

Friday July 12

Theoretical perspectives on an excitonic insulator Ta_2NiSe_5

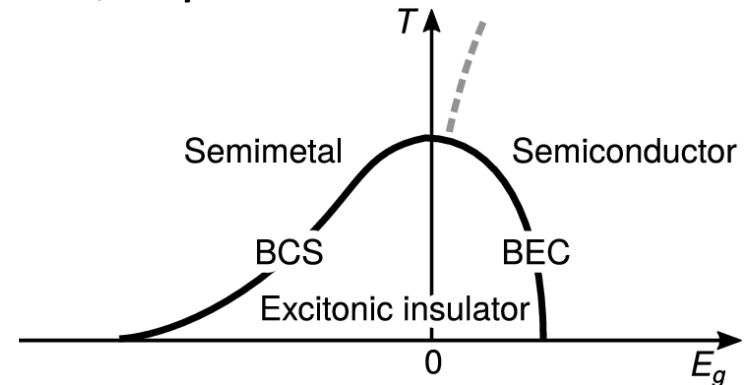
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K. Sugimoto *et al.*,
PRL **120**, 247602 (2018)



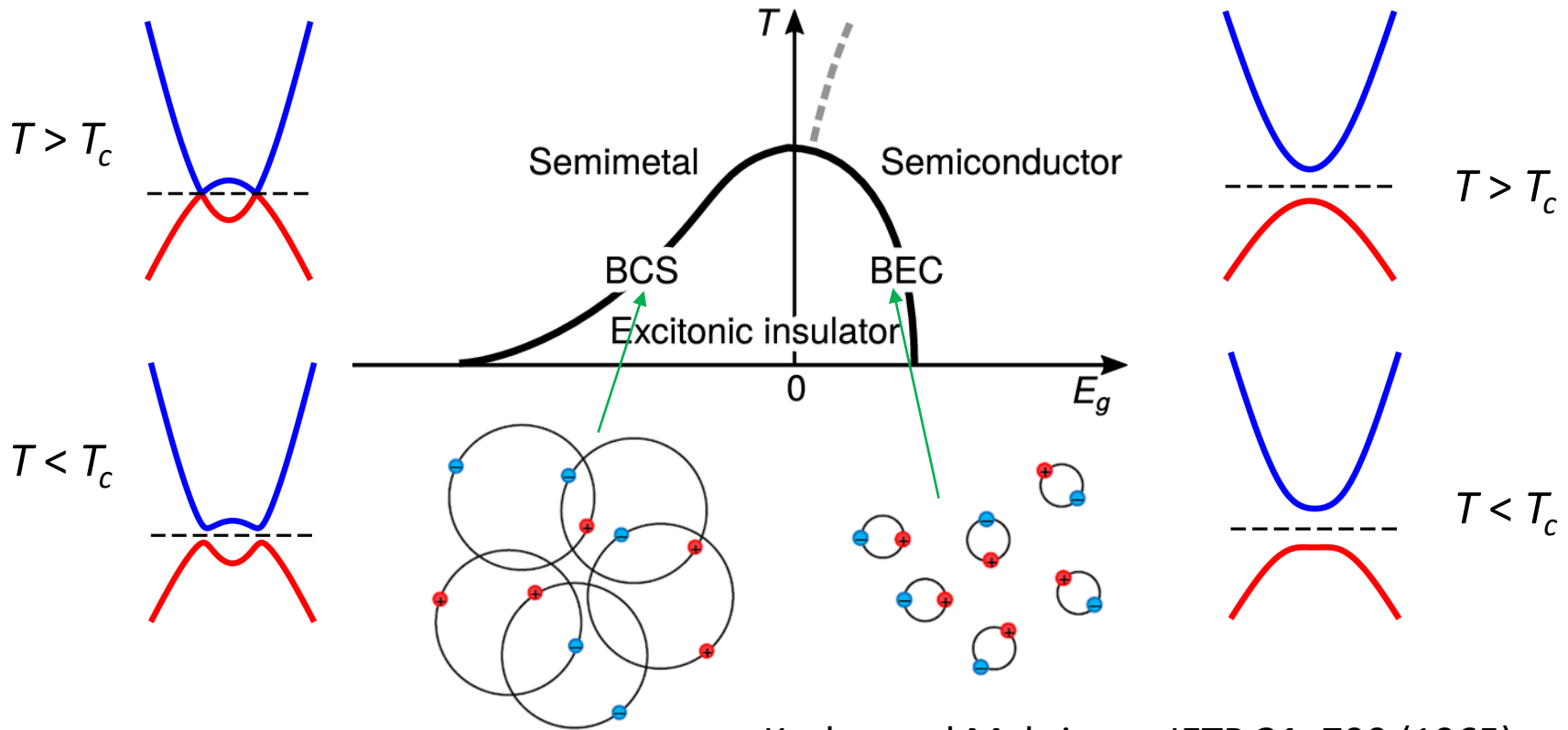
Collaborators

K. Sugimoto (Keio Univ., Tokyo)
S. Nishimoto (TU & IFW, Dresden)
T. Kaneko (Columbia Univ., NY)

Experimental information

Sawa, Takagi, Mizokawa, Okazaki, ...

Conventional phase diagram



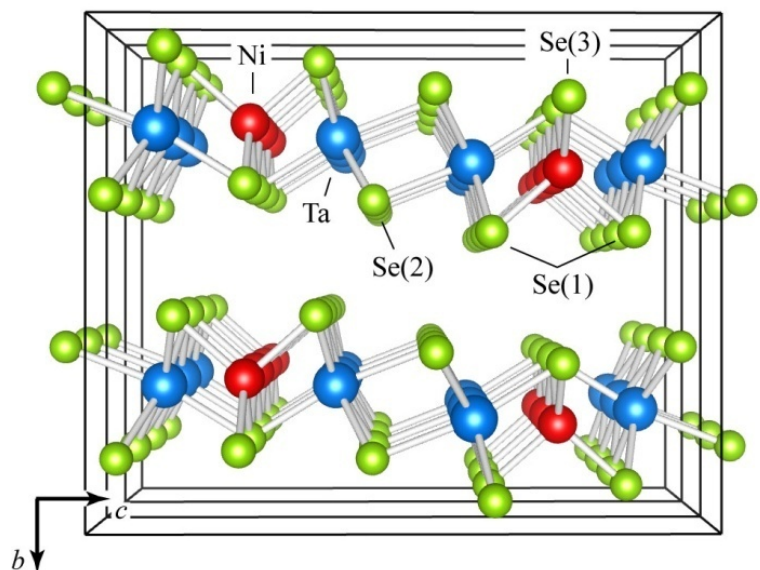
Kozlov and Maksimov, JETP **21**, 790 (1965)

Ta_2NiSe_5 should be in the BEC regime
because it is semiconducting in the entire temperature range observed.

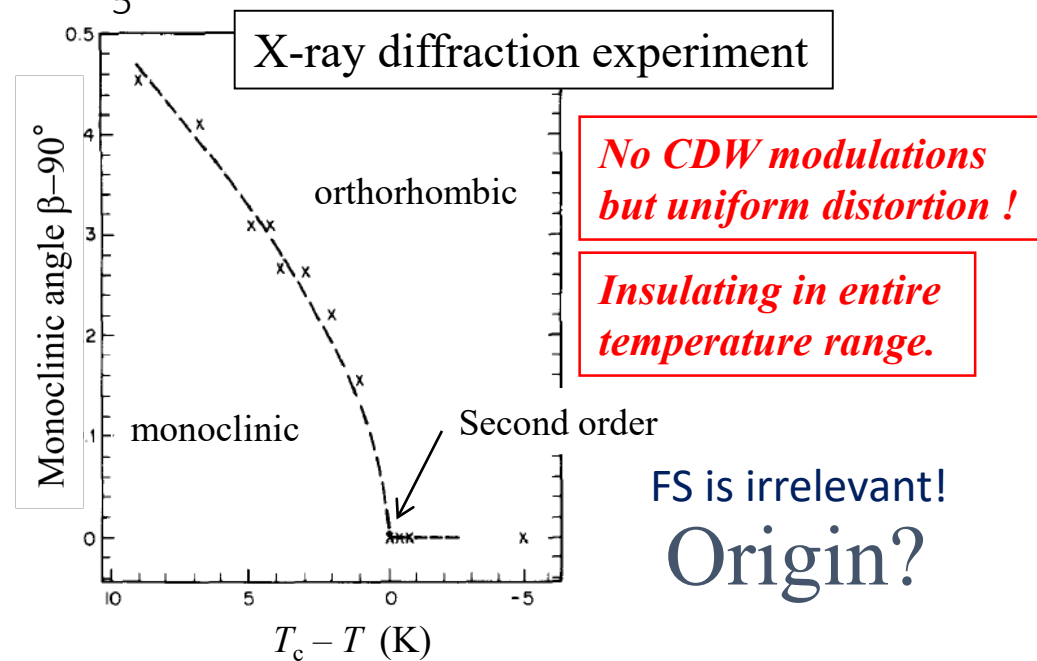
This conventional picture is in fact seriously violated in Ta_2NiSe_5 .

Sugimoto, Nishimoto, Kaneko, and Y.O., PRL **120**, 247602 (2018).

Structural phase transition of Ta_2NiSe_5

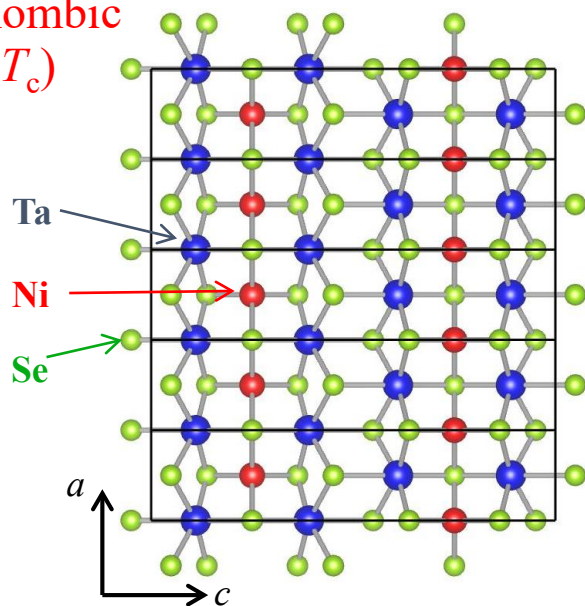


Sunshine and Ibers, Inorg. Chem. **24**, 3611 (1985).



