Quantum optics and magnetism in 2D materials

Atac Imamoglu

ETH Zurich

Presented by: Martin Kroner

Motivation:
A new regime of cavity-QED with nonperturbative interactions between photons/polaritons and itinerant electrons or magnons?
Outline

1) **Strong light-matter coupling in semiconducting transition metal dichalcogenide (TMD) monolayers:**
   - Realization of an atomically thick mirror using monolayer MoSe$_2$
   - Observation of robust exciton-polaritons

2) **Strong exciton-electron interaction in TMDs:**
   - *exciton-polarons* as elementary many-body optical excitations
   - nonequilibrium dynamics of a mobile quantum impurity

3) **Giant valley/spin susceptibility in monolayer MoSe$_2$**
A new class of 2D materials: Transition metal dichalcogenides (TMD)

Formula: MX$_2$
M = Transition Metal
X = Chalcogen

Layered materials

Effective monolayer

Electrical property | Material
---|---
Semiconducting | MoS$_2$, MoSe$_2$, WS$_2$, WSe$_2$, MoTe$_2$, WTe$_2$
Semimetallic | TiS$_2$, TiSe$_2$
Ferromagnetic | Cr$_3$I$_3$, CrBr$_3$
Metallic, CDW, Superconducting | NbSe$_2$, NbS$_2$, NbTe$_2$, TaS$_2$, TaSe$_2$, TaTe$_2$
Transition metal dichalcogenides (TMD)

- Monolayer TMD has a honeycomb lattice
- Unlike graphene, inversion symmetry is broken
- Valley semiconductor: physics in $\pm K$ valleys

$$H_K = v \begin{pmatrix} \Delta/2\sqrt{3} & p_x - ip_y \\ p_x + ip_y & -\Delta/2\sqrt{3} \end{pmatrix}$$

(Unlike graphene, 2-band model only provides a qualitative description)

- Monolayers of TMDs can be combined with other 2D materials to make van der Waals heterostructures with novel properties
For this talk: a 2D charge tunable valley semiconductor with strongly bound excitons - emphasis on interactions

Pioneering work: Heinz, Xu
Magnetic 2D materials

(Mak & Shan)

- Magnetic material with a bandgap of 2 eV
- Exchange fields up to 13 T in WSe2
Photoluminescence (PL) from 2D materials

- Due to strong Coulomb interactions, electrons and holes form strongly bound states before they recombine: PL is dominated by decay of an exciton.

Exciton binding energy:
- ~10 meV, band-gap ~1.5 eV (GaAs)
- ~0.5 eV, band-gap ~2.0 eV (TMD)
Photoluminescence (PL) from 2D materials

- Due to strong Coulomb interactions, electrons and holes form strongly bound states before they recombine: PL is dominated by decay of an exciton or a trion if QW has localized electrons.

Exciton binding energy:
- \( \approx 10 \text{ meV, bad-gap } \approx 1.5 \text{ eV (GaAs)} \)
- \( \approx 0.5 \text{ eV, band-gap } \approx 2.0 \text{ eV (TMD)} \)

Exciton+electron form a trion, an H-like molecule with binding energy:
- \( \Delta \approx 1 \text{ meV (GaAs)} \approx 25 \text{ meV (TMD)} \)
Photoluminescence (PL) from 2D materials

- Due to strong Coulomb interactions, electrons and holes form strongly bound states before they recombine: PL is dominated by decay of an exciton or a trion if QW has localized electrons.

Exciton linewidth of MoSe2 in hBN is comparable to the radiative decay rate.
Implications of strong exciton binding
≡ small Bohr radius $a_B$

- TMD excitons couple very strongly to resonant photons:
  - ultrafast /sub-ps radiative decay rate ($\sim 1/a_B^2$) $\Gamma_{rad} \sim 1.5$ meV
  - strong reversible coupling to cavities ($\sim 1/a_B$) $g \sim 10$-40 meV

- State-of-the art TMD monolayers have nearly radiative decay limited exciton linewidths
  - resonant coherent light scattering - not incoherent absorption!
Monolayer MoSe$_2$: atomically thin mirror?

