2D Materials and Hybrids: Hybrid Quasiparticles in Quantum Materials

771. WE-Heraeus-Seminar

24 Sep - 28 Sep 2022
Physikzentrum
Bad Honnef/Germany

WILHELM UND ELSE
HERAEUS-STIFTUNG

Introduction

The Wilhelm und Else Heraeus-Stiftung is a private foundation that supports research and education in science with an emphasis on physics. It is recognized as Germany's most important private institution funding physics. Some of the activities of the foundation are carried out in close cooperation with the German Physical Society (Deutsche Physikalische Gesellschaft). For detailed information see https://www.we-heraeus-stiftung.de

Aims and scope of the 771. WE-Heraeus-Seminar

Quasiparticles and their mutual interactions form the basis for quantum technologies and integrated opto-/electronic circuits with embedded novel functionalities. It is therefore important to understand the underlaying correlations including coherence properties and their dynamics on a very fundamental level. Material platforms hosting such quasiparticles and potentially allowing the implementation of integrated circuitries with intelligent or quantum functionalities are two dimensional (2D) materials and their hybrids formed from e.g. coupling with plasmonic structures. Two-dimensional materials exhibit a rich set of quasiparticle excitations, together with exciting spin and valley properties and topological non-trivial materials. 2D materials such as the intensively studied class of transition metal dichalcogenides (TMDCs), tetradymites, and further 2D materials with distinct topological properties have emerged as a platform to realize, understand and tailor quantum hybrid structures. They can be combined with arbitrary substrates, be twisted, and be assembled into heterostructures. Those 2D materials can be interfaced with plasmonic films and nanostructures to fundamentally study weak and strong interactions, as well as energy and information transfer between the different quasiparticles and for the realization of hybrid quasiparticles that can be integrated in functional units. The aim of the seminar is bringing together a group of excellent internationally well-established researchers working on 2D materials, hybrids and their quasiparticles from various points of views using different characterization tools, materials, or theoretical methods to foster synergy. Young researchers at the PhD or PostDoc levels will be encouraged to actively participate with poster presentations, at a panel discussion and a synergy workshop session.

Scientific Organizers:

Prof. Dr. Ursula Wurstbauer — WWU Münster, Germany
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Prof. Dr. Nahid Talebi — Universität zu Kiel (CAU), Germany
E-mail: talebi@physik.uni-kiel.de

Introduction

Administrative Organization:

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Martina Albert

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Registration:

Martina Albert (WE Heraeus Foundation)
at the Physikzentrum, reception office
Saturday (17:00 h – 20:00 h) and Sunday morning

Program

Program

Saturday, 24 September 2022

17:00 – 20:00	Registration	
18:30 – 19:30	*BUFFET SUPPER*	
19:30 – 19:40	Scientific organizers	**Welcome note**
19:40 – 20:25	Asger Mortensen	**Exploring nonlocal electrodynamics: Interfacing metals and 2D materials**

Sunday, 25 September 2022

08:00	*BREAKFAST*	
09:00 – 09:45	Kai Rossnagel	**Quasi-electron imaging of quasi-2D materials**
09:45 – 10:30	Christoph Kastl	**Spin-orbit control in graphene-based van der Waals interfaces**
10:30 – 10:50	Iris Niehues	**Nanoscale mapping of carrier density in intercalated 2D semiconductors by IR and THz nanoscopy**
10:50 – 11:10	*COFFEE BREAK*	
11:10 – 11:55	Jana Zaumseil	**Tuning carbon nanotubes emission with quantum defects and cavity-hybridization**
11:55 – 12:40	Christoph Lienau	**Communication on the Nanoscale: Strong coupling and Multidimensional Spectroscopy**
12:40	*LUNCH*	

Program

Sunday, 25 September 2022

14:00 – 14:45	Antonietta de Sio	**Ultrafast nonadiabatic dynamics in optoelectronic materials revealed by two-dimensional electronic spectroscopy**
14:45 – 15:30	Nahid Talebi Sarvari	**Interaction of Electron Beams with 2D Materials and Hybrids**
15:30 – 15:50	Jonas Bauer	**Probing excitonic population dynamics by nonlinear optical wave mixing in monolayer WSe2**
15:50 – 16:20	*COFFEE BREAK*	
16:20 – 17:05	Fabio Caruso	**Phonons out of equilibrium: from ab-initio theory to pump-probe experiments**
17:05 – 17:50	Michael Bauer	**Non-equilibrium carrier dynamics and carrier-phonon interaction in 2D quantum materials**
17:50 – 18:10	Deividas Sabonis	**Nonlocal signatures of hybridization between quantum dot and Andreev bound states**
18:30	*DINNER*	
20:00	**Networking / Panel discussion**	

Program

Monday, 26 September 2022

08:00	*BREAKFAST*	
09:00 – 09:45	Giancarlo Soavi	**Nonlinear optics with 2D materials**
09:45 – 10:30	Thomas Heine	**Topological 2D polymer/graphene heterostructures**
10:30 – 10:50	Zeinab Ebrahimpour	**THz nonlinear optical conductivity in bulk black phosphorus**
10:50 – 11:10	*COFFEE BREAK*	
11:10 – 11:55	Rainer Hillenbrand	**Strong coupling between molecular vibrations and phonon polaritons in van der Waals materials**
11:55 – 12:40	Javier García de Abajo	**Quantum optical phenomena in 2D material heterostructures**
12:40	*LUNCH*	
14:00 – 14:45	**Poster Flash Presentations**	
14:45 – 14:55	**Conference photo**	
14:55 – 15:50	**Postersession (with coffee)**	
16:00 – 18:30	**Excursion (hike / walk)**	
18:45 – 19:00	**Poster Awards**	
19:00	*HERAEUS DINNER at the Physikzentrum* *(cold & warm buffet, with complimentary drinks)*	

Program

Tuesday, 27 September 2022

08:00	*BREAKFAST*	
09:00 – 09:45	Doris Reiter	**Theory of spatiotemporal carrier dynamics in 2D semiconductors**
09:45 – 10:30	Luiz Tizei	**Fast electron spectroscopies to probe optical excitations in 2D materials**
10:30 – 10:50	Shubhadeep Biswas	**Strong light-field controlled valleytronics**
10:50 – 11:10	*COFFEE BREAK*	
11:10 – 11:55	Harald Giessen	**Topological plasmonics: Ultrafast vector movies of plasmonic skyrmions on the nanoscale**
11:55 – 12:40	Anika Schlenhoff	**Using image-potential states as a quantum probe of atomic-scale spin textures**
12:40	*LUNCH*	
14:00 – 14:45	Joshua Robinson	**Semiconductors and Metals at the Atomic Limit**
14:45 – 15:30	Alexander Steinhoff (online)	**Theory of exciton dynamics in atomically thin semiconductors**
15:30 – 15:50	Nihit Saigal	**Collective Excitations and Degenerate Phases in TMDC Bilayers**
15:50	*COFFEE BREAK*	

Program

Tuesday, 27 September 2022

16:20 – 17:05	Alexander Weber-Bargioni	**Engineering heterostructures in 2-D solids to define new localized quantum states with quantum coherent properties**
17:05 – 17:50	Paolo Sessi	**2D ferroics explored via scanning tunneling microscopy and spectroscopy**
17:50 – 18:10	Julian Klein	**Magnetically correlated defects in a quasi-1D electronic van der Waals magnetic semiconductor**
18:30	*DINNER*	
20:00 – 20:30	**Posterprize Talks**	
	(10 min each)	

Wednesday, 28 September 2022

08:00	*BREAKFAST*	
09:00 – 09:20	Shen Zhao	**Observation of bright hybridized interlayer exciton in MoTe2/MoSe2 heterobilayers**
09:20 – 10:05	Christoph Koch	**Exploration of (hybrid) quasiparticles in quantum materials using high-resolution electron energy-loss spectroscopy**
10:05 – 10:50	Frank Koppens (online)	**Cryogenic photocurrent nanoscopy of twisted graphene**
10:50 – 11:00	Scientific Organizers	**Closing remarks**
11:00 – 11:30	***End of the seminar and FAREWELL COFFEE / Departure***	

Posters

Posters

Mohammad Adnan	**Role of a 2D inorganic-organic hybrid semiconductor in controlling the stability of 3D perovskites**
María Barra-Burillo	**Microcavity phonon polaritons: from the weak to the ultrastrong phonon-photon coupling regime**
Cian Bartlam	**Orientation Dependent Coupling in Type-II Perylene-TMD Heterostructures**
Jonas Bauer	**Probing excitonic population dynamics by nonlinear optical wave mixing in monolayer WSe2**
Shubhadeep Biswas	**Strong light-field controlled valleytronics**
Fatemeh Davoodi	**Tailoring Plasmon-Exciton Interactions in Nanometer-Thick Gold–WSe2 Hybrid Structure using Cathodoluminescence Spectroscopy**
Zeinab Ebrahimpour	**THz nonlinear optical conductivity in bulk black phosphorus**
Zeinab Ebrahimpour	**EuPRAXIA@SPARC_LAB FELs: Brilliant Soft X-Ray Pulse Sources for Spectroscopy and Imaging**
Khairi Elyas	**The Fabrication of nanostructured van der Waals Heterostructures using He Ion Beam Patterning**
Stefan Heiserer	**Observation and Dynamics of Interlayer Excitons in Perylene Bisimide-MoS2 Heterostructures**

Posters

Jakob Henz	**Light matter interaction and quantum confinement in 2D polar metals**
Julian Klein	**Magnetically correlated defects in a quasi-1D electronic van der Waals magnetic semiconductor**
Sebastian Klimmer	**All-optical polarization and amplitude modulation of second harmonic generation in atomically thin semiconductors**
Johannes Krause	**Enhancing photoluminescence and Raman signals in TMDC monolayers via plasmonic nanostructures**
Iris Niehues	**Nanoscale mapping of carrier density in intercalated 2D semiconductors by IR and THz nanoscopy**
Hossein Ostovar	**Optically detected ionic gate spectroscopy and exciton manifolds in highly doped WS2**
Borislav Polovnikov	**Signatures for localized and remote trions in MoSe2 / WS2 heterostructures**
Marina Quijada	**Nonlinear plasmonic response to circularly polarized light**
Deividas Sabonis	**Nonlocal signatures of hybridization between quantum dot and Andreev bound states**
Deividas Sabonis	**Planar Josephson Junctions under Microwave Irradiation**

Posters

Nihit Saigal	**Collective Excitations and Degenerate Phases in TMDC Bilayers**
Kabyashree Sonowal	**Valley spin acoustic resonance in monolayer MoS_2**
Shen Zhao	**Observation of bright hybridized interlayer exciton in MoTe2/MoSe2 heterobilayers**

Abstracts of Lectures

(in alphabetical order)

Probing excitonic population dynamics by nonlinear optical wave mixing in monolayer WSe_2

Jonas M. Bauer[1], Lijue Chen[1,2], Philipp Wilhelm[1], Sebastian Bange[1], John M. Lupton[1], Kai-Qiang Lin[1]

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Monolayer semiconductors are emerging platforms for strong nonlinear light-matter interaction, due to their giant oscillator strength of tightly bound excitons. Recently, we reported the existence of a new excitonic species, the high-lying exciton (HX), in monolayer WSe_2. The HX appears at around twice the energy of the band-edge A-exciton, forming a ladder-type excitonic three-level system.

