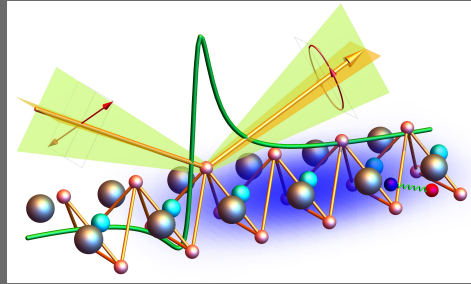


Giant exciton Fano resonances in Ta_2NiSe_5



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SOLID STATE RESEARCH
STUTTGART**



Acknowledgements



T. I. Larkin, R. Dawson, M. Höppner, and B. Keimer

T. Takayama*, A.N. Yaresko, A. W. Rost*, and H. Takagi*

Y. F. Lu and M. Isobe - *crystal growth*

** also @ Institute for Functional Materials
and Quantum Technology, Uni of Stuttgart*



K. A. Kikoin



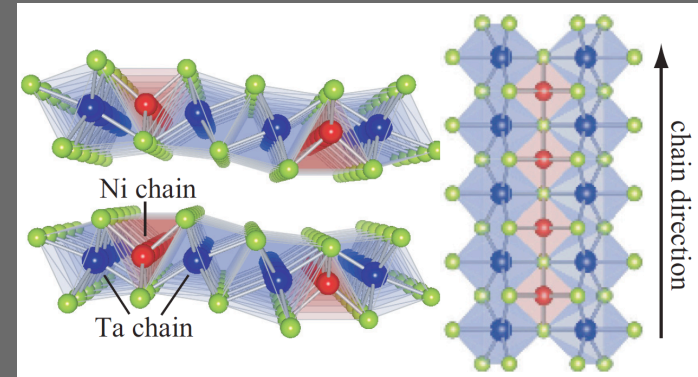
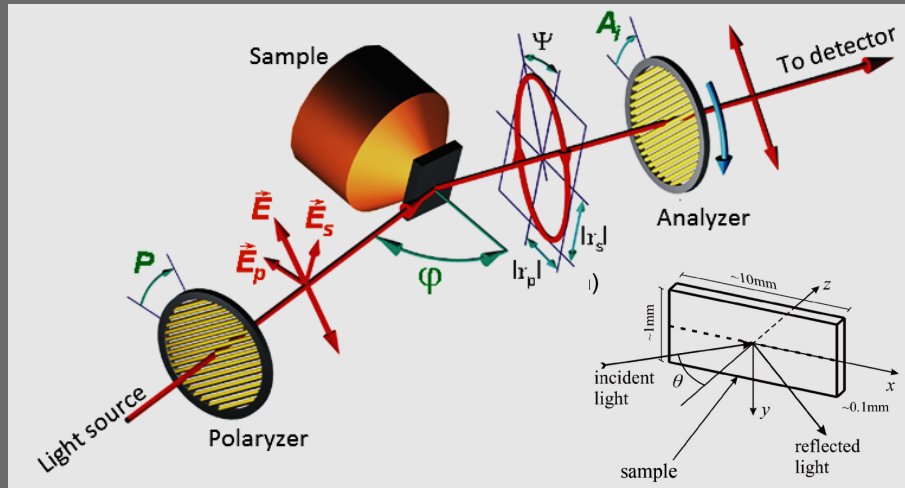
*School of Physics and Astronomy,
Tel-Aviv Uni, Israel*



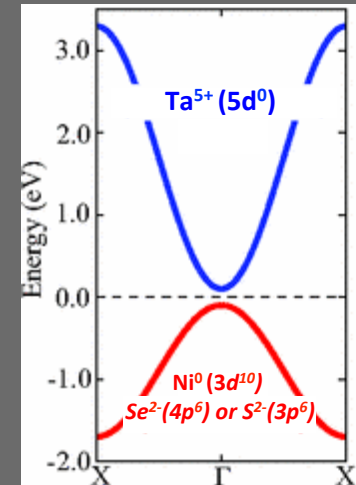
YF Lu...AVB, and H. Takagi, Nature Commun. 8, 14408 (2017)
TI Larkin..., and AVB, PRB 95, 195144 (2017)
TI Larkin..., and AVB, **Editors' Suggestion** PRB 98, 125113 (2018)
T. I. Larkin, PhD Thesis, University of Stuttgart, 2016

Electrodynamics of quasi-1D Ta₂NiSe₅ vs. Ta₂NiS₅

wide-band THz-UV ellipsometry:

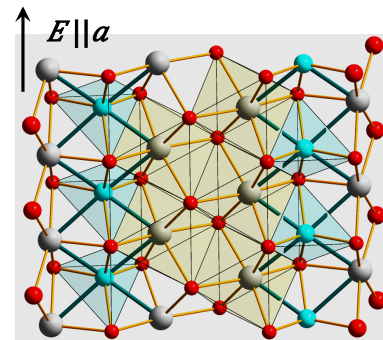
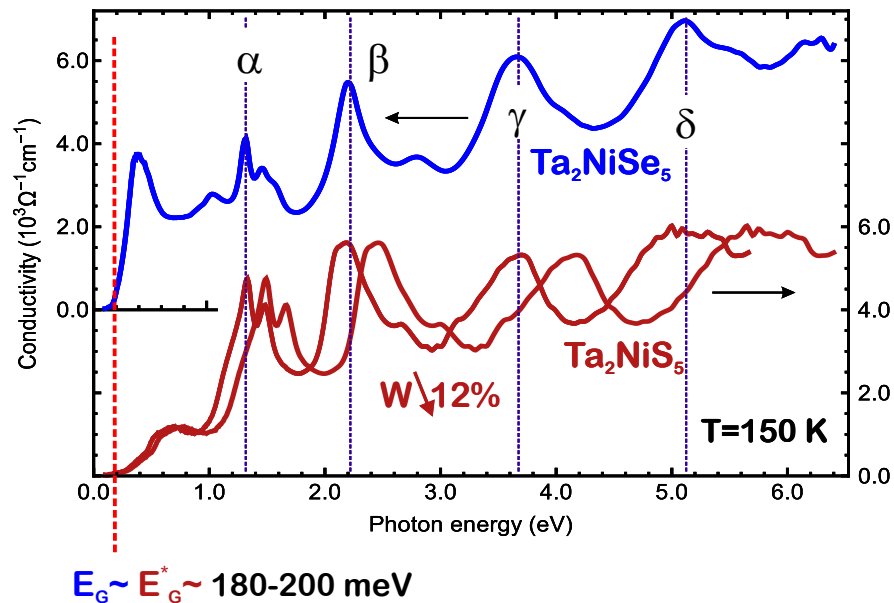


$$\hat{\epsilon} = \begin{pmatrix} \epsilon_{xx} & 0 & 0 \\ 0 & \epsilon_{yy} & 0 \\ 0 & 0 & \epsilon_{zz} \end{pmatrix} : \epsilon_1^a(\omega, T) = \frac{\omega}{4\pi} \epsilon_1^a(\omega, T)$$



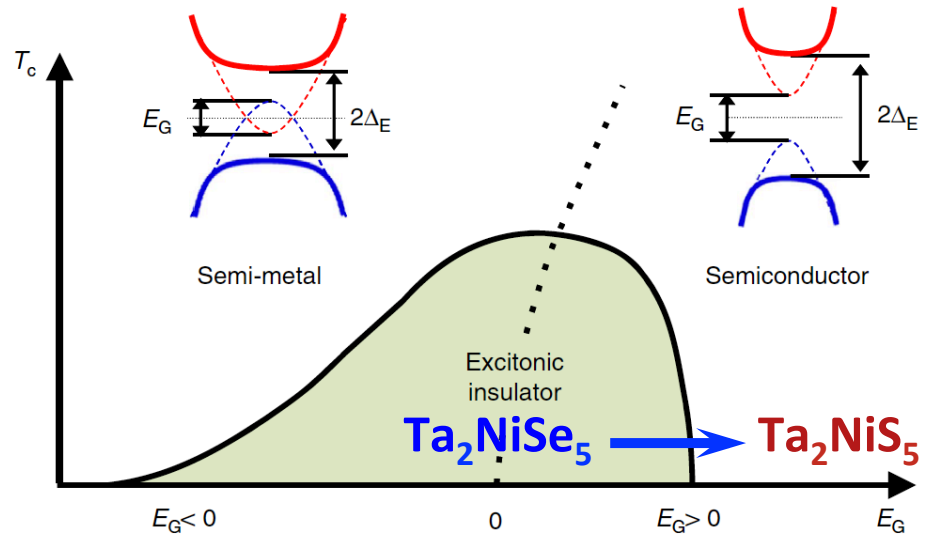
K. Seki *et al.*, PRB 90 155116 (2014)

Optical conductivity of Ta_2NiSe_5 vs. Ta_2NiS_5



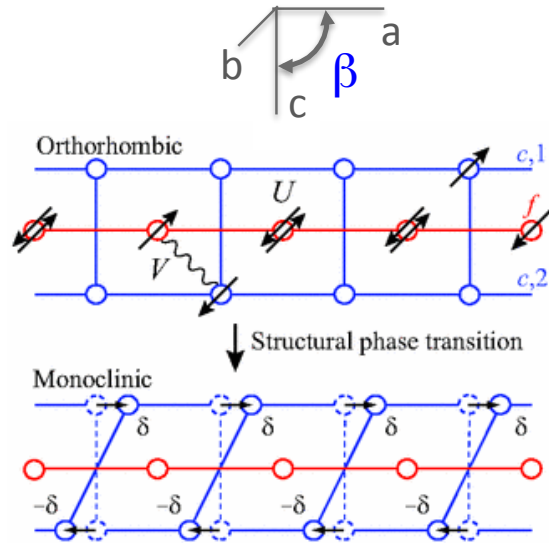
TI Larkin..., and AVB, PRB 95, 195144 (2017)

