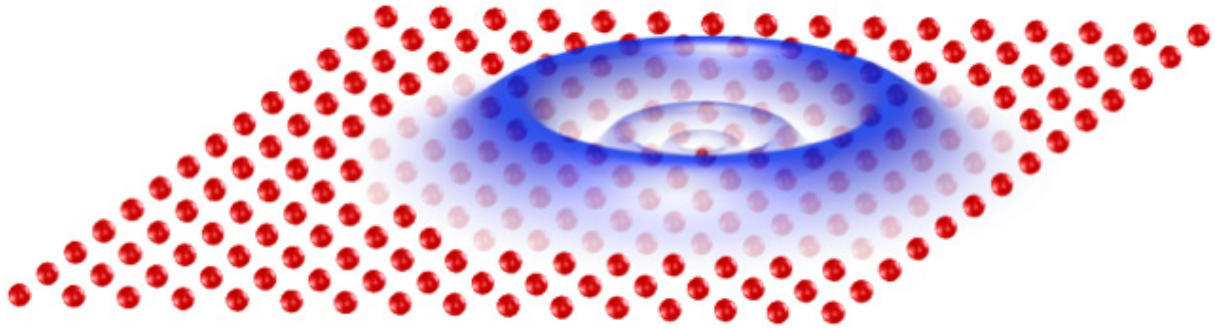
5th International Workshop

on

RYDBERG EXCITONS IN SEMICONDUCTORS

10-12 MAY 2022



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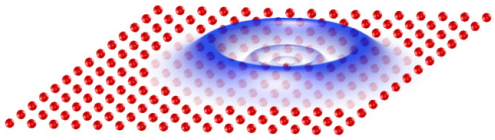
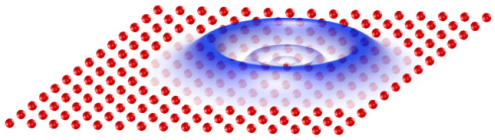


INVITED TALKS

Hamid Ohadi, University of St. Andrews, UK

Rydberg polaritons in a Cu2O microcavity

Giant Rydberg excitons with principal quantum numbers as high as n = 30 have been observed in cuprous oxide (Cu2O), a semiconductor in which the exciton diameter can become as large as ~1 μm. The giant dimension of these excitons results in excitonic interaction enhancements of orders of magnitude. Rydberg exciton–polaritons, formed by the strong coupling of Rydberg excitons to cavity photons, are a promising route to exploit these interactions and achieve a scalable, strongly correlated solid-state platform. However, the strong coupling of these excitons to cavity photons has remained elusive. Here, by embedding a thin Cu2O crystal into a Fabry–Pérot microcavity, we achieve strong coupling of light to Cu2O Rydberg excitons up to n = 6 and demonstrate the formation of Cu2O Rydberg exciton–polaritons. These results pave the way towards realizing strongly interacting exciton–polaritons and exploring strongly correlated phases of matter using light on a chip.

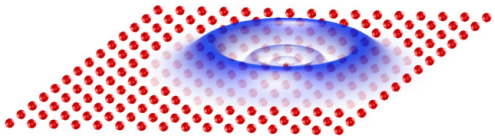


Ajit Srivastava, Emory University, Atlanta, US

Interacting Excitons and Electrons in vdW Heterostructures

Atomically thin semiconductors, such as transitional metal dichalcogenides (TMDs), have recently come to the forefront of research in materials physics. This is largely due to the ease with which they can be combined into artificially engineered heterostructures that exhibit emergent electronic and optical properties. Enhanced Coulomb interactions in the atomically thin limit result in stable excitons and their neutral of charged complexes. Furthermore, van der Waals (vdW) heterostructures can host interlayer excitons with a permanent electric dipole moment which gives rise to a system of interacting excitons.

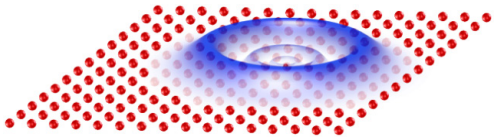
In this talk, I will begin by highlighting some unique properties of TMD excitons. Next, I will talk about few-body and many-body interactions amongst them, which can be exploited for realizing on-demand quantum matter in a driven-dissipative setting. Finally, I will show how trapped dipolar excitons can serve as high-resolution quantum sensors of correlated electronic phenomena.



Stephen Lynch, Cardiff University, UK

Unlocking the Potential of Cuprous Oxide for Quantum Technologies

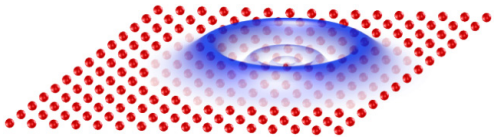
The landmark 2014 paper by Kazimierczuk et al. demonstrating high principle quantum number excitons in cuprous oxide reinvigorated interest in this extraordinary crystalline material. Coincident with an era where funding opportunities for quantum technologies are rapidly growing, our research field has perhaps its best chance ever to benefit from significant investment. Our challenge now is to convince funders that there is a realistic pathway towards industrial exploitation. There are remarkable parallels with the battle fought by scientists working on the negatively charged nitrogen vacancy centre in natural diamond gemstones in the early 2000s. While it remains the case that the best samples are of natural origin, there is a grave danger that cuprous oxide will be merely viewed as an interesting laboratory curiosity by the wider quantum technologies community. To accelerate progress, we need to close the feedback loop where material markers identified through rapid characterisation techniques correlate with the observation of high principle quantum number excitons. In this talk, I will discuss our progress in this direction, and outline some new research directions.



