Giant exciton Fano resonances in Ta$_2$NiSe$_5$

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Acknowledgements

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YF Lu...AVB, and H. Takagi, Nature Commun. 8, 14408 (2017)
TI Larkin..., and AVB, PRB 95, 195144 (2017)
TI Larkin..., and AVB, *Editors’ Suggestion* PRB 98, 125113 (2018)
Electrodynamics of quasi-1D Ta$_2$NiSe$_5$ vs. Ta$_2$NiS$_5$

wide-band THz-UV ellipsometry:

\[
\hat{\epsilon} = \begin{pmatrix}
\epsilon_{xx} & 0 & 0 \\
0 & \epsilon_{yy} & 0 \\
0 & 0 & \epsilon_{zz}
\end{pmatrix}
\]

\[
\epsilon^a_1(\omega, T) = \frac{\omega}{4\pi} \sigma_1(\omega, T)
\]

K. Seki et al., PRB 90 155116 (2014)
Optical conductivity of Ta$_2$NiSe$_5$ vs. Ta$_2$NiS$_5$

$E_g \sim E'_g \sim 180$-200 meV

TI Larkin..., and AVB, PRB 95, 195144 (2017)
Excitonic vs. semiconducting band gap

YF Lu...AVB, and H. Takagi, Nature Commun. 8, 14408 (2017)
Excitonic vs. semiconducting band gap

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

YF Lu...AVB, and H. Takagi, Nature Commun. 8, 14408 (2017)
Control of $T_C$ by changing $E_G$

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

Optical conductivity and band structure calculations

DFT-based calculations using the modified semilocal exchange (Becke-Johnson) potential with an appropriate choice of the parameters following the approach of “Accurate Band Gaps of Semiconductors and Insulators with a Semilocal Exchange-Correlation Potential” by Tran and Blaha (PRL, 102, 226401, 2009)

K Sugimoto et al., PRL, 120, 247602 (2018)
Electron-phonon interaction

\[ \omega_j(T), \Gamma_j(T) \sim \frac{1}{e^{(\hbar \omega_{ph}/k_B T)} - 1} \]

\[ \omega_{ph}^{Se} = 13 \pm 17 \text{ meV} \]

\[ \omega_{ph}^S = 21 \pm 24 \text{ meV} \]

\[ \frac{\omega_{ph}^S}{\omega_{ph}^{Se}} = \sqrt{\frac{m_{Se}}{m_S}} \]
Absorption edge

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

0.7$e$/Ni

**Ta$_2$NiS$_5$**

0.35$e$/Ni

[Graphs showing the absorption edge and conductivity for Ta$_2$NiSe$_5$ and Ta$_2$NiS$_5$.]
Optical gap in $\text{Ta}_2\text{NiSe}_5$

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

Conductivity ($10^3 \Omega^{-1}\text{cm}^{-1}$)

Perrmittivity

Photon energy (eV)

Optical peak position (eV)

ARPES peak position (eV)

Optical peak width (eV)

ARPES peak width (eV)

Temperature (K)

Y Wakisaka et al., PRL 103, 026402 (2009)
JSNM 25, 1231 (2012)

K. Seki et al., PRB 90 155116 (2014)
Optical gap in $\text{Ta}_2\text{NiSe}_5$

$0.17 \text{ eV} \approx 2 \times E_a^\text{DC}$
Exciton doublet

Ta$_2$NiSe$_5$

$\Delta E^{\text{ex}}_{2,1} = 0.1 \text{ eV}$

$E_b = \frac{m^*/m}{\varepsilon^2} \times \text{Ry} = \alpha(\Delta E^{\text{ex}}_{2,1}), \quad \alpha \geq 1$

Ta$_2$NiS$_5$

$\Delta E^{\text{ex}}_{2,1} = 0.16 \text{ eV}$
Exciton Fano resonances

3.2 Fano Resonances as a General Feature of the Optical Absorption in Low-Dimensional Semiconductors

S. Glutsch, Springer (2013)

Epitaxial layer of GaAs in a magnetic field of 6 T

S Bar-Ad et al., PRL, 78, 1363 (1997)
Exciton Fano resonances

$\Delta \sigma_1(\omega)$

$\Delta \varepsilon_1(\omega)$

exciton self-trapped in phonon fields + el–h continuum

$\Delta \sigma_1(\omega)$

$\Delta \varepsilon_1(\omega)$
Exciton Fano resonances

\[ \varepsilon_1(\omega) + 4\pi i\sigma_1(\omega)/\omega = A \hat{G} A + \bar{\varepsilon}_{bg}(\omega), \]

