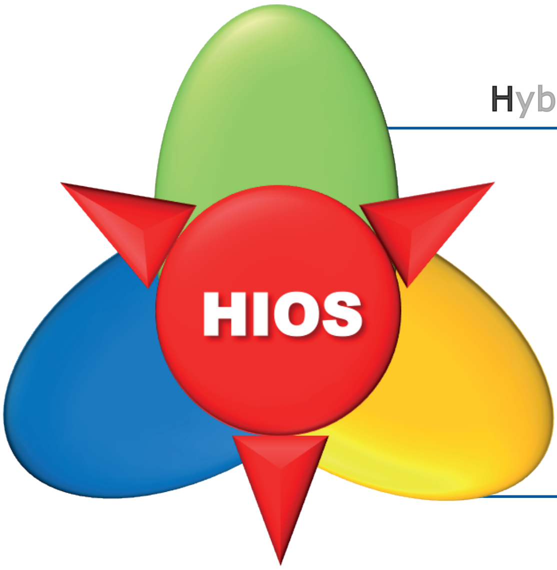
Gefördert durch
DFG Deutsche
Forschungsgemeinschaft

HUMBOLDT-UNIVERSITÄT ZU BERLIN

Collaborative Research Centre 951



Hybrid Inorganic/Organic Systems for Opto-Electronics

Young Researcher Workshop 2021

Online Conference 11.10. — 12.10.2021

Book of Abstracts

TU berlin

Universität Potsdam

Freie Universität Berlin

MAX-PLANCK-GESELLSCHAFT
FRITZ-HABER-INSTITUT

HZB Helmholtz
Zentrum Berlin

HIOS Young Researcher Workshop 2021
Workshop Program

Monday (11.10.2021)

Opening Remarks
Short Overview of the CRC 951
10:00 – 10:15
Prof. Dr. Norbert Koch
(Spokesperson of the CRC 951, A08, Z01, Z03)

Session 01
10:15 – 11:15
Chair: Alberto Eljarrat

Boubacar Tanda Bonkano (B09)

Qiang Wang (A08)

Dominik Christiansen (B12)

Meysam Raoufi (B07)

11:15 – 11:30
Coffee Break

Session 02
11:30 – 12:45
Chair: Ana M. Valencia

Ignacio Gonzalez Oliva (B11)

Hanen Hamdi (B16)

Matheus Jacobs (B16)

Nicolas Zorn Morales (B03)

Daniel Steffen Rühl (B03)

Lunch Break
12:45 – 13:45

Session 03
13:45 – 15:15
Chair: Matheus Jacobs

Alaa Akkoush (A13)

Jennifer Hartfiel (A14)

Björn Kobin (A03)

J. Rika Simon (A14)

Juan Pablo Guerrero (B16)

Jannis Krumland (B16)

Tuesday (12.10.2021)

Session 04
10:00 – 11:30
Chair: Martin Rothe

Alberto Eljarrat (A12, Z02)

Dmitrii Maksimov (A13)

Manuel Katzer (B12)

Hélène Seiler (B17)

Alexander Neef (B17)

Johannes Müller (A12)

11:30 – 12:30
Lunch Break

Session 05
12:30 – 14:15
Chair: Hélène Seiler

Niko Nikolay (B18)

Martin Rothe (B02)

Ana M. Valencia (B16)

Hu Lin (A06)

Max Heyl (B14)

Elsie Bowen-Dodoo (B09)

Fabian Gärisch (B14)

Contents

Important information at a glance

ZOOM link

The workshop will be held via ZOOM. Please click on the link below for direct access to the corresponding ZOOM room or use the provided log-in data. The login data will not change during the workshop.

ZOOM link	Meeting ID	Password
Link	662 0029 0337	085481

Please make sure to download the latest version of ZOOM. More technical information on ZOOM can be found below.

General Information

After entering the ZOOM room, your microphone and camera will be deactivated. We kindly ask you to keep both deactivated at all times, unless the moderators ask you to activate them. The speakers will share their screens for their presentations. For the best possible view, we recommend to hide participants without video and to activate the side-by-side mode.

Discussions

Your questions and comments are highly welcome after the talk. For this, you have the following two options:

- **Use the "raise hand" feature:** you can ask your question in person after the talk by using the "raise hand" feature. The chair will ask you to activate your microphone and camera. After your question was answered, please deactivate your microphone and camera again and lower your hand.
- **Ask your question in the chat:** you can write down your question at any time in the chat. The session chair will read your question aloud after the talk. Please refrain from writing to the speaker directly. If possible, start writing your question during the talk.

Communication with other participants

During the breaks, we can host a private break out room for you to have discussions with other participants. If you wish to have a private break out room with other participants, please send Christian Pugatschow or Jule Budnick a private message via the chat function.

Zoom basics

Audio/Video: By clicking on the buttons "Join Audio" and "Start Video" in the lower left corner of the ZOOM window you can activate and deactive your video/audio, respectively. Please keep both deactivated during the oral presentations unless you recieve a notification to do otherwise.

Screen Sharing: To share your screen, click on the green button "Share Screen" in the lower middle of the ZOOM window.

The "raise hand" feature: To raise your hand, click on the "Reactions" button in the lower right side of the ZOOM window. A popup window with several reactions will open. In the bottom of this window, you can find the "raise hand" button.

The Chat function: To chat with other participants, click on the "Chat" button next to the "Share Screen" button in the lower middle of the ZOOM window. The chat window will open. In the lower area of the chat window is the input field. Right above the input field, you can choose if you want to address your message to all participants or only to certain participants.

Technical advice for using ZOOM

Please make sure to use the latest ZOOM version. If possible, use a LAN connection instead of WiFi, especially if you plan to activate your camera or audio. This notably improves your video and audio quality. If possible, try to minimize background noise when activating your audio and avoid rooms with a large echo. Your audio quality can be further enhanced by using a headset.