Jörg Main, University of Stuttgart, DE

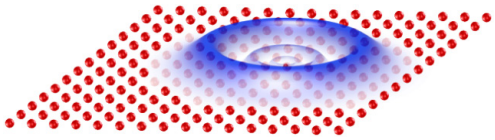
Semiclassical approaches to Rydberg excitons in cuprous oxide

The experimental work of M. Bayer's group at the TU Dortmund [1, 2] has shown the existence of highly excited exciton states in cuprous oxide in a regime, where the correspondence principle is valid and semiclassical methods can be applied. The short-range correlations between the exciton states can be analyzed with statistical methods. Magnetoexcitons break all anti-unitary symmetries and the Hamiltonian leads to the nearest-neighbor-spacing statistics of a Gaussian unitary ensemble [2, 3]. By changing the energy and the orientation of the crystal relative to the magnetic eld axis the system is ideally suited to study the crossover between GOE, GUE, and Poissonian statistics [4]. The long-range correlations between quantum states are related to the periodic orbits of the underlying classical system by semiclassical theories [5, 6]. However, due to additional spin degrees of freedom, viz. the quasispin and hole spin in the band-structure terms of the Hamiltonian, the existence of an exciton dynamics in semiconductors similar as for the electron in the hydrogen atom is not obvious. For highly excited Rydberg excitons the characteristic timescale of the spin dynamics is short compared to that of the exciton dynamics in coordinate space. Therefore, we use an adiabatic approach for the exciton dynamics by assuming that the fast spin dynamics reacts instantly to a change in the slow relative motion [7], in analogy to, e.g., the Born-Oppenheimer approximation for molecules. To the quantum system we apply a scaling technique in a way that the eigenvalues of the Schrödinger equation correspond to an eective
Planck constant. The classical exciton dynamics does not depend on this eective Planck constant, and thus stays the same for all states of the scaled spectrum [8]. By computing and comparing quantum mechanical and semiclassical Fourier transform recurrence spectra of cuprous oxide, we show that the quantum mechanical recurrence spectra exhibit peaks, which, by application of semiclassical theories, can be directly related to classical periodic exciton orbits. The application of semiclassical theories to exciton physics requires the detailed analysis of the classical exciton dynamics, including



two-dimensional orbits in symmetry planes of the crystal and fully three-dimensional orbits, which strongly deviate from hydrogenlike Keplerian orbits. The good agreement between the quantum and semiclassical recurrence spectra clearly illustrates the existence and meaningfulness of a classical exciton dynamics and provides a deeper physical interpretation and understanding of the band-structure splittings of excitons.

[1] T. Kazimierczuk, D. Fröhlich, S. Scheel, H. Stolz, and M. Bayer, Nature 514, 343 (2014).
[2] M. Aßmann, J. Thewes, D. Fröhlich, and M. Bayer, Nature Materials 15, 741 (2016).
[3] F. Schweiner, J. Main, and G. Wunner, Phys. Rev. Lett. 118, 046401 (2017).
[4] F. Schweiner, J. Main, and G. Wunner, Phys. Rev. E 95, 062205 (2017).
[5] M. V. Berry and M. Tabor, Proc. R. Soc. Lond. A 349, 101 (1976).
[6] M. C. Gutzwiller, Chaos in Classical and Quantum Mechanics (Springer, New York, 1990).
[7] J. Ertl, P. Rommel, M. Mom, J. Main, and M. Bayer, Phys. Rev. B 101, 241201(R) (2020).
[8] J. Ertl, M. Marquardt, M. Schumacher, P. Rommel, J. Main, and M. Bayer, Phys. Rev. Lett., submitted.



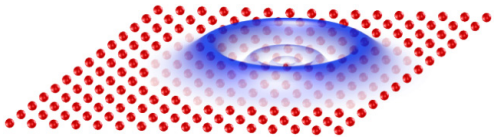
Dirk Semkat, University of Greifswald, DE

Electron-hole plasma influence on Rydberg excitons: Consequences for the interpretation of measurements

In spite of their close analogy to their atomic counterpart, Rydberg excitons [1] cannot be prepared without a solid state environment which strongly influences their properties. A prominent example is the electron-hole plasma. The plasma-induced changes of exciton-line positions and widths, and the bandedge position have been analyzed recently within a quantum many-body-theoretical approach [2, 3, 4, 5]. We discuss the consequences of the predicted effects for the analysis of measured transmission spectra and the interpretation of their features [6]. Furthermore, we show that the plasma influence is not only interesting to study properties of Rydberg excitons. Instead, we can "turn the tables" and use Rydberg excitons as testing probes to measure parameters of ultralow-density electron-hole plasmas like density and temperature.

References

[1] T. Kazimierczuk, D. Fröhlich, S. Scheel, H. Stolz, and M. Bayer, Nature 514, 343 (2014).
[2] J. Heckötter, M. Freitag, D. Fröhlich, M. Aßmann, M. Bayer, P. Grünwald, F. Schöne, D. Semkat, H. Stolz, and S. Scheel, Phys. Rev. Lett. 121, 097401 (2018).
[3] D. Semkat, H. Fehske, and H. Stolz, Phys. Rev. B 100, 155204 (2019).
[4] D. Semkat, H. Fehske, and H. Stolz, Eur. Phys. J. Spec. Top. 230, 947 (2021).
[5] H. Stolz and D. Semkat, J. Phys.: Condens. Matter 33, 425701 (2021).
[6] H. Stolz, D. Semkat, R. Schwartz, J. Heckötter, M. Aßmann, W.-D. Kraeft, H. Fehske, and M. Bayer, Phys. Rev. B 105, 075204 (2022).



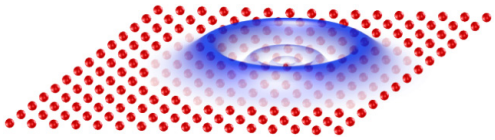
Sylwia Zielińska-Raczyńska, Bydgoszcz University of Science and Technology, PO

Rydberg excitons in nanoscale Cu2O systems

The dynamically developing eld of Rydberg exciton physics reached the maturity level where the focus of the studies is moving from the bulk medium towards nanostructures. The fabrication techniques are just entering the stage where consistent synthesis of high-quality Cu2O nanostructures becomes possible[1,2]. Therefore, it is a perfect moment to develop a theoretical description of nonlinear eects in lowdimensional RE structures. With solid and particularly exible theoretical approach based on real density matrix, one can analyse these new results and provide valuable insight on optimal experimental setups and possible applications of Rydberg excitons in Cu2O nanostructures.

On a fundamental level, depending on the number of dimensions that are reduced, we dene zerodimensional quantum dots, one-dimensional quantum wires, two-dimensional quantum wells and nally three-dimensional bulk crystal. Each of the systems is characterized with appropriate connement potential. In the case of quantum dots, we have provided some preliminary results that agree with the general tendencies found in other semiconductors [3] (Fig. 1a) and successfully explained anomalous broadening of excitonic lines in quasi-spherical nanoparticles [4] (Fig. 1b). In bulk medium, the inclusion of polaritonic eects on the crystal boundaries explains the divergence of the oscillator strengths from the precise $n^{-}2$ relation (Fig. 1c). During the talk I will also discuss the electro-optical spectra of Rydberg excitons in quantum wells for two dierent conguration of an external electric elds [5].

