HL 2: 2D Semiconductors and van der Waals Heterostructures I

The session covers the photonic properties and corresponding devices made from 2D semiconductors and van der Waals heterostructures.

Time: Monday 9:30–13:00 Location: H15

HL 2.1 Mon 9:30 H15

Probing strong electron-phonon coupling in graphene by resonance Raman spectroscopy with infrared excitation energy — •Simone Sotgiu[1,2], Tommaso Venanzi[1], Lorenzo Graziotto[1], Francesco Macheda[1], Taoufiq Ouaj[2], Elena Stellino[1], Bernd Beschoten[2], Christoph Stampfer[2], Francesco Mauri[1], and Leonetta Baldassarre[1] — [1]Department of Physics, Sapienza University of Rome, Rome, Italy — [2]JARA-FIT and 2nd Institute of Physics, RWTH Aachen University, Aachen, Germany

Resonance Raman spectroscopy (RRS) has been a key asset to study the interplay between electronic and vibrational properties of graphene. We report on RRS measurements with an excitation photon energy down to 1.17 eV on mono (MLG) and bilayer (BLG) graphene, to study how low-energy carriers interact with lattice vibrations. Thanks to the excitation energy close to the Dirac point, we unveil in the MLG a giant increase of the intensity ratio between the double-resonant 2D and 2D* Raman peaks with respect to graphite [1]. In BLG, the low excitation energy hampers some of the resonant Raman processes giving rise to the 2D peak. Consequently, the subfeatures composing the 2D mode are spectrally more separated with respect to visible excitations. We compare experimental measurements on BLG with ab initio theoretical calculations and we trace back such modifications on the joint effects of probing the electronic dispersion close to the band splitting and enhancement of electron-phonon matrix elements [2]. [1] T. Venanzi et al., Phys. Rev. Lett. 2023, 130, 256901 [2] L. Graziotto et al., Nano Lett. 2024, 24, 1867

HL 2.2 Mon 9:45 H15

Nonlinear Probing of Ultrafast Bandgap Modulations in Atomically Thin Semiconductors — •Sebastian Klimmer[1,2], Thomas Lettau[3], Jan Wilhelm[4,5], Dragomir Neshev[2], and Giancarlo Soavi[1] — [1]Institute of Solid State Physics, Friedrich Schiller University Jena, Jena, Germany — [2]ARC Centre of Excellence for Transformative Meta-Optical Systems, Department of Electronic Materials Engineering, Research School of Physics, The Australian National University, Canberra, Australia — [3]Institute of Condensed Matter Theory and Optics, Friedrich Schiller University Jena, Jena, Germany — [4]Regensburg Center for Ultrafast Nanoscopy (RUN), University of Regensburg, Regensburg, Germany — [5]Institute of Theoretical Physics, University of Regensburg, Regensburg, Germany

Bandgap modulations are pivotal for semiconductor based applications, typically achieved by permanent modifications of material composition or size. Recently, dynamic, ultrafast bandgap modulations have been demonstrated in transition metal dichalcogenides, exploiting the valley exclusive optical Stark (OS) and Bloch-Siegert (BS) shifts. However, their detection requires state of the art two-color pump-probe schemes. Here, we show a simplified method by measuring the second-harmonic (SH) power dependence in monolayer WSe_2. The fundamental beam (FB) blue-shifts the bandgap by OS/BS shifts, which scale linearly with intensity, resulting in a enhanced (reduced) SH signal, depending on the detuning $E_g - 2\hbar\omega_{FB}$. Our experimental data are fully explained by an analytical SBE model, which allows us to extract values for the transition dipole element and dephasing time.

HL 2.3 Mon 10:00 H15

Detecting out-of-plane polarized luminescence in TMDs with laser-written waveguide circuits — •Alina Schubert[1], Karo Becker[1], Rico Schwartz[1], Andreas Thies[2], Alexander Szameit[1], Matthias Heinrich[1], and Tobias Korn[1] — [1]Institut für Physik, Universität Rostock, Rostock, Germany — [2]2Ferdinand Braun Institut, Leibnitz Institut für Höchstfrequenztechnik, Berlin, Germany

Photoluminescence (PL) spectroscopy is used widely to investigate the outstanding excitonic properties of two-dimensional crystals. However, since these measurements are typically performed in vertical incidence, the sample is excited with light polarized in the plane of its layer. Despite, these detection schemes systematically neglect the out-of-plane polarized components of the emitted signal that propagate along the layer and therefore are only accessible by side-on detection.

Here, a side-on detection is achieved by using femtosecond laser direct written waveguides in fused silica glass as substrates [1]. By inscribing the waveguides directly under the surface of the glass, interactions of the waveguide's evanescent field and the sample are enabled in a controlled fashion. Therefore, the sample can be excited due to its proximity to the waveguide, while, in turn, the excitonic emission can be coupled into the waveguide and subsequently be detected.

In this work, the in-plane propagating luminescence in TMDs is analyzed with a particular focus on determining influences on the signal resulting from combining TMDs and surface waveguides.

[1] A. Szameit et al., J. Phys. B.: At. Mol. Opt. Phys., 43 (2010).

HL 2.4 Mon 10:15 H15

Current-induced second-harmonic generation in monolayer graphene-devices — •Nele Tornow[1], Friederike Renz-Wieland[1], Omid Ghaebi[1], and Giancarlo Soavi[1,2] — [1]Institute of Solid State Physics, Friedrich Schiller University Jena, Jena, Germany — [2]Abbe Center of Photonics, Friedrich Schiller University Jena, Jena, Germany

In pristine and isolated graphene, second-order nonlinear optical processes are, within the dipole approximation, forbidden due to the invariance under space inversion symmetry. Few studies have suggested that inversion symmetry can be broken by in-plane currents, leading to a measurable second-harmonic generation (SHG) [1,2]. In particular, investigations have shown that interfacial charge trapping between a monolayer graphene and silicon dioxide (SiO_2) on silicon substrate results in electrically tunable SHG when driving in-plane currents [2]. To further verify this hypothesis, we study current-tunable second-harmonic emission in two graphene based devices: one high-quality double-encapsulated hexagonal boron nitride/graphene field-effect transistor and one with a graphene-SiO_2 interface, finding SHG only in the latter one.

Our findings provide insights into the origin of current-tunable SHG and the mechanisms underlying space inversion symmetry breaking in graphene.

[1] Dean, J. et al., Appl. Phys. Lett. 26, 261910 (2009)
[2] An, Y. et al., Nano Lett. 13, 2104-2109 (2013)

HL 2.5 Mon 10:30 H15

Electron-phonon coupling across the TMD/hBN van der Waals interface — •Gianmarco Gatti[1,2], Christophe Berthod[3], Julia Issing[2], Michael Straub[2], Salony Mandloi[2], Yann Alexaniann[2], Anna Tamai[2], and Felix Baumberger[2] — [1]Department of Physics and Astronomy, Aarhus University, Aarhus, Denmark — [2]Department of Quantum Matter Physics, University of Geneva, Geneva, Switzerland — [3]Department of Theoretical Physics, University of Geneva, Geneva, Switzerland

The lattice mismatch at van der Waals interfaces is widely used to engineer single particle band structures supporting complex quantum phases in two dimensions. Electronic states in van der Waals heterostructures may also couple to bosonic excitations in an adjacent substrate. These interfacial interactions are difficult to identify, and it is commonly assumed that they play a marginal role. Here, we reveal that quasiparticles in monolayer transition metal dichalcogenides (TMDs) are dressed by a remote cloud of phonons in the adjacent hexagonal boron nitride slab. Using angle resolved photoemission, we identify replica bands in the TMDs which are a clear fingerprint of a long-range electron-phonon interaction. We develop a modified Fröhlich model that shows quantitative agreement with the experimental spectral functions and discuss the properties of this model. Our analysis shows that remote electron-phonon coupling is a generic property of interfaces with hBN. This has implications for electron mobilities in 2D materials, for superconductivity and possibly for moiré correlated phases.

HL 2.6 Mon 10:45 H15

Polaron spectroscopy of many-body systems — •Ivan Amelio — Université Libre de Bruxelles, Brussels, Belgium

When an impurity is immersed in a many-body background, it is

dressed by the excitations of the bath, and forms "a polaron".

As a result, the injection spectrum of the impurity carries the hallmarks of the correlations present in the bath. This physics is relevant for excitons optically injected in a few layer heterostructure, or for cold atomic mixtures.

In this talk, we will first review the basic theoretical framework and recent experimental progress.

Then, we will theoretically analyze a few cases of correlated many-body states: the impurity injection spectra are predicted to display peculiar features, that allow to distinguish whether the bath features BCS pairing, charge density waves, topological phases, the BKT transition, etc.

15 min. break

HL 2.7 Mon 11:15 H15

Ultrafast All-Optical Probe of Broken Time-Reversal Symmetry in Monolayer WSe_2 — •Paul Herrmann[1], Sebastian Klimmer[1], Thomas Lettau[1], Till Weickhardt[1], Anastasios Papavasileiou[2], Ksenia Mosina[2], Zdeněk Sofer[2], Jan Wilhelm[3], and Giancarlo Soavi[1] — [1]Friedrich Schiller University Jena, Germany — [2]University of Chemistry and Technology, Prague, Czech Republic — [3]University of Regensburg, Germany

The combination of broken space inversion and preserved time-reversal symmetry (TRS) underlies the spin-valley degree of freedom in monolayer transition metal dichalcogenides (TMDs). Introduction of an imbalance between the energy degenerate, but non-equivalent valleys (local extrema of the bandstructure) at the ±K points of the Brillouin zone breaks TRS. We probe broken TRS and a valley imbalance on ultrashort time scales by comparing the second harmonic (SH) intensity for a circularly *vs.* linearly polarized fundamental beam (FB). By numerically and analytically solving the semiconductor Bloch equations, we show that a two-photon resonant right/left circularly polarized FB interacts exclusively with the ±K valley. Thus, a circularly polarized FB probes the C_{3h} wave vector group of the ±K valleys, in contrast to a linearly polarized FB probing the D_{3h} group of the Γ point. This difference between wave vector groups at K and Γ fully captures and explains the experimentally measured deviation from the otherwise expected ratio of 2 in the circular *vs.* linear SH intensities.

HL 2.8 Mon 11:30 H15

Layer number sensitive Raman modes of atomically thin layered MoS_2 — •Henry Hübschmann[1], Gerhard Berth[1], Klaus Jöns[1], Katharina Burgholzer[2], and Alberta Bonanni[2] — [1]PhoQS Institute, CeOPP and Department of Physics, Paderborn University, Paderborn, Germany — [2]Johannes Kepler University Linz, Linz, Austria

The material group of TMDCs like molybdenum disulfide has gained great attention in the fields of quantum technologies over the last decade due to their particular electronic and optical properties [1]. 2D-MoS_2 has found many applications in optoelectronics and photonics, where the tunable electronic band gap exhibiting strong strutural dependency is an essential feature [2]. Here Raman spectroscopy represents the method of choice for the layer number identification of such 2D structures [3].
This work deals with layer structure sensitive phonon modes of mechanically exfoliated 2D-MoS_2 utilizing Raman analysis. Regarding the two main phonon modes occuring we successfully observed corresponding Raman shifts for monolayer to nine-layer configurations showing a specific dependency on the layer number, enabling the unambiguous determination of the layer number. Besides the two dominant vibrations many other phonon modes are identified, assigned to the symmetry and analyzed in the same manner. Within our comprehensive study we found other structure sensitive phonon modes, showing specific dependency on the layer number, expanding the set of Raman modes for the investigation of 2D-MoS_2 and its properties.

HL 2.9 Mon 11:45 H15

Universal and ultrafast probe of broken time-reversal symmetry — •Florentine Friedrich[1], Paul Herrmann[1], Sebastian Klimmer[1], Zdeněk Sofer[2], Shridhar Sanjay Shanbhag[3], Jan Wilhelm[3], and Giancarlo Soavi[1] — [1]Institute of Solid State Physics, University of Jena, Germany — [2]Department of Inorganic Chemistry, University of Chemistry and Technology Prague, Czech Republic — [3]Institute of Theoretical Physics and Regensburg Center for Ultrafast Nanoscopy, University of Regensburg, Germany

Time-reversal symmetry (TRS) defines some of the most fundamental properties of condensed matter, such as the relation between energy, spin and Berry curvature, therefore influencing features like topology or non-trivial spin textures. Two-dimensional transition metal dichalcogenides (TMDs) offer the possibility to engineer TRS and space-inversion symmetry independently, rendering them excellent model systems. TRS in TMDs can be lifted *via*, *e.g.* valley-selective bandgap opening. Nonlinear optics (NLO) as a probe of TRS is non-invasive and ultrafast; however, detection *via* second harmonic generation is limited to non-centrosymmetric systems. In this work, we make the nonlinear all-optical probe of broken TRS universal by utilizing polarization-resolved third harmonic generation (THG), which is always present also in centro-symmetric crystals. As a proof of principle, we probe broken TRS in a TMD monolayer, by using elliptically polarized light, leading to valley and spin asymmetric gap opening. We then test our method on a TMD bilayer, probing broken TRS in a centrosymmetric system with THG for the first time.

