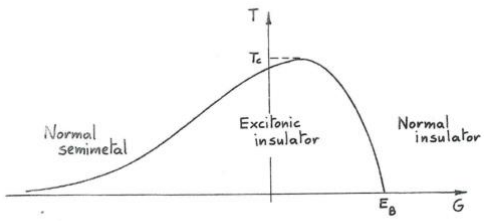
Excitonic and competing orders in low-dimensional materials



June 21, 2021 - June 23, 2021
Online meeting, hosted by CECAM-HQ

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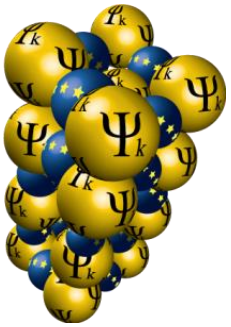

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1. Description

In the last twelve months, the field of the excitonic insulator has moved extremely fast. This research has its origin in a heretic prediction formulated more than 50 years ago by a group of visionary physicists, including Leonid Keldysh and Walter Kohn [1-3]: if a narrow-gap semiconductor, or a semimetal with slightly overlapping conduction and valence bands, failed to fully screen its intrinsic charge carriers, then excitons - electron-hole pairs bound together by Coulomb attraction - would spontaneously form. This would destabilize the ground state, leading to a reconstructed 'excitonic insulator'- a condensate of excitons at thermodynamic equilibrium. This chimeric phase shares fascinating similarities with the Bardeen-Cooper-Schrieffer superconductor: a distinctive broken symmetry, inherited by the exciton character, and collective modes of purely electronic origin. Its observation was deterred for many decades by the trade-off between competing effects in the semiconductor: as the size of the energy gap decreases, favoring spontaneous exciton generation, the screening of the electron-hole interaction increases, suppressing the exciton binding energy. In the last two years, mounting evidence [4-12] has been accumulating in low-dimensional materials, as they combine optimal band structures, poor screening behavior, truly long-ranged interactions, and giant excitonic effects (see also the list of recent literature maintained at www.nano.cnr.it/index.php?mod=men&id=196). This was the topic of our first CECAM Workshop in September 2018.

The last year has witnessed mounting indications that the most promising excitonic insulator candidates are 2d materials. These candidates however exhibit also other kinds of order: a variety that includes topological insulators [4,13-19], ferroelectrics [19,20], unconventional superconductors [21-23], often depending on tiny variations of tunable parameters, such as doping, pressure, strain. This has introduced new urgent and far-reaching questions, concerning the role of excitonic correlations in a plethora of allegedly unrelated phenomena, whose interplay is just beginning to be explored. At the same time, the long-term challenge of establishing the excitonic insulator through the signatures of macroscopic quantum coherence is attracting renewed interest in this class of materials.

By collecting the key actors of theoretical and experimental research, who are spread among different communities, this Workshop aims at in-depth analysis of common themes and novel challenges, both theoretical and computational, to progress our understanding of interacting systems in low dimensions.

Key References

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2. Program

Day 1 - Monday June 21st 2021

- 14:15 to 14:30 - Welcome & Introduction
- 14:30 to 14:55 - **Allan H. MacDonald**
  In praise of indirectness: the phenomenology of bilayer exciton condensates
- 14:55 to 15:20 - **Shahal Ilani**
  Visualizing the quantum phases of magic angle graphene
- 15:20 to 15:45 - **Daniele Varsano**
  Evidence of ideal excitonic insulator in MoS_2 under pressure
- 15:45 to 16:00 - Coffee break
- 16:00 to 16:25 - **Kamran Behnia**
  Is the field-induced insulating state in graphite a BEC of excitons?
- 16:25 to 16:50 - **Matteo Calandra**
  Light-tunable charge density wave orders in $MoTe_2$ and WTe_2 single layers
- 16:50 to 17:15 - **David Cobden**
  Evidence for equilibrium excitons in monolayer WTe_2
- 17:15 to 18:00 - Discussion (chair: P. Abbamonte)

Day 2 - Tuesday June 22nd 2021

- 14:30 to 14:55 - **Peter Abbamonte**
  Collective excitations of the correlated metal Sr_2RuO_4
- 14:55 to 15:20 - **Antoine Georges**
  Is Ta_2NiSe_5 an excitonic insulator?
- 15:20 to 15:45 - **Stefan Kaiser**
  Light-driven coherence in excitonic insulators
- 15:45 to 16:45 - Poster session
- 16:45 to 17:10 - **Ludger Wirtz**
  The carbon chain revisited: excitonic-insulator instability and Peierls distortion
- 17:10 to 17:35 - **Sandro Sorella**
  Insulating (excitonic?) phases in the honeycomb lattice Hubbard-Holstein model
- 17:35 to 18:00 - **Gianluca Stefanucci**
  Coherent versus incoherent excitons: stability, time-dependent ARPES spectrum and Floquet topological phases

