



ICP2DC4

4TH INTERNATIONAL CONFERENCE ON PHYSICS OF TWO-DIMENSIONAL CRYSTALS 2019

**International Center for Polaritonics
Westlake University**

Mediterranean Institute of Fundamental Physics

10th – 15th June 2019, CHINA

ICP 2DC4'19



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Presentation



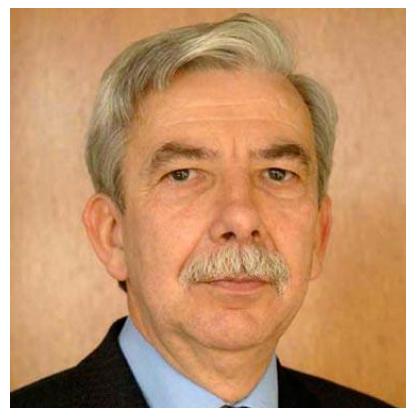
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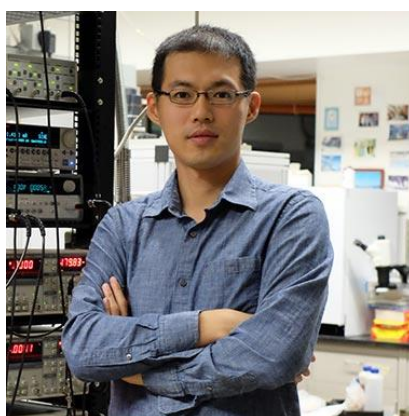
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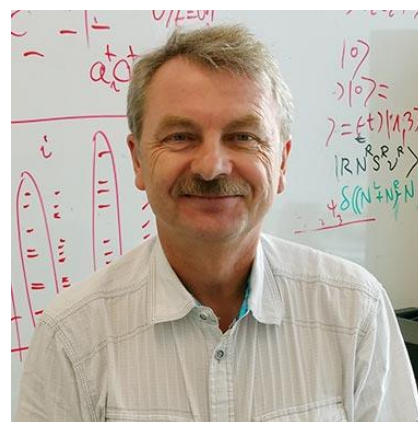
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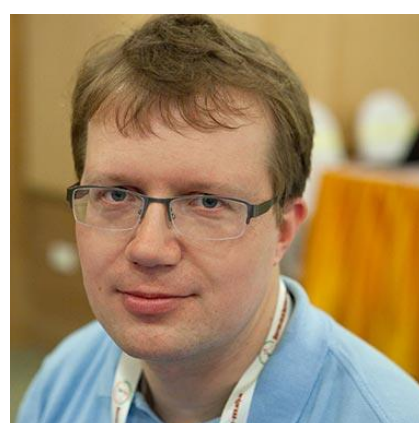
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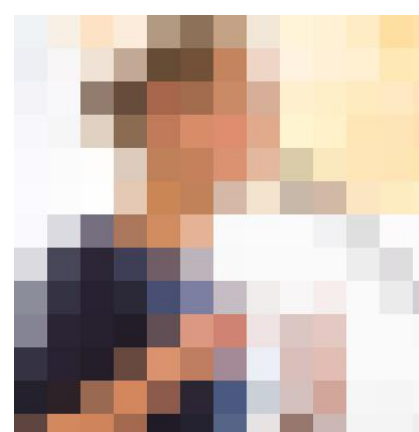
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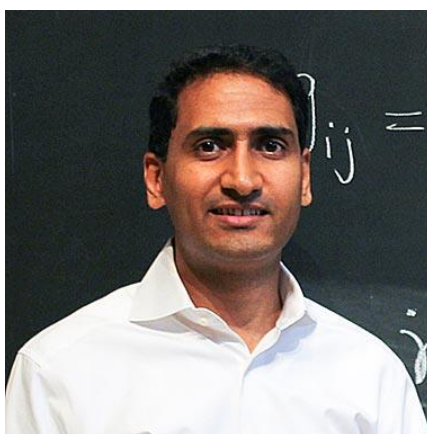
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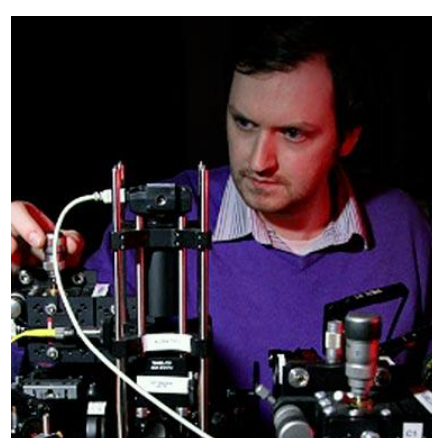
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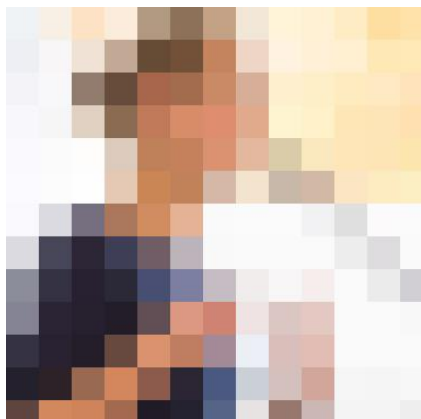
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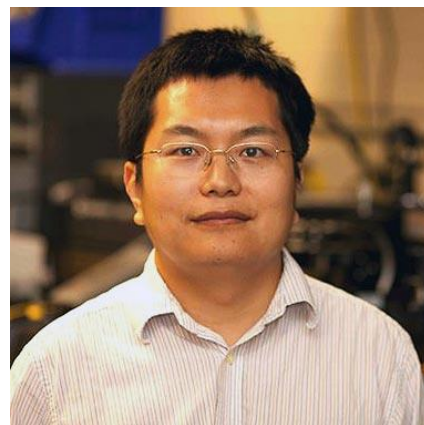
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Westlake University, China

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General Information

Welcome Reception: 10 June 2019

Bar in the lobby of Zhejiang Hotel
No. 278, Santaishan Road, Hangzhou

Conference Venue: 11-14 June 2019

Osmanthus Room
Conference Center
Zhejiang Hotel
No. 278, Santaishan Road, Hangzhou

Poster Session: 11 June 2019

Osmanthus Room
Conference Center
Zhejiang Hotel
No. 278, Santaishan Road, Hangzhou

Conference Lunch: 11-14 June 2019

Lilac Café Main
Building 2F
Zhejiang Hotel
No. 278, Santaishan Road, Hangzhou

Conference Banquet: 12 June 2019

Lotus Room
Zhejiang Hotel
No. 278, Santaishan Road, Hangzhou

Hotel Layout Plan



Program

Day 1 - Tuesday June 11, 2019

09:00 to 09:15	Alexey Kavokin Chairman Opening Ceremony
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Photonic Crystals and Structures Session Chair: Alexey Kavokin

09:20 to 10:00	Yasuhiko Arakawa Plenary Speaker <i>Excitons in quantum dots for advanced light sources with photonic nanostructures</i>
10:00 to 10:30	Zhanghai Chen Invited Speakers <i>t.b.d.</i>
10:30	Break

Technology of Thin Crystals Session Chair: Brian Gerardot

11:00 to 11:30	Takashi Taniguchi Invited Speaker <i>Synthesis of boron nitride and other 2D single crystals under high pressure</i>
11:30 to 12:00	Kazuhiko Hara Invited Speaker <i>Improvement in the luminescence property of hexagonal boron nitride grown by CVD on a c-plane sapphire substrate</i>
12:00 to 12:30	Yuhuai Liu <i>Two-dimensional h-BN multilayer grown on AlN by metalorganic vapor phase epitaxy</i>
12:30	Lunch

Defects and Quantum dots in 2D layers Session Chair: Marek Potemski

14:00 to 14:30	Rudolf Bratschitsch Invited Speaker <i>Single-photon emitters in 2D materials</i>
14:30 to 15:00	Brian Gerardot Invited Speaker <i>Valley-Dependent Properties of Moiré Trapped Interlayer Excitons</i>
15:00 to 15:30	Ajit Srivastava Invited Speaker <i>Single photons, phonons and spins atomically thin WSe₂</i>
15:30 to 15:45	Zai-Quan Xu <i>Fabrication and electrically tuning of ultra-pure quantum emitters in hBN</i>

15:45 to 16:00	Julian Klein <i>Atomistic defect states as quantum emitters in monolayer MoS₂</i>
16:00	Break

Quantum Materials and Graphene I

16:15 to 16:45	Alexey Kavokin Invited Speaker <i>Free Standing Monoatomic Carbon Chains</i>
16:45 to 17:15	Jiun-Haw Chu Invited Speaker <i>Strain-tuned topological phase transition in ZrTe₅</i>
17:15 to 17:45	Tatiana Shubina Invited Speakers <i>Exciton states and radiative properties of chalcogenide nanotubes and related nanostructures</i>

POSTER SESSION

VIP DINNER

Day 2 - Wednesday June 12, 2019

Quantum Materials and Graphene II

Session Chair: Takashi Taniguchi

09:00 to 09:30	Hongqi Xu Invited Speaker <i>Semiconductor InSb nanolayers: A new platform for developments of quantum and topological devices</i>
09:30 to 10:00	Annika Kurzmam Invited Speakers <i>Electrostatically-Defined Quantum Dots in Bilayer Graphene</i>
10:00 to 10:15	Bin Cao <i>Unbiased, non-equilibrium photo-carrier transport in the quantum Hall regime</i>
10:15	Break

Excitons in TMD I

Session Chair: Rudolf Bratschitsch

10:30 to 11:00	Xiaodong Xu Invited Speaker <i>t.b.d.</i>
11:00 to 11:30	Alexey Chernikov Invited Speaker <i>Dielectric disorder and exciton propagation in 2D semiconductors</i>
11:30 to 11:45	Kai-Qiang Lin <i>Electromagnetically induced transparency in second-harmonic generation from monolayer WSe₂</i>
11:45	Break

Theory I

Session Chair: Andreas Knorr

12:00 to 12:30	Pawel Hawrylak Invited Speaker <i>Optical properties of 2D crystals: band nesting, exciton spectrum, robust trions and laser cooling</i>
12:30 to 13:00	Iann Gerber Invited Speaker <i>Beyond standard density functional theory investigations of optical properties of transition metal dichalcogenides based structures</i>
13:00	Lunch

Excitons in TMD II

Session Chair: Alexey Chernikov

14:30 to 15:00	Marek Potemski Invited Speaker <i>Direct bandgap excitons in atomically thin semiconductors</i>
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15:00 to 15:15	Jonas Gael Roch <i>Spin-Polarized Electrons in Monolayer MoS₂</i>
15:15 to 15:45	Shiwei Wu Invited Speaker <i>Unveiling the Defect Structure of Localized Excitons in WSe₂ Monolayer</i>
15:45	Break

Theory II

Session Chair: Pawel Hawrylak

16:15 to 16:45	Friedhelm Bechstedt Invited Speaker <i>Quantum spin Hall phase versus quantized spin Hall conductivity?</i>
16:45 to 17:15	Alexander Steinhoff Invited Speaker <i>Biexciton fine structure in monolayer transition metal dichalcogenides</i>
17:15 to 17:45	Andreas Knorr Invited Speaker <i>Theory of ultrafast excitonic intervalley dynamics in TMDs: interplay of dark states, phonons and Coulomb exchange</i>
17:45 to 18:00	Pierre Gilliot <i>Exciton Fine-Structure in Transition-Metal Dichalcogenides Mono-Layers</i>

CONFERENCE BANQUET

Day 3 - Thursday June 13, 2019

TMD Polaritons I

Session Chair: Christian Schneider

09:00 to 09:30	Paul Walker Invited Speakers <i>Polariton nonlinear interactions - from GaAs waveguides to transition metal dichalcogenide microcavities</i>
09:30 to 09:45	Mateusz Król <i>Valley polarization of exciton-polaritons in monolayer WSe₂ in a tunable microcavity</i>
09:45	Break

Perovskites

Session Chair: Antonio Polimeni

10:00 to 10:30	Su Rui Invited Speaker <i>Optical Properties and Strong Light-Matter Coupling in 2D Perovskite Crystals</i>
10:30 to 11:00	Jochen Feldmann Invited Speaker <i>Halide Perovskite Nanocrystals: from platelets to supercrystals</i>
11:00 to 11:15	Michał Baranowski <i>Optical spectroscopy of 2D perovskites in high magnetic fields</i>

EXCURSION

Day 4 - Friday June 14, 2019

TMD polaritons II Session Chair: Alexander Högele

09:00 to 09:45	Atac Imamoglu Plenary Speaker <i>Many-body optical excitations in solid-state systems</i>
09:45 to 10:15	Jie Gu Invited Speakers <i>Control of light-matter interaction in Van der Waals materials</i>
10:15 to 10:30	Kyoung-Duck Park <i>Probing nanoscale light-matter interactions in atomically thin semiconductors</i>
10:30	Break

Excitons in TMD III Session Chair: Evgeny Alexeev

11:00 to 11:30	Alexander Högele Invited Speaker <i>Cavity and magnetic field control of interlayer excitons in van der Waals heterobilayers</i>
11:30 to 11:45	Roman Cherbunin <i>Kerr rotation study of indirect excitons in $WSe_2 / Mo_{0.5}W_{0.5}Se_2$ heterostructures</i>
11:45 to 12:15	Philipp Merkl <i>Internal structure and ultrafast dynamics of interlayer excitons in van der Waals hetero-bilayers</i>
12:15 to 13:00	Round-Table

Round-Table Collaboration East-West in 2D Physics

13:00	Lunch
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TMD polaritons III Session Chair: Paul Walker

14:00 to 14:15	Yan Xue <i>Generation and memory of a temporal pulse train in a polariton condensate</i>
14:15 to 14:30	Vasily Kravtsov <i>Strong coupling of excitons in $MoSe_2$ and optical bound states in the continuum</i>
14:30 to 14:45	Matthias Wurdack <i>Hybrid polariton condensation in a $MoSe_2$-GaAs Tamm device</i>

14:45

Break

Excitons in TMD IV
Session Chair: Atac Imamoglu

15:15 to 15:45

Evgeny Alexeev | Invited Speaker
Controlling the optical properties of van der Waals heterostructures by interlayer rotation

15:45 to 16:15

Antonio Polimeni | Invited Speaker
Controlled micro/nano-dome formation in proton-irradiated bulk transition-metal dichalcogenides

16:15 to 16:45

Cedric Robert | Invited Speaker
Control of the exciton radiative lifetime in van der Waals heterostructures

16:45 to 17:00

Florian Sigger
Hybridized indirect excitons in MoS_2/WS_2 heterobilayers

Announcement for next ICP2DC
Alexey Kavokin



Oral Abstracts

Excitons in quantum dots for advanced light sources with photonic nanostructures

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Semiconductor quantum dots (QDs) in photonic crystals (PhCs) are one of the most fascinating platforms for exploring the physics of light matter interactions in the solid state as well as for developing novel quantum optoelectronic devices. The PhC structure enables an advanced control of the vacuum field, resulting in a substantial modification of its interaction with the excitons in the QDs. We have been working on excitons and polaritons in QDs with 1D, 2D and 3D PhC nanocavities for investigating light-matter interaction and light sources application.

In this presentation, we discuss advances in excitons and polaritons in the QDs with photonic nanostructures, including time-domain measurement of the vacuum Rabi oscillation in a QD-nanocavity coupled system. We also discuss quantum dot light sources with QDs, such as topological nanocavity lasers, nanowire lasers and single-photon sources integrated on a CMOS silicon photonic chip.

Synthesis of boron nitride and other 2D single crystals under high pressure

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The attractive potential of hexagonal boron nitride (hBN) as wide-band gap materials was realized after obtaining high quality single crystals by using high pressure synthesis process [1]. Taking advantage of the highly luminous properties of hBN, a stable operation of far ultraviolet - plane-emission device was demonstrated [2]. It is also emphasized that hBN exhibits superior properties as a substrate of graphene devices [3].

In order to realize these newly developed potential of hBN crystals, more precise insight for its quality control is important. Although the major impurities affects the optical properties of hBN are carbon and oxygen, the details of their contribution are still the subject of study. For the application of graphene's substrates, some unknown point defects in hBN are considered to still affect its device qualities. In order to figure out this issue, this paper focus on a spatial distribution of residual carbon impurity within hBN single crystals and modification of their properties depend upon post heat treatments.

Another issue is to fabricate fine hBN crystals with high quality via conventional route. Although high pressure synthesis process has an advantage to use reactive alkali-base solvent such as Ba-BN, search for the alternative synthesis route without pressure is important for the practical application of hBN. Since hBN is thermodynamically stable at high temperatures and at atmospheric pressure, it should be possible to obtain high-quality hBN crystals at atmospheric pressure by using an appropriate solvent. Ni or Co-base metal base solvents seem useful to obtain high quality hBN crystals [4], though the yield of the crystals is less than those of high pressure process.

On the other hands, liquid phase crystal growth process is also applicable for other 2D materials such as graphite, black phosphor (BP) and TMDs. Also, controlling of boron and nitrogen isotope ratio (^{10}B , ^{11}B and ^{15}N) in hBN and cBN crystals can be now carried out by methathesis reaction under HPHT.

In this paper, recent studies for synthesis of BN and other 2D single crystals under high pressure with respect to impurity control will be reported. Particularly the present issue to improve properties of BN further is to eliminate residual carbon and oxygen impurities. Furthermore, recent challenge for obtaining boron and nitrogen isotope control of BN crystals will also be introduced.

[References]

- [1] T. Taniguchi, K. Watanabe, J. Cryst. Growth, **303**, 525 (2007).
- [2] K. Watanabe, T. Taniguchi, A. Niiyama, K. Miya, M. Taniguchi, Nature Photonics **3**, 591 (2009).
- [3] C.R. Dean, A.F. Young, I. Meric, C. Lee, W. Lei, S. Sorgenfrei, K. Watanabe, T. Taniguchi, P. Kim, K.L. Shepard, J. Hone, Nature Nanotechnology, **5**, 722 (2010).
- [4] Y. Kubota, K. Watanabe, O. Tsuda, T. Taniguchi, Science, **317**, 932 (2007).

Fundamental Insights And Perspectives Into Novel 2d Anisotropic Materials

Sefaattin Tongay

Anisotropic 2D materials (pseudo-1D crystals) are a new class of materials in which atoms are confined in 2D but are arranged in a way that they form 1D chain-like features running across one specific lattice direction. They exhibit direction and polarization dependent properties that allow for a new degree of freedom, which are particularly attractive for a number of advanced photonic, optics, and optoelectronic applications. In a sense, they offer unique properties that fall between traditional 1D and 2D material systems. This talk summarizes recent advances made in pseudo-1D material synthesis, characterization, fundamental understanding, and applications by the team led by Prof. Tongay at Arizona State University. Special emphasis will be given to recent projects by Tongays team using novel growth methods, one-of-a-kind spectroscopy, microscopy, scanning transmission electron microscopy (STEM), high-resolution TEM (HRTEM), and spectroscopy facilities at Arizona State University.