\[ A = (A_e, A_1, A_2) \]

\[ G_e^{-1} = \omega_e^{2} + i \gamma_{e,1,2} - i \varepsilon_1(\omega) - \omega^2 \]

\[ \hat{G}^{-1} = \begin{pmatrix} G_e^{-1} & i k_1 \omega & i k_2 \omega \\ i k_1 \omega & G_1^{-1} & 0 \\ i k_2 \omega & 0 & G_2^{-1} \end{pmatrix} \]

\[ q = 1 \]

\[ \Delta \sigma_1(\omega) \]

\[ \Delta \varepsilon_1(\omega) \]
Giant oscillator strength

effective number of electron per Ni atom contributing to the quantum interference effect

"Giant oscillator strength associated with exciton complexes"

Rashba antenna effect

Quantum Cavity Effect

\[ \phi_{ex/ph} \sim \frac{R^3}{V_{u.c}} f_{ex} \]

"Giant oscillator strength associated with exciton complexes"

Exciton-phonon complexes

$$f^{\text{ex/ph}}(Se) >> f^{\text{ex/ph}}(Se)$$

$$R^{\text{ex/ph}}(Se) >> R^{\text{ex/ph}}(Se)$$

consistent with long-range monoclinic distortions, related to formation of the excitonic insulator state in Ta$_2$NiSe$_5$
Phonons in $\text{Ta}_2\text{NiS}_5$

orthorhombic Cmcm

in-gap soft electronic mode
0.5% of the total spectral weight of the el-h transitions across the direct gap

TI Larkin..., and AVB, PRB 98, 125113 (2018)
Phonons in $\text{Ta}_2\text{NiSe}_5$

Monoclinic distortion allows bidirectional ac-plane activity of $B_{1u}/B_{3u}$ modes, as they are assigned in the orthorhombic $C2/c$ distortion.

### Table II

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<thead>
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<th>Experiment</th>
<th>Calculation</th>
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<td>$c$ axis</td>
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TI Larkin..., and AVB, PRB 98, 125113 (2018)
Phonons in Ta$_2$NiSe$_5$

monoclinic C2/c

7B$_{1u}$ + 4B$_{3u}$ $\rightarrow$ 11B$_u$

1: $\beta_1 (B_1)$ 4.37 (4.71) meV
2: $\beta_1 (B_3)$ 11.65 (14.08) meV
3: $\beta_1 (B_3)$ 12.89 (10.58) meV
4: $\beta_1 (B_3)$ 17.40 (16.74) meV
5: $\beta_1 (B_3)$ 19.27 (21.05) meV
6: $\beta_1 (B_3)$ 20.51 (18.61) meV
7: $\beta_1 (B_3)$ 22.23 (22.49) meV
8: $\beta_1 (B_3)$ 26.13 (25.89) meV
9: $\beta_1 (B_3)$ 28.01 (26.10) meV
10: $\beta_1 (B_3)$ 29.27 (30.05) meV
11: $\beta_1 (B_3)$ 35.64 (34.63) meV

TI Larkin..., and AVB, PRB 98, 125113 (2018)
Phonons in $\text{Ta}_2\text{NiSe}_5$

monoclinic C2/c

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

$7B_{1u} + 4B_{3u} \rightarrow 11B_u$

TI Larkin..., and AVB, PRB 98, 125113 (2018)
Phonons in Ta$_2$NiSe$_5$

monoclinic C2/c

TD-THz

synchrotron-based ellipsometry

7B$_{1u}$ + 4B$_{3u}$ → 11B$_u$
Optical gap filling in Ta$_2$NiSe$_5$

monoclinic C2/c → orthorhombic Cmcm

TI Larkin, AB et al., PRB 98, 125113 (2018)

TD-THz

mid-IR ellipsometry

TI Larkin..., and AVB, PRB 95, 195144 (2017)
In Ta$_2$NiSe$_5$, the giant spectral weight of the exciton Fano resonances due to antenna emission of large exciton-phonon complexes implies that they are largely extended, overlapping and span the entire crystal, preserving the translational symmetry of the lattice.

In Ta$_2$NiS$_5$, on the other hand, the localized exciton-phonon complexes cause local distortions and do not develop a long-range order.

This difference reflects the different nature of the ground states of these two compounds, in good agreement with the EI hypothesis in Ta$_2$NiSe$_5$ but not in Ta$_2$NiS$_5$. Comparative studies of Ta$_2$NiSe$_5$ and Ta$_2$NiS$_5$, are required to confirm the EI hypothesis.