Session 1

Speaker: Boubacar Tanda Bonkano
Project: B9
Office Number: 030 8413 5126
E-Mail: tandabonkano@fhi-berlin.mpg.de

Excited state dynamics of terrylene

Boubacar Tanda Bonkano, Samuel Palato, Sergey Kovalenko, Lukas Gierster, Björn Kobin,
Stefan Hecht, Julia Stähler

Abstract

In hybrid solar cells, the separation of the electron-hole pairs after photogeneration is a key aspect of the light harversting functionality. This requires, in hybrid inorganic/organic systems, the use of molecules that have excited states with sufficiently long lifetime to allow charge separation. In order to investigate terrylene molecules, we performed two types of time-resolved spectroscopy, time-correlated single photon counting (TCSPC) and broadband transient absorption (TA) using a white light continuum probe. The steady state absorption and emission spectra of terrylene monomers in solution show mirrored lineshapes. Both the TCSPC and TA consistently show a decay of 3.7 ns for the excited state S1. Thin terrylene film shows absorption bands strongly ($>$ 0.5 eV) blue-shifted due to H-aggregation. Photoexcitation of the H-aggregate leads to the formation of an induced monomer-like absorption band at 2.3 eV. The analysis of this spectral feature, enlightened by several works reported in the literature, indicates that it originates from an interference between Frenkel exciton state and charge-tansfer (CT) state. A phenomenological fit model helps to extract and attribute a time constant of 150 ps to the aggregate ground state bleach recovery while the CT state relaxes into the dark state of the H-band within 230 ps. These insights will contribute to a better understanding and control of terrylene-based systems, beneficial for applications.

Speaker: Qiang Wang
Project: A8
Office Number: 030 2093 7538
E-Mail: qwanghu@physik.hu-berlin.de

Thickness-dependent energy level alignment of terrylene molecules on WS_2 substrate

Qiang Wang

Abstract

Monolayers of TMDCs are emerging two-dimensional (2D) semiconductors with outstanding properties, such as superior stability, flexibility and strong light-matter coupling. Using TMDCs combined with conjugated organic materials featuring strong light-matter coupling, novel electronic and opto-electronic properties may result from such hybrid inorganic/organic systems (HIOS). We synthesized 2D continuous monolayer WS_2 on sapphire through chemical vapor deposition (CVD) with precursors of tungsten oxide and sulphur, promoted with sodium tungstate. The band structure was first determined by angle resolved photo emission spectroscopy (ARPES). Then, the electronic and optical properties of the inorganic/organic hybrid structure WS_2/terrylene are studied. It is determined by photo-emission spectroscopy that the ionization potentials of the hydrocarbon molecule, terrylene, shows a reduction of 0.5 eV as the molecular orientation switching from lying-down (monolayer) to standing-up (multi-layer) with respect to the 2D WS_2 substrate. The interface shows a type-II energy level alignment. Photoluminescence and absorption spectra reveal an additional excitonic band of interlayer energy transfer at longer wavelength range of about 1.54 eV.

Speaker: Dominik Christiansen
Project: B12
Office Number: 030 314 27658
E-Mail: dominik.christiansen@tu-berlin.de

Excitonic insulator in molecular functionalized atomically-thin semiconductors

Dominik Christiansen, Malte Selig, Mariana Rossi, Andreas Knorr

Abstract

It is theoretically expected that if the exciton binding energy of semiconductors at thermodynamic equilibrium exceeds the single-particle bad gap a new phase arises: the excitonic insulator [1]. This phase is characterized by a spontaneous formation of excitons leading to a correlated ground state of 1s excitons without optical. Although theoretically predicted, compelling experimental evidence is still missing [2,3]. Here, we propose that hybrids of monolayer transition-metal dichalcogenides and organic molecules are ideal candidates for the realization of the excitonic insulator. Focusing on the exemplary candidate WS_2-F6TCNNQ, we calculate the electronic phase diagram as function of the experimentally relevant parameters band gap (tunable by applied electric voltage), temperature, and dielectric environment. Additionally, we discuss the optical characterization of the electronic phases (semiconductor, semi-metal and excitonic insulator) with far-infrared to terahertz spectroscopy.

References

[1] D. Jerome, T. M. Rice, W. Kohn, Phys. Rev. 158, 462 (1967)
[2] Z. Jiang, Y. Li, S. Zhang, W. Duan, Phys. Rev. B 98, 081408(R) (2018)
[3] arXiv preprint arXiv:2104.07032 (2021)

Speaker: Meysam Raoufi
Project: B7
Office Number: 0331 977 5633
E-Mail: Raoufi@uni-potsdam.de

Influence of energy landscape on charge transfer state formation at MoS_2/organic hybrid interfaces

Meysam Raoufi, Sreelakshmi Chandrabose, Nicolas Zorn Morales, Sylke Blumstengel, Dieter Neher

Abstract

Two-dimensional transition metal dichalcogenides (TMDCs) exhibit excellent optical properties. This has encouraged the solar cell community to combine TMDCs with organic semiconductors to form heterostructures with tailorable properties that possess advantages of both materials [1, 2, 3]. A crucial initial step for designing TMDC-based heterojunctions is tuning energy level alignment to achieve sufficient charge transfer.

Here, we systematically investigate the energy level alignment at the hybrid layer stacks towards the optimization of TMDC exciton dissociation in to charge transfer state. We fabricate bilayer samples of monolayer of MoS_2 and transparent hole transfer materials with different HOMO energies in order to tune energy offset at the heterojunction and, hence, the charge transfer (CT) energy.

The Steady state photoluminescence microscopy studies, confirm exciton quenching for the bilayers with more efficient exciton dissociation and thereby charge transfer for ones with lower CT energy. In parallel, Charge transfer mechanism in bilayers from ultrafast transient absorption spectroscopy resolve long-lived signal which are assigned to charge transfer state. In consistent with photoluminescence study, efficient exciton dissociation happen for energetically aligned systems. These results serves a nice platform for optimization of the bilayer towards realization of efficient solar cells.

References

[1] Z, Chengmei, et al., J. Phys. Chem. Lett. 10 (2018).
[2] K, Tika R., et al., ACS 28 (2019).
[3] T. R. Kae, et al., ACS Nano 17, 164 (2017).
[4] T. A. Shastry, et al., ACS Nano 10, 10573 (2016).