Improvement in the luminescence property of hexagonal boron nitride grown by CVD on a c-plane sapphire substrate

**Kazuhiko Hara,^{1,2,*} Naoki Umehara,² Kohei Shima,³ Kazunobu Kojima³
and Shigefusa F. Chichibu³**

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Hexagonal boron nitride (h-BN), which has a graphite-like crystal structure with stacked honeycomb sheets of boron and nitrogen, is expected to be one of the key materials in the future electronics devices [1]. Its unique crystallographic property, excellent electric insulation and efficient luminescence at 215 nm have proven the potential as a substrate and an insulating layer for Van der Waals heterostructures, a light emitting material for the deep ultraviolet region and so on. Aiming at the development of such devices, we have paid attention to the growth of h-BN thin films on a c-plane sapphire substrate by chemical vapor deposition (CVD) with BCl₃ and NH₃ as boron and nitrogen sources, respectively. This source combination, which has been used to fabricate pyrolytic BN, is expected to be suitable for fast growth of h-BN on a large-size substrate. We were also motivated by the observation of intrinsic excitonic emission at room temperature from h-BN films grown on a nickel substrate by CVD with the same source combination [2].

The CVD apparatus used in this study consisted of a horizontal hot-wall reactor tube made of BN ceramics with an inner diameter of 40 mm and a high-temperature tubular furnace with graphite heaters [3]. The typical growth temperature was 1200 °C. We have found that the growth pressure (P_g) has a major impact on the sample properties. The morphology of the grown film was improved by reducing P_g from atmospheric pressure to 20 kPa. The samples grown at 20 kPa consisted of columnar grains with a flat top surface, some of which coalesced each other, whereas those with rough surfaces were only grown at atmospheric pressure. Low pressure growth also achieved the formation of single crystal h-BN films with uniform out-of- and in-plane orientations.

Furthermore, it was found that the growth at 5 kPa improves the cathodoluminescence (CL), resulting in the observation of intrinsic exciton emission at room temperature (RT). It should be noted that its spectral width, 5 nm in FWHM, is almost same as that observed for the high quality bulk crystal fabricated by a high-pressure and high-temperature solution technique [4]. The fine structures corresponding to the indirect excitonic line and its phonon replicas were clearly observed at 12 K. The CL images at RT indicated that the intrinsic exciton emission was observed only from the columnar crystal grains, revealing high crystalline quality of the columnar grains. On the other hand, the deep luminescence at 350 nm was observed from the entire surface with relatively high intensities from the valley region between the columnar grains, where the randomly oriented grains were formed. Attempts to further improve the film quality will be reported in the presentation.

This work was supported in part by JSPS KAKENHI Grant Nos. 18K04231, JP16H06427 and 17H02907 by MEXT.

References

- [1] A. K. Geim and I. V. Grigorieva, *Nature* **499**, 419 (2013). [2] O. Tsuda, K. Watanabe and T. Taniguchi., *Jpn. J. Appl. Phys.* **46**, L287 (2007). [3] N. Umehara, A. Masuda, T. Shimizu, I. Kuwahara, T. Kouno, H. Kominami, and K. Hara, *Jpn. J. Appl. Phys.* **55**, 05FD09 (2016).
- [4] K. Watanabe and T. Taniguchi, *Int. J. Appl. Ceram. Technol.* **8**, 977 (2011).

Two-dimensional h-BN multilayer grown on AlN by metalorganic vapor phase epitaxy

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Hexagonal boron nitride (h-BN), an isostructural material to graphene, is a wide bandgap semiconductor of interest for electronics, deep UV photonics and two-dimensional heterostructures. A scalable technique of metal-organic vapor phase epitaxy (MOVPE) has been utilized to epitaxial growth of III-V compounds including h-BN. However, sapphire and Si, the most common substrates for semiconductors, are not very suitable for deposition of h-BN, which requires growth temperatures in the range of 1500 °C. SiC would be one choice and indeed has been used for h-BN epitaxy. AlN could be an alternative option, as it is also thermally stable up to 1600 °C even under ammonia, optical transparent until 200 nm, in theory smaller lattice mismatch than sapphire to h-BN, and thus is perfectly compatible with BN epitaxy by MOVPE. The growth of h-BN on AlN provides important knowledge to integration of h-BN into AlN- or high Al-content AlGaN-based UV devices¹. H-BN as sacrificing layer and grown on AlN may further increase the versatility in the transfer technique since AlN can be grown on a wide variety of substrates. Due to high thermal stability and exfoliation property, it could also serve as protective coating material for ion-implantation and high temperature post-healing process in AlN.

In this work, h-BN layers were directly grown on AlN/sapphire template by MOVPE. The layers were formed with different cycles and each alternately supplies NH₃ for 2 sec and triethylboron for 1 sec. The growth pressure and nominal V/III ratio were maintained at 3.85 kPa and 3000, respectively. The impact of flow rates, process temperatures, and supply time on h-BN growth were comprehensively investigated. At initial growth stage, the lateral 2D growth rate of nuclei is ~25 nm/min and the vertical growth rate (nuclei height) ~0.3 nm/min, indicating the process is not self-limiting. On those nuclei, highly ordered 2D h-BN layers were grown as confirmed by X-ray diffraction (XRD) and transmission electron microscopy. The nucleation and growth mechanism will be further discussed in the conference.

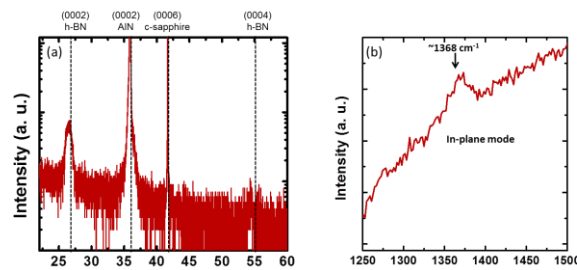


Figure 1: (a) XRD 2θ-ω measurement of a 40 nm thick h-BN layer grown on AlN/sapphire template (b) and corresponding Raman spectrum. Both confirm 2D h-BN formation with well-ordered lattice.

Acknowledgements

This work is partially supported by JSPS KAKENHI #17J05229, National Institute for Materials Science, Strategic International Collaborative Research Program (SICORP) of Japan Science and Technology Agency (JST), MOST-SKRD (2016YFE0118400), and KPSTHP (172102410062). The authors would like to thank Professor Hideto Miyake for providing AlN/sapphire templates.

References

- [1]. H. X. Jiang and J. Y. Lin, Semicond. Sci. Technol. 29, 084003 (2014) 084003.

Single-photon emitters in 2D materials

Rudolf Bratschitsch^{1,*}

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Graphene is known as a prototypical two-dimensional material with unique physical properties. However, the difficulty of creating an optical band gap stimulated the search for other monolayer materials.

Atomically thin transition metal dichalcogenides serve as a promising new material class for opto-electronics. In contrast to thicker crystals, monolayers of MoS₂, WS₂, MoSe₂, and WSe₂ exhibit prominent photoluminescence. Recently, we have discovered bright and stable single-photon emitters in single layers of WSe₂ [1], which renders atomically thin semiconductors also interesting for quantum optics. In my talk, I will show that these quantum light sources are strain-induced and demonstrate deterministic positioning of the emitters on the nanoscale [2]. Furthermore, I will present single-photon emission from the layered monochalcogenide semiconductor GaSe [3] and provide evidence that the incorporated non-classical light sources are also strain-induced. Next, I will demonstrate that these single photons originating from GaSe emitters can be routed in dielectric waveguides on a photonic chip [4]. Finally, I will discuss the nature and prospects of newly discovered single-photon emitters in hBN [5].

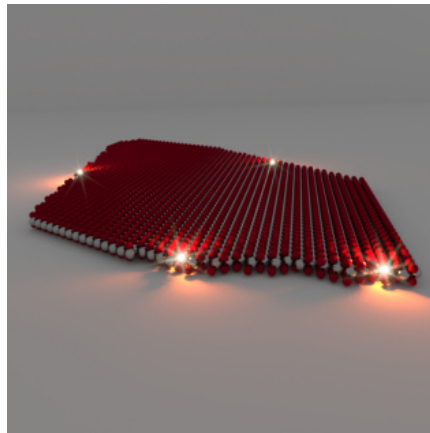


Figure 1: Artistic impression of a WSe₂ monolayer with several single-photon emitters.

References

- [1] P. Tonndorf et al., *Optica* **2**, 347 (2015).
- [2] J. Kern et al., *Advanced Materials* **28**, 7101 (2016).
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- [5] D. Wigger et al., *2D Materials* **6**, 035006 (2019).

Valley-Dependent Properties of Moiré Trapped Interlayer Excitons

Mauro Brotons-Gisbert, Hyeonjun Baek, Dale Scerri, and Brian D. Gerardot

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Van der Waals (vdW) heterostructures, in which a wide range of unique atomic layers can easily be combined, offer novel prospects to engineer and manipulate quantum confined states. I will present evidence for quantum confined states engineered by the periodic potential landscape of interlayer valley excitons in a vdW heterostructure. Here, the moiré potential of a twisted MoSe₂/WSe₂ heterostructure traps interlayer excitons, and we show that the trapped interlayer excitons inherit the magneto-optical properties of the type-II band-structure. Finally, we will discuss the ability to charge the trapped states with electrons or holes in tunable electronic devices. These results highlight new opportunities to engineer quantum confined spins in vdW heterostructures.

Title: Single photons, phonons and spins atomically thin WSe₂

Abstract: Monolayer transition metal dichalcogenides (TMDs), such as WSe₂, are atomically thin semiconductors with a “valley” degree of freedom, which can be optically addressed, thus opening up exciting possibilities for “valleytronics”. Recently, naturally occurring single quantum emitters, believed to be excitons trapped in shallow potentials, were reported in TMDs. They seem to inherit the valley degree of freedom from the host TMD and owing to their longer lifetimes, appear promising for quantum information processing applications.

In this talk, I will begin by highlighting some unique properties of TMDs excitons which result from the off-Gamma-point origin of the constituent single particle electronic states. After describing the basic properties of quantum dots in TMDs [1], I will present evidence for quantum entanglement between chiral phonons of the 2D host and single photons emitted from the quantum dots [2]. I will also present evidence for optical initialization of single spin-valley in trapped, positively charged WSe₂ excitons [3]. Due to quenching of electron-hole exchange, which is the main mechanism of valley-mixing in neutral excitons, the valley lifetime of excess spin-valley is prolonged to at least nanoseconds timescale. Our work extends the field of two-dimensional valleytronics to the level of single spin- valleys, with implications for quantum information and sensing applications.

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Fabrication and electrically tuning of ultra-pure quantum emitters in hBN

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Solid-state, quantum emitters (QEs) are critical components for future quantum photonics with potential applications in quantum technologies. Hexagonal boron nitride (hBN) has recently found to host bright and optically stable QEs at room temperature¹. Despite all the efforts so far²⁻³, reliable techniques to prepare highly pure QEs in large scale has yet shown, which hinder the development of applications in practical devices. In this work, we first demonstrate chemical vapor deposition growth of large-area, few layer hBN films that host large quantity of QEs, 85% of which have a zero-phonon line (ZPL) at $580\pm 10\text{nm}$.⁴ We then develop two methods to process hBN after transfer which significantly improve SPEs in hBN films. The emitters exhibit narrow linewidths of $\sim 3\text{nm}$ and photon purity in excess of 90%. Finally, we demonstrate tuning of the ZPL wavelength using ionic liquid devices over a spectral range of up to 15 nm--the largest obtained to date from any solid-state QEs. Our work lays a foundation for producing high-quality emitters in an ultra-compact 2D material system, and paves the way for deployment of hBN SPEs in scalable on-chip photonic and quantum devices.

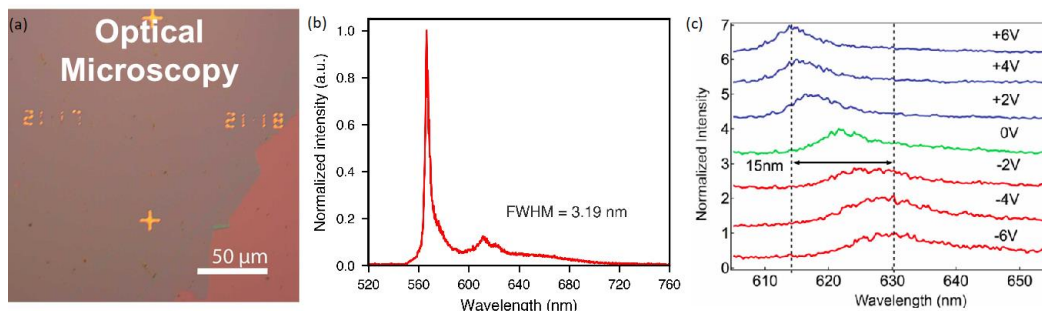


Figure 1: (a) Optical image of the as-transferred hBN films on a Si substrate. (b) A PL spectrum of QE shown a bandwidth of 3nm. (c) Normalized PL spectra of a QE at gate voltages noted displaying a shift of 15 nm. The spectra are offset for clarity.

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Atomistic defect states as quantum emitters in monolayer MoS₂

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We demonstrate the deterministic generation of single defect emitters in a monolayer MoS₂ van der Waals heterostructure. [1] We bombard monolayer MoS₂ with helium ions to generate optically active defect luminescence [2] and encapsulate the defective MoS₂ within hBN to greatly enhance optical quality. [3] The encapsulation of the defective MoS₂ reveals narrow spatially localized spectral lines X_L (see Fig. 1(a)) that exhibit emission that is redshifted by 90 - 220 meV with respect to the neutral 2D exciton (see Fig. 1(b)). We spectroscopically investigate single emitters by performing photoluminescence excitation spectroscopy and temperature dependent measurements. The line shape reveals a strong asymmetry resembling the interaction with LA/TA phonons. Employing the independent Boson model to our emission lines, we find that the emitters are spatially localized to a length scale of 2 nm (see Fig. 1(c)). We attribute the emission to atomistic defects induced by the helium ion bombardment and discuss their origin in the light of scanning tunneling microscopy measurements. [4] Our work paves the way towards a controlled and deterministic generation of single quantum emitters in monolayer TMDC van der Waals heterostructures.

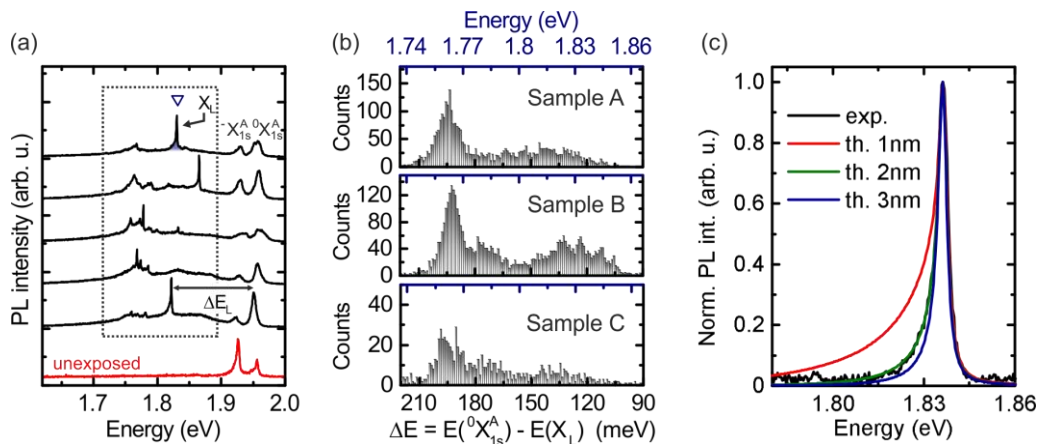


Figure 1: (a) Low temperature (10K) PL spectra of bombarded and unbombarded hBN/MoS₂/hBN. (b) Emission distribution of X_L for three samples. (c) Independent Boson model fit to a typical X_L emitter.

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Semiconductor InSb nanolayers: A new platform for developments of quantum and topological devices

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Semiconductor InSb nanowires have been demonstrated as one of the most promising materials systems for realizing topological superconducting structures in which Majorana bound states can be created and manipulated [1-3]. For achieving quantum computing with Majorana bound states, an efficient scheme for braiding Majorana bound state needs to be developed. In this respect, proposals of using branched InSb nanowires and two-dimensional InSb planar structures have been envisioned [4,5]. In this talk, I will report on our recent developments in epitaxial growth of free-standing InSb nanoplates and in building quantum devices and superconducting Josephson junction devices with these InSb nanoplates [6-8]. These InSb nanoplates were grown by molecular beam epitaxy (MBE) and exhibits excellent structural and transport properties [6]. The advantages of employing these InSb nanoplates include flexibilities of transferring them to desired substrates for device fabrication and of directly contacting them with different metals and superconductors. Several quantum devices have been fabricated using our MBE-grown InSb nanoplates and have been studied by transport measurements. In particular, we will demonstrate realizations of first InSb nanoplate quantum dot devices [7] and first Al-InSb nanoplate-Al Josephson junction devices [8]. Perspectives of achieving topological quantum devices will also be presented and discussed.

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Strain-tuned topological phase transition in ZrTe_5

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Band insulators with time reversal symmetry can be classified into normal insulators (NI), weak topological insulators (WTI) and strong topological insulators (STI) based on their Z_2 topological indices. Changing Z_2 indices requires closing and reopening the bandgap, and topologically distinct insulating phases are separated by a gapless Dirac or Weyl semimetal phase. The van der Waal layered material ZrTe_5 is a prototypical example that the emergence of massive 3D Dirac fermions is due to its proximity to the STI-WTI phase boundary. In this talk, I will demonstrate that a STI-WTI topological phase transition can be induced by applying uniaxial stress to ZrTe_5 , and make the case that the in-situ tunable strain is a powerful tool to study and control topological materials. In addition, due to the low carrier density in this material, the lowest Landau level can be reached within a few tesla of magnetic field. I will describe the unusual magnetotransport behavior in the quantum limit magnetic fields.

Exciton states and radiative properties of chalcogenide nanotubes and related nanostructures

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Nanotubes (NTs) of transition metal dichalcogenides (TMD) attract an increasing interest due to possible applications in nanoelectronics and nanophotonics. The radiation ability of NTs has been discovered recently [1,2], in spite of the fact that they were synthesized long enough ago. Their planar allotrope of a monolayer thickness demonstrates bright photoluminescence (PL) related to direct bandgap excitons, while multilayered structures exhibit the rather weak emission of indirect exciton states. In common opinion, the multiwalled NTs should exhibit similar behavior. Contrary to that, we observe the bright PL from the single TMD NTs (MoS₂ and WS₂) with diameters of 0.2 - 2 microns, whose walls contain several dozens of monolayers.

In contrast to the flakes, synthesized during the same process, the NTs display predominantly the emission of direct excitons. PL temperature dependencies in the NTs and flakes differ by the decrease or rise, respectively, in PL intensity with increasing temperature. However, they have a common feature that is the suppression of the indirect exciton transitions at low temperatures. This has been observed in the flakes that were exfoliated from a high-quality bulk crystal as well. The threshold appearance of the indirect-exciton emission is suggestive of a spin-forbidden lowest state in indirect transitions.

