped P3HT. Here, we show that these nitrile frequency shifts can be understood as resulting from the vibrational Stark effect. F4TCNQ– serves as a sensitive vibrational Stark probe and its nitrile stretches provide a direct read-out of the locally-experienced electric fields from the nearby polaron on the doped polymer backbone. Thus, the vibrational spectroscopy of F4TCNQ– in doped P3HT provides direct experimental access to the degree of polaron coherence on the nearby polymer. This provides the first direct experimental measurement of polaron delocalization in doped conjugated polymer films, and verifies theoretical models relating polaron coherence to the shape of the doped polymer's mid-IR electronic absorption spectrum. We then show that we can control the extent of polaron delocalization by using specially-designed dopants that break the coulomb interaction between the polaron and the dopant counterion.

Chiral coupling in hybrid plasmonic nanostructures

Oliver Benson

Nanooptik, Institut für Physik, Humboldt-Universität zu Berlin, Berlin (Germany)

Spin-momentum locking is a peculiar effect in the near-field of guided optical or plasmonic modes. It can be utilized to map the spinning or handedness of electromagnetic fields onto the propagation direction. This motivates a method to probe the circular dichroism of an illuminated chiral object. In this presentation we introduce local, sub-diffraction limited chiral coupling of light and propagating surface plasmon polaritons in a self-assembled system of a gold nanoantenna and a silver nanowire [1]. We perform characterization of our systems through correlated electron microscopy, energy-dispersive X-ray spectroscopy, dark-field, and fluorescence imaging. Three-dimensional numerical simulations support the experimental findings. We discuss the potential of the hybrid system for as an ultracompact sensor for chiral response far below the diffraction limit.

[1] Self Assembly of Plasmonic Nanoantenna-Waveguide Structures for Subdiffractional Chiral Sensing, M. Rothe, Y. Zhao, J. Müller, G. Kewes. C. T. Koch, Y. Lu and O. Benosn, ACS Nano 15, 351 (2021)

Poster session

The Poster session will take place in the
Erwin-Schrödinger-Zentrum at Campus Adlershof, Berlin:
Room 0’101, Rudower Chaussee 26, 12489 Berlin.

01 Emergence of a singly-occupied state of *p*-terphenly-based thiols bound to sulphur defects on MoS_2/Au(111)

J. Rika Simon[1], Dmitrii Maksimov[2], Juan Pablo Guerrero Felipe[1], Paul Wiechers[1], Christian Lotze[1], Ana M. Valencia[3,4], Caterina Cocchi[3,4], Björn Kobin[4], Stefan Hecht[4], Mariana Rossi[2] and Katharina J. Franke[1]

[1]Freie Universität Berlin, Berlin (Germany)
[2]MPI for the Structure and Dynamics of Matter, Hamburg (Germany)
[3]Carl von Ossietzky Universität Oldenburg, Oldenburg (Germany)
[4]Humboldt-Universität zu Berlin, Berlin (Germany)

The combination of transition-metal dichalcogenides (TMDCs) and organic molecules into hybrid inorganic-organic systems is a field gathering much interest in recent years. The use of submonolayers of the TMDC MoS_2 as a decoupling layer in an STM junction is already well established and allows highly resolved $\mathrm{d}I/\mathrm{d}V$ spectra. But 2D materials also have drawbacks: Their properties are highly dependent on their local structure, because defects influence their properties severely.

Here we utilise these defects by anchoring the thiol-based molecule CF3-3P-SH (trifluoromethyl-*p*-terphenyl-thiol) into purposely created top-layer sulphur point defects in MoS_2 on Au(111). One end-group of the anchored molecule is bound to the defect, allowing it to rotate around the anchoring point. On such molecules we observe a Kondo resonance. *Ab initio* molecular dynamics simulations show the emergence of a singly-occupied molecular state near E_F depending on the configuration of the molecule with respect to the surface, which in turn gives rise to the observed Kondo resonance.

02 The geometries of the point defects under study for MX_2 M=W and X=Se, S. AddX is adatom X, VX/M is mono X/M vacancy, VX2 is di-vacancies at the top and bottom coincident lattice sites and VX22 is di-vacancies at neighboring sites.

Alaa Akkoush[1], Mariana Rossi[1]

[1]Institut für Chemie and IRIS Adlershof, Humboldt-Universität zu Berlin, Berlin (Germany)

Defects can strongly influence the electronic, optical and mechanical properties of 1D materials. However, their stability and distribution under different conditions of temperature, pressure and strain are not well characterized from an atomistic perspective. We have investigated the structural and electronic properties, as well as the thermodynamic stability of point defects (vacancies and adatoms) in monolayer transition metal dichalcogenides MX_2 with $M = Mo/W$ and $X = S/Se$, through density-functional theory (DFT) simulations with hybrid exchange correlation functional, as implemented in the all-electron package FHI-aims [1]. These calculations are carried out using a supercell approximation to model localised defects using periodic boundary conditions. We show quantitatively that X adatom is most favorable in rich X conditions while in poor X environment X monovacancy is most favorable. Interestingly, an interplay between adatom and divacancies takes place as temperature increases. To gauge the importance of vibrational free energy contributions on the engineering of gap states in the 2D monolayers, we compare the formation energies of point defects with an adsorbed F6TNAP at various thermodynamic conditions.