Session 2

Speaker: Ignacio Gonzalez Oliva
Project: B11
Office Number: 030 2093 66368
E-Mail: gonzalez@physik.hu-berlin.de

Hybrid excitations at the interface between a MoS_2 monolayer and organic molecules

Ignacio Gonzalez Oliva, Fabio Caruso, Pasquale Pavone, Claudia Draxl

Abstract

First-principles calculations based on density functional theory (DFT) and many-body perturbation theory are used to investigate the electronic and optical properties of the interface between a MoS_2 monolayer and organic molecules, i.e, pyrene and pyridine. To understand how the constituents interact, attention is dedicated to changes in the level alignment [1]. G_0W_0 calculations show that the electronic levels of both components are renormalized when dynamical screening effects are included. Consequently, the level alignment of our hybrid systems changes from type-II to type-I, with the HOMO of both molecules located below the valence band maximum of MoS_2. The character and spatial distribution of optical excitations are obtained by the solution of the Bethe-Salpeter equation (BSE). Three different types of excitations are identified: MoS_2-like excitons, charge-transfer excitons, and hybrid excitons. The emergence of charge-transfer excitons is observed at the pyrene@MoS_2 interface, whereas hybrid excitations are predominant at the pyridine@MoS_2 interface. All calculations are performed using exciting, an all-electron full-potential code, implementing the family of linearized augmented plane wave plus local orbitals methods [2].

References

[1] C. Draxl, D. Nabok, and K. Hannewald, Acc. Chem. Res. 47, 3225 (2014).
[2] A. Gulans et al., J. Phys. Condens. Matter 26, 363202 (2014).

Speaker: Hanen Hamdi
Project: B16
Office Number: 030 2093 66411
E-Mail: hanen.hamdi@uni-oldenburg.de

First-principles calculations of phonon properties of pyrene@MoS_2 and pyrene@$MoSe_2$

Hanen Hamdi, Caterina Cocchi

Abstract

The growing interest in hybrid inorganic/organic interfaces formed by conjugated molecules adsorbed on transition-metal dichalcogenide monolayers has stimulated a number recent studies on the electronic properties of these compounds (see, e.g. Ref [1]). However, little is known about the phonon characteristics of these hybrid materials, which are crucial for a full understanding of their photophysical properties. In this work, we use first-principles calculations based on density-functional (perturbation) theory, to study the hybrid system formed by the pyrene molecule adsorbed on either MoS_2 or $MoSe_2$ monolayers. We find that, in these hybrid materials, new phonon modes are activated, compared to those of the individual constituents. Furthermore, we analyze the role of the interactions between the molecule and the substrate in determining the infrared activity of the combined system.

References

[1] J. Krumland and C. Cocchi, submitted (2021); arXiv 2109.09433

Speaker: Matheus Jacobs
Project: B16
Office Number: 030 2093 66411
E-Mail: jacobs@physik.hu-berlin.de

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

Matheus Jacobs, Jannis Krumland, Caterina Cocchi

Abstract

In the last years, the interest in transition metal dichalcogenide monolayers have grown enormously due to their unique electronic structure and light-matter coupling properties. Combining these materials with carbon conjugated molecules can give rise to new materials with enhanced opto-electronic performance, specially when excited by coherent radiation. In the framework of real-time time-dependent density functional theory, we investigate the ultrafast chargecarrier dynamics at the interface formed by pyrene molecules physisorbed on a $MoSe_2$ monolayer. By monitoring the effect of the incident pulse intensity on the energy and the electron transfer on the hybrid heterostructure, we identify a striking nonlinear response of the system, which in turn impacts the charge-carrier dynamics and the nature of cahrge transfer from the inorganic to the organic components.

Speaker: Nicolas Zorn Morales
Project: B3
Office Number: 030 2093 82532
E-Mail: nicolas.zorn.morales@hu-berlin.de

Excited state transitions between J-aggregates and transition metal dichalcogenide monolayers

Nicolas Zorn Morales

Abstract

Combination of semiconducting monolayer (ML) transition metal dichalcogenides (TMDC) with conjugated organic molecules provides a route to improve and expand the functionality of TMDC MLs in opto-electronic and photonic devices. If properly designed, molecular layers with strong light matter coupling can enhance, control, and spectrally tune the absorption and emission of light of such heterostructures. Previously, resonant energy transfer from PTCDA to MoS_2 was employed to enhance the photoluminescence yield of ML-MoS_2. The reverse transfer, namely the transfer of electron hole pairs from a TMDC-ML, acting as donor, to an organic acceptor layer remains an open issue. With sharp, well defined optical features in combination with a high photoluminescence quantum yield, J-aggregates are promising candidates to study these excited state interactions. Here we present our recent findings on this topic.

Speaker: Daniel Steffen Rühl
Project: B3
Office Number: 030 2093 82532

Recent progress in exfoliated monolayer device applications

Daniel Steffen Rühl

Abstract

Transition metal dichalcogenide (TMDC)-based monolayer devices have been gaining attention in the last decade due to their promise for the world of semiconductors by providing easily accessible atomically thin and flat platforms. In my talk I will discuss different ML-TMDC devices. Firstly, MoS_2 and WS_2 based devices operating in an aqueos environment will be presented. These devices can be applied in sensing applications due to their high transconductance and sensitivity to environmental changes, which might be further increased with the help of organic molecules. Secondly, I will show the applications of MoS_2 in a photodetector. Here we were able to increase the photon conversion efficiency by the addition of a thin phthalocyanine layer to the inorganic TMDC by about two orders of magnitude. The TMDC in these experiments where thinned down to their monolayer form applying gold mediated exfoliation. The challenges for device performance resulting from such an approach will be discussed.