Transmission electron microscopy studies have revealed that the NT walls are not homogeneous but consist of domains comprising 4-7 monolayers, the thickness of whose and mutual shift (chirality) are controlled by the internal strain. Thus the optical properties can be discussed in the context of the formalism of excitons in the chiral stacks [3]. Such architecture promotes the localization of excitons within isolated spatial regions. In other words, the condition for exciton confinement is rather 2D than 3D.

A micron-sized tube can act as an optical resonator supporting whispering gallery modes (WGMs) with the quality factor of several hundreds. The observed sharp peaks of WGMs in the emission spectra are strongly polarized along NT axis (TM polarization). The reason of that is the TE modes possess low quality factor in the spectral range of interest. Theoretical consideration shows that the polarization of excitation affects the initial distribution of excitons in the NT walls, which can be probed using different detection regimes. At oblique light incidence on the NT, the coupling between optical modes and direct exciton is possible with the formation of exciton-polariton, propagating within the NT walls. However, the observation of such polariton needs the tubular structure of higher quality, synthesized by either chemical reaction or grown by molecular beam epitaxy. The first attempts towards exploiting of the latter method are done. In particular, it is demonstrated that such method can create nanostructures of different shapes varied from nanoplatelets to nanotubes, and that some of them can radiate strongly polarized narrow PL lines. In general, our studies open a way towards combination of an effective emitter and a resonator in single tubular structure.

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Free Standing Monoatomic Carbon Chains

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Being monoatomic chains of carbon atoms, carbynes represent ultimate one-dimensional crystals. Unfortunately, free standing carbon chains are instable in vacuum, and fluctuations almost necessarily lead to their deformation and folding. The synthesis of free-standing carbon chains remains one of the Holy Graals of nano-physics and nano-chemistry [1,2]. Here we report on the experimental realization of long straight free-standing carbon chains stabilized by metallic nanoparticles. We synthesize linear carbon chains (carbynes) in a colloidal solution, then deposit them on a surface and study free standing carbyne films with the transmission electron microscopy (TEM). TEM images of the carbon threads with golden nanoparticles attached to their ends demonstrate straight monoatomic carbon chains with full lengths of over 100 nm and linear parts of about 12 nm length (52 atoms), in average. Parallel linear chains of carbon atoms form a quasi one-dimensional crystal structure with a lattice constant of 0.256 nm as confirmed by the X-ray diffraction measurements. This method paves the way to fabrication of an ultimate one-dimensional crystal: a monoatomic carbon wire. Currently, we are performing the low-temperature photoluminescence measurements to reveal excitonic features in the optical spectra of carbynes. The spectra taken at the resonant excitation at 390 nm show the appearance of ultranarrow peaks forming regular triplet groups in the range of 430-490 nm. We cautiously attribute these spectral features to the excitonic transitions and phonon replica in carbon chains of different lengths.

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Electrostatically-Defined Quantum Dots in Bilayer Graphene

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Graphene is a promising candidate for future nano-electronic devices including building blocks for quantum information processing. Reasons are the expected long spin lifetimes and high carrier mobility. Recent improvements in fabrication technologies for graphene nanostructures, namely, the encapsulation between boron nitride, edge-contacting, graphite back-gates and the use of electrostatic gating of bilayer graphene, have leveraged the quality of quantum dots to such an extent, that few-electron or -hole quantum dots have been realized that are comparable to the best devices in gallium arsenide [1].

Here we confine charge carriers laterally by applying strong displacement fields forcing charge carriers to flow through a narrow channel (see inset in Fig. 1). In transport direction, charge carriers are confined by pn-junctions forming natural tunnel barriers, thus creating a p-type quantum dot coupled to n-type leads, or vice versa. In this ambipolar system, we can realize both single electron and single hole occupation of the respective quantum dots showing charging energies on the order of 5 meV (see Fig.1). In addition, we can use our design to form multi-dots [2,3].

We use finite bias spectroscopy to study and identify the single-particle and many-body ground- and excited states (marked with arrows in Fig. 1) of electrostatically-defined quantum dots in bilayer graphene trapping only one or two charge carriers [4]. While the properties of the material bear similarities to carbon nanotubes silicon because of the two-fold valley and spin-degeneracies, the results of our experiments allow us to propose a remarkably clear level scheme for two-particle spectra, in which the spin- and valley-entanglement, as well as exchange interactions play a crucial role. With this level scheme at hand, future experiments can investigate spin- and valley-coherence and relaxation times, which are key parameters to be compared to other material systems.

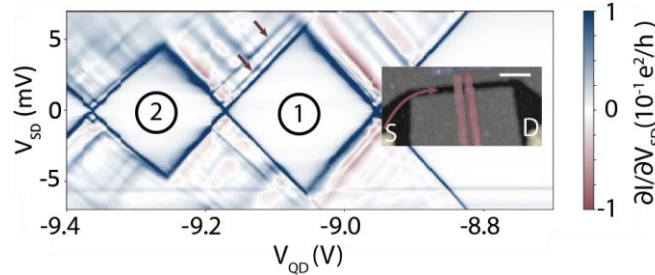


Figure 1. Coulomb diamond measurement for a quantum dot that confines holes. Since the quantum dot can be completely depleted, we can label each diamond with the occupation number of the quantum dot. Inset: AFM image of the quantum dots. The white scale bar has a width of 100 nm.

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Unbiased, non-equilibrium photo-carrier transport in the quantum Hall regime

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In integer quantum Hall systems, optical excitation can be used to investigate a variety of physics; for example, the Landau-level density of states through absorption, and non-equilibrium physics through the generation of excited electrons and holes. Optical techniques are attractive because the length scale associated with photo-excited charge carries lies between that of local probes and global transport measurements, perhaps revealing phenomena in a meso-scale environment. In addition, optical approaches are particularly attractive in Dirac materials where the Landau-level spacing is nonlinear, allowing for more restrictive selection rules for optical transitions.

Here, we discuss results from our photocurrent investigations of optically-excited electrons and holes in BN encapsulated graphene in the integer quantum Hall regime [1]. With no external sample bias between the Ohmic contacts, we observe that the photocurrent oscillates as a function of Fermi level, revealing the Landau-level quantization and the relaxation dynamics of non-equilibrium carriers, see Figure 1. We can distinguish between non-equilibrium carrier diffusion directly to the electrical contacts or directly towards edge states. These oscillations are present even in low magnetic fields, where conductance measurements do not show quantum Hall plateaus. We observe that the photocurrent oscillations are different for Fermi levels near and distant from the Dirac point. Our observation qualitatively agrees with a model that assumes the photocurrent is dominated by chiral edge transport of non-equilibrium carriers and that the chirality depends on the carrier type and curvature of the occupied band. Our experimental results are consistent with electron and hole chiralities being the same when the Fermi level is distant from the Dirac point, and opposite when near the Dirac point.

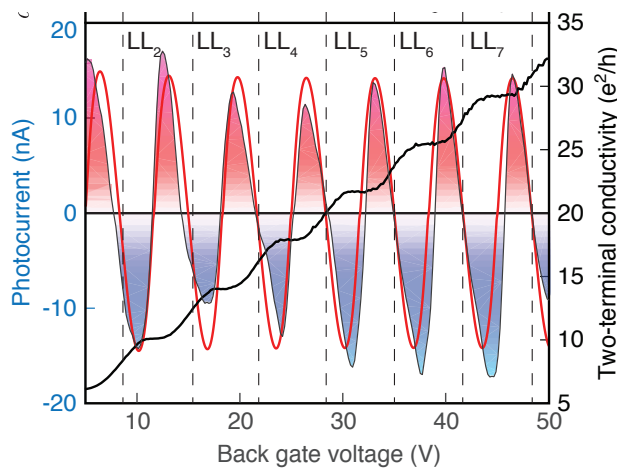


Figure 1. Oscillations of the photocurrent induced by the Landau quantization (4T) as a function of back-gate voltage. **Right axis (Black):** conductivity in the quantum Hall regime. The Dirac point corresponds to -7 V. **Left axis:** Zeros of the strong photocurrent oscillations correspond to half-fillings (vertical dashed lines) and integer-fillings of Landau levels for $LL_n \geq 4$. Red line: Photocurrent calculation.

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Dielectric disorder and exciton propagation in 2D semiconductors

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Semiconducting two-dimensional materials are found to exhibit a number of highly intriguing phenomena, including spin-valley locking, efficient light-matter coupling, and strong Coulomb interaction. Spin-valley physics in particular remain in the focus of the ongoing research, further motivated by a variety of recently available heterostructures to tune and harness the associated phenomena. This strongly motivates increased attention to the consequences of environmental influences and the role of proximity effects for the key properties of local electronic structure.

Here, I will present an alternative, fundamental source of disorder in 2D materials based entirely on the local changes of the Coulomb interaction due to fluctuations of the external dielectric environment. The consequences for optical and transport properties of excitons and free charge carriers will be discussed. The importance of dielectric homogeneity in heterostructures will be further highlighted, demonstrated by spatial mapping of the fluctuations and by identifying the elimination of the dielectric disorder as a main consequence of material's encapsulation. Finally, I will present direct, time-resolved monitoring of exciton transport in 2D semiconductors, discuss the influence of disorder, and address strong non-linearities beyond linear regime.

Electromagnetically induced transparency in second-harmonic generation from monolayer WSe₂

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Electromagnetically induced transparency (EIT) occurs in atomic systems and shows versatile applications in slow-light generation, gain without inversion and optical quantum-information processing [1, 2]. We demonstrate a cavity-free, atomic-like EIT effect in single-layer crystals of WSe₂, probed by exploiting the intrinsic second-harmonic generation (SHG) arising from the breaking of inversion symmetry [3]. Under conditions of double resonance of the driving and radiated field with the fundamental transitions, the SHG spectrum bifurcates. The feature follows a pump-wavelength-dependent spectral anticrossing given in Figure 1, accurately described by a ladder-type three-level model. Crucially, the SHG power-law exponent diverges from the canonical value of 2 to follow a Fano-like dispersion with wavelength. This signature of quantum interference is retained at room temperature, opening up opportunities in solid-state quantum nonlinear optics for optical mixing and gain without inversion.

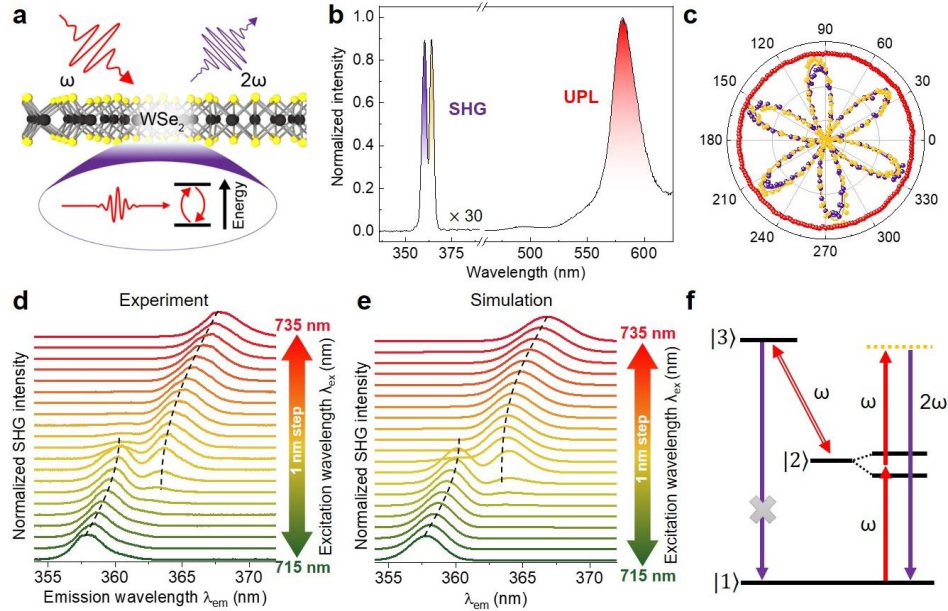


Figure 1: Quantum interference in the second-harmonic generation (SHG) of single-layer WSe₂ at 5 K. **a**, Illustration of SHG from a single layer of WSe₂. **b**, Typical emission spectrum of monolayer WSe₂ encapsulated by hexagonal boron nitride (hBN) measured under excitation at 724 nm, showing bifurcated SHG centred at 362 nm and an additional upconversion photoluminescence (UPL) feature at 581 nm. **c**, The intensity of the two SHG peaks (blue and yellow) and the UPL (red) copolarized to the incident laser as the polarization of the driving field is rotated with respect to the crystal axis. Dependence of the normalized SHG spectrum on pump wavelength in **d**, experiment and **e**, simulation, revealing anticrossing behaviour of the SHG peaks. **f**, Ladder-type three-level model of resonant SHG and the quantum-interference pathways. [3]

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Optical properties of 2D crystals: band nesting, exciton spectrum, robust trions and laser cooling.

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We discuss the electronic and optical properties of monolayer 2D hexagonal crystals, transition metal dichalcogenites (TMDCs). The ab-initio calculations [1-3] establish monolayer TMDCs as direct gap semiconductors. In order to develop a better understanding of the electronic properties and their response to external magnetic field a tight binding model involving Mo and W metal d-orbitals and sulfur dimer S₂ p orbitals is developed based on input from ab-initio calculations [2]. The role of d- and p-orbitals, nearest and next nearest neighbor hopping and the origin of Q-points and their role in band-nesting is clarified. The effective tight binding model is further reduced to the massive Dirac Fermion model at K-points which allows introduction of the magnetic field and explicit derivation of valley-selective light-matter interaction [4]. Using the microscopic wavefunctions the direct and exchange Coulomb matrix elements are computed [5]. The exciton spectrum is obtained from the solution of Bethe-Salpeter equation. The role of screening, valleys and band-nesting on the exciton spectrum is described [5]. We next discuss robust trions and the role of electron-phonon interaction [6], the possibility a Valley Polarized Electron Gas (VPEG) [3] and potential for laser cooling of 2D crystals [7].

*Work in collaboration with M. Bieniek, M. Korkusinski, L. Szulakowska, J. Jadczak, L. Bryja, I.Ozfidan, P. Potasz, A. Delgado, A. Petrou and G. Kioseoglou.

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Beyond standard density functional theory investigations of optical properties of transition metal dichalcogenides based structures

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The present talk will review our recent work advances on optical properties calculations of transition metal dichalcogenide based structures using beyond standard DFT method. Indeed the use of the so-called *GW*+BSE approach is mandatory, since the optical properties of two-dimensional TMD monolayers such as MoS₂ or WSe₂ are dominated by excitons, Coulomb bound electron-hole pairs [1]. As a first example, we will show that the sign and the amplitude of the splitting between bright and dark exciton states can be determined, showing the influence of the spin-orbit coupling on the optical spectra and demonstrating the strong impact of the combined intra-valley Coulomb exchange term and effective mass changes on the dark-bright exciton fine structure splitting [2]. Using the same methodology we have recently reported the existence of interlayer excitons in MoS₂ bilayer structures also observed experimentally [3].

Finally, screening effects due the presence of hexagonal-BN surrounding layers will be presented, since we have calculated the dependence of both the quasi-particle gap and the binding energy of the neutral exciton ground state E_b as a function of the hBN layer thickness. This study demonstrates that the effects of screening at this level of theory are more short-ranged than it is widely believed. The encapsulation of a WSe₂ monolayer by three sheets of hBN (~ 1 nm) already yields a 20 % decrease of E_b whereas the maximal reduction is 27% for thick hBN [4]. Similar calculations have been performed in the case of a WSe₂ monolayer deposited on stacked hBN layers. These results will be compared to the recently proposed Quantum Electrostatic Heterostructure approach.

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Direct bandgap excitons in atomically thin semiconductors

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The energy structure of direct bandgap excitons which appear at K-points of the Brillouin zone in mono- and few-layers of semiconducting transition metal dichalcogenides (S-TMD) will be reviewed.

The s-type Rydberg series of bright excitonic states in S-TMD monolayers will be shown to follow a simple energy ladder $\epsilon_n = -Ry^*/(n + \delta)^2$, $n=1, 2, \dots$, which is well accounted by using the modified Kratzer potential to describe the Coulomb interaction in a non-uniform dielectric medium [1].

Next, the optical activity, the magnetic brightening and polarization properties of the so called dark excitons (pairs of electrons and holes with opposite spin) in tungsten based S-TMD monolayers will be presented. The scheme of probing and manipulating the valley degree of freedom of dark excitons will be discussed [2].

Finally, the fine structure of K-excitons in S-TMD multilayers will be examined with the results of experimental and theoretical studies of bi- and tri-layer MoS₂. Particular attention will be focused on properties of inter-layer excitons which appear in S-TMD multilayers, in addition to more conventional intra-layer excitons [3]

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Spin-Polarized Electrons in Monolayer MoS₂

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We present experimental evidence for a spin-polarized electronic ground-state in a two-dimensional semiconductor, monolayer MoS₂ [1].

In a two-dimensional electron gas (2DEG), Coulomb effects dominate over single-particle effects in the limit of low electron-density. In this regime, the average inter-electron distance is larger than the Bohr radius in the host material. In gallium arsenide and silicon 2DEGs, electrons are localized at these low electron densities such that Coulomb effects tend to be obscured. New opportunities arise in transition-metal dichalcogenides (TMDs). A TMD represents a natural realization of a 2DEG. Significantly, the extremely small TMD Bohr-radius suggests that Coulomb effects play an important role at experimentally relevant electron-densities.

We investigate the optical absorption of a monolayer MoS₂, a member of the optically active TMD-family. The band edges are located at the K and K' points of the Brillouin zone. At realistic electron densities, there are four relevant conduction bands [2,3]: two at the K point and two at the K' point. The two bands at K, equivalently the two bands at K', carry opposite spin and are split by the spin-orbit interaction. MoS₂ is special among the TMDs in that the conduction-band spin-orbit splitting is small compared to the typical Fermi levels achieved by gating. In a single-particle picture, the electronic ground-state is formed by a near-equal filling of the four available conduction bands. We present an experiment that overturns this single-particle picture [1]. In a magnetic field applied perpendicular to the 2DEG, we find that the electrons have the same spin. Two of the four bands are occupied – the two with the same spin but at different valleys. This is a striking result: the creation of a spontaneous spin-polarization in a 2DEG.

We probe the electronic ground-state at various electron densities using optical absorption, a local, non-invasive, spin- and valley-resolved tool. At the smallest electron densities, our gated device gives a close-to-ideal exciton linewidth. On increasing the electron concentration, we observe a strong exciton-Fermi sea interaction. Specifically, we observe two low-energy trion-resonances for photons with one polarization, yet only very weak features in the other polarization. We interpret the spectra with the well-established exciton-polaron picture of optical absorption in a 2DEG [4-6]. We are led to the conclusion that the electrons are spin-polarized.

We propose that inter-valley electron-electron exchange is responsible for the spontaneous creation of a spin-polarized ground-state in electron-doped monolayer MoS₂. Even though the K and K' points are far apart in phase-space, the Bohr radius is so small that inter-valley Coulomb scattering is significant [7]. The Mermin-Wagner theorem prohibits a spin polarization for isotropic spins. This suggests that the small spin-orbit interaction is crucial to the explanation.

