University of Zurich[UZH]

Time-resolved photoemission of the excitonic insulator phase in a low dimensional material

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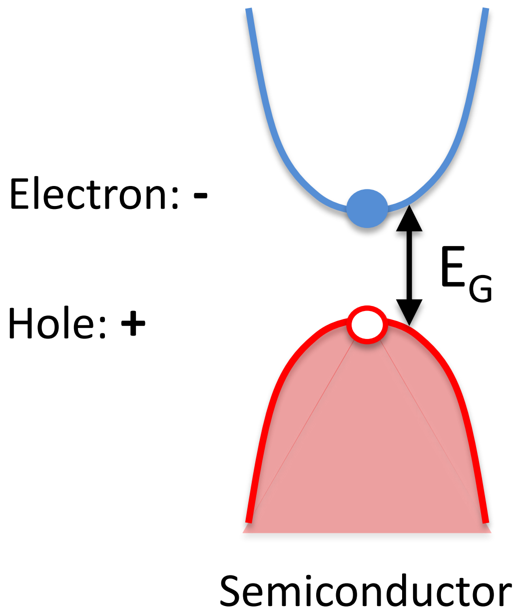
International School and Workshop on Electronic Crystals ECRYS-2017

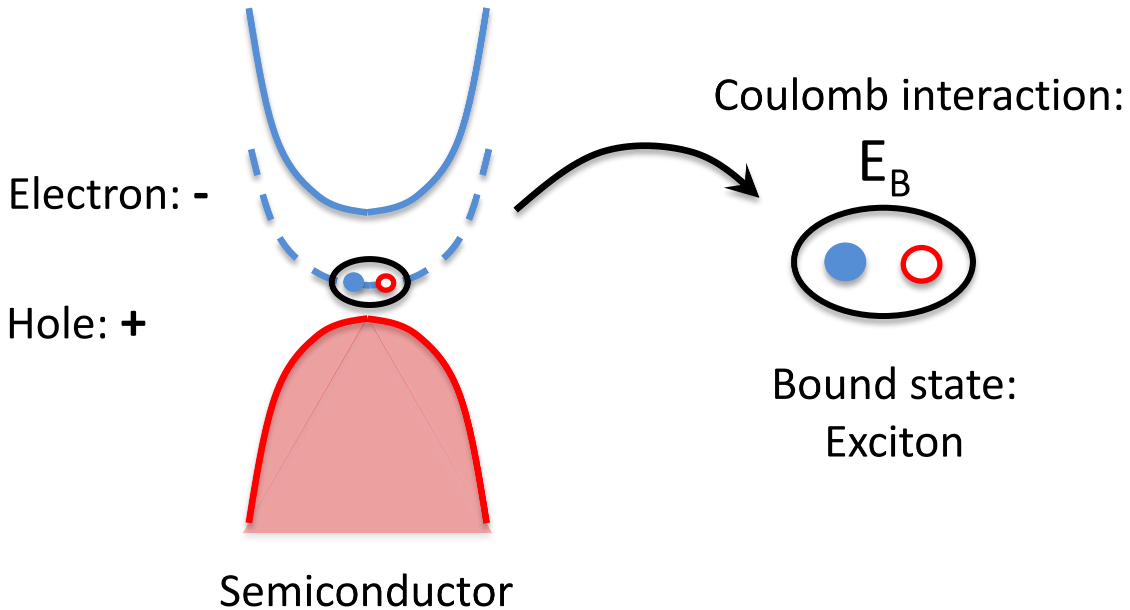
The excitonic insulator phase in solid state

The excitonic insulator phase: principle



The excitonic insulator phase in solid state

The excitonic insulator phase: principle

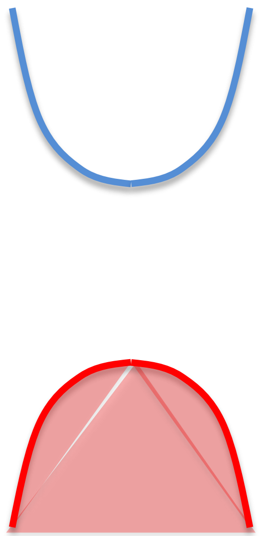


The excitonic insulator phase in solid state

The excitonic insulator phase: principle

Electron: **-**

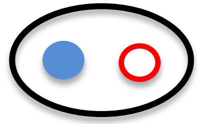
Hole: **+**



Semiconductor

Coulomb interaction:

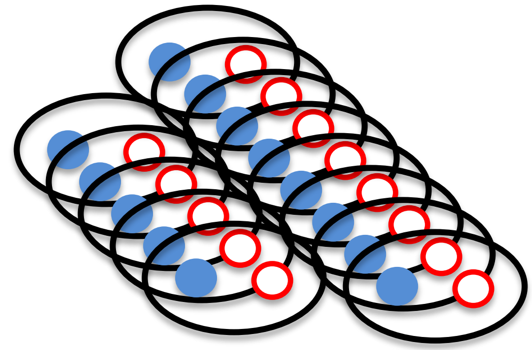
E_B



Bound state:
Exciton

if $E_B > E_G$

At low temperatures, $T < T_c$:



More and more excitons,
Longer lifetime

Spontaneous condensation of excitons ⟷ Macroscopic coherent state of excitons

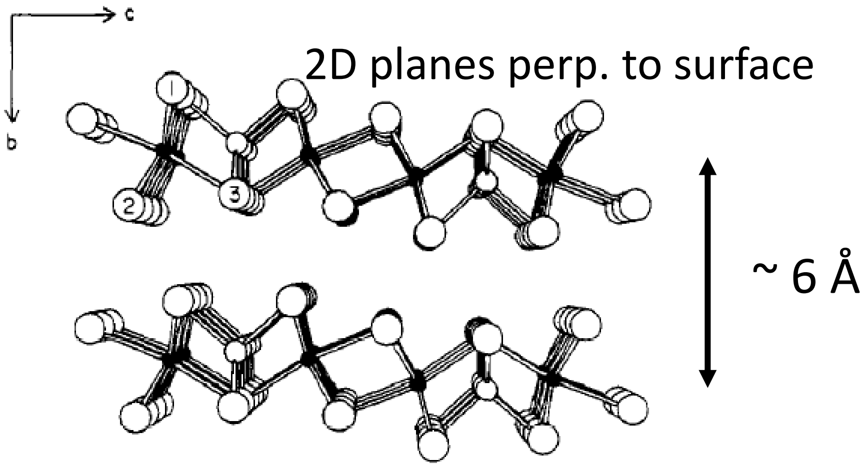
➡ Semiconductor-Insulator phase transition

(similar to the condensation of electron Cooper pairs in superconductivity)

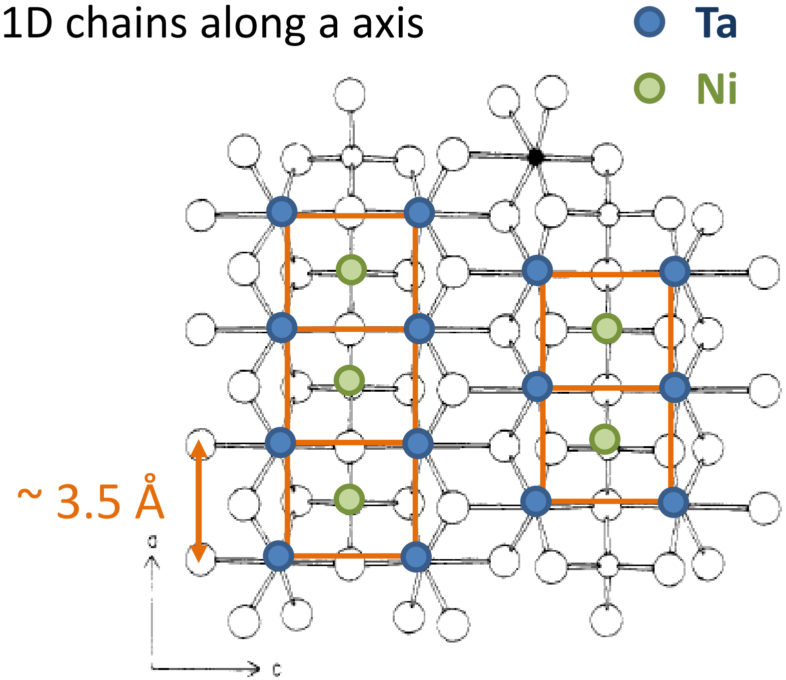
N.F. Mott, Phil. Mag. **6**, 287 (1961)
L.V. Keldysh and Y.V. Kopaev, Sov. Phys. Solid State **6**, 2219 (1965)
D. Jérome, T.M. Rice and W. Kohn, Phys. Rev. **158**, 462 (1967)