Session 3

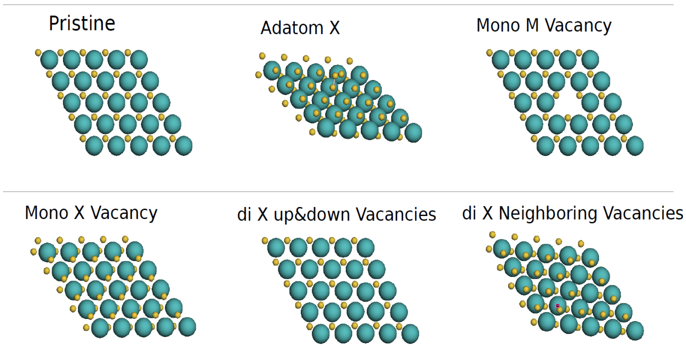
Speaker: Alaa Akkoush
Project: A13
Office Number: 040 8998 88328
E-Mail: akkoush@fhi-berlin.mpg.de

The stability of point defects in 2D monolayer transition metal dichalcogenides and their impact on the electronic structure

Alaa Akkoush, Mariana Rossi

Abstract

Defects are always created during the growth and exfoliation of materials especially monolayers, influencing the electronic, optical and mechanical properties of materials, such as the electronic band structure and exciton dynamics [1]. Hence, our goal is to build a framework in order to achieve better identification and characterization of defect states in monolayer TMDC-based devices, such as photo-detectors and photo-transistors [2]. 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 (TMDCS) such as MoS_2, $MoSe_2$, WS_2 and WSe_2 through ab-initio simulations within the framework of density functional theory (DFT) using FHI-aims [3]. These calculations are carried out using supercell approximation to model localised defects in a periodic host using periodic boundary conditions. Five types of intrinsic point defects were investigated (see figure). We show that chalcogen vacancies are the most favorable point defects in comparison to metal mono vacancy, and the X adatom has the lowest formation energy compared to mono and di X vacancies. Moreover, there is no significant difference in the formation energy of di vacancies of different configurations (up &down and neighbouring) for Mo based systems; however, for W based TMDCs the up&down configuration is more favourable. We then discuss how different thermodynamic conditions change this picture.



References

[1] C. Freysoldt, et al., Rev. Mod. Phys. 86, 253–305 (2014).
[2] W. Choi, et al., Materials Today. 20(3), 116-130 (2017).
[3] V. Blum, et al., Comp. Phys. Comm. 180, 2175–219 (2009).

Speaker: Jennifer Hartfiel
Project: A14
Office Number: 030 838 52813

STM and AFM investigation of MoS_2 on Sapphire

Jennifer Hartfiel, J. Rika Simon, Christian Lotze, Areej Aljarb, Vincent Tung, Patrick Amsalem, Norbert Koch, Katharina J. Franke

Abstract

The STM is a well-suited tool for investigating the local density of states (LDOS). It has proven useful for charactering 2D-materials such as MoS_2 [1] and defects [2,3] as well as atomic [5] and molecular adsorbates [6,7]. Due to the requirement of a conducting substrate, most work has been done for 2D-materials on a metal substrate. However, this has the effect that the metal states hybridize with the MoS_2 [8,9]. To avoid this effect, we aim to investigate MoS_2 on an insulating substrate. The challenge here is the contacting of the MoS_2 monolayer [10] to obtain a conducting circuit for STM measurements. We have tried different approaches for contacting the MoS_2 monolayer on sapphire which turned out inefficient. We ascribe the challenges of establishing good contacts to large Schottky barriers [11]. Using atomic force microscopy (AFM), we find a large monolayer of MoS_2 with very few defects and without grain boundaries. The MoS_2 layer can be charged by approaching the surface with an applied bias voltage which corroborates the absence of contacts to a metallic lead.

References

[1] N. Krane, Surface Science 678, 136 (2018)
[2] B. Schuler, PRL 123, 076801 (2019)
[3] E. Mitterreiter, Nat. Commun. 12, 3822 (2021).
[4] T. Le Quang, 2D Mater. 5, 035034 (2018)
[5] S. Trishin, arXiv:2105.01176 (2021)
[6] N. Krane, ACS Nano 12, (2018)
[7] Z. Song, ACS Nano 11, (2017)
[8] N. Krane, Nano Lett. 16, (2016)
[9] A. Bruix, PRB 93, (2016)
[10] A. Allain, Nat. Mater. 14, (2015)
[11] S. Park, ACS Nano 15, 14794, (2021)

Speaker: Björn Kobin
Project: A3
Office Number: 030 2093 7451
E-Mail: kobinbjo@hu-berlin.de

Attempts towards TMDC surface functionalization, photoswitches with charges, and custom synthesis of organic molecules

B. Kobin, J. Schwarz, T. Schultz, R. Wang, N. Koch, S. Hecht

Abstract

Our recent activities in the frame of HIOS cover three research areas. In the first one, TMDC surface functionalization via coordinative bonds is attempted. Inspired by a report describing binding of metal acetates to multilayer TMDCs accompanied by a strong sulfur 2p core level shift in XPS,[1] we explore the capability of metal complexes as anchoring groups for TMDC-functionalization. Although, we cannot reliably reproduce the core level shifts presented in [1] with monolayer TMDCs it still seems possible to form layers of metal complexes on TMDCs.

Some arylazotetracyanocyclopentadienides are excellent photoswitches comprising a permanently charged moiety. They can be isomerized by visible light from trans to cis and reverse.[2] In the second part of the talk, synthesis and photochemical properties of different derivatives with anchoring groups for surface functionalization are presented. Particularly, some thiol functionalized derivatives show good photoswitching properties in solution and in solid state.

Finally, a short overview on the activities of the custom synthesis part of service project Z1 is given.

References

[1] C. Backes, et al., Angew. Chem. Int. Ed. 54, 2638 (2015).
[2] Y. Garmshausen, Dissertation, Humboldt-Universität zu Berlin (2019)

Speaker: J. Rika Simon
Project: A14
Office Number: 030 838 56052
E-Mail: rika.simon@fu-berlin.de

p-Terphenyl-based thiols bound to top-layer sulphur defects on MoS_2/Au(111) investigated by STM

J. Rika Simon, Paul Wiechers, Christian Lotze, Björn Kobin, Stefan Hecht, Katharina J. Franke

Abstract

The transition-metal dichalcogenide (TMDC) MoS_2 has been well established as a decoupling layer in an STM junction, where submonolayers allow for highly resolved dI/dV spectra. These enable us to distinguish between e.g. different rotamers of a molecule [1]. But there are also drawbacks: The properties of these 2D materials are highly dependent on their local structure, because defects influence their properties severely. For instance, sulphur point defects in MoS_2 show a localised resonance around the Fermi energy, not present in pristine MoS_2.