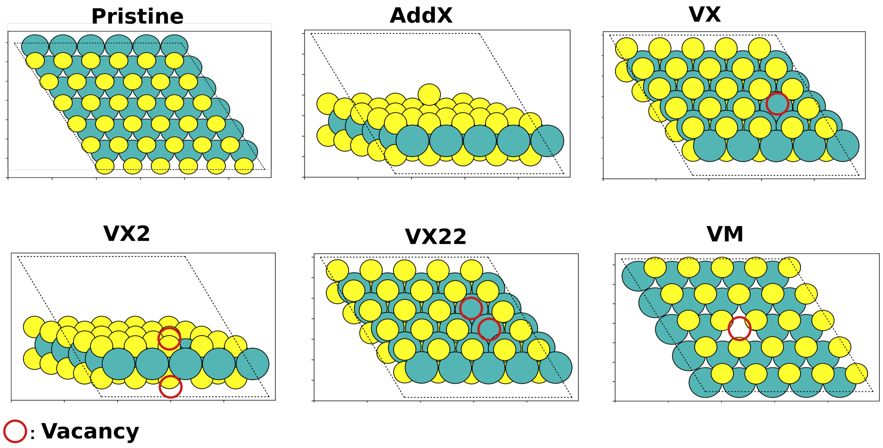


Figure 4.1

[1] V. Blum et al., Comp. Phys. Comm. 180, 2175–219 (2009)

03 Laser-induced ultrafast charge-carrier dynamics in pyrene-doped $MoSe_2$ monolayer

Matheus Jacobs[1], Jannis Krumland, Caterina Cocchi[1]

[1]Institut für Physik and IRIS Adlershof, Humboldt-Universität zu Berlin, Berlin (Germany)
[2]National Centre for Scientific Research "Demokritos", Athens (Greece)

In the last years, the interest in transition metal dichalcogenide monolayers have grown enormously due to their particular electronic structure and light-matter coupling properties. Combining these systems with physisorbed carbon conjugated molecules [1] can give rise to new systems with enhanced opto-electronic performance, specially when excited by resonant time-dependent electric fields. In the framework of real-time time-dependent density functional (RT-TDDFT) theory, we investigate the ultrafast charge-carrier dynamics at the interface formed by pyrene molecules physisorbed on a $MoSe_2$ monolayer [2]. By monitoring the effect of the incident pulse intensity on the excitation energy and number of excited electrons, we identify a striking nonlinear response of the system, which in turn impacts the charge transfer. Moreover, in order to further analyze the role of vibronic interactions, we have been joining forces with project A13, by integrating classical- and quantum nuclear trajectories into RT-TDDFT simulations. First results indicate a rather minor impact of vibronic coupling in the ultrafast charge-carrier dynamics and transfer in this hybrid system in comparison with other hybrid interfaces studied previously [3].

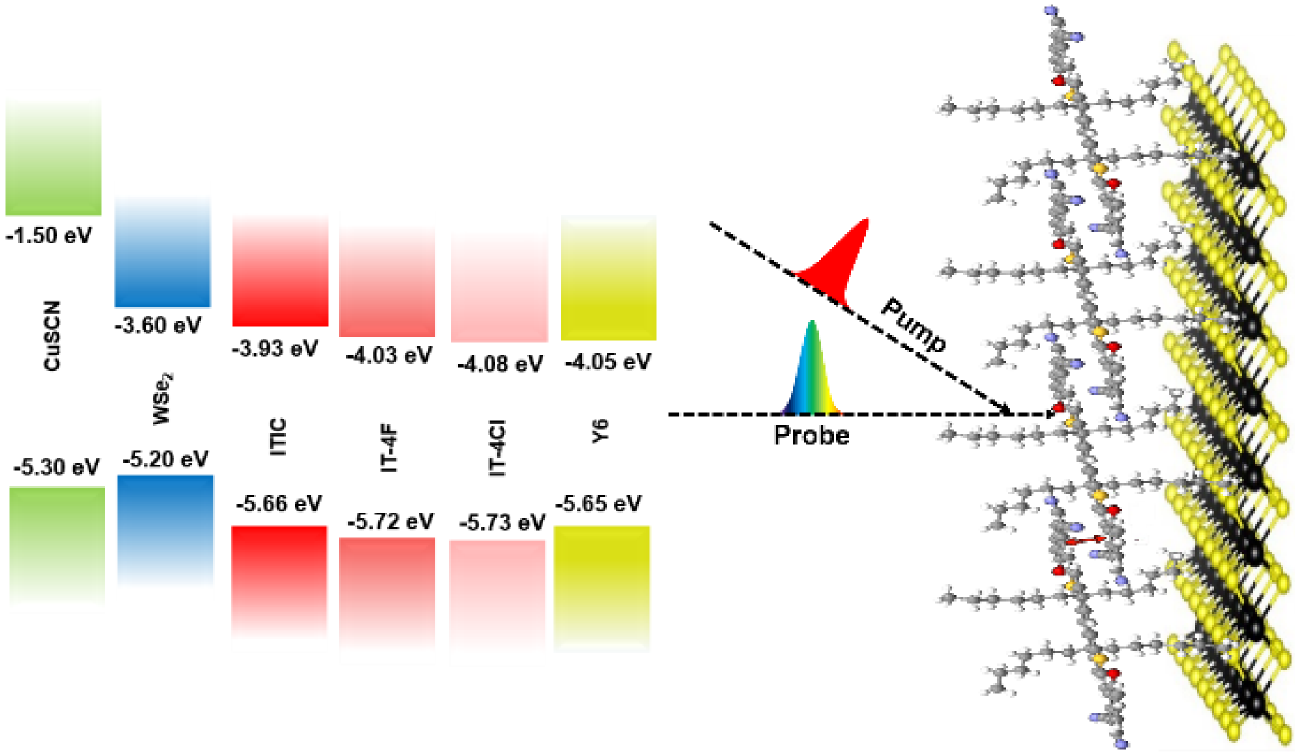
[1] J. Krumland and C. Cocchi, Electronic Structure 3, 044003 (2021).
[2] M. Jacobs, J. Krumland, and C. Cocchi, ACS Appl. Nano Mater. 5, 5187 (2022).
[3] M. Jacobs, J. Krumland, A. M. Valencia, H. Wang, M. Rossi, and C. Cocchi, Adv. Phys. X 5, 1749883 (2020).