The complex dynamics and enhanced nonlinear properties of systems containing Rydberg excitons indicate that Cu2O might become one of the most versatile, scalable and tunable platform for quantum computing technologies.



BOOK OF ABSTRACTS

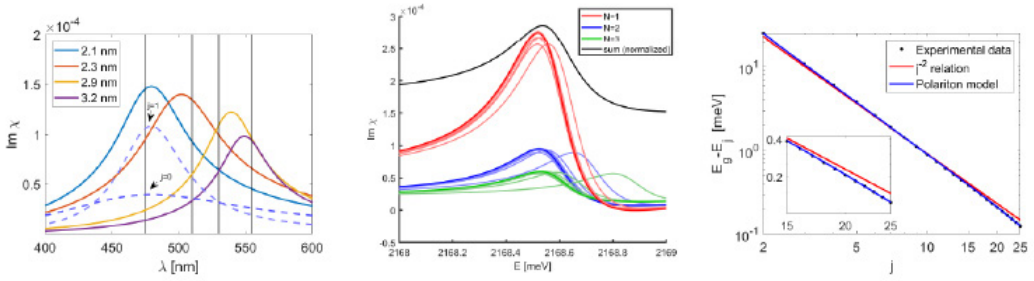
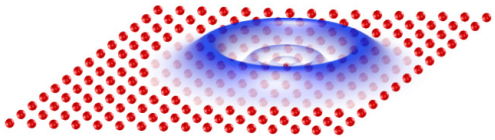


Figure 1: a) Comparison of quantum dot absorption spectrum peaks with their experimental locations (black lines). b) Absorption lines originating from connement states N = 1; 2; 3, in an ensemble of nanoparticles of various sizes, overlapping to form a single, broadened line. c) Calculated oscillator strengths of excitonic resonances, compared with experimental data from [6].

[1] S. Steinhauer, M. Versteegh, S. Gyger, A. Elshaari, B. Kunert, A. Mysyrowicz, V. Zwiller, Nature Communications Materials 1, 11, 2-7 (2020).
[2] M. Takahata, K. Tanaka, and N. Naka, Phys. Rev. B 97, 205305 (2018).
[3] D. Ziemkiewicz, K. Karpiński, G. Czajkowski, S. Zielińska-Raczyńska, Phys. Rev. B 101, 205202 (2020).
[4] K. Orfanakis, S. Rajendran, H. Ohadi, S. Zielińska-Raczyńska, G. Czajkowski, K. Karpiński, D. Ziemkiewicz, Phys. Rev. B 103, 245426 (2021).
[5] D. Ziemkiewicz, K. Karpiński, G. Czajkowski, S. Zielińska-Raczyńska, Phys. Rev. B 104, 075303 (2021).
[6] T. Kazimierczuk, D. Frölich, S. Scheel, H. Stolz, and M. Bayer, Nature 514, 344 (2014).

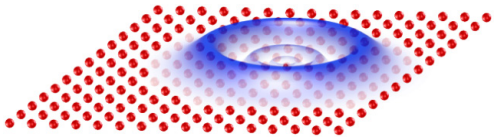


Liam Gallagher, University of Durham, UK

Microwave-optical coupling via Rydberg excitons in Cu2O

We present our recent experimental results demonstrating exciton-mediated coupling between optical and microwave fields in cuprous oxide [1]. Rydberg excitonic states are studied using one-photon (absorption) and SHG (second harmonic generation) spectroscopy techniques. In absorption spectroscopy we observe up to n=17 and find the microwave field causes a significant change in absorption throughout the exciton spectrum. However, in one-photon spectroscopy the Rydberg exciton states sit on a non-resonant background which is not modified by the microwave field.
SHG spectroscopy provides a tool for studying Rydberg excitons without a non-resonant background [2]. When a microwave field is applied during SHG spectroscopy, a four-wave mixing process can occur, leading to the appearance of sidebands on the second harmonic; demonstrating the microwave field can coherently modulate an optical carrier. The observations are in good agreement with a model based on the microwave-driven electric dipole transitions between Rydberg states of opposite parity. We show that with a simple microwave antenna, it is possible to reach a regime where the microwave coupling (Rabi frequency) is comparable to the linewidth of the Rydberg exciton states. The results provide an additional way to manipulate excitonic states, and open up the possibility of a cryogenic microwave to optical transducer based on Rydberg excitons.

[1] L. A. P. Gallagher et al., Phys. Rev. Res. 4 013031 (2022)
[2] J. Rogers et al., Phys. Rev. B 105 (11), 115206 (2022)



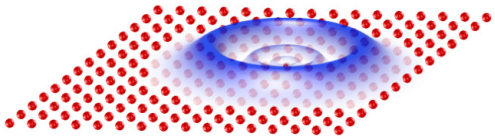
Tomasz Smoleński, ETH, Zûrich, CH

Optical sensing of strongly correlated electronic states in atomically-thin materials

When the strength of Coulomb interaction E_C between itinerant electrons in a two-dimensional system becomes significantly larger than the kinetic energy E_F, the electrons start to develop strong correlations. A paradigm phase that is expected to emerge in this regime is an electronic Wigner crystal (WC) [1], in which the electrons spontaneously form a periodic lattice mimicking that of the real crystals. In order for such a crystallization to occur in the absence of the magnetic field $B = 0$, the ratio of the two energy scales $r_s = E_C/E_F$ must exceed 30 [2]. Owing to severe difficulties in satisfying this condition for conventional semiconductors (e.g., GaAs), prior experimental studies of electronic solid have been mainly focused on the electrons occupying a single Landau level under the influence of a strong external B-field, where the kinetic energy is almost completely quenched. In this regime, the WC competes with other correlated phases, e.g., fractional quantum Hall (FQH) states, resulting in striking oscillations of longitudinal resistance at low filling factors [3].

Recently, atomically-thin transition metal dichalcogenides (TMDs) have emerged as a highly-tunable experimental platform that unlocks the access to uncharted territories of strongly correlated electron physics. This is due to reduced dielectric screening and large carrier effective masses, which endow TMD monolayers with excellent optical properties and give rise to strong inter-electron interactions enabling to reach rs values being more than an order of magnitude larger than that for the GaAs at comparable electron densities. In this talk, I will review our recent optical investigations of landmark correlated phases in charge-controlled TMD-based van der Waals heterostructures.