Physical properties of Ta_2NiSe_5

TNS is also a low-dimensional materials: quasi-1D chains along a axis



Di Salvo et al., J. Less. Comm. Metals, 116, 51 (1986)



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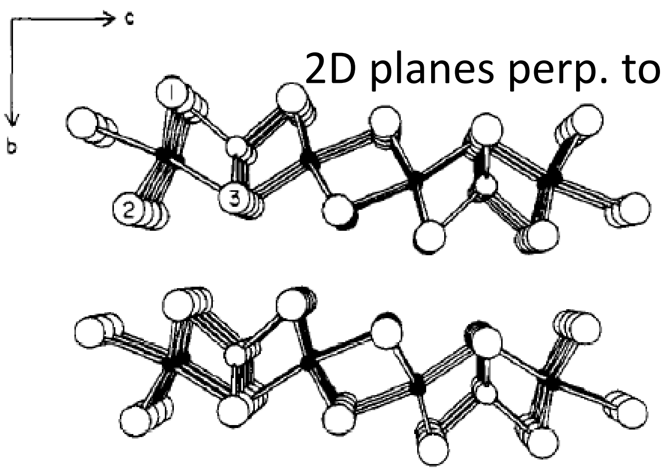
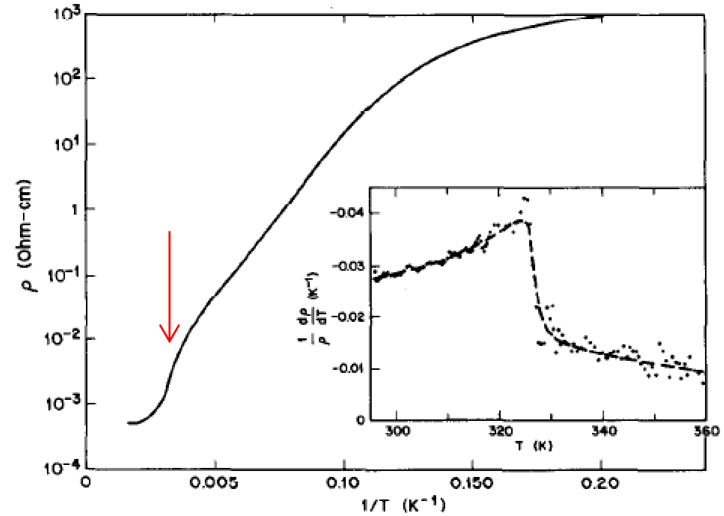
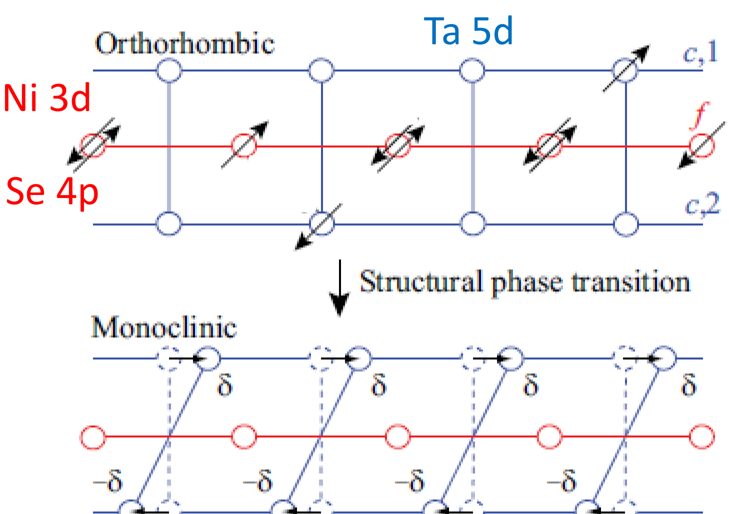


Di Salvo et al., J. Less. Comm. Metals, 116, 51 (1986)



Electrical resistivity along chains



Kaneko et al., PRB 87, 035121 (2013)

- **Semiconductor-semiconductor transition at 328 K accompanied by a structural transition:** orthorhombic -> monoclinic

Electronic structure of Ta_2NiSe_5

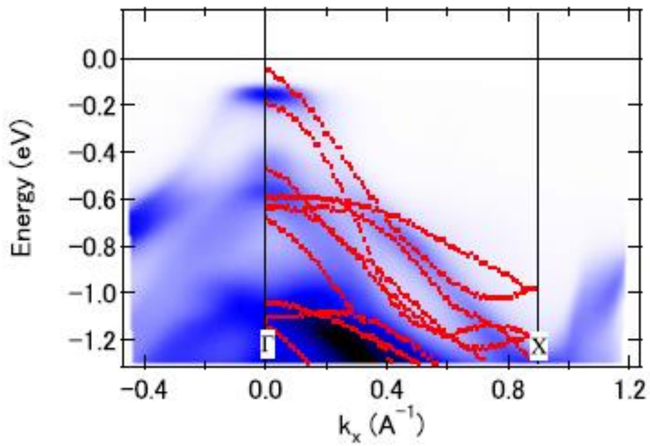
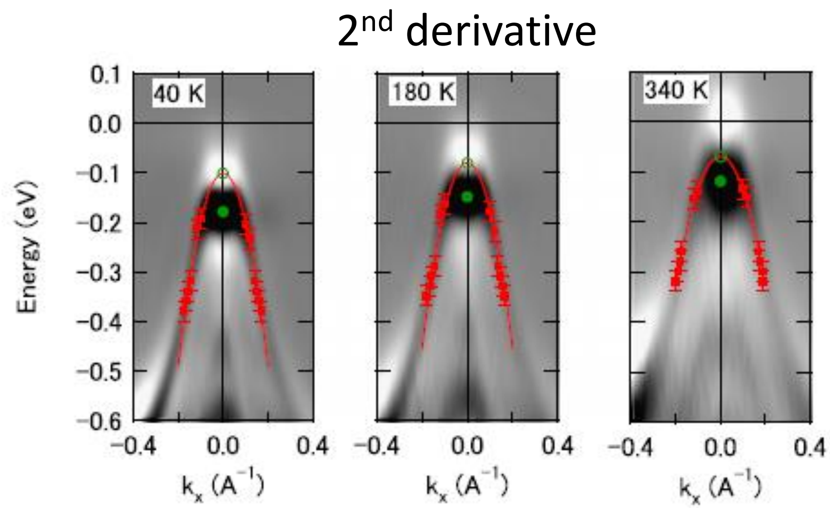
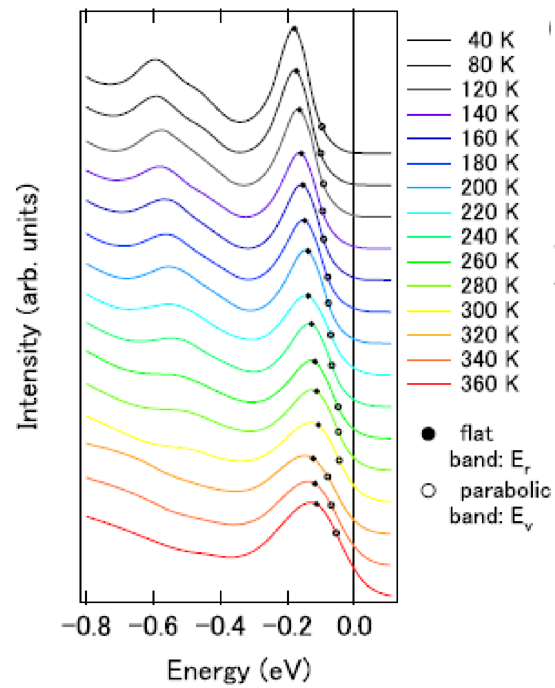


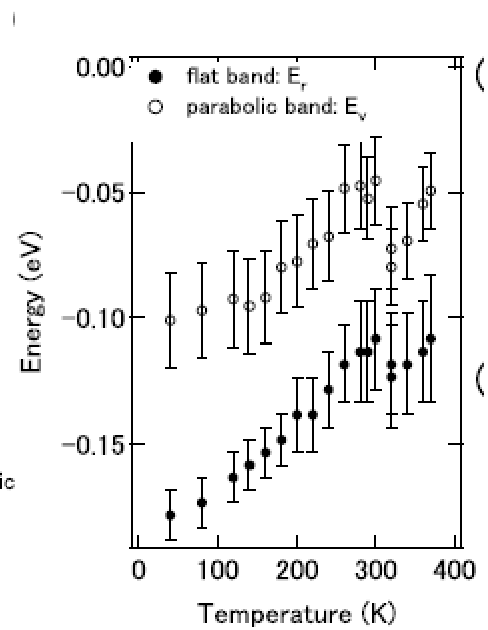
Fig. 1 Intensity plots of in-plane band dispersion of Ta_2NiSe_5 at 40 K measured with photon energy $h\nu = 23$ eV along the chain direction $\Gamma - X$ (**a**), and perpendicular to the chain direction $\Gamma - Z$ (**b**)