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Unveiling the Defect Structure of Localized Excitons in WSe₂ Monolayer

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Abstract:

Understanding and controlling defects in crystalline materials is an everlasting theme in material science and engineering. For two dimensional materials at atomic limit, defects are more influential and have come to the forefront in the pursuit of electronic and optoelectronic applications. In particular, intrinsic defects in semiconducting transition metal dichalcogenide (TMD) monolayers lower the carrier mobility and photoluminescence quantum yield despite of their direct semiconducting bandgap. Furthermore, defects in WSe₂ monolayer host localized excitons that could behave as single quantum emitters (SQEs) at low temperature. However, the exact nature of these defects remains elusive. In this talk, I will introduce our recent progress in study of the point defect in TMDs. By using low temperature scanning tunneling microscopy and spectroscopy, in corroboration with density functional theory calculations, we unveiled the atomic structure of the most abundant defects in WSe₂ monolayer grown by chemical vapor deposition. Instead of chalcogen vacancies that prevail in other TMD materials, such defects in WSe₂ arise from single tungsten vacancies, leading to the hole (p-type) doping and modifying electronic structure locally (~1.5 nm). Moreover, we found these defects to dominate the optical emission of WSe₂ monolayer at low temperature, providing the first real-space peek into bound excitons. In the end of the talk, I will also discuss the relationship between the bound exciton trapped by the tungsten vacancy and the SQEs. We conjecture that the tungsten vacancy defect is a likely candidate, at least a precursor, for SQEs in the WSe₂ monolayer.

Quantum spin Hall phase versus quantized spin Hall conductivity?

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The quantum spin Hall (QSH) phase is a quantum state of matter, proposed to exist in two-dimensional (2D) semiconductors with inverted band structure, i.e., topological insulators (TIs). Such a quantum phase should exhibit a spin Hall (SH) conductance while the charge Hall conductance vanishes. Since no external magnetic field is present, it is due to the (strong) spin-orbit interaction (SOI). As central questions we investigate if such a QSH phase is indeed related to a quantized static SH conductivity, as indicated by the characterization as quantum phase, and how the answer depends on translational and point-group symmetry.

As central quantities we study the SH conductivity $\sigma_{xy}^z(\omega)$ via the Kubo formula [1] and the Z_2 topological invariant via the parity or Wannier charge center method [2,3] by means of *ab initio* calculations of relativistic electronic structures of infinite 2D sheet crystals. The dynamical quantity is illustrated for atomically thin germanene-based systems [4]. The third-rank tensor of the static SH conductivity is computed for three $Z_2=1$ classes: Hexagonal honeycomb atomic layers made by group-IV elements, germanene (Ge) and stanene (Sn), and their chemically functionalized, e.g. hydrogenated (GeH), iodinated (GeI) or fluorinated (SnF), counterparts [4]. Square and rectangular 2D Bravais lattices are investigated for the model systems 1S- and 1T'-MoS₂ as well as -WS₂ [5]. The influence of destroying the inversion symmetry, e.g. by vertical electric fields, is also studied.

The inverted band structures with a fundamental gap due to SOI are strongly modified by atomic functionalization. As a consequence, we observe drastic changes in the frequency dependence of $\sigma_{xy}^z(\omega)$. For hexagonal 2D TIs the static SH conductivity $\sigma_{xy}^z(0)$ for z-spin orientation exhibits quantization with e^2/h , the reciprocal von Klitzing constant, as shown in Fig. 1 [4,5]. This value is hardly influenced by temperature or Fermi level position. While for 2D square 1S crystals the quantization, clearly observable for hexagonal TIs, is less influenced, the conductivity value tends to zero for rectangular 1T' systems (see Fig. 2). Lifting the inversion symmetry, the quantization is violated also for hexagonal sheets. We conclude that the QSH phase is not generally characterized by a quantized SH conductivity, only in high-symmetric cases. This fact is explained in terms of spin conservation.

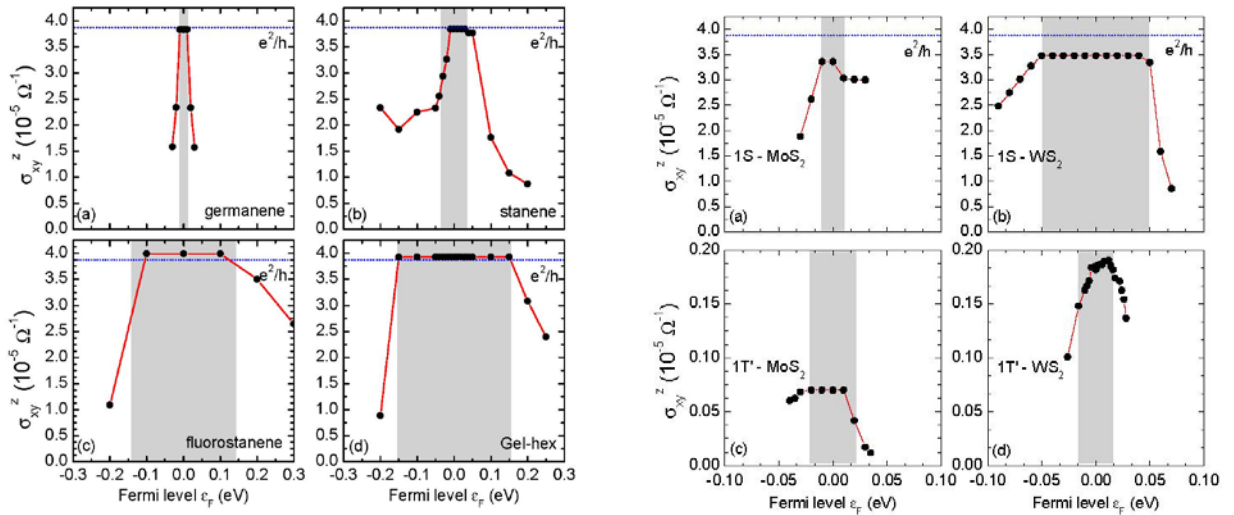


Fig. 1/2: Static spin Hall conductivity versus Fermi energy.

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Biexciton fine structure in monolayer transition metal dichalcogenides

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The optical properties of atomically thin transition metal dichalcogenide (TMDC) semiconductors are shaped by the emergence of correlated many-body complexes due to strong Coulomb interaction. Exceptional electron-hole exchange predestines TMDCs to study fundamental and applied properties of Coulomb complexes such as valley depolarization of excitons and fine-structure splitting of trions.

Biexcitons in these materials are less understood and it has been established only recently that they are spectrally located between exciton and trion. We show that biexcitons in monolayer TMDCs exhibit a distinct and rich fine structure on the order of meV due to electron-hole exchange. [1] Ultrafast pump-probe experiments on monolayer WSe₂ reveal decisive biexciton signatures and a fine structure in excellent agreement with a microscopic theory, as shown in Figure 1. We provide a pathway to understand the complex spectral structure of higher-order Coulomb complexes in TMDCs going beyond the usual classification scheme in terms of four-particle configurations.

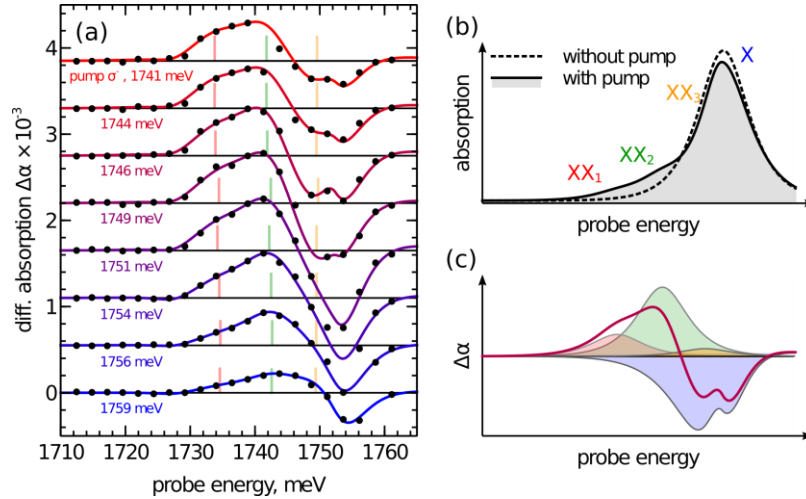


Figure 1: Differential absorption of monolayer WSe₂ on sapphire from theory and experiment. (a) Experimental data at zero time delay (filled black circles) for different pump-pulse energies are compared to fits based on the predictions of our theoretical model. (b) Fit function for absorption spectra without and with pump at 1746 meV. (c) Differential absorption from the fitting curves in (b).

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Theory of ultrafast excitonic intervalley dynamics in TMDs: interplay of dark states, phonons and Coulomb exchange

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In monolayer transition metal dichalcogenides (TMDs), the interplay of spin-orbit interaction and circular dichroism enables valley and spin selective optical excitation of excitons. In this talk, we develop, based on a fully excitonic picture [1], a theoretical description of the excitation, coherent dynamics and relaxation pathways of TMD excitons on ultrashort time scales [2]. The analysis includes the optical preparation of excitons, the subsequent formation of polarons and momentum dark excitons due to exciton-phonon coupling [3,4] as well as excitonic coupling due to Coulomb interaction (exchange interaction, mean-field and biexcitonic states) [1,5]. As experimental observables we address phonon sidebands in absorption, pump-probe signals and photoelectron emission. Some results are illustrated and discussed in Fig. 1:

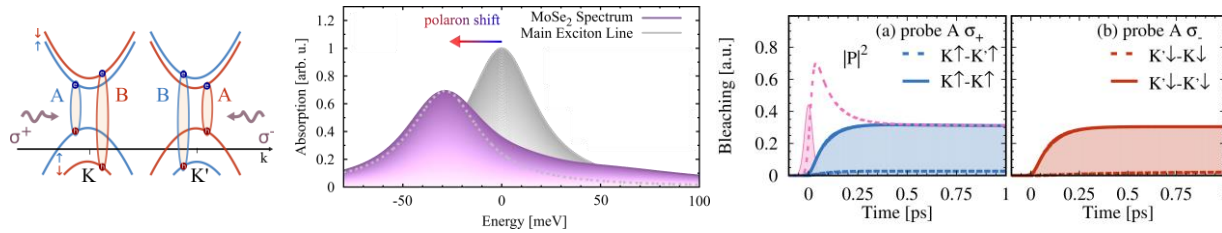


Figure 1: (Left) Illustration of the MoSe₂ band structure at the corners of the Brillouin zone including optical selection rules. (Middle) Non-Markovian linear absorption spectrum of monolayer MoSe₂ (at 300 K) showing phonon sidebands and a clear polaron red-shift (purple) compared to the Markovian limit (grey). (Right) Bleaching of the A σ_+ (a) and A σ_- (b) transition after optically pumping the A σ_+ transition. Despite contributions from direct valley excitons and the excitonic coherence, also momentum space indirect excitons account for the bleaching of both transitions.

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Exciton Fine-Structure in Transition-Metal Dichalcogenides Mono-Layers

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In this paper, we discuss theoretically the exciton fine-structure in transition-metal dichalcogenides (TMD) mono-layers such as MoSe_2 . While bulk MoSe_2 is an indirect-gap material, MoSe_2 mono-layers (which have D_{3h} point-group symmetry) possess a direct energy gap at the $K_{\pm}$ -points of the two-dimensional Brillouin zone. Depending on their coupling to the electromagnetic radiation field optical active (bright) and inactive (dark) exciton states can be formed. Thus, two exciton series (called "A" and "B" series) are observed in optical measurements.

In order to discuss their fine structure, we define excitons in the product space of electron- and hole states, including the lowest conduction band (LCB) and the uppermost valence band (UVB) where, at the Γ -point, they are both spin degenerate. All other states are neglected. On this basis exciton states are constructed and analyzed in the framework of an invariant expansion of a model Hamiltonian: The spin-orbit coupling in the conduction- and valence band is first simulated by introducing an effective magnetic field, giving rise to a splitting of the electron- and hole states outside the Γ -point. Then the Coulomb electron-hole exchange-interaction is introduced into the exciton Hamiltonian. It is due to the fact that electron and hole are indistinguishable particles in the exciton problem. In D_{3h} crystal symmetry this electron-hole exchange-interaction (named "intra-valley exchange") has two different contributions: A first term accounts for an energy re-normalization of the different dark- and bright-exciton states in both series. A second term does not influence the bright states but affects only the dark states of both series, which become mixed. The importance of this mixing depends also on the spin-orbit coupling within the bands. As a result, exchange interaction and spin-orbit coupling of the conduction-band electrons lift together by different amounts the degeneracy of the bright- and dark states.

These calculations (starting from the Γ -point) can be extended up to the $K_{\pm}$ points at the edges of the Brillouin zone. In transition-metal dichalcogenides mono-layers the wave-vector group the $K_{\pm}$ points is C_{3h} . K_+ and K_- points being separated by a reciprocal lattice vector, they are equivalent points of the Brillouin zone. Their corresponding wave-vectors are connected to each other by time-reversal symmetry. In this situation, the exchange interaction introduced above leads also to exchange interaction between electrons and holes from different $K_{\pm}$ valleys. The latter is called "inter-valley exchange interaction". Outside the $K_{\pm}$ points this results into a mixing of exciton states belonging to different valleys and leads to an inter-valley transfer of electron-hole excitation that has been studied e. g. in the framework of $\mathbf{k.p}$ perturbation theory. (See Ref. [1])

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Polariton nonlinear interactions - from GaAs waveguides to transition metal dichalcogenide microcavities.

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Polaritons are the hybrid quasi-particles formed by strong coupling of light to semiconductor optical excitations such as GaAs quantum well excitons or TMD excitons or trions. Their excitonic part provides nonlinear optical interactions many orders of magnitude larger than in weakly-coupled semiconductor devices [1]. In this presentation I will discuss polaritons nonlinear interactions in different systems which, nevertheless, are governed by very similar underlying physics.

In GaAs polariton waveguides we observe dramatic nonlinear-optical effects such as solitons [1,2] and spatio-temporal optical continuum generation [3] at unprecedented low power, opening the door to rich new physics in a convenient chip-scale device. The hybrid photon-exciton nature of polaritons strongly affects nonlinear dynamics compared to those in simple Kerr nonlinear systems.

Meanwhile, strong coupling between TMD flakes and microcavity and waveguide photons have been demonstrated [4,5] so that polariton nonlinearity could be exploited using monolayer active materials. In an open-access microcavity we observe MoSe2 exciton- and trion-polaritons and find that the trion-polariton nonlinearity can be orders of magnitude larger even than in GaAs exciton-polariton systems, paving the way to even lower power nonlinear-optical devices.

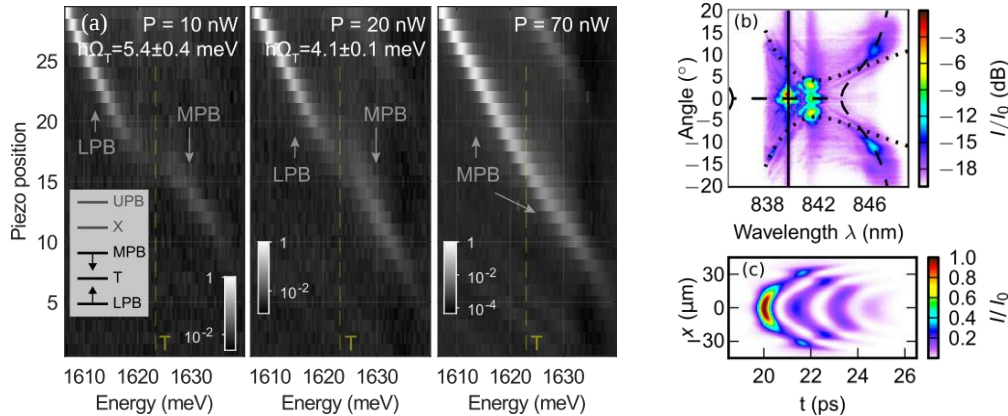


Figure 1(a). Polariton-trion coupling in an MoSe2 flake is quenched resulting in nonlinear shift of the optical frequency at ultra-low power. (b) Broad spatio-temporal continuum generated from a 2 pico-second (ps), 0.22nJ pulse in a GaAs-based polariton waveguide [3]. (c) Simulation of a sub-ps pulse train formed by nonlinear self-modulation of a single 2ps pulse in a GaAs polariton waveguide [3].

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Valley polarization of exciton-polaritons in monolayer WSe₂ in a tunable microcavity

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Excitons in thin layers of semiconducting transition metal dichalcogenides such as MoS₂, WS₂, MoSe₂ and WSe₂ due to their high binding energy and oscillator strength are perfect systems for investigation of light–matter interactions.

In this work we demonstrate a tunable optical cavity incorporating tungsten diselenide (WSe₂) monolayer. Our open cavity consists of two dielectric TiO₂/SiO₂ distributed Bragg reflectors (DBRs). One of them is mounted directly to the piezoelectric chip, which allows for precise control over the intermirror distance, thus energy of the cavity mode. Exfoliated WSe₂ single-layers are placed directly on the surface of the DBR. For optimal coupling the DBRs are specially designed to ensure the antinode of the electric field at the position of the flake. Our setup allows for angle resolved experiments with a control over the cavity mode energy over 100 meV.

We report observation of the strong coupling regime between excitons in WSe₂ and the cavity photons. An angle-resolved reflectance and photoluminescence spectra measured at 5 K are presented in Fig. 1. Both spectra show two anticrossing upper and lower polariton branches with the Rabi splitting of 26 meV and up to 5 splitting-to-linewidth ratio.

In this system we investigated polariton valley polarization with circularly-polarized excitation laser beam resonant with the WSe₂ exciton. Tunability of the photonic mode allowed us to probe a wide spectral range and intersect polariton modes with other excitonic transitions such as dark and charged excitons. We observed that the degree of circular polarization of the emitted light strongly depends on polaritons energy, but is almost constant for different wavevectors. We demonstrate that depending on the intersecting resonance we can retain up to 40% of initial laser polarization or completely lose it.

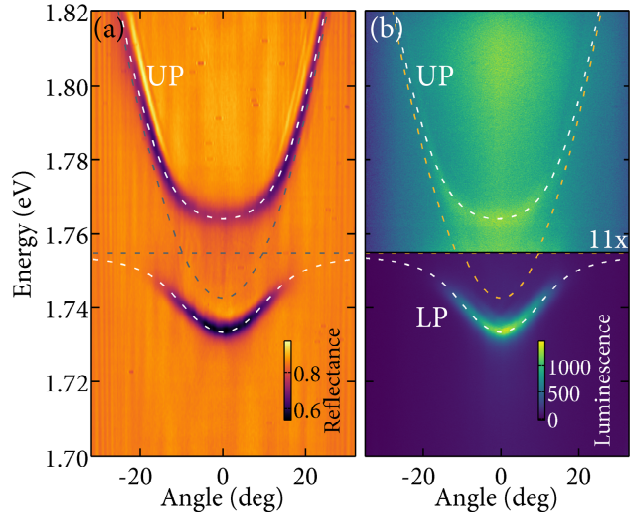


Figure 1: Angle-resolved (a) reflectance and (b) photoluminescence spectra illustrating a strong coupling between excitons in a WSe₂ monolayer and the cavity photons at 5 K.