In the first part, I will present direct evidence that the electrons in a $MoSe_2$ monolayer at densities $< 3\times10^{11}$ cm^{-2} form a WC at B = 0 [4]. This is uncovered by our novel spectroscopic technique allowing us to detect a long-range charge order in an electronic state through the periodic potential it generates for the excitons [5]. In the presence of this potential, the excitons undergo a Bragg diffraction, which gives rise to the emergence

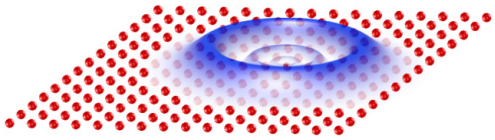


of a Bragg-umklapp transition in the reflectance spectrum that heralds the formation of the WC state. I will also show that the phase transition between this solid state and an electron liquid in the presence of an external *B*-field is associated with a sizable change in the electron spin-valley relaxation dynamics, as revealed by our time-resolved pump-probe experiments [6].

The second part of the talk will be devoted to optical spectroscopy of correlated electronic phases in graphene. Although these phases have been extensively investigated in prior transport studies, they have remained thus far optically inaccessible due to the lack of graphene bandgap. I will show that they can be all-optically probed with the use of Rydberg excitons in a proximal TMD monolayer that is separated from the graphene by a few-layer-thick spacer of hexagonal boron nitride. Owing to large Bohr radii of Rydberg excitons, their energies and spectral weights are sensitively dependent on the compressibility of adjacent graphene electrons. This allows us to sense the formation of incompressible FQH states in graphene with a similar sensitivity to that of state-of-the-art transport tools [7].

References:

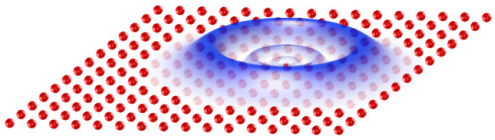
[1] E. Wigner, Phys. Rev. 46, 1002 (1934).
[2] N. D. Drummond and R. J. Needs, Phys. Rev. Lett. 102, 126402 (2009).
[3] H. Jiang, R. Willett, H. Stormer, D. Tsui, L. Pfeiffer, and K. West, Phys. Rev. Lett. 65, 633 (1990).
[4] T. Smoleński, P. E. Dolgirev, C. Kuhlenkamp, A. Popert, Y. Shimazaki, P. Back, X. Lu, M. Kroner, K. Watanabe, T. Taniguchi, I. Esterlis, E. Demler, and A. Imamoğlu, Nature 595, 53-57 (2021).
[5] Y. Shimazaki, C. Kuhlenkamp, I. Schwartz, T. Smoleński, K. Watanabe, T. Taniguchi, M. Kroner, R. Schmidt, M. Knap, A. Imamoğlu, Phys. Rev. X 11, 021027 (2021).
[6] T. Smoleński, K. Watanabe, T. Taniguchi, M. Kroner, and A. Imamoğlu, Phys. Rev. Lett. 128, 127402 (2022).
[7] A. Popert, Y. Shimazaki, M. Kroner, K. Watanabe, T. Taniguchi, A. Imamoğlu, and T. Smoleński, arXiv:2111.12887 (2021).



Georg Bruun, Aarhus University, DK

Light-induced topological superconductivity in transition metal dichalcogenide monolayers

Monolayer transition metal dichalgonenides (TMDs) host deeply bound excitons and their interactions with itinerant electrons represent an exciting new quantum many-body Bose-Fermi mixture. Here, we demonstrate that electrons interacting with a Bose-Einstein condensate (BEC) of exciton-polaritons can realise a two-dimensional topological px+ipy superconductor. Using strong coupling Eliashberg theory, we show that this is caused by an attractive interaction mediated by the BEC, which can overcompensate the repulsive Coulomb interaction between the electrons. The hybrid light-matter nature of the BEC is crucial for achieving this, since it can be tuned to reduce retardation effects and increase the mediated interaction in regimes important for pairing. We finally calculate the critical temperature and discuss how the great flexibility of TMDs can be used to observe the elusive topological superconducting phase.

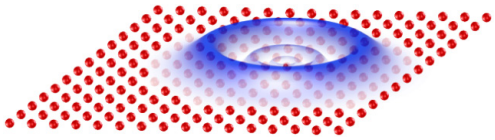


Chun Hung Lui, University of California, Riverside, US

Optical spectroscopy of novel excitonic states in 2D semiconductors and moiré superlattices

Two-dimensional (2D) semiconductors, such as $MoSe_2$ and WSe_2, host robust excitonic states with remarkable properties for novel applications. We investigate the excitonic states in the 2D semiconductors by optical spectroscopy. They exhibit a panoply of excitonic states, including ground and excited Rydberg excitonic states. In the presence of a Fermi sea, the interaction between the excitons and the Fermi sea can give rise to the exciton-polaron states. When two different monolayers are stacked together, they can form moiré superlattices. The moiré superlattices can significantly modify the excitonic properties, giving rise to moiré trions with distinctive optical signatures. These results establish 2D semiconductors as an excellent platform to explore novel excitonic physics and applications.

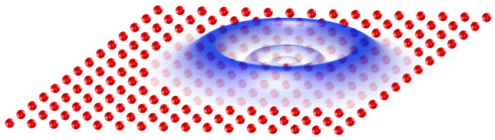
Biography: Prof. Chun Hung (Joshua) Lui joined the Department of Physics and Astronomy at the University of California Riverside as an Assistant Professor in July 2015. Before that, he was a postdoc at the Massachusetts Institute of Technology (MIT). He obtained his Ph.D. in physics at Columbia University in 2011. His research group investigates novel quantum phenomena in two-dimensional materials and heterostructures by optical spectroscopy combined with nanofabrication.