Excitonic vs. semiconducting band gap

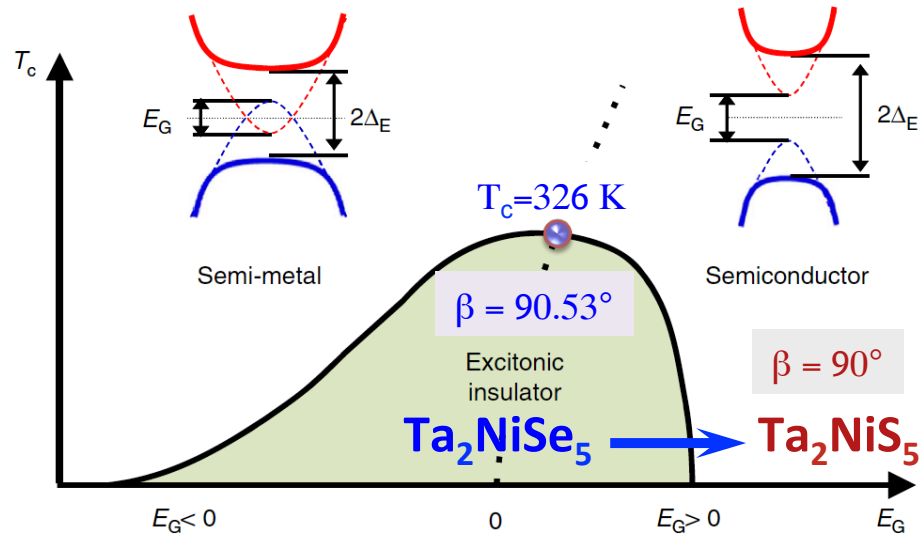


YF Lu...AVB, and H. Takagi, Nature Commun. 8, 14408 (2017)

Excitonic vs. semiconducting band gap

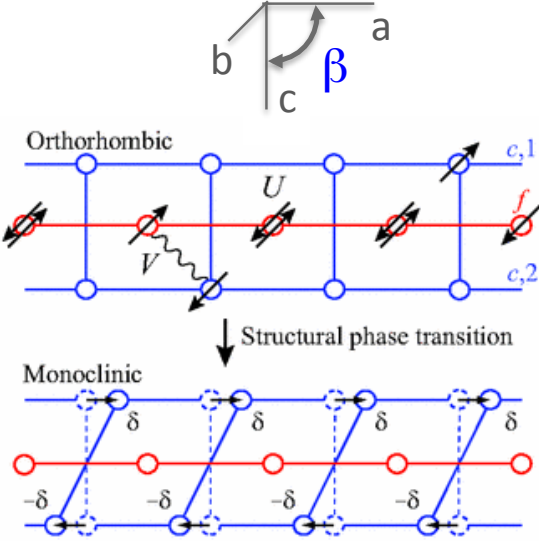


T Kaneko et al, PRB, 87, 035121 (2013)

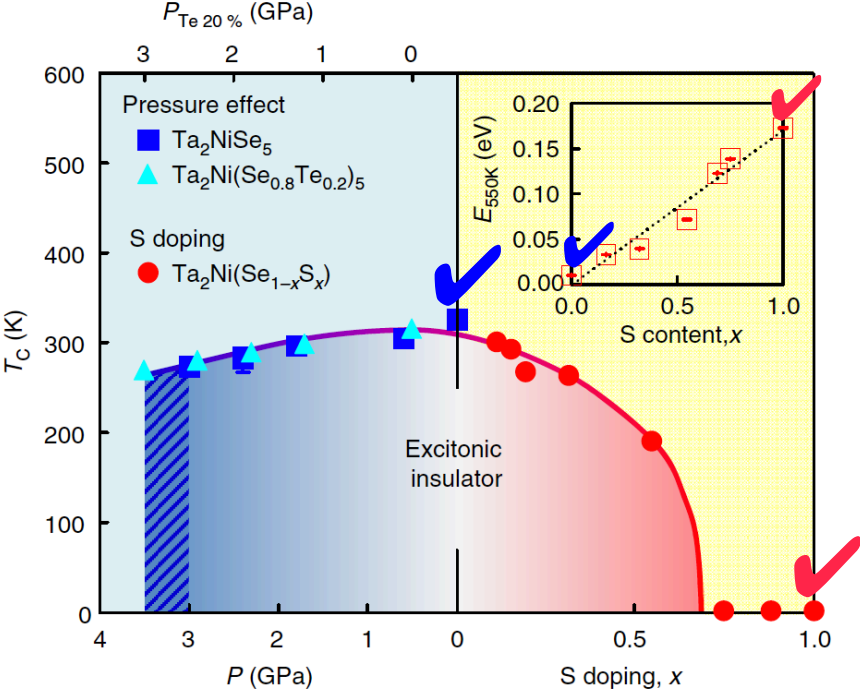


YF Lu...AVB, and H. Takagi, Nature Commun. 8, 14408 (2017)

Control of T_C by changing E_G



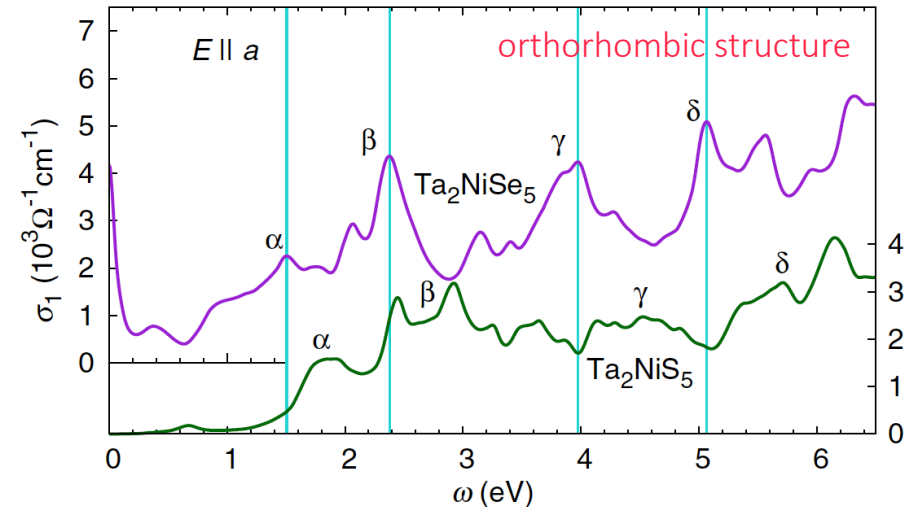
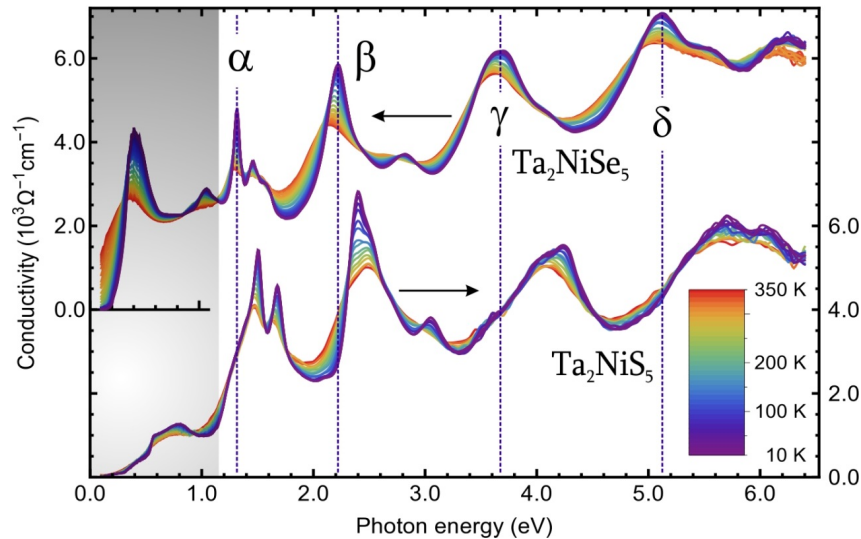
T Kaneko et al, PRB, 87, 035121 (2013)



YF Lu...AVB, and H. Takagi, Nature Commun. 8, 14408 (2017)

Optical conductivity and band structure calculations

DFT-based calculations using the modified semilocal exchange (Becke-Johnson) potential with an appropriate choice of the parameters



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

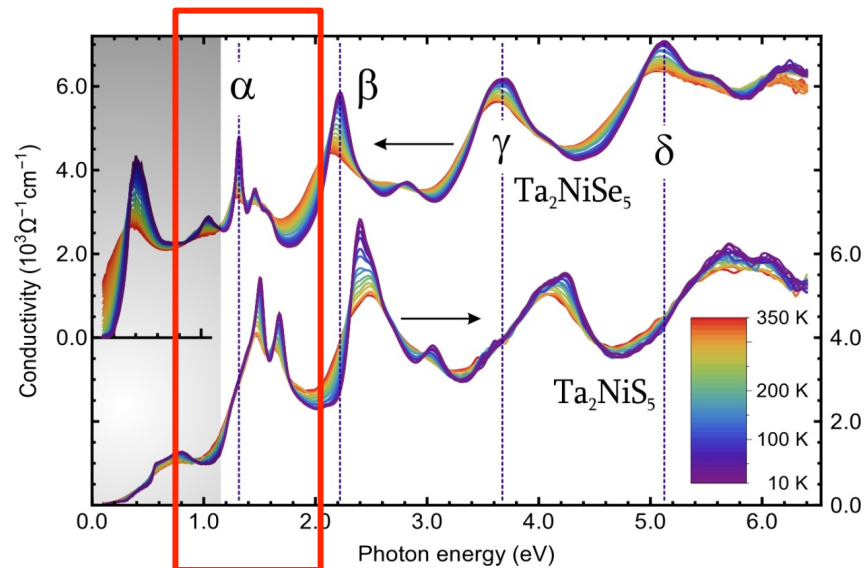
... following the approach of
"Accurate Band Gaps of Semiconductors and Insulators
with a Semilocal Exchange-Correlation Potential"