Excitonic quantum interference emerges in second-harmonic generation (SHG) in monolayers[1] and twisted bilayers[2].

Here we apply time-resolved nonlinear spectroscopy to probe the excitonic dynamics in such excitonic three-level system[3]. We find that a significant time difference between two light pulses is necessary for optimal sum-frequency generation (SFG) and four-wave mixing (FWM) when one of the pulses is in resonance with an excitonic transition. The experimental results are explained by numerical calculations based on density-matrix formalism, which provides insights to the coherent exciton dynamics on a femtosecond scale.

References

[1] K.-Q. Lin et al., Nat. Phys. **15**, 242-246 (2019).

[2] K.-Q. Lin, J. M. Bauer et al., Nat. Commun. **12**, 1553 (2021).

[3] J. M. Bauer et al., Nat. Photon. accepted (2022). Preprint available at Research Square, DOI: 10.21203/rs.3.rs-1347888/v1.

Non-equilibrium carrier dynamics and carrier-phonon interaction in 2D quantum materials

M. Bauer[1, 2]

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[2] *Kiel Nano, Surface and Interface Science KiNSIS, Kiel University, Kiel, Germany*

In my presentation, I will present results of two recent time- and angle resolved photoemission (TRARPES) studies on ultrafast processes in 2D transition metal dichalcogenides (TMDC).

In a singly oriented film of single-layer WS_2 deposited on a Au(111) surface we studied the spin- and valley-selective photoexcitation and decay of free carriers at the K and K'-points [1]. Our results reveal that in the valence band maximum an ultimate valley polarization of free holes of 84% can be achieved upon excitation with circularly polarized light at room temperature. For the photoexcited free electrons in the conduction band minimum, we observe a significantly smaller valley polarization. Clear differences in the carrier dynamics between electrons and holes imply intervalley scattering processes into dark states being responsible for the efficient depolarization of the excited electron population. The observed characteristic timescale for this process match very well values predicted from theory for the formation of momentum forbidden intervalley dark excitons in W-based SL TMDCs due to electron-phonon interaction.

In the second example I will discuss results on the electron-phonon interaction in the Weyl-semimetal Td-WTe_2 as probed in a TRARPES experiment following the excitation of coherent phonons [2]. We can show that a phonon-frequency selective analysis of the experimental data provide high-resolution information on strengths and types of couplings of the individual modes to the electronic bands. I particularly will discuss results on the transient modulation of a Dresselhaus-type spin splitting of electronic bands driven by the selective coupling of an interlayer shear mode of the layered compound. The latter results reveal real-time insights into electron-phonon coupled processes that are of vital importance for a light-driven topological phase transition in Td-WTe_2.

References

[1] H. Beyer, *et al.*, Phys. Rev. Lett. **123**, 236802 (2019).
[2] P. Hein, *et al.*, Nat. Commun. **11**, 2613 (2020).

Strong light-field controlled valleytronics

S Mitra[1,2], Á Jiménez-Galán[3,4] , M Neuhaus[1,2], O Smirnova[3,5] , M Ivanov3,[6,7] , M F Kling[1,2,8,9] and S Biswas[1,2 *]

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[3] Max-Born-Institute, Berlin, Germany
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[6] Department of Physics, Humboldt University, Berlin, Germany.
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[8] SLAC National Accelerator Laboratory, Menlo Park, CA, USA.
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The fusion between modern quantum material science with the ability to control sub-cycle light fields promises ultrafast light-field electronics, which can potentially operate million times faster than state-of-the-art electronics. Inversion-symmetry broken 2d-quantum materials, e.g., monolayer hexagonal boron nitride (hBN) or transition metal dichalcogenides (TMDs), possess valley degrees of freedom associated with the local minima in the band structure [1]. Their A) selective excitation, B) controlled inter-valley switching, and C) readout of the excited quantum states are the prerequisites for their use in information processing. Attempts to control valley polarization so far relied on resonant excitation [2], along with valley switching with strong terahertz fields [3].

Here, following the recent theoretical prediction [4], we demonstrate all-optical nonresonant strong field control of all the three major steps (A-C). We used two-color bi-circular ultrashort laser fields to control the selective valley excitation in hBN and then probe the quantum valley states via third harmonic generation ellipsometry. The non-resonant pump field modifies the topological properties of the band structure by inducing Halden-type [5] complex next nearest neighbor hopping, which manifests the breaking of time-reversal symmetry. This hopping interaction controls the valley asymmetry and can be manipulated by varying the phase delay between the two colors of the bi-circular field. In agreement with the theoretical predictions, the experimental data shows selective enhancement of valley population as a function of the two-color phase delay. These results demonstrate a new regime of valleytronics and lightinduced topology in quantum materials

References

[1] J. R. Schaibley et al. Nat. Rev. Mater **1**, 16055 (2016)
[2] K. F. Mak et al. Nat. Nanotech., **557**, 76–80 (2012)
[3] F. Langer et al. Nature **557**, 76–80 (2018)
[4] Á. Jiménez-Galán et al. 2020 Nat. Photon. **14**, 728 – 732 (2020)
[5] F. D. M. Haldane Phys. Rev. Lett. **61**, 2015 – 2018 (1988)

Phonons out of equilibrium: from ab-initio theory to pump-probe experiments

F. Caruso

[1]*University of Kiel*

The ultrafast lattice dynamics under non-equilibrium conditions encodes detailed information on the fundamental interaction mechanisms in solids and their time scales. This talk will focus on the ab-initio modelling of non-equilibrium phonon dynamics based on the time-dependent Boltzmann equation (TDBE) and its application to the study of 2D and layered materials [1].

The TDBE approach enables to predict transient changes of the phonon population with unprecedented time and momentum resolution, and it constitutes a valuable tool to identify the fingerprints of non-thermal states of the lattice in pump-probe experiments. I will illustrate how predictive ab-initio simulations can be combined with femto-second electron diffuse scattering to surgically dissect and identify timescales, decay pathways, and interaction mechanisms which underpin the lattice thermalization in black phosphorus [2] and monolayer MoS2 [3].

References

[1] Caruso, J. Phys. Chem. Lett. **12**, 1734 (2021)
[2] Seiler et al., Nano Lett. **21**, 6171 (2021).
[3] Britt et al, Nano Lett. **22,** 4718 (2022).

Ultrafast nonadiabatic dynamics in optoelectronic materials revealed by two-dimensional electronic spectroscopy

Antonietta De Sio[1]

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The dynamics of non-equilibrium excitations upon light-matter interaction in functional photoactive materials are governed by the details of the electronic structure, by its coupling to the vibrational degrees of freedom, by the specific structural arrangement and possible disorder in the condensed phase. All these microscopic properties ultimately determine the flow of energy and the motion of charges upon photoexcitation. They are therefore of primary importance for the rationale design and for efficient application of such materials in e.g., light-harvesting and energy conversion, sensors, light-emission, as well as for the control of such dynamics on the nanoscale.

In this talk, we present some of our recent results probing strong exciton-phonon couplings and nonadiabatic dynamics in technologically relevant materials for optoelectronics and discuss how two-dimensional electronic spectroscopy (2DES) can provide detailed new insight into these dynamics and underlying couplings [1-5]. Specifically, using 2DES we reveal and track wave packet motion through intermolecular conical intersections in molecular aggregates and phonon-driven exciton Rabi oscillations in halide perovskites, and discuss their potential role for steering energy transport on the nanoscale.

References

[1] A. De Sio et al., Nature Commun. **7**, 13742 (2016)
[2] A. De Sio et al, Phys. Chem. Chem. Phys. **19**, 18813-18830 (2017)
[3] X.T. Nguyen et al, J. Phys. Chem. Lett. **10**, 5414 (2019)
[4] A. De Sio et al, Nature Nanotechnol. **16**, 63-68 (2021)
[5] X.T. Nguyen et al, submitted (2022)

THz nonlinear optical conductivity in bulk black phosphorus

Z. Ebrahimpour[1,2] N. Adhlakha[2], P. Di Pietro[2], J. Schmidt[2], S. Lupi[3] and A. Perucchi*[2]

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[3] CNR-IOM and Dipartimento di Fisica, Universit`a di Roma Sapienza, Roma, Italy

Black phosphorus (BP), as a new member of the 2D materials family, has recently attracted much interest due to its high carrier mobility and unique strong in-plane anisotropic physical properties. Its promising optical, electrical, and thermal conductance are significant advantages for optoelectronic and photonic applications. BP's bandgap is direct and strongly thickness dependent, spanning from 0.3 eV in bulk to 2 eV in monolayer, although it always remains direct. Moreover, the band structure of bulk BP exhibits non-symmetric conduction and valence bands, the energy-dependent electron velocities and energy-momentum dispersion [1][2].

The advent of high-field terahertz (THz) sources has paved way for the study of conduction electron nonlinear optical responses, such as saturable absorption, in condensed matter systems. The connection between the fundamental properties of material and nonlinear THz propagation could well be evidenced by observation of highly nonequilibrium excited states of electrons induced by the THz field.

Here, we present the investigation of the optical nonlinearity caused by conduction electrons in bulk BP. Experimentally, we observed a high incident THz field dependent optical conductivity derived from an increase in transmission. The TeraFERMI superradiant THz beamline at the FERMI free-electron-laser facility was used for the THz field dependent measurements [3]. A FDTD method together with the model of the ballistic motion of conduction electrons could describe the intense THz pulse-induced transparency observed in the bulk BP in good semi-quantitative agreement with the experimental results [4]. According to the presented model, under the intense THz field, the electrons are accelerated to the highly nonparabolic regions of the conduction band energy of BP in the first Brillouin zone. Due to the anisotropy in the conduction band of BP, the strength and the onset of the observed effect in the armchair and zigzag structural directions are different.

References

[1] D. Correas-Serrano et al, J. Opt. **18**, (2016)
[2] A. Montanaro et al., Nat. Commun. **13** (2022)
[3] N. Adhlakha et al., Condens. Matter **5** (2020)
[4] S. Yu, et al, Phys. Rev. B **95** (2017)

Quantum optical phenomena in 2D material heterostructures

Javier García de Abajo[1,2]

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Atomically thin materials have emerged as a robust platform for manipulating and exploiting light at the nanoscale thanks to a wide variety of polaritonic modes, ranging from plasmons in highly doped graphene to excitions in transition metal dichalcogenides and photons in ionic insulators. The electromagnetic behavior of these modes can be well understood in terms of effective surface conductivities, which in addition can capture their strong dependence on temperature and external static electric and magnetic fields. Recent advances have also been produced in the synthesis of thin noble-metal films [1], which open new possibilities for exploring entirely new regimes of nanometallic plasmonics. In this talk, I will overview the general characteristics of the optical response of these materials, which can be understood in terms of simple theoretical descriptions. We will also cover more sophisticated models, aiming at exploring genuinely quantum-mechanical effects. We will further overview recent advances in the control of the ultrafast optical response and nonlinear optics, as well as the potential application of these materials in light modulation, quantum-optics, and optical sensing. The in/out coupling problem between external light and polaritons of short wavelength remains as a major challenge, for which we will propose innovative solutions. The presentation will conclude with some emerging directions in the design of polaritonic materials, including the intriguing possibility of exploiting quantum phase effects [2].