Armando Genco, The Polytechnic University of Milan, IT

Rydberg excitons dynamics and interacting dipolar excitons and polaritons in atomically thin semiconductors

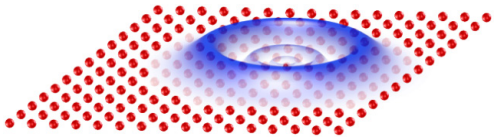
The reduced Coulomb screening in monolayers of Transition Metal Dichalcogenides (TMDs) grants high binding energies (up to 0.5 eV) and large oscillator strengths to excitons, enhancing the stability of their many-body complexes. Cryogenic temperatures and encapsulation in hBN narrow the excitonic linewidths, unveiling a hydrogen-like Rydberg series of excitonic states below the free particle bandgap. Moreover, the recently emerged exciton hybridization in TMD bilayers gives rise to dipolar excitons with high oscillator strength. All of those excitonic degrees of freedom can be exploited to achieve high nonlinearities, key for accessing collective quantum phenomena in polariton systems, when TMDs are strongly coupled to light.
We employ hybridized interlayer excitons (hIXs) in bilayer MoS_2 to achieve highly nonlinear effects relying on their dipolar nature. Compared to excitons in MoS_2 monolayers, hIXs exhibit about 8 times higher nonlinearity, which is further strongly enhanced when hIXs and intralayer excitons, sharing the same valence band, are excited simultaneously. This gives rise to a highly nonlinear regime which we describe theoretically by introducing the concept of hole crowding. Owing to the hIX large oscillator strength, we also observe strong nonlinear interactions of dipolar polaritons (dipolaritons) in bilayers placed in optical microcavities. Besides, we probe the ultrafast dynamics of excited Rydberg excitons by exciting hBN-encapsulated WSe_2 monolayers with femtosecond pulses, then changing the pump-probe delay time. 2s excitons revealed an ultrafast formation, similar to 1s states, while showing significantly longer relaxation times crucial for increased interactions. In transient reflectivity, the spectra of both 1s- and 2s-excitons showed additional low-energy features, ascribed to their many-body complexes, with distinct decay times.
The rich dynamics of Rydberg excitons and the strongly interacting behavior of dipolar excitons and polaritons explored in our studies offers novel insights on the many-body physics in atomically thin semiconductors and opens up wide opportunities for accessing extreme nonlinear quantum phenomena in such systems.



Valentin Walther, Harvard University, US

The route to quantum light via semiconductor excitons

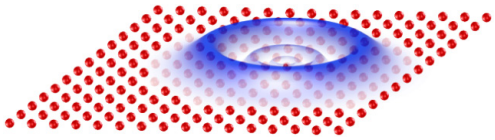
Manipulating light down to the level of single photons is an important goal of quantum optics. Semiconductor excitons afford a convenient integrable platform for this purpose. However, the optical nonlinearities of such systems are typically too weak to induce significant quantum correlations. Here, we present new strategies for this purpose that are effective in the presence of strong dissipation. These possibilities are opened up by high-lying excitonic Rydberg states with unusually strong interactions. We discuss recent progress with Rydberg excitons in bulk and two-dimensional systems along with their specific challenges, which we show can be overcome with new theoretical proposals. The results promise record photon correlations and, ultimately, provide a positive outlook on quantum optics with semiconductor excitons.



Na Young Kim, University of Waterloo, CA

Temperature Study of Rydberg Excitons in Cu_2O

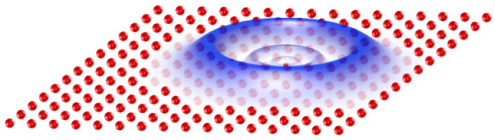
We aim to search high-temperature solid-state materials for promising quantum optical technologies. Fascinated by beautiful scaling behavior of Rydberg excitons in Cu_2O below 2 K, we are curious how high Rydberg exciton states are robust at higher temperatures. Thus, we perform temperature-dependent absorption spectroscopic measurements between 4 K and 100 K to identify the yellow and green Rydberg exciton resonance states. We present our experimental results with systematic scaling law studies at varying temperatures.



Hadiseh Alaeian, Purdue University, US

From Dipolar to Rydberg Photonics: Harnessing Atom-Atom Interactions

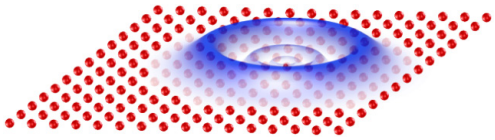
Light-induced atom-atom interactions at densities higher than 1 atom per cubic wavelength give rise to density shifts and broadenings. When confined in less than a wavelength size, such dipolar interaction leads to collective blockade phenomena, which mostly have been studied in the context of strongly interacting Rydberg states.
Here we study these phenomena for low-lying excited atomic states confined in thin atomic clouds that are generated via the pulsed Light-Induced Atomic Desorption (LIAD) technique. For the first few nanoseconds, the transient light-induced dipolar interaction of the low-lying lines of Rubidium leads to shifts and broadenings well beyond the well-known Lorentz-Lorenz limit. In the second experiment, we benefit from highly controllable fields of Nano-photonic devices to manipulate the many-body dipolar interactions. We interface the atoms with the tightly-confined field of a slot waveguide, where the Purcell enhancement modifies the interactions and the shifts, further. The latter experiments are done at telecom wavelength where one can integrate the collective quantum effects such as the blockade to create deterministic on-demand single-photon emitters. Towards the end of my talk, I will introduce our novel quantum material, thin-film cuprite, that allows us to realize strongly interacting Rydberg excitons in a solid-state platform that is inherently suitable for scalable and integrable quantum photonic technologies.



Stephan Steinhauer, KTH Royal Insitute of Technology, Stockholm, SE

Recent advances on the growth of mesoscale Cu_2O and the spectroscopic characterization of Rydberg exciton states

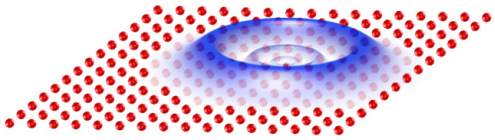
Cuprous oxide (Cu_2O) is a unique metal oxide semiconductor that can act as host material for optically excited Rydberg states with high principal quantum numbers. Although several growth methods have been reported for fabricating Cu_2O thin films and single crystals, state-of-the-art quantum optics experiments still focus on natural bulk crystals from geological samples fortuitously found in mines, which have shown the highest Rydberg states in Cu_2O found to date. Here, I will present recent progress at KTH Royal Institute of Technology on the growth of synthetic micro- and nanocrystalline Cu_2O structures with high crystalline quality and low defect density. It will be demonstrated how growth via thermal oxidation of copper (deposited on single-crystalline substrates or standard silicon substrates by physical vapour deposition) can be tailored for achieving size- and site-controlled Cu_2O crystals, epitaxial growth as well as two-dimensional structures. Furthermore, I will discuss spectroscopic characterization of Rydberg exciton states via photoluminescence excitation (PLE) spectroscopy. The employed method is based on scanning the laser excitation wavelength over the Rydberg resonances and recording the luminescence of 1s orthoexcitons. First, results on natural bulk Cu_2O crystals will be summarized, which show that this technique is well-suited for observing giant Rydberg excitons with principal quantum numbers up to n=30. Next, PLE measurements on mesoscale Cu_2O will be presented, discussing distinct spectral features and their interpretation. Eventually, future directions related to Cu_2O integration with photonic circuits and devices will be outlined.