Here we show preliminary results of anchoring the thiol-based molecules CF3-3P-SH (trifluoromethyl-*p*-terphenyl-thiol) and 3P-SH (*p*-terphenyl-thiol) into purposely created top-layer sulphur point defects in MoS_2 on Au(111). One end-group of the anchored molecules is bound to the defect, allowing them to rotate around the anchoring point upon excitation with the STM tip. Electronically, the local structure of the MoS_2 greatly influences the molecules. The localised resonance of the sulphur point defects remains present in most cases despite the saturation with the anchored molecule. It seems to be localised not only at the anchoring point but along the whole molecule. Furthermore, we observe vibronic states of the molecules inside the MoS_2 gap that point to strong vibrational modes.

References

[1] N. Krane, C. Lotze, G. Reecht, L. Zhang, A. L. Briseno, and K. J. Franke., ACS Nano 12.11, 11698 (2018)

Speaker: Juan Pablo Guerrero
Project: B16
Office Number:
E-Mail: guerreroj97@zedat.fu-berlin.de

Electronic properties of MoS_2 monolayer doped by donor, acceptor, and aromatic molecules

Juan Pablo Guerrero, Ana M. Valencia, Jannis Krumland, Caterina Cocchi

Abstract

The electronic properties of hybrid inorganic-organic interfaces are critically influenced by the level alignment across the heterostructure and by possible hybridization effects that occur therein. In turn, these properties are determined by the nature of the molecular dopants and by their arrangements. In the framework of (hybrid) density functional theory, we investigate the electronic structure of a single sheet of MoS_2 covered by monolayers of planar molecules such as pyrene, tetrathiafulvalene, and bithiophene, which are known to act as donors, as well as with the acceptors 7,7,8,8-tetracyanoquinodimethane and its tetrafluorinated counterpart. Our results show that all considered heterostructures exhibit a type II level alignment with negligible charge transfer at the interface. However, in the electronic structure of the systems, the signatures of electron or hole doping to the MoS_2 can be identified.

Speaker: Jannis Krumland
Project: B16
Office Number:
E-Mail: krumlanj@physik.hu-berlin.de

Conditions for electronic hybridization between transition-metal dichalcogenide monolayers and physisorbed carbon-conjugated molecules

Jannis Krumland

Abstract

We present a first-principles study on electronic hybridization in inorganic-oraganic interfaces composed of monolayer transition-metal dichalcogenides (TMDCs; molybdenum and tungsten disulfide and diselenide) and exemplary carbon-conjugated molecules such as pyrene and perylene. By means of band-structure unfolding techniques applied to hybrid density-functional theory calculations including spin-orbit coupling, we achieve an intuitive and clear description of electronic interaction between the inorganic and organic components of the heterostructures. From atom-projected band structures, we are able to rationalize the strong mixing between the valence states of the TMDC and the molecular orbitals. We additionally clarify why the highest occupied orbital couples with the TMDC bands only very weakly, regardless of the composition of the interface. The proposed analysis based on band structure unfolding lends itself for computationally efficient and yet reliable predictions of electronic interactions in more complex hybrid interfaces including larger molecules harvesting visible radiation.

Session 4

Speaker: Alberto Eljarrat
Project: A12, Z2
Office Number: 030 2093 7734
E-Mail: aeljarrat@physik.hu-berlin.de

STEM-EELS characterization capabilities for HIOS

Alberto Eljarrat

Abstract

In this talk we will review the capabilities offered by scanning transmission electron microscopy (STEM) by looking at results obtained in the framework of projects Z2 and A12.

In this project, we employ a Nion HERMES dedicated STEM instrument recently installed at the IRIS building that offers state-of-the-art aberration corrected imaging and monochromated electron energy-loss spectroscopy (EELS). Additionally, this instrument is equipped with a Dectris ELA hybrid-pixel direct-detection camera and an in-situ liquid nitrogen cooling holder. We are thus able to perform a wide array of experiments, including chemical and structural characterization at the nanoscale, and measurement of EELS features related to optical and vibrational excitations.

Mapping the ionization edges observed in core-loss EELS, chemical and structural characterization at the nanoscale is achieved. In collaboration with several groups from the HIOS project, we have applied this characterization to plasmonic systems that mix carbon carbon based nano-structures with metallic content (iron, vanadium). We have also examined plasmonic systems based on metallic nano-rods (gold, silver) with organic cover, and bidimensional materials such as MoS_2. In these cases, the plasmonic and excitonic properties of these systems were also measured at the same instrument.

We design and employ advanced analysis algorithms, such as relativistic Kramers-Kronig analysis that allows to recover the complex dielectric function information from EELS. Additionally, we are performing tests measuring with high-energy resolution at cryogenic temperatures in order to reveal hidden excitonic and vibrational properties.

Speaker: Dmitrii Maksimov
Project: A13
Office Number: 0176 7519 7703
E-Mail: maksimov@fhi-berlin.mpg.de

Conformation prediction for flexible molecules at surfaces using ab initio random structure search

Dmitrii Maksimov

Abstract

Finding stable molecular adsorbate structures (in isolation or forming layers) is especially difficult for weakly bound systems or flexible adsorbates. To address this issue using ab initio potentials, we created a random structure search program that explicitly considers molecular flexibility and location with respect to 1D (e.g. ions), 2D (e.g. surfaces) or 3D (e.g. solids) static references [1]. The search method is combined with a structure optimization algorithm that uses preconditioners specifically tailored to accelerate the optimization of van der Waals bonded structures and is capable of dealing with significant structural changes. We demonstrate that these methods improve geometry optimization performance and show the ability of the package to anticipate the structure of molecules adsorbed on TMDC monolayers.

References

[1] https://github.com/sabia-group/gensec

Speaker: Manuel Katzer
Project: B12
Office Number: 030 314 23764
E-Mail: manuel.katzer@physik.tu-berlin.de

Förster-type energy transfer between molecules and atomically thin semiconductors

Manuel Katzer, Malte Selig, Sviatoslav Kovalchuk, Kyrylo Greben, Kirill Bolotin, Andreas Knorr

Abstract

Interfaces of dye molecules and two-dimensional transition metal dichal-cogenides (TMDCs) are promising candidates for optoelectronic applications since they combine the large molecular optical dipoles and spectral tunability with high carrier mobilities in TMDCs [1]. At such interfaces, Förster energy transfer is one key mechanism for excitation transfer and has many intriguing technical applications [2].