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Optical Properties and Strong Light-Matter Coupling in 2D Perovskite Crystals

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Research on lead halide semiconductors with perovskite lattices is a rapidly growing field semiconductor physics and optics. They have great potential for low-cost yet efficient solar cells and light-emitting devices. Their optical properties can be tuned by tailoring the chemical composition and/or the nanostructure spatial dimension with very high precision and high quality. Moreover, the hybrid nature and soft lattice of organic-inorganic lead halide perovskites render their structural changes and optical properties susceptible to external driving forces such as temperature and pressure, remarkably different from conventional semiconductors. Herein, we study the optical properties and strong light-matter coupling in high-quality two-dimensional perovskite crystals. We report multiple intrinsic optical transitions originating from the radiative recombination of coupled electron-hole pairs (excitons). An extraordinary fine-structure splitting has been experimentally resolved on these atomically thin semiconductors thanks to the unusual Coulomb interaction. This fine-structure splitting and anisotropic polarized emission are supported by first-principle calculations. The optical spectra exhibit a large tunability of up to 320 meV within a moderate pressure range of below 3.5 GPa, while the quantum-yield remains constant. Such a large tunable range originates from the anisotropic structural deformation that effectively modulates the quantum confinement effect by 250 meV *via* barrier height lowering. Moreover, due to the large binding energy and oscillator strengths, two-dimensional perovskites are ideal candidates for studying strong light-matter interaction. We further successfully demonstrate room-temperature strong coupling in exfoliated 2D perovskite semiconductors embedded into a planar microcavity, exhibiting a large energetic splitting-to-linewidth ratio of 34.2. Our findings advocate a considerable promise of 2D perovskite to explore not only fundamental optical properties but also quantum phenomena such as Bose-Einstein condensation, superfluidity and exciton-polariton networks.

Halide Perovskite Nanocrystals: from platelets to supercrystals

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I will report on the optical properties of metal halide perovskite nano-platelets with controllable thickness down to one monolayer [1]. Pronounced quantum confinement effects, large excitonic binding energies and comparably high radiative recombination rates have been found, all depending on the number of monolayers present in the respective nano-platelets. Ultrafast optical experiments provide further insight into characteristic charge carrier and spin relaxation scenarios in 2D perovskites. Finally, the assembly of halide perovskite nanocrystals into ordered supercrystals leads to remarkable changes of their linear and nonlinear optical properties [2,3].

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Optical spectroscopy of 2D perovskites in high magnetic fields

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

The 2D perovskites are often regarded as perfect quantum wells, because they are not plagued by interfacial roughness or intermixing characteristic of epitaxially grown quantum wells. The well layers consists of planes of lead-halide octahedra, separated by organic spacers. The research of this natural perovskite quantum wells is in infancy stage and their structural, dielectric, optical, and excitonic properties remain to be explored especially influence of organic spacers. Despite bandedge states are composed of lead and halide orbitals organic ligands affects the band structure and electron-phonon coupling via distortion of the octahedral cages. The optical properties of these materials are dominated by strong electron-hole attraction resulting from dielectric confinement. Very often this materials exhibits complex absorption and emission spectrum with many sidebands which origin rises controversy since they can be attributed to phonon replicas or bound excitonic states. In this work we address this issue for $(C_nH_{2n+1}NH_3)_2PbI_4$ (with $n=4,6,8,10,12$) by means of optical spectroscopy in magnetic field up to 67T. We show that complex absorption features shift parallel in magnetic field indicating phonons related nature of the observed side bands. Moreover our studies supported by modeling, unambiguously demonstrate that the reduced mass of exciton change about 30% upon phase transition. This is reflected by 3 time decrease of diamagnetic shifts coefficient for low temperature crystal phase. Since the phase transition occurs close to room temperature our founding can provide additional way for 2D perovskite properties engineering via moderate cooling achievable by Peltier coolers.

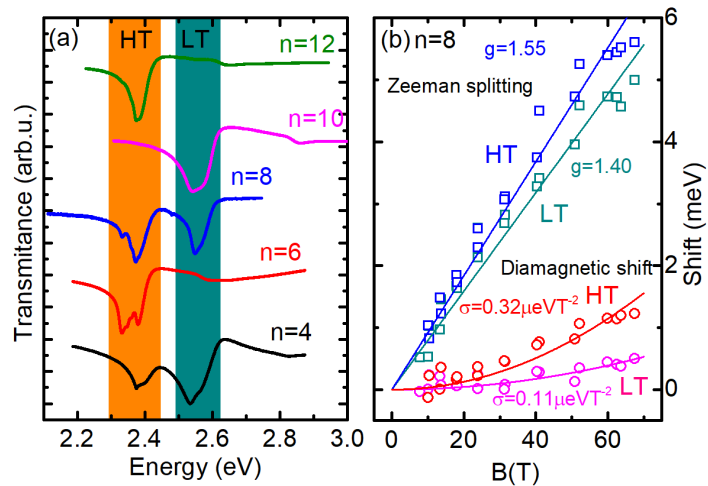


Figure 1(a) Transmittance spectrum of investigated samples at 4K. Low (LT) and frozen high (HT) absorption features can be observed in the spectrum. (b) Zeeman splitting and diamagnetic shift of high and low temperature phase of the crystals with $n=8$.

Many-body optical excitations in solid-state systems

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Two dimensional materials provide new avenues for synthesizing compound quantum systems. Monolayers with vastly different electric, magnetic or optical properties can be combined in van der Waals heterostructures which ensure the emergence of new functionalities; arguably, the most spectacular example to date is the observation of strong correlations and low electron density superconductivity in Moire superlattices obtained by stacking two monolayers with a finite twist angle. Optically active monolayers such as molybdenum diselenide provide a different "twist" as they allow for investigation of nonequilibrium dynamics in van der Waals heterostructures by means of femtosecond pump-probe measurements. Moreover, interactions between electrons and the elementary optical excitations such as excitons or polaritons, provide an ideal platform for investigation of quantum impurity physics, with possibilities to probe both Fermi- and Bose-polaron physics as well as mixtures with tunable density of degenerate fermions and bosons.

After introducing the framework we use to describe many-body optical excitations in van der Waals heterostructures, I will describe two recent developments in the field. The first experiment uses pump-probe measurements to demonstrate how exciton-electron interactions beyond the non-self-consistent T-matrix approximation lead to optical gain by stimulated cooling of exciton-polaron-polaritons. The second experiment shows that a tri-layer system, consisting of two semiconducting monolayers separated by an insulating layer, could lead to hybridization of intra- and inter-layer excitons. The latter has potential applications ranging from strongly interacting polaritons to reaching Feshbach resonance condition in exciton-electron scattering.

CONTROL OF LIGHT-MATTER INTERACTION IN VAN DER WAALS MATERIALS

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Two-dimensional (2D) Van der Waals materials have emerged as a very attractive class of optoelectronic material due to the unprecedented strength in its interaction with light. In this talk I will discuss approaches to enhance and control this interaction by integrating these 2D materials with microcavities. I will first discuss the formation of strongly coupled half-light half-matter quasiparticles (microcavity exciton-polaritons) and their spin-optic and electrical excitation and control in the 2D transition metal dichalcogenide (TMD) systems. Prospects of realizing condensation and few photon nonlinear switches using Rydberg states in TMDs will also be discussed. Finally, I will talk about room temperature single photon emission from hexagonal boron nitride and the prospects of developing deterministic quantum emitters using them.

Probing nanoscale light-matter interactions in atomically thin semiconductors

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Many classes of two dimensional (2D) quantum systems, e.g., transition metal dichalcogenide (TMD) monolayers, have recently emerged as potential candidates for novel optoelectronic and photonic devices. Hence, understanding the physical properties and light-matter interactions of 2D quantum systems are of fundamental significance. In this talk, I will introduce a hybrid nano-opto-mechanical tip-enhanced spectroscopy and imaging approach combining tip-enhanced Raman scattering (TERS) and tip-enhanced photoluminescence (TEPL) to probe the heterogeneous optical responses of TMD monolayer at nanoscale defects and control the local bandgap of crystal through atomic force local strain manipulation as shown in Fig. 1(a) [1]. This approach is further extended to probe and control the radiative emission of dark excitons [2] and localized excitons [3]. Based on nano-tip enhanced spectroscopy with $\sim 600,000$ -fold PL enhancement induced by the plasmonic Purcell effect and few-fs radiative dynamics of the optical antenna tip, we can directly probe and actively modulate the dark exciton and localized exciton emissions in time ($\sim$ ms) and space (<15 nm) at room temperature as shown in Fig. 1(b-c).

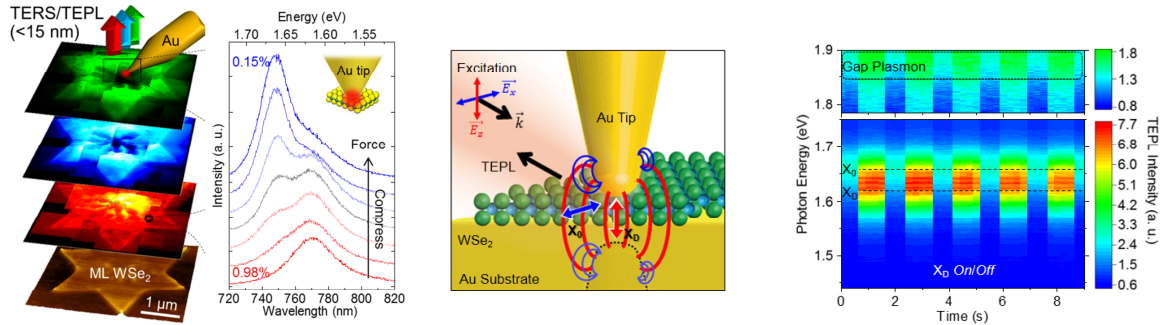


Figure 1: (a) Illustration of a hybrid nano-opto-mechanical tip-enhanced spectroscopy and imaging approach. (b) Schematic of tip-enhanced photoluminescence spectroscopy. Selective excitation and probing of the transition dipole moments of dark (out-of-plane) and bright (in-plane) excitons by polarization control. (c) Time-series TEPL response (X_D , X_0 and gap plasmon PL) in discrete On/Off switching of X_D emission with discrete tip-sample distance variation between 1 nm and 5 nm.

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Valleytronics in the moiré of 2D semiconductors

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ABSTRACT

Van der Waals stacking of the 2D semiconductors into vertical layered structures is a powerful approach towards designer quantum materials that can combine and extend the exotic properties of the building blocks. Ubiquitous to these vdW heterostructures is the formation of moiré pattern due to the inevitable lattice mismatch and twisting between the layers. In 2D semiconducting transition metal dichalcogenides where the band edge carriers are described by valley-spin locked massive Dirac fermions, we show that the moiré offers unprecedented opportunities for engineering magnetic and topological properties for valley-spintronic controls in heterobilayer [1,2] and homobilayer [3] semiconductors.

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Cavity and magnetic field control of interlayer excitons in van der Waals heterobilayers

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Transition metal dichalcogenides (TMDs) have emerged as novel two-dimensional semiconductors with a direct band gap in their monolayer limit¹ and remarkable opto-valleytronic properties². While extensive research has been dedicated to TMD monolayers in the past decade, more recent work has focused on novel aspects provided by combinations of dissimilar monolayers in stacks of van der Waals heterobilayers. Vertical MoSe₂-WSe₂ heterobilayers, for example, host layer separated excitons with finite dipole moments and long radiative lifetimes³ that can be subjected to moire effects in twisted heterobilayer systems⁴.

In our experiments, we studied the cryogenic photophysics of excitons in as-grown moire-free MoSe₂-WSe₂ heterobilayers in cavity-modified photonic environments and in response to external magnetic field. We demonstrate cavity-control of the radiative decay channels associated with different types of interlayer excitons, and quantify the respective light-matter coupling strengths⁵. Moreover, we discuss the distinct roles of bright and dark interlayer excitons for the dynamics of photoexcited spin orientation in the presence of external magnetic fields⁶. The findings will be compared with observations made on exfoliation-stacked MoSe₂-WSe₂ heterostructures with different orientation angles.

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Kerr rotation study of indirect excitons in $\text{WSe}_2 / \text{Mo}_{0.5}\text{W}_{0.5}\text{Se}_2$ heterostructures

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Van der Waals heterostructures based on transition metal dichalcogenide monolayers are new promising objects of research in semiconductor physics. By choosing the material of semiconductor layers, the mutual orientation of the crystallographic axes of different layers and the distances between them, the optical transition energy and the lifetime of photoexcited states can be varied over a wide range, which allows engineering of various photoelectric devices based on them. The complex structure of spin sublevels associated with the presence of several degenerate valleys and strong spin-valley interaction requires separate study of the dynamics of the spin degrees of freedom of charge carriers in such heterostructures.

In this work, the spin relaxation of excitons in a heterostructure based on $\text{WSe}_2 / \text{Mo}_{0.5}\text{W}_{0.5}\text{Se}_2$ monolayers was studied using the method of micro-Kerr rotation. A strong difference in the spin relaxation time of photogenerated excitons between the WSe_2 monolayer and the heterostructure was found. We conclude that such an increase in the spin relaxation time in the heterostructure is associated with the excitation of an indirect exciton, when the electron and hole are localized in different layers of the heterostructure. Since the decay rate of the signal of Kerr rotation is determined by the sum of the recombination and spin relaxation rates, this means that the one and the other decreases tenfold. The change in the recombination rate is due to the spatial separation of electrons and holes, and the decrease in the spin relaxation rate is due to the stabilization of the hole spin by a strong spin-orbit interaction. The rate of depolarization of the electron spins does not change in this case, which manifests itself as a rapid decrease in the signal on the scale of the first picoseconds.

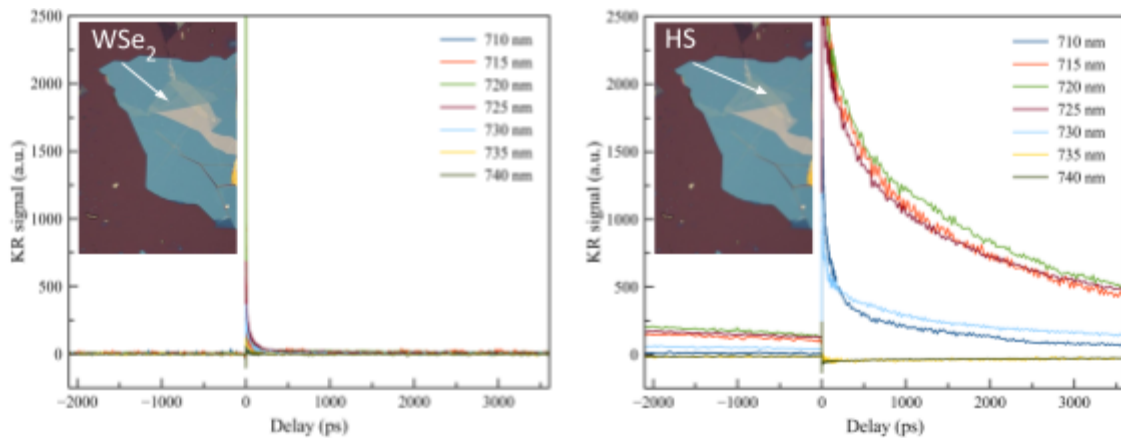


Figure 1: Kerr rotation signal as a function of excitation wavelength measured on single layer of WSe_2 (left) and $\text{WSe}_2 / \text{Mo}_{0.5}\text{W}_{0.5}\text{Se}_2$ heterostructure (right).

Internal structure and ultrafast dynamics of interlayer excitons in van der Waals hetero-bilayers

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In heterostructures of transition metal dichalcogenides, electrons and holes residing in adjacent monolayers can bind into spatially indirect excitons¹. These interlayer bound pairs have attracted tremendous interest due to their strong promise for novel optoelectronics², valleytronics³ and moiré-induced nanodot lattices⁴. Since these quasiparticles couple only weakly to light, their binding energies have not been directly measured yet. Here we introduce a direct ultrafast access to Coulomb correlations acting between monolayers. For the prototypical case of WSe₂/WS₂ hetero-bilayers, we optically excite 1s A excitons in the WSe₂ monolayer (Fig. 1a, orange pulse). Phase-locked mid-infrared (MIR) pulses (Fig. 1a, red waveform) subsequently interrogate characteristic intra-excitonic transitions independently of the center-of-mass momentum of the excitons or interband optical selection rules^{5,6}. Our measurements reveal a novel transition⁷ between 1s and 2p orbitals of interlayer excitons at an energy of 67 ± 6 meV (Fig. 1b, upper panel). This value coincides with our numerical solution of the Wannier equation (Fig. 1c) and implies a binding energy⁷ of itinerant interlayer excitons as 126 ± 7 meV. Since the complex-valued spectral response functions of inter- and intralayer excitons differ strongly (Fig. 1b), we can sensitively track the ultrafast evolution of the respective exciton densities (Fig. 1d) by monitoring the MIR response as a function of the pump-probe delay time t_{pp} . Interestingly, intralayer excitons photoinjected into the WSe₂ monolayer transform into interlayer species by direct electron tunnelling without a strong intermediate phase of unbound electron-hole pairs. Depending on the stacking angle of the individual monolayers, intra- and interlayer species coexist on picosecond scales and relax into quantum confined states in moiré-induced nanodots.

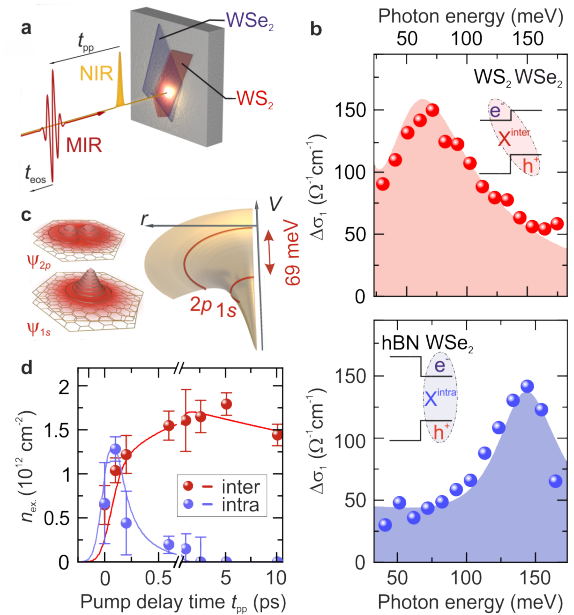


Fig. 1: **a**, Time-resolved near-infrared (NIR) pump – mid-infrared (MIR) probe spectroscopy of a WSe₂/WS₂ heterostructure on a diamond substrate. The NIR pump pulse resonantly injects 1s A excitons in the WSe₂ monolayer, whereas the MIR probe pulse samples the dielectric response of the heterostructure. **b**, Pump-induced change of the real part of the optical conductivity $\Delta\sigma_1$ revealing 1s-2p transitions of interlayer excitons (upper panel) and intralayer excitons (lower panel). Insets: band alignments of the heterostructures. **c**, Interlayer Coulomb potential and wavefunctions of the 1s and 2p interlayer excitons in real space. **d**, Intra- and interlayer exciton densities as a function of pump-probe delay time t_{pp} for a WSe₂/WS₂ heterostructure with a twist angle of $\theta = 5^\circ$. Solid lines: results of the microscopic theory.

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generation and memory of a temporal pulse train in a polariton condensate

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Exciton polaritons in semiconductor microcavity constitute the archetypal system to investigate condensate in solid state materials with room temperature. Together with the consideration of microscale size, polariton condensates appears promising for polaritonic devices for all-optical information and communication processing. To implement these quantum technologies, quantum memory to store and recall the quantum state on demand is one of the most important and challenging issues. In polariton condensates, however, due to the inherently dissipative property and non-equilibrium character, only several researches [1,2] around the manipulation of solitons and vortices are explored. With the investigation of temporal coherence [3,4] in polariton condensate, the study of polariton memory appears possible but remains still unaddressed.

Here we address the quantum memory based on temporal pulse train of phase locking in microcavity polariton condensate. Two significant outcomes are: (a) by firstly exploiting the quantum interference in time-frequency domain, which has been reported only in spatial domain in prior works [5,6], we generate the temporal pulse train with tunable periodicity around several picoseconds in polariton condensate. The results are helpful not only for the signal storage mentioned following but also for the study of polariton laser. (b) Based on the temporal pulse train of π jump phase, we implement the memory with high efficiency and fidelity in polariton condensate, which is checked to be independent of the material parameters and experimentally accessible within a quasi-1D microcavity. It allows the very first observation of quantum memory in a polariton condensate.

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Strong coupling of excitons in MoSe₂ and optical bound states in the continuum

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Optical bound states in the continuum (BICs) are peculiar resonances that can exist in photonic crystals. Characterized by diverging giant Q-factors and field enhancement due to their non-radiative character, as well as formation of momentum-space vortices, BICs are promising candidates for applications in sensing, lasing, and nonlinear optical devices [1]. As planar photonic crystals can be interfaced with 2D transition metal dichalcogenides (TMDs) [2], BICs can be efficiently coupled to TMD excitons creating polaritons.

We study experimentally and theoretically exciton-polaritons formed in the strong coupling regime between TMD excitons and BICs [3]. Our hybrid structures (Fig. 1a) consist of Ta₂O₅/SiO₂/Si photonic crystal slabs supporting BIC resonances at Γ point in the momentum space (Fig. 1b), hBN spacer layer, and monolayer MoSe₂.

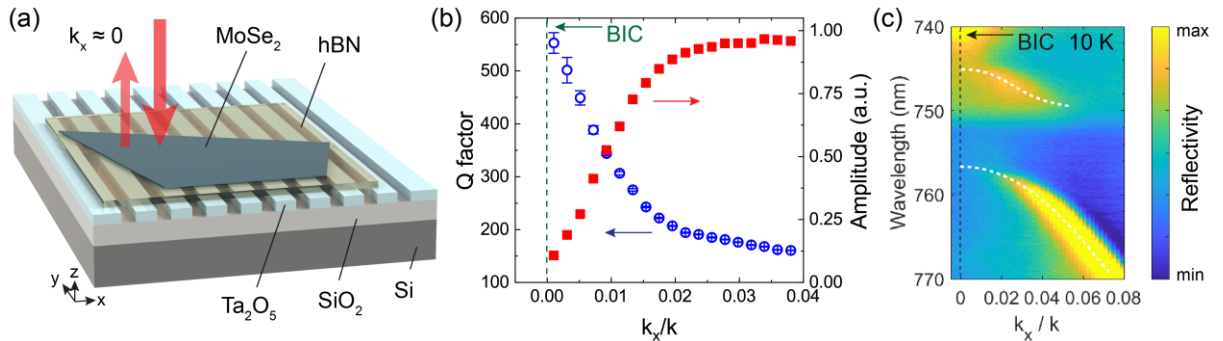


Figure 1: (a) schematic of hybrid structure for strong coupling of MoSe₂ excitons and optical bound states in the continuum, (b) amplitude and Q-factor of the photonic crystal mode, (c) resulting polariton dispersion.

We observe the strong coupling regime via angle-resolved reflectivity (Fig. 1c) and photoluminescence measurements, with Rabi splitting on the order of 20 meV at resonance between MoSe₂ neutral exciton and BIC. Through temperature- and power-dependent measurements we investigate and control the coupling strength, exciton-polariton lifetime, and nonlinear polaritonic interaction. Our results open ways to create and manipulate exciton polaritons with potentially long and tunable lifetimes in hybrid planar structures with straightforward electrical control through gating. Further, strong coupling of BICs and TMD excitons can enable polariton lasers with momentum-space vortex structure.

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Hybrid polariton condensation in a MoSe₂-GaAs Tamm device

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We present the design and realization of a hybrid MoSe₂-GaAs Tamm structure operating in the strong coupling regime (Fig1a). This device combines the unique physics inherent to transition metal dichalcogenide (TMDC) monolayers (e.g. spin-valley locking) with the mature III-V device platform in optoelectronics and polaritonics. We observe collective strong coupling of the different kinds of excitons (Wannier type excitons in GaAs and strongly bound valley excitons in the MoSe₂ monolayer) to the same cavity mode and the formation of hybrid polaritons with their three characteristic dispersion branches (Fig1b) via angle resolved PL and reflection measurements [1]. That manifests the first successful observation of this new kind of quasiparticle.

Furthermore, we observe bosonic condensation in this device driven by the excitons hosted in the atomically thin layer of MoSe₂, visualized via power dependent PL measurements at T = 4.2 K (Fig1c). Additionally we demonstrate that the effects of spin-valley locking are conserved in the condensate [2]. Our work paves the way towards highly efficient, ultra-compact polariton-based light sources and valleytronic devices hosting bosonic quantum fluids in atomically thin materials which we hope can be ultimately operated at room temperature.

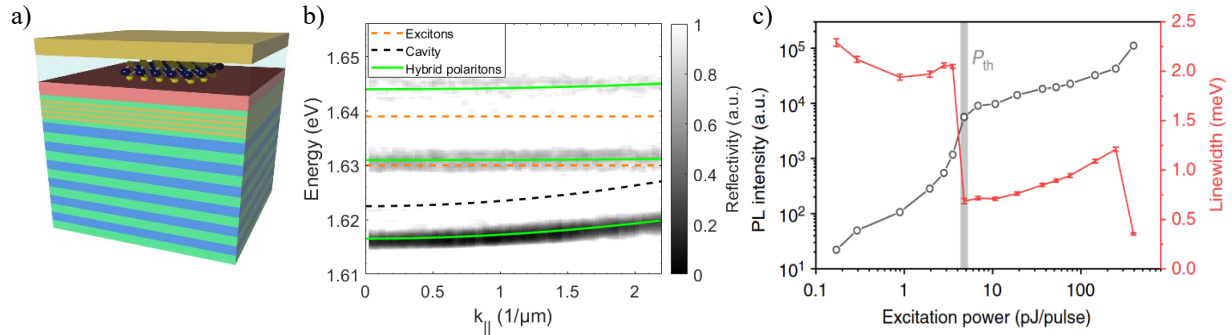


Figure 1: a) Schematic illustration of the hybrid structure with an integrated MoSe₂-monolayer and GaAs QWs. b) Angle-resolved reflectivity measurement on the device at T = 140 K. c) Intensity (black) and linewidth (red) of the hybrid polariton PL at $k_{||} = 0$ as a function of the excitation power at T = 4.2 K.

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Controlling the optical properties of van der Waals heterostructures by interlayer rotation

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The absence of strict lattice matching requirement in van der Waals heterostructures allows for coupling between monolayer crystals with incommensurate lattices and arbitrary mutual orientation. The introduction of rotational misalignment has been shown to modify mechanical and electronic coupling between the layers¹, as well as give rise to long-range periodic variation of the local atomic registry known as moiré superlattice. The latter has attracted considerable research interest in recent years due to its ability to significantly alter the energy spectrum of heterostructures, leading to the formation of topological, superconducting, and quantum-confined states²⁻⁴.

We demonstrate that rotational alignment can be used to efficiently and systematically control the optical properties of van der Waals heterostructures built of incommensurate transition metal dichalcogenide monolayers. We investigate the effects of interlayer rotation by studying a large number of MoSe₂/WS₂ and MoSe₂/MoS₂ heterobilayers with various interlayer orientations, fabricated from either CVD-grown or mechanically exfoliated monolayers. We show that in MoSe₂/WS₂ heterostructures, the near-degeneracy of the conduction band edges leads to twist-angle-dependent hybridization between intra- and interlayer excitons, which manifests as a gradual redshift of the photoluminescence peaks of the hybridized exciton states of up to 60 meV⁵. For heterostructures in which the monolayer pairs are nearly aligned, resonant mixing of the electron states leads to pronounced effects of the geometrical moiré pattern of the heterostructure on the dispersion and optical spectra of the hybridized excitons. While in MoSe₂/MoS₂ heterobilayers the hybridization between the intra- and interlayer exciton states is suppressed by the large band offset, the mutual layer orientation strongly affects the properties of interlayer exciton, leading to 100 meV variation of its emission energy. Our findings underpin new strategies for band structure engineering in semiconductor devices based on van der Waals heterostructures.

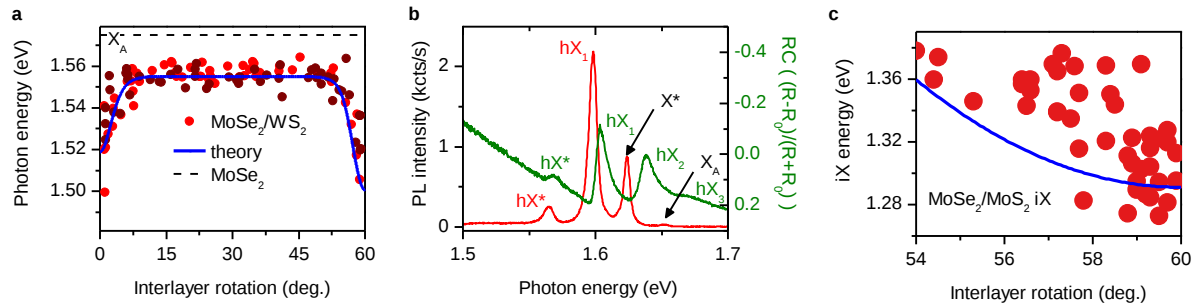


Figure 1 (a) Variation of the hybridised exciton photoluminescence energy with twist angle, measured at room temperature from two substrates containing MoSe₂/WS₂ heterobilayers composed of CVD-grown monolayers (b) Low-temperature photoluminescence (red) and reflectance contrast (green) spectra of an aligned MoSe₂/WS₂ heterobilayer, fabricated from mechanically exfoliated monolayers and fully encapsulated in hBN. The reflectance contrast spectrum shows four features originating from the optically-bright moiré miniband states of hybridized excitons. (c) Variation of the interlayer exciton emission energy in MoSe₂/MoS₂ heterostructures with mutual layer orientation approaching anti-alignment.

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Controlled micro/nano-dome formation in proton-irradiated bulk transition-metal dichalcogenides

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At the few-atom-thick limit, transition-metal dichalcogenides (TMDs) exhibit strongly interconnected structural and optoelectronic properties. The possibility to tailor the latter by controlling the former is guaranteed to have a great impact. Here, we present a route toward the patterning of TMDs based on the effects of proton irradiation on the structural and electronic properties of bulk WS₂, WSe₂, WTe₂, MoS₂, MoSe₂ and MoTe₂. Suitable irradiation conditions let protons penetrate the top layer and trigger the hydrogen evolution reaction just beneath the first chalcogen-metal-chalcogen basal plane. This results in the blistering of one-monolayer thick domes, which stud the crystal surface and locally turn the dark bulk material into an efficient light emitter. These stable (>2-year lifetime), robust domes host strong, non-trivial strain fields that modify profoundly the electronic band structure of the curved monolayer planes. The domes can be produced with well-ordered positions and sizes tunable from the nanometer to the micrometer scale, with important prospects in nanomechanics, pressure sensing and nanophotonics, as well as for the strain engineering of two-dimensional materials.

Control of the exciton radiative lifetime in van der Waals heterostructures

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Optical properties of atomically thin transition metal dichalcogenides are controlled by robust excitons characterized by a very large oscillator strength [1,2,3].

Encapsulation of monolayers such as MoSe₂ in hexagonal boron nitride (hBN) yields narrow optical transitions approaching the homogeneous exciton linewidth [4,5]. We demonstrate that the exciton radiative rate in these van der Waals heterostructures can be tailored by a simple change of the hBN encapsulation layer thickness as a consequence of the Purcell effect [6].

The time-resolved photoluminescence measurements together with cw reflectivity and photoluminescence experiments show that the neutral exciton spontaneous emission time can be tuned by one order of magnitude depending on the thickness of the surrounding hBN layers. The inhibition of the radiative recombination can yield spontaneous emission time up to 10 ps. These results are in very good agreement with the calculated recombination rate in the weak exciton-photon coupling regime. The analysis shows that we are also able to observe a sizeable enhancement of the exciton radiative decay rate.

Understanding the role of these electrodynamical effects allow us to elucidate the complex dynamics of relaxation and recombination for both neutral and charged excitons.

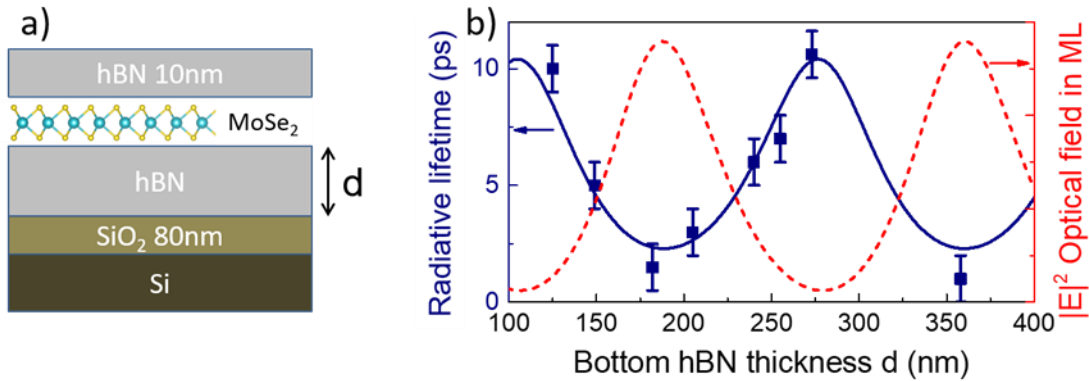


Figure 1: a) Sketch of a MoSe₂ monolayer encapsulated into hBN layers and deposited on a SiO₂/Si substrate. b) Measured radiative lifetime of the neutral exciton (square points) as a function of the bottom hBN thickness. Short (long) lifetimes are reported when the monolayer is located at antinode (node) of the optical field.

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Hybridized indirect excitons in MoS₂/WS₂ heterobilayers

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

The work is supported by the Deutsche Forschungsgemeinschaft (DFG) via excellence clusters NIM and eConversion as well as DFG projects WU 637/4-1 and HO3324/9-1.

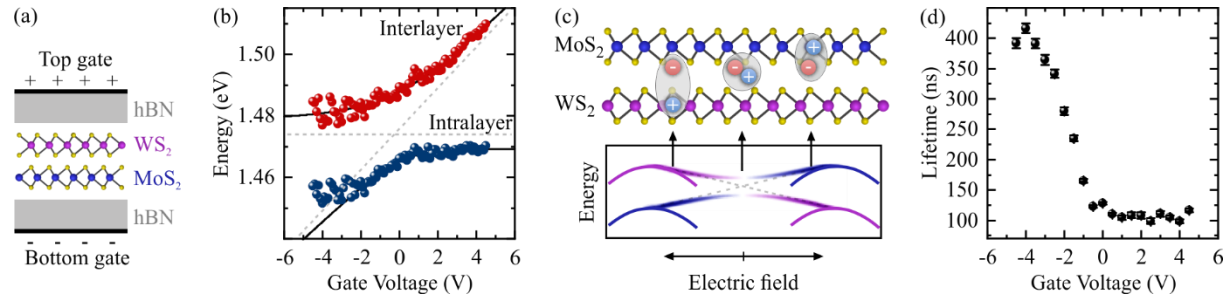


Fig.1: (a) Schematic depiction of the MoS₂/WS₂ heterobilayer field effect structure. (b) Field dependent level anti-crossing of the energetically lowest interband transitions from the Σ -point to the K-point at a bath temperature of 10K. (c) Proposed model for the level repulsion. The hole at the K-point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

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Poster Abstracts

Comparison of density functional theory methods in layered MS₂ compounds under compression

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We report high-pressure Raman-scattering measurements on the transition-metal dichalcogenide (TMDC) compound HfS₂. The aim of this work is twofold: (i) to investigate the high-pressure behavior of the zone-center optical phonon modes of HfS₂ and experimentally determine the linear pressure coefficients and mode Grüneisen parameters of this material; (ii) to test the validity of different density functional theory (DFT) approaches in order to predict the lattice-dynamical properties of HfS₂ under pressure. For this purpose, the experimental results are compared with the results of DFT calculations performed with different functionals, with and without van der Waals (vdW) interaction corrections. We find that DFT calculations within the generalized gradient approximation (GGA) properly describe the high pressure lattice dynamics of HfS₂ when vdW interactions are taken into account. In contrast, we show that DFT within the local density approximation (LDA), which is widely used to predict structural and vibrational properties at ambient conditions in 2D compounds, fails to reproduce the behavior of HfS₂ under compression. Similar conclusions are reached in the case of MoS₂. This suggests that large errors may be introduced if the compressibility and Grüneisen parameters of bulk TMDCs are calculated with LDA. Therefore, the validity of different approaches to calculate the structural and vibrational properties of bulk and few-layered vdW materials under compression should be carefully assessed. [1]

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Strain-induced localization of interlayer excitons in a van-der-Waals heterostructure

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Interlayer excitons (IX) in van-der-Waals heterostructures are currently subject to intense study [1-4]. For example, they constitute an interacting Bosonic system in the solid state whose properties can be widely tuned by electric fields [1], the choice of materials and the microscopic separation and rotational alignment of the individual layers. [2-4]. In this contribution we use local strain to localize IX in a WSe₂-MoSe₂-heterobilayer. Hereby, we place it onto a dielectric substrate patterned with arrays of 130 nm (width) x 90 nm (height) micropillars using viscoelastic stamping. Confocal luminescence measurements performed at low temperature (10 K) reveal clear IX emission around 1.37 eV, that blueshifts by ~30 meV upon increasing the excitation level over six orders of magnitude from 0.03 W/cm² to 50 kW/cm². In contrast, at the pillar sites the IX emission for the lowest excitation levels studied is accompanied by sharp line emission ~50 to 100 meV below the IX and attributed to localized IX (denoted LIX on fig 1). Upon increasing the pumping level, additional emission peaks emerge in the vicinity of LIX. We identify these sharp emission features as arising from multiple, interacting IX localized within a local strain potential by excitation-power-dependent experiments (fig. 2). Our results provide information about exciton-exciton interactions within the strain potential.