Arturo Camacho Guardian, National Autonomous University of Mexico, MX

Moiré-induced non-linearities: From multi-photon resonances to translational symmetry breaking in driven-dissipative moiré systems

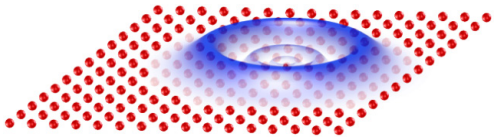
Moiré lattices formed from semiconductor bilayers host tightly localised excitons that simultaneously couple strongly to light and possess a large dipole moment enabling the formation of strongly interacting moiré polaritons. In this talk, we will show that the moiré platforms enable the realization of a new form of polaritons that exhibit strong optical non-linearities controlled by the underlying discrete character of the matter excitations. We will demonstrate the emergence of multi-photon resonances, “discrete” bi-stabilities, the appearance of states with broken translational symmetry, and discuss the role of free carriers on the optical response.



Thomas Boulier, Laboratoire de Physique de l'Ecole Normale Supérieure (LPENS), Paris, FR

Self-Kerr effect in the Cu_2O Yellow Rydberg series

I will present recent results whereby the self-Kerr coefficient (nonlinear optical index n_2) is measured, along with the absorption, across the yellow Rydberg series of copper oxide under resonant excitation. We found that n_2 is resonantly enhanced near each excitonic state and increases rapidly with the principal quantum number n, as expected from the line narrowing. While the nonlinearity is enormous at low light intensity, we found it saturates quickly, so that the total nonlinear phase shift across the crystal remains moderate whatever the pump power and whatever the state used. Interestingly, the saturation intensity decreases as n^{-7}, indicating that the nonlinear optical response is limited by the Rydberg blockade mechanism. This work represents a first step towards exploiting Rydberg excitons for high refractive optical nonlinarities.



Marc Aßmann, Technical University of Dortmund, DE

Rydberg Exciton Impurity interactions

Charged impurities in semiconductor structures are known to result in local electric fields and charge noise which may in turn cause significant spectral fluctuations of exciton lines. These are highly detrimental for any kind of precision spectroscopy or application in quantum technologies. Suppressing them has been an important step in reaching transform limited spectral lines, e.g., for quantum dots [1].
For Rydberg excitons, charged impurities and residual charges in the system create shifts of the band gap and result in undesirable blockade effects which limit the observability of states with large principal quantum number [2]. The influence of such static charges on the Rydberg exciton spectrum has been studied thoroughly theoretically and good agreement has been found. In this talk, I will discuss the other side of this interaction:
In which way may can propagating Rydberg excitons influence the state of charged impurities?
To this end, we performed time-resolved pump probe measurements in bulk Cu_2O at low pump powers where the enhanced probe beam absorption acts as a signature of impurities changing their charge state. We find that even small amounts of Rydberg excitons introduced by pump powers on the order of 1 µW have a tremendous impact on the impurity landscape and "purify" the crystal, resulting in a more pristine transmission spectrum. The interaction shows the characteristics of charge-induced dipole interactions and scales with the principal quantum number of the pumped excitons as $n^{3.5}$. As a complementary experimental approach, I will discuss the influence of the purifying effect on the photoluminescence of excitons localized at impurities.
Our results imply that Rydberg excitons may open up the possibility to suppress the detrimental influence of impurities in a wide range of semiconductor systems, including other materials and low-dimensional systems such as TMDCs.

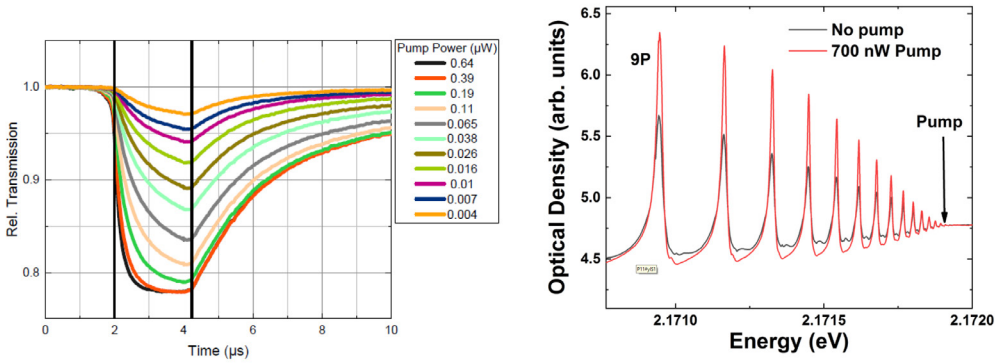
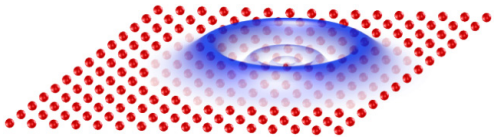


Figure 1: Left: Purifying dynamics after pulsed excitation with a duration of 2 microseconds for n_{pump}=15 and n_{probe}=9. Right: CW purifying spectrum at a power of 700 nW.

References

[1] J. Houel et al., Phys. Rev. Lett. 108, 107401 (2012).
[2] J. Heckötter et al., Phys. Rev. Lett. 121, 097401 (2018).
[3] S.O. Krüger et al., Phys. Rev. B 101, 235204 (2020).