F Tran and P Blaha, PRL, 102, 226401 (2009)

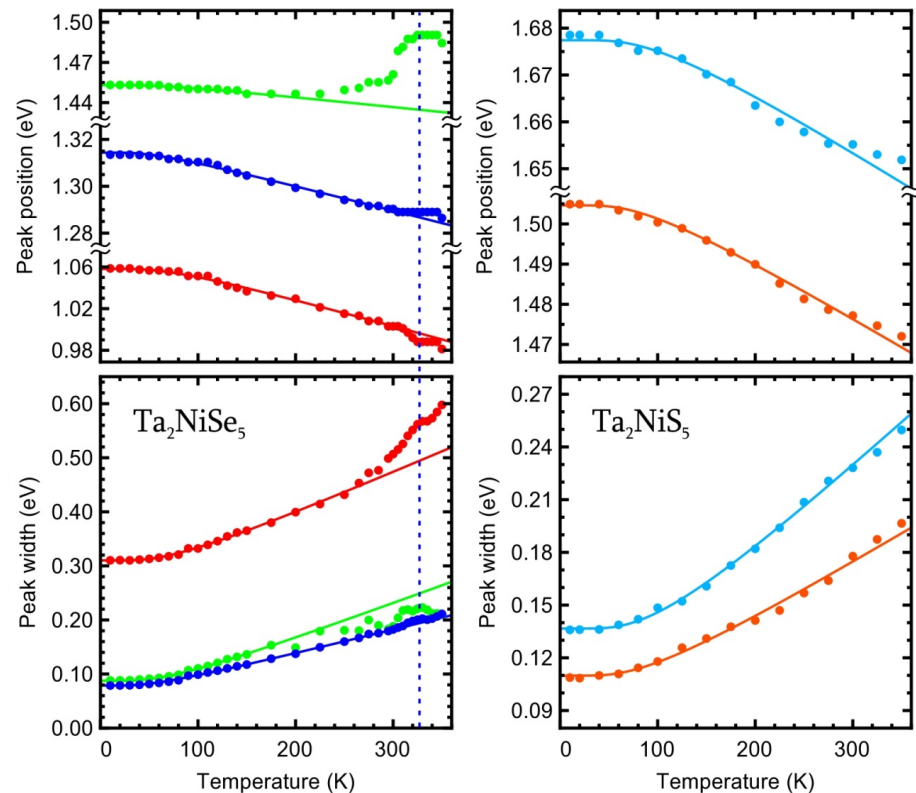
Times Cited: 1,835



Electron-phonon interaction



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



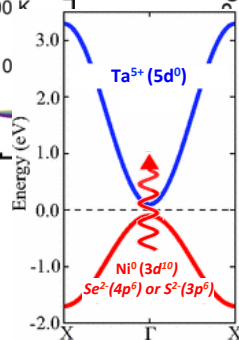
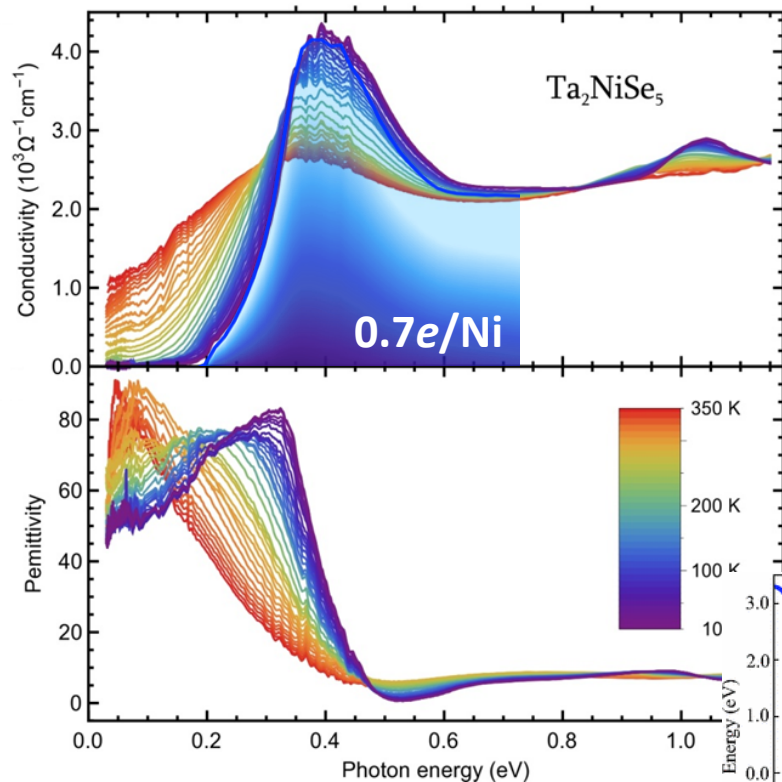
$$\omega_{ph}^{Se} = 13 \div 17 \text{ meV}$$

$$\omega_{ph}^S = 21 \div 24 \text{ meV}$$

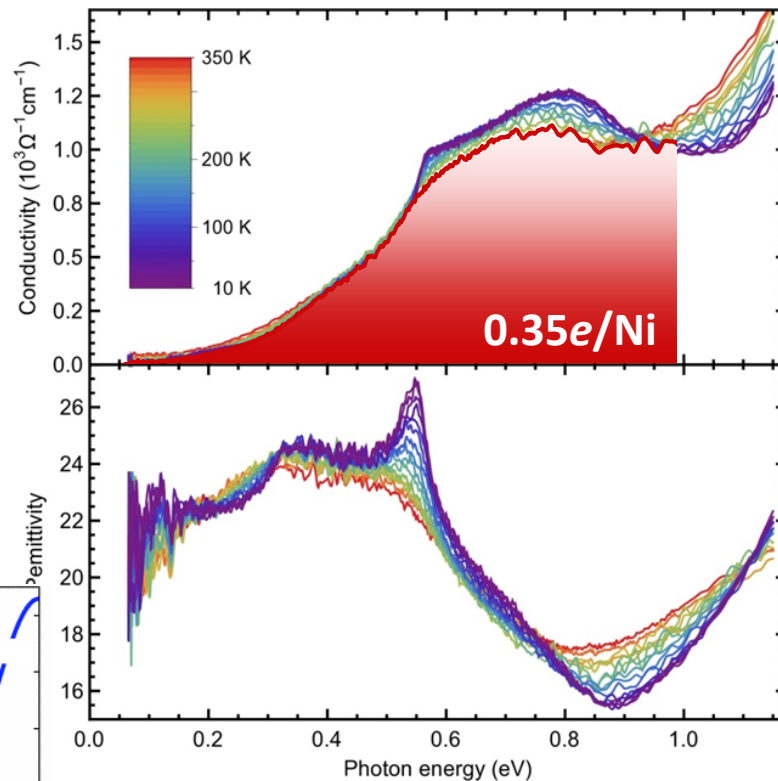
$$\frac{\omega_{ph}^S}{\omega_{ph}^{Se}} = \sqrt{\frac{m_{Se}}{m_S}}$$

Absorption edge

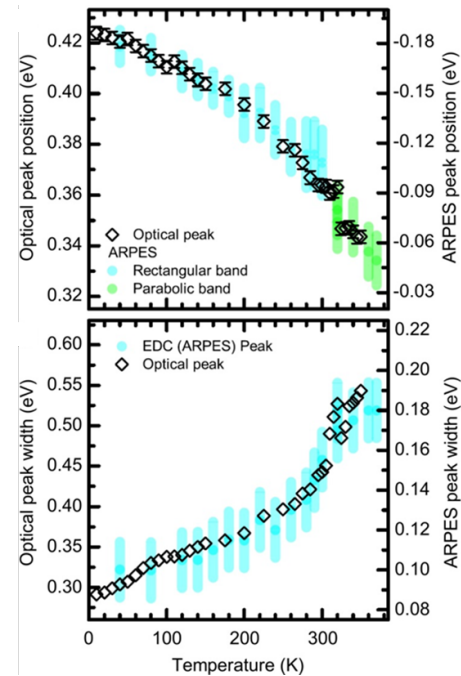
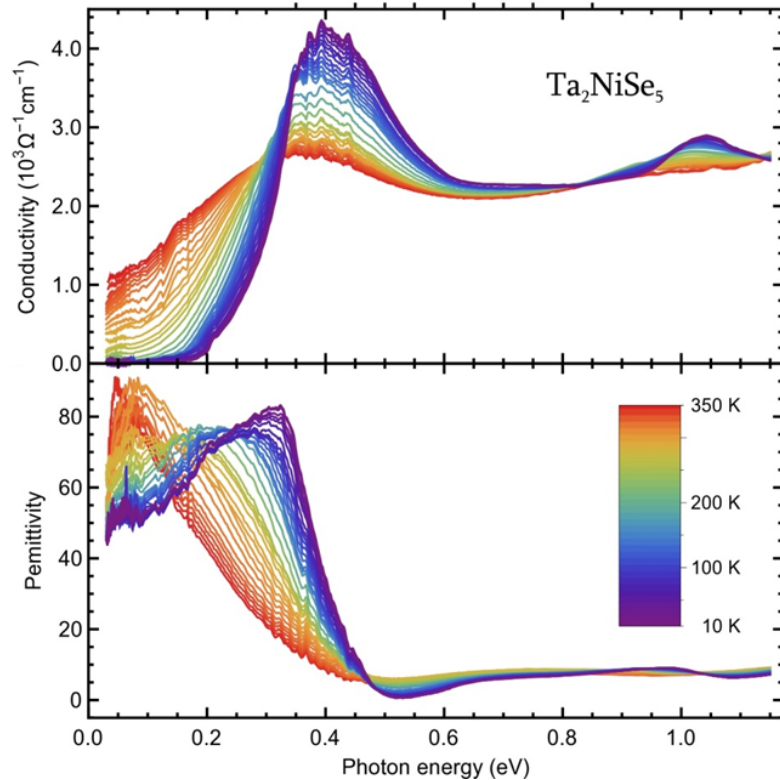
Ta₂NiSe₅