In a joint theory-experiment study, we report microscopic calculations of the Förster induced transition rate from dye molecules to a TMDC layer and discuss the corresponding optical signatures, with excellent agreement to the experimental data. The theoretic approach is based on microscopic Bloch equations which are solved self-consistently together with Maxwells equations [3], incorporating the sample geometry within the Rytova-Keldysh framework. Our study reveals the dominating influence of dark TMDC excitons.

References

[1] Jariwala et al., Nat. Mater. 16, 170 (2017)
[2] Dagher et al., Nat. Nanotech. 13, 925–932 (2018)
[3] Selig et al., Phys. Rev. B 99, 035420 (2019)

Speaker: Hélène Seiler
Project: B17
Office Number: 030 8413 5148
E-Mail: seiler@fhi-berlin.mpg.de

Ultrafast vibrational dynamics of TMDC/organic heterostructures in momentum space

Hélène Seiler, Patrick Hildebrandt, Daniela Zahn, Benedikt Haas, Johannes Müller, Xiaomin Xu, Soohyung Park, Thomas Vasileiadis, Yoav William Windsor, Yingpeng Qi, Norbert Koch, Christoph T. Koch, Ralph Ernstorfer

Abstract

Non-radiative dissipation pathways are ubiquitous in nanoscale materials and are the dominant loss channels in most opto-electronic devices. Here we show how femtosecond electron diffraction can provide direct insights into these channels in a MoS$_2$/pentacene heterostructure. Before the couplings between the inorganic and organic components of the heterostructure are analyzed, we first investigate the two materials separately. In a previous study we have provided an in-depth analysis of the nuclear dynamics in pentacene single crystals [1]. In this talk we focus on recent results obtained on the TMDC side. Specifically, we employ an approach where the energy flows in photo-excited MoS$_2$ are probed with time- and momentum-resolution [2]. Having gained a detailed view of the vibrational dynamics in the individual components, we are now equipped to study the couplings specific to the heterostructure.

References

[1] H. Seiler, M. Krynski, D. Zahn, S. Hammer, Y. W. Windsor, T. Vasileiadis, J. Pflaum, R. Ernstorfer, M. Rossi, H. Schwoerer, Sci. Adv. 7(26), eabg0869 (2021).
[2] H. Seiler, D. Zahn, M. Zacharias, P.-N. Hildebrandt, T. Vasileiadis, Y. W. Windsor, Y. Qi, C. Carbogno, C. Draxl, R. Ernstorfer, F. Caruso, Nano Lett. 21(14), 6171–6178 (2021).

Speaker: Alexander Neef
Project: B17
Office Number: 030 8413 5135
E-Mail: neef@fhi-berlin.mpg.de

Orbital-resolved observation of singlet fission

Alexander Neef, Samuel Beaulieu, Sebastian Hammer, Shuo Dong, Tommaso Pincelli, Julian Maklar, Martin Wolf, Laurenz Rettig, Jens Pflaum, Ralph Ernstorfer

Abstract

Great strides have been made in observing ultrafast molecular processes on their intrinsic length and time scales. Some techniques directly image molecular motion [1], while others access spatial information in momentum space [2]. Ideally, these techniques should allow to follow the dynamics of electrons and atoms in time, space and energy. Experiments so far have shown potential but exploiting them to unravel major mysteries in ultrafast molecular processes has not been achieved. Here we investigate ultrafast singlet fission – the process of a singlet exciton splitting into two triplet excitons [3] – in crystalline pentacene by using time- and angle-resolved photoemission spectroscopy. Using insights from orbital tomography, we were able to match singlet and triplet excitons to their orbital character based on their respective momentum maps. Furthermore, we separated the dynamics of states with different orbital character and thereby gained mechanistic insights about singlet fission.

References

[1] Cocker, Peller et al., Nature 539, 263–267 (2016)
[2] Wallauer et al., Science 371, 1056-1059 (2021)
[3] Smith & Michl, Chem. Rev. 110, 6891–6936 (2010)

Speaker: Johannes Müller
Project: A12
Office Number: 030 2093 82512
E-Mail: Johannes.mueller@physik.hu-berlin.de

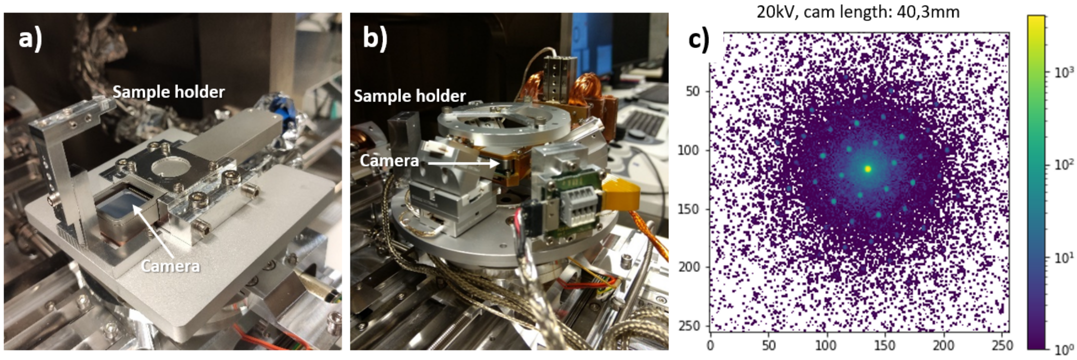
Enabling imaging of electron beam sensitive hybrid materials

Johannes Müller, Julian Schmehr, Jin Cao, Mara Voiculescu, Benedikt Haas, Sherjeel Shabih,
Julian Becker, Christoph T. Koch

Abstract

We modified our scanning electron microscope (SEM) to acquire diffraction patterns of thin samples in transmission. By analyzing the acquired diffraction patterns, we can investigate crystallographic and material properties on mm^2-scale with a spatial resolution of less than 5nm, for example how organic and inorganic components of HIOS are orientated with respect to each other. The SEM platform offers low acceleration voltages (0.5-30kV) reducing knock-on beam damage to the sample and increasing elastic scattering yielding stronger contrast compared to higher electron energies. We can map with up to 2000 diffraction patterns per second while scanning the electron beam over the sample, giving the A12 project a fast and easy to use system to investigate HIOS and attracting interest from microscope manufacturers to commercialize this method.