In-plane momentum conservation ensures that for radiatively broadened 2D exciton resonance, incident resonant light experiences perfect 100% specular reflection.

- High reflection or extinction of transmission on resonance only possible for spontaneous emission broadened excitons
- Equivalent to a single atom coupled to a 1D reservoir/waveguide

Theory: Zeytinoglu et al. arxiv 1701.08228; related work on atomic arrays: Adams & Lukin-Yelin groups
Exciton dispersion

- Incident photons with in-plane momentum $k$ generate excitons with identical momentum (translational invariance)
- Secondary field generated by excitons interferes with the external field to modify transmission and lead to reflection
- Only excitons within the light cone couple to light; disorder scatters excitons to dark states – leads to real absorption
Realization of an atomically thin mirror

- 90% extinction of transmission
- 45% peak monolayer reflection

→ Demonstrates predominantly radiatively broadened excitons

(see also Kim-Lukin-Park & Shan-Mak results - up to 80% reflection)
Realization of an atomically thin mirror

→ For pure radiative decay, we should have $R + T = 1$, for all $\omega_{\text{inc}}$
→ Also, for pure inhomogeneous broadening $R + T = 1$, for all $\omega_{\text{inc}}$
→ $R + T$ lineshape is non-Lorentzian – due to scattering into $k \neq 0$
A suspended atomically thin mirror (Mak & Shan)

A new paradigm for optomechanics
Cavity-polaritons with 2D materials

- Tunable vacuum field strength and long cavity lifetime allowing for high-precision spectroscopy
- Versatile platform for cavity-QED with any material system
Strong coupling regime

- Large normal mode splitting: $\Omega_R = 17$ meV – new elementary excitations: exciton-polaritons

- Maximum reported splitting $> 50$ meV

Earlier results:
Menon, Tartakovskii
Rydberg blockade analog

Rydberg EIT/blockade

- Short exciton lifetime is irrelevant: the decay rate of polaritons is determined exclusively by their cavity nature – the more exciton like the polariton is, the longer the decay time
Contrary to common wisdom, it is not possible to observe a (sharp) trion peak in absorption or emission from an ideal degenerate 2DES.

- Direct formation of a trion in absorption is \( \propto \) to the probability of finding a \( k \sim 0 \) exciton in a strongly bound trion \( \propto (k_{\text{photon}} a_B)^2 \).
- The radiative decay of a \( k=0 \) (lowest energy) trion has to produce a \( k=0 \) electron - Pauli-blocking when \( E_F > 0 \) (not the case for localized electrons).
Optical excitations out of a 2DES

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  - Direct formation of a trion in absorption is \( \propto \) to the probability of finding a \( k \sim 0 \) exciton in a strongly bound trion \( \propto (k_{\text{photon}}a_B)^2 \)
  
  - The radiative decay of a \( k=0 \) (lowest energy) trion has to produce a \( k=0 \) electron - Pauli-blocking when \( E_F > 0 \) (not the case for localized electrons)

- This talk: proper description of the optical excitation spectrum is provided by many-body excitations termed exciton-polarons

  \[ \rightarrow \text{exciton as a finite-mass impurity in 2DES} \]
Electrical control of optical properties

- A van der Waals heterostructure incorporating a graphene layer on top of hBN/MoSe2/hBN layers allow for controlling charge density
- Ideal for investigating exciton-electron interactions
Carrier density dependent reflection

- Sharp increase in conductance indicates free carriers
- Reflection is strongly modified as electrons or holes injected
Carrier density dependent reflection & emission (PL)

Electron doped regime

- Sharp increase in conductance indicates free carriers
- Absorption & emission are different for high electron density
Fermi energy dependence of absorption spectrum

Horitontal line-cut: absorption (blue) + PL (green)
Fermi energy dependence of absorption spectrum

- Repulsive polaron
- Attractive polaron
Fermi energy dependence of the spectrum

Due to exciton-electron interactions, elementary optical excitations are exciton-polarons

\[ |\psi\rangle = \alpha |\text{bare exciton}\rangle + \beta |\text{exciton dressed with electron screening cloud}\rangle \]

\[ \downarrow \text{ensures strong light coupling} \]

\[ \downarrow \text{leads to red (attr. pol.) or blue (rep. pol.) shifted resonance} \]
Trion vs. attractive polaron