04 Probing the charge transfer mechanism at WSe2/non-fullerene acceptor interfaces

Sreelakshmi Chandrabose[1], Meysam Raoufi[1], Dieter Neher[1,2]

[1]Institute of Physics and Astronomy, University of Potsdam, Berlin (Germany)

Two-dimensional transition metal dichalcogenides (TMDCs) can be combined with other materials such as organic small molecules to form hybrid van der Waals heterostructures [1]. Recently, non-fullerenes, a novel class of organic electron acceptors have been widely studied owing to their attractive properties including tunable bandgap, strong absorption coefficients in the visible to near infrared (NIR) regime and excellent exciton diffusion lengths exceeding 30 nm [2]. This motivates to integrate them with 2D TMDCs to novel devices that benefits from advantageous properties of both materials. This study presents a systematic investigation of the charge transfer mechanism at the interfaces of monolayer tungsten diselenide (WSe2) and a series of best performing non-fullerene acceptors (NFAs) (Y6, ITIC, IT-4F, IT-4Cl) using ultrafast transient absorption spectroscopy (TAS). Upon selectively exciting the NFA, evidence of sub-picosecond hole transfer is obtained from the appearance of WSe2 ground state bleach along with the prolongation of electron related signals in the acceptors. The results are in consistent with the strong photoluminescence quenching observed in the bilayers. For ITIC combined with WSe2, the recombination of the generated free carriers is found to populates the low-lying triplet state (T1) of ITIC. Importantly, this unwanted process is effectively suppressed in the bilayer with the fluorinated and chlorinated derivatives, due to a modified triplet level alignment [3]. We observe a similar scenario when using CuSCN as an exciton quencher for the acceptor materials, meaning that the triplet state is not induced by the high spin-orbit coupling of WSe2. Our results highlight the delicate balance between excitons and charges in such inorganic/NFA devices.



[1] 1. Kafle, T.R. et.al, ACS Nano., 11, 10184-10192 (2017).
[2] Firdaus, Y. et al, Nat Communications., 11, 5220 (2020).
[3] Souza, J.P.A. et al, The Journal of Physical Chemistry A., 126 (8), 1393-1402 (2022).

05 Organic Synaptic Diodes Based on Polymeric Mixed Ionic-Electronic Conductors

Fabian Gärisch[1], Giovanni Ligorio[1], Patrick Klein[2], Michael Forster[2], Ullrich Scherf[2] and Emil J. W. List-Kratochvil [1,3]
[1]Institut für Chemie, Institut für Physik & IRIS Adlershof, Humboldt-Universität zu Berlin, Berlin (Germany)
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Due to the limitations of contemporary computer architecture, the implementation of machine learning functionality in the form of hardware is highly desirable. In biological neural systems, learning and signal processing are achieved by communication between neurons, mediated by time-dependent ion flux in the synapses. Integrating an equivalent ion-dependent operating principle in neuromorphic devices can deliver an energy efficient and powerful technology. Here a device known as a light- emitting electrochemical cell (LEC) is revisited and modified, exploiting its ability to modulate current through ion accumulation/depletion at the electrodes, turning it into a biomimetic organic synaptic diode. The LEC, a two-terminal device, is based on an organic mixed ionic-electronic conducting polymer that serves as the active material for conducting lithium ions as well as charge carriers. In the here-presented device the ionic conduction properties are modified by cryptand molecules, able to reversibly capture ions. The device can be reliably switched between states for at least 100 cycles without visible degradation and displays state retention for multiple minutes. The applicability for neuromorphic applications is further demonstrated by exploring frequency-dependent plasticity and paired-pulse facilitation behaviour in the millisecond range. The solution-processable polymer material, combined with the simple two-terminal architecture of the presented neuromorphic device, open up a range of possibilities regarding the fabrication of artificial neural networks.