Day 3 - Wednesday June 23rd 2021

- 14:30 to 14:55 - **Jan Kunes**
  Superfluid, solid and supersolid in two-orbital Hubbard model
- 14:55 to 15:20 - **Enrico Da Como**
  Dynamics of excitonic and mott charge density waves in transition metal dichalcogenides
- 15:20 to 15:45 - **Friedhelm Bechstedt**
  Excitonic effects, infrared absorbance, and quantum spin Hall phase in Xenes and related 2D crystals
- 15:45 to 16:00 - Coffee break
- 16:00 to 16:25 - **Peter Littlewood**
  Novel dynamical phase transition in exciton-polariton condensates: and a generalization to other multi-component non-reciprocal interacting systems
- 16:25 to 16:50 - **Simone Latini**
  Collective behaviour in quantum materials: microscopic origin of the phase transitions in Ta_2NiSe_5 and cavity induced ferroelectricity in SrTiO3
- 16:50 to 17:15 - **Lucia Reining**
  The collective nature of excitons
- 17:15 to 18:00 - Discussion (chair: A. MacDonald)
- 18:00 to 18:10 - Closing Word

3. Abstracts

Coherent versus incoherent excitons: stability, time-dependent ARPES spectrum and Floquet topological phases

Gianluca Stefanucci
University of Rome, Italy

TBA

Collective behaviour in quantum materials: microscopic origin of the phase transitions in Ta_2NiSe_5 and cavity induced ferroelectricity in $SrTiO_3$

Simone Latini
MPI Hamburg, Germany

The possible existence of an excitonic-insulator phase in Ta_2NiSe_5 has attracted a remarkable interest in this material and related compounds. The origin of the structural phase transition in Ta_2NiSe_5 has been rationalized in terms of crystal symmetries breaking driven by both electronic correlation and lattice distortion. However, the role of structural and electronic effects has yet to be disentangled. In this talk I will present an extensive investigation of the electronic and phononic effects involved in the phase transition in Ta_2NiSe_5 and Ta_2NiS_5 by means of extensive first- principles calculations based on increasingly accurate density functionals and many-body theory methods [1]. I will show that, despite the difference in electronic behaviour, the two materials share a common structural origin of the phase transition which can be assigned to a phononic instability. A total energy landscape analysis in the broken symmetry phase shows no tendency towards a purely electronic instability and we find that in Ta_2NiSe_5 a sizeable lattice distortion is needed to open a bandgap. Importantly, I will discuss how, our calculations are compatible all experimental observations and recent experimental Raman findings. I will show that the microscopic origin of the structural phase transition in both Ta_2NiSe and TaNiS is the same.
While our first-principle analysis suggests that a collective excitonic transition is unlikely to drive the structural phase transition in Ta_2NiSe_5 I will propose an alternative route to alter the collective behaviour of a material by means of light trapped in a cavity. In an optical cavity, the confinement of light on a small region of space can result in strong light-matter coupling and hence alter the properties of the material altogether. Here I will demonstrate, based on first principles calculations, that the strong light-matter coupling in a cavity can induce a change of the collective phase from paraelectric to ferroelectric in the ground state of $SrTiO_3$ [2]. This is a light-matter-hybrid ground state which can only exist because of the coupling to the vacuum fluctuations of light, a photo-ground state. The atomistic simulations show the nuclear quantum fluctuations of the $SrTiO_3$ lattice, the ones responsible for the quantum paraelectricity of the $SrTiO_3$, can be suppressed by coupling to the vacuum fluctuations of light in a process that is analogous to dynamical localisation. I will further demonstrate that the effect of cavity induced localization extends to finite temperatures, even when thermal lattice fluctuations overcome the quantum ones. These findings ultimately demonstrate how exposing a material to confined light can change fundamental properties of its ground state as far as its crystal structure.