Our newly acquired direct detection camera has single electron sensitivity reducing sample damage and possible contaminations further. This new camera should allow us to implement ptychography, which will improve the spatial resolution even more. We are also now able to map electric fields in the SEM and are planning to realize diffraction in reflection, which will allow us to investigate samples on top of substrates without the otherwise necessary sample preparation.



a) First setup using a direct detection camera from X-Spectrum GmbH. b) Second setup using a scintillator based camera with a hexapod for 6 axis sample control. c) Diffraction pattern of a MoS2 multi-layer using the direct detection camera (log scale in counted electrons).

Session 5

Speaker: Niko Nikolay
Project: B18
Office Number: 030 2093 4991
E-Mail: nikolay@physik.hu-berlin.de

Polarization sensitive correlations of single photon emitters in *h*-BN

Niko Nikolay, Florian Böhm, Fridtjof Betz, Günter Kewes, Noah Mendelson, Sven Burger,
Igor Aharonovich, Oliver Benson

Abstract

Optically active color centers in hexagonal boron nitride are promising candidates as single photon sources. Therefore, they have been extensively studied in recent years. Their atomic origin is still unknown, so the experiments presented in this talk shed light on the underlying level structure. We will show that two spectra differing in their polarization contribute to the fluorescence of the observed single photon emitter. Based on these results, we then present polarization-sensitive photon correlation measurements and compare them to a multilevel rate equation model.

Speaker: Martin Rothe
Project: B2
Office Number: 030 2093 4934
E-Mail: rothe@physik.hu-berlin.de

Degradation assessment of hybrid plasmonic nanowires

Martin Rothe, Yuhang Zhao, Henry Halim, Yan Lu, Oliver Benson

Abstract

Near-future high-end devices will consist of hybrid inorganic-organic nanophotonic circuits. Many of their building blocks, e.g. junctions, routers, couplers, or sensors, will rely on interactions between photo-emitters and waveguides, resonators, or antennas. A limiting factor for the operation of the device is the lifetimes or stability of its building blocks. A typical weak spot is the photo-bleaching of the optically coupled organic emitters, even though this problem is often omitted in discussions and proof-of-principle demonstrations.

We have studied single hybrid core / shell perylene diimide (PDI) / silver nanowires as a building block, which guides optical waves due to the tightly-confined propagating surface plasmon modes. The functionality ranges from being a versatile waveguide and resonator[1], to plasmon routing and chiral sensing[2]. During its operation of spectrally converting propagating plasmons, we recorded leaking fluorescence light to determine the bleaching process of the encapsulated PDI molecules. Based on our universal analytical model for 1D optical waveguides and spatio-temporal fluorescence measurements, we derived that the bleaching process of the PDI molecules is triggered by a single-photon absorption – proportional to the fluorescence process. Consequently, we measured that the yield of fluorescence plasmons is a fixed quantity independent of the operation conditions, without perturbing the operation of the building block.

References

[1] M. Rothe, Y. Zhao, G. Kewes, Z. Kochovski, W. Sigle, P. A. van Aken, C. Koch, M. Ballauff, Y. Lu, O. Benson, Sci. Rep. 9, 3859 (2019)
[2] M. Rothe, Y. Zhao, J. Müller, G. Kewes, C. T. Koch, Y. Lu, O. Benson, ACS Nano 15, 351 (2021)

Speaker: Ana M. Valencia
Project: B16
Office Number:
E-Mail: ana.valencia@physik.hu-berlin.de

Charge-transfer excitons in pentacene:perfluoropentacene co-crystal

Ana M. Valencia, Darius Günder, Michele Guerrini, Tobias Breuer, Gregor Witte, Caterina Cocchi

Abstract

Disclosing the nature of optical excitations in organic co-crystals is a challenge due to the complex interplay between the structural arrangement of the molecules and long-range electronic interactions in these systems. For these reasons, the application of isolated cluster models is highly questionable. By means of density functional theory and many-body perturbation theory, we investigate the electronic and optical properties of the triclinic pentacene: perfluoropentacene co-crystal. The band-structure analysis indicates that the highest-occupied and the lowest-unoccupied states are mainly localized on different molecules, suggesting the presence of a charge-transfer exciton at the lowest energy, which is indeed revealed in the dielectric tensor computed from the solution of the Bethe-Salpeter equation. We demonstrate that this feature cannot be reproduced by a molecular cluster model where the first excitation is unambiguously polarized along the short molecular axis. Comparison with corresponding measurements clarifies the importance of adequate periodic treatment of molecular co-crystals to correctly reproduce the character of their excitations [1].

References

[1] D. Günder, A. M. Valencia, et al., in preparation.

Speaker: Hu Lin
Project: A6
Office Number: 030 2093 7714
E-Mail: linhu621@physik.hu-berlin.de

Reversible switching of charge transfer at the graphene–mica interface with intercalating molecules

Hu Lin, José D. Cojal González, Nikolai Severin, Igor M. Sokolov, Jürgen P. Rabe

Abstract

Understanding and controlling charge transfer through molecular nanostructures at interfaces is of paramount importance, particularly for electronic devices but also for contact electrification or in bio-electronics. We demonstrate here reversible switching of charge transfer at graphene-mica interface by molecularly thin layers of fluid molecules intercalating the interface. While water intercalating the interface blocks charge transfer [1] [2], organic molecules like ethanol filling initially dry interface, or diffusively replacing water do not block or restore the charge transfer at the interface respectively. We propose a charge transfer model between occupied mica trap states and electronic states of graphene, offset by the electrostatic potentials produced by the molecular dipole layers, as supported by molecular dynamics simulations. Our work facilitates the understanding of the influence of molecular intercalants onto charge transfer at interfaces.

References

[1] J. Shim, C. H. Lui, T. Y. Ko, Y. J. Yu, P. Kim, T. F. Heinz, S. Ryu, Nano Lett. 12, 648-54 (2012).
[2] H. Lin, A. Schilo, A. R. Kamoka, N. Severin, I. M. Sokolov, J. P. Rabe, Phys. Rev. B 95, 195414 (2017).