- Valence band dispersing at Γ, mostly along chain direction
- Flat top of the VB shifts down at lower temperatures
- Gap opening gradually below 328 K

Wakisaka et al., J. Supercond. Nov. Magn.25, 1231 (2012)



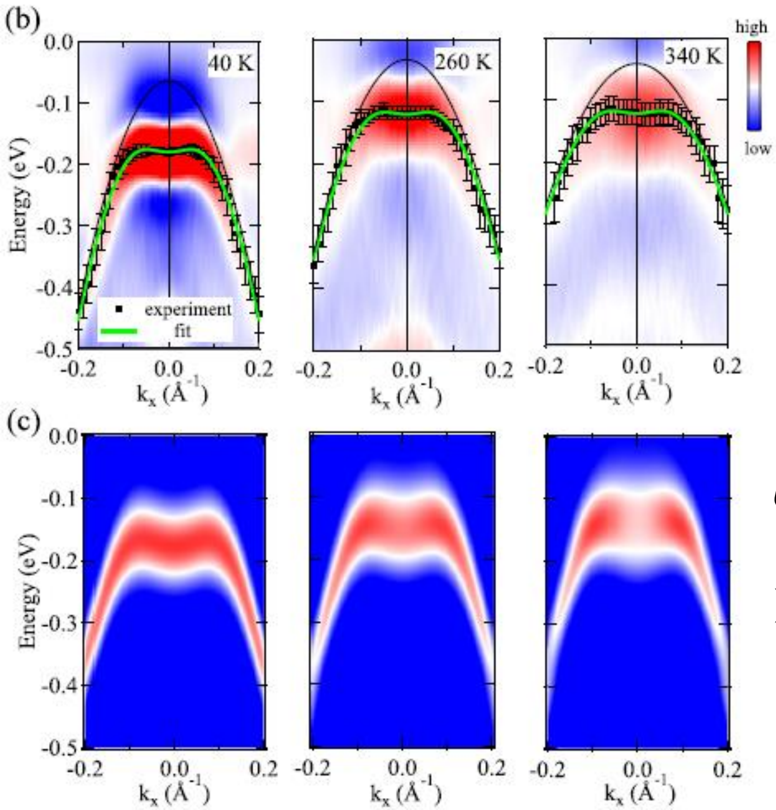


PHYSICAL REVIEW B 90, 155116 (2014)

Excitonic Bose-Einstein condensation in Ta_2NiSe_5 above room temperature

K. Seki,[1] Y. Wakisaka,[2] T. Kaneko,[1] T. Toriyama,[1] T. Konishi,[3] T. Sudayama,[2] N. L. Saini,[4] M. Arita,[5] H. Namatame,[5] M. Taniguchi,[5,6] N. Katayama,[7] M. Nohara,[8] H. Takagi,[9] T. Mizokawa,[2] and Y. Ohta[1]

We show that finite temperature variational cluster approximation (VCA) calculations on an extended Falicov-Kimball model can reproduce angle-resolved photoemission spectroscopy (ARPES) results on Ta_2NiSe_5 across a semiconductor-to-semiconductor structural phase transition at 325 K. We demonstrate that the characteristic temperature dependence of the flat-top valence band observed by ARPES is reproduced by the VCA calculation on the realistic model for an excitonic insulator only when the strong excitonic fluctuation is taken into account. The present calculations indicate that Ta_2NiSe_5 falls in the Bose-Einstein condensation regime of the excitonic insulator state.

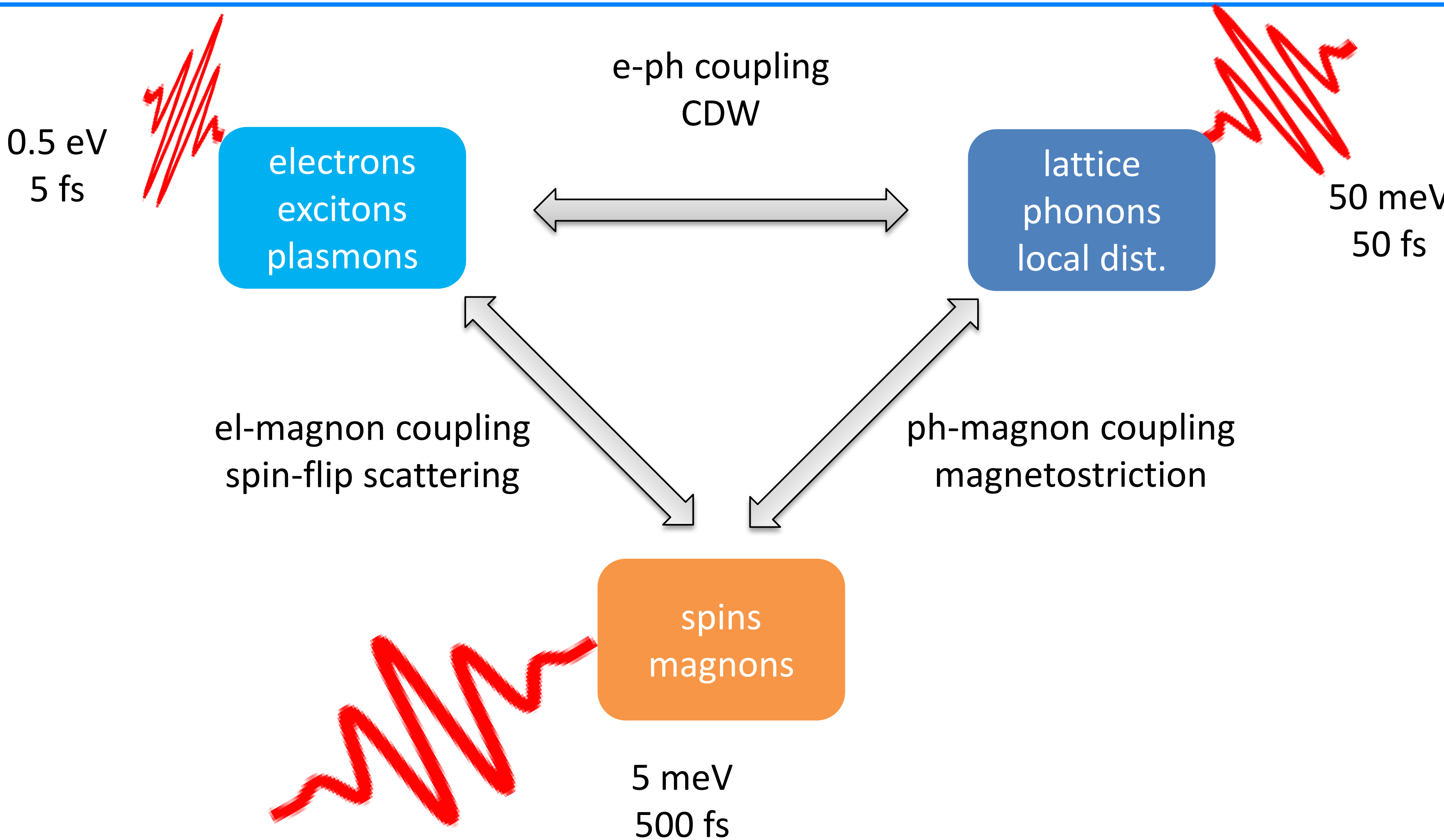


$$\mathcal{H} = -\sum_{\delta=x,y,z} t_c^\delta \sum_{\langle ij\rangle}(c_i^\dagger c_j + \mathrm{H.c.}) + (D/2-\mu)\sum_i n_{ic} - \sum_{\delta=x,y,z} t_f^\delta \sum_{\langle ij\rangle}(f_i^\dagger f_j + \mathrm{H.c.}) + (-D/2-\mu)\sum_i n_{if} + U\sum_i n_{ic}n_{if}, \quad (1$$

U: interorbital Coulomb repulsion between electrons

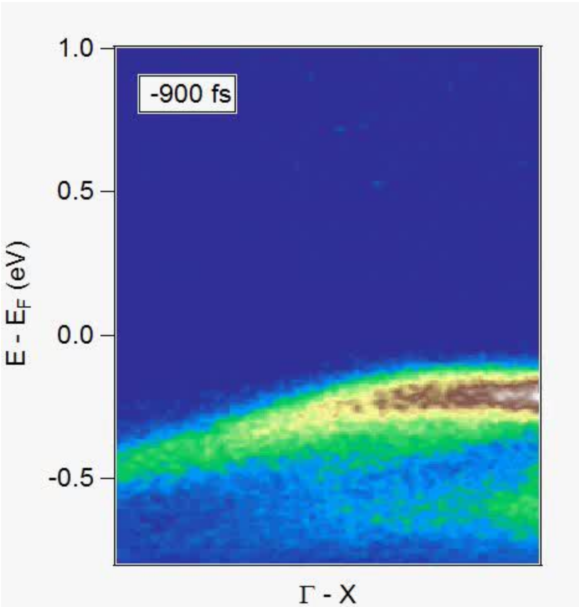
➢ excitonic insulator phase at LT (BEC regime)

Probing correlations in condensed matter by ultrafast techniques

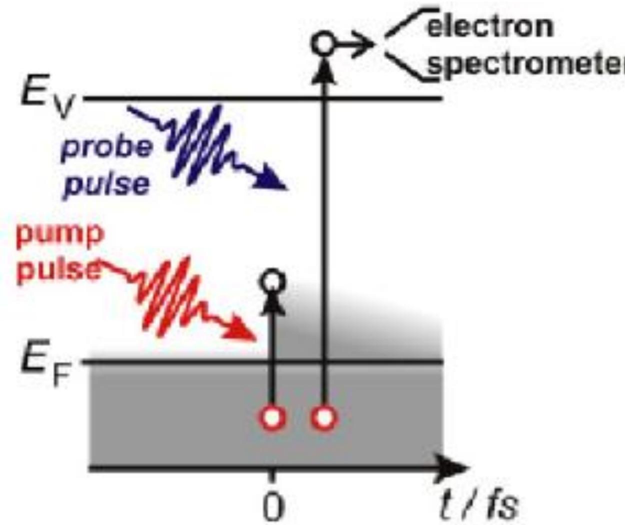


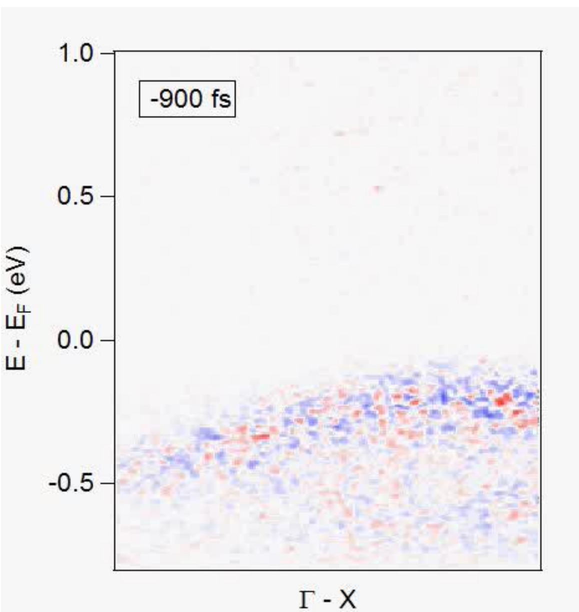
- Different DOFs respond on different timescales
- Generation of non-equilibrium states during the first hundreds fs

Time-resolved ARPES on Ta_2NiSe_5



Raw data





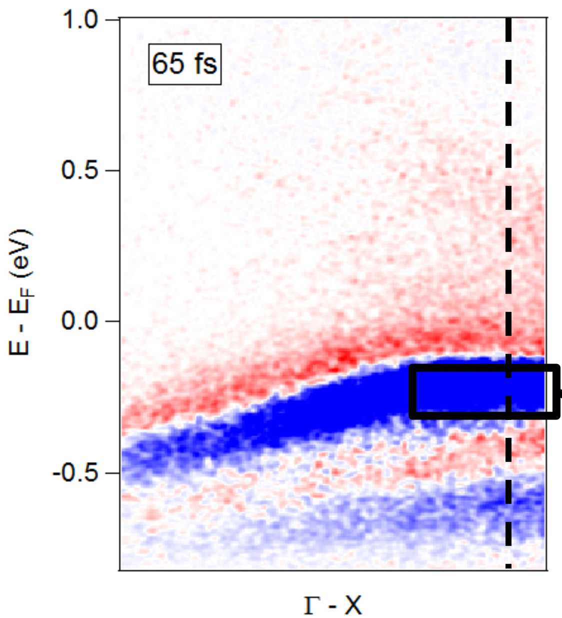
Difference data

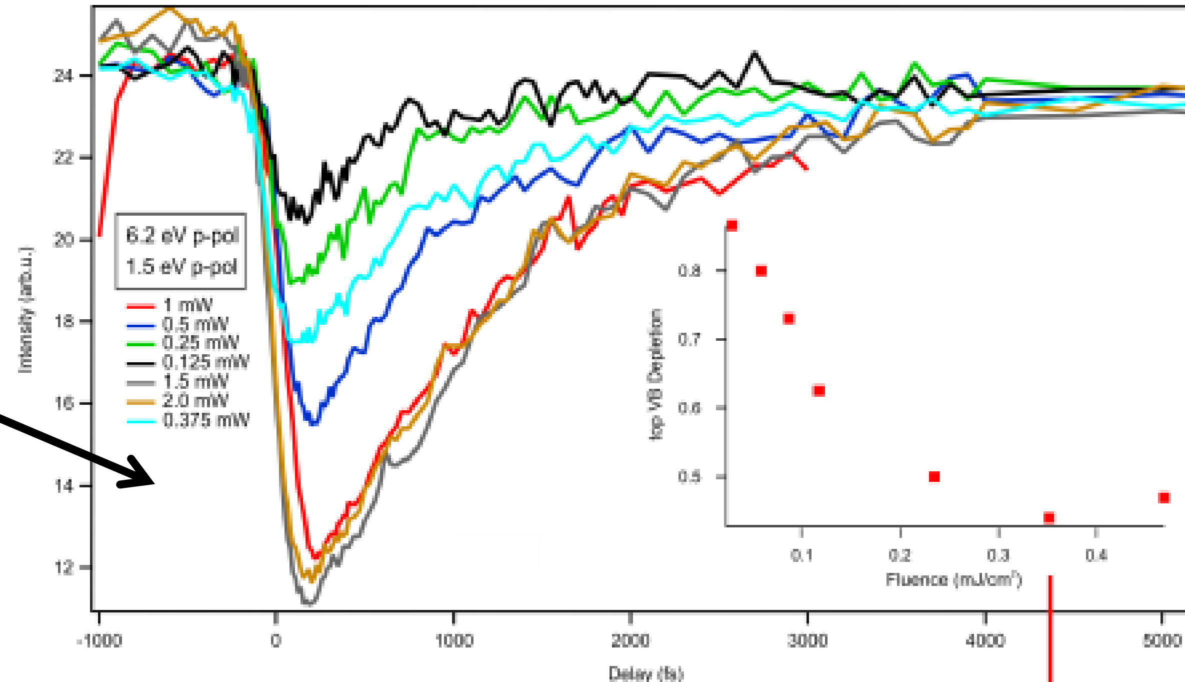
- Exciting with 800 nm (1.55 eV)
- Probing with 6.2 eV
- Performed at 40 kHz
- Sample kept at 110 K << 328 K

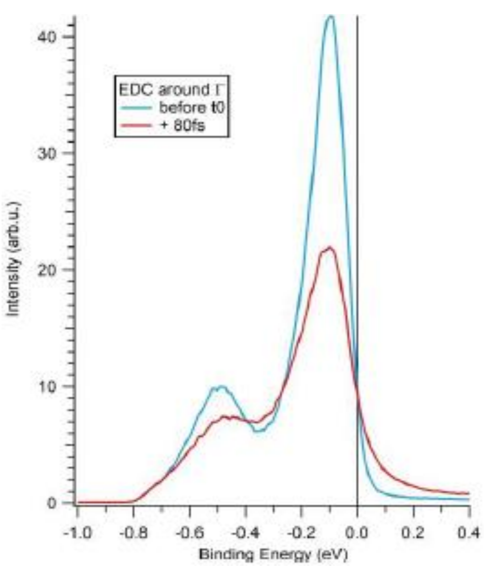
$I(t) – I(t<0)$

Time-resolved ARPES: Transient saturation at Γ

1PPE spectrum at 100K







min intensity of top VB population around t0

⇩

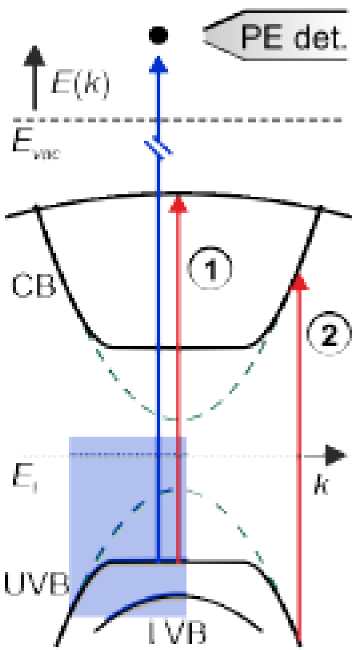
at 100 K saturation at

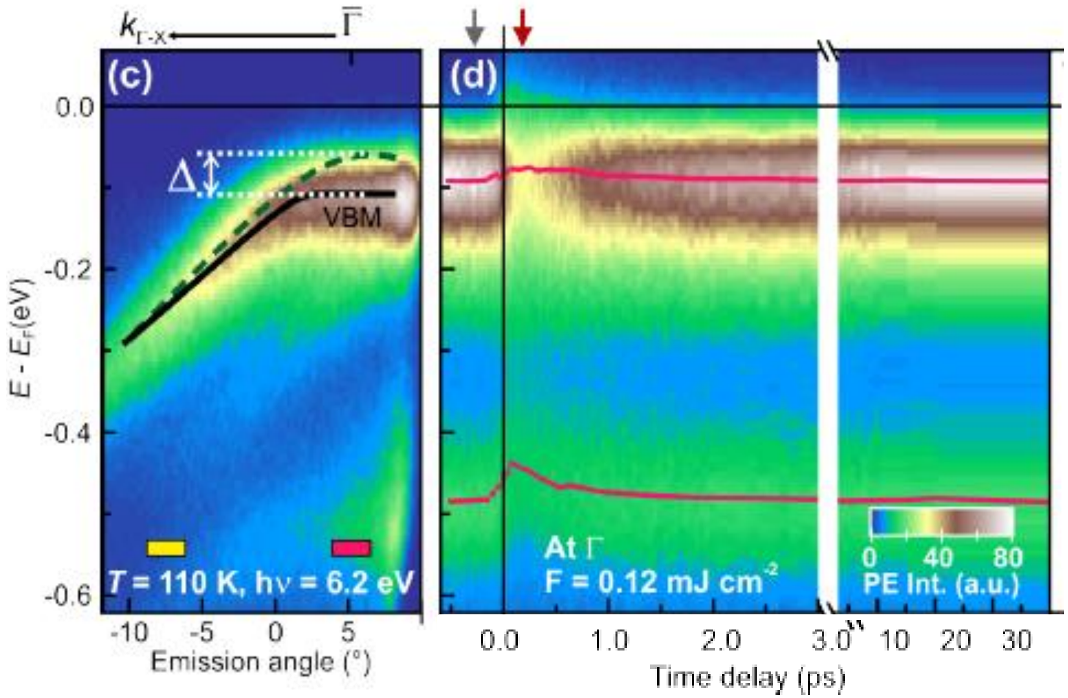
F_{sat} ~ 0.3 mJ/cm^2

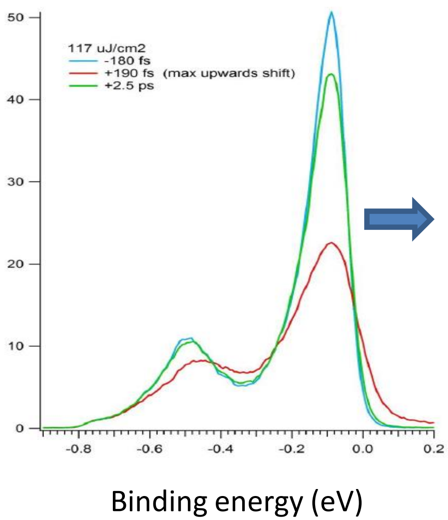
⇩

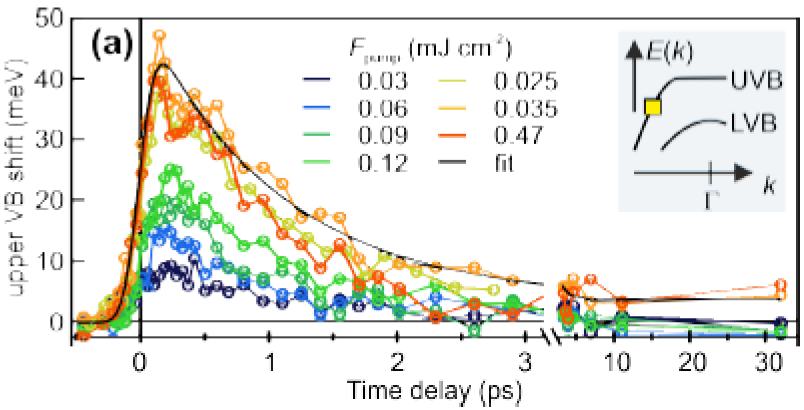
similar to optical F_c !

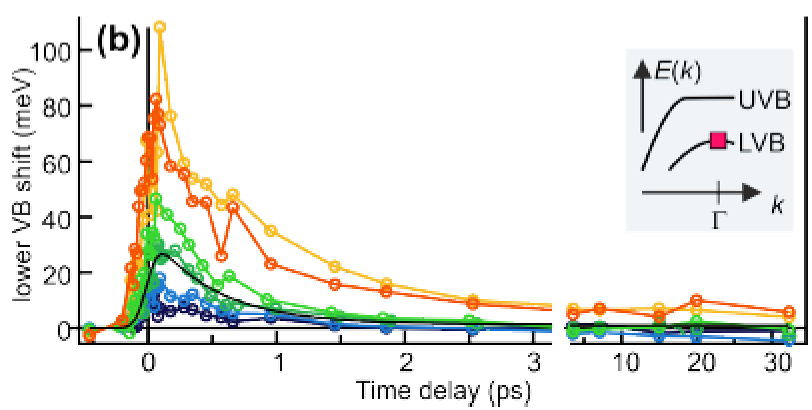
Time-resolved ARPES: Shrinking of the gap