[1] L. Windgätter, M. Rösner, G. Mazza, H. Hübener, A. Georges, A. J. Millis, S. Latini, and A. Rubio (2021)
[2] S. Latini, D. Shin, S. A Sato, C. Schäfer, U. De Giovannini, H. Hübener and A. Rubio (2021)

Collective excitations of the correlated metal Sr_2RuO_4

Peter Abbamonte
University of Illinois, United States

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Dynamics of excitonic and Mott charge density waves in transition metal dichalcogenides

Enrico Da Como
University of Bath, United Kingdom

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Evidence for equilibrium excitons in monolayer WTe_2

David Cobden
University of Washington, United States

TBA

Evidence of ideal excitonic insulator in MoS_2 under pressure

Daniele Varsano
CNR, Italy

Spontaneous condensation of excitons is a long-sought phenomenon analogous to the condensation of Cooper pairs in a superconductor. It is expected to occur in a semiconductor at thermodynamic equilibrium if the binding energy of the excitons - electron (*e*) and hole (*h*) pairs interacting by Coulomb force - overcomes the band gap, giving rise to a new phase: the “excitonic insulator” (EI). Transition metal dichalcogenides are excellent candidates for the EI realization because of reduced Coulomb screening, and indeed a structural phase transition was observed in few-layer systems. However, previous work could not disentangle to which extent the origin of the transition was in the formation of bound excitons or in the softening of a phonon. Here [1] we focus on bulk MoS_2 and demonstrate theoretically that at high pressure it is prone to the condensation of genuine excitons of finite momentum, whereas the phonon dispersion remains regular. Starting from first-principles many-body perturbation theory, we also predict that the self-consistent electronic charge density of the EI sustains an out-of-plane permanent electric dipole moment with an antiferroelectric texture in the layer plane: At the onset of the EI phase, those optical phonons that share the exciton momentum provide a unique Raman fingerprint for the EI formation. Finally, we identify such fingerprint in a Raman feature that was previously observed experimentally, thus providing direct spectroscopic confirmation of an ideal excitonic insulator phase in bulk MoS_2 above 30 GPa.

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Excitonic effects, infrared absorbance, and quantum spin Hall phase in Xenes and related 2D crystals

Friedhelm Bechstedt
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The rediscovery of graphene and its peculiar physical properties have caused intense research on two-dimensional (2D) sheet crystals. Hexagonal graphene-like, but buckled 2D crystals of other group-IV elements, Xenes such as silicene, germanene and stanene, are predicted to be ‘wonder’ materials since their honeycomb structure is responsible for Dirac electrons. Spin-orbit coupling (SOC) increasing along the group IV of the periodic table opens a fundamental gap. The accompanying band inversion gives rise to topological and quantum spin Hall insulators, respectively.
In this talk important consequences of the linear band structure, the small SOC-induced gap, and the low screening on two-particle electron-hole excitations in frequency-dependent optical and spin Hall conductivities are discussed:

1. The linear bands have enormous consequences for the IR optical properties. Independent of the group-IV element and the degree of hybridization the universal absorbance is ruled by the Sommerfeld fine structure constant [1].

2. Just above the Dirac cones, in the visible spectral range, the optical properties are significantly influenced by excitonic effects [2]. Saddle-point excitons appear near the M1 van Hove singularity [3,4].
3. Spin-orbit interaction not only modifies the optical properties by band gap opening [5]. It makes the 2D crystals to topological insulators which exhibit a quantization of the static spin Hall conductivity [6, 7].
4. Because of the reduced screening the nominal band-edge exciton binding energy exceeds the gap suggesting an excitonic insulator (EI) phase [8]. Vertical electric fields may induce a phase transition between EI and trivial phase.

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In praise of indirectness: the phenomenology of bilayer exciton condensates

Allan H. MacDonald
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Insulating (excitonic?) phases in the honeycomb lattice Hubbard-Holstein model

Sandro Sorella
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Is Ta_2NiSe_5 an excitonic insulator?

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Is the field-induced insulating state in graphite a BEC of excitons?

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Light-driven coherence in excitonic insulators

Stefan Kaiser
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Light-matter phenomena in quantum materials
Angel Rubio
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Light-tunable charge density wave orders in $MoTe_2$ and WTe_2 single layers
Matteo Calandra
University of Trento, Italy

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Novel dynamical phase transition in exciton-polariton condensates: and a generalization to other multi-component non-reciprocal interacting systems
Peter Littlewood
University of Chicago, United States

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Superfluid, solid and supersolid in two-orbital Hubbard model
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TU Wien, Austria

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The carbon chain revisited: excitonic-insulator instability and Peierls distortion
Ludger Wirtz
Université du Luxembourg, Luxembourg

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The collective nature of excitons
Lucia Reining
CNRS-École Polytechnique, France

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Visualizing the quantum phases of magic angle graphene
Shahal Ilani
Weizmann Institute, Israel

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4. Posters

Competitive screening and band gap renormalization in n-type monolayer transition metal dichalcogenides