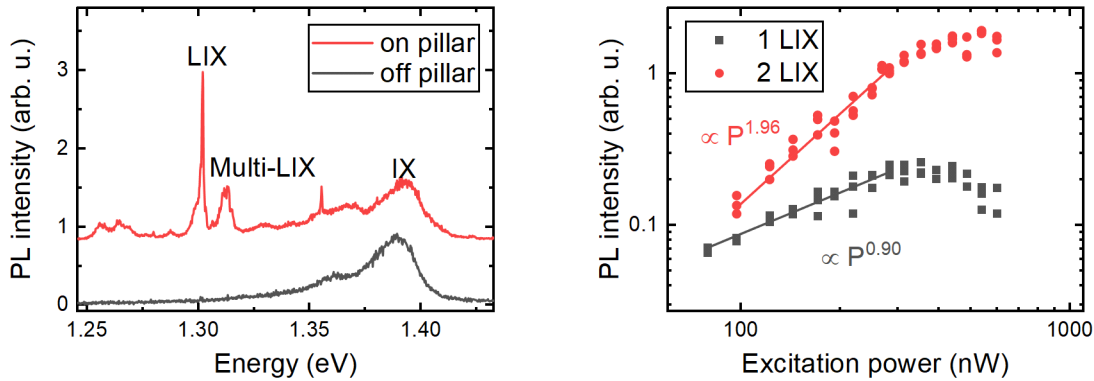


Figure 1: Left: Low-temperature PL spectrum of a strained WSe₂-MoSe₂-heterobilayer at a pillar site. The spectrum features the characteristic IX emission at 1.39 eV (also present off pillar) and strongly red-detuned emission from localized interlayer excitons (LIX). Right: PL intensities of the localized single and double interlayer exciton as a function of excitation power. Before saturation, the characteristic linear and quadratic power dependence are observable.

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2D metasurfaces based on the atomic carbon wires: the carbynes

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We synthesize macroscopically long linear carbon chains (carbynes) in a colloidal solution, then deposit them on a surface. The method is based on the formation of carbon threads by laser ablation in a colloid accompanied by the stabilization of resulting linear carbon chains by golden nanoparticles. We observe the signatures of the polyynic allotrope of carbyne in the photoluminescence and Raman spectra of the solutions. We deposit the synthesized carbyne threads on fused quartz glass substrates. The transmission electron microscopy images of the deposited threads with golden nanoparticles attached to their ends demonstrate the straight linear parts of the monoatomic carbon chains varying in length between 8 and 24 carbon atoms. This method paves the way to fabrication of an ultimate one-dimensional crystal: a monoatomic carbon wire.

Till now, stable freestanding samples of straight carbon chains have not been realised, to the best of our knowledge. Their synthesis appears to be a formidable challenge as, in general, infinite one-dimensional atomic chains are unstable in vacuum. Fluctuations prevent formation of ideal one-dimensional crystals, according to the Landau theorem [1]. In this context, many works have been devoted to the artificial stabilization of carbyne with use of heavy anchor atomic groups [2], confinement of carbon chains in double-wall carbon nanotubes [3], their pinning to metallic surfaces etc. The synthesis of free-standing carbon chains remains one of the Holy Graals of nano-physics and nano-chemistry. If realized these one-dimensional crystals would exhibit unique mechanical, optical and electronic properties [4].

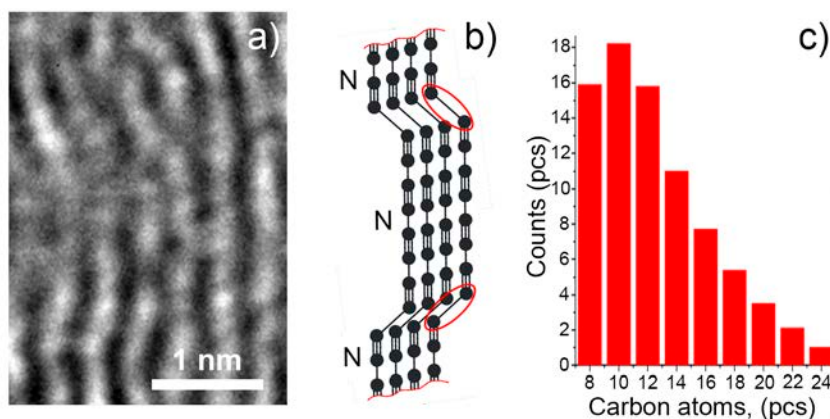


Figure 1: Deposited chains investigation. a) TEM image of the studied carbyne film. b) shows schematically the atomic structure of deposited carbon chains, c) shows the extracted length distribution of the deposited carbon chains.

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Abrupt changes in the Raman spectra of MnPS₃ below Néel temperature

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The world of 2D materials is rich in materials that are characterised with diverse physical properties. Ample phenomena, well-known in condensed matter physics of bulk specimen, have been thoroughly investigated in 2D limit, often uncovering features arising explicitly due to reduced dimensionality. Until recently, emergence of magnetic order in atomically thin films was questioned and constituted a strikingly missing aspect. Due to the progress in sample fabrication technology, it is currently clear that many materials preserve the magnetic order even in monolayer form [1, 2]. This finding empowered researchers with new routes to develop the fundamental understanding of 2D magnetism from the ground up.

One of the simple, yet impactful, observations is that the appearance of magnetic order below a critical temperature may influence the strength of inter-atomic bonds, which determine the structure of the crystal. A trivial consequence of that is the renormalisation of the fundamental lattice parameters, such as lattice constant and interlayer spacing. However, more intricate scenarios have been considered, including a reconfiguration of the atoms arrangement into a new crystallographic structure [3]. Here, we will investigate these notions based on the measurements of Raman spectra of bulk MnPS₃. The photon scattering processes involving emission of a phonon and/or a magnon provide a direct insight into the adaptation of the crystal structure to the emergence of magnetic interactions and appearance of novel low-energy excitations. MnPS₃ is a material that displays antiferromagnetic in-plane coupling between magnetic moments of neighboring Mn²⁺ ions with Néel temperature of 78 K [4]. As shown in **Figure 1**, the abrupt changes in the Raman spectra are observed around the critical temperature. The colour map demonstrates a temperature evolution of a resonance in the scattering spectrum, whose energy and line shape are starkly different in the nonmagnetic and the magnetic state of the sample.

The systematic optical study of MnPS₃ crystals will be demonstrated with the emphasis on Raman scattering. The tentative interpretation of the experimental data will be used to discuss the interplay between the existence of magnetic order and the optical response of the samples. In the perspective of the growing interest in magnetic layered materials, such basic exploration is important to build fundamental understanding, which in the future may act as a foundation for further research.

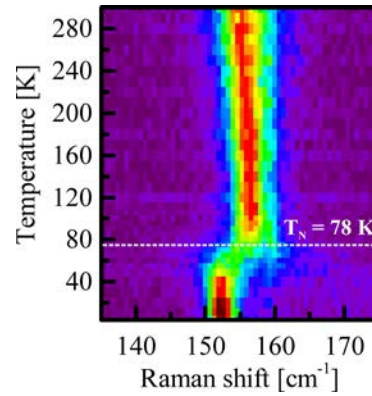


Figure 1. Temperature evolution of Raman spectrum of bulk MnPS₃ crystal. The 155 cm⁻¹ peak exhibits a drastic change at the critical temperature, indicative of alternation of crystal structure due to appearance of magnetic order.

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Epitaxial CaF₂: a Route towards Scalable 2D Electronics

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Two-dimensional (2D) semiconductors can potentially provide a means to overcome the limitations of Si technologies by enabling more than Moore FETs with several nanometer dimensions. However, this target cannot be realized without scalable insulators, which should simultaneously maintain high quality of the interface with 2D channel and low gate leakages for equivalent oxide thicknesses (EOT) below 1nm. Unfortunately, the insulators currently used in 2D FET prototypes do not satisfy these stringent requirements. For instance, oxides known from Si technologies (e.g. SiO₂, Al₂O₃, HfO₂) are amorphous when grown in thin layers, and hBN has unfavorable dielectric parameters and unfortunate band offsets to most 2D semiconductors.

To overcome this bottleneck, we suggest the use of epitaxial fluorite (CaF₂) as an insulator for 2D electronics. Few-nanometers thin CaF₂ layers [1] can be grown on Si(111) by molecular beam epitaxy (MBE) and form F-terminated inert surface with no dangling bonds [2]. This results in a high-quality quasi van der Waals interface with 2D materials (Fig.1a). At the same time, owing to good dielectric parameters ($\epsilon = 8.43$, $E_G=12.1\text{eV}$), the tunnel leakages through CaF₂ are lower than through most high-k oxides with equal EOT [3], not to mention SiO₂ and hBN.

Recently we fabricated functional CVD-MoS₂ FETs with MBE-grown CaF₂ insulators of record-small thickness only about 2 nm (EOT less than 1nm). Already in the first bare channel prototypes we achieve ultra-low leakage currents (Fig.1b), on/off current ratios of up to 10⁷ and subthreshold swings down to 90 mV/dec (Fig. 1c). At the same time, our devices exhibit almost no hysteresis, which is likely due to the virtually defect-free MoS₂/CaF₂ interface.

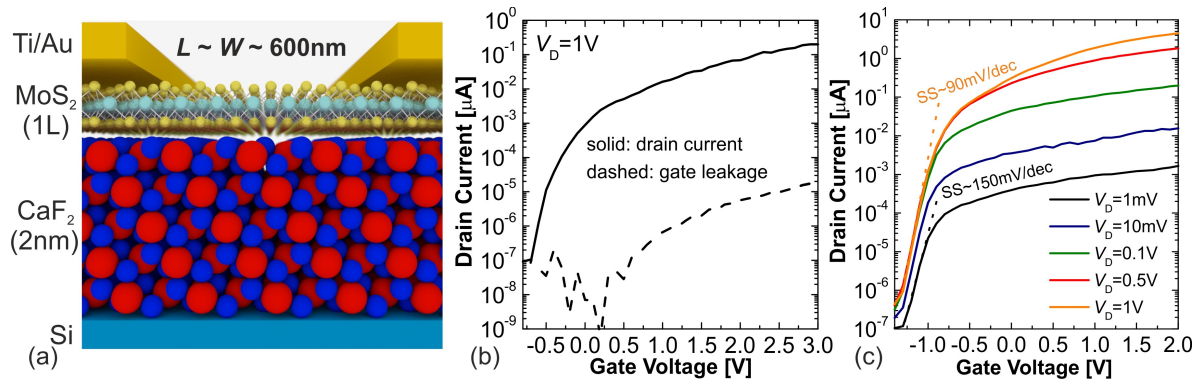


Figure 1: (a) Schematic layout of our device with quasi van der Waals CaF₂/MoS₂ interface. (b) Measured drain current and gate leakage. (c) Typical set of the gate transfer characteristics at different drain voltages.

In summary, we extended the natural stacking properties of 2D materials towards quasi van der Waals heterostructures. Demonstration of competitive MoS₂ FETs with 2nm CaF₂ insulators presents a route towards fully scalable 2D electronics. Our results are especially valuable assuming recent reports on epitaxial growth of MoSe₂ [2] and MoTe₂ [4] on CaF₂(111) crystals.

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Exciton Polariton Lasing in Cesium Lead Halide Perovskites

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Strong light-matter coupling has been reported in a wide range of organic and inorganic semiconductors, while demonstrations of the polariton condensation are limited within a handful of semiconductors at both low and room temperatures. In inorganic materials, polariton condensation significantly relies on sophisticated epitaxial growth, while organic active media usually suffer from large threshold density. Considerable efforts have been devoted to hybrid organic-inorganic perovskite materials, as they combine the advantages of both inorganic and organic materials. However, up to now, only the room-temperature strong coupling regime was observed in such materials [1,2]. The all-inorganic cesium lead halide perovskites are part of a new class of perovskite materials that are currently drawing attention to the photonics community. The epitaxy-free fabrication combined with their excellent optical gain properties with large exciton binding energy and oscillator strength, tunable emission band from UV to NIR, and better optical stability under high laser flux illumination [3], promise further important technological developments. In this talk, I will first present our results on polariton lasing at room temperature in two-dimensional CsPbCl₃ nanoplatelets embedded in a planar microcavity. These effects are unambiguously evidenced by superlinear power dependence, macroscopic ground state occupation, blueshift of the ground state emission, narrowing of the linewidth and the build-up of long-range spatial coherence. Then, I will present our latest results on realizing polariton lasing in various kinds of polariton lattice, including Lieb lattice, zigzag lattice and so on. Our findings pave the way for utilizing perovskite polariton condensate for realizing photonic quantum simulators and polaritonic devices operating at room temperature.

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Prospects and limitations of monolayer transition metal dichalcogenides as laser gain media

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The unique properties of semiconducting monolayers of transition metal dichalcogenides (TMD) have stirred excitement across physics and chemistry communities. Now, these new materials have moved into the center of interest for optoelectronic device applications, such as nanoscale lasers with ultra-low threshold currents. Despite being atomically thin, the high internal quantum efficiency and strong absorbance renders single atomic layers capable of providing sufficient gain to achieve lasing [1, 2].

Here, I give first insight into the prospects of TMD-based nanolasers and the intrinsic limitations of the gain material that arise from the many-body physics in the presence of excited carriers [3]. Strong excitation induces band-structure renormalizations that trigger a direct-to-indirect band-gap transition, leading to a rollover for the material gain and, thus, limiting the excitation regime in which laser operation is possible. The rollover can be seen in Fig. 1 for three different materials (solid lines) in comparison to logarithmic gain models (dashed lines) with no intrinsic mechanism limiting the gain. Addressing applications in optoelectronics, we combine the material-realistic gain calculations with a rate-equation approach to specify requirements to achieve lasing with the commonly used TMD semiconductors and discuss consequences for experimentally accessible laser characteristics. Finally, a quantum-optical investigation reveals that the transition to coherent emission – associated with the autocorrelation function $g^{(2)}(0)$ approaching a value of 1 – is offset to the laser threshold in the input-output characteristics, delaying the actual threshold and possibly mitigating the benefits of TMDs as laser gain materials [4].

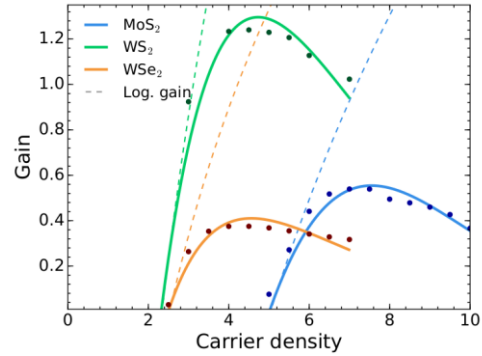


Figure 1: Gain from a single layer of different TMD materials (solid lines) show an intrinsic gain rollover limiting the maximum gain. Logarithmic gain models (dashed lines) do not capture this material realistic properties.

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Quantum-dot-like states in molybdenum disulfide nanostructures due to the interplay of local surface wrinkling, strain, and dielectric confinement

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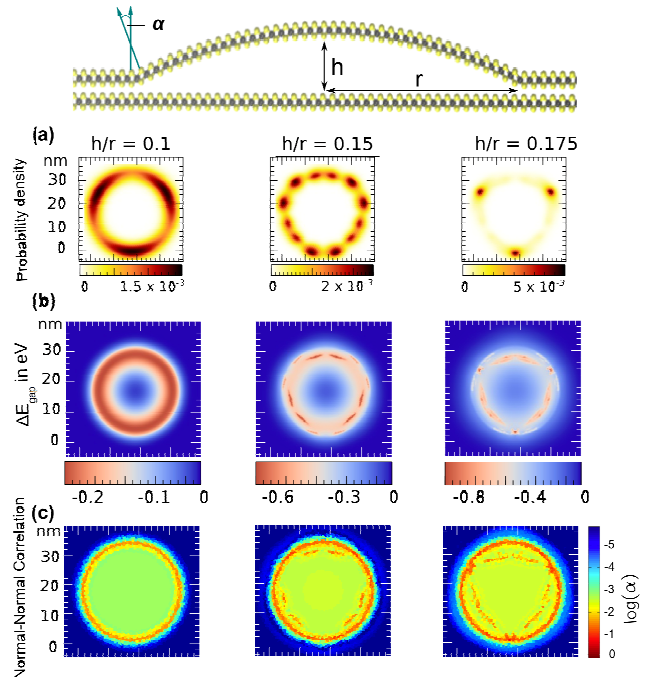
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Quantum light sources in solid-state systems are of major interest as a basic ingredient for integrated quantum device technologies. In particular, transition metal dichalcogenides (TMDs) have been found to be an excellent material for opto-electronic applications. While several experiments prove the existence of single-photon emission, open questions remain regarding its microscopic origin.

We show how changes of the dielectric environment and a non-continuous deformation of the TMD surface lead to strong carrier localization on the nm scale [1]. Effects on this length scale cannot be described using continuum methods, as the underlying atomic structure of the material becomes a determining factor. By using a combined atomistic valence force-field and million-atom supercell tight-binding approach we identify the relevant effects and provide insight into the mechanism of quantum-dot-like carrier localization, as shown in Figure 1. The interplay of local surface wrinkling, strain-induced confinement, and local changes of the dielectric environment is demonstrated for the example of nanobubble nanostructures [2] that form when monolayers are deposited on substrates or other two-dimensional materials.

Figure 1: Formation of quantum-dot-like states in MoS₂ nanobubble nanostructures.

(a) Top view of the probability densities for the electronic states, which are responsible for the strongest optical transition. The degree of localization is controlled by the aspect ratio h/r . Strong localization is obtained for $h/r=0.175$ and corresponds to the experimentally reported value in Ref. [2]. **(b)** Strain-induced band-gap changes are responsible for the localized carrier confinement. Dielectric screening leads to an additional repulsive potential in the center of the nanobubble with band-gap changes on the order of 100 meV (not shown). **(c)** Strain pockets originate from local wrinkling of the surface on the nm scale as revealed by the average angle α between normal vectors of neighboring unit cells varying up to 6° for the largest aspect ratio.



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Distinctive signatures of the spin- and momentum-forbidden dark exciton states underlying in the temperature-dependent photo-luminescence of WSe₂ monolayers

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Transition-metal dichalcogenide monolayers (TMDC-MLs) have recently drawn a great deal of attention because of the strong light-matter interaction and the optically selective valley features. [1,2] Unlike the bulk counterpart, atomically thin TMDC-MLs possess the direct band-gap opened at the two distinctive K and K' valleys, which are revealed as a new and optically accessible spin-like degree of freedom being the base of the new quantum application of valleytronics. With both spin and valley degrees of freedom, the low-lying excitonic spectra of photo-excited TMDC-MLs exhibit complex and physically rich excitonic fine structures, comprising the intra-valley bright exciton states and various intra- and inter-valley dark ones as well. According to the violated optical selection rules, the latter are classified as two types, i.e. spin- and momentum-forbidden dark excitons. Because of the optical invisibility, those two types of dark exciton are hardly measured and even distinguished in conventional optical spectroscopies. [3] In this work, we will present a theoretical and computational investigation of the exciton fine structures of WSe₂ monolayers by solving the Bethe-Salpeter equation (BSE) in the tight-binding theory established from the density-functional-theory based first principle computation. As the main result, we reveal the distinctive signatures of the spin- and momentum-forbidden exciton states of a WSe₂-ML underlying in the temperature-dependent photo-luminescence and explicitly derive the guiding principle to distinguish the energetic locations of the two types of dark excitons. [4]

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Magnetic field induced valley polarization of singlet and triplet trions in monolayer WS₂

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The direct-bandgap character, strong direct and exchange Coulomb interactions as well as significant effects of spin-orbit coupling in electronic bands of monolayers of semiconducting transition metal dichalcogenides make these systems an interesting platform for studying two-dimensional physics. Optical response of these atomically thin materials is dominated by excitonic complexes, such as neutral-, charged- and bi-excitons, which appear in multiple configurations of electrons and holes from different valleys and with different spins.