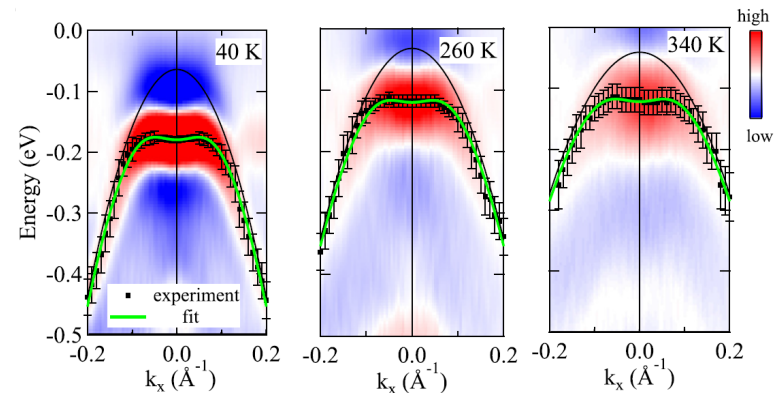
Ta₂NiS₅



Optical gap in Ta₂NiSe₅

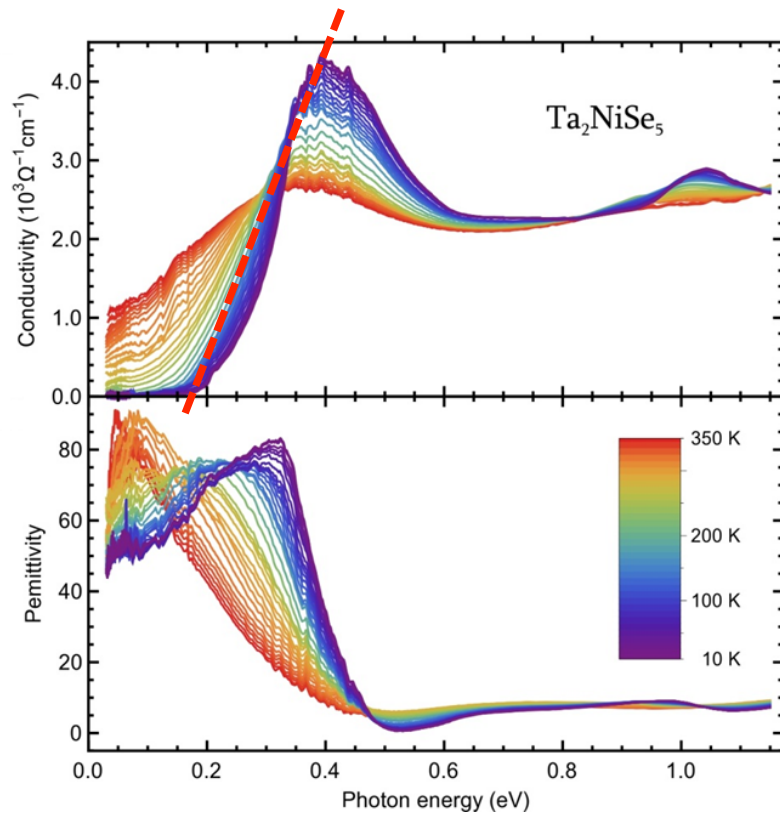


Y Wakisaka *et al.*,
PRL 103, 026402 (2009)
JSNM 25, 1231 (2012)

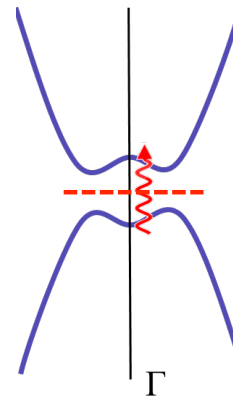


K. Seki *et al.*, PRB 90 155116 (2014)

Optical gap in Ta₂NiSe₅



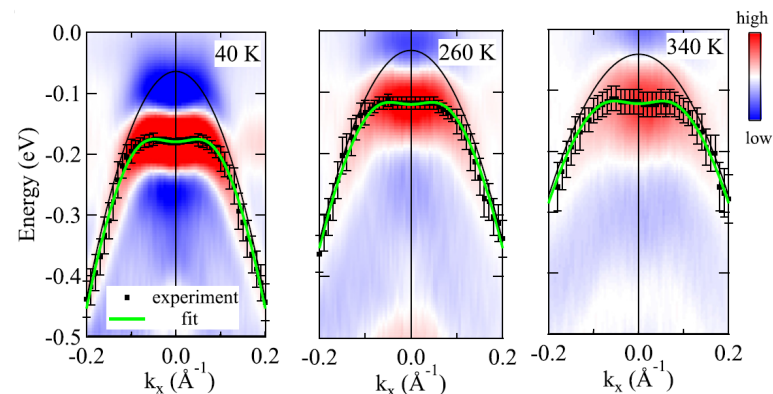
Ta⁵⁺ (5d⁰)



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

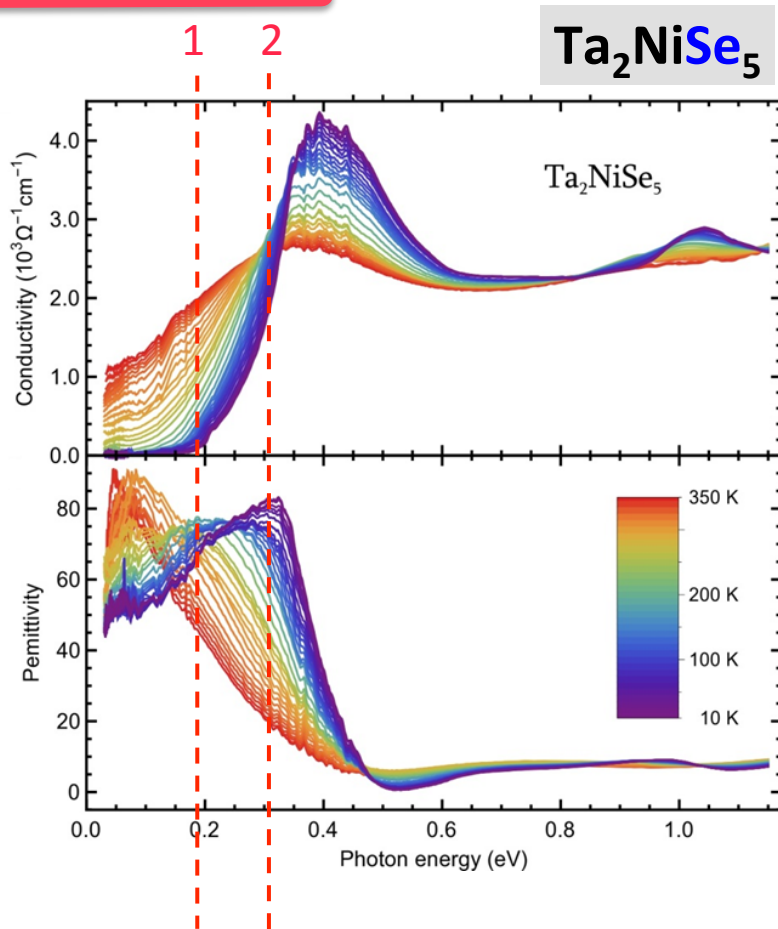
Ni⁰ (3d¹⁰)

Se²⁻ (4p⁶)