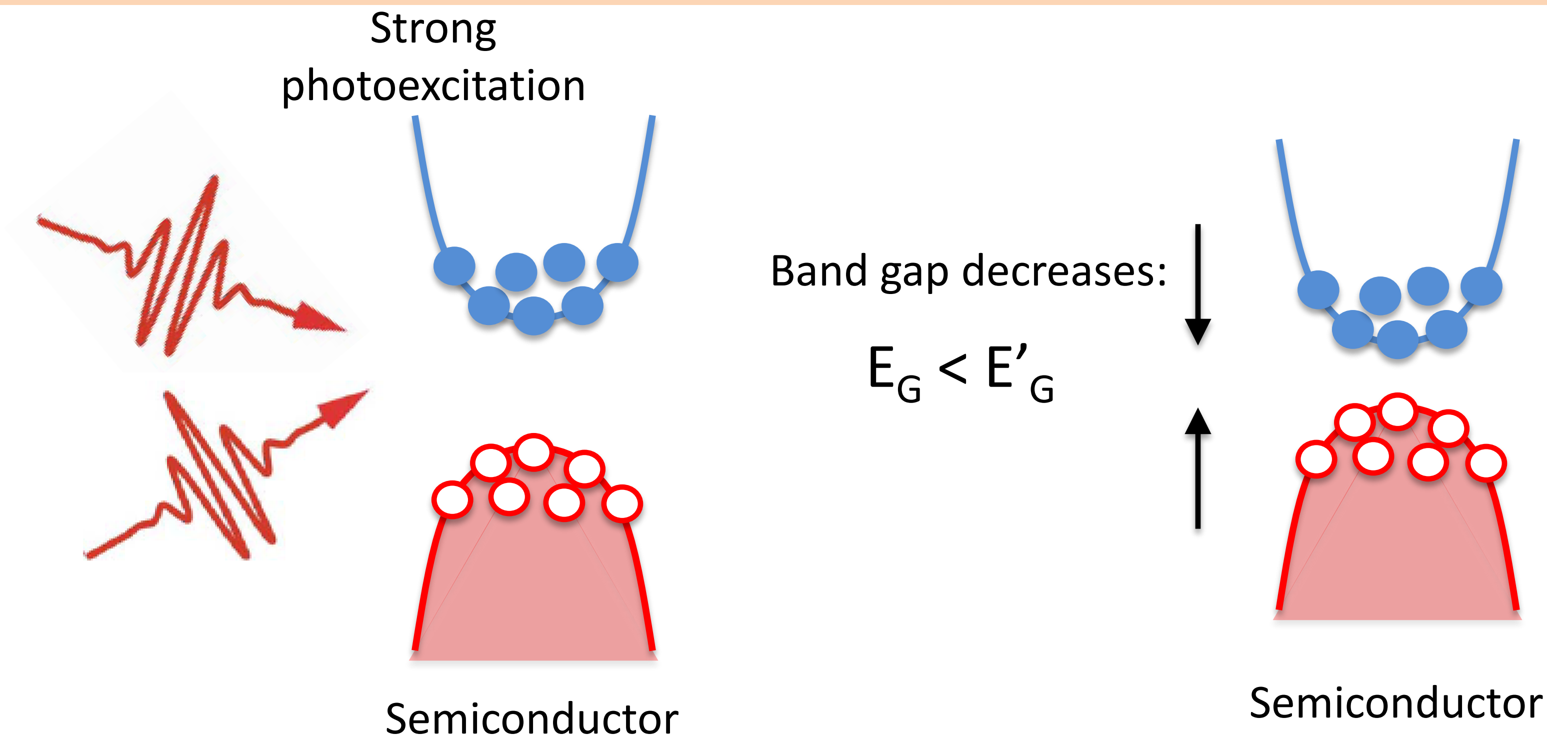








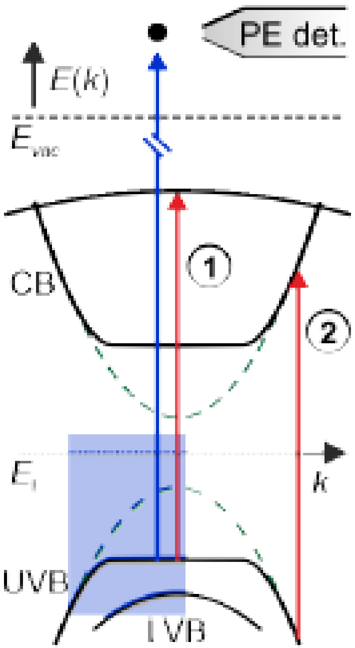
Behavior of a semiconductor under strong IR excitation

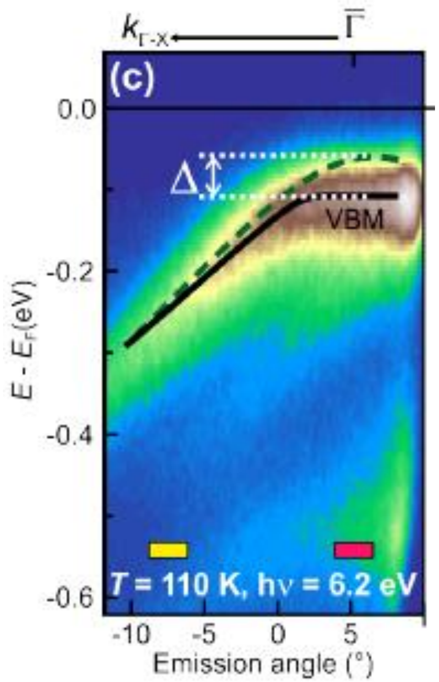


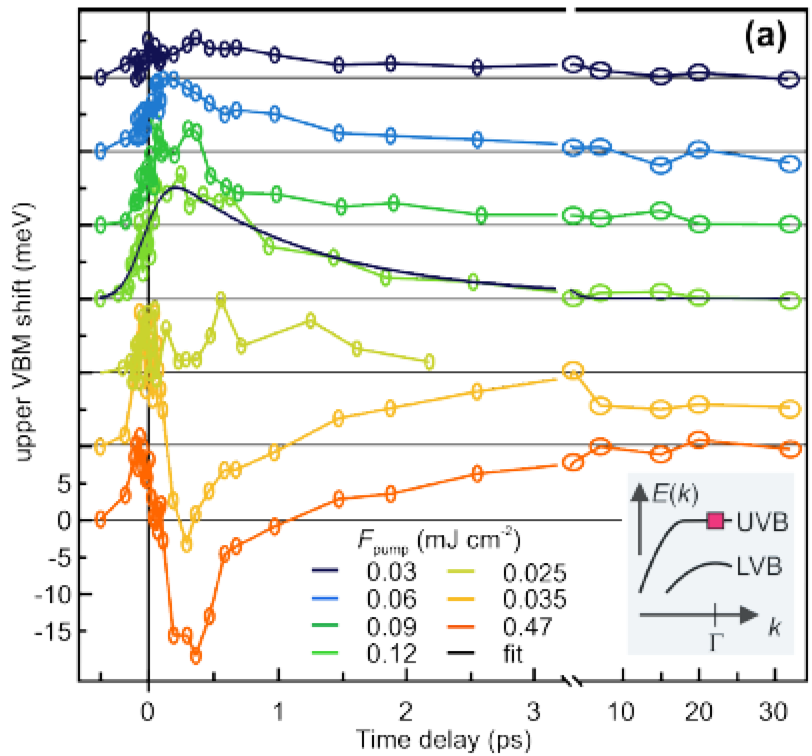
Band gap renormalization: the excited charge carriers enhance the screening of Coulomb interaction

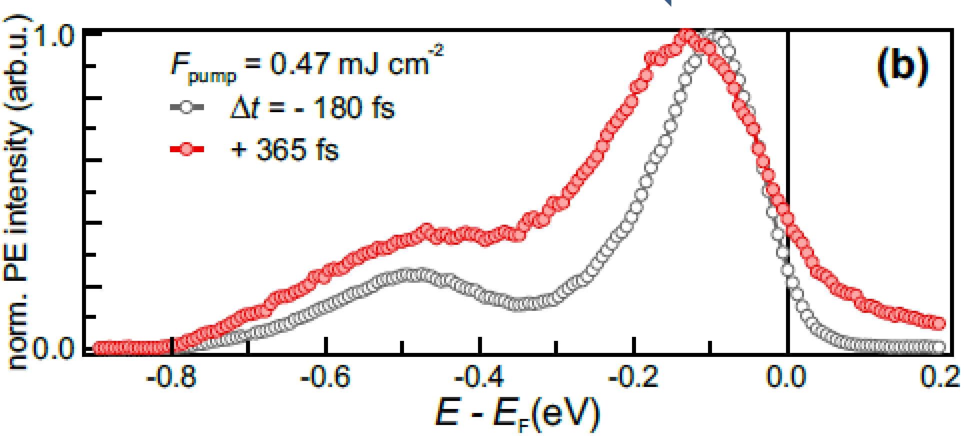
- Leads to a reduction of Hartree and Self-Energy: reduction of band gap
  (cf. eg. Oschlies et al., PRB 45, 13741 (1992))

Time-resolved ARPES: gap enhancement at Γ









<u>At low fluence $F < F_{sat}$:</u>

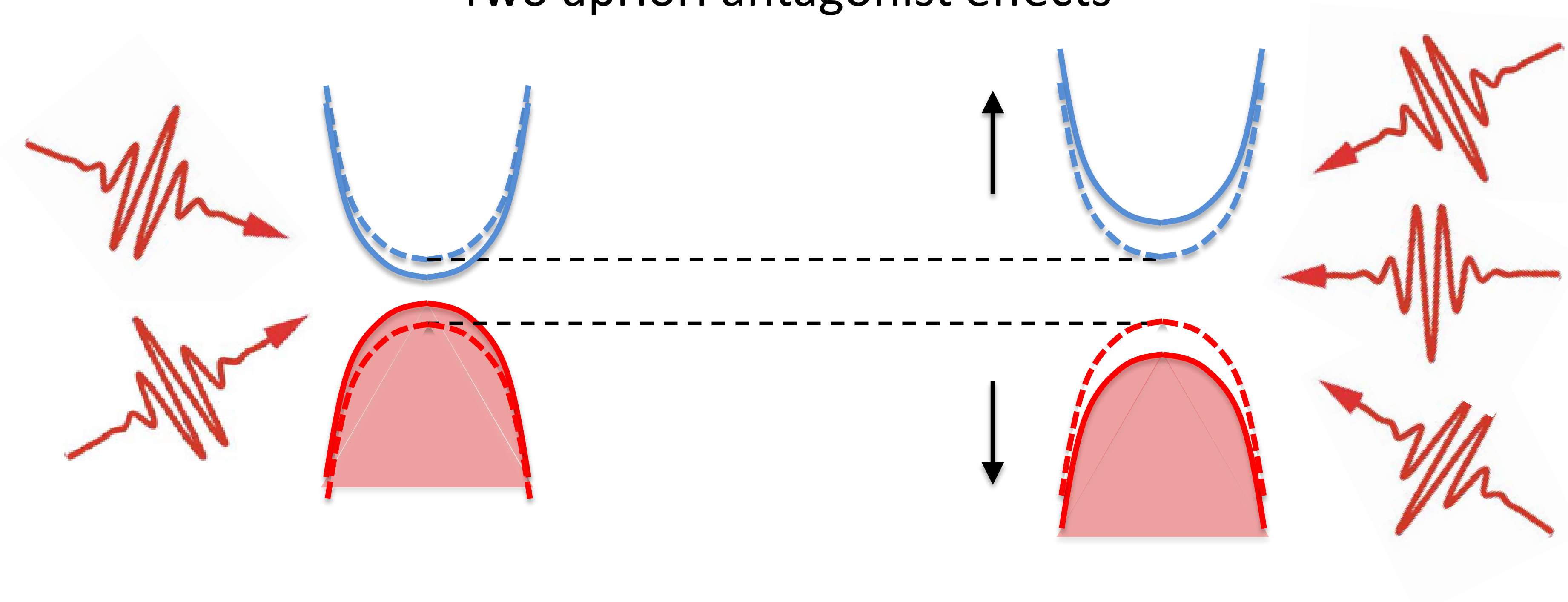
- Gap shrinks (usual for semiconductor)