CONTRIBUTED TALKS

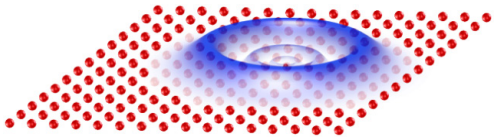
Katharina Brägelmann, M. Harati, B. Panda, J. Heckötter, M. Aßmann, Dortmund Technical University of Dortmund, DE

Neutralisation of detrimental effects on the Rydberg exciton absorption spectrum

We report on the neutralisation of charged impurities by excitation of Rydberg excitons with surprisingly small laser powers. Rydberg excitons are highly excited states in Cu_2O with principal quantum numbers of up to $n = 30$ [1] with extensions in µm range. The wellknown theories propose an n^{-3} scaling for both oscillator strengths and linewidths of the excitons. Usually the highest states ($n = 16$ and above) show some deviation from those theories, as oscillator strengths are smaller and linewidths are wider than expected, which leads to an reduced absorption of these states. Those deviation are known to stem from the presence of charged impurities in the material [2]. Here, we show a way to increase the absorption and to minimize the deviations mentioned above. This increase of absorption happens when the system is pumped in an extremly narrow energy region around the band gap (less than 1 meV) with very small powers of only 0.1 – 10 µW. This effect incidates a 'purification' of the illuminated volume as the naturally charged impurities have less detrimental impact on the high excitonic states. This research contributes to a deeper understanding of impurity - exciton interactions.

References

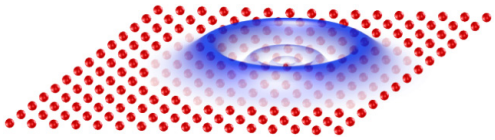
[1] M. A. M. Versteegh, S. Steinhauer, J. Bajo, T. Lettner, A. Soro, A. Romanova, S. Gyger, L. Schweickert, A. Mysyrowicz, and V. Zwiller, Phys. Rev. B 104, 245206 (2021).
[2] S. O. Krüger, H. Stolz, and S. Scheel, Phys. Rev. B 101 (2020).



Florian Morawetz, University of Rostock, DE

Towards calculating Rydberg excitons in quantum wells

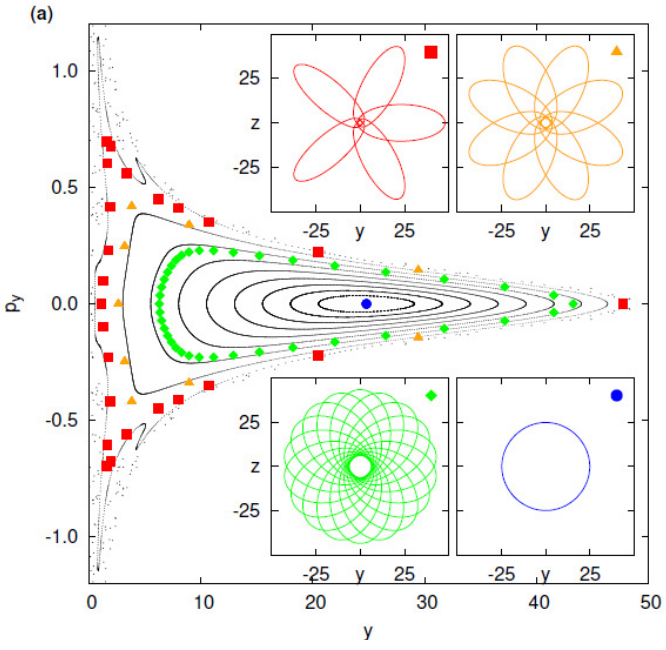
In this talk, rst approaches towards calculating the spectrum of Rydberg excitons in quantum wells are shown. Due to recent advancements in manufacturing high-quality thin samples of cuprous oxide, the need for a microscopic description of the connement eects on highly excited (Rydberg) excitons is apparent. The dierent connement regimes are initially introduced. Corresponding limiting cases, i.e. quasi-2D and bulk lead to well-known analytic solutions with hydrogen-like wave functions. Based on those limits, two dierent methods for a complete basis expansion are derived to calculate the excitonic spectrum. For dierent quantum well lengths the obtained states can then directly be classied by means of the quantum numbers of the corresponding limiting cases.



Jan Ertl,[1] M. Marquardt,[1] M. Schumacher,[1] P. Rommel,[1] J. Main,[1] and M. Bayer[2]
[1]University of Stuttgart, DE, [2]Technical University of Dortmund, DE

Signatures of Exciton Orbits in Quantum Mechanical Recurrence Spectra of Cuprous Oxide

The seminal work by T. Kazimierczuk et al. [1] has shown the existence of highly excited exciton states in a regime, where the correspondence principle is applicable and quantum mechanics turns into classical mechanics. This naturally raises the question for an interpretation of exciton spectra in terms of classical exciton dynamics. The exciton orbits exhibit a secular motion of Kepler ellipses on near-integrable tori in phase space [2]. The connection of classical exciton dynamics to quantum spectra is given by semiclassical formulas for the density of states where each classical orbit provides sinusoidal uctuations [3, 4]. In the recurrence spectrum, i.e. the Fourier transform of the density of states individual contributions of classical exciton orbits can be uncovered, providing an intuitive description of excitons in cuprous oxide.



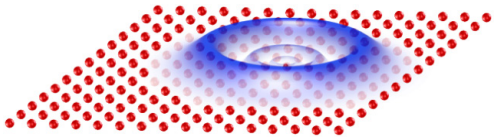
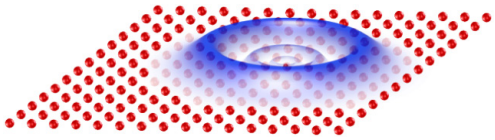


FIG. 1. Poincaré surface of section of the orbits in the (x = 0)-plane at n_0 = 5. Dominating regular tori are surrounded by a small chaotic region. The insets show selected periodic orbits on rational tori marked with colored symbols.

[1] T. Kazimierczuk et al., Nature 514, 343 (2014).
[2] J. Ertl et al., PRB 101, 241201(R) (2020).
[3] M. C. Gutzwiller, Chaos in Classical and Quantum Mechanics, Springer, New York, 1990.
[4] M. V. Berry and M. Tabor, Proc. R. Soc. Lond. A 349, 101 (1976).

