Theoretical perspectives on an excitonic insulator Ta$_2$NiSe$_5$

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Contents

1. Introduction
2. Theoretical aspects of Ta$_2$NiSe$_5$
3. New perspectives  
   3.1 Optical conductivity  
   3.2 Novel insulator state
4. Summary

Collaborators

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Experimental information

Sawa, Takagi, Mizokawa, Okazaki, ...

K. Sugimoto *et al.*,  
Conventional phase diagram

Ta$_2$NiSe$_5$ 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$_2$NiSe$_5$.

Structural phase transition of Ta$_2$NiSe$_5$


Orthonhombic monoclinic

$T_c = 328$ K

$\beta = 90.59^\circ$

at $T = 150$ K.

X-ray diffraction experiment

No CDW modulations but uniform distortion!

Insulating in entire temperature range.

Origin?
Band structure calculations

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.

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

WIEN2K (FLAPW, GGA)
Structural optimization for the orthorhombic structure

Belong to different irreducible representations at the $\Gamma$ point of BZ. No hybridization occurs!
Effective model and mean-field analysis

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

Singlet exciton of Ni $3d$ hole and Ta $5d$ electron

Excitonic Bond Order formation ($Q = 0$)

Orthorhombic monoclinic

Orthorhombic

Monoclinic

$T_c = 328$ 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 $\Gamma$-X direction, $T = 40$ K

Flat band with double-peak structure!

Spontaneous hybridization

Also see: Wakisaka et al., PRL 103, 026402 (2009)
Other quantities of \( \text{Ta}_2\text{NiSe}_5 \)

1. Specific heat
   Sugimoto \textit{et al.}, PRB \textbf{93}, 041105(R) (2016).

2. Elastic shear constant
   Sugimoto \textit{et al.}, PRB \textbf{93}, 041105(R) (2016).

3. Quantum interference (NMR and ultrasonic attenuation)
   Sugimoto \textit{et al.}, PRB \textbf{93}, 041105(R) (2016).

4. Diamagnetic susceptibility
   Sugimoto and Y.O., PRB \textbf{94}, 085111 (2016)

\textit{So far, so good!}
Comparative study of optical conductivity: $\text{Ta}_2\text{NiSe}_5$ vs. $\text{Ta}_2\text{NiS}_5$

- **Anomalous peak!**

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

Larkin et al., PRB 95, 195144 (2017)
Comparison between $\text{Ta}_2\text{NiSe}_5$ and $\text{Ta}_2\text{NiS}_5$

$\text{Ta}_2\text{NiSe}_5$ ... No phase transition is observed, even under high pressures.

$\text{Ta}_2\text{NiS}_5$ ... a simple semiconductor

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

$\text{Ta}_2\text{NiS}_5$ ... a semimetal?!

$\text{Ta}_2\text{NiSe}_5$ ... a strange insulator

Tran and Blaha, PRL 102, 226401 (2009)
Optical conductivity in $\text{Ta}_2\text{NiSe}_5$ and $\text{Ta}_2\text{NiS}_5$

**Theory by mBJ with $c = 1.5$**

(a) $E \parallel a$

(b) $E \parallel c$

**Experiment**

Experimental data from Larkin et al., PRB 95, 195144 (2017); Larkin, PhD Thesis, Univ. Stuttgart (2016)
Origin of the anomalous peak?

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.

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

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

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

**Ta$_2$NiSe$_5$**

**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

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$\text{Ta}_2\text{NiSe}_5$

Three-chain Hubbard model

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

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

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

$\mu$-metal 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”.

Phase diagrams

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

Photoexcitation reveals the noninteracting band structure.
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 $\text{Ta}_2\text{NiS}_5$ is a simple semiconductor, $\text{Ta}_2\text{NiSe}_5$ is a band-overlap semimetal, even in the mBJ calculations.

3. An effective model calculation for $\text{Ta}_2\text{NiSe}_5$ 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 $\text{Ta}_2\text{NiSe}_5$ 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 $\text{Ta}_2\text{NiSe}_5$