<u>At large fluence $F > F_{sat}$:</u>

- Gap at Γ is enhanced: very unusual!

Two-gap behavior of Ta_2NiSe_5

Two apriori antagonist effects



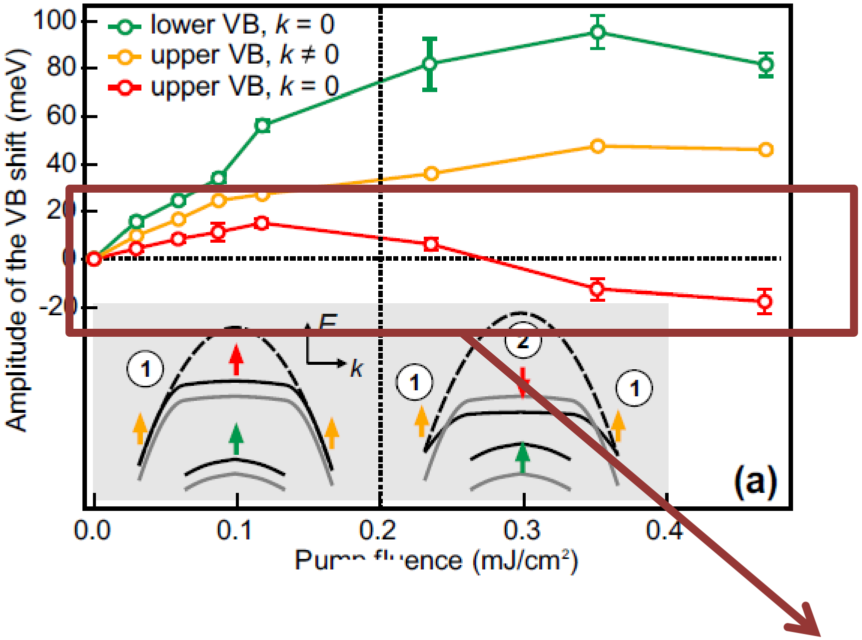
Semiconductor band gap renormalization

Correlated band gap enhancement

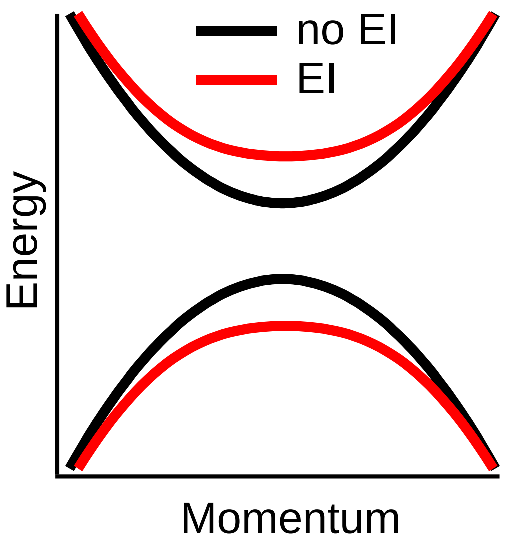
How to reconcile these two effects? ....

Non-equilibrium enhancement of the exciton condensate

Schematic picture of the transient process:



equilibrium situation



2 competing effects: BGR and … ?

Non-equilibrium enhancement of the exciton condensate

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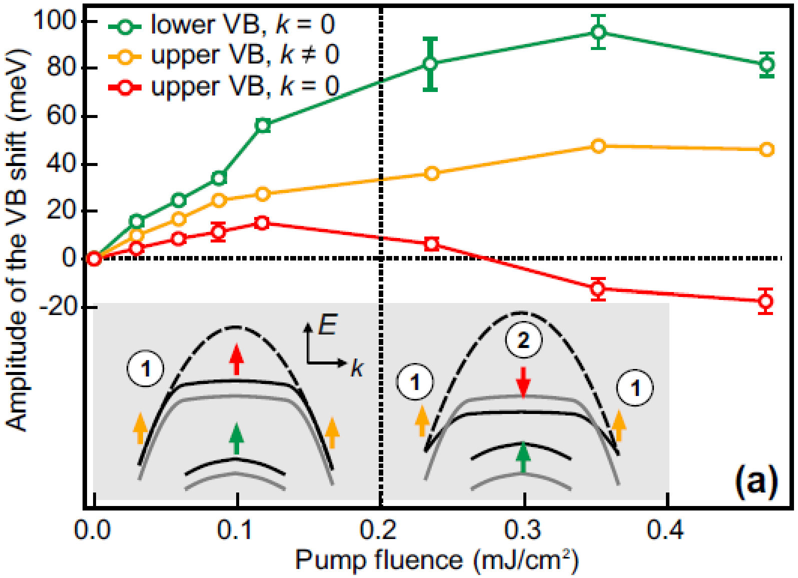
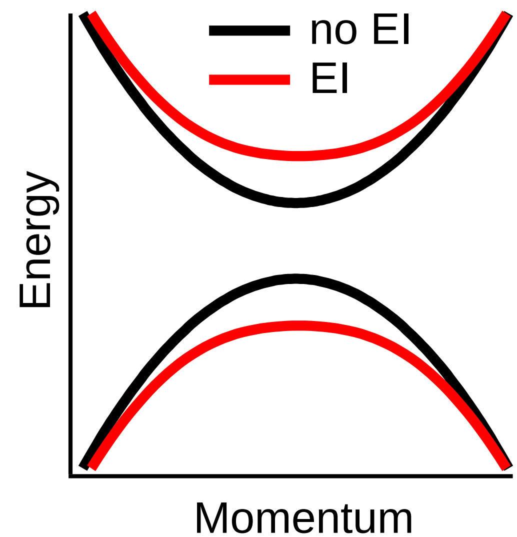
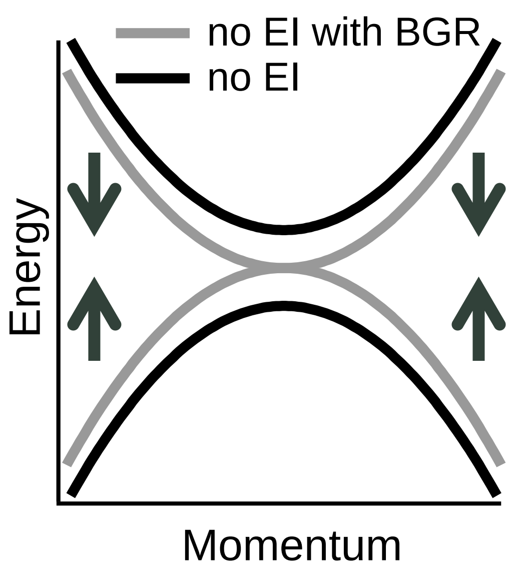