Di Salvo *et al.*, J. Less-Common Met. **116**, 51 (1986).

orthorhombic
($T > T_c$)

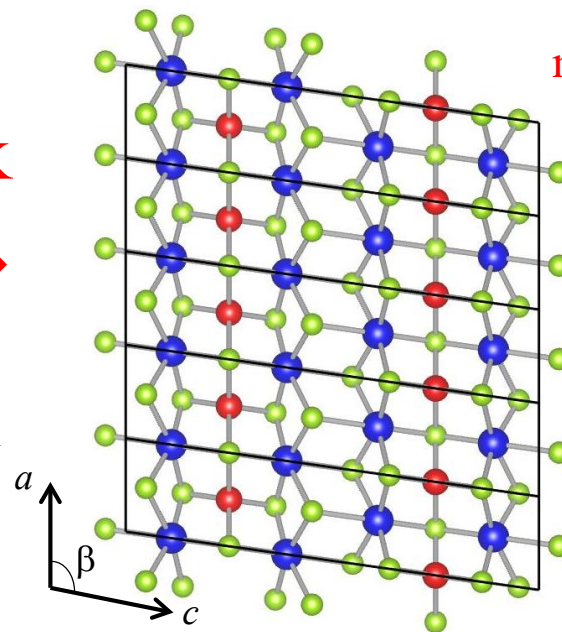


$T_c = 328$ K



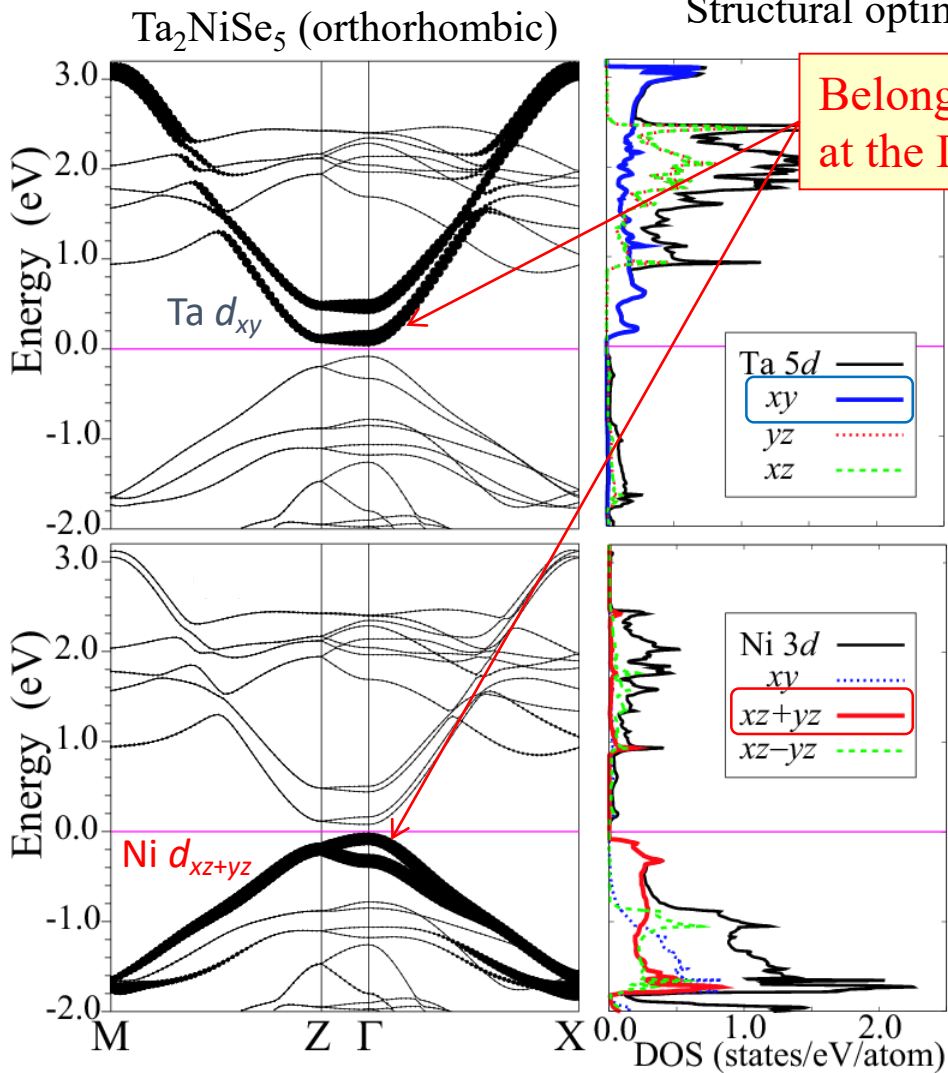
$\beta = 90.59^\circ$
at $T = 150$ K.

monoclinic
($T < T_c$)

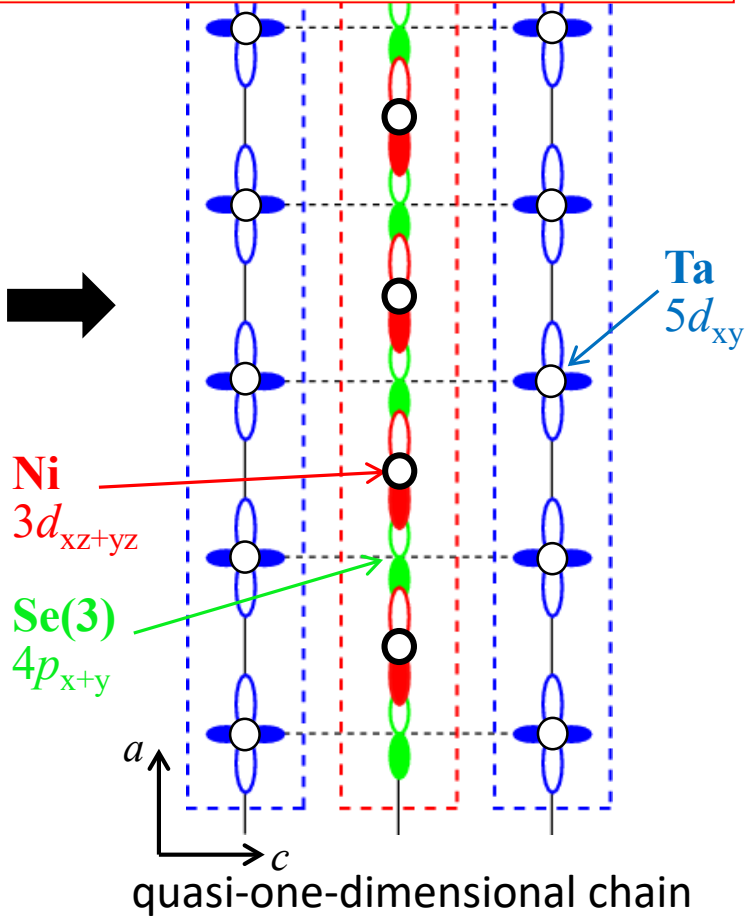


WIEN2K (FLAPW, GGA)

Structural optimization for the orthorhombic structure



Belong to different irreducible representations at the Γ point of BZ. No hybridization occurs!

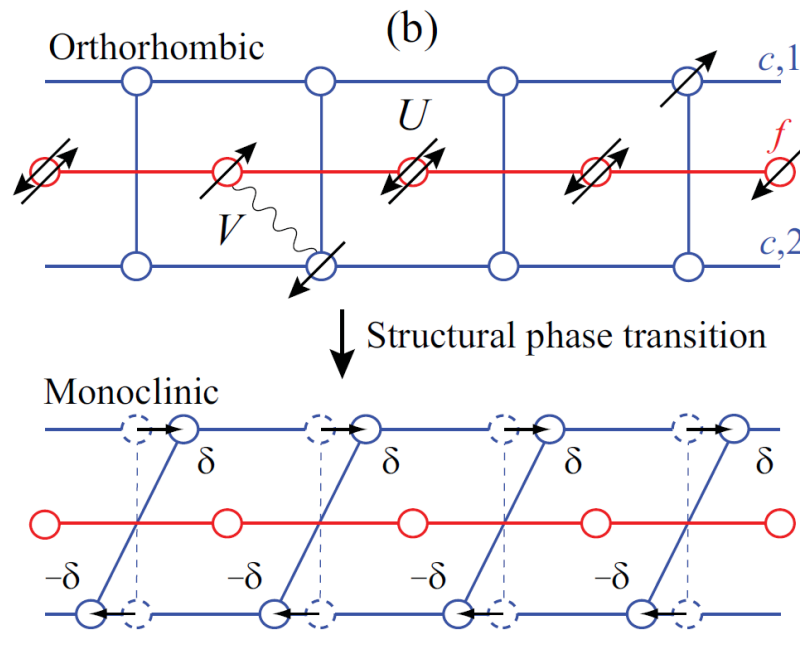
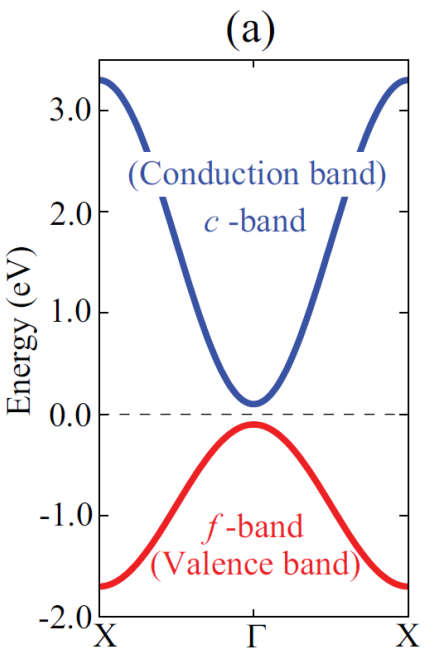


A chain made of the $3d_{xz+yz}$ orbitals of Ni and $4p_{x+y}$ orbitals of Se forms the valence band. Two chains made of the $5d_{xy}$ orbitals of Ta form the conduction bands.

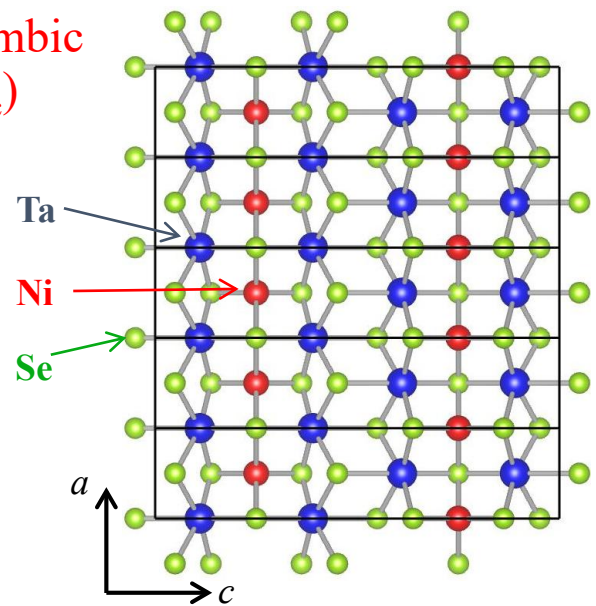
Effective model and mean-field analysis

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

Excitonic Bond Order
formation ($Q = 0$)