HL 2.10 Mon 12:00 H15

Ultrafast pump-probe spectroscopy of WSe_2 multilayer bubbles — •Jens-Christian Drawer, Henri Melchert, Marti Struve, Christian Schneider, and Martin Esmann — Carl von Ossietzky Universität Oldenburg, Oldenburg, Germany

Heterostructures of van der Waals materials have recently emerged as a versatile platform for the tailored generation and detection of coherent acoustic phonons in the GHz up to THz frequency range [1]. In this work, we investigate WSe_2 layers of different thicknesses by ultrafast pump-probe spectroscopy and observe thickness-dependent breathing-type acoustic phonon modes up to 850 GHz in frequency in the bilayer case. Under suitably chosen conditions for the dry-gel stamping preparation, 40 nm thick WSe_2 flakes form nanobubbles. For these bubbles, our pump-probe measurements reveal a significant improvement in the quality factor of their fundamental 29 GHz acoustic Fabry-Pérot mode from $Q = 13.9 \pm 0.7$ to $Q = 141 \pm 5$. We then tailor these high-Q modes by encapsulating the bubbles with hBN, introducing new spectral features attributed to coupled acoustic modes between WSe_2 and hBN. For example, our approach may enable the ultrafast modulation of strain-defined quantum emitters found in WSe_2.

[1] Yoon, Y. et al.. Nature 631, 771–776 (2024).

HL 2.11 Mon 12:15 H15

Polarized optical contrast spectroscopy of in-plane anisotropic van-der-Waals heterostructures — Ernst Knöckl[1,2], •Alexandre Bernard[1,2], Alexander Holleitner[1,2], and Christoph Kastl[1,2] — [1]Walter Schottky Institute and Physics Department, TU Munich, Garching, Germany — [2]Munich Center for Quantum Science and Technology, München, Germany

To properly exploit interfacial symmetry breakings in van der Waals (vdW) heterostructures, giving rise to emergent behaviors, it is crucial to determine the symmetry axes of the individual layers.

I will discuss polarized optical contrast spectroscopy as a simple and non-destructive approach to characterize the crystalline anisotropy and orientation of 2D materials in vdW heterostructures. We developed a 3D-printed motorized polarization module compatible with typical microscope platforms, allowing broadband polarization-resolved reflectance spectroscopy. We investigated the in-plane birefringence of exfoliated MoO3 thin films (optically transparent) and few-layer WTe2 crystals (semi-metallic). We compared the measured spectra to a model based on a transfer matrix formalism.

Contrasting with other polarization-sensitive approaches, such as Raman or second harmonic generation spectroscopy, this method requires orders of magnitude less excitation power densities, avoiding degradation of delicate layers. Furthermore, it allows quick and simple polarization-sensitive absorbance measurements to resolve anisotropic excitonic properties in symmetry-breaking heterostructures or anisotropic semiconductors.

HL 2.12 Mon 12:30 H15

Microscopic comparison of TMD and QW laser capabilities — •Tommy Schulz, Daniel Erben, Alexander Steinhoff, and Frank Jahnke — Institut für theoretische Physik, Bremen, Germany

The lasing capabilities of monolayer transition metal dichalcogenides (TMDs) are compared to quantum wells (QWs). For material-realistic calculations of the optical gain we connect tight-binding bandstructures for TMDs and $\vec{k} \cdot \vec{p}$ band structures for QWs and the respective interaction matrix elements with state of the art many-body theory. The semiconductor Bloch equations are solved for highly excited ma-

terials, where Coulomb interaction is treated selfconsistently on a GW level together with carrier-phonon interaction. While TMDs provide larger material gain, they also exhibit large shifts of the peak gain with increasing high excitation densitiy, thereby limiting the lasing capabilities.

HL 2.13 Mon 12:45 H15

Strong coupling between light confined in a dielectric nanocavity and excitons in a monolayer TMDC — •Frederik Schröder[1,2], Paweł Wyborski[1], Meng Xiong[1,2], George Kountouris[1,2], Battulga Munkhbat[1], Martijn Wubs[1,2], Philip T. Kristensen[1,2], Jesper Mørk[1,2], and Nicolas Stenger[1,2] — [1]Department of Electrical and Photonics Engineering, Technical University of Denmark, 2800 Kgs. Lyngby, Denmark — [2]NanoPhoton - Center for Nanophotonics, Technical University of Denmark, Ørsteds Plads 345A, DK-2800 Kgs. Lyngby, Denmark

Enhancing light-matter interactions with optical nanocavities is essential for many applications, such as nanolasers and quantum technologies. Recently, advances in the design and fabrication of extreme dielectric confinement (EDC) cavities enabled the confinement of electromagnetic fields in InP on the tens of nanometer scale without being limited by absorption losses [1]. We demonstrate experimentally strong light-matter interactions between excitons in a single layer of $MoTe_2$ and light confined in an EDC nanocavity. The avoided crossing of the system is verified with both photoluminescence and reflection measurements. The observed Rabi-energy of 10.1 meV exceeds the averaged losses in the system [2]. These results pave the way for future studies on nonlinearities at the single-photon level [3].

[1] M. Xiong et al., *Opt. Mater. Express*, 14, 397 (2023), [2] F. Schröder et al., *in preparation*, [3] E. V. Denning et al., *Phys. Rev. Res.*, 4, L012020 (2022)