06 Surface doping of rubrene single crystals by molecular electron donors and acceptors

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Organic semiconductors have shown great promise for sustainable (opto)electronic applications within the past decade. Organic light-emitting diodes are already commercially available, and organic photovoltaics (OPVs) and field-effect transistors (OFETs) are also rapidly advancing, with power conversion efficiency of OPV reaching over 18% and carrier mobility in OFETs higher than 1 cm^2/Vs, respectively. However, achieving further improved device performance requires continued optimization of certain aspects of interfacial electronic energy level alignment, such as improving charge injection by minimizing the energetic barriers and creating charge accumulation layers at the interface. Accordingly, this work seeks to zoom in on the mechanisms of interface engineering by considering the surface doping of rubrene single-crystals by molecular electron donors and acceptors. Rubrene is a benchmark example of a high carrier mobility semiconductor, as its charge transport is mediated primarily through electronic bands and not via hopping. In this regard, to effectively tune the energy levels without compromising the band structure, doping should be non-destructive to the crystalline surface of rubrene. Our angle-resolved photoemission results show that deposition of molecular p-type and n-type dopants, $Mo(tfd\text{-}CO_2Me)_3$ and CoCp2respectively, on rubrene causes a corresponding shift of the valence bands, accompanied by the formation of free charge carriers in rubrene, while the electronic band parameters remain unperturbed.

07 Electronic Interactions of Molecular Adsorbates on hBN and MoS2 monolayers

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The design of low-dimensional organic-inorganic hybrid interfaces for the next generation of optoelectronic applications requires an extensive understanding of the electronic interactions and properties in these systems. In this work [1], we present a first-principles study based on density-functional theory inspecting the structural, energetic, and electronic properties of five molecular donors and acceptors adsorbed on freestanding hexagonal boron nitride (hBN) and molib- denum disulfide (MoS_2) monolayers. All considered heterostructures are stable, due to the crucial contribution of dispersion interactions, which are maximed by the overall flat arrangement of the physisorbed molecules on both substrates. The level alignment of the hybrid systems depends on the characteristics of the constituents. On hBN, both type-I and type-II heterostructures may form, depending on the relative energies of the frontier orbitals with respect to the vacuum level. On the other hand, all MoS_2 based hybrid systems always exhibit a type-II level alignment, with the molecular frontier orbitals positioned across the energy gap of the semiconductor. The electronic structure of the hybrid materials is further determined by exploring the projected density of states. There, it is found a localization of frontier orbitals within the band gap and hybridization, between the organic and inorganic compounds, for those levels which form further in the conduction and valence bands. These results provide important indications for the design of novel low-dimensional hybrid materials with suitable characteristics for optoelectronics.

[1] Melani G., Guerrero-Felipe J. P. et al, Phys. Chem. Chem. Phys., 24, 16671-16679 (2022).

08 Synthesis and Application of Hybrid Plasmonic Nanoparticles

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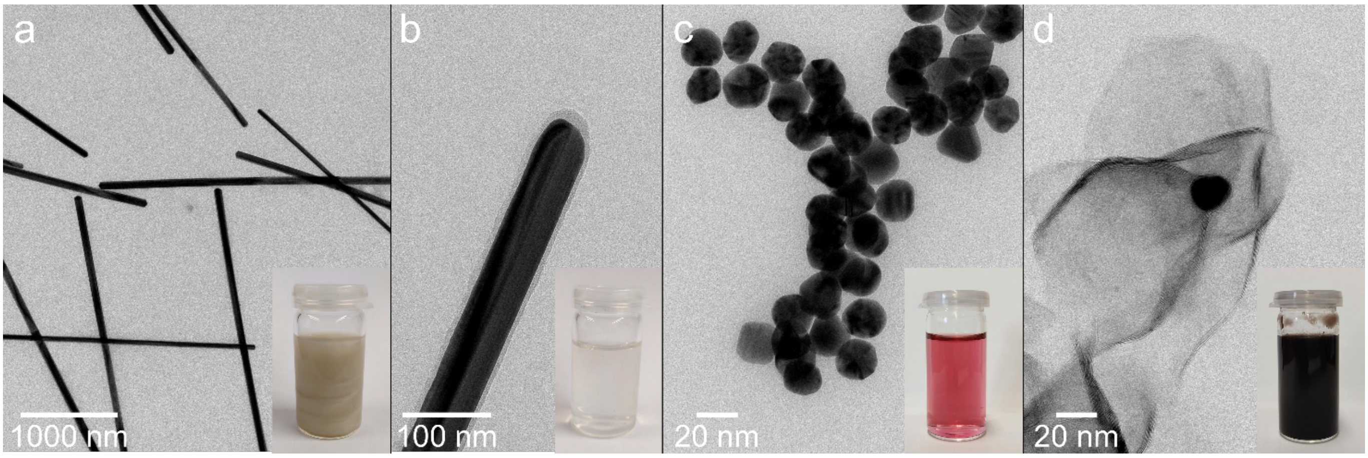
Plasmonic nanostructures can be made by physical top-down approaches or chemical bottom-up approaches. The bottom-up approach typically yields particles with higher quality surfaces, which is often detrimental to the plasmonic properties. Having plasmonic nanomaterials in particle form also allows a variety of surface modifications/coatings, enabling tailored properties and applications. Here I present two hybrid plasmonic nanomaterials I am currently working on: silver nanowires coated with a thin layer of silica and gold nanospheres/nanorods coated with sheets of molybdenum disulphide. In our recent works, we optimized the 8 nm silica coating [1] and embedded organic fluorophores in this shell, so we can use the nanowires as a plasmonic waveguide / plasmon converter for subdiffractional chiral sensing [2]. Furthermore, we analysed the photobleaching process of the dyes during the operation of a single converter [3]. Meanwhile we are currently examining the potential of the coated gold nanomaterials for CO2 reduction, with results on the way.