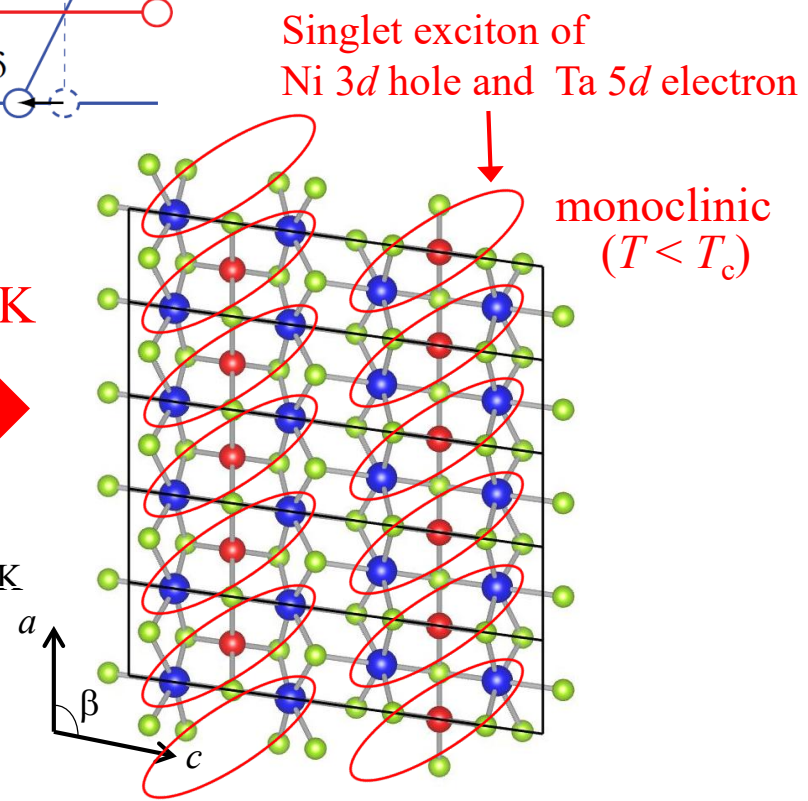
orthorhombic
($T > T_c$)



$T_c = 328$ K

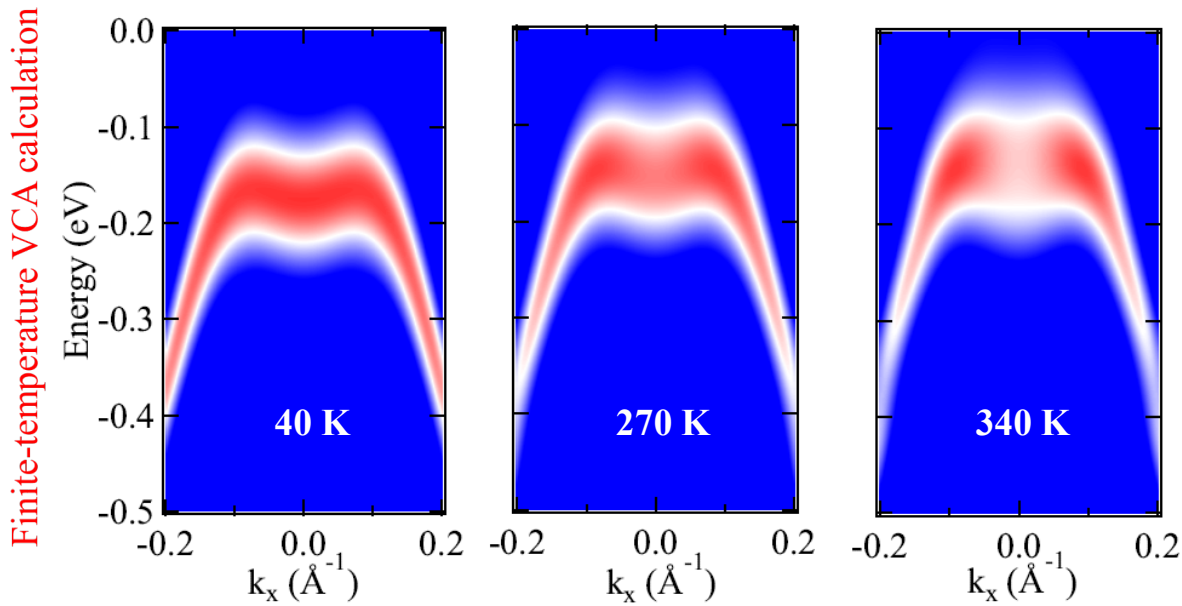
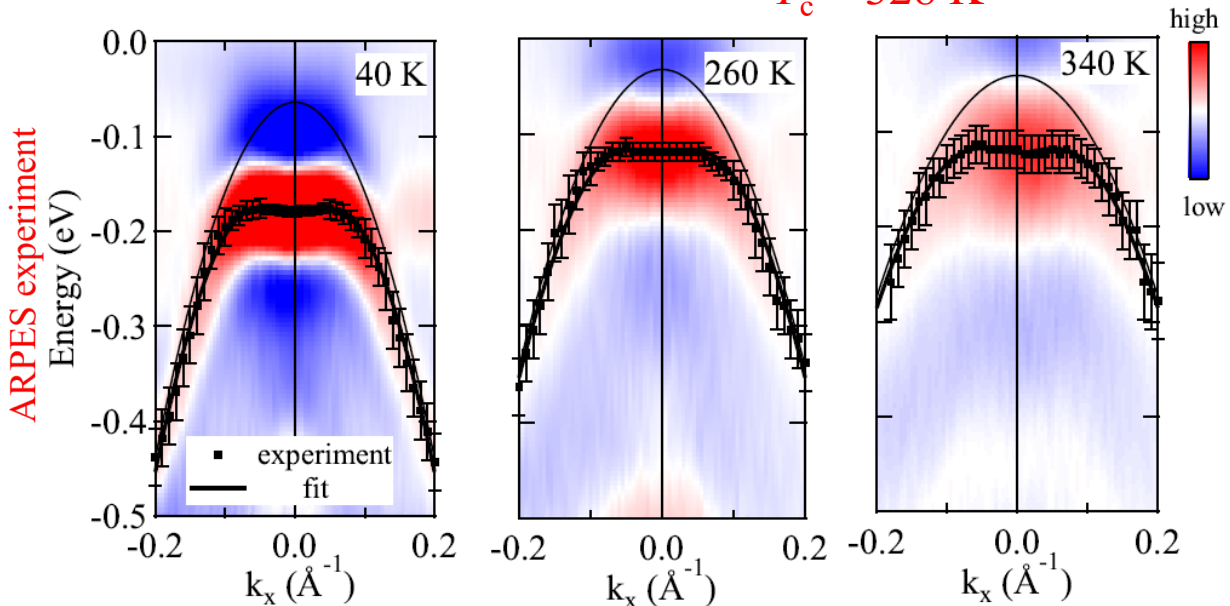


$\beta = 90.59^\circ$
at $T = 150$ K

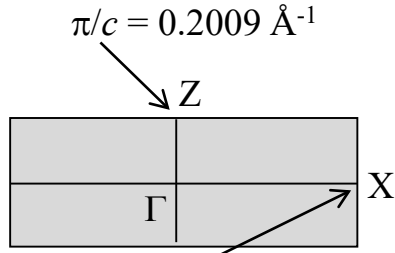
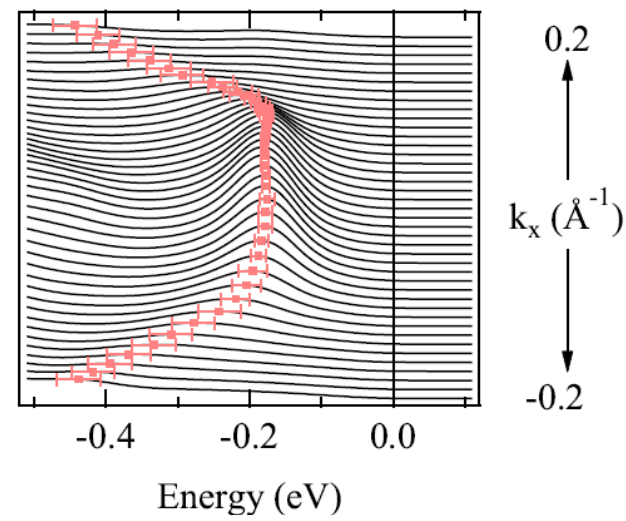


ARPES: comparison with experiment

Seki *et al.*, PRB **90**, 155116 (2014). $T_c = 328$ K



Along Γ -X direction, $T = 40$ K



Γ X: chain direction

Spontaneous hybridization

Flat band with **double-peak** structure!

Also see: Wakisaka *et al.*, PRL **103**, 026402 (2009)

Other quantities of Ta₂NiSe₅

1. Specific heat

Sugimoto *et al.*, PRB **93**, 041105(R) (2016).

2. Elastic shear constant

Sugimoto *et al.*, PRB **93**, 041105(R) (2016).

3. Quantum interference (NMR and ultrasonic attenuation)

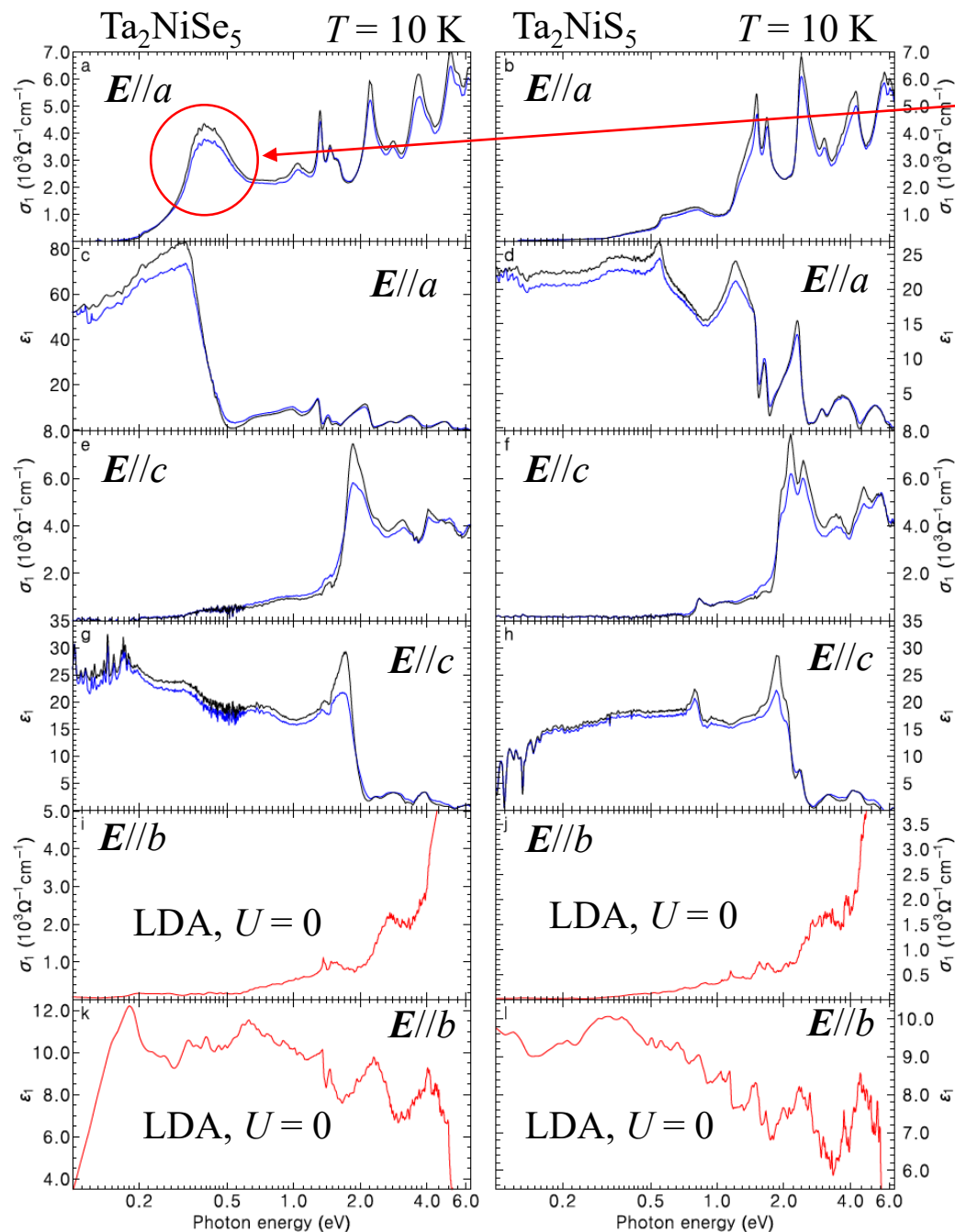
Sugimoto *et al.*, PRB **93**, 041105(R) (2016).