S. Samaneh Ataei, Ali Sadeghi
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Gate induced carriers impact the many-body interactions in monolayer Transition Metal Dichalcogenides (TMDs) by modifying the screened Coulomb potential and renormalizing the band gap, thus influencing the strong excitonic effects in these materials. Using the GW approximation and a plasmon pole theory to model the carrier induced plasmons in the frequency-dependent part of the screening, we accurately calculate the band gap renormalization of the electron doped monolayer MoS2 and WS2. The excitonic states of the low doped systems are calculated by solving the Bethe-Salpeter equation. Our results clarify the competition between screening and band gap renormalization. An exact cancellation occurs between the reduced band gap and the exciton binding energy for doped monolayer WS2, in good agreement with previous experimental results. In contrast, the exciton energy of doped monolayer MoS2 blueshifts by tens of meV. We show the role of the electronic band structure of the monolayers in the calculated exciton energies. Our results could be generally expanded to other monolayer TMDs and is helpful for quantitatively engineering the opto-electronic devices with desired features.

Exciton g-factors of van der Waals heterostructures from first principles calculations

Tomasz Woźniak[1], Paulo Faria Junior[2], Andrey Chaves[3], Gotthard Seifert[4], Jens Kunstmann[4]
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[3]Universidade Federal do Ceará, Fortaleza, Brazil
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External fields are a powerful tool to probe optical excitations in materials. The linear energy shift of an excitation in a magnetic field is quantified by its effective g-factor. Here we show how exciton g-factors and their sign can be determined by converged first principles calculations. We apply the method to monolayer (1L) excitons in semiconducting transition metal dichalcogenides (TMDs) and to interlayer excitons in $MoSe_2/WSe_2$ heterobilayers and obtain excellent agreement with recent experimental data. The precision of our method allows to assign measured g-factors of optical peaks to specific transitions in the band structure and also to specific regions of the samples. This revealed the nature of various, previously measured interlayer exciton peaks. We further show that, due to specific optical selection rules, g-factors in van der Waals heterostructures are strongly spin and stacking-dependent. The presented approach can potentially be applied to a wide variety of semiconductors [1].
The method was successfully applied to bigger excitonic complexes: trions, their phonon replicas and biexcitons in 1L WS_2. It also overcomes the currently used simple models, yielding g-factors of individual electrons and holes in perfect agreement with their experimental values [2].

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Excitonic effects on two-dimensional transition-metal dichalcogenide monolayers: impact on solar cell efficiency

Alexandre Cavalheiro Dias[1], Helena Bragança[2], João Paulo A. de Mendonça[1], Juarez L. F. Da Silva[1]
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The search for 2D systems for applications in solar cells has continuously challenged our community. In this work we performed a screening of 2D monolayers composed by transition metals from group IV-XI and the chalcogenides S, Se and Te. We combine DFT, MLWF-TB and BSE formalism to investigate the electronic, optical and excitonic properties of these systems, using a machine learning correlation analysis of the generated data to identify non-trivial correlations between optical and excitonic properties. From the 72 2H-TMD monolayers, phonon calculations show 22 stable systems, which 14 are semiconductors. Our results shows that the presence of excitons affects band alignment and PCE. We find high-efficiency vdW heterostructures of solar cells and observe a strong linear correlation between exciton energy and electronic band gap for the stable semiconductors.

Excitonic vs mott insulator in carbon nanotubes: a proposed experimental test

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Ultraclean, undoped carbon nanotubes are always insulating, even when the gap predicted by band theory is zero. The residual, intrinsic gap is thought to have a many-body origin, associated with either a Mott or an excitonic phase. Whereas the two scenarios are fundamentally different, as they are driven by the short- and long-range part of Coulomb interaction, respectively, a conclusive experiment has been missing so far. Here we propose as a unique fingerprint of the excitonic insulator the presence of a cusp in the evolution of the gap with the axial magnetic field, close to the gap minimum. On the contrary, the Mott phase exhibits a featureless, rounded profile. The non-analytic spike originates from the extreme sensitivity of electron-electron interactions to the Aharonov-Bohm gap modulation, as we demonstrate by combining a first-principles analysis of screening with extensive model calculations for tubes of different size and chirality.

Excitons and excitonic insulator in WTe_2: insight from theory

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We present here the details of the theory reported in the talk by David Cobden: 'Evidence for equilibrium excitons in monolayer WTe_2'.

5. Participant list

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6. Additional Information

CECAM website for this workshop:
https://www.cecam.org/workshop-details/21

Notes: