

Interplay of magnetism and band structure in EuCd_2As_2

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23.02.2023.

02.23.2023.

Low-energy degrees of freedom



Topological materials

2007: 2D topological insulators

M. König et al., *Science* 318, 766 (2007)

2008: 3D topological insulators

D. Hsieh et al., *Nature* 452, 970 (2008)

2012: 3D topological crystalline insulators

P. Dziawa et al., *Nature Mater.* 11, 1023 (2012)

2014: Symmetry-protected 3D Dirac semimetals

Z. K. Liu et al., *Science* 343, 864 (2014)

2015: 3D Weyl semimetals

B. Q. Lv et al., *Nature Phys.* 11, 724 (2015)

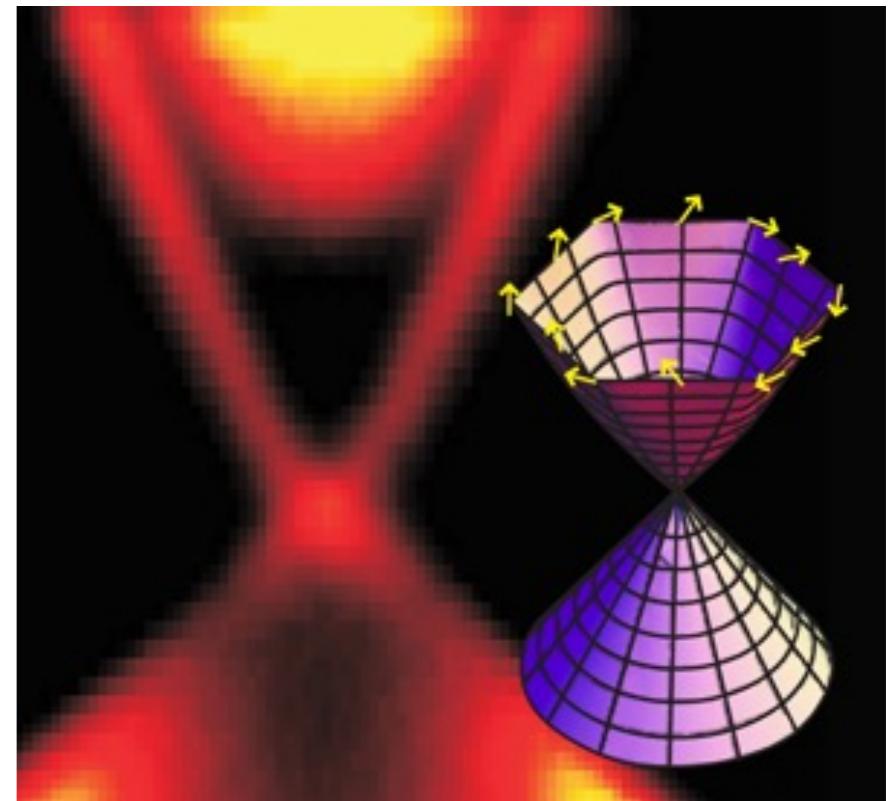
2016: Nodal line/loop Dirac semimetals

Wu, Y. et al., *Nature Phys.* 12, 667 (2016)

2019: Multifold massless electrons

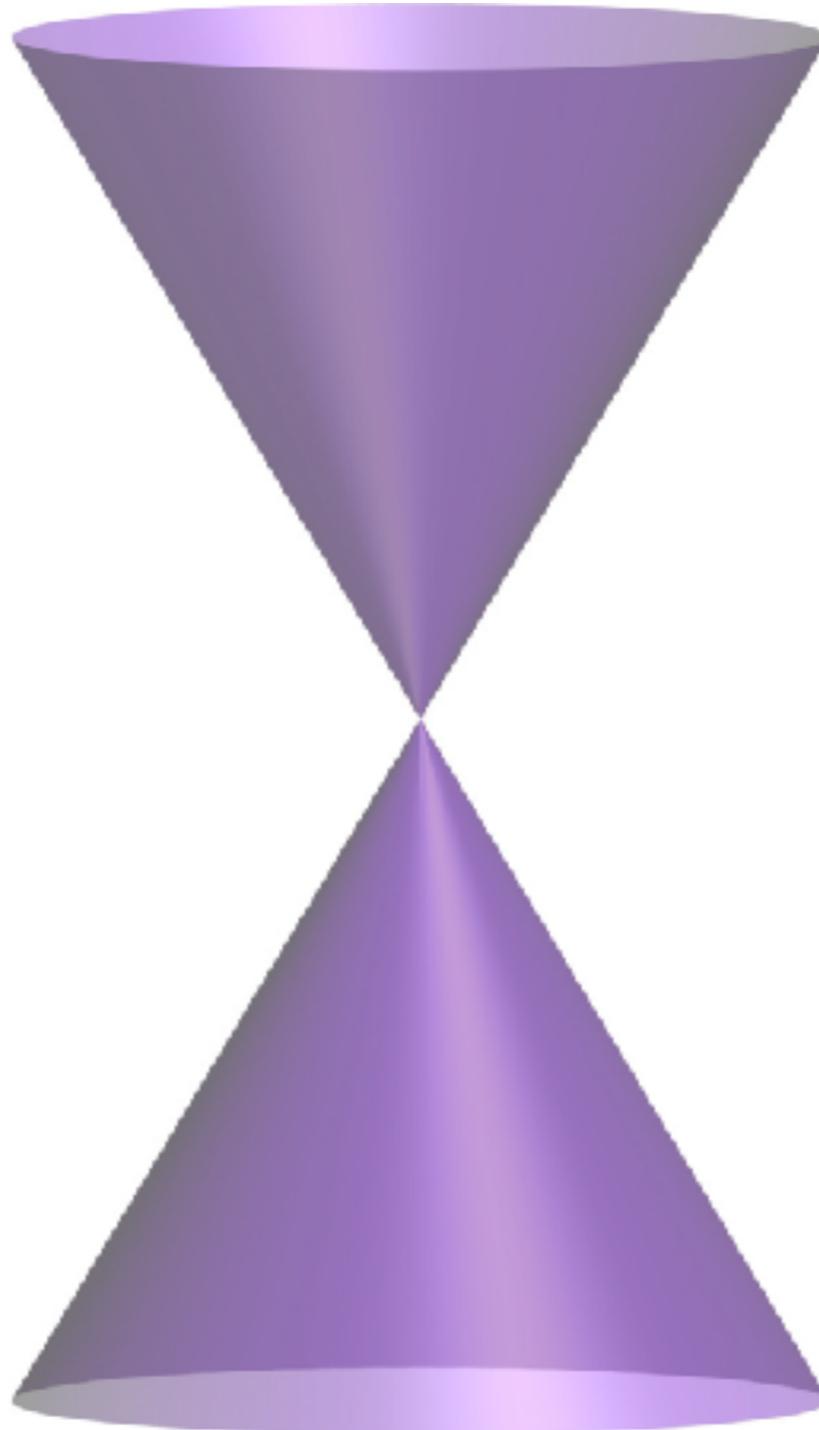
D. S. Sanchez et al., *Nature* 567, 501 (2019)

2023: ...



M. Z. Hasan and C. L. Kane, *RMP* 82, 3045 (2010)

Topological semimetals



conical bands

small energy scales

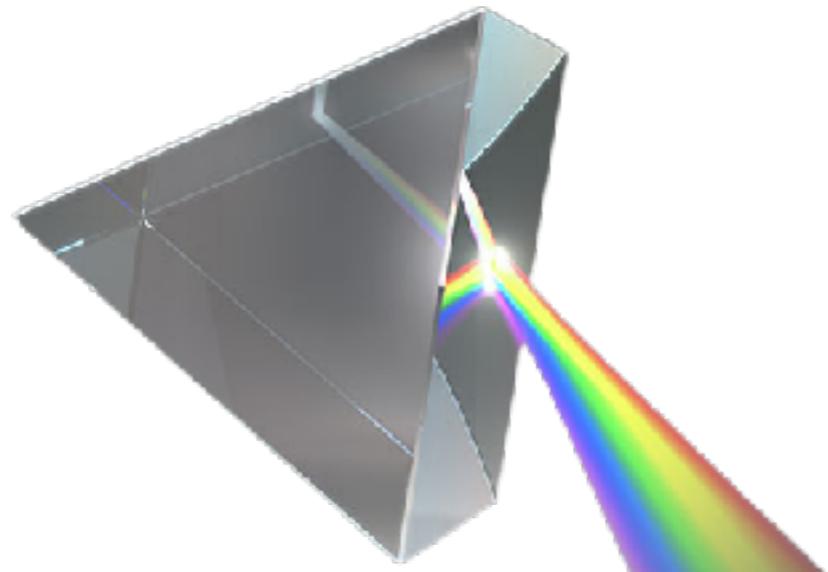
can be tuned
externally

exotic quasiparticles:
eg Weyl fermions

can be coupled
to magnetism

correlations

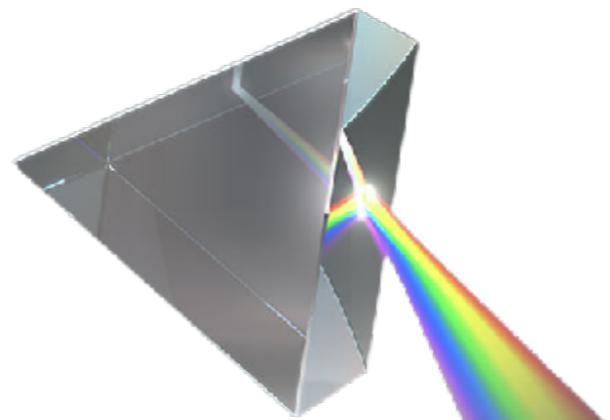
Our techniques



infrared spectroscopy



high-pressure
IR spectroscopy



magneto-optics

Infrared spectroscopy

Volume I. July-August, 1893. Number 1.

THE PHYSICAL REVIEW.

A STUDY OF THE TRANSMISSION SPECTRA OF
CERTAIN SUBSTANCES IN THE INFRA-RED.

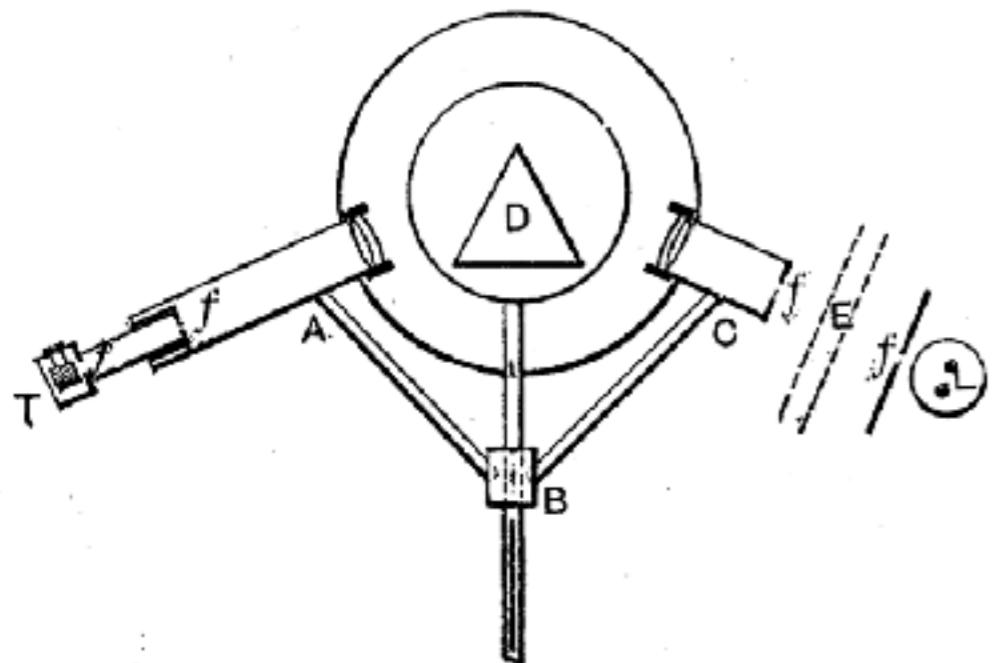


Fig. 1.

Infrared spectroscopy

Volume I. July-August, 1893. Number 1.

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A STUDY OF THE TRANSMISSION SPECTRA OF CERTAIN SUBSTANCES IN THE INFRA-RED.

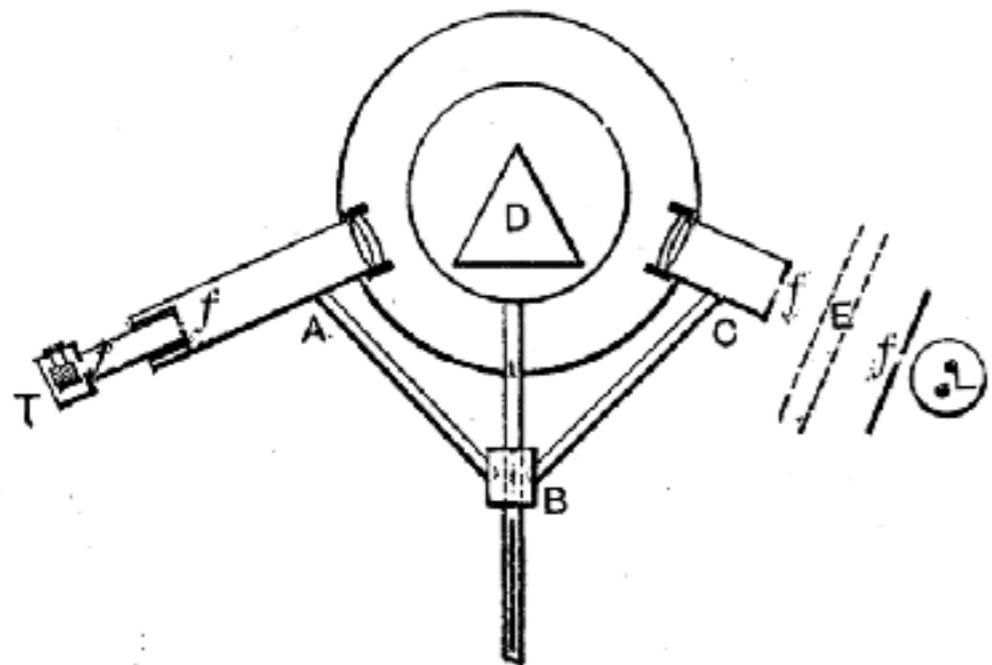
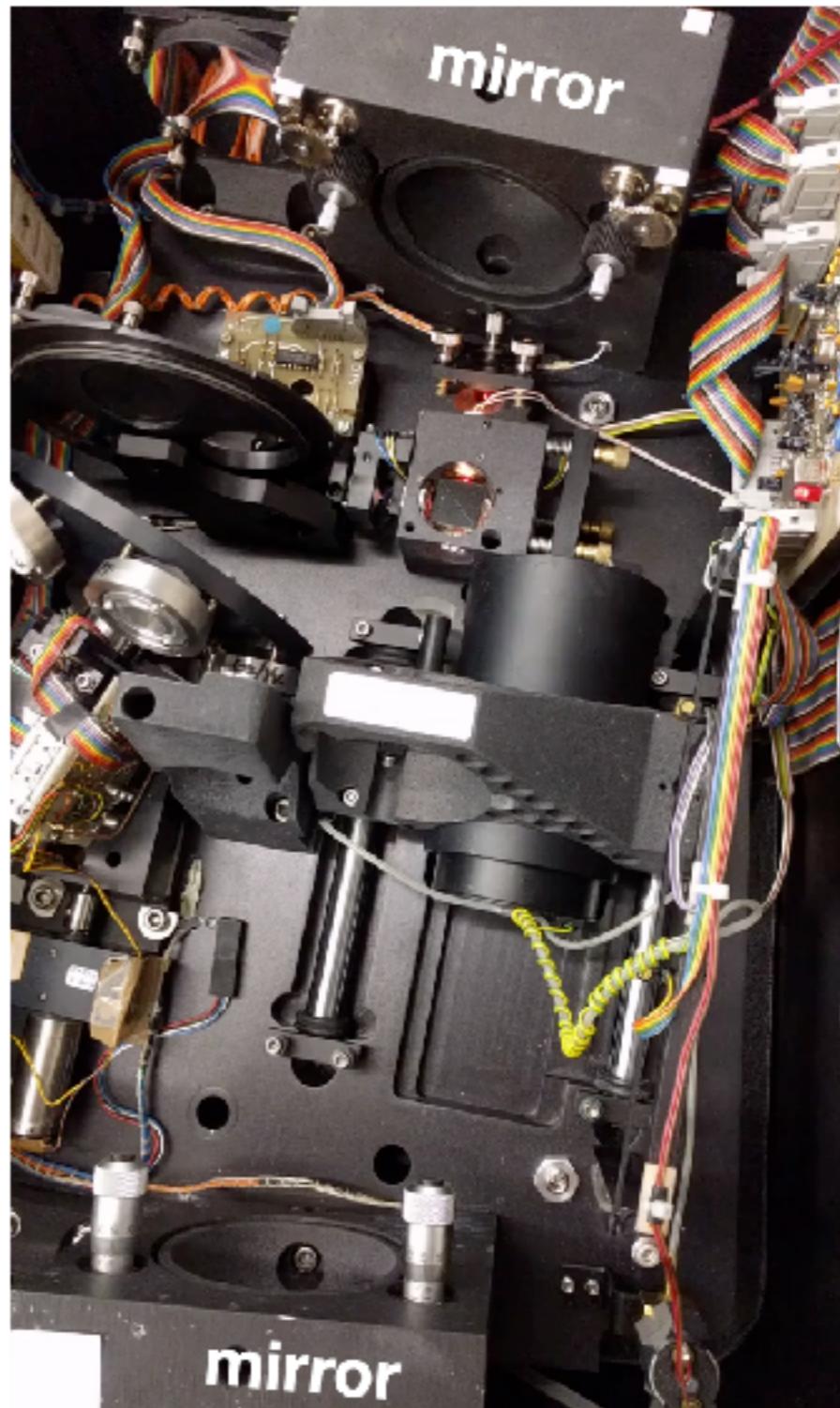
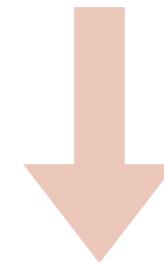


Fig. 1.

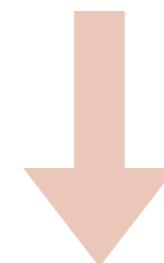


Infrared spectroscopy

$$R = \left| \frac{E_R}{E_0} \right|^2 = \left| \frac{1 - \sqrt{\varepsilon}}{1 + \sqrt{\varepsilon}} \right|^2$$



Kramers-Kronig relations



Complex optical conductivity

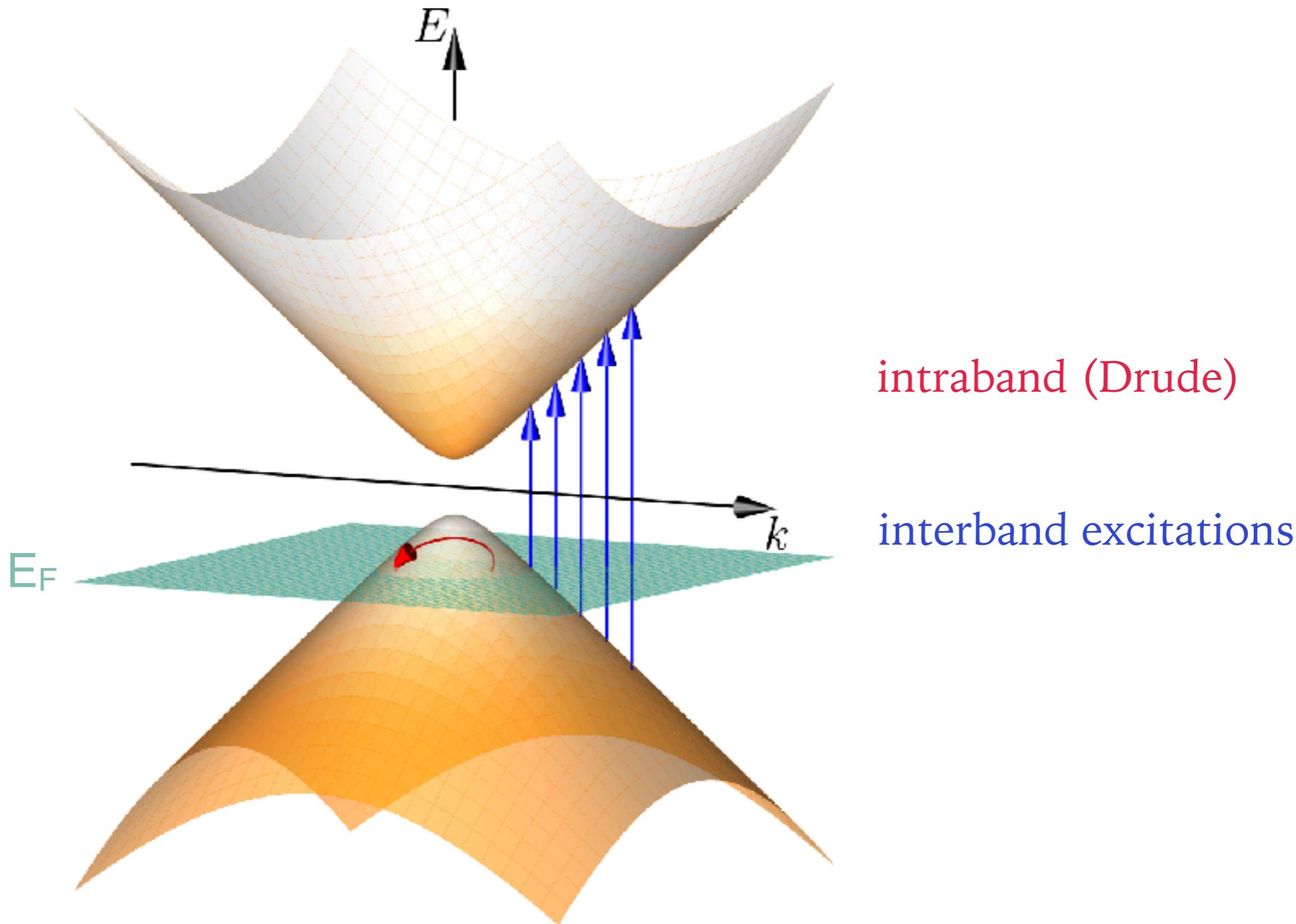
$$\sigma(\omega) = \sigma_1(\omega) + i\sigma_2(\omega)$$

$I(\omega)$

$R(\omega)$

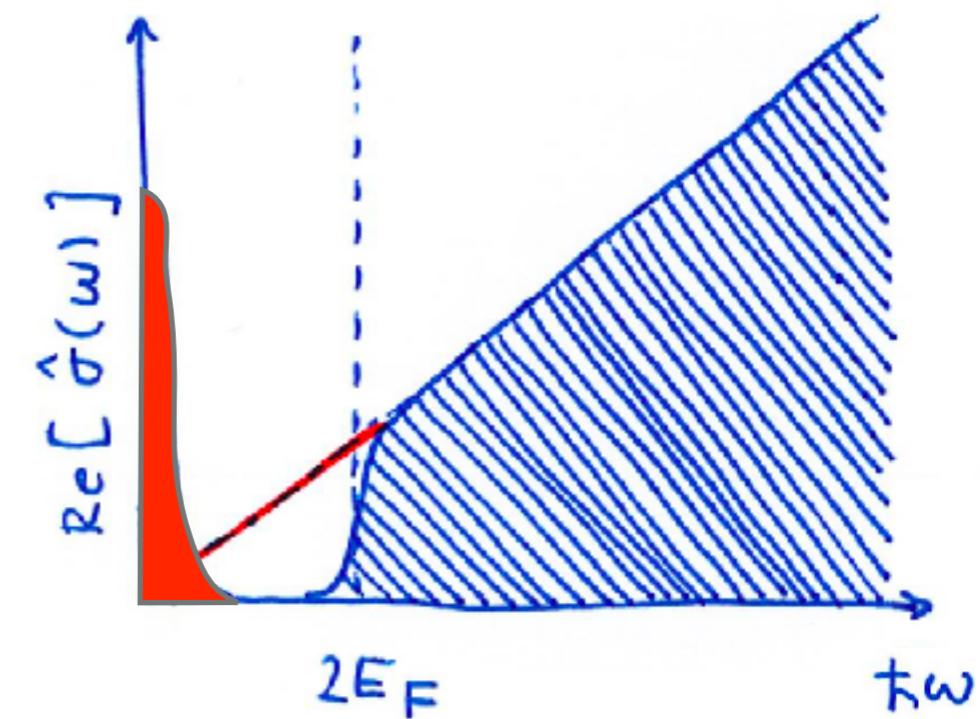
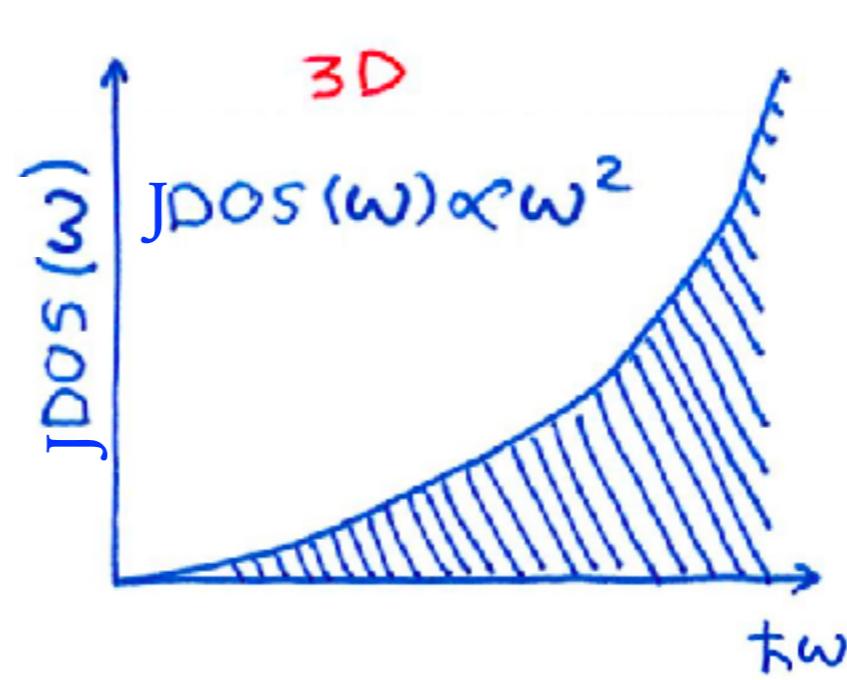
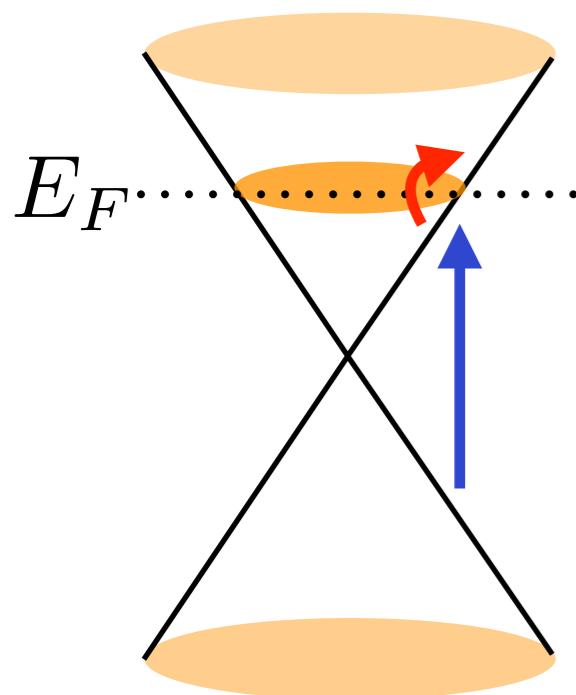


Optical spectroscopy tells about jDOS



Optical conductivity tells about jDOS

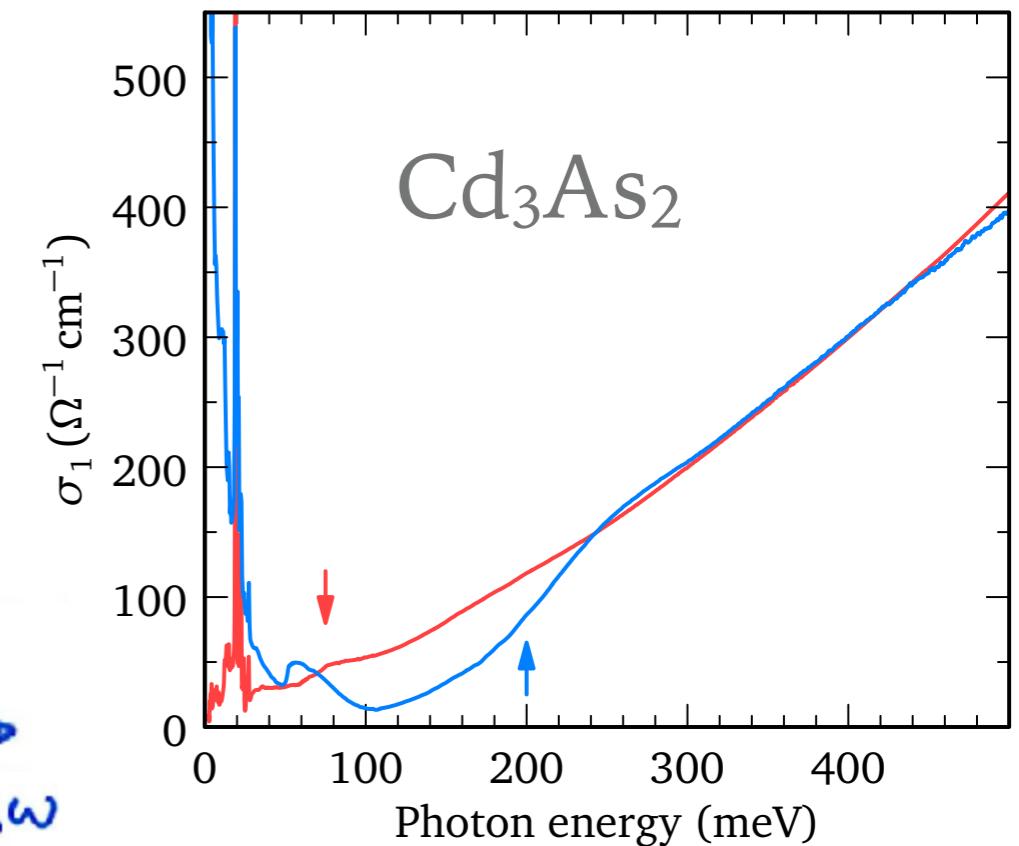
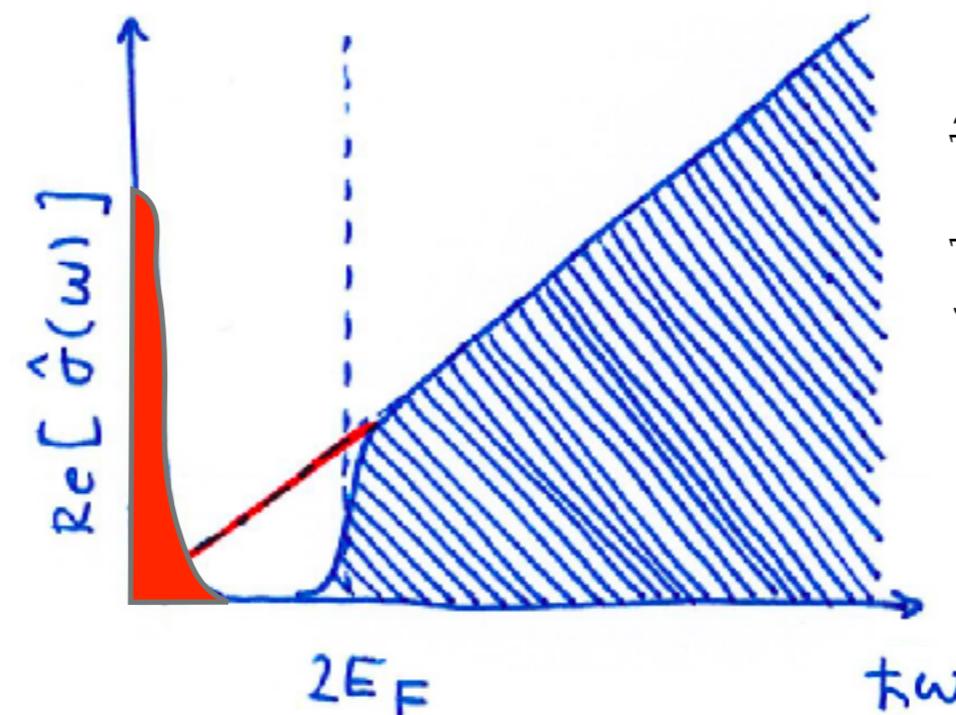
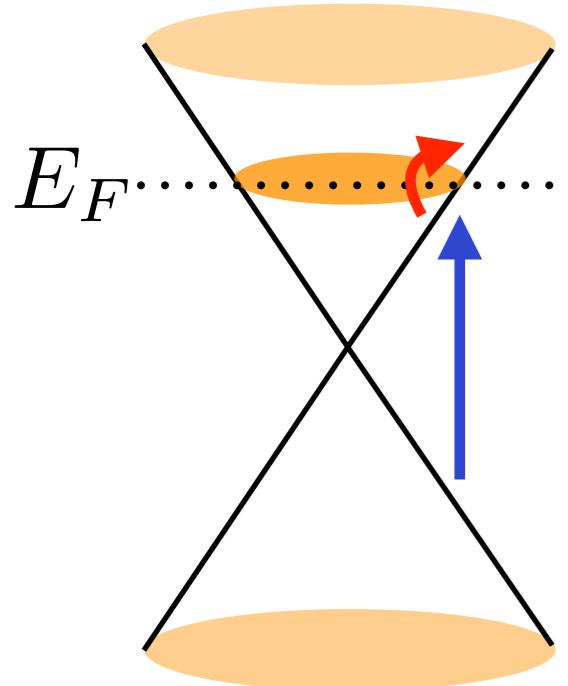
3D linear bands



$$\sigma_1(\omega) \propto \frac{jDOS}{\omega}$$

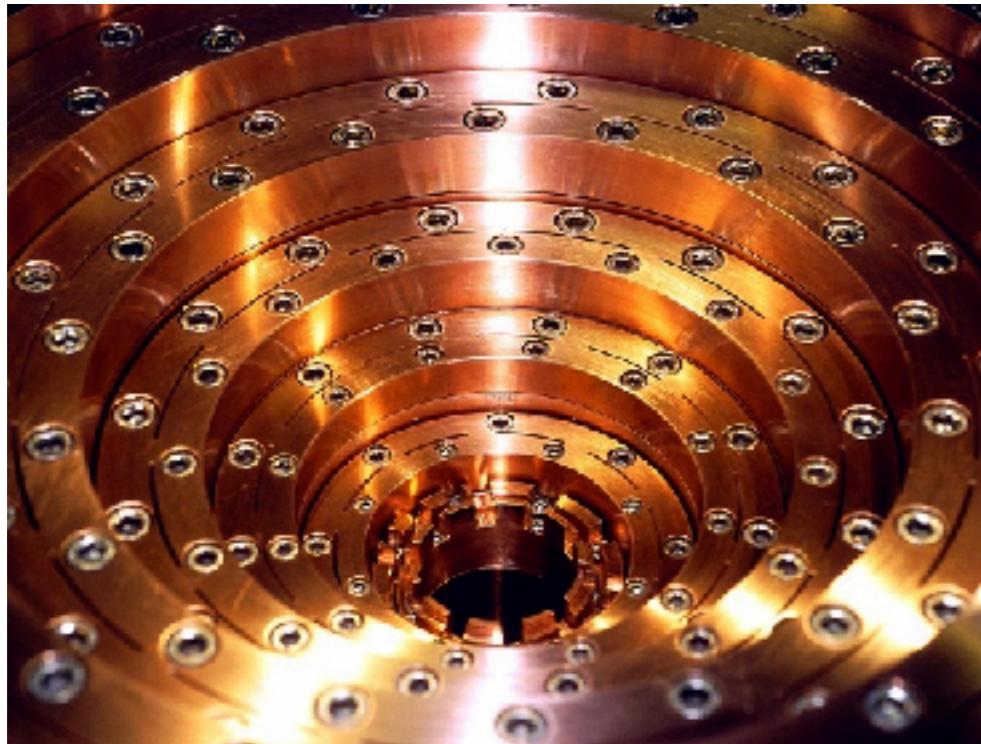
Optical conductivity tells about jDOS

3D linear bands



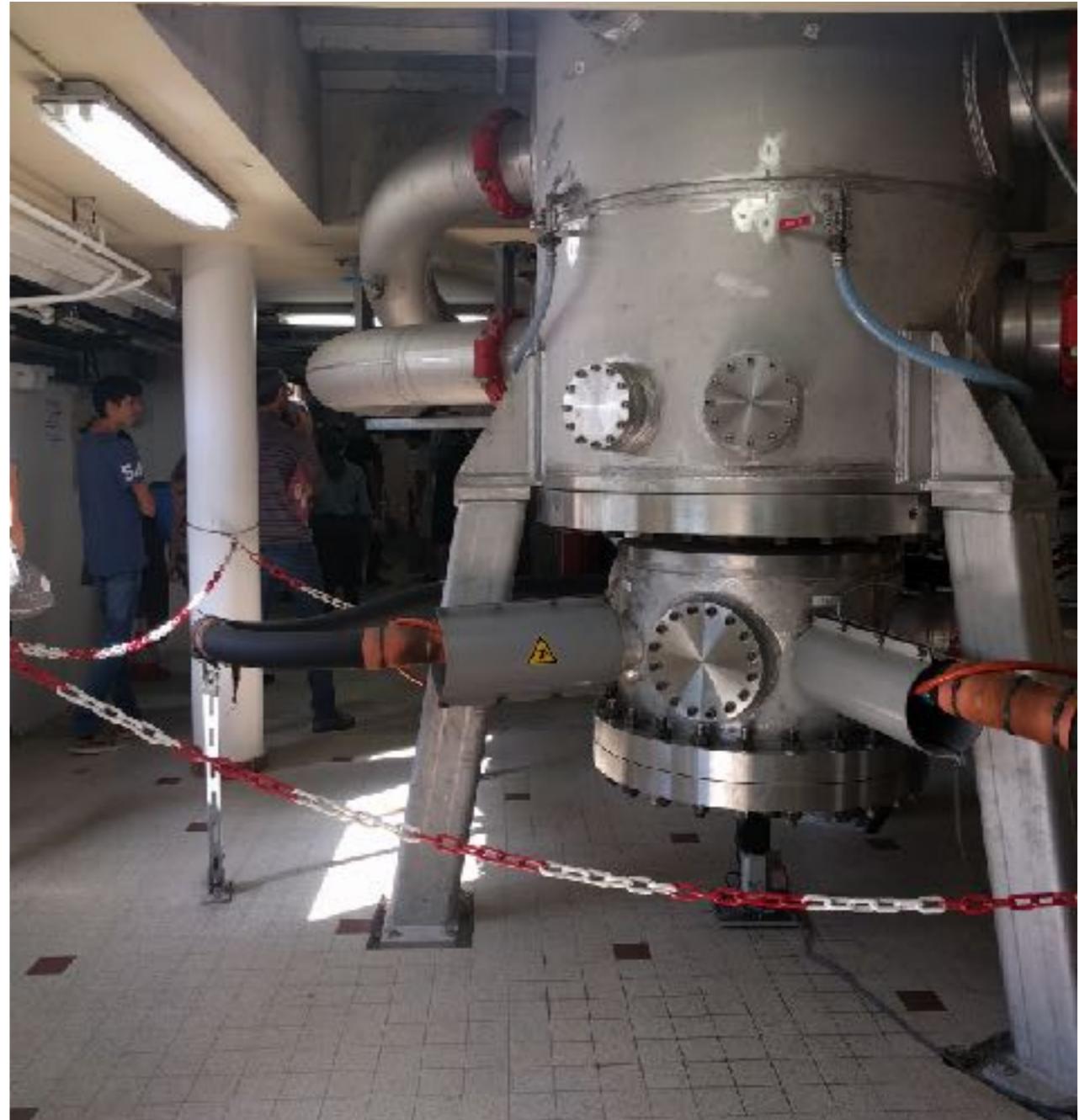
$$\sigma_1(\omega) \propto \frac{jDOS}{\omega}$$

Also in high magnetic fields

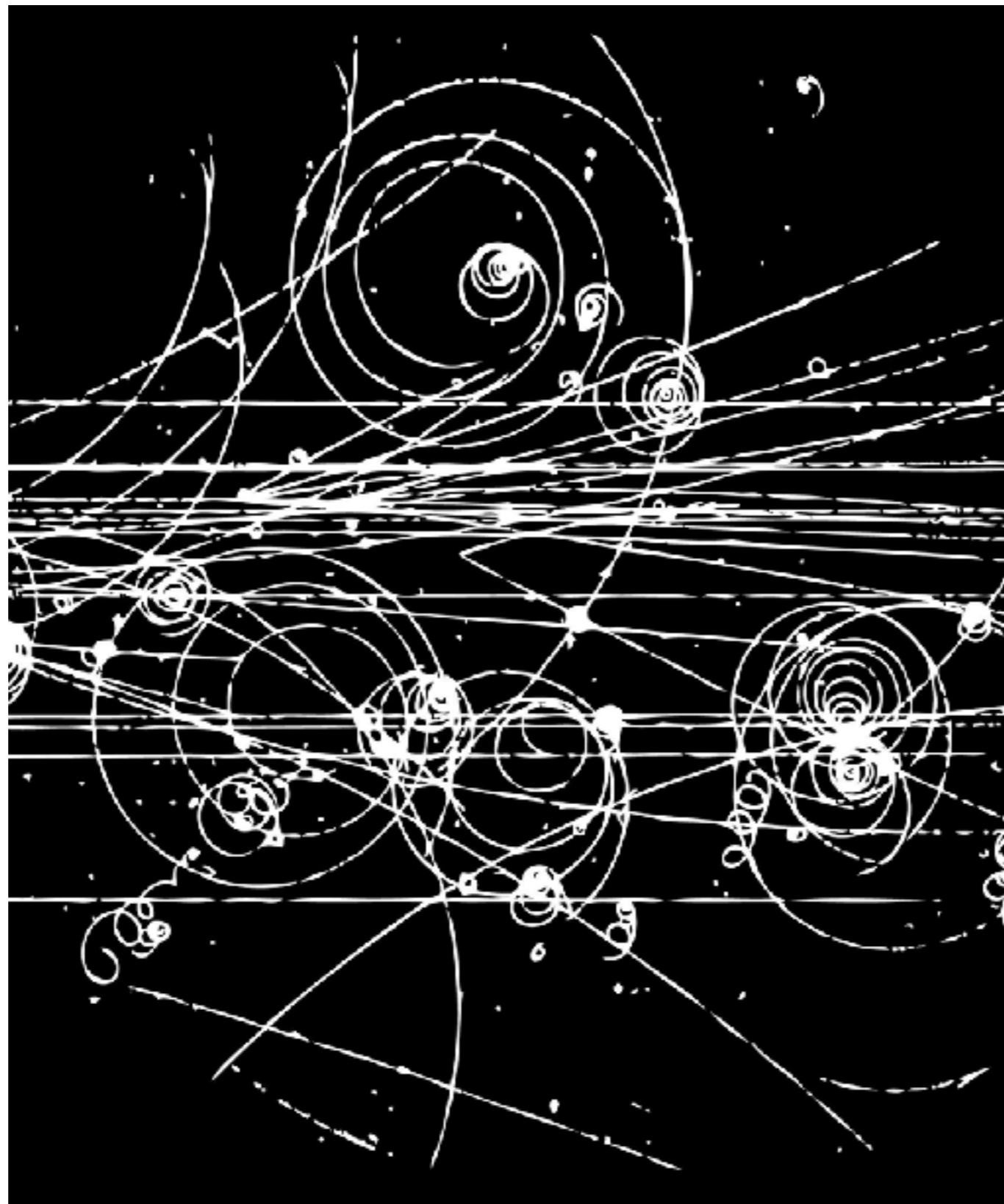


14'000 amps through
4 copper alloy coils

25 MW for 36 T
static magnetic field



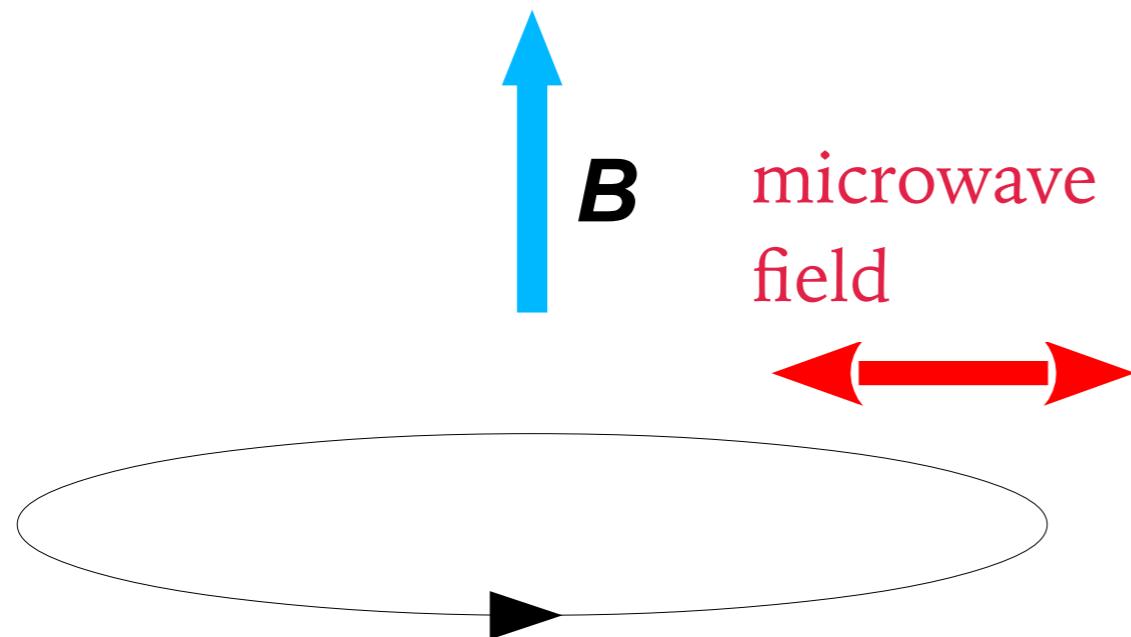
Cyclotron motion



CERN bubble chamber (1960)

Cyclotron resonance

Classical picture

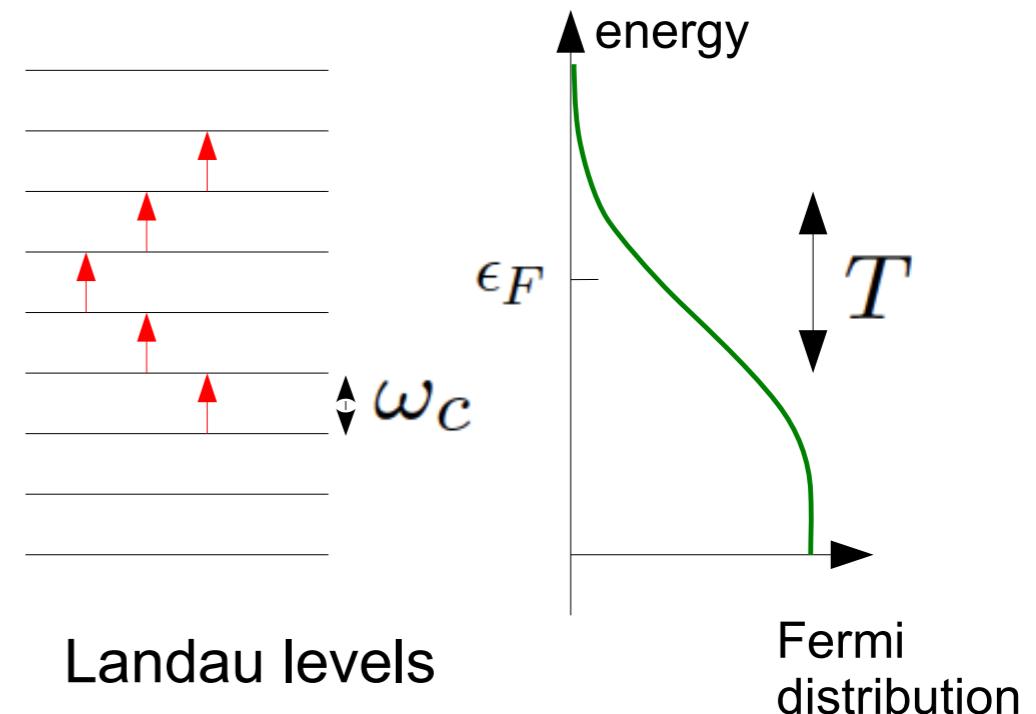


$$\frac{d\mathbf{p}}{dt} = \frac{e}{c} [\mathbf{v} \times \mathbf{B}] + \mathbf{F}_0 \cos \omega t$$

Cyclotron motion
with the frequency ω_c

Resonance
when $\omega = \omega_c$

Quantum picture



Landau levels

Transitions
between Landau
levels

Cyclotron resonance = resonant absorption of light at the cyclotron frequency

Observation of Cyclotron Resonance in Germanium Crystals*

an the simple magnetron. The angular rotation frequency in a crystal is

$$\omega_L = (eH)/(m^*c), \quad (1)$$

where m^* is the appropriate effective mass; thus the experiment determines the effective mass directly.



G. Dresselhaus, A.F. Kip, C. Kittel, Phys. Rev. 92, 827 (1953).

M.L. Cohen, AIP Conference Proceedings 772, 3 (2005).

Observation of Cyclotron Resonance in Germanium Crystals*

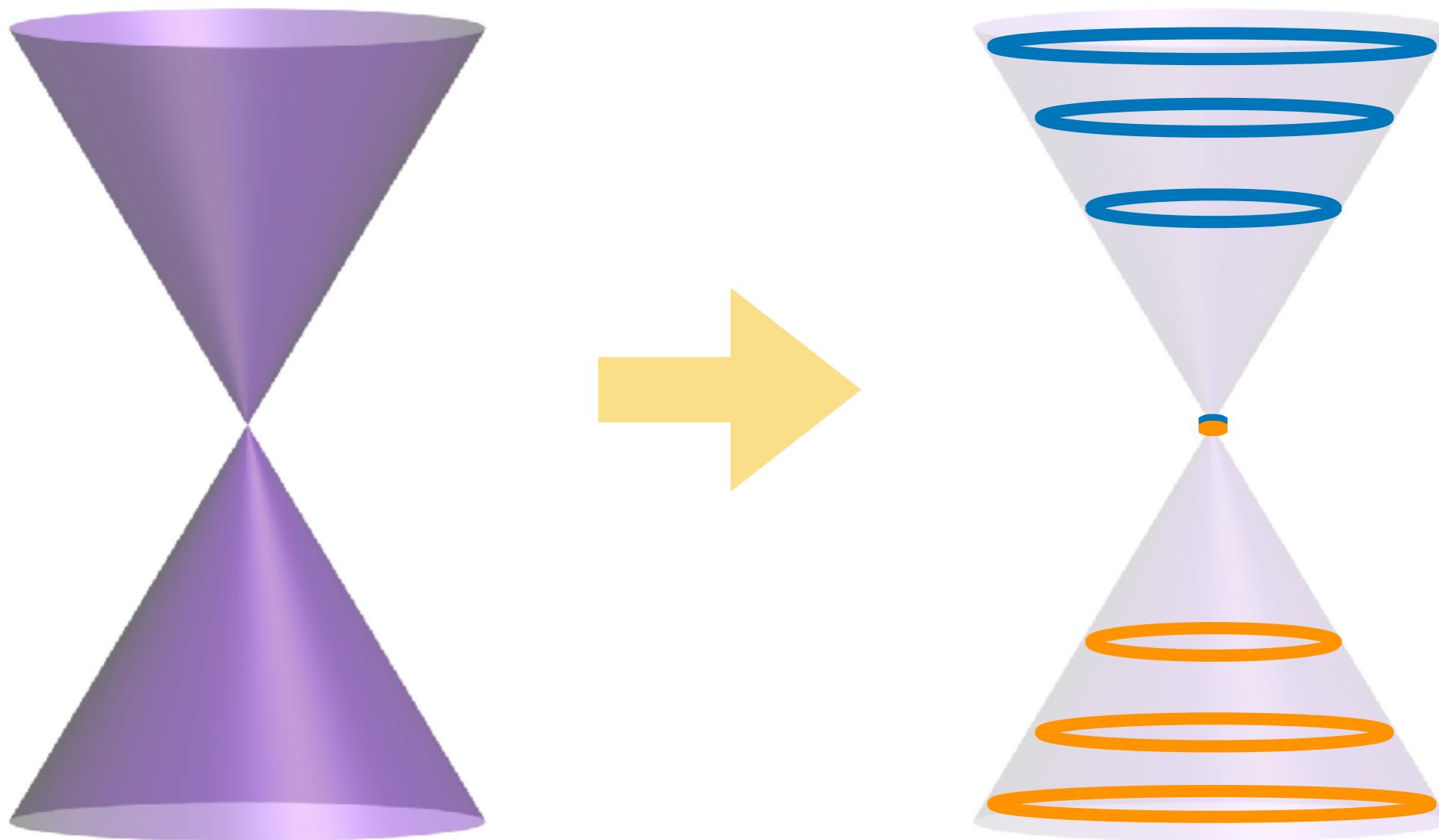
"The first experimental observation of cyclotron resonance in Ge was very exciting. A.F. Kip was at the controls of the spectrometer while Kittel and I, along with other members of the research group, were offering lots of advice and encouragement in the background.



G. Dresselhaus, A.F. Kip, C. Kittel, Phys. Rev. 92, 827 (1953).

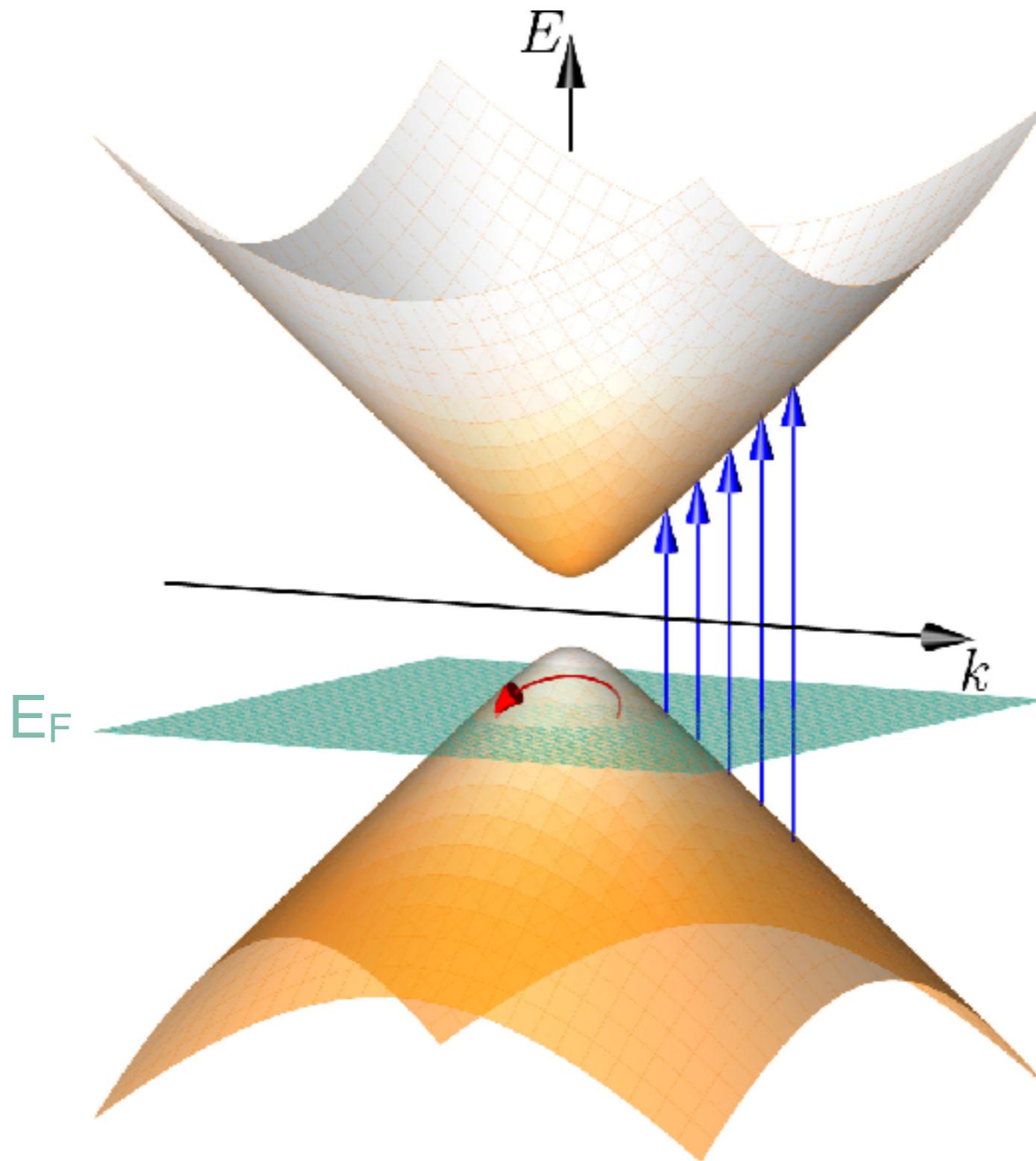
M.L. Cohen, AIP Conference Proceedings 772, 3 (2005).

Nonequidistant Landau levels



The nature of interband transitions is preserved.
Precise access to very low energy features.

Optical spectroscopy tells about jDOS



intraband (Drude) —
Cyclotron resonance

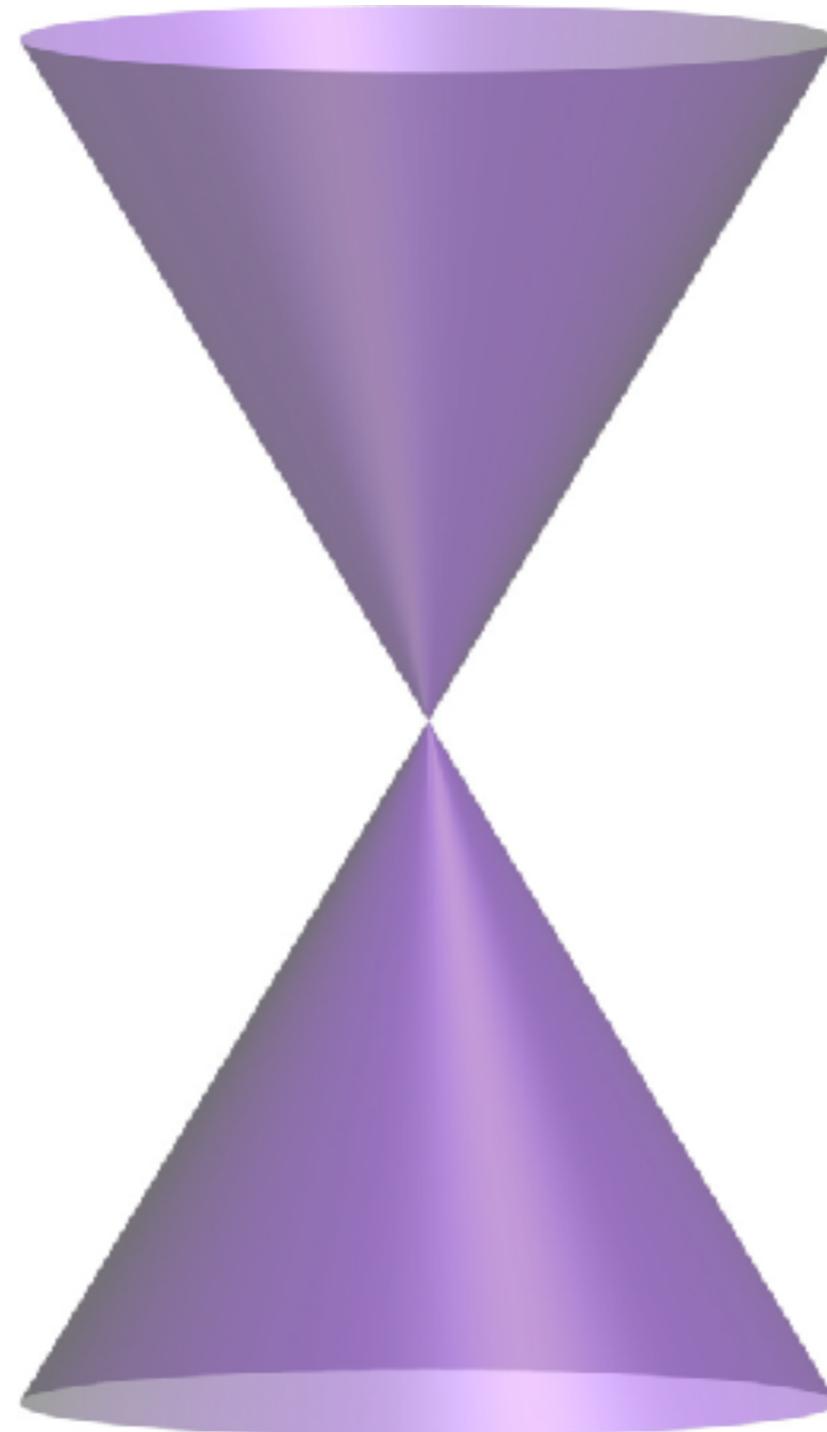
interband excitations —
interband inter-Landau level
transitions

Topological semimetals

conical bands

small energy scales

can be tuned
externally

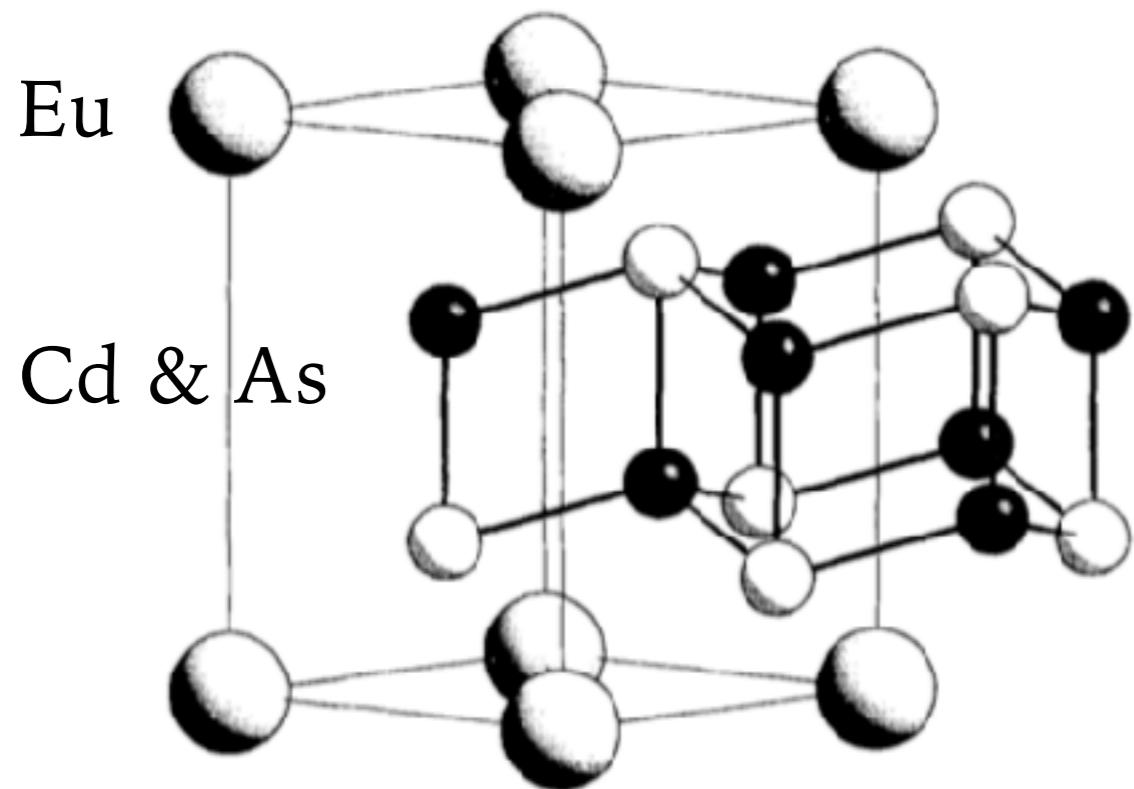


exotic quasiparticles:
eg Weyl fermions

can be coupled
to magnetism

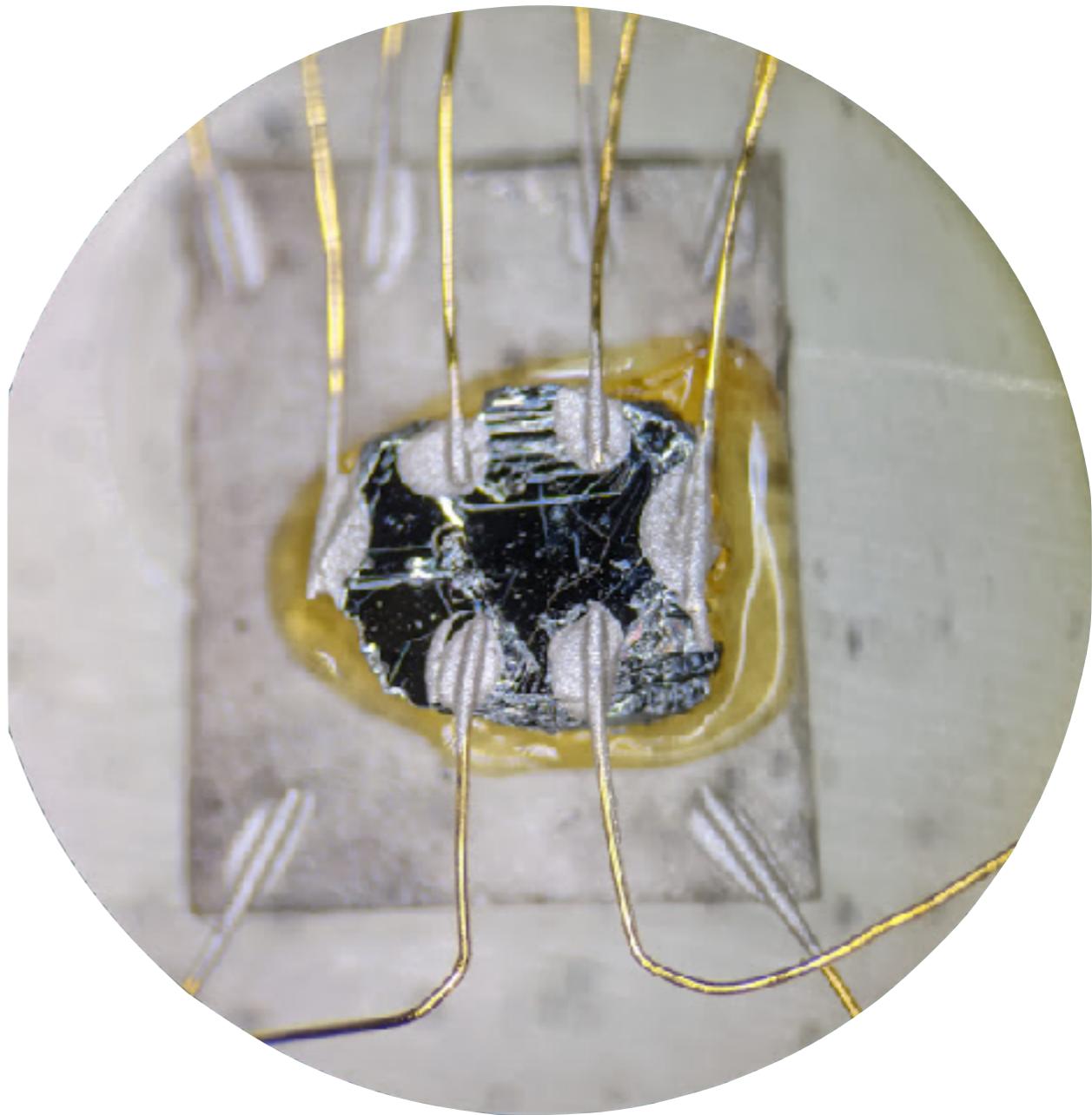