4. Diamagnetic susceptibility

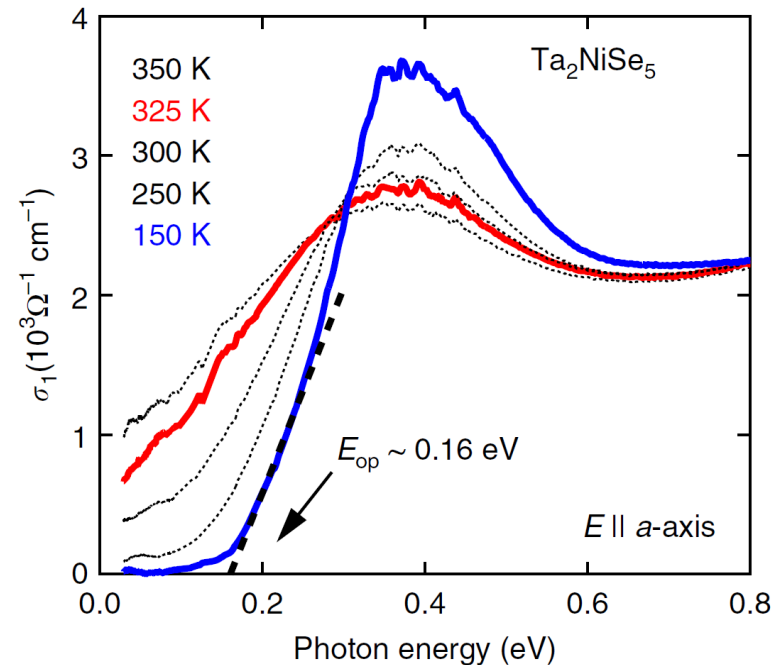
Sugimoto and Y.O., PRB **94**, 085111 (2016)

So far, so good!

Comparative study of optical conductivity: Ta_2NiSe_5 vs. Ta_2NiS_5



Anomalous peak!

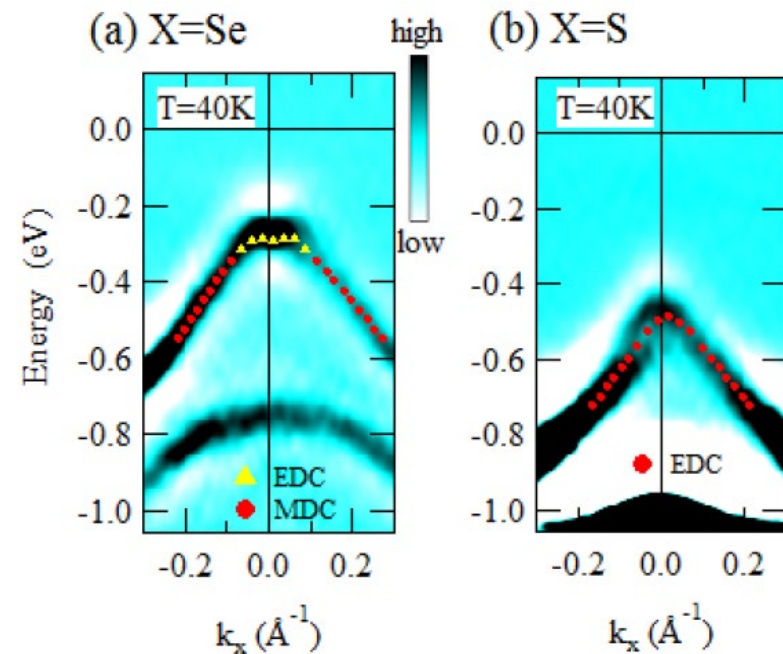
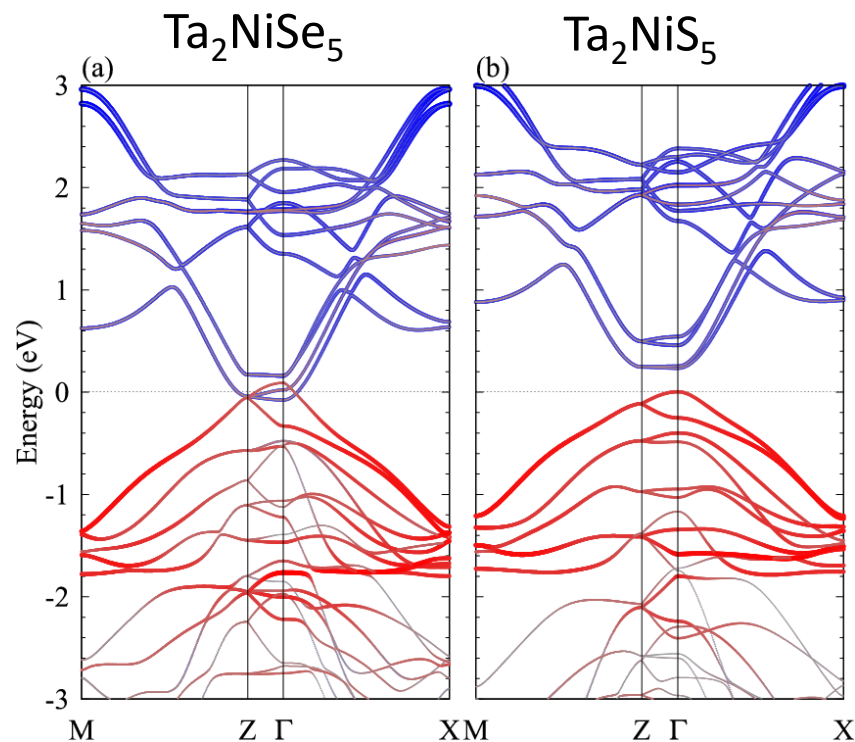


Y. F. Lu *et al.*,
Nat. Commun. **8**, 14408 (2017)

The peak appears
both below and above T_c

Larkin, PhD Thesis, Univ. Stuttgart (2016)
Larkin *et al.*, PRB **95**, 195144 (2017)

Comparison between Ta_2NiSe_5 and Ta_2NiS_5



Mizokawa *et al.*

Ta_2NiS_5 ... No phase transition is observed, even under high pressures.

We use the modified Becke-Johnson (mBJ) exchange potential, assuming $c = 1.5$.

Tran and Blaha, PRL **102**, 226401 (2009)

Ta_2NiS_5 ... a simple semiconductor

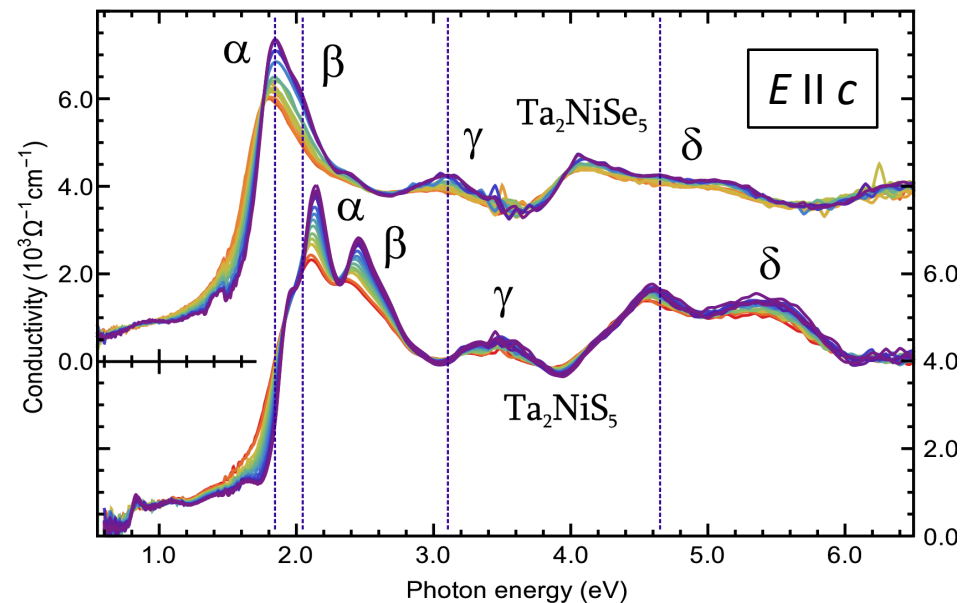
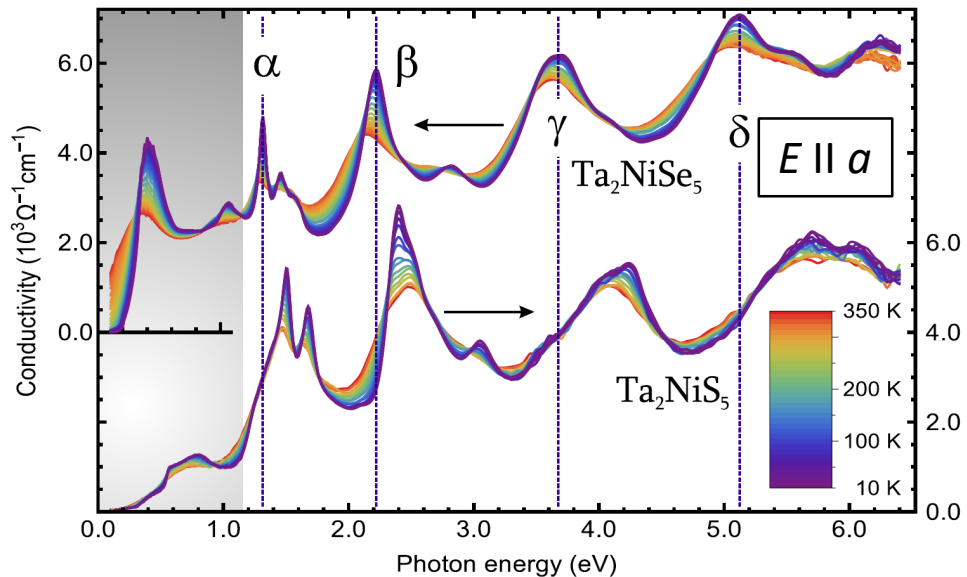
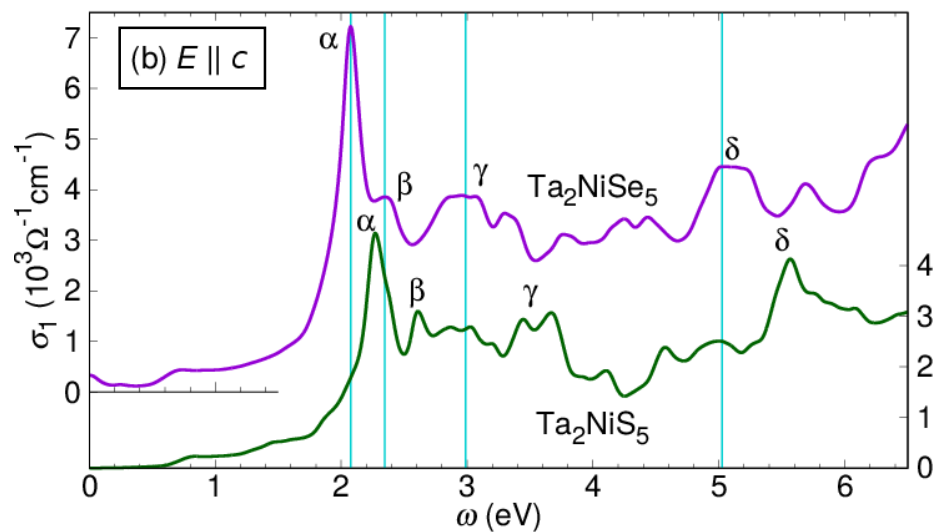
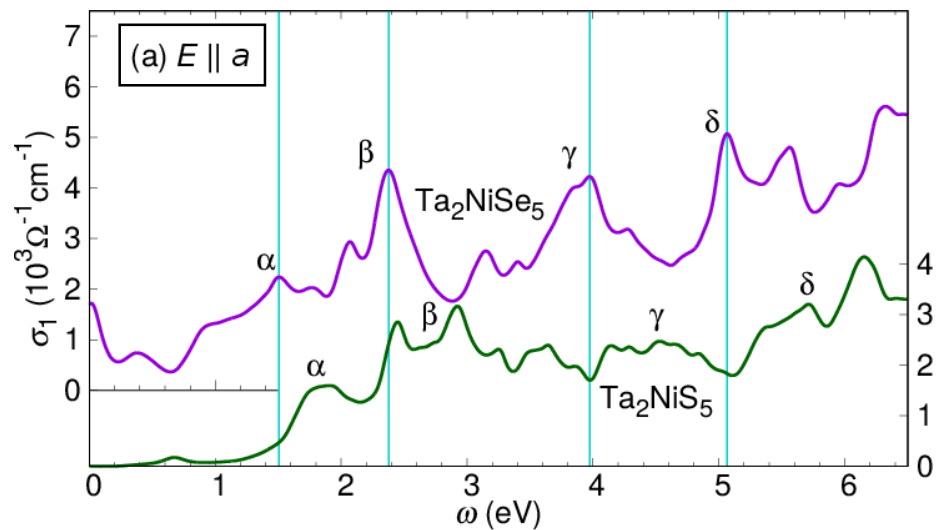
Ta_2NiSe_5 ... a semimetal?!