We have studied valley polarization of singlet and triplet states of negative trions in WS₂ monolayer (ML) encapsulated in hexagonal boron nitride, by means of helicity resolved reflectance contrast (RC) and photoluminescence (PL) experiments in magnetic fields up to 14 T. Due to the high quality of the investigated ML, we were able to resolve both the PL and RC signals from the singlet (intravalley, T_S) as well as triplet (intervalley, T_T) states of the negative trions (see Fig. 1). We found that with increasing magnetic field the related transition resonances become almost fully valley polarized in the RC spectrum at 14 T with the same values of the polarization degrees, but of opposite signs, what confirms their electrons' spins configurations. This allows us to estimate the electron concentration of the order of $\sim 10^{11} \text{ cm}^{-2}$ in our sample. However, the magnetic field evolutions of the trion emissions are entirely different. The polarization degree of the T_S emission is comparable to the one obtained from the RC experiment, while the corresponding polarization degree of T_T emission is of about 3 times smaller. The found difference in an influence of magnetic field on the RC and PL signals can be explained in terms of relaxation processes of both electrons in the conduction band as well as whole trion complexes.

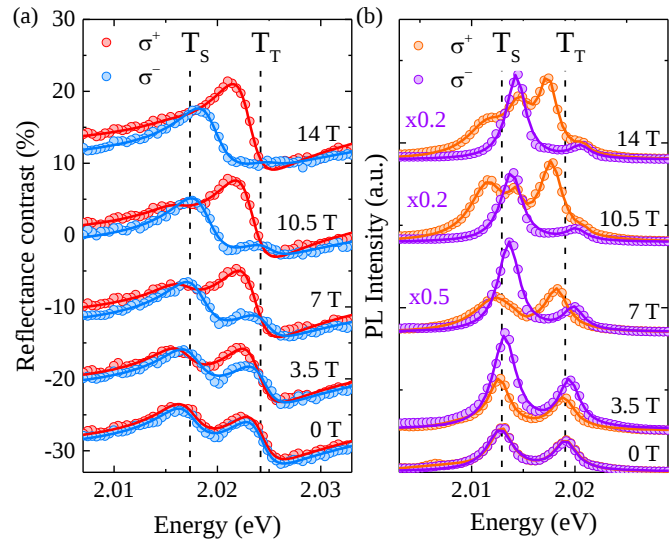


Figure 1: Circular-polarization-resolved low temperature ($T=4.2 \text{ K}$) (a) reflectance contrast, (b) photoluminescence spectra, of singlet and triplet trions measured on the WS₂ monolayer encapsulated in hBN as a function of the magnetic field.

Gate dependent emission from WSe₂/MoS₂ hetero-bilayer

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We observe the photoluminescence of interlayer excitons in nearly aligned WSe₂/MoS₂ hetero-bilayers. The type II band alignment between WSe₂ and MoS₂ monolayers drives electrons from WSe₂ to MoSe₂ and holes in the opposite direction, giving rise to interlayer excitons with spatially separated electrons and holes. This configuration is manifested by photoluminescence excitation spectrum of interlayer excitons, which shows resonance with monolayer WSe₂ and MoSe₂ exciton peaks. We fabricate WSe₂/MoS₂ hetero-bilayer FET device to manipulate the doping density and electric field by gate voltage. The photoluminescence of interlayer exciton is shifted with gate voltage at a rate of 0.15 meV/V. We also observe localized sharp emission in the spectral range of interlayer excitons at the temperature of 4K. The polarization of those sharp peaks can be either co- or cross-polarized with circularly polarized incident laser. The g factor of 6.7 provides further evidence for interlayer feature of those sharp peaks [1].

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Coupling of Atomically Thin Semiconductors to Plasmonic Nanoantennas

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Combining nano-optical systems with optically active two-dimensional materials has recently emerged as a fascinating topic to achieve new optical functionalities at the nanoscale [1]. In this contribution, we present investigations of light-matter interactions between transition metal dichalcogenide (TMD) monolayers and lithographically defined gold bowtie nanoantennas. By performing 3D-FDTD calculations, we tuned the design of the bowtie nanoantennas to match the dipolar resonance with the fundamental exciton transitions in a proximal MoSe₂ monolayer. Fabricated bowtie nanoantennas show quality factors of $Q = 5$ and sub-10nm feed-gaps with estimated mode volumes as small as $V_m = 2000\text{nm}^3$. Typical differential reflectance spectra recorded from individual TMD-bowtie nanostructures at room temperature reveal low- and high-energy peaks separated by a dip at the energy of the uncoupled exciton. To elucidate the nature of characteristic spectral features, we use the coupled oscillator model [2], which result in coupling constants at zero detuning of $g = 55\text{ meV}$. This places our hybrid system in the weak-coupling regime with spectra exhibiting Fano-like behavior. Furthermore, we demonstrate active control of the optical response by varying the polarization of the excitation light. The methods developed in our work contribute to on-demand realization of optimally coupled TMD-nanoantenna systems that can be site-selectively addressed. This type of nanostructure could pave the way for on-chip actively controlled hybrid devices operating at elevated temperatures.

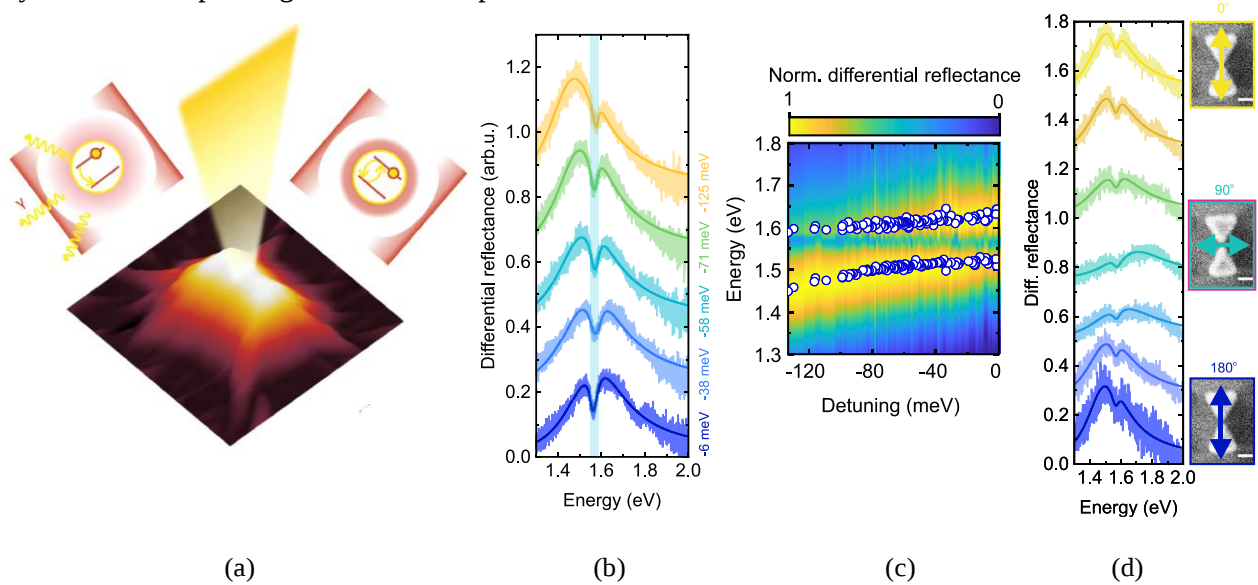


Figure 1: (a) Schematic representation of a TMD-bowtie hybrid nanostructure. (b)(c) Differential reflectance spectra recorded from single nanoantennas ordered by detuning to the exciton transition. Data reveals an anti-crossing-like behaviour. (d) Control of the optical response by tuning the polarization of the excitation light.

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Experimental study of electronic band structure of bulk and atomically thin $\text{Mo}_{1-x}\text{W}_x\text{Se}_2$

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Photoacoustic (PA) and photoreflectance (PR) spectroscopy have been applied to study the indirect and direct band gap for bulk and atomically thin molybdenum tungsten diselenide alloys $\text{Mo}_{1-x}\text{W}_x\text{Se}_2$ – with $x=0, 0.014, 0.19, 0.37, 0.51, 0.71, 1$. The bulk samples performed using the flux zone growth were obtained from 2Dsemiconductors company, whereas atomically thin samples were obtained by mechanical exfoliation. The lateral size of the studied monolayers were on the order of $10\mu\text{m}$ what allowed to conduct micro-photoreflectance experiments.

It is shown that the indirect band gap can be determined by PA technique while the direct optical transitions can be probed by modulated reflectance techniques, in this case PR, which are not sensitive to indirect transitions. On base of the experiments performed for bulk samples the energy of indirect (I) and direct (A, B and C) excitonic transitions were obtained. Furthermore also for monolayer samples the energy of A and B excitonic transitions were determined and compared with the results of photoluminescence measurements. The energy of abovementioned transitions for bulk samples, i.e. I, B and C follow well known Vegard's law without a need to take into account bowing parameter which was required in the case of A excitonic transition. Additionally, the changes of the electronic band structure, of bulk and atomically thin samples, when going from MoSe_2 to WSe_2 observed experimentally were compared with the results of DFT calculations.

FORM OF PRESENTATION: POSTER

Probing direct optical transitions of ReS₂ and ReSe₂ by means of high-pressure photoreflectance spectroscopy

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Keywords: transition-metal dichalcogenides, photoreflectance, DAC, van der Waals, excitons

Amongst all transition metal dichalcogenides (TMDCs), ReX₂ (X= S, Se) has received special interest due to its large in-plane anisotropic properties, and small gap (≈ 1.2 eV). These properties arise from its reduced crystal symmetry and make ReX₂ appealing for novel optoelectronic devices such as photodetectors or field-effect transistors, extending the optoelectronic applications of TMDCs into the near-infrared range.¹ Owing to its large technological interest the optoelectronic properties has been extensively investigated by modulated spectroscopies at different temperatures and polarizations.² More recently, angle-resolved photoemission spectroscopy measurements shed new light into the electronic band structure,^{3,4} but the origin of all excitonic transitions is not fully understood and an accurate assignation of all excitonic transitions is highly desirable. In this regard, high-pressure photoreflectance is a powerful technique that allow determining the pressure dependence of the direct optical transitions.

Here we provide an experimental and theoretical study of the electronic band structure of ReS₂ and ReSe₂ at high hydrostatic pressures. Our high-pressure photoreflectance experiments are analyzed in terms of *ab initio* calculations within density functional theory. We report the pressure dependence of the two most dominant excitonic transitions, which exhibit a negative pressure coefficient in contrast to group-6 TMDCs like MoS₂ where it is positive. These transitions are assigned to the Z k-point of the Brillouin zone and other k-points located away from high-symmetry points. The origin of the pressure coefficients of the measured direct transitions is discussed in terms of orbital analysis of the electronic band structure and van der Waals interlayer interaction. We find that ReX₂ does not exhibit a strong electronic decoupling, in agreement with recent angle-resolved photoemission studies. Finally, the anisotropic optical properties are studied at high pressure by means of polarization-resolved photoreflectance measurements.

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Exciton Localization in 2D MoS₂ by Introducing Atomic Defects

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Defects and their spatial distribution are essential factors for local modulation of electronic and photoluminescence properties of 2D materials. Missing atoms in semiconductors are exciton-trapping sites, moreover, excitons can bind to impurity atoms or can be trapped in a potential well created by local strain or structural defects. In this context, introducing defects with a focused ion beam (FIB) is of particular interest by giving the opportunity for manipulation of 2D materials on the nanometer scale. Here, two aspects are to be discussed. First, we investigate how focused gallium ion irradiation affects the intrinsic luminescence and vibrational properties of atomically thin MoS₂. Defects were introduced by scanning the Ga⁺ probe over a certain area. The amount of defects was controlled by varying the Ga⁺ dose starting from $2 \cdot 10^{11}$ ions/cm² until the PL signal was completely vanished ($2 \cdot 10^{13}$ ions/cm²). After Ga⁺ irradiation, the micro-photoluminescence measurements at T = 4 K show that the A exciton emission was suppressed and a new prominent emission feature, a bound exciton (D) located at 1.75 eV, emerges. This broad emission feature is red-shifted by $\Delta E \sim 170$ meV with respect to the neutral exciton X emission and remains active at elevated temperatures. Second, we will focus on the emission linewidth. Encapsulating monolayer MoS₂ into hBN drastically reduces the inhomogeneous contribution to the exciton linewidth [1]. The possibilities of tailoring optically active defect centers in 2D MoS₂ to even host single-photon emitters will be discussed.

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Ionic permeability and interfacial doping of graphene on SiO₂

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Graphene has been extensively used as electrodes in various electrochemical applications, thanks to its outstanding electrical and mechanical properties and superior chemical stability. Despite its relevance for these various applications, the effect of electrolyte solutions on the electronic properties, among which specifically the conductivity (σ) of graphene, and in return how graphene can sense the ions in the electrolyte, remain poorly understood. In a few available studies using graphene field effect transistors (FETs) for sensing various cations, conflicting results have been reported [1, 2].

Here employing optical pump-THz probe (OPTP) spectroscopy as a contact-free, all optical means, we investigate the impact of a series of cations with various solvation radius (based on chloride salt XCl, with X = Li⁺, Na⁺, K⁺, Ca²⁺) on the electronic properties. We demonstrate a strong pure electrostatic n-doping effect (over 200 meV) in graphene induced by metal cations in the electrolyte. Furthermore, we study the kinetics of the doping process: it takes 10's of minutes to achieve full doping, and the doping time is critically depending on the solvation radius of ions with the trend of: K⁺ < Li⁺ < Na⁺ < Ca²⁺. Our results can be rationalized by assuming that the ionic doping process involves the permeation of cations through defects in graphene, and interfacial diffusion of cations between graphene and the supported substrate SiO₂. This process is controlled by the interplay between the size, density of defects (atomic defects, pores, or grain boundaries), and the cation solvation radius. Our report presents not only a new tool to monitor the ionic dynamics at graphene and potentially other materials interfaces, but also offers new insight into graphene's cation-sensing mechanism, and highlights the importance and impact of cation permeability through graphene and the ion interfacial dynamics on the graphene's electronic properties and thus its ionic sensibility.

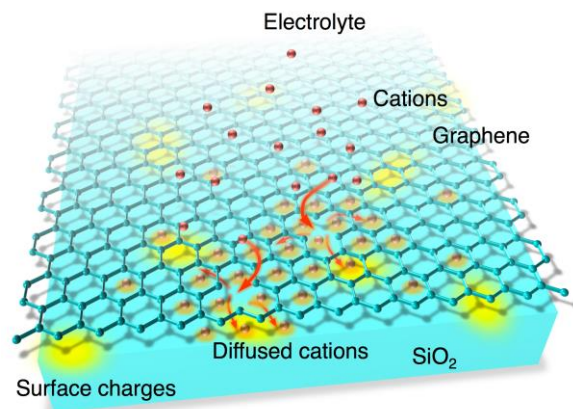


FIGURE 1. Illustration of cation permeate through graphene sheet into the graphene-SiO₂ interface and the interfacial doping of graphene.

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CHARACTERIZATION OF COMMERCIAL LUMINESCENT POWDERS

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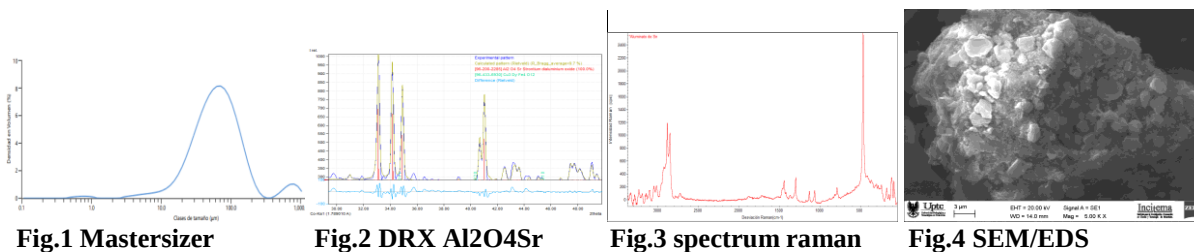
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This study reports the characterization of commercial strontium aluminates by x-ray diffraction, scanning electron microscopy, raman spectroscopy and visible UV spectrophotometry. For its possible application as mechanoluminescent coating. [3]

Strontium aluminates doped with rare earths were reported by Matsuzawa in 1996 [2] for the first time and since then it has become the photoluminescent material by excellence. For the study commercial luminescent powders were used with a particle size measured by the mastersizer 3000 which yields a particle size in the range of 100 μm to 20 μm (fig.1) and a specific surface area of 195.6 m^2/kg , for structural analysis was used an X-ray diffractometer (fig.2), PANalytical X'Pert PRO PW500 with cobalt cathode, the spectrum was analyzed with the match!3 software, and gives us a monoclinic structure with unit cell parameters $a = 8.4470 \text{ \AA}$ $b = 8.8160 \text{ \AA}$ $c = 5.1630 \text{ \AA}$ $\beta = 93.420$ with formula $\text{Al}_2\text{O}_4\text{Sr}$, the district electron microscopy was performed on the scanning electron microscope MEB ZEISS LSM 800 in which the particle size and composition of $\text{Al}_2\text{O}_4\text{Sr}$ were checked by EDS and Dy doping (Fig.4.) in the raman spectrum(Fig.3) is observed the peak in 465cm^{-1} , is associated to the torsional vibration of the O-AL-O bond, associated to the formation of strontium aluminate, this bond can be associated to the monoclinic phase, dopings were found in the peaks 117,244,340,383,423,558 cm^{-1} corresponding to the cubic phase of Eu_2O_3 , 143,328,370 cm^{-1} and 462 cm^{-1} Dy_2O_3 [1] in the visible uv spectrum, the wavelength of absorbance and transmittance of strontium aluminate powders is checked where the lowest energy passing through the material is given between 970 nm to 980 nm and a maximum absorbance at 980 nm.



Due to its micro structural characteristics, it is concluded that strontium aluminate was manufactured at high temperatures due to the absence of other characteristic phases such as Al_2O_3 and SrCO_3 , which persists at lower manufacturing temperatures than 1000°C . The particle size is affected by transport, but presents characteristics that agree with industrial manufacturing, which is done by synthesis by solid state and has temperature parameters between 1300 and 1900°C and a particle size between 20 and $100 \mu\text{m}$, essential characteristics for use as a coating.

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