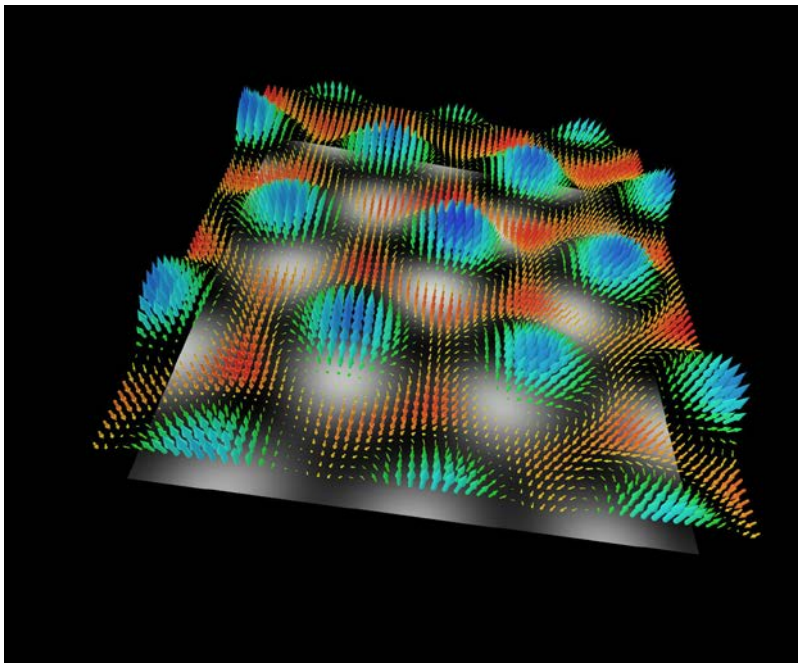
References

[1] V. Mkhitaryan *et al*. (in preparation).
[2] V. Di Giulio *et al*., Nat. Commun. (in press).

Topological plasmonics: Ultrafast vector movies on the nanoscale

Harald Giessen
4th Physics Institute and Research Center SCoPE, University of Stuttgart, Germany

We present an ultrafast vector microscope with 10 nm spatial and subfemtosecond temporal resolution which is capable of mapping all three vector components of the electric field as well as the magnetic field of light on nanophotonic structures. As first application, we record and analyze the temporal evolution of plasmonic skyrmions and the skyrmion number on a nanostructured gold surface.



References

Ultrafast vector imaging of plasmonic skyrmion dynamics with deep subwavelength resolution
T. Davis, D. Janoschka, P. Dreher, B. Frank, F. Meyer zu Heringdorf and H. Giessen
Science **368**, eaba6415 (2020).

Revealing the subfemtosecond dynamics of orbital angular momentum in nanoplasmonic vortices
G. Spektor, D. Kilbane, A. Mahro, B. Frank, S. Ristok, L. Gal, P. Kahl, D. Podbiel, S. Mathias, H. Giessen, F. Meyer zu Heringdorf, M. Orenstein and M. Aeschlimann
Science **355**, 1187 (2017).

The chemistry way towards quasiparticle physics

T. Heine

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Quasiparticles and collective excitations have been in the focus of condensed matter physics of the past decades. Their in-depth exploration has led to a significantly better understanding of the nature of matter in a quite general sense, beyond the boundaries of the materials-oriented disciplines. Following Carlo Beenakker, by taking “what nature offers” and to deeply explore the intriguing physics inherent to known and newly developed materials has led to the discovery and ultimately the deliberate control of many exotic phenomena, including topological insulators, Majorana fermions, skyrmions and polaritons, to name just a few. The impact of this research is not restricted to condensed matter physics or the related emerging field of quantum materials, it indeed drives our very fundamental understanding of matter.
In my talk I will demonstrate a path beyond “what nature offers”, extending the realm of quantum materials to “what is mathematically possible and chemically feasible”. I will show that the proper and precise arrangement of molecular building blocks in regular lattices, linked together by strong bonds with controlled electronic interactions, opens the door to advance electronics and optoelectronics, to manifest exotic physics with Dirac points and flat bands, and to establish new chemical concepts that allow us to face current global challenges. I will bridge our work on two-dimensional semiconductors to the rather novel class of crystalline two-dimensional polymers, a materials class that relies solely on the compounds of organic chemistry, and thus is abundant, non-toxic and potentially bio-compatible.

References

[1] M. A. Springer, T.-J. Liu, A. Kuc, T. Heine, Chem. Soc. Rev. **49**, 2007 (2020). 2007-2019F. Author, Journal **volume**, page (year)

[2] Y. Jing, X. Zhu, S. Maier, T. Heine, Trends in Chemistry, in press (2022).

Strong coupling between molecular vibrations and phonon polaritons in van der Waals materials

R. Hillenbrand[1,2]

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Phonon polaritons - light coupled to optical lattice vibrations - in 2D materials can exhibit ultra-short wavelengths, long lifetimes and strong field confinement, which allows for manipulating infrared light at the nanometer scale. Here, we demonstrate how they can be utilized to achieve vibrational strong coupling with nanoscale amounts of organic molecules. To that end, we employ far-field spectroscopy and real-space nanoimaging of ultra-confined infrared phonon polariton in hexagonal boron nitride (h-BN) nanostructures and layers that are adjacent to molecule layers [1,2].

References

[1] M. Autore et al., Light Sci. Appl. **7**, 17172 (2018)
[2] A. Bylinkin et al., Nat. Photon. **15**, 197 (2021)

Spin-orbit control in graphene-based van der Waals interfaces

C. Kastl[1]

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2 Munich Center for Quantum Science and Technology, München, Germany.

Experimental control of local spin-charge interconversion is of primary interest for spintronics. Van der Waals (vdW) heterostructures enable such functionality by design when combining graphene with a spin-orbit coupled material. Such heterostructures promise high electron mobilities, non-trivial spin texture, and gate-tunability of electronic properties rendering them candidates for all-electrical control of (proximity-induced) spin phenomena.

I will discuss exemplarily interfaces between graphene and Bi_2Te_2Se featuring a lattice-matched, commensurate stacking, where proximity effects have been predicted to impart an anisotropic and electronically tunable spin texture. By polarization-resolved photocurrent measurements, we find a circular photogalvanic effect which is drastically enhanced at the Dirac point of the proximitized graphene. We attribute the gate-tunability to the proximity-induced interfacial spin structure, which could be exploited for, e.g., spin filters [1].
Typically, electric spin valve experiments are employed to read-out such devices in non-local geometries, while leaving the local interplay between the interface symmetry and local charge flow across the heterointerface unexplored. Taking graphene/WTe_2 heterointerfaces as model system, we utilize magneto-optical Kerr microscopy to probe the local, current-induced spin polarisation. Even for a nominal in-plane transport, substantial out-of-plane spin accumulation is induced by a corresponding out-of-plane current flow. We present a theoretical model which fully explains the electric switching and spatial distribution of the Kerr signal as a result of a gate-tunable, non-linear anomalous Hall effect in the heterostructure. Our results highlight the potential of optoelectronic methods for the local read-out of spin-charge interconversion and spin-orbit coupling [2].

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Magnetically correlated defects in a quasi-1D van der Waals magnetic semiconductor

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Van der Waals (vdW) layered magnets are exceptional materials for studying low-dimensional magnetism. [1] A particularly attractive material is the air stable magnetic semiconductor CrSBr [2] with an energy band gap of ~1.6eV and high Néel temperature (132K) with excitons and charge transport correlated with a rich magnetic phase diagram. [3, 4] Atomic-level defects are generally attractive candidates for engineering artificial magnetic textures [5] but remain mostly unexplored.

First, we show that CrSBr is electronically a quasi-1D quantum material in the bulk dominating all quasiparticles and their mutual interactions. [6] Second, we show that CrSBr hosts optically active defects that are correlated with the magnetic phase diagram. [7] The optical emission properties in multilayer CrSBr flakes at low temperature (4K) are excellent, with narrow linewidths (1meV) of both exciton and defect emission owing to the quasi-1D electronic character and the absence of inhomogeneity like strain usually present in supported monolayer material. The optically active defects sense the local magnetic environment and somewhat correlate with a defect-related magnetic phase at low temperature. We discuss potential exchange coupling mechanisms of the defects and their interactions also considering the quasi-1D electronic structure.

CrSBr is a fascinating material that offers experimentally clean and pronounced optical excitations of excitons and defects all correlated with the magnetic phase diagram. This opens several avenues for optically studying tailor-made correlated magnetic phases. [5]

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Exploration of (hybrid) quasiparticles in quantum materials using high-resolution electron energy-loss spectroscopy

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Exploring and fully comprehending the nature of (hybrid) quasiparticles requires their characterization in energy-, momentum-, and real space with highest possible resolution. The transmission electron microscope (TEM) is a very versatile tool for exploring opto-electronic and structural properties of nanostructures down to individual atoms and (point) defects. When equipped with a monochromator, electron energy loss spectroscopy (EELS) in a TEM can resolve excitations of optically bright and dark surface plasmon resonances [1-3], quasiparticle states within a 2D electron gas [4], excitons [5], phonons [6], as well as their interactions [7,8].
After reviewing some recent results of the characterization of quasiparticles by EELS, I will present our observations of the spatially constrained modification of the excitonic spectrum of the hybrid system MoS2/Au. Areas of MoS2 that are covered by small Au islands feature an excitonic peak related to the MoS2/Au interface, likely due to the strong dielectric screening for excitons in MoS2 layer in close contact with the metal [9].

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Cryogenic photocurrent nanoscopy of twisted graphene

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Twisted two-dimensional materials are formed by stacking two layers with a small twist angle. This causes an interference pattern in the atomic lattice called a moiré pattern, which affects the electronic and optical properties dramatically. The first studies on twisted graphene near the “magic angle” of 1.1° revealed strongly correlating states and topological features, making it a host of tunable exotic phases that may shed light on the origins of unconventional superconductivity.

These phenomena motivate us to study the optical properties of twisted materials on the nanoscale. To this end, we use cryogenic (10K) near-field optical microscopy and near-field photocurrent mapping. This technique allows for 20-nm resolution spatial mapping of the local optical properties, collective excitations and Seebeck coefficient. Our observations allow us to relate the spatial variations of correlated electronic states to spatial variations of the twist angle. Moreover, we observe spatial anisotropies, which shed light on the microscopic mechanisms of symmetry breaking.

Communication on the Nanoscale: Strong coupling and Multidimensional Spectroscopy

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The function of essentially all present and future quantum devices, from quantum computers over quantum sensors to photocatalytic systems or solar cells, relies on the transport of energy, charges and spins on ultrafast time (femtoseconds) and exceedingly short length scales (nanometers). Usually, these dynamics are governed by such a complex interplay between electronic and nuclear motion that is challenging to visualize them. Our understanding is therefore quite limited and we often rely on oversimplified, particle-like transport models for describing dynamics and function on the nanoscale.

In my talk, I will introduce and discuss several systems in which this classical, particle-like transport regime breaks down and the wave-like coherent transport of energy and charge becomes dominant, even in disordered nanostructures and at room temperature [1-4]. I will report on an exceptionally powerful, emerging experimental technique, two-dimensional electronic spectroscopy [5], for probing such transport processes. Specifically, I will describe what 2DES can tell us about coherent couplings between excitons and surface plasmon polaritons, the elementary optical excitations of a large class of artificially designed nanostructures comprising metals and semiconductors. By improving the time resolution and sensitivity in 2DES, we resolve coherent energy exchange (strong coupling) and two-particle excitations in such hybrid systems for the first time. The experiments illustrate a conceptually new approach for understanding how nanostructures “talk to each other”.

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Exploring nonlocal electrodynamics: Interfacing metals and 2D materials

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Plasmonic phenomena in metals are commonly explored within the framework of classical electrodynamics and semiclassical models for the interactions of light with free-electron matter. The more detailed understanding of mesoscopic electrodynamics at metal surfaces is, however, becoming increasingly important for both fundamental developments in quantum plasmonics [1] and potential applications in emerging light-based quantum technologies [2]. While this intuitively calls for a full quantum description of plasmon-enhanced light-matter interactions, recent discoveries suggest how classical electrodynamics may still suffice if appropriately dressed by quantum-corrected mesoscopic boundary conditions — surface-response formalism [3, 4, 5, 6]. The talk will address several cases, where mesoscopic electrodynamic effects matter: plasmonemitter interactions [7], electronic surface states in crystalline materials [8], and plasmonpolariton interactions in graphene-on-metal structures [9]. Finally, prospects for probing electrodynamics of correlated electron materials are discussed [10].

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Nanoscale mapping of carrier density in intercalated 2D semiconductors by IR and THz nanoscopy

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Intercalation of 2D semiconductors, such as MoS_2, with molecules is interesting because it drastically changes the electric, optical, and magnetic properties of the host crystal. To observe and understand the intercalation process and its impacts on the material properties, it is crucial to gain knowledge about the molecule distribution in the semiconductor.

Here, we use near field techniques to show that Tetraethylammonium (TEA) molecule intercalation of MoS_2 bulk crystals locally changes the conductivity. The techniques are based on elastic light scattering at an atomic force microscope tip, employing monochromatic laser illumination or broadband illumination from an IR supercontinuum laser. Acting as an optical antenna, the tip converts the illuminating field into a strongly concentrated near field at the very tip apex (nanofocus). Recording of the tip-scattered field as a function of sample position (monochromatic illumination) yield nanoscale-resolved IR/THz images (s-SNOM), while Fourier-transform spectroscopy of the tip-scattered field (broadband illumination) allows for nanoscale IR point spectroscopy (nano-FTIR). In the s-SNOM images we find a drop of the amplitude signals with increasing frequency, and a change of the phase contrast, resembling a Drude-like response, while the pristine MoS_2 shows no changes with frequency. Furthermore, the amplitude and phase images of the intercalated MoS_2 flakes are not homogeneous, indicating a spatial variation of the local conductivity, i.e., the carrier concentration. In addition, we use nano-FTIR to measure the molecular vibrations showing the presence and amount of the TEA molecules.

Generally, our work shows the potential of IR/THz nanoimaging as a noninvasive technique to simultaneously map the carrier concentration together with molecular vibrations, thus allowing for correlating the presence of molecules with the conductivity of the system.

Theory of spatiotemporal carrier dynamics in 2D semiconductors

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Monolayers of transition metal dichalcogenides (TMDCs) are two-dimensional (2D) semiconductors, which can host zero-dimensional (0D) potentials induced for example by strain. When carriers impinge on such localized potential, they can scatter into the bound states of the potential mediated via phonons. Because of the interplay of different dimensionalities and geometries, a non-trivial spatio-temporal dynamics builds up. A crucial aspect is that carrier capture happens only locally, i.e., only when carriers are in the vicinity of the potential [1,4]. The uncaptured density shows a refraction pattern, which can be monitored by a second localized potential in the 2D plane [2]. On the other hand, the captured carriers can display an oscillatory behaviour. Using multiple wave packets these oscillations can be controlled showing linear and even circular movements. These different movements can be mapped onto an electronic Poincare sphere [3].
In this talk, we will review the theoretical methods to describe such spatio-temporal dynamics, which requires a non-diagonal treatment of the density matrix. We start by looking at the dynamics of electronic wave packets impinging on a localized potential. We then extend our study to the dynamics of optically excited excitons and discuss the role of polarons for the capture [4].
Modelling the spatio-temporal behaviour of the carrier dynamics can be helpful for exploring new physics and designing future devices.

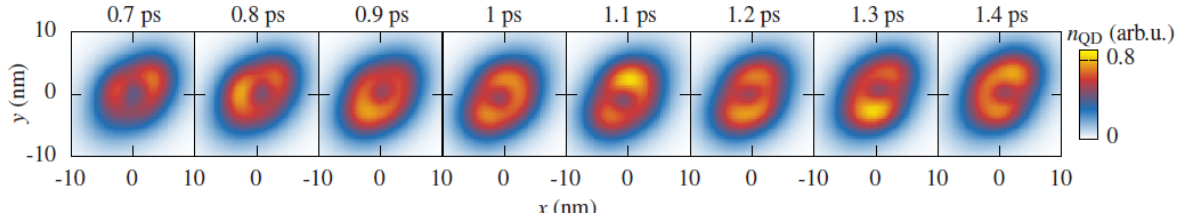

Figure: Snapshots of the dynamics of the captured density inside a 0D potential [1]

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Semiconductors and Metals at the Atomic Limit

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Abstract

The last decade has seen an exponential growth in the science and technology of two-dimensional materials. Beyond graphene, there is a huge variety of layered materials that range in properties from insulating to superconducting that can be grown over large scales for a variety of electronic devices and quantum technologies, such as topological quantum computing, quantum sensing, and neuromorphic computing. In this talk, I will discuss recent breakthroughs in two-dimensional atomic layer synthesis and properties, including understanding how substrate and layer thickness impacts doping of 2D materials to tune them from n- to p-type. Subsequently, I will discuss recent breakthroughs in the realization of unique 2D forms of traditional 3D metals. I will introduce a novel synthesis method, dubbed confinement heteroepitaxy (CHet), that utilizes graphene to enable the creation of atomically thin metals, enabling a new platform for creating artificial quantum lattices with atomically sharp interfaces and designed properties. By shrinking these traditional metals to atomically thin structures, we find that their properties are completely different than their bulk counterparts, lending themselves to unique quantum and optical applications not possible before.

Quasi-electron imaging of quasi-2D materials

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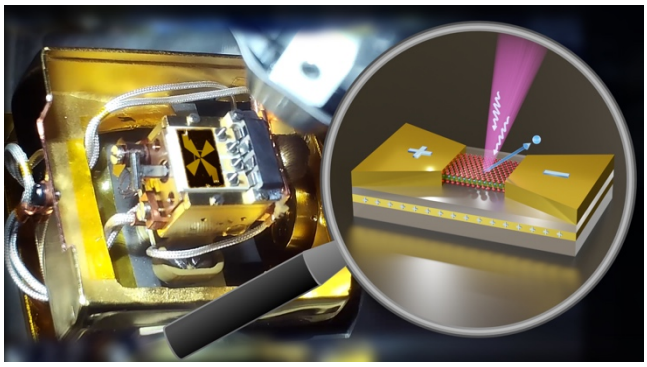
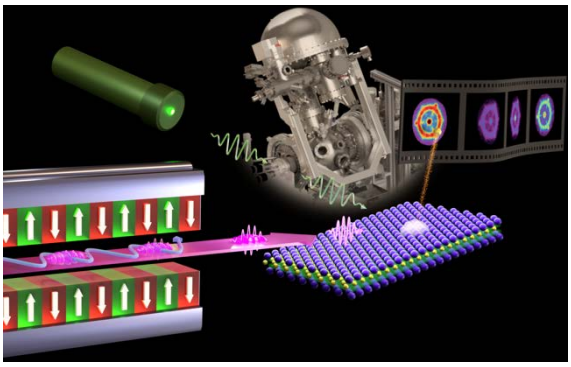
Quasi-electrons are a powerful concept for theoretically describing quantum materials and explaining their electronic properties. Quasi-electrons represent well-defined particle-like elementary excitations in interacting electron systems, whose properties are (very) different from those of bare, noninteracting electrons. Through various interactions with each other and with other elementary excitations, quasi-electrons acquire a finite lifetime and a modified energy spectrum, leading to a broadened and renormalized energy-momentum dispersion relation (as captured by the single-particle spectral function).

The primary experimental tool for measuring quasi-electron band structures is angle-resolved photoelectron spectroscopy (ARPES), which has evolved into a most powerful multidimensional energy-momentum imaging technique due to continuous advances in photon source and electron detector technology. In particular, ARPES has recently been extended with high spatial and temporal resolution using nanofocused and femtosecond-pulsed photon beams in the extreme ultraviolet to soft x-ray range. Intriguingly, these innovative modalities of nano-ARPES and ultrafast ARPES have opened up direct investigations of nonequilibrium quasi-electron dynamics: on the one hand in device-like structures under electrical operating conditions and on the other hand in quantum materials and heterostructures in real time at fundamental time scales.

Here, we give an overview of the current possibilities in nanoscopic and femtostroboscopic quasi-electron imaging using ARPES. Our focus will be on results obtained from quasi-2D materials of the transition-metal dichalcogenide family.





Nonlocal signatures of hybridization between quantum dot and Andreev bound states

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We investigate local and nonlocal signatures of hybridization between a quantum dot state and an extended Andreev bound state (ABS) in a gate-defined InAs nanowire with multiple side probes. When a quantum dot in one of the side probes was hybridized with an ABS in the nanowire, a characteristic spectroscopic pattern was observed both locally, i.e., in the probe with the quantum dot, and nonlocally, in the tunnel conductance of a remote probe. Nonlocal signatures of hybridization reveal the extended nature of the ABS

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Collective Excitations and Degenerate Phases in TMDC Bilayers

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Twisted bilayers of 2D materials such as graphene and TMDCs can host many body physics and correlated phases including superconductors and Mott insulators.[1,3] The moiré potential depending on the twist angle or lattice constant mismatch between two monolayers is exploited to simulate Mott-Hubbard physics.[2] The interfacial electronic band alignment in these systems gives rise to interlayer excitons (IX) where the electron and hole reside in different layers. IX provide a promising platform to study correlated physics such as exciton condensation due to their prolonged lifetimes and large binding energies.[3] We investigate exciton ensembles in a $MoSe_2$ and WSe_2 hetero-bilayer encapsulated in hBN, using photoluminescence (PL) and resonant inelastic light scattering down to millikelvin temperatures. The signatures of collective and degenerate behavior manifest in spectral narrowing and drastically reduced blueshift of the emission energy in dependence of excitation intensity. We further observe highly resonant low-frequency light scattering spectra that indicates coupled collective excitations of the electronic, excitonic and lattice degree of freedom.

We acknowledge financial support via DFG WU 637/4-2, 7-1 and SPP2244.

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Using image-potential states as a quantum probe of atomic-scale spin textures

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Atomic-scale magnetism in ultra-thin metallic films or in moiré structures of 2D-hybrid materials raise expectations for potential spintronic applications, demanding for atomic-scale, spin-sensitive, but yet robust techniques for sensing and manipulation. Image-potential states are unoccupied electronic states in front of polarizable surfaces, serving as a quantum probe for various properties like the surface work function [1] and the electron reflectivity. On magnetic surfaces, they are spin-polarized. They can be locally studied by means of spin-polarized scanning tunnelling microscopy (SP-STM) and spectroscopy. Spin-polarized electrons tunnel resonantly from the magnetic tip via the spin-polarized image-potential states (sp-IPSs) into the surface, resulting in a magnetic image contrast mediated by these states [2].

As I will show, sp-IPSs exhibit the same local spin quantization axis as the spin texture of the underlying sample surface, even when it rotates on the atomic scale [2]. Our SP-STM experiments on non-collinear spin textures in ultra-thin metallic films demonstrate that spin-polarized resonant electron tunneling via sp-IPSs allows for atomic-scale spin-sensitive imaging in real space at tip-sample distances of up to 8 nm, providing a loophole from the hitherto existing dilemma of losing spatial resolution when increasing the tip-sample distance in a scanning probe setup [3]. Experimental results will be discussed in terms of the IPSs spin-splitting as well as in terms of the atomic-scale nature of the resonant tunneling condition. Technically applicable to a variety of material systems, our experiments on iron-intercalated graphene (Gr) on an Ir substrate show that the IPSs are sensitive to the interlayer coupling of Gr to the underlying ferromagnet resulting in their laterally varying spin-polarization thereby serving as probe of the atomic-scale magnetism within the moiré unit cell of the ferromagnet-graphene hybrid system [4].

When the electrons relax from the sp-IPS into the surface, a spin-transfer torque is exerted on the sample that can be exploited for thermally-assisted magnetization switching [5]. Using the IPSs for locally probing and manipulation of magnetism, our approach qualifies for a spin-sensitive read-write technique with ultimate lateral resolution, potentially opening a pathway towards future technical applications.

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2D ferroics explored via scanning tunneling microscopy and spectroscopy

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Ferroic materials are characterized by the emergence of spontaneous polarization, magnetization, or strain below a certain transition temperature, the Curie point. In most materials, the ordering temperature is progressively reduced by reducing the size of the system, a mechanism which can lead to the complete disappearance of ferroic order in low dimensions. This idea has been recently challenged by the discovery of 2D ferroic materials. Not only it has been shown that ferroic order can be preserved once materials are thinned down to their ultimate limit but, even more remarkably, it has been reported that 2D ferroic order can emerge at temperatures significantly higher than those in the respective 3D counterparts. In this talk, I will discuss how the combination of molecular beam epitaxy with scanning probe microscopy allows to create and characterize several 2D ferroic materials and heterostructures. In particular, I will report on the discovery of ferroelectricity stable above room temperature in monolayer SnSe. I will demonstrate how the use of scanning tunneling microscopy and spectroscopy allows to track various ferroic phenomena: polarization direction, temperature dependence, domains evolution, as well as the emergence of complex structures such as ferroelectric vortices. I will subsequently report on the structural, electronic and magnetic properties of 2D magnets. Finally, I will show how 2D magnets can be proximitized to 2D superconductors to create hybrid magnetic-superconducting heterostructures, which I will discuss as possible platforms for engineering unconventional superconductivity.

Nonlinear optics with 2D materials

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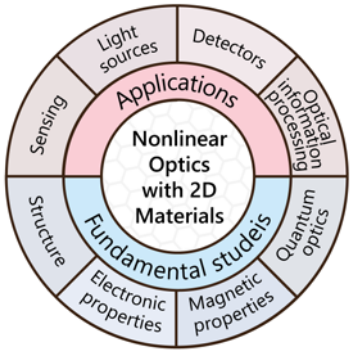

Nonlinear optics is paramount in several fields of science and technology with applications in frequency conversion, entangled photon generation, self-referencing of frequency combs, crystal characterization, sensing, and ultra-short light pulse generation and characterization. In recent years, layered materials and related heterostructures have attracted huge attention in this field, due to their large nonlinear optical susceptibilities, their ease of integration on photonic platforms, and their two-dimensional nature which relaxes the phase-matching constraints and thus offers a practically unlimited bandwidth for parametric nonlinear processes.

In this talk I will discuss some of our recent results in the field of nonlinear optics with two-dimensional materials including strategies for high-speed electrical [1] and all-optical [2] tunable frequency converters, nanoscale lasing [3], nonlinear valleytronics and nonlinear optical integrated devices for gas sensing [4] and logic operations [5].

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Microscopic Theory of Exciton-Exciton Annihilation in Two-Dimensional Semiconductors

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Auger-like exciton-exciton annihilation (EEA) is considered the key fundamental limitation to quantum yield in devices based on excitons in two-dimensional (2d) materials. Since it is challenging to experimentally disentangle EEA from competing processes, guidance of a quantitative theory is highly desirable. The very nature of EEA requires a material-realistic description that is not available to date. In my talk, I will present a many-body theory of EEA based on first-principle band structures and Coulomb interaction matrix elements that goes beyond an effective bosonic picture [1].

The presented approach consistently takes into account all electron-hole correlations on a two-particle level. We apply our theory to quantify EEA coefficients in hBN-encapsulated monolayer MoS_2 and analyze how different Bloch states contribute to the exciton-exciton scattering. We obtain an EEA coefficient on the order of 10^{-3} cm^2s^{-1} at room temperature, suggesting that carrier losses are often dominated by other processes, such as defect-assisted scattering. We also study the influence of temperature and dielectric environmental screening on EEA, finding an inverse temperature dependence that is much stronger than the dependence on substrate dielectric constants. Our studies open a perspective to quantify the efficiency of intrinsic EEA processes in various 2d materials in the focus of modern materials research.

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Interaction of Electron Beams with 2D Materials and Hybrids

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Electron beams have been widely applied to investigate the nanooptical excitations in plasmonic structures. However, only a few studies have explored the possibility to use electron beams for probing strong-coupling effects in hybrid plasmon-exciton systems [1, 2]. Here, I will show that electron beams can probe the so-called self-hybridization effect, which happens due to the strong interaction of the photonic modes of a thin film with excitons. Particularly, I elaborate on the role of the Cherenkov radiation [3], and show how strong-coupling effect can be used to tailor the band structure of plasmonic crystals [4]. Finally, I will discuss our recent experimental results, where the sequential interaction of the electron beams with a structured electron-driven photon source [5] and a thin WSe_2 flake can be used to retrieve the relative intensity and phase of the electron-induced cathodoluminescence scattered to the far field.

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Fast electron spectroscopies to probe optical excitations in 2D materials

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Spectroscopies with focused free electrons beams (kinetic energy 30-300 keV) have the unique benefit of probing “bulk” materials with nanometer or atomic resolution. Among them, three have direct application to nano-optics: electron energy loss/gain spectroscopies (EELS/EEGS) and cathodoluminescence (CL).

In this seminar, I will describe recent advances on the use of EELS and CL to understand the nanometer-scale properties of 2D materials and their heterostructures. These techniques are close counterparts of optical extinction and off-resonance photoluminescence. In h-BN/WS_2/h-BN heterostructures, the measured EELS and CL spectra (Fig. 1) closely resemble those of 1-R-T (R and T the optical reflectivity and transmission). We have used these, coupled to electron imaging, diffraction, and chemical mapping, to unveil these structures behavior under influence of their support (in this case, amorphous carbon), where the charged to neutral exciton emission rate varies significantly.

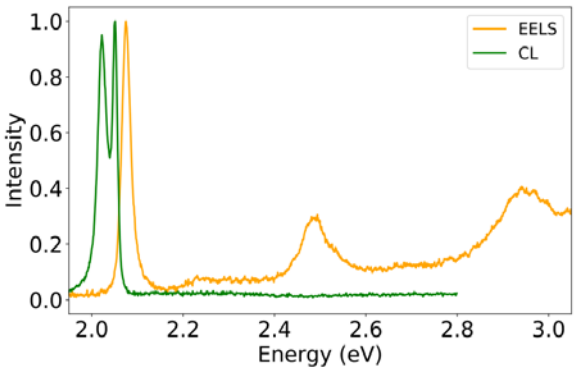

Fig. 1: a EELS and CL spectra of WS_2 encapsulated in h-BN.

h-BN encapsulation is necessary to achieve these materials’ optical response (emission and absorption linewidth) comparable to optical experiments. Surprisingly, experiments with different substrates show that monolayer roughness (as in the case of suspended layers) is not the only or main the source of absorption linewidth increase: trapped charges and surface cleanness also play key roles.

The physics behind TMD monolayer excitation is still unknown. I will discuss how temporal coincidence EELS-CL experiments will help us in unveiling these processes.

Engineering heterostructures in 2-D solids to define new localized quantum states with quantum coherent properties

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We explore how atomically sharp hetero structures such as 2-D stacks, 1-D boundaries within a 2-D material, and 0- D vacancies and substitutes in 2-D materials create new protected states that host quasiparticle excitations using photo low temperature Scanning Tunneling Microscopy, near field optical microscopy, and low temperature time resolved optical spectroscopy.

The concept of quantum coherence is well established in fields such as atomic physics and quantum optics. In solid state systems, quantum coherence plays a critical role defining new phenomena such as coherent quasiparticle transport or coherent emission. However, in the solid state, the meaning of quantum coherence of a quasiparticle excitation is often not straightforward due to undefined decoherence channels, coherence time noise and the difficulty to directly access their associated quantum properties experimentally. While environmental effects are difficult to characterize in 3D systems, coherent quasiparticle transport or emission in 2D solids can be directly and non-destructively analyzed, manipulated experimentally and fully captured with ab-intio methods. Understanding how atomically precise heterogeneities in 2-D solids can create localized and protected states that host quasiparticle excitations and how to control their interaction with their environment is at the core of my research interest.

In the first part of my presentation I will focus on coupled excitonic quasiparticles. We explored the formation and recombination channels of interlayer excitons formed in WS2 / WSE2 stacks and as that may relate to the recently reported formation of excitonic Bose Einstein Condensates. In combination of coupling excitons to plasmonic cavities we map out the local emission of dark excitons with preliminary evidence of measuring their coherent transport. We were also able to show strong coupling between excitons and plasmon polaritons to form a new quasiparticle excitation in for of a plexcitons and report on their transport properties as well. In the second part of my presentation I will focus on next generation quantum emitters in 2-D hosts characterized by photo STM with atomic resolution an ab initio greens function approach simulations. We show how 2-D MoSe2 and 2-D WS2 carry a zoo of intrinsic point defects that modify substantially electronic properties, such as individual S vacancies that create two level systems within the band gap with extremely high spin orbit coupling. These S defects can be artificially induced, and mediate single photon emission via optical stimulation as well as electric stimulation. We show how C-H for S or Se substitutes in 2-D WS2 and 2-D WSe2 form locally charged hydrogen-like states. Upon deprotonation the localized carbon radical hosts a localized deep in gap state with a net spin and electron-phonon coupling that bears strong similarities to NV color centers in diamond, but in this case with atomistic control.

Tuning Carbon Nanotube Emission with Quantum Defects and Cavity-Hybridization

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Polymer-wrapping has enabled the sorting and purification of large amounts of semiconducting and monochiral single-walled carbon nanotubes (SWNTs) that can be applied in a wide range of (opto-)electronic devices with outstanding charge transport properties. Their near-infrared absorption and emission properties (large oscillator strength, narrow linewidth, small Stokes shift and high exciton binding energy) are ideal for strong-light matter coupling, *i.e.*, the hybridization of excitons with cavity photons and thus the formation of exciton-polaritons in Fabry-Pérot cavities [1]. We have demonstrated optically and electrically pumped near-infrared exciton-polaritons in metal-clad microcavities with dense films of (6,5) SWNTs including voltage-dependent tuning between weak, strong and ultrastrong coupling [2,3]. Transient absorption measurements on such strongly-coupled cavities even provide evidence for polariton-mediated biexciton transitions in SWNTs [4].

Furthermore, the controlled covalent functionalization of SWNTs has enabled the creation of specific luminescent sp^3-defects with characteristically red-shifted emission [5]. Mobile excitons are funneled to few defect sites with longer photoluminescence lifetimes leading to higher overall quantum yields. For chiral (6,5) SWNTs the wavelength of the defect emission is governed by the precise binding configuration, which has enabled the controlled creation of even deeper defect states for single-photon emission at room temperature [6]. Such covalently functionalized SWNTs can also be integrated in strongly-coupled microcavities without changing the polariton branch structure. However, radiative pumping via these emissive defects leads to an up to 10-fold increase of the polariton population [7].

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Observation of bright hybridized interlayer exciton in MoTe2/MoSe2 heterobilayers

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Heterostructures of atomically thin transition metal dichalcogenide (TMDC) with strong exciton transitions provide the unique opportunity to rationally tailor the optical properties of solids. Recently, theory [1] and experiment [2] show that in a TMDC heteorobilayer when the conduction band edges of the two constituent layers are near-degeneracy, the electrons could delocalize over both layers, producing new resonant hybridized excitonic states that inherit the spectral properties of both intra- and interlayer excitons. Here we report on the cryogenic optical spectroscopy results of MoTe2/MoSe2 heterobilayers, which are predicted to be a suitable candidate for studying the hybridization effect. We observe a strong interlayer exciton transition with high oscillator strength in aligned MoTe2/MoSe2 samples. This hybridization effect makes aligned MoTe2/MoSe2 heterobilayer have much brighter interlayer exciton emission than the other non-resonant TMDC heterobilayers with much larger band offsets. In addition, g-factors and optical selection rules of these hybridized excitonic states in MoTe2/MoSe2 heterobilayers are discussed as well as doping and twist angle dependence of the degree of hybridization. Our findings pave the way for understanding and engineering the rich exciton physics in TMDC heterostructures and the attractive emission energy (1.1 eV) of the interlayer exciton of MoTe2/MoSe2 provides possibilities for integration with silicon photonics.

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Abstracts of Posters

(in alphabetical order)

Role of a 2D inorganic-organic hybrid semiconductor in controlling the stability of 3D perovskites

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Three-dimensional metal halide perovskites have emerged as popular optoelectronic materials due to their narrow emission linewidth and very high photoluminescence quantum efficiency owing to their high defect tolerance. Perovskite materials have been utilized in photovoltaic applications and demonstrated exceptional efficiency of ~24% over the past few years. But their instability is one of the challenges, because under operating conditions including moisture and heat, they degrade very fast into their inorganic and organic constituents due to the weak hydrogen bonding between organic cation and PbI_2 octahedron [1].

Two-dimensional inorganic-organic (2D IO) hybrid semiconductors have a layered structural arrangement and lead to the formation of multiple quantum well structures with a high exciton binding energy. These 2D IO hybrid semiconductors with weak van der Waals interactions are suitable candidates in enhancing the stability of perovskites. We investigate the laser-induced phase segregation effect and the dynamic evolution of room-temperature photoluminescence (PL) from mixed halide $CH_3NH_3Pb(Br_{0.5}I_{0.5})$ colloidal nanoparticles. These 3D perovskites are surface functionalized with long alkylammonium based 2D IO hybrids, which act as an effective shield and enhance their stability. UV laser-assisted compositional changes are observed in PL when the nanoparticles are surface-functionalized with different halide based long alkylammonium moieties ($C_8H_{17}NH_3X$; X = Cl, Br or I) [2]. Photo-assisted bandgap engineering may find interesting applications in developing advanced optoelectronic devices and creating colorful solar designs.

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Microcavity phonon polaritons: from the weak to the ultrastrong phonon-photon coupling regime

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Strong coupling between molecular vibrations and microcavity modes has been demonstrated to affect physical and chemical properties of the molecular material, for example, inducing changes in chemical reactivity [1] or triggering phase transitions [2]. On the other hand, the coupling between lattice vibrations (phonons) in inorganic materials and microcavity modes has been much less studied, but it may offer intriguing possibilities for fundamental studies and applications. This can be expected because phonons influence various intrinsic properties of materials [3], [4]. The lack of studies might be related to the difficulty to fabricate high-quality thin crystal layers of arbitrary thickness and place them inside the microcavities. Here, we take advantage of the Van der Waals compound hexagonal boron nitride (hBN), in order to obtain layers of any thickness via mechanical exfoliation [5]. In particular, in our work we embed thin layers of hBN into classical microcavities in order to couple its transversal optical (TO) phonon with the first cavity mode. We demonstrate the evolution from weak to ultrastrong phonon-photon coupling by increasing the hBN thickness from a few nanometers to a fully filled cavity. Remarkably, strong coupling is achieved for hBN layers as thin as 10 nm, and ultrastrong coupling for hBN layers thicker than 100 nm. Further, the ultrastrong coupling in fully filled cavities yields a polariton dispersion matching that of phonon polaritons in bulk hBN, highlighting that the maximum light-matter coupling in microcavities is limited to the coupling strength between photons and the bulk material. Tunable cavity phonon polaritons could become a versatile platform for studying how the coupling strength between photons and phonons may modify the properties of polar crystals.

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Orientation Dependent Coupling in Type-II Perylene-TMD Heterostructures

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2D transition metal dichalcogenides (TMDs) are an emerging class of materials that possess a range of novel optical and quantum properties. Band alignment between organic molecules and TMD crystals enables the formation of hybrid heterostructure materials which can host interlayer excitons between the different layers, potentially increasing the excitonic lifetime, making these systems more widely applicable in quantum devices.[1] Critical to the interaction of these materials is the orientation of the molecular orbitals on the TMD surface. Here we present the optical and physical characterisation of perylene bisimide (PBI)-bilayer MoS2 heterostructures at both room and low (4 K) temperatures. We describe the influence of molecular orientation of the PBI on the optical properties of this system, identifying regions of high and low orbital coupling. Analysis of lattice strain by statistical Raman mapping and the charge transfer using photoluminescence measurements gives insight into the role of molecular coupling and hybrid exitonic states in these systems.

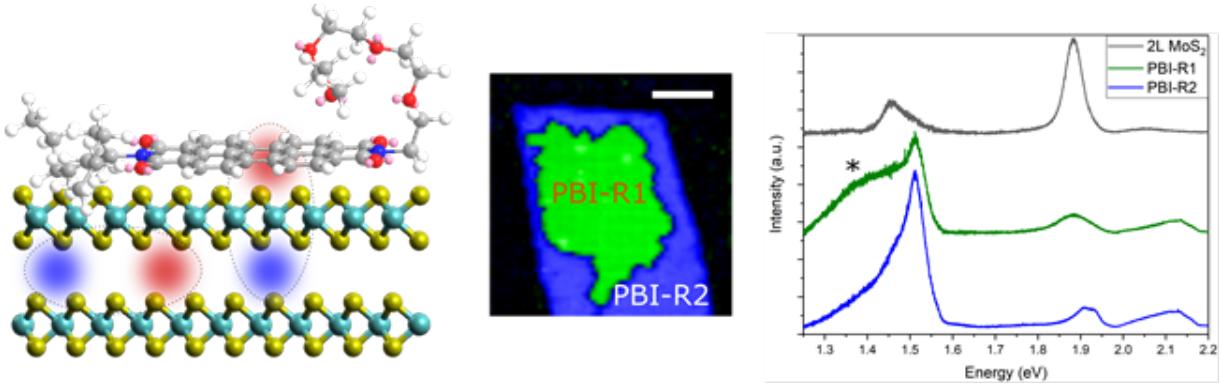

Figure 1: Left – schematic illustrating the presence of both interlayer and intralayer excitons in organic-TMD heterostructures. Centre – Raman map highlighting variations in the crystallographic alignment of the organic PBI molecule on a MoS_2 surface (5 µm scale). Right – Low temperature photoluminescence spectra highlighting the spectral variations between the two regions and an unfunctionalised bilayer (* denotes the energy of the interlayer exciton).

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Probing excitonic population dynamics by nonlinear optical wave mixing in monolayer WSe_2

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Monolayer semiconductors are emerging platforms for strong nonlinear light-matter interaction, due to their giant oscillator strength of tightly bound excitons.

Recently, we reported the existence of a new excitonic species, the high-lying exciton (HX), in monolayer WSe_2. The HX appears at around twice the energy of the band-edge A-exciton, forming a ladder-type excitonic three-level system.

Excitonic quantum interference emerges in second-harmonic generation (SHG) in monolayers[1] and twisted bilayers[2].

Here we apply time-resolved nonlinear spectroscopy to probe the excitonic dynamics in such excitonic three-level system[3]. We find that a significant time difference between two light pulses is necessary for optimal sum-frequency generation (SFG) and four-wave mixing (FWM) when one of the pulses is in resonance with an excitonic transition. The experimental results are explained by numerical calculations based on density-matrix formalism, which provides insights to the coherent exciton dynamics on a femtosecond scale.

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Strong light-field controlled valleytronics

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The fusion between modern quantum material science with the ability to control sub-cycle light fields promises ultrafast light-field electronics, which can potentially operate million times faster than state-of-the-art electronics. Inversion-symmetry broken 2d-quantum materials, e.g., monolayer hexagonal boron nitride (hBN) or transition metal dichalcogenides (TMDs), possess valley degrees of freedom associated with the local minima in the band structure [1]. Their A) selective excitation, B) controlled inter-valley switching, and C) readout of the excited quantum states are the prerequisites for their use in information processing. Attempts to control valley polarization so far relied on resonant excitation [2], along with valley switching with strong terahertz fields [3].

Here, following the recent theoretical prediction [4], we demonstrate all-optical nonresonant strong field control of all the three major steps (A-C). We used two-color bi-circular ultrashort laser fields to control the selective valley excitation in hBN and then probe the quantum valley states via third harmonic generation ellipsometry. The non-resonant pump field modifies the topological properties of the band structure by inducing Halden-type [5] complex next nearest neighbor hopping, which manifests the breaking of time-reversal symmetry. This hopping interaction controls the valley asymmetry and can be manipulated by varying the phase delay between the two colors of the bi-circular field. In agreement with the theoretical predictions, the experimental data shows selective enhancement of valley population as a function of the two-color phase delay. These results demonstrate a new regime of valleytronics and lightinduced topology in quantum materials

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Tailoring Plasmon-Exciton Interactions in Nanometer-Thick Gold−WSe_2 Hybrid Structure using Cathodoluminescence Spectroscopy

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Transition-metal dichalcogenides (TMDCs) with their exciton-dominated optical behavior emerge as promising materials for realizing strong light−matter interactions and exciton polaritons (EP) in the visible range and at ambient conditions. Recently, optical micro-spectroscopy and scanning near-field optical microscopy have been employed to investigate thin TMDC waveguides in near-field region [1]. However, because of missing high spatial resolution of spectroscopic investigation at broad energy, the underlying coupling mechanisms remain largely unclear. Moreover, combining TMDCs such as WSe_2 with structured metals such as a gold plasmonic lattice and the energy confining ability of plasmon polaritons in metals below the diffraction limit allows for further enhancing and tailoring the interactions between EPs, photons, as well as plasmonic Bloch modes (PBMs) [2, 3]. Herein, through the extensive analysis of numerical investigations and cathodoluminescence spectroscopy (CL) measurements, we demonstrate the progressive enhancement in interactions from strong exciton–photon couplings in free-standing WSe_2 flakes due to the self-hybridization effect up to ultra-strong coupling between the excitons of WSe_2 flakes and PBMs supported by thin gold films including a square array of holes [4, 5]. The CL hyperspectral images of the multilayer structure, demonstrate ultra-strong interactions between excitons and PBMs and the emergence of plexcitonic photonic flat bands that can be further controlled via the thickness of the WSe_2 flakes. The energy splitting and the formation of two distinct band gaps as the consequence of mode repulsion between the oscillators, i.e., plasmons and excitons in plexcitonic structures are promising for the development of new applications including ultrasensitive sensors and nonlinear optics.