Speaker: Max Heyl
Project: B14
Office Number: 030 2093 7552
E-Mail: max.heyl@hu-berlin.de

How to exfoliate millimeter-scale MoS_2 Monolayers with gold and their transfer onto arbitrary substrates

Max Heyl, Daniel Steffen Rühl, Fabian Gärisch, David Burmeister, Thorsten Schultz, Sebastian Pallasch, Sylke Blumstengel, Giovanni Ligorio, Norbert Koch, Emil J. W. List-Kratochvil

Abstract

The efforts to reach the monolayer limit of transition metal dichalcogenides (TMDCs), is motivated by a whole set of interesting properties accompanying two-dimensional TMDCs, e.g., indirect-to-direct band gap transitions, 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, where reliable transducing elements to convert small potential changes into large current signals are required. 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. Gold proved to be suitably adhesive for exfoliation of several 2D materials, including TMDCs. 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. The aim of this talk is to guide through this process to unlock large-scale high-quality monolayers in as many collaborating labs as possible.

Speaker: Elsie Bowen-Dodoo
Project: B9
Office Number: 030 8413 5128
E-Mail: bowendodoo@fhi-berlin.mpg.de

Investigating the formation dynamics of hybrid interface excitons in ZnO-based hybrid systems

E. Bowen-Dodoo, L. Gierster, J.-C. Deinert, S. Vempati, Y. Garmshausen, S. Hecht, J. Stähler

Abstract

In optoelectronic applications, the best of both worlds comes from hybrid systems consisting of an inorganic material combined with a functional organic molecular adlayer. This project seeks to provide a detailed understanding of the formation dynamics of hybrid interface excitons, by employing time-resolved two-photon photoemission (TR-2PPE) spectroscopy. ZnO $(10\bar{1}0)$ serves as the inorganic substrate, and p-quinquephenyl-pyridine (5P-Py) acts as the organic molecule adlayer. A giant work function reduction, $\Delta\Phi$ of up to 2.1 eV is observed upon 5P-Py monolayer adsorption on ZnO. An in-gap state below the Fermi level is also observed. Intramolecular excitation as excitation pathway, is exploited for study. A strong signature lasting about 350 fs is observed at 1.6 eV above the Fermi level, and this is attributed to electrons populating the LUMO after excitation. Two intensity transients are also observed at later delays after the LUMO has decayed to the ZnO conduction band. They build up within 100(10) ps and exhibit lifetimes of several μs. Further investigations relate these intensity transients to hybrid exciton formation.

Speaker: Fabian Gärisch
Project: B14
Office Number: 030 2093 82530
E-Mail: fabian.gaerisch@hu-berlin.de

Organic synaptic diodes based on polymeric mixed ionic-electronic conductors

Fabian Gärisch

Abstract

Neuromorphic devices are likely to be the next evolution of computing, allowing to implement machine learning within hardware components. In biological neural systems, learning and signal processing are achieved by communication between neurons through time-dependent ion flux in the synapses. Integrating such ion-mediated operating principles in neuromorphic devices could deliver an energy efficient and powerful technology. Here we revisit and modify a device known as a light-emitting electrochemical cell, exploiting its ability to modulate current through ion accumulation/depletion at the electrodes and turn it into an organic synaptic diode. This two-terminal device is based on an organic mixed ionic-electronic conducting polymer that serves as active layer for conduction of lithium ions as well as charge carriers. 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 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 polymeric nature, combined with the simple two-terminal architecture of the presented neuromorphic device opens up a range of possibilities regarding the fabrication of artificial neural networks.

List of Names

Last Name	First Name	Session Number	Time	Project Number	Page Number
Akkoush	Alaa	3	Monday, 13:45	A13	18
Bowen-Dodoo	Elsie	5	Thursday, 13:45	B9	37
Christiansen	Dominik	1	Monday, 10:45	B12	9
Eljarrat	Alberto	4	Tuesday, 10:00	A12	25
Gärisch	Fabian	5	Tuesday, 14:00	B14	38
Gonzalez Oliva	Ignacio	2	Monday, 11:30	B11	12
Guerrero	Juan Pablo	3	Monday, 14:45	B16	22
Hamdi	Hanen	2	Monday, 11:45	B16	13
Hartfiel	Jennifer	3	Monday, 14:00	A14	19
Heyl	Max	5	Tuesday, 13:30	B14	36
Jacobs	Matheus	2	Monday, 12:00	B16	14
Katzer	Manuel	4	Tuesday, 10:30	B12	27
Kobin	Björn	3	Monday, 14:15	A3	20
Krumland	Jannis	3	Monday, 15:00	B16	23
Lin	Hu	5	Tuesday, 13:15	A6	35
Maksimov	Dmitrii	4	Tuesday, 10:15	A13	26
Müller	Johannes	4	Tuesday, 11:15	A12	30
Neef	Alexander	4	Tuesday, 11:00	B17	29
Nikolay	Niko	5	Tuesday, 12:30	B18	32
Raoufi	Meysam	1	Monday, 11:00	B7	10
Rothe	Martin	5	Tuesday, 12:45	B2	33
Rühl	Daniel Steffen	2	Monday, 12:30	B3	16
Seiler	Hélène	4	Tuesday, 10:45	B17	28
Simon	J. Rika	3	Monday, 14:30	A14	21
Tanda Bonkano	Boubacar	1	Monday, 10:15	B9	7
Valencia	Ana M.	5	Tuesday, 13:00	B16	34
Wang	Qiang	1	Monday, 10:30	A8	8
Zorn Morales	Nicolas	2	Monday, 12:15	B3	15

Contact

If you are experiencing technical problems, the administration of the CRC 951 will gladly try to help you. You can contact the administration via:

ZOOM: Write a private message to Christian Pugatschow or Jule Budnick

E-mail: sfb951@physik.hu-berlin.de

Phone: +49 30 20 93 66 380

+49 30 20 93 66 374

Imprint

Chair:

Prof. Dr. Norbert Koch

Humboldt-Universität zu Berlin
Institut für Physik
Brook-Taylor-Straße 6
12489 Berlin

Organisation:

Christian Pugatschow, Jule Budnick

Humboldt-Universität zu Berlin
Collaborative Research Centre 951
Zum Großen Windkanal 2
12489 Berlin