- **Trion**: H-like bound state of an exciton and electron
- Assume an exciton at \( r = 0 \)

**Trion**
- Excess charge
- Net charge = \(-e\)
- Small oscillator strength

**Attractive Polaron**
- Excess charge
- Charge neutral!
- Strong optical coupling

**Ansatz describes polaron & trion**

\[
|\Psi\rangle = \left( x_0^\dagger \phi_0 + \sum_{k > k_F, q < k_F} \phi_k c_k^\dagger c_q x_{k-q}^\dagger \right) |0_x\rangle |FS\rangle
\]
- simple Chevy Ansatz captures the repulsion of the two (polaron) resonances remarkably well (no fit parameters for splitting)

- The overall blue shift of the excitonic resonances due to phase space filling, screening and bandgap renormalization is a fit parameter.

(Sidler et al., Nat. Phys. 2017, Efimkin-MacDonald PRB 2017)
Strong cavity coupling

Monolayer is depleted of free electrons: only exciton resonance is visible: \( \Omega_R = 18 \text{ meV} \)

exciton-polaritons

Fermi energy \( E_F < E_T, \Omega_R \): both attractive & repulsive polarons are observable

exciton-polaron-polaritons
Strong cavity coupling

Monolayer is depleted of free electrons: only exciton resonance is visible: $\Omega_R = 18 \text{ meV}$

Fermi energy $E_F < E_T$, $\Omega_R$: both attractive & repulsive polarons are observable

Observation of polaron physics with an ultra-light mass (polariton) impurity

$E_F \sim E_T$, $\Omega_R$: only attractive-polaron polariton is observable: $\Omega_R = 7 \text{ meV}$

(Sidler et al. Nat. Phys. 2017)
New physics and applications

• Transport of polaritons (dressed with electrons) using external electric & magnetic fields (with F. Pientka, R. Schmidt & E. Demler)

• Polaritons mediating attractive interactions between electrons: light induced superconductivity (V. Ginzburg, W. Little, A. Kavokin)

• Polaritons dressed with anyons in FQHE regime

• Electrons mediating interactions between polaritons: a new method to enhance photon-photon interactions?
Giant spin susceptibility of TMD monolayers

- In comparison to GaAs, TMD monolayers exhibit:
  - large effective mass (small kinetic energy)
  - reduced screening (large exchange energy gain from spin/valley polarization)

- Itinerant ferromagnetism?
Resonant optical detection of electron valley polarization

- Trion and attractive polaron formation is only possible if the exciton and the electron occupy different valleys ↔ inter-valley trion.
- If electrons are valley polarized, trion formation and polaron absorption/emission is observed only for a single polarization.

Intra-valley trion in MoSe$_2$ is triplet – and is not bound.
Gate voltage dependent reflection at $B=7\,T$

- For $100\,V > V_{\text{gate}} > 70\,V$, $\sigma^+$ reflection is dominated by attractive polaron whereas that of $\sigma^-$ by exciton
- The electron density needed to observe $\sigma^-$ attractive polaron line is $1.6 \times 10^{12}\, \text{cm}^{-2}$
- In the absence of interactions, this would have required $g_{\text{elec}} = 38!$
For $B = 7T$, electrons are valley polarized in $-K$ valley and holes are valley polarized in $+K$ valley.

Hole-polaron PL is a factor of 2.5 stronger than electron-polaron!
How do we understand the B=7 Tesla Photoluminescence spectrum?
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trion formation is not possible since there are no electrons in -K valley and same-valley (triplet) trion is not bound.

\[ \text{trion formation with an electron in } K \text{ valley} \]
Super-paramagnetic response of MoSe$_2$

- In contrast to GaAs, MoSe$_2$ has a high electron mass and reduced screening: exchange energy gain from valley/spin polarization could exceed kinetic energy cost - towards Stoner instability?

- No magnetization for $B=0$T
  - but saturation for $B>5$T
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