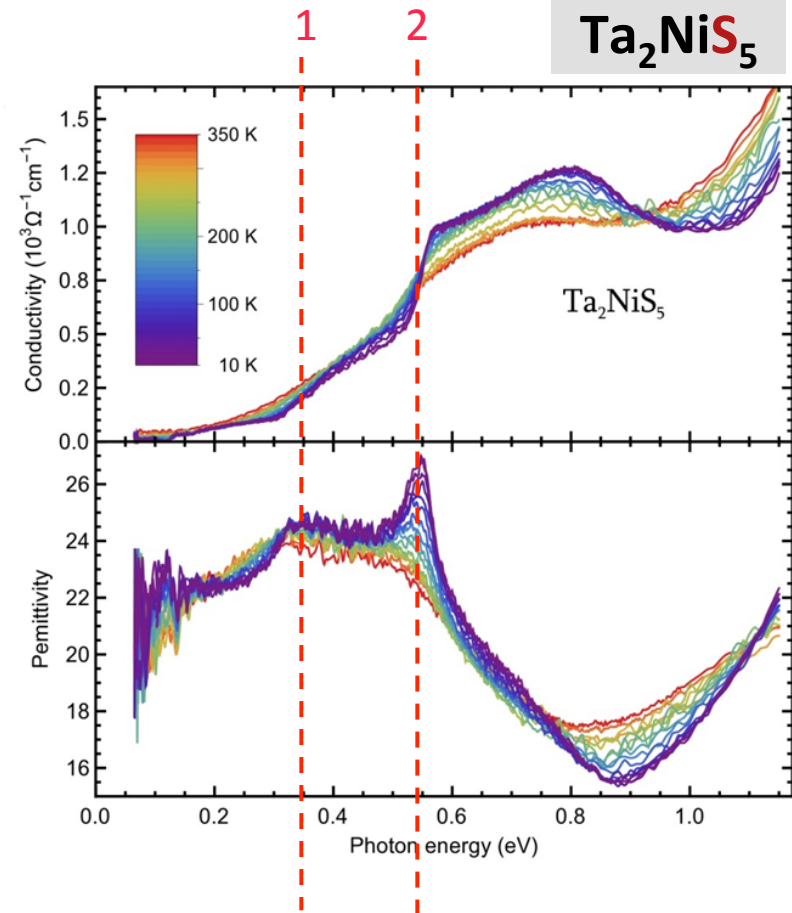
K. Seki *et al.*, PRB 90 155116 (2014)

Exciton doublet



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

$$E_b = \frac{m^*/m}{\epsilon^2} \times Ry = \alpha(\Delta E_{2,1}^{\text{ex}}), \alpha \geq 1$$

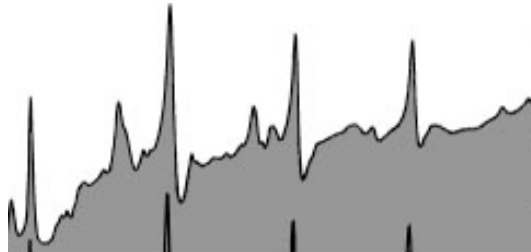
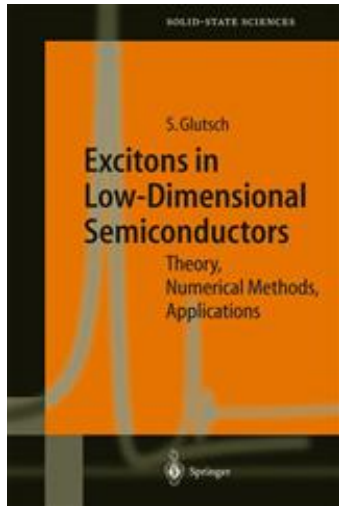


$$\Delta E_{2,1}^{\text{ex}} = 0.16 \text{ eV}$$

Exciton Fano resonances

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

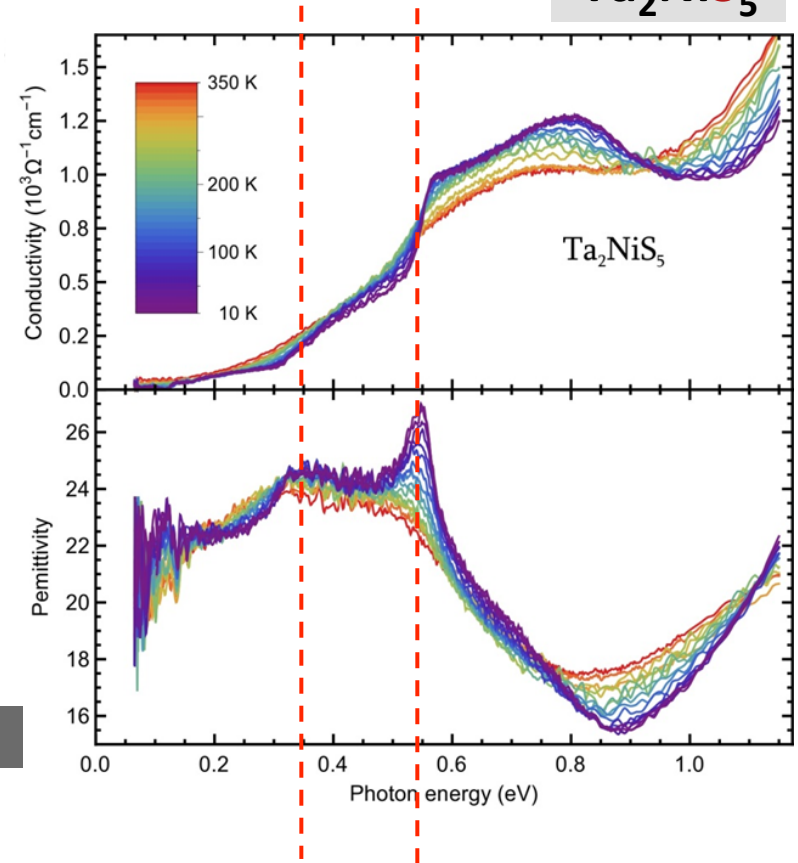
S. Glutsch, Springer (2013)



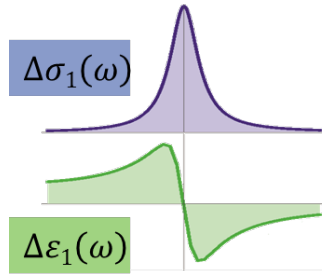
Epitaxial layer of GaAs in a magnetic field of 6 T

S Bar-Ad *et al.*, PRL, 78, 1363 (1997)

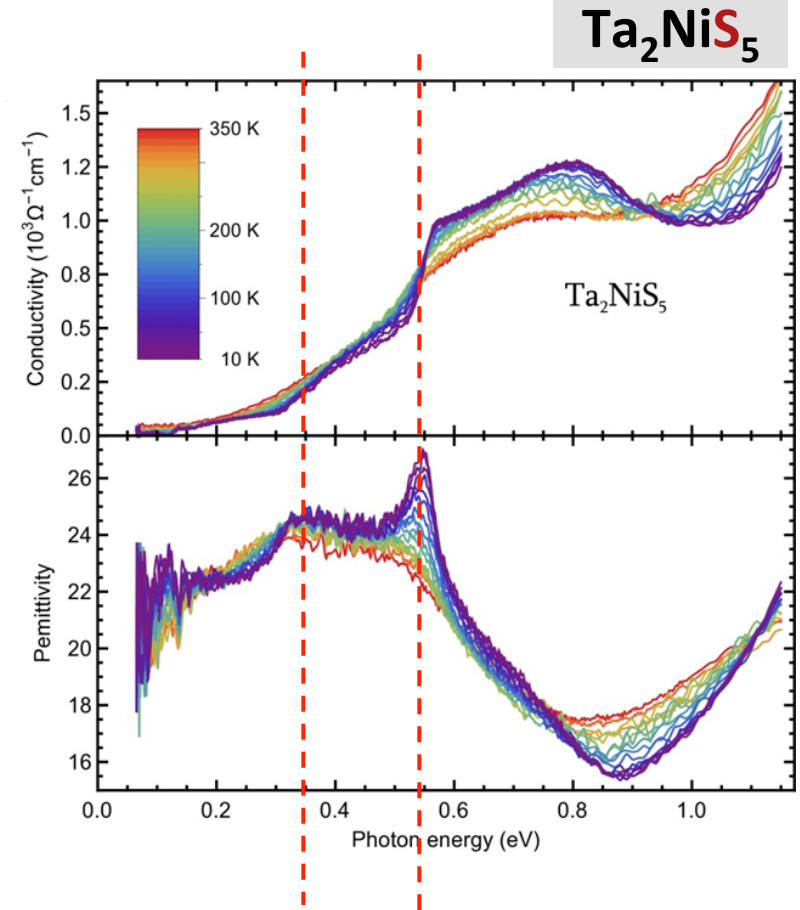
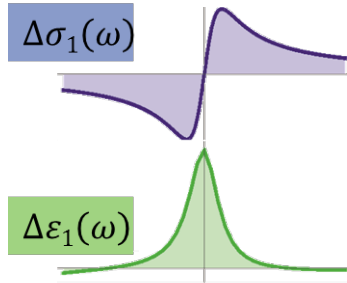
Ta₂NiS₅



Exciton Fano resonances



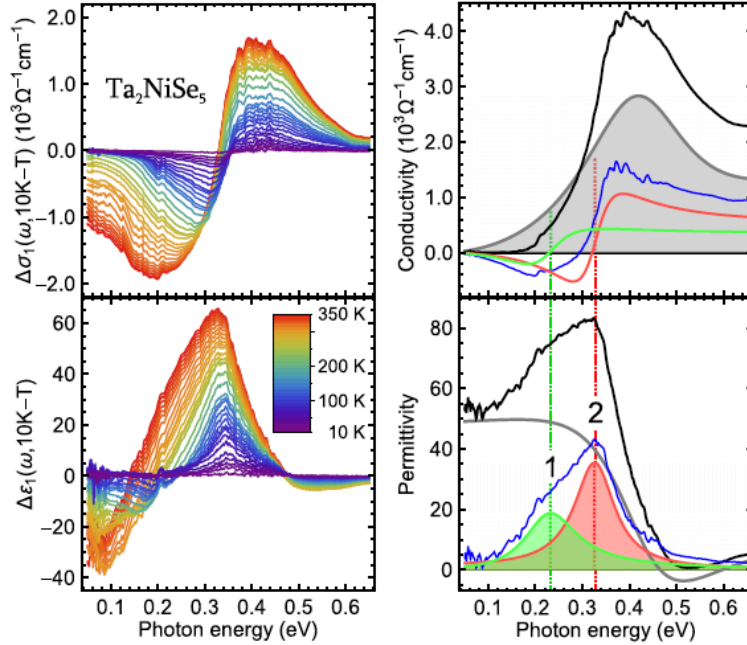
exciton self-trapped
in phonon fields
+
el-h continuum



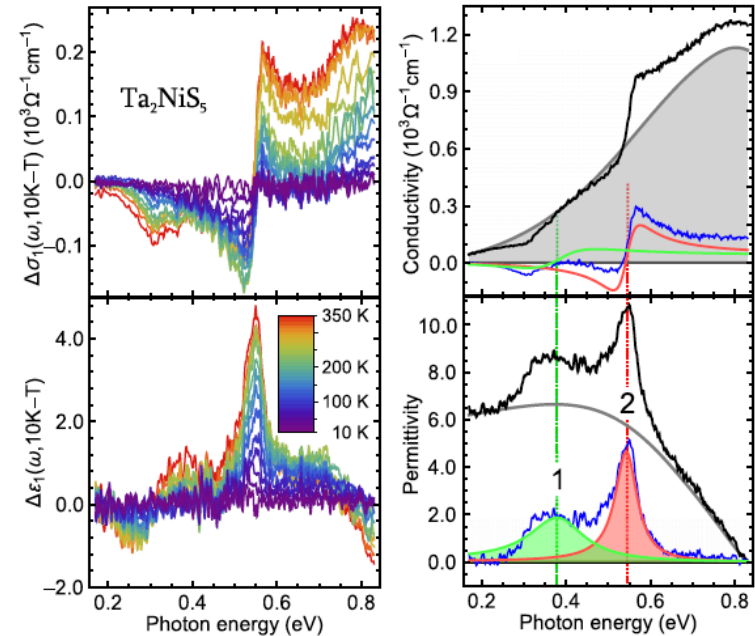
Exciton Fano resonances

TI Larkin..., and AVB, PRB 95, 195144 (2017)

Ta₂NiSe₅



Ta₂NiS₅

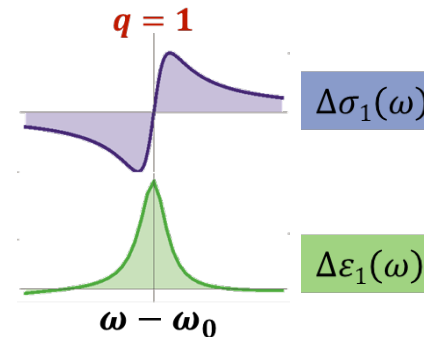


$$\varepsilon_1(\omega) + 4\pi i\sigma_1(\omega)/\omega = \mathbf{A}\hat{\mathbf{G}}\mathbf{A} + \tilde{\varepsilon}_{\text{BG}}(\omega),$$

$$\mathbf{A} = (A_e, A_1, A_2)$$

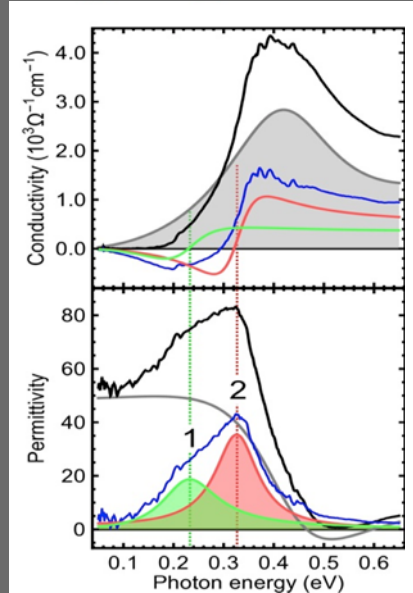
$$G_{e,1,2}^{-1} = \omega_{e,1,2}^2 - i\gamma_{e,1,2}\omega - \omega^2$$

$$\hat{\mathbf{G}}^{-1} = \begin{pmatrix} G_e^{-1} & ik_1\omega & ik_2\omega \\ ik_1\omega & G_1^{-1} & 0 \\ ik_2\omega & 0 & G_2^{-1} \end{pmatrix}$$



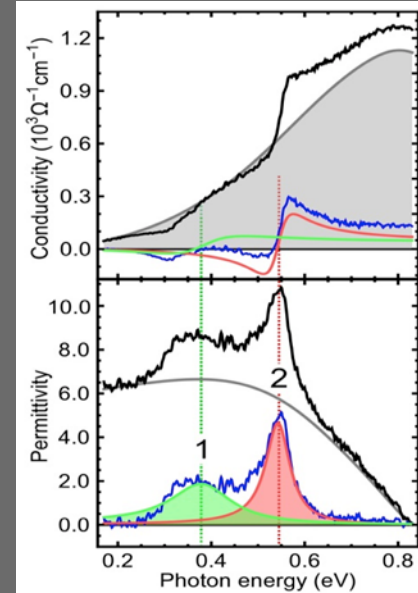
Giant oscillator strength

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



Ta_2NiSe_5

0.26
 e/Ni



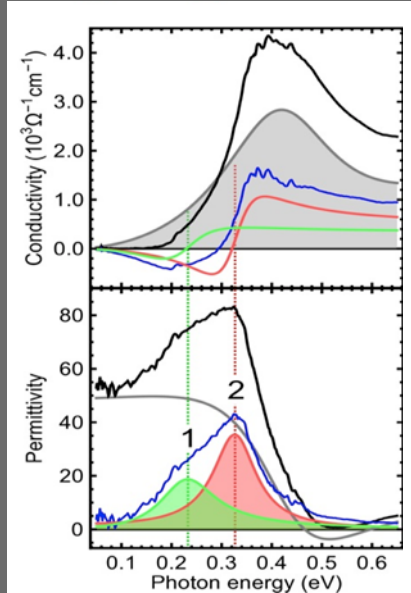
Ta_2NiS_5

0.024
 e/Ni

“Giant oscillator strength associated
with exciton complexes”

EI Rashba, Sov. Phys. Semicond, 8, 807 (1974)

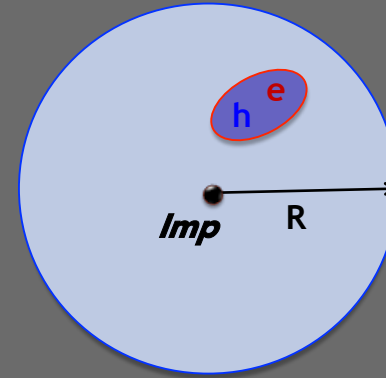
Rashba antenna effect



Ta₂NiSe₅

**0.26
e/Ni**

Quantum Cavity Effect



$$f^{ex/ph} \sim \frac{R^3}{V_{u.c}} f^{ex}$$

“Giant oscillator strength associated with exciton complexes”

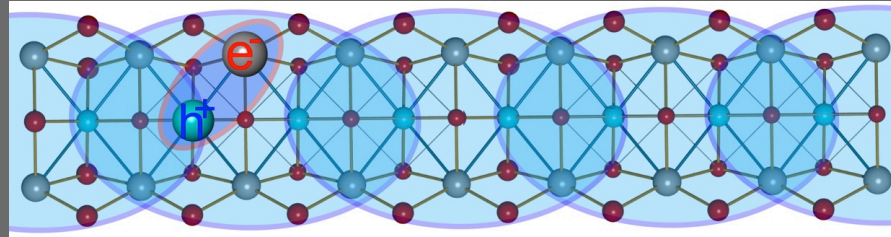
EI Rashba, Sov. Phys. Semicond, 8, 807 (1974)

Exciton-phonon complexes

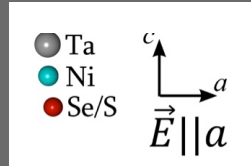
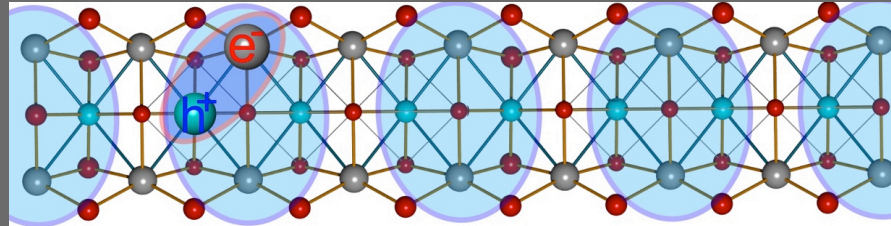
$$f^{ex/ph}(Se) \gg f^{ex/ph}(Se)$$