TEM images of plasmonic nanomaterials and photographs of their dispersion (inset). a) silver nanowires. b) silver nanowires coated with silica. c) gold nanospheres. d) gold nanospheres coated with sheets of molybdenum disulphide.

[1] M. Rothe, Y. Zhao, G. Kewes, et al. Scientific reports 9.1 (2019): 1-12.
[2] M. Rothe, Y. Zhao, J. Müller, et al. ACS nano 15.1 (2020): 351-361.
[3] M. Rothe, Y. Zhao, H. Halim, et al. Optics Continuum 1.8 (2022): 1730-1740.

09 Infrared-active acoustic-optical phonon modes in two dimensional organic/inorganic interfaces

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Physics phenomena at hybrid inorganic/organic interfaces formed by conjugated molecules adsorbed on transition-metal dichalcogenide (TMDC) monolayers have stimulated intense research activity showcasing for instance the non-trivial electronic coupling between the two type of components [1].

In this work, we investigate by means of density functional perturbation theory, the phonon modes in a prototypical two-dimensional hybrid interface formed by a MoSe2 monolayer decorated with pyrene molecules. Our results reveal the presence of an emergent libration IR-active phonon mode caused from the coupling of an optical phonon of the TMDC with an acoustic one in the molecule corresponding to a rotation around its central axis. This results opens unprecedented perspectives in nanoscopic mechanism engineering particularly the generation of molecular nanorotors on 2D substrates.

[1] Jannis Krumland and Caterina Cocchi, Electron. Struct. 3 044003 (2021).

10 Scanning Tunneling Microscopy investigation of intrinsic defects of 1T-$TiSe_2$

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he electronic properties of 2D materials are strongly influenced by single atomic defects or adsorbates. In particular, top layer vacancies can also be used as molecular anchor points. 1T-$TiSe_2$ has been reported to be either a semiconductor with a very small indirect band gap or a semimetal with a very small indirect band overlap [1]. Atomic defects have been found to locally dope the 1T-$TiSe_2$ [2]. The Scanning Tunneling Microscope (STM) is a well-suited tool for investigating the local density of states (LDOS) and therefore for probing these local perturbations by Scanning Tunneling Spectroscopy (STS). In our work we characterize the different defect species by means of STM and STS, where we can distinguish top from bottom layer Selenium vacancies and such where the Selenium was substituted by a different element, based on [2].

[1] J. Rasch, PRL 101, 237602 (2008).
[2] B. Hildebrand, PRL 112, 197001 (2014).

11 Gold-Mediated Exfoliation of millimeter-scale MoS_2 Monolayers and their use in high transconductance Electrolyte Gated Field Effect Transistors

Max Heyl[1], Daniel Steffen Rühl[1], Sarah Grützmacher[1], Fabian Gärisch[1], Sylke Blumstengel[1], Giovanni Ligorio[1], Emil J. W. List-Kratochvil[1,2]

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The efforts to reach the monolayer limit of transition metal dichalcogenides (TMDCs), are motivated by a whole set of interesting properties accompanying two-dimensional TMDCs, e.g., rich exciton physics, and the "only-surface-no-bulk" character. This extreme surface-to-volume ratio makes devices from monolayers exceptionally sensitive to changes in their surroundings, giving potential for applications in sensing devices. However, the isolation of high-quality, large-area TMDC monolayers still poses a key challenge. On this note, scalable gold-mediated exfoliations attracted broad attention to supersede the traditional scotch tape method. Herein, a gold-mediated exfoliation based on a low-temperature annealing is presented. The exfoliation achieves scaling with parent crystal areas, placing it on par with previously reported methods. The process is benchmarked using MoS_2 as prototypical TMDC and millimeter-scale monolayers are successfully exfoliated and transferred. With large-scale high-quality monolayers unlocked, we show that electrolyte gated devices operated in an aqueous environment provide for promising transducing elements for (bio)sensor applications. Looking forward, these results pave the way for additional neuromorphic functions co-implemented in the sensing platform, enabled by the tuneability of the stimuli response by simply modifying the monolayer interface.

12 Influence of molecular films intercalating interfaces between WS_2 and MoS_2 monolayers and mica substrate onto charge transfer across the interface

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We investigate here how intercalation of molecularly thin layers of water, ethanol and tetrahydrofuran (THF) into the interface between tungsten disulfide (WS2) or molybdenum disulfide (MoS2) monolayers and mica substrate affect photoluminescence (PL) and differential reflectivity () of the WS2 monolayers. We exfoliate WS2 monolayers onto mica substrate in dry nitrogen environment, and record photoluminescence from the flakes first under dry nitrogen, and afterwards during exposure to vapors of the liquids. Molecularly thin layers of liquid molecules intercalate the interface between MoS2 or WS2 and mica upon exposures of the samples to the respective liquid vapors, as confirmed by AFM imaging. Due to the intercalation, there is a noticeable shift in the energy of the PL maximum with the amount and direction of the shift depending on the liquid used. The peak position becomes red shifted by intercalation of THF and blue shifted by the intercalation of ethanol and even more blue shifted by the intercalation of water, with a difference in exciton peak position between THF and water of about 80 meV. Intercalation of water and ethanol is reversible as well as their influence on the peak positions, for this reason we assume intercalation of molecular layers does not affect WS2 monolayer strain. The conclusion is supported by almost no shift of the absorption peak of the neutral exciton of WS2 monolayers revealed by the reflectance spectra. Therefore, we attribute the shift in PL peak position to charge doping which affects the ratio between charged (trion) and neutral exciton emission. The superposition of these two components in different relative amounts results in an apparent shift of the peak maximum. The charging of the TMDC is due to the mica surface and is screened by the liquid layers, most effective by the most polar solvent (Water) and least effective by the least polar solvent (THF). The findings presented here are allow to get deeper insight into the mechanisms of how molecularly thin films intercalating interfaces regulate charge transfer across the interfaces.

13 Large perpendicular field in bilayer TMD via hybrid molecular gating

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We consider structures in which bilayer TMDs are sandwiched between a layer of molecules and Si gate. We show that these structures allow increasing, by a factor of 2, maximum electric field achievable in this 2D material. This in turn, allows reaching electric field greater than 0.25 V/nm. In MoS_2 we show avoided crossing behavior between interlayer and B excitons, and demonstrate previously unresolvable splitting of A exciton that arises from coupling to the second, higher energy interlayer. This allows us to restore positions of two intra- and two inter- layer excitons , and calculate the coupling strength, further contributing to our understanding of MoS_2, and potentially other TMDs.

14 Resolution of Intramolecular Dipoles and a Push-Back Effect of Individual Molecules on a Metal Surface

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Molecules consisting of a donor and an acceptor moiety can exhibit large intrinsic dipole moments. Upon deposition on a metal surface, the dipole may be effectively screened and the charge distribution altered due to hybridization with substrate electronic states. We study Ethyl-Diaminodicyanoquinone molecules on a Au(111) surface with scanning tunneling microscopy (STM) and non-contact atomic force microscopy (nc-AFM). By mapping the local contact potential difference (LCPD) over the flat lying molecules we show that the intrinsic dipole moment of the molecules persists upon adsorption on the Au(111) surface. Density functional theory calculations reveal that the dipole moment is even increased on the metal substrate as compared to the gas phase. Overall, the LCPD value is decreased by several tens of meV with respect to the bare metal as a consequence of the so-called push-back or cushion effect [1].

[1] S. Trishin, et al., J. Phys. Chem. C, 126, 17, 7667 (2022).

15 Spectroscopic and charge transport properties of X/γ-rays detectors based on $MAPbBr_3$ single crystals

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Organic-inorganic perovskites have shown to be promising materials with great potential for use in X- and γ-radiation detectors due to their relatively high atomic number and thus high stopping power, superior carrier life-time product, tunable band gap, and low temperature fabrication process. In this study, ITO/PTAA/$MAPbBr_3$/C_{60}/BCP/Cu detectors based on $MAPbBr_3$ single crystals are investigated. The spectra of isotopes ^{241}Am (59 keV) and ^{137}Cs (662 keV) were experimentally measured at room temperature and different values and polarities of bias voltages. The charge transport mechanisms within the entire voltage range are described in terms of the well-known theoretical models. The comparison of calculations results with the experimental data allowed us to determine the main parameters of the crystals and barrier structures. The optimization of technological conditions of the formation of contacts to the $MAPbBr_3$ crystals is necessary to decrease the leakage current through the barrier structures, and thus improve the charge collection and energy resolution of the detectors.

16 Level alignment of hybrid inorganic/organic interfaces on the cheap

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The combination of organic molecules with two-dimensional inorganic substrates offers promising routes for producing novel materials with enhanced opto-electronic characteristics. In particular, transition metal dichalcogenide (TMDC) monolayers and carbon-conjugated molecules represent suitable platforms especially in light of their stability and electronic properties [1,2,3]. Yet, the fully atomistic description of such systems becomes increasingly demanding as the size of the molecules increase. To this end, the recently implemented implicit method LayerPCM [4] offers an advantageous trade-off between accuracy and computational costs [5]. In this work, we adopt this method to compute the bandgap and the absorption optical spectra oligoacene molecules of increasing lengths considering various dielectric substrates. Using the same method, level alignments and bandstructures of hybrid systems of phthalocyanines on TMDC monolayers are additionally investigated tuning the exact-exchange parameter of the PBE0 hybrid functional. Comparison with GW calculations validates the adopted approach.

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[2] J. Krumland and C. Cocchi, Electron. Struct. 3, 044003 (2021).
[3] M. Jacobs, J. Krumland, and C. Cocchi, ACS Appl. Nano Mater. 5, 5187 (2022).
[4] J. Krumland, G. Gil, S. Corni, and C. Cocchi, J. Chem. Phys. 154, 224114 (2021).
[5] C. Cocchi, M. Guerrini, J. Krumland, N.T Nguyen, A. M. Valencia, submitted to J. Phys. Mater. (2022); arXiv:2208.09168.

17 Computational studies of structure formation and dynamic properties of organic molecules at hybrid inorganic/organic interfaces

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Hybrid structures of organic molecules and inorganic semiconductors combine the favorable properties of each material class into conjugates with great application potential. The organic parts offer a vast diversity in terms of chemistry and structure. Combined with inorganic substrates, onto which the organic molecules can be deposited as thin films, Hybrid Inorganic/Organic Structures (HIOS) with well-tailored properties can be created that cannot be realized with either material class alone. This Poster will give an overview of the work in our group to theoretically describe the deposition and thin film growth of organic molecules on inorganic substrates using a multi-scale strategy combining quantum density functional theory, all-atom molecular dynamics and Langevin dynamics simulations, and classical diffusion theory.

18 Fast photo response from hybrid monolayer MoS_2/organic interface

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As a direct-band-gap transition semiconductor with high carrier mobility, monolayer metal dichalcogenides (TMD) are promising materials semiconductor for photodetection. However, the persistent photoconductance (PPC) that originates from hole capture by deep traps causes a slow response speed[1,2]. Herein, we apply frequency modulated photocurrent spectroscopy to investigate the photoresponse in hybrid samples based on monolayer MoS_2 covered with an ultrathin layer of phthalocyanine (H_2Pc) molecules. Compared to neat MoS2 the MoS_2/H_2Pc bilayer exhibits an improved photoresponse speed and responsivity, which we assign to efficient interfacial charge transfer, suppressing the PPC in MoS_2 as alternative route for hole transfer. The frequency dependent photocurrent data can be consistently explained by the Havriliak–Negami (HN) equation, from which the carrier lifetime and the steady state photoresponse is deduced [3]. By performing the experiments under steady state background, we conclude about the dominant charge recombination process. The strategy presented here based on interfacing monolayer of MoS_2 with organic molecules serves a nice platform for optimization of TMD-based photodetection toward ultrafast speed and high sensitivity.

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[2] Furchi, Marco M., et al. Nano letters, 2014, 14, 6165.
[3] Epshtein, O., et al. Physical Review B, 2001, 63, 2.

19 Reversible training of waveguide-based AND/OR gates for optically driven artificial neural networks using photochromic molecules

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Artificial neural networks (ANNs) are inspired by the biological nervous system. The high performance of such ANNs is achieved through the dynamic change of the synaptic weights by applying self-optimizing learning algorithms. Despite the simple operations for each single element in an ANN, a network with a huge number of simulated elements consumes lots of computing capacity using von Neumann computer architectures. To overcome this issue, neuromorphic devices facilitate the design of hardware ANNs that emulate the synaptic functions. Here we demonstrate the viability of such an approach using photonic waveguides in combination with a photochromic diarylethene (DAE) molecule. By positioning and irradiating DAE molecules on single waveguides, we modulate the intensity and thereby emulate the plasticity of the synaptic weights. To run the photonic device as an ANN we firstly characterize the modulation range and encode a learning procedure accordingly. As the proof of concept, we operate a y-shaped waveguide performing basic AND/OR logic gate functions, with the capability to switch between these two gate functions by using specific training sets.

20 Work Function and Energy Level Alignment Modification at Ti_3C_2 MXene Surfaces and Interfaces Using Strong Organic Donor/Acceptor Molecules

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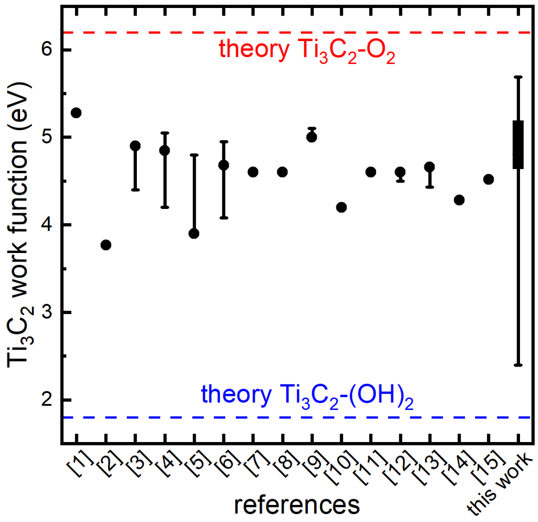
The family of two dimensional MXenes, with Ti_3C_2 being the most prominent member, shows a large variety of properties and therefore potential for a manifold of applications. One of them is the application as an electrode in organic light emitting diodes or field effect transistors. In these cases, the work function of the Ti_3C_2 MXenes plays an important role for the energy level alignment to the subsequently deposited organic layer, as it directly determines the electron and hole injection barriers. Means to change the Ti_3C_2 work function are therefore desirable. Here it is demonstrated that by using thin layers of strong organic donor/acceptor molecules, the work function of Ti_3C_2 can be tuned over a wide range of more than 3 eV. Thereby, the energy level alignment to a subsequently deposited organic semiconductor can be tuned all the way from HOMO-level pinning (minimal hole injection barrier) to LUMO-level pinning (minimal electron injection barrier), as desired for the respective application. Furthermore, it is shown that a fully oxygen terminated surface (Ti_3C_2-O_2) does not lead to an extraordinary high work function, in contrast to theoretical predictions.