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EuPRAXIA@SPARC_LAB FELs: Brilliant Soft X-Ray Pulse Sources for Spectroscopy and Imaging

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Soft x-ray free electron laser (FEL) technology allows ultrafast electrical and structural dynamic investigation of various materials, interacting linearly and nonlinearly with femtosecond and high-flux x-ray pulses. Soft x-ray FEL technological advancements are leading to more compact, efficient, and high-quality sources, resulting in significant innovation. In this manner, and as part of the leading EuPRAXIA project, EuPRAXIA@SPARC_LAB intends to develop a unique high-gradient plasma accelerated first fifth-generation free electron laser (FEL) source facility that is cost-effective in terms of both size and expense for the entire facility [1]. Two beamlines will deliver ultra-bright photon pulses for experiments to the user community. One is a seeded FEL named ARIA in the VUV range (50-180 nm) [3], while the other one is a SASE soft x-ray FEL in the water window (4-6 nm) named AQUA. [2]. This poster presents an introduction to the under-construction facility and gives an overview of the foreseen applications of these photon sources, which include different methods of imaging and spectroscopy. They can provide information about the structure and dynamical behavior of a variety of samples, including biological samples, soft matters such as polymers and inorganic materials. for instance, the electronic state of excited molecules in the vicinity of conical intersections can be visualized.

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The Fabrication of nanostructured van der Waals Heterostructures using He Ion Beam Patterning

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Polaritons in two-dimensional materials exhibit enhanced light-matter interactions, which makes them interesting for low-loss, highly confined light transport. A polariton is a quasiparticle that combines a photon with a dipole-carrying excitation in matter and is strongly dependent on the type and geometry of the material. The hybridization of polaritonic modes in different 2D materials may provide strong localization of plasmonic excitations with long propagation distances of phonon modes [1]. By modifying the geometry of van der Waals (vdW) heterostructures at the nanoscale, we tune hybrid polaritonic modes.

We fabricate and patterning of heterostructures based on single crystalline gold or silver flakes, graphene, and hexagonal boron nitride (hBN). For dry transfer we used polydimethylsiloxane (PDMS) and poly(propylene) carbonate (PPC) films due to their strong adhesion to 2D materials at room temperature. Therewith, single-layer to few-layer 2D materials were successfully transferred onto thin electron transparent membranes of silicon nitride.

To modify the geometry of the heterostructures at the nanoscale, a Zeiss Orion Nanofab microscope is then used for patterning by He and/or Ne ion beam milling. As polaritonic modes are not only strongly influenced by geometry, but also by material quality, an important step in the study is therefore to investigate different currents, acceleration voltages, and ion types to determine what damaging effects they have on the crystalline lattice and the corresponding m material response. The optimization of the patterning routines is carried out with the help of FIB-o-Mat, which provides complete control over the beam path [2].

In the following step, monochromated, low-loss scanning transmission electron microscopy (STEM), electron energy-loss spectroscopy (EELS) [3] is used to map the optical properties of the fabricated heterostructures, and the results are compared to near-field optical methods.

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Observation and Dynamics of Interlayer Excitons in Perylene Bisimide-MoS2 Heterostructures

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Transition metal dichalcogenides (TMDs) are materials being investigated for next generation semiconductor devices in various fields due to the relative ease of tuning the electronical and optical properties of these materials by various methods. By exploiting the noncovalent functionalization of MoS_2 with perylene derivates, organic-inorganic heterostructures can be formed[1] which can exhibit hybrid excitonic states between the two materials, similar to those seen in TMD-TMD heterostacks.[2]

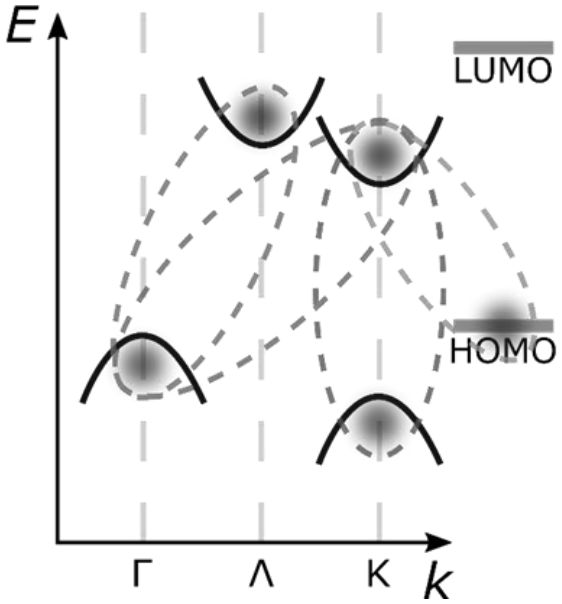

Figure 1: Schematic energy levels of PBI-functionalized bilayer MoS_2 showing type-II alignment of the heterostructure and the position of the interlayer exciton

In our study, low temperature photoluminescence (PL) measurements provide insights into the interlayer charge transfer between bilayer MoS_2 and perylene bisimide (PBI). The observation of interlayer excitons between the two materials, through power-dependent PL studies, confirms type-II band alignment of the heterostack indicating hole transfer from MoS_2 valence band into PBI HOMO after photoexcitation. This charge transfer is shown to be strongly dependent on orbital alignment of the PBI molecule on the MoS_2 surface resulting in saturation of charge transfer in weakly coupled regions.

Our work provides insights into the charge carrier dynamics of organic-inorganic heterostructures, with particular relevance to applications for optoelectronic and excitonic devices.

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Light matter interaction and quantum confinement in 2D polar metals

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Important for all (quantum) optical technologies is the manipulation of the light-matter interaction to achieve a high level of control, particularly in technologically relevant solid-state nanomaterials. Atomically thin two-dimensional layered materials receive great interest because of their unique properties. A novel class of atomically thin materials such as 2D polar metals such as 2D gallium or 2D indium and their ternary alloys that exhibit fascinating properties like superconductivity [1,2] strong nonlinear optical properties emerging by giant second harmonic generation [3] and epsilon near zero behavior in the visible and NIR range [4]. The layer dependence of the energies of localized plasmons due to interband transition strongly suggest quantum confined 2D metal films.

In this paper, we will introduce the rather new class of 2D materials, 2D polar metals and focus on their linear optical response measured by imaging spectroscopic ellipsometry in dependence of the temperature covering room temperature to 800mK. At low temperatures emergent behavior such as superconductivity occurs such that we are able to monitor the dielectric functions across the phase transitions. Interestingly, while 2D Ga turns into a superconductor at low temperatures and 2D In remains metallic, 2D Ag turns into a semiconductor in the monolayer limit [5] that is reflected in its altered light mater interaction described by the dielectric functions [6].

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Magnetically correlated defects in a quasi-1D van der Waals magnetic semiconductor

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Van der Waals (vdW) layered magnets are exceptional materials for studying low-dimensional magnetism. [1] A particularly attractive material is the air stable magnetic semiconductor CrSBr [2] with an energy band gap of ~1.6eV and high Néel temperature (132K) with excitons and charge transport correlated with a rich magnetic phase diagram. [3, 4] Atomic-level defects are generally attractive candidates for engineering artificial magnetic textures [5] but remain mostly unexplored.

First, we show that CrSBr is electronically a quasi-1D quantum material in the bulk dominating all quasiparticles and their mutual interactions. [6] Second, we show that CrSBr hosts optically active defects that are correlated with the magnetic phase diagram. [7] The optical emission properties in multilayer CrSBr flakes at low temperature (4K) are excellent, with narrow linewidths (1meV) of both exciton and defect emission owing to the quasi-1D electronic character and the absence of inhomogeneity like strain usually present in supported monolayer material. The optically active defects sense the local magnetic environment and somewhat correlate with a defect-related magnetic phase at low temperature. We discuss potential exchange coupling mechanisms of the defects and their interactions also considering the quasi-1D electronic structure.

CrSBr is a fascinating material that offers experimentally clean and pronounced optical excitations of excitons and defects all correlated with the magnetic phase diagram. This opens several avenues for optically studying tailor-made correlated magnetic phases. [5]

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All-optical polarization and amplitude modulation of second harmonic generation in atomically thin semiconductors

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Nonlinear optics is of paramount importance in several fields of science and technology. This is particularly true in the case of second harmonic generation (SHG), which is commonly used for frequency conversion, self-referencing of frequency combs, crystal characterization, sensing, and ultra-short pulse characterization. Large efforts have been devoted in the last years to realizing electrical and all-optical modulation of SHG in two-dimensional (2D) materials, which are ideal for novel nonlinear integrated devices. They provide in this respect distinct advantages thanks to their ease of integration on photonic platforms [1] and their atomically thin nature, which relaxes phase matching constraints and thus offers a practically unlimited bandwidth for nonlinear optical effects [2]. In this work, we propose a new approach to broadband all-optical modulation of SHG in 2D materials. Our concept is based only on symmetry considerations and thus it is applicable to any material of the D_{3h} symmetry group and with deep sub-wavelength thickness, such as all monolayer transition metal dichalcogenides. With this approach we demonstrate a 90° rotation of the polarization of the emitted SHG on a time-scale limited only by the fundamental pulse duration [3]. In addition, this ultrafast polarization switch can be immediately applied to realize all-optical SH amplitude modulation with depth of close to 100%. Our results outperform any previous work on all-optical SHG modulation [4,5] in terms of modulation speed, modulation depth and SHG bandwidth.

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Enhancing photoluminescence and Raman signals in TMDC monolayers via plasmonic nanostructures

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Novel 2D thin film materials garnered a great interest in the recent years. The family of the Transition metal dichalcogenide (TMDCs) among those materials are especially appealing because of their indirect-direct bandgap transition and the possibility to stack different composites of the TMDCs on top of each other to achieve so called heterostructures.

Herein, we present a technique to define nanostructures on a SiO_2 wafer using thermal scanning probe lithography. Further, we deposit individual TMDC monolayer flakes on top of the metallized structures via the deterministic transfer technique. We characterize our flakes utilizing photoluminescence and Raman measurements, reporting major enhancements effects (~20x) in PL and minor enhancement (~3x) in Raman signals.

(a) (b)

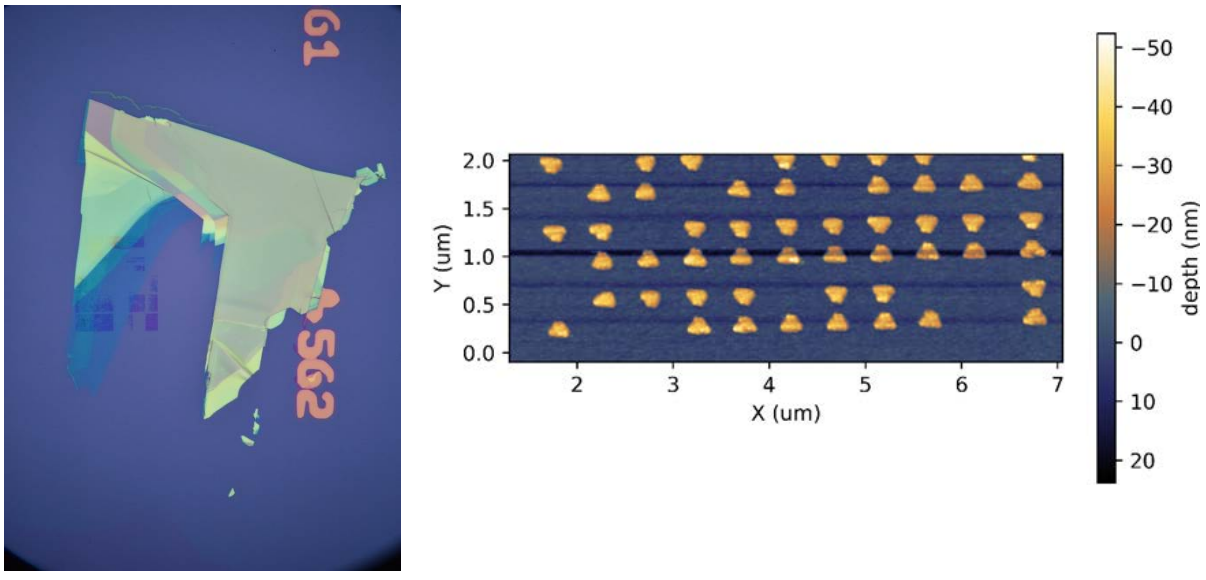

Figure 1. (a) MoS_2 flake with monolayer and bilayer on top of nanostructures (b) Thermal scanning probe lithography image of the gold nanostructures

Nanoscale mapping of carrier density in intercalated 2D semiconductors by IR and THz nanoscopy

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Intercalation of 2D semiconductors, such as MoS_2, with molecules is interesting because it drastically changes the electric, optical, and magnetic properties of the host crystal. To observe and understand the intercalation process and its impacts on the material properties, it is crucial to gain knowledge about the molecule distribution in the semiconductor.

Here, we use near field techniques to show that Tetraethylammonium (TEA) molecule intercalation of MoS_2 bulk crystals locally changes the conductivity. The techniques are based on elastic light scattering at an atomic force microscope tip, employing monochromatic laser illumination or broadband illumination from an IR supercontinuum laser. Acting as an optical antenna, the tip converts the illuminating field into a strongly concentrated near field at the very tip apex (nanofocus). Recording of the tip-scattered field as a function of sample position (monochromatic illumination) yield nanoscale-resolved IR/THz images (s-SNOM), while Fourier-transform spectroscopy of the tip-scattered field (broadband illumination) allows for nanoscale IR point spectroscopy (nano-FTIR). In the s-SNOM images we find a drop of the amplitude signals with increasing frequency, and a change of the phase contrast, resembling a Drude-like response, while the pristine MoS_2 shows no changes with frequency. Furthermore, the amplitude and phase images of the intercalated MoS_2 flakes are not homogeneous, indicating a spatial variation of the local conductivity, i.e., the carrier concentration. In addition, we use nano-FTIR to measure the molecular vibrations showing the presence and amount of the TEA molecules.

Generally, our work shows the potential of IR/THz nanoimaging as a noninvasive technique to simultaneously map the carrier concentration together with molecular vibrations, thus allowing for correlating the presence of molecules with the conductivity of the system.

Optically detected ionic gate spectroscopy and exciton manifolds in highly doped WS_2

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Semiconducting two-dimensional transition metal dichalcogenides such as WS_2 excel as due to their exciton dominated light-matter interaction even at room temperature (RT) that is highly tunable by external stimuli such as, dielectric environment, strain or doping [1]. The disentanglement of single and many particle properties in these 2D semiconductors as well as their dependencies on high carrier concentration is challenging to experimentally study by pure optical means. In this work, we demonstrate optical signatures for Fermi-edge singularities in optical emission measurements when the Fermi energy is aligned with conduction band or valence band edges in electrolyte gated measurements. This enables capacitance spectroscopy to get direct access to the single particle band gap E_{gap} and to simultaneously estimate the exciton binding energies of weakly doped WS_2 monolayers [2]. Combined imaging spectroscopic ellipsometry and photoluminescence spectroscopies spanning large n- and p- type doping with charge carrier densities up to 1×10^{14} cm^{-2} allows to study screening phenomena and doping-dependent evolution of the rich exciton manifold. We show that the two most prominent emission bands in photoluminescence experiments are due to the recombination of spin forbidden and momentum forbidden charge neutral excitons activated by phonons. The observed interband transitions are redshifted and drastically weakened under electron and hole doping.

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Signatures for localized and remote trions in $MoSe_2$ / WS_2 heterostructures

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Transition Metal Dichalcogenides (TMDs), a family of layered van der Waals (vdW) materials, have attracted growing interest over the last decade. In their monolayer limit TMDs present direct semiconductors with bandgaps in the visible range, and their large effective electron and hole masses allow for strongly bound excitons that dominate their coupling to light all the way up to room temperature. By vertically stacking multiple monolayers into vdW-heterostructures one can engineer their physical properties through parameters such as the choice of materials, the rotation angle between the different layers or their dielectric environment, turning TMDs into a promising platform for fundamental experiments in solid state physics. In this work we present spectroscopic data on the TMD heterobilayer $MoSe_2$/WS_2, a structure with closely aligned conduction band edges and an emergent Moiré superlattice due to the lattice constant mismatch between the two monolayers. We use low temperature Photoluminescence (PL) and Reflection Contrast (DR) measurements at varying doping levels and out-of-plane magnetic fields to map out the band structure of this structure, and provide evidence for localization of excitons that report on strongly correlated states in both electron and hole doping regimes.

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Nonlinear plasmonic response to circularly polarized light

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Varying polarization and orbital momentum of incident light offers unprecedented opportunities to explore light-matter interactions. This holds particularly true in nanoplasmonics, where optical fields can be controlled at the nanoscale, leading to chiral and nonlinear effects in unprecedentedly small volumes, thus triggering a fundamental and practical interest on the topic. Despite many works have tackled the linear response of plasmonic structures, the fundamental quantum-mechanical analysis of the nonlinear response induced by a circularly polarized electromagnetic field has not been attempted so far.

Here we use time-dependent density functional theory (TDDFT) to quantum-mechanically address the nonlinear polarization and nonlinear near fields induced in an infinite metallic nanowire by a circularly polarized incident electromagnetic pulse. The electric field of the pulse rotates within the 2D (x,y) plane perpendicular to the nanowire z-axis. Retardation effects can be neglected as the size of the nanoparticle is much smaller that the optical wavelength. From our calculations, we reveal the dominance of the surface non-linearity that results in an induced charge density at the n-th harmonic characterized by the excitation of a multipole surface charge density of order n rotating along the nanowire axis at the fundamental driving frequency. We further discuss the role of the surface and bulk nonlinearities, as well as the polarization of the associated far- and near-fields. We believe that our results shed light to important aspects of the nonlinear response in plasmonic nanostructures triggered by circularly polarized light.

Planar Josephson Junctions under Microwave Irradiation

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An understanding of the response of planar JJs to microwave irradiation is crucial in the context of ABS control and engineering. We investigate the response of planar Josephson junctions (JJs) in an InAs/Al heterostructure [1] under microwave irradiation. We perform spectroscopy of Andreev bound states (ABSs) using a superconducting probe with a gate-tunable tunnelling barrier. On increasing the power of the microwave field, we observe replicas of conductance features separated by the photon energy of the microwave field. This observation was recently interpreted as evidence of Floquet-Andreev modes in the junction [2]. By tuning the tunnelling barrier, we show that, in our devices, photon assisted tunnelling across the barrier [3] is the dominant cause of ABS replication.
An asymmetric SQUID geometry allows to measure the current-phase relation of the JJ without the influence of the probe. The response to microwave irradiation can be well described by an adiabatic theory up to intermediate powers [4]. Deviation at large powers is consistent with a non-thermal distribution of ABS occupation [5], without invoking a Floquet-Andreev model.

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Collective Excitations and Degenerate Phases in TMDC Bilayers

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Twisted bilayers of 2D materials such as graphene and TMDCs can host many body physics and correlated phases including superconductors and Mott insulators.[1,3] The moiré potential depending on the twist angle or lattice constant mismatch between two monolayers is exploited to simulate Mott-Hubbard physics.[2] The interfacial electronic band alignment in these systems gives rise to interlayer excitons (IX) where the electron and hole reside in different layers. IX provide a promising platform to study correlated physics such as exciton condensation due to their prolonged lifetimes and large binding energies.[3] We investigate exciton ensembles in a $MoSe_2$ and WSe_2 hetero-bilayer encapsulated in hBN, using photoluminescence (PL) and resonant inelastic light scattering down to millikelvin temperatures. The signatures of collective and degenerate behavior manifest in spectral narrowing and drastically reduced blueshift of the emission energy in dependence of excitation intensity. We further observe highly resonant low-frequency light scattering spectra that indicates coupled collective excitations of the electronic, excitonic and lattice degree of freedom.

We acknowledge financial support via DFG WU 637/4-2, 7-1 and SPP2244.

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Valley spin-acoustic resonance in monolayer MoS_2

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Monolayer MoS_2 is a strong candidate material to study spin-valley coupled physics in 2D materials. Its unique band structure comprises of spin-split subbands crossing each other at finite momenta with opposite spin orientation in both the valleys. When exposed to Rayleigh surface acoustic waves, strain induced pseudomagnetic fields couple with spin resulting in spin-phonon interaction. We theoretically predict the occurrence of spin acoustic resonance accompanied by acoustoelectric current due to spin-flip transitions between the spin-split subbands. We calculate the transition probabilities, obtain the conditions for observing spin acoustic resonance and calculate and study the behavior of the acoustoelectric current. On breaking time reversal symmetry, both the spin acoustic resonance and acoustoelectric current becomes valley sensitive paving way for acousto-electric spectroscopy of valley selective phenomena.

Observation of bright hybridized interlayer exciton in MoTe2/MoSe2 heterobilayers

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Heterostructures of atomically thin transition metal dichalcogenide (TMDC) with strong exciton transitions provide the unique opportunity to rationally tailor the optical properties of solids. Recently, theory [1] and experiment [2] show that in a TMDC heteorobilayer when the conduction band edges of the two constituent layers are near-degeneracy, the electrons could delocalize over both layers, producing new resonant hybridized excitonic states that inherit the spectral properties of both intra- and interlayer excitons. Here we report on the cryogenic optical spectroscopy results of MoTe2/MoSe2 heterobilayers, which are predicted to be a suitable candidate for studying the hybridization effect. We observe a strong interlayer exciton transition with high oscillator strength in aligned MoTe2/MoSe2 samples. This hybridization effect makes aligned MoTe2/MoSe2 heterobilayer have much brighter interlayer exciton emission than the other non-resonant TMDC heterobilayers with much larger band offsets. In addition, g-factors and optical selection rules of these hybridized excitonic states in MoTe2/MoSe2 heterobilayers are discussed as well as doping and twist angle dependence of the degree of hybridization. Our findings pave the way for understanding and engineering the rich exciton physics in TMDC heterostructures and the attractive emission energy (1.1 eV) of the interlayer exciton of MoTe2/MoSe2 provides possibilities for integration with silicon photonics.

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