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

Ta₂NiSe₅

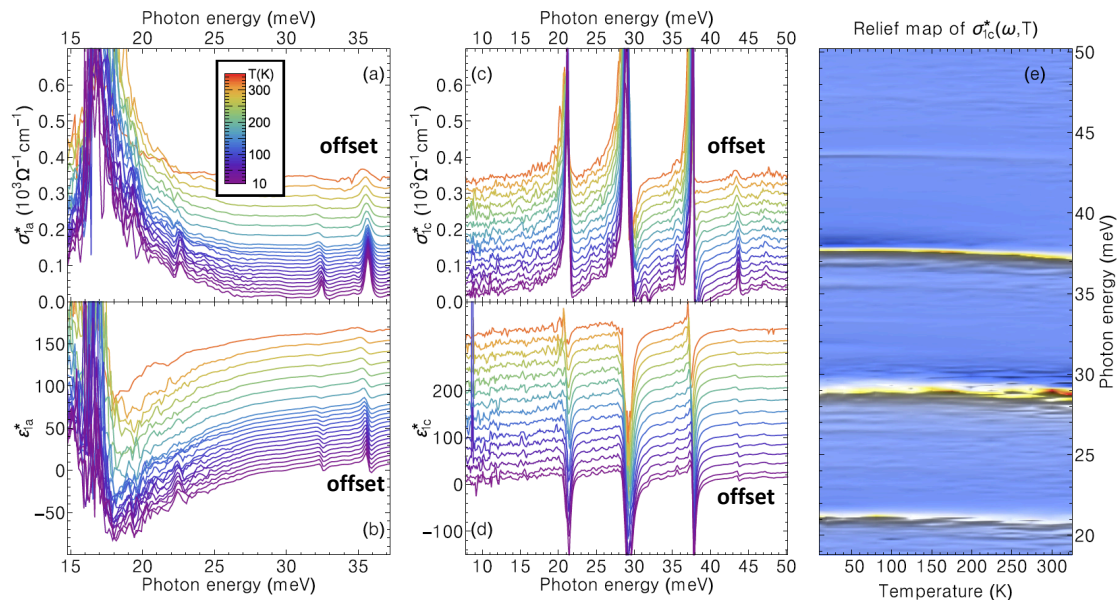


Ta₂NiS₅



consistent with long-range monoclinic distortions, related to formation of the excitonic insulator state in Ta₂NiSe₅

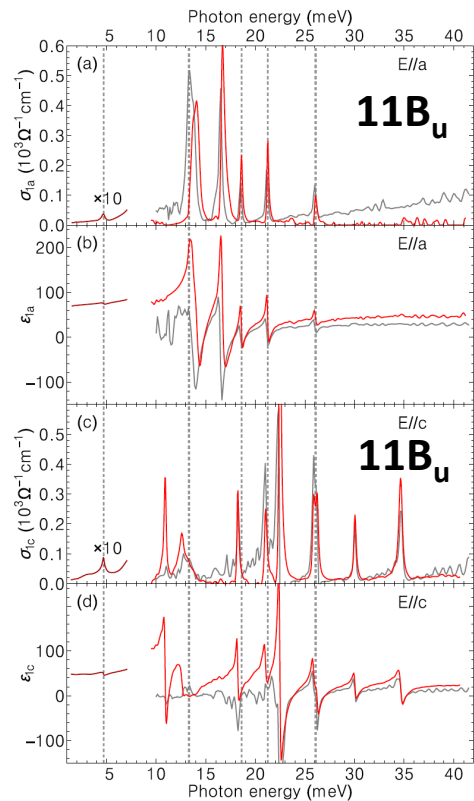
orthorhombic Cmcm



0.5% of the total spectral weight of
the el-h transitions across the direct gap

Phonons in Ta₂NiSe₅

monoclinic C2/c



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

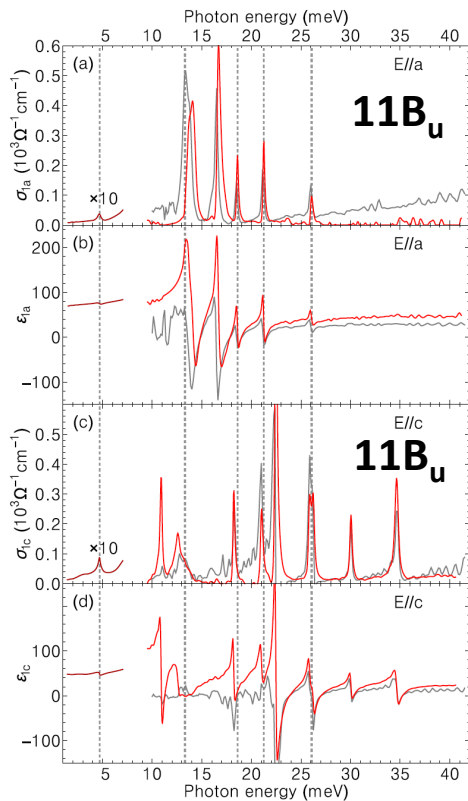
TABLE II. The observed phonon modes in Ta₂NiSe₅ for the two polarizations and the calculated eigenfrequencies of the 11 B_u modes along with a possible assignment. The last column indicates the dominant character (Ch.) of the modes according to orthorhombic symmetry.

Experiment				Calculation		
N	ω_0 (meV)	$\Delta\varepsilon$	γ (meV)	N	ω_0 (meV)	Ch.
<i>a</i> axis				B_u		
1	4.71	0.55	0.37	1	4.37	B_{1u}
2	14.00	10.92	0.62	2	11.65	B_{3u}
4	16.74	6.21	0.40	3	12.89	B_{1u}
6	18.61	0.95	0.20	4	17.40	B_{3u}
5	21.26	0.93	0.20	5	19.27	B_{1u}
9	26.10	0.28	0.29	6	20.51	B_{3u}
<i>c</i> axis				7	22.25	B_{1u}
				8	26.13	B_{1u}
1	4.71	0.55	0.37	9	28.01	B_{3u}
3	10.88	6.90	0.31	10	29.27	B_{1u}
2	12.67	8.32	1.13	11	35.64	B_{1u}
6	18.24	1.56	0.22			
5	21.05	1.14	0.27			
7	22.49	4.39	0.25			
8	25.89	0.71	0.26			
9	26.22	0.95	0.33			
10	30.05	0.54	0.29			
11	34.63	0.96	0.45			

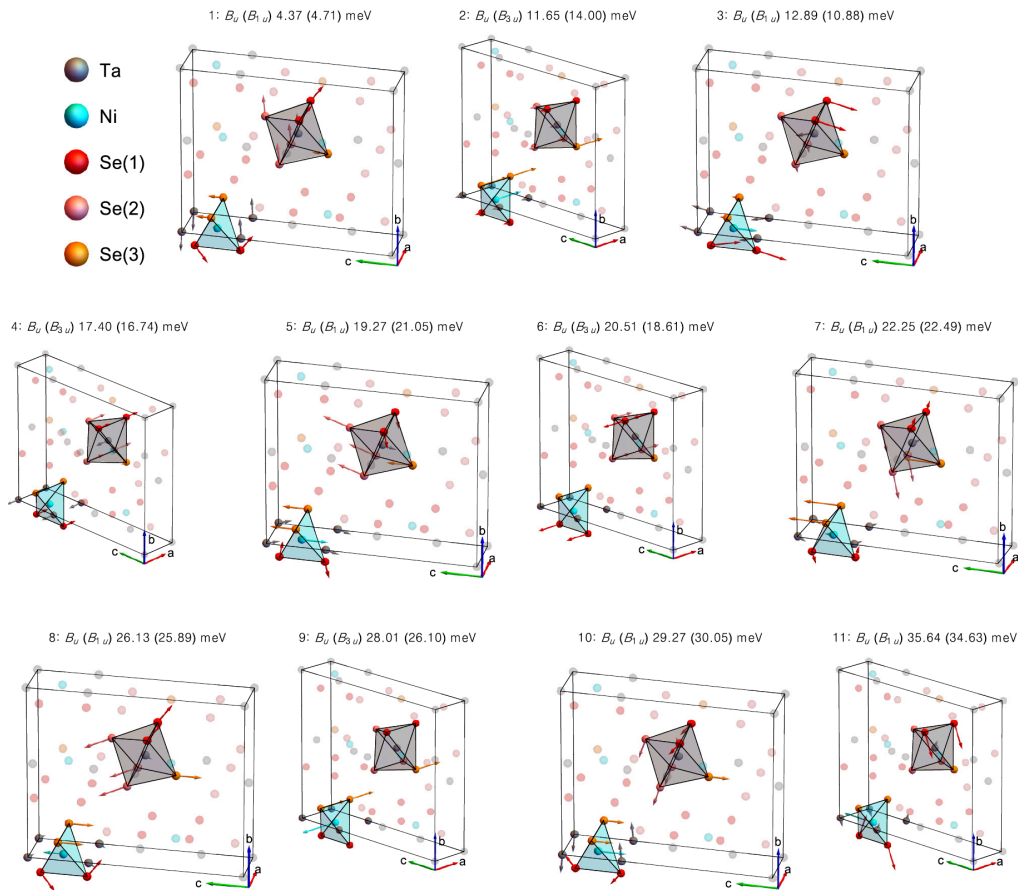
Monoclinic distortion allows bidirectional ac-plane activity of B_{1u}/B_{3u} modes, as they are assigned in the orthorhombic