21 Direct observation of ultrafast lattice distortions during exciton-polaron formation in lead-halide perovskite nanocrystals

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The microscopic origin of slow carrier cooling in lead-halide perovskites remains debated, and has direct implications for applications. Slow carrier cooling has been attributed to either polaron formation or a hot-phonon bottleneck effect at high excited carrier densities ($> 10^{18}$ cm^{-3}). These effects cannot be unambiguously disentangled from optical experiments alone. However, they can be distinguished by direct observations of ultrafast lattice dynamics, as these effects are expected to create qualitatively distinct fingerprints. To this end, we employ femtosecond electron diffraction and directly measure the sub-picosecond lattice dynamics of weakly confined CsPbBr3 nanocrystals following above-gap photo-excitation. The data reveal a light-induced structural distortion appearing on a time scale varying between 380 fs to 1200 fs depending on the excitation fluence. We attribute these dynamics to the effect of exciton-polarons on the lattice, and the slower dynamics at high fluences to slower hot carrier cooling, which slows down the establishment of the exciton-polaron population. Further analysis and simulations show that the distortion is consistent with motions of the [PbBr$_3$]-octahedral ionic cage, and closest agreement with the data is obtained for Pb-Br bond lengthening. Our work demonstrates how direct studies of lattice dynamics on the sub-picosecond timescale can discriminate between competing scenarios, thereby shedding light on the origin of slow carrier cooling in lead-halide perovskites.

22 Structural and electronic properties of transition metal dichalcogenide nanobubbles and nanoripples

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The application of strain to transition metal dichalcogenide (TMDC) monolayers is a promising way to tune the electronic properties of these materials leading to their application, among others, as single-photon emitters. The resulting nanostructures, having the form of nanoripples or nanobubbles, exhibit significant variations in their electronic properties in comparison with the pristine monolayers, as our ab initio calculations based on density-functional theory (DFT) clearly demonstrate. Using band unfolding techniques [1], we rationalize the effects of curvature and strain on the band structure of the materials. This analysis opens the door to computationally cheaper DFT investigations on curved TMDC monolayers, based on more accurate starting points in the relaxation process.

[1] J. Krumland and C. Cocchi, Electron. Struct. 3, 044003 (2021).

23 Coherent & Incoherent Energy transfer in ML-WS_2-metal and ML-WS_2-organic heterostructures

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Van der Waals heterostructures consisting of 2D transition metal dichalcogenides (TMDCs), organic molecules, and metallic films/nanostructures hold the promise to benefit from synergy effects that enhance optoelectronic properties of such systems, for example, to manipulate and enhance light emission. Studies on energy transfer (ET) in such systems have until now mostly focused using a TMDC as an acceptor. In this contribution, we demonstrate incoherent ET from a monolayer (ML) of WS_2 to an organic dye molecule (ATTO 725) as well as a coherent ET between a ML WS_2 and a surface plasmon polariton (SPP) of a thin silver film. The energy transfer from WS_2 to ATTO 725 was confirmed by time correlated single photon counting (TCSPC), photoluminescence (PL) microspectroscopy, and PL excitation (PLE) spectroscopy. TCSPC shows a reduced PL lifetime of the A exciton of WS_2 after deposition of the organic dye suggesting an ET efficiency of about 70%. The PLE spectrum of the WS_2/ATTO 725 hybrid sample recorded at the emission energy of the dye molecule reproduces clearly the absorption spectrum of the WS2 monolayer proving that the exciton energy is transferred to the dye as an acceptor. Since any type of energy transfer happens on length scales less than 10 nm, these results indicate that organic dye molecules may well be used as local probes for mapping optical response of TMDCs beyond the optical resolution limit. It also could pave the way for the fabrication of single photon sources. Coupling of excitons of ML-WS_2 with propagating SPPs of a thin Ag film is studied by spectroscopic ellipsometry in total internal reflection geometry. Anticrossing and Rabi splitting provide evidence for strong coupling between the A exciton of ML-WS_2 and the SPP while B and C excitons couple only weakly. In such planar configuration, electric field tuning of the optical response of TMDCs could be employed to actively control of the exciton-plasmon coupling strength for the construction of novel plasmonic devices.

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