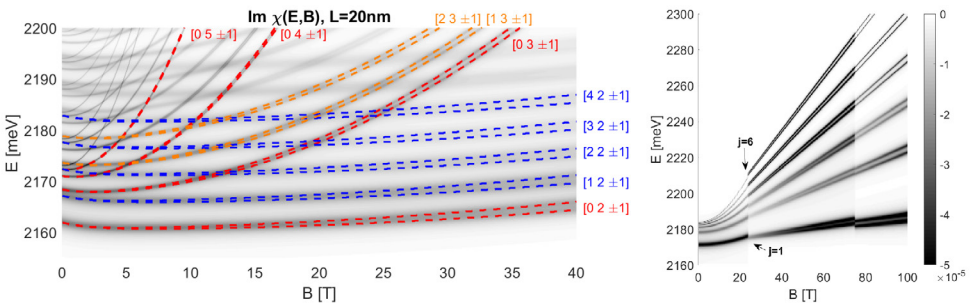


Karol Karpinski, Bydgoszcz University of Science and Technology, PO

Magneto-optical properties of low dimensional Rydberg systems

The extraordinary properties of Rydberg excitons include an extraordinary strong response to a magnetic eld, which attracted a lot of attention of various experimental and theoretical groups [1,2]. It has been recognized that the large number of absorption lines observed in excitonic media could not be interpreted on the grounds of a simple hydrogen-like model [2] and a full description of complex valence band structure and crystal symmetry is needed. Real density matrix approach provides such a description [3].

The previously developed RDMA framework for description of quantum wells can be extended to include connement potential of a quantum well [4]. A complicated interplay of eld-dependent energy shift, Zeeman eect and the appearance of multiple absorption lines corresponding to connement states leads to a highly complex spectrum (Fig. 1a). Furthermore, an unique advantage of Rydberg excitons over Rydberg atoms is the fact that due to their lower binding energy, one can observe the system in low-, intermediate- and high-eld regime in realistic experimental conditions. The division into three regimes is based on the ratio of exciton binding energy to the magnetic eld energy. Such a division into three regimes can be seen on the Fig. 1b. Two dierent geometric congurations (Voigt and Faraday) of the system are discussed.



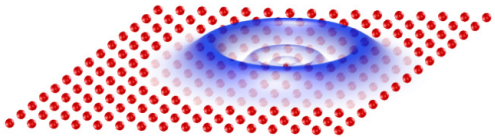
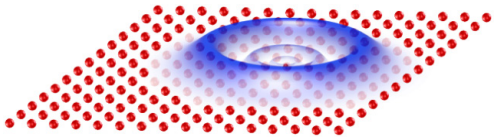


Figure 1: a) Absorption spectrum of Cu_2O quantum well in magnetic eld; numbers in parenthesis are connement, principal and magnetic quantum numbers. b) Absorption spectrum in the weak, intermediate and strong eld regime.

[1] S. Artyukhin, D. A. Fishman, C. Faugeras, M. Potemski, A. Revcolevschi, M. Mostovoy, P. H. M. van Loosdrecht, Sci Rep 8, 7818 (2018).
[2] F. Schweiner, J. Main, G. Wünner, M. Freitag, J. Heckötter, C. Uihlein, M. Assmann, D. Fröhlich, M. Bayer, Phys. Rev. B 95, 035202 (2017).
[3] S. Zielińska-Raczyńska, D. A. Fishman, C. Faugeras, M. P. Potemski, P. H. M. van Loosdrecht, K. Karpiński, G. Czajkowski, D. Ziemkiewicz, New J. Phys. 21, 103012 (2019).
[4] D. Ziemkiewicz, G. Czajkowski, K. Karpiński, S. Zielińska Raczyńska, Phys. Rev. B 103, 035305 (2021).



BOOK OF ABSTRACTS

POSTER CONTRIBUTIONS

Anindya Sundar,[1,2] Sai Kiran Rajendran,[3] Lorenzo Scarpelli,[2] Thomas Volz,[2] and Hamid Ohadi[1],
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Towards strong coupling of Rydberg excitons in Cu_2O to photons in fully tunable micro-cavities

Binodbihari Panda, Julian Heckötter, Katharina Brägelmann, Mariam Harati, Marc Aßmann, Manfred Bayer, University of Dortmund, DE
Impurity-Rydberg Exciton Interaction in Cu_2O

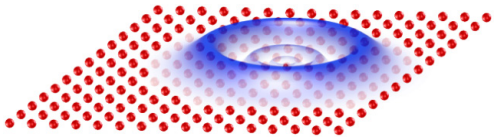
Jan Kumlin, Aarhus University, DK
Quantum Optics with Rydberg Superatoms

Julian Heckötter, B. Panda, K. Brägelmann, M. Harati, M. Aßmann, M Bayer, University of Dortmund, DE
D excitons and the role of intermediate peaks in Rydberg exciton spectra in Cu_2O

Lida Zhang, Aarhus University, DK
Chiral and coherent manipulation of photonic quantum states with Rydberg arrays

Patric Rommel,[1] J. Main,[1] A. Farenbruch,[2] J. Heckötter,[2] M. Aßmann,[2] D. Yakovlev,[2] and M. Bayer[2],
[1]University of Stuttgart, DE, [2]Technical University of Dortmund, DE
Fine structure of the S- and D-excitons in cuprous oxide

Simon Panyella Pedersen, Aarhus University, DK
Quantum nonlinear optics in atomic dual arrays



BOOK OF ABSTRACTS

David Ziemkiewicz, Bydgoszcz University of Science and Technology, PO

Towards microwave technologies with Rydberg excitons and plasmons

Esben Rohan Christensen,[1] A. Camacho-Guardian,[2] A. Imamoglu,[3] M. Wouters,[4] G. M. Bruun,[1 ‘ 5] and I. Carusotto[6],
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Microscopic theory of light-enhanced interactions of dipolaritions

Aleksi Julku, Aarhus University, DK

Non-local interactions and supersolidity of moiré excitons

Karol Karpinski, Bydgoszcz University of Science and Technology, PO

Magneto-optical properties of low dimensional Rydberg systems

Jan Ertl,[1] M. Marquardt,[1] M. Schumacher,[1] P. Rommel,[1] J. Main,[1] and M. Bayer[2],
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Signatures of Exciton Orbits in Quantum Mechanical Recurrence Spectra of Cuprous Oxide

Katharina Brägelmann, M. Harati, B. Panda, J. Heckötter, M. Aßmann, Dortmund Technical University of Dortmund, DE

Neutralisation of detrimental effects on the Rydberg exciton absorption spectrum