Phonons in Ta₂NiSe₅

monoclinic C2/c

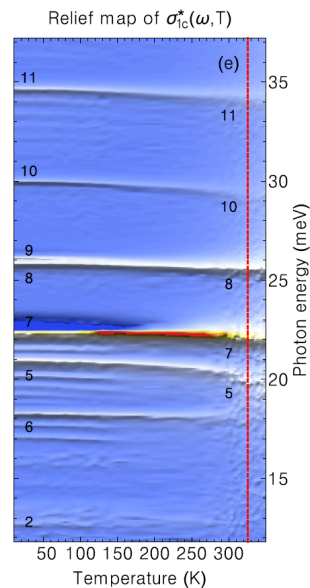
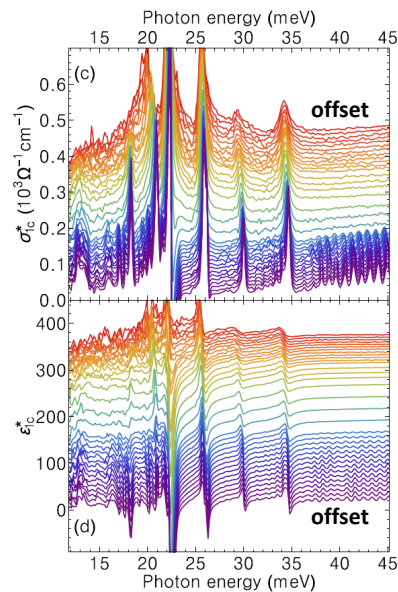
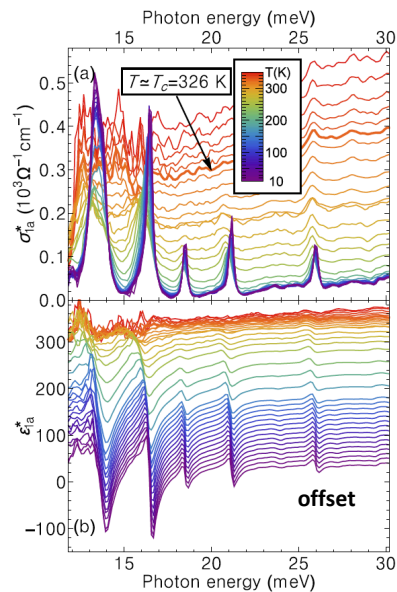
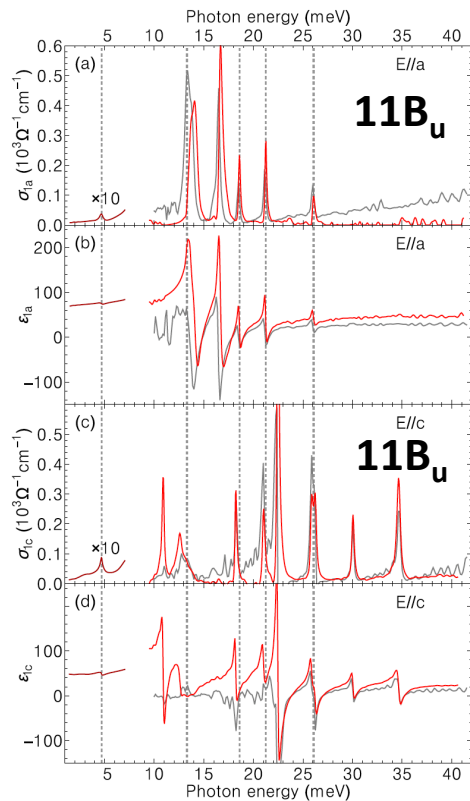


$$7B_{1u} + 4B_{3u} \longrightarrow 11B_u$$



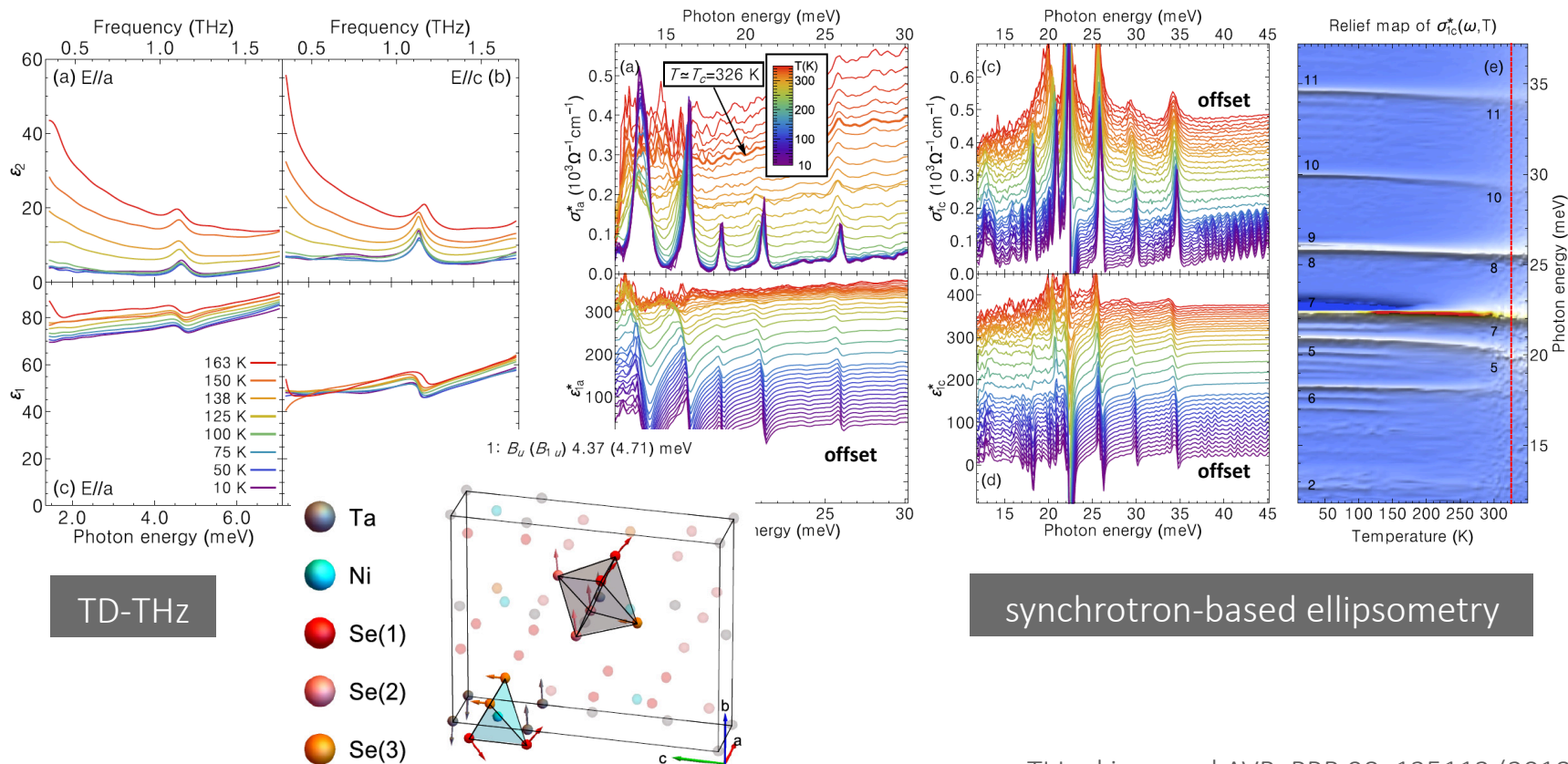
Phonons in Ta₂NiSe₅

monoclinic C2/c



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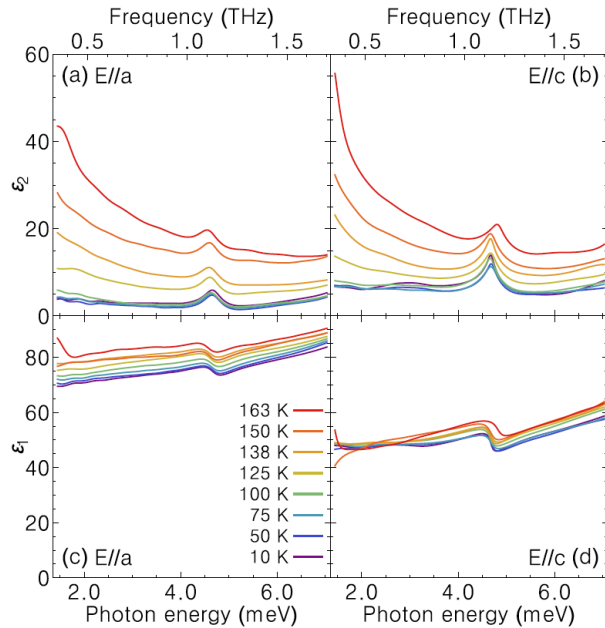
monoclinic C2/c



TI Larkin..., and AVB, PRB 98, 125113 (2018)

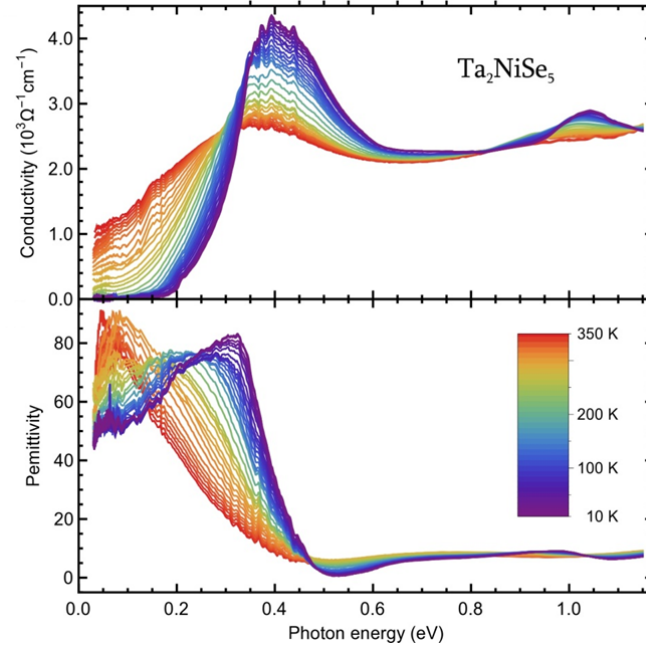
Optical gap filling in Ta₂NiSe₅

monoclinic C2/c $\longrightarrow$ orthorhombic Cmcm



TD-THz

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



mid-IR ellipsometry

TI Larkin..., and AVB, PRB 95, 195144 (2017)

Conclusions

Comparative studies of Ta_2NiSe_5 and Ta_2NiS_5 , are required to confirm the EI hypothesis.

- In Ta_2NiSe_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_2NiS_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_2NiSe_5 but not in Ta_2NiS_5 .