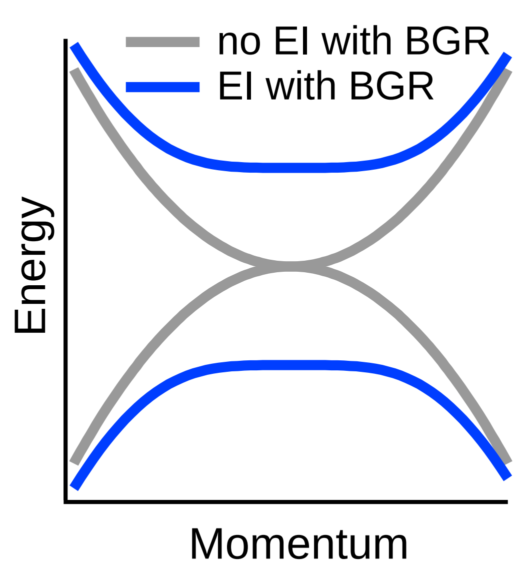
equilibrium situation



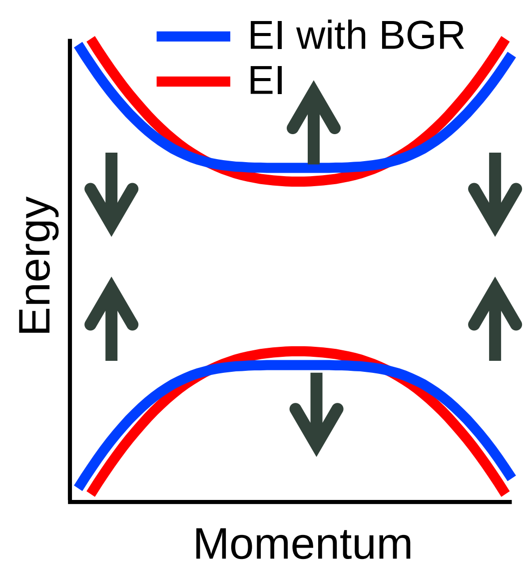
1) BGR of the bare (no EI) case:



2) Switch on EI:



3) Resulting shifts:



Non-equilibrium enhancement of the exciton condensate

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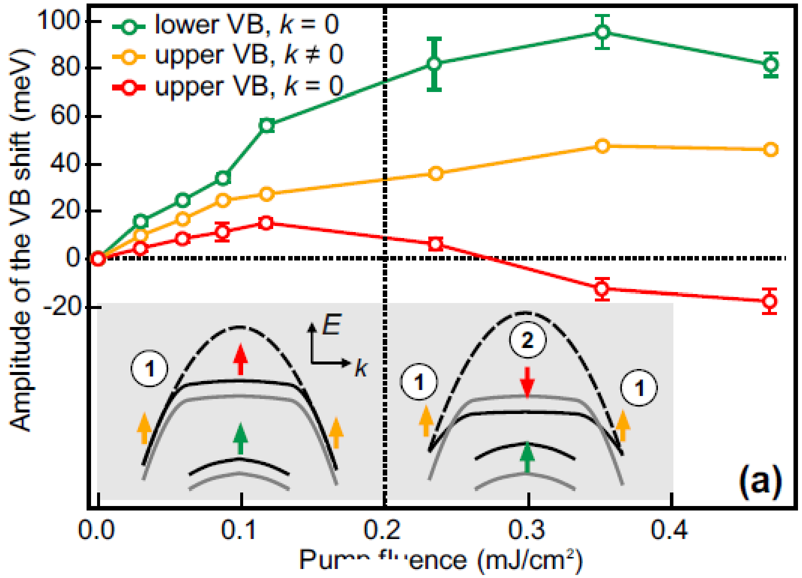

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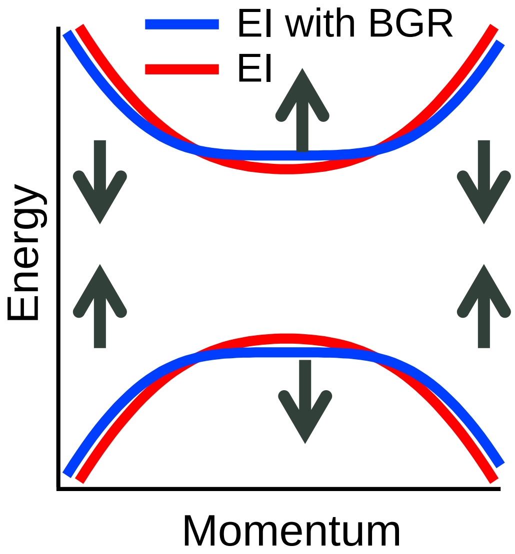


Competing processes for $F > F_{sat}$ (1):

a) The **overall semiconductor band gap is decreased** due to the enhanced screening: **band gap renormalization**

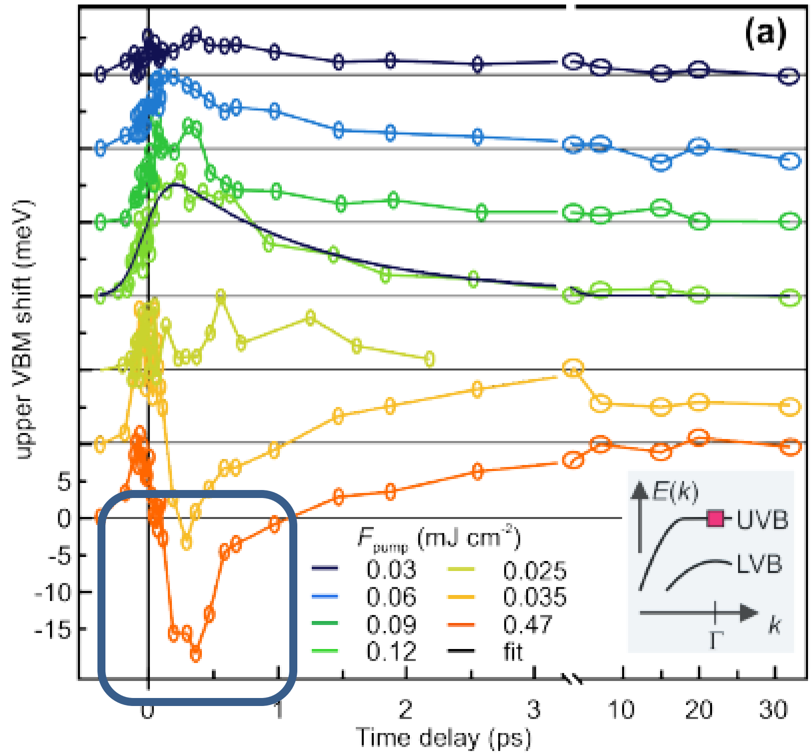
b) The **excitonic insulator band gap at Γ is increased** due to:
- Process a) brings valence band and conduction band closer to each other: EI renormalization is stronger for a **frozen exciton condensate**
- This results in a net band gap increase at Γ

Conclusions

- **Saturation** of the photoinduced depletion in the topmost valence band reveals a **critical fluence F_{sat}**
- At low fluences, $F < F_{sat}$, we observe a global **band gap renormalisation** (shrinking of semiconductor gap)
- At higher fluences, $F > F_{sat}$, the **excitonic insulator gap at Γ is enhanced upon pumping**:

Out-of-equilibrium state:

excitonic condensate **order is transiently enhanced** during a few 100s fs



Mor et al., PRL 119, 086401 (2017)

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M. Eckstein

ARPES at University of Waseda (Tokyo, Japan):
T. Mizokawa

Ta_2NiSe_5 samples (University of Tokyo, Japan):
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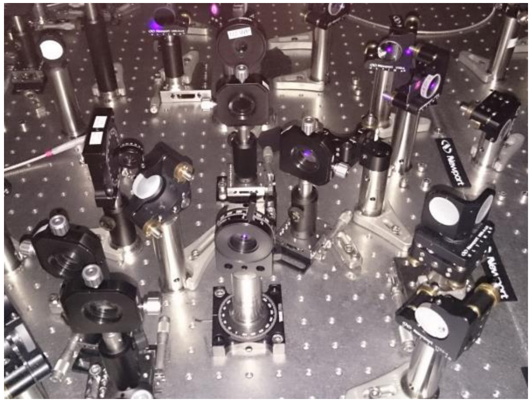

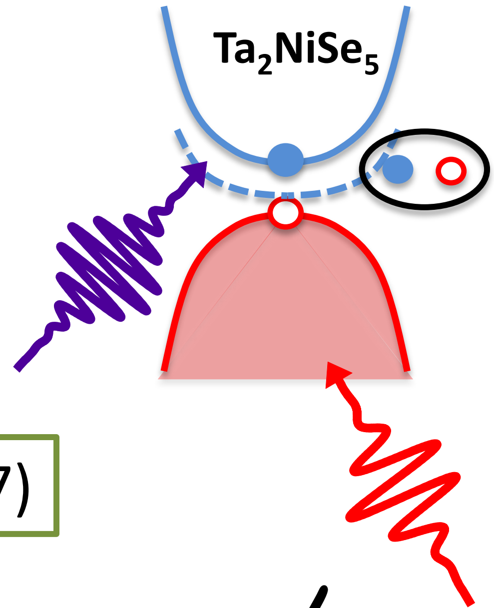

Mor et al., PRL 119, 086401 (2017)

Thank you for your attention!