Strange insulator

Optical conductivity in Ta₂NiSe₅ and Ta₂NiS₅

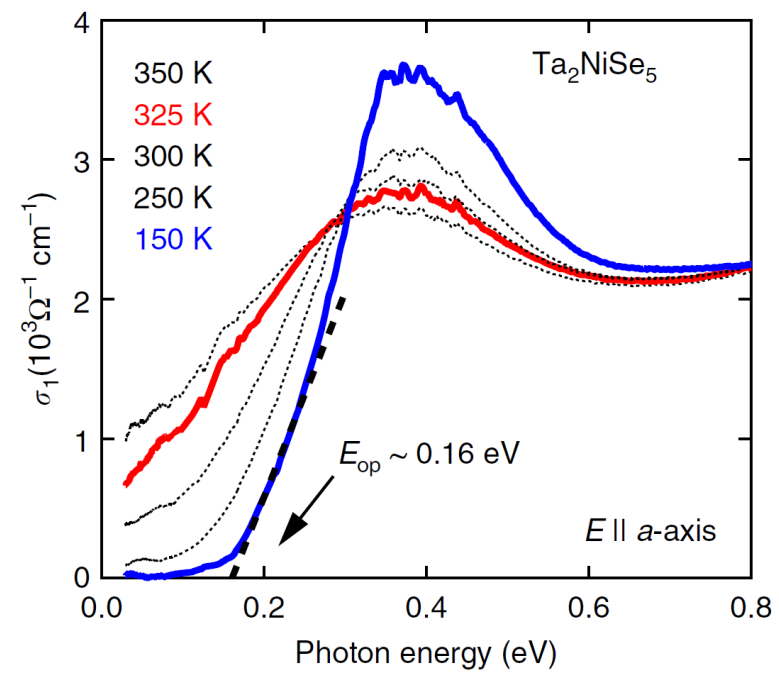
Experiment

Theory by mBJ with $c = 1.5$



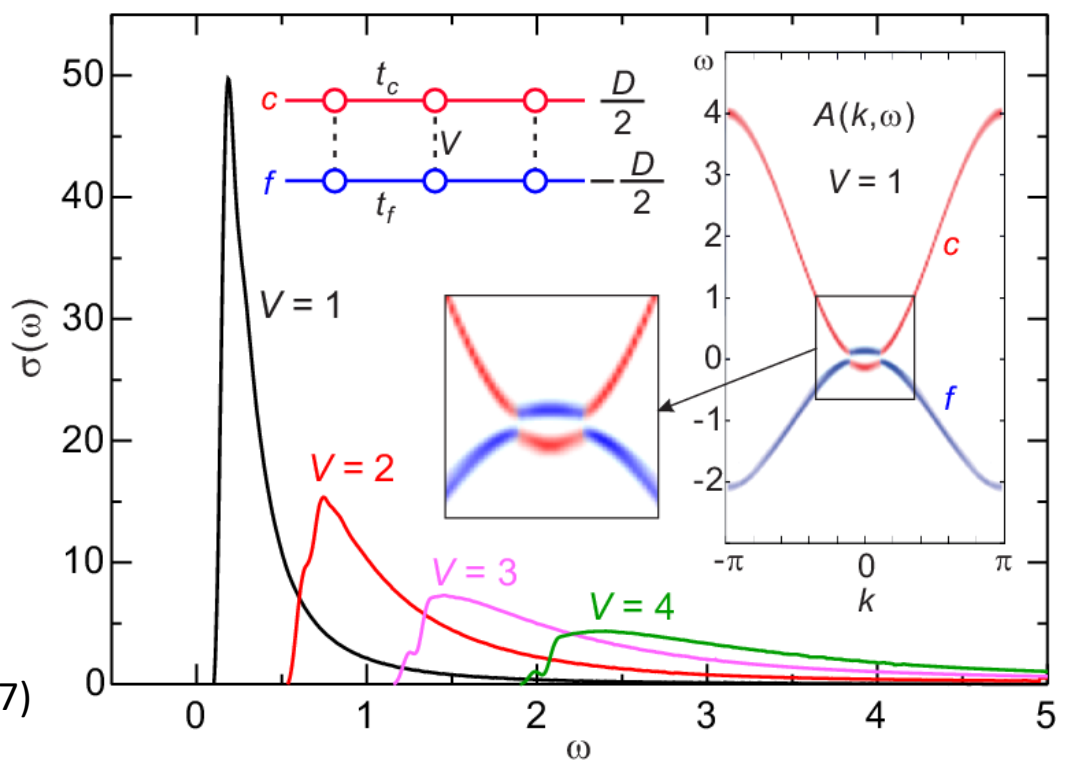
Experimental data from
Larkin *et al.*, PRB **95**, 195144 (2017); Larkin, PhD Thesis, Univ. Stuttgart (2016)

Origin of the anomalous peak?

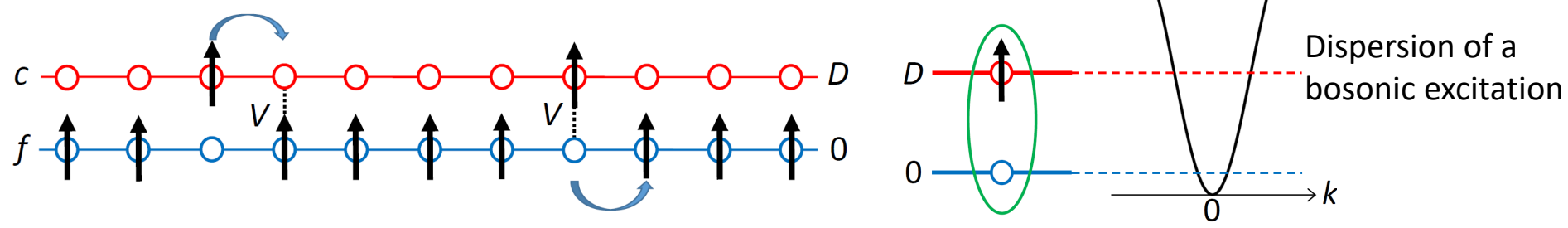


Y. F. Lu *et al.*, Nat. Commun. **8**, 14408 (2017)

Dynamical DMRG by S. Nishimoto

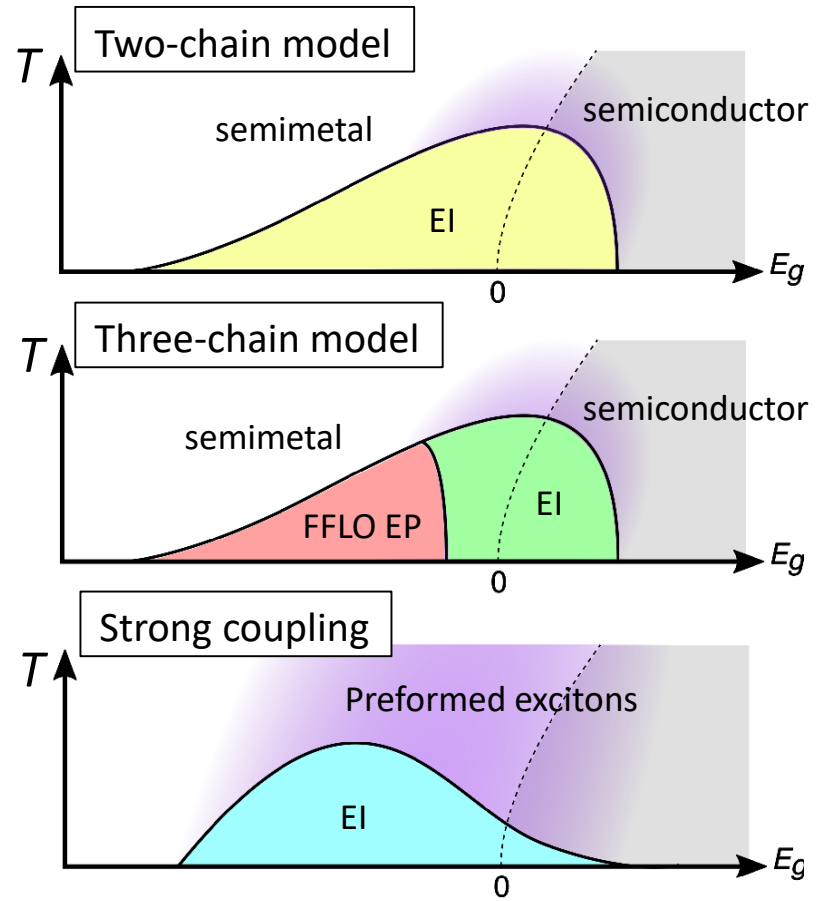
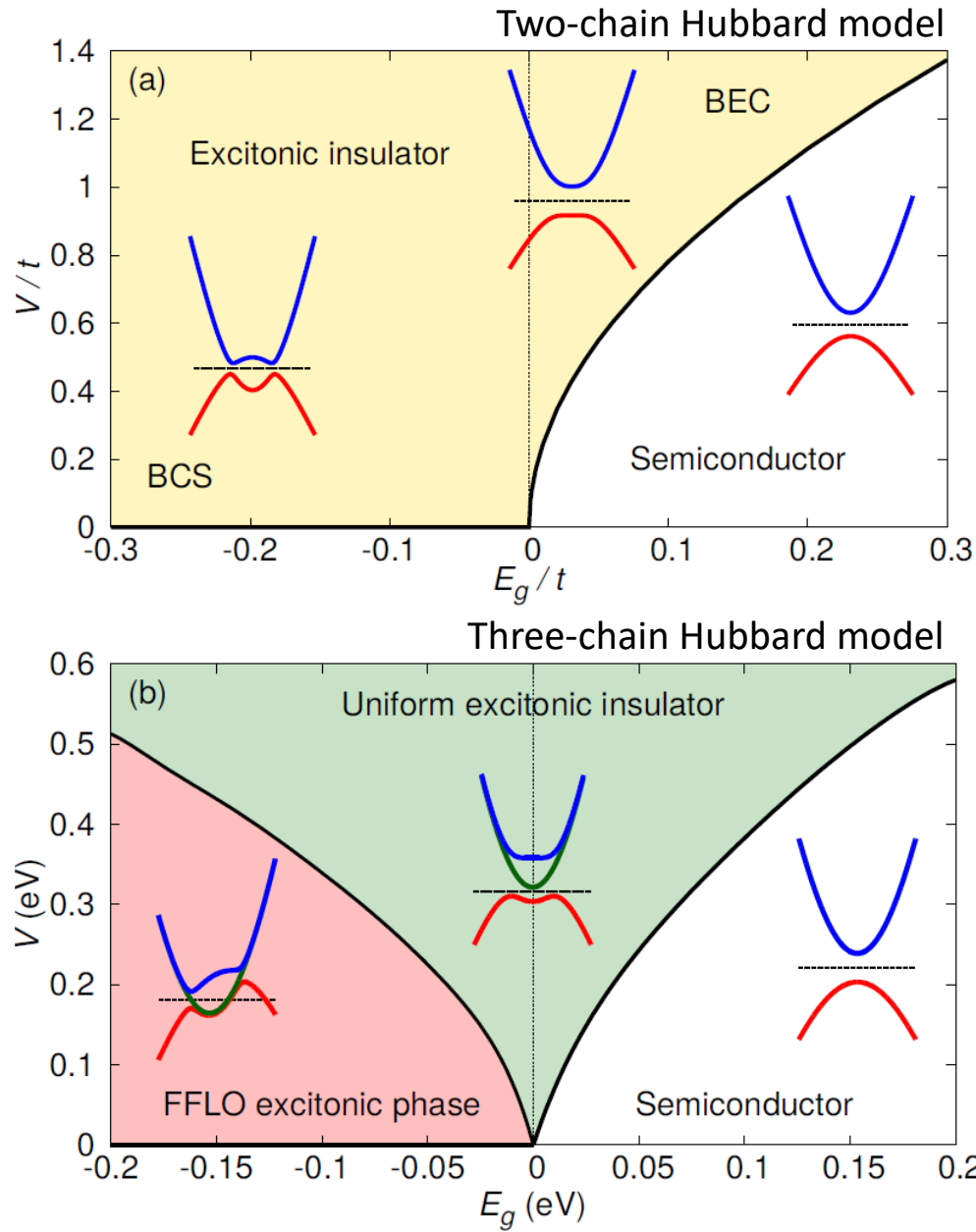


Assume a *semimetallic* state in a spinless two-chain model



A peak appears in $\sigma(\omega)$ at $\omega \approx V$ and band gap of the size V opens, irrespective of whether the long-range BEC occurs or not.

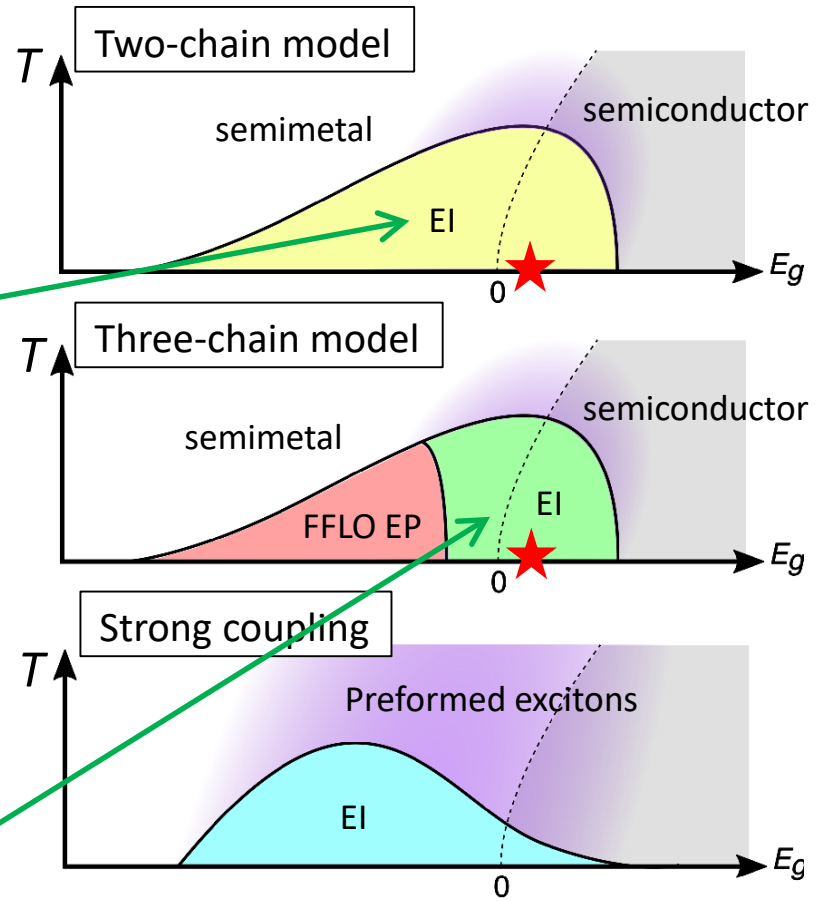
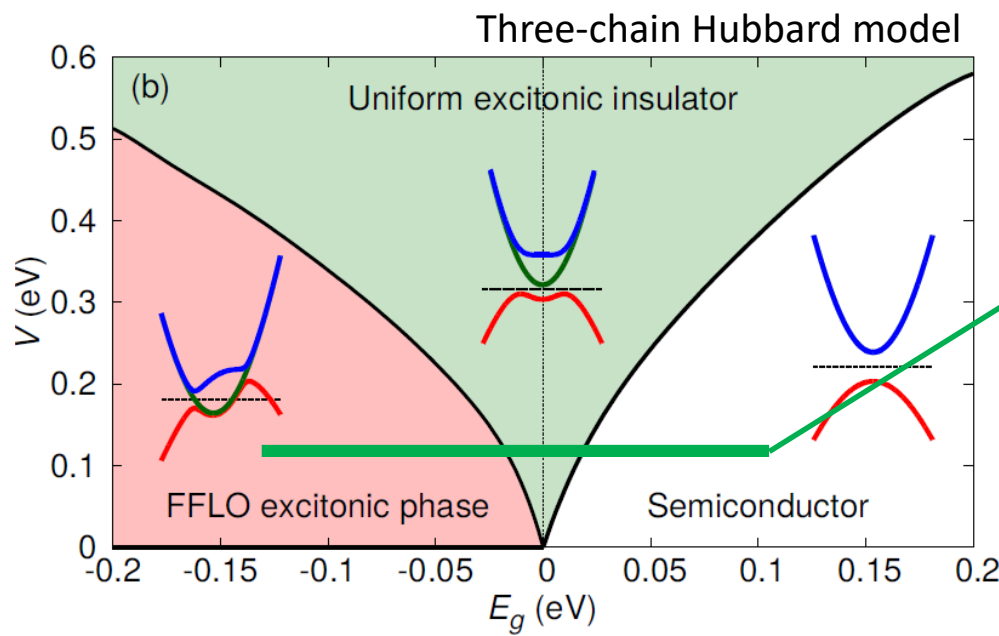
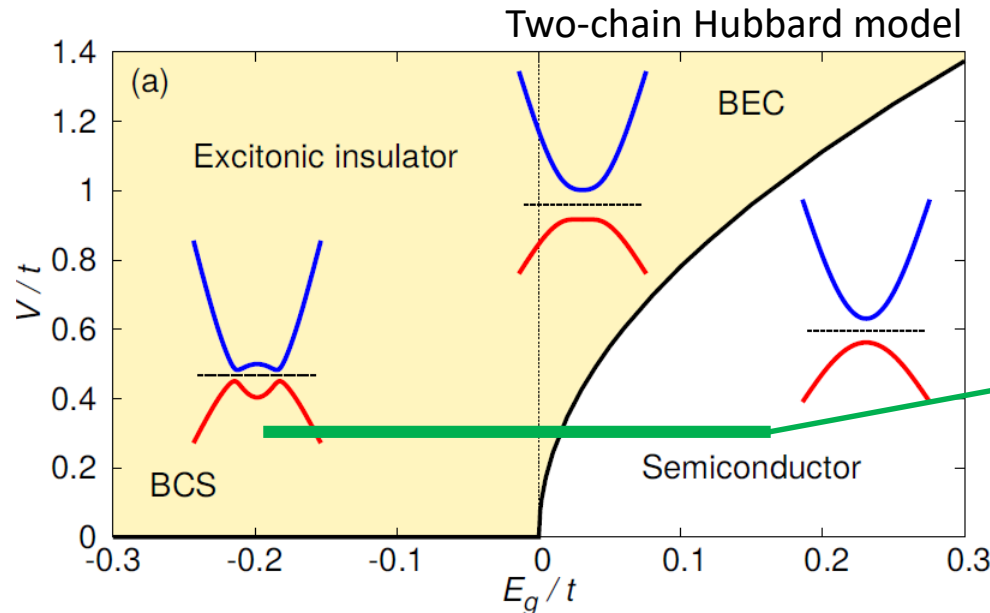
Phase diagrams



cf. FFLO EP:
Yamada *et al.*, JPSJ **85**, 053703 (2016).

Without FS nesting in 2D, the PM state appears when $E_g < 0$ and V is small.

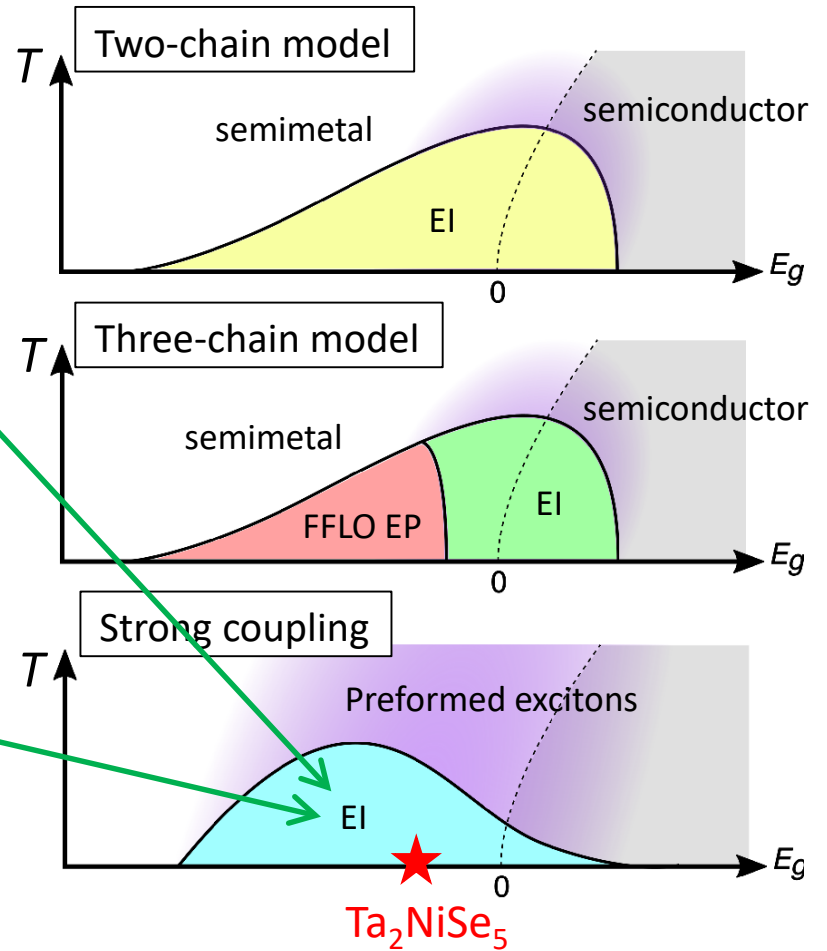
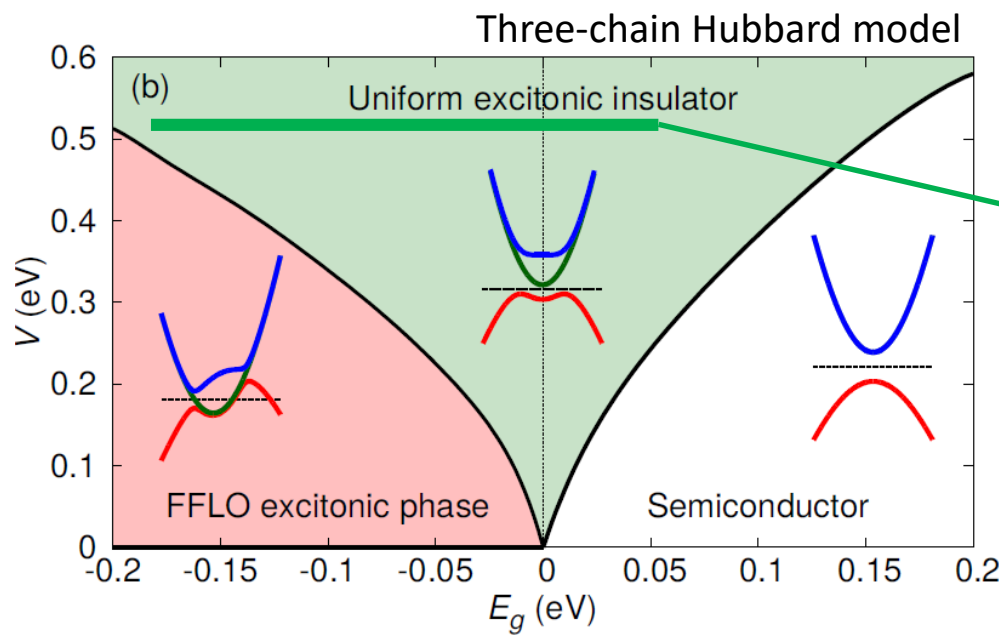
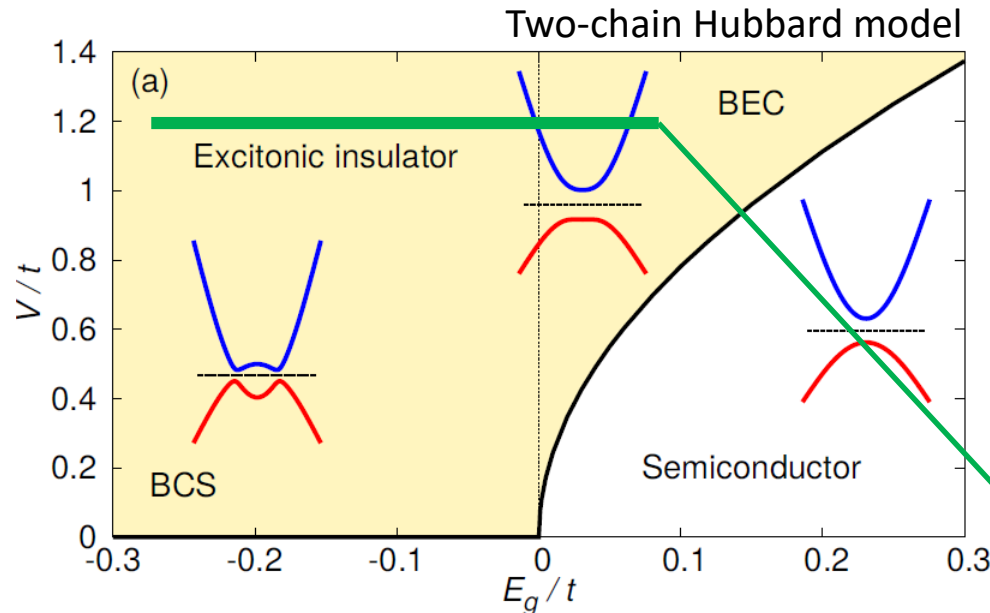
Phase diagrams



cf. FFLO EP:
Yamada *et al.*, JPSJ **85**, 053703 (2016).

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Phase diagrams



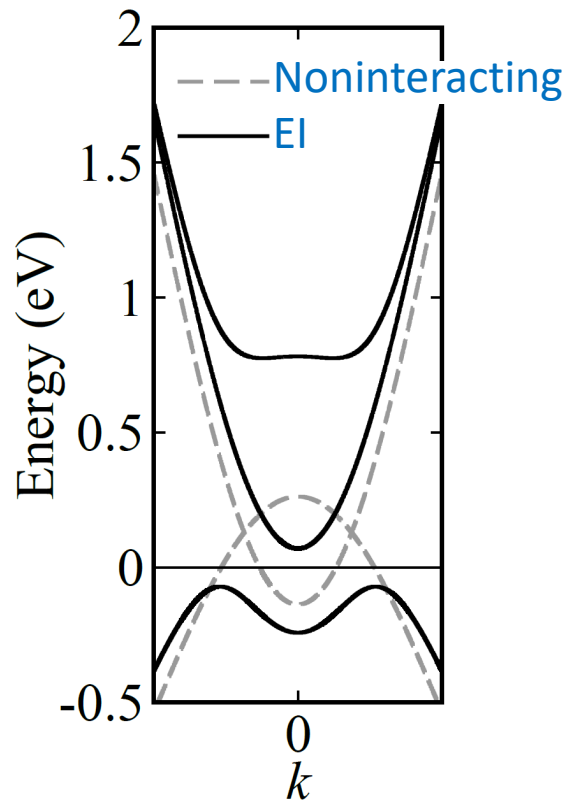
$T > T_c$
A state of preformed excitons with band gap and peak in $\sigma(\omega)$

Without FS nesting in 2D, the PM state appears when $E_g < 0$ and V is small.

A novel insulator state caused by the strong electron-hole attraction



Three-chain Hubbard model



Theoretical predictions:

1. Noninteracting band is semimetallic
2. Strong electron-hole attraction
3. Real-space pairing and bond-order formation
4. A peak in $\sigma(\omega)$ appears and band gap opens
5. A state of preformed pairs above T_c

Experimental evidences:

1. Semiconducting in the entire temperature range
2. Large peak in $\sigma(\omega)$ at $\omega \approx 0.4$ eV
3. Double-peak structure in the ARPES spectrum
4. Photo-induced semimetallic state ?
5. Pressure-induced metallic/superconducting state ?

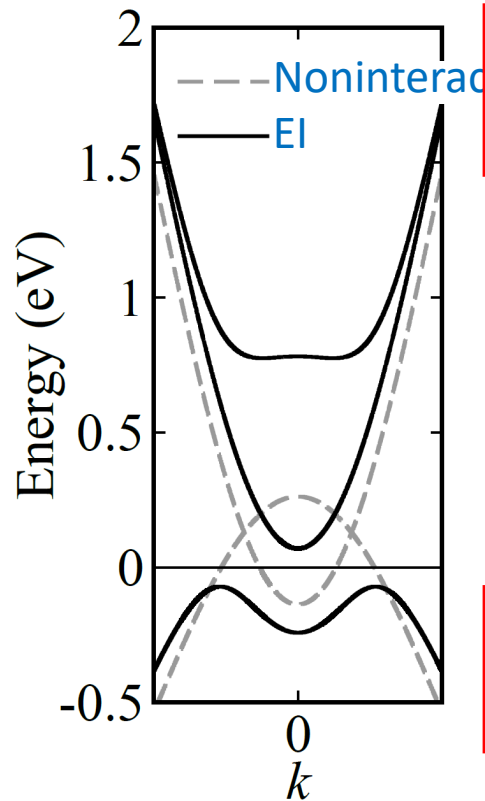
may be contrasted with

Mott insulator state caused by the strong electron-electron repulsion

A novel insulator state caused by the strong electron-hole attraction



Three-chain Hubbard model



Systems of **even** numbers of electrons per site.
 $T > T_c \dots$ insulating without excitonic condensation.

1. Near-space pairing and bond-order formation
4. A peak in $\sigma(\omega)$ appears and band gap opens
5. A state of preformed pairs above T_c

Both of these insulators become metallic if the electron correlations are weakened.

Experimental evidences.

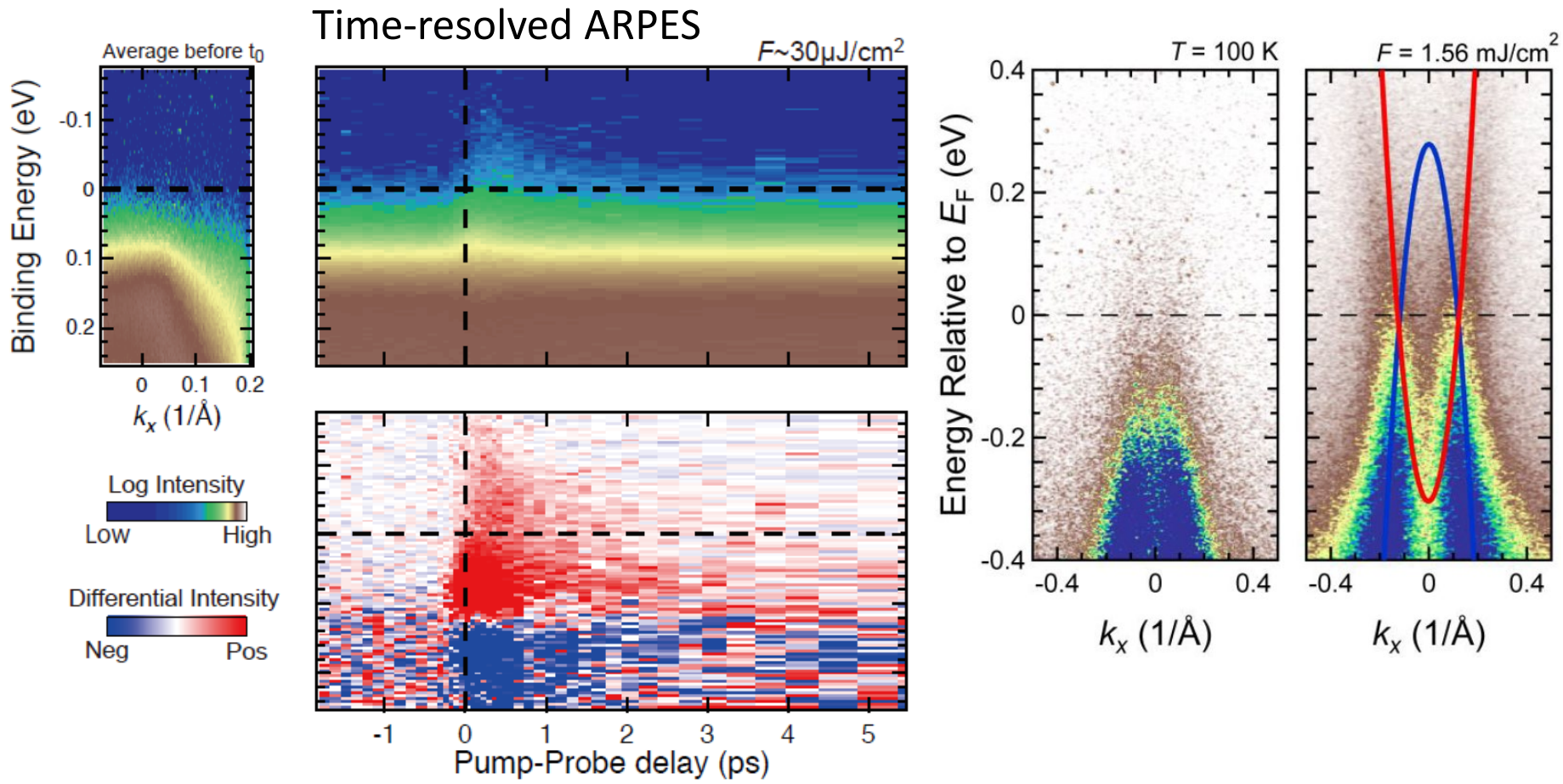
1. Semiconducting in the entire temperature range
2. Large peak in $\sigma(\omega)$ at $\omega \approx 0.4$ eV

Systems of **odd** numbers of electrons per site.
 $T > T_c \dots$ insulating without magnetic order.

may be contrasted with

Mott insulator state caused by the strong electron-electron repulsion

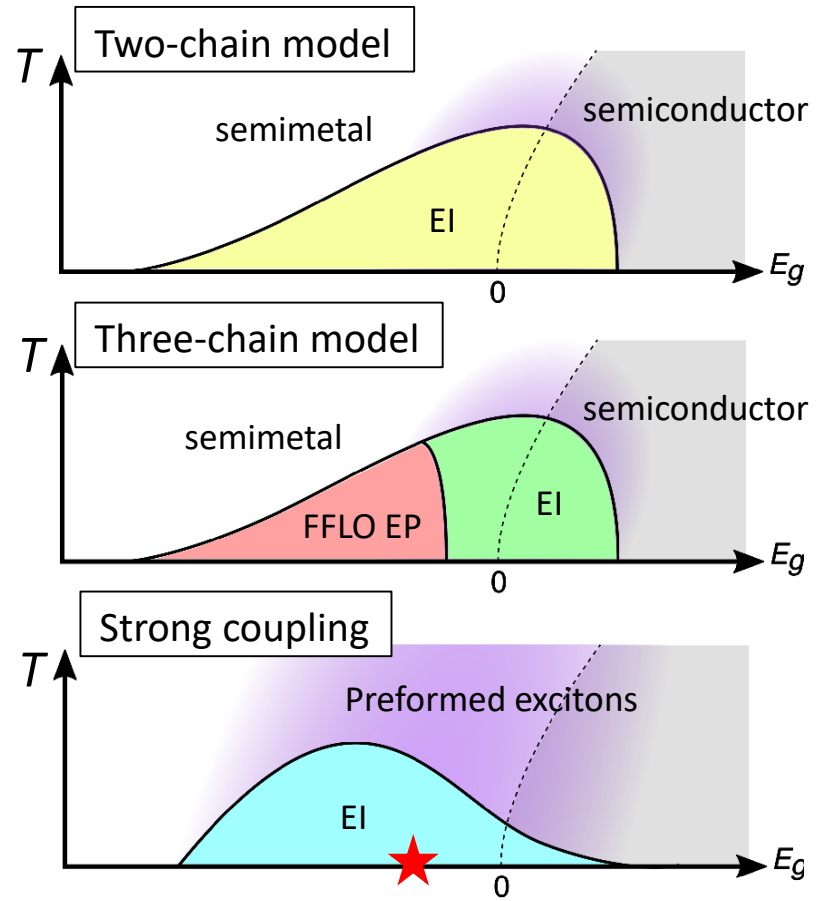
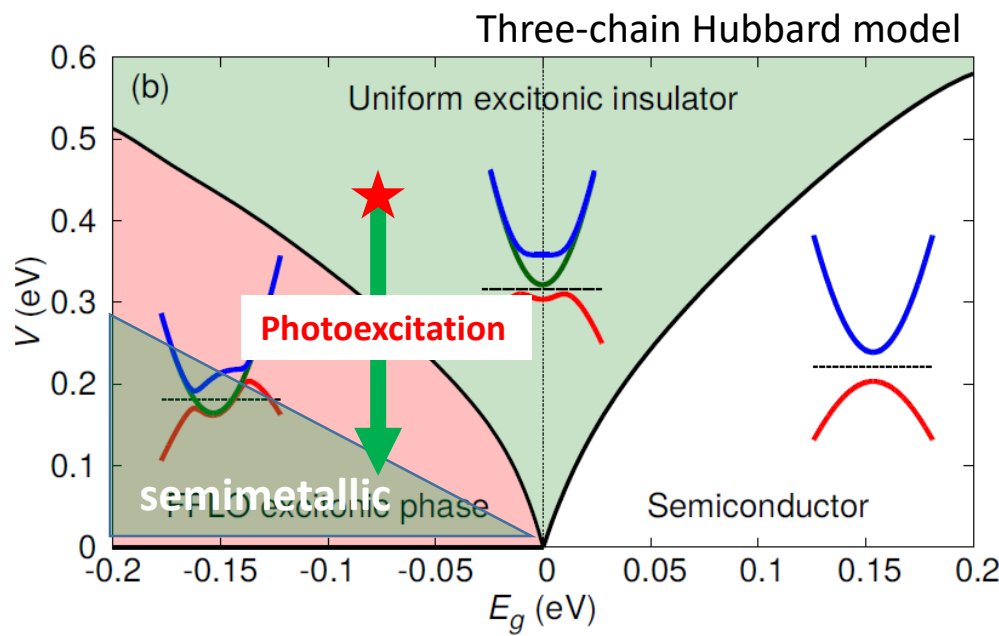
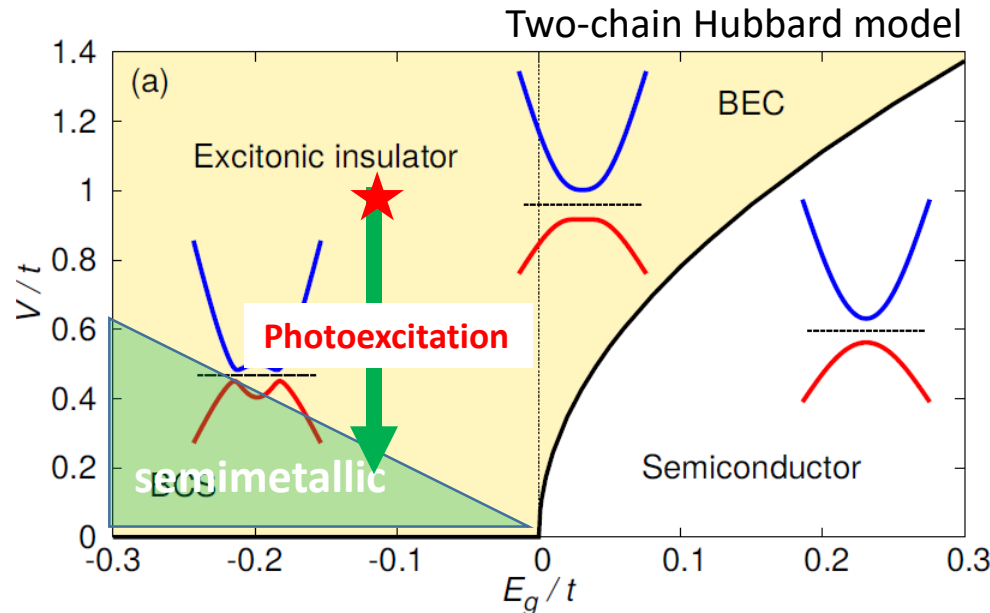
Realization of *semimetallic* state: an intriguing example



Photoexcited carriers screen the Coulomb interaction. Then, we could virtually observe the “noninteracting band dispersions”.

K. Okazaki *et al.*, “Photo-induced semimetallic states realized in electron-hole coupled insulators” Nat. Commun. **9**, 4322 (2018)

Phase diagrams



Photoexcitation reveals the noninteracting band structure.

Without FS nesting in 2D, the PM state appears when $E_g < 0$ and V is small.

Summary

1. The DFT calculation of $\sigma(\omega)$ explains the high-energy spectral features very well.
2. However, for low energies, we find that, while Ta_2NiS_5 is a simple semiconductor, Ta_2NiSe_5 is a band-overlap semimetal, even in the mBJ calculations.
3. An effective model calculation for Ta_2NiSe_5 showed that the large peak appears in $\sigma(\omega)$ and the band gap opens in the single-particle spectrum, which are due to V .
4. The ground state of Ta_2NiSe_5 is an excitonic insulator of the BEC type, despite the fact that the noninteracting band structure is a band-overlap semimetal.
5. A novel insulator state caused by the strong electron-hole attraction is predicted, which may be contrasted with a Mott insulator state caused by the strong electron-electron repulsion.

K. Sugimoto, S. Nishimoto, T. Kaneko, and Y.O.,
Strong Coupling Nature of the Excitonic Insulator State in Ta_2NiSe_5
Phys. Rev. Lett. **120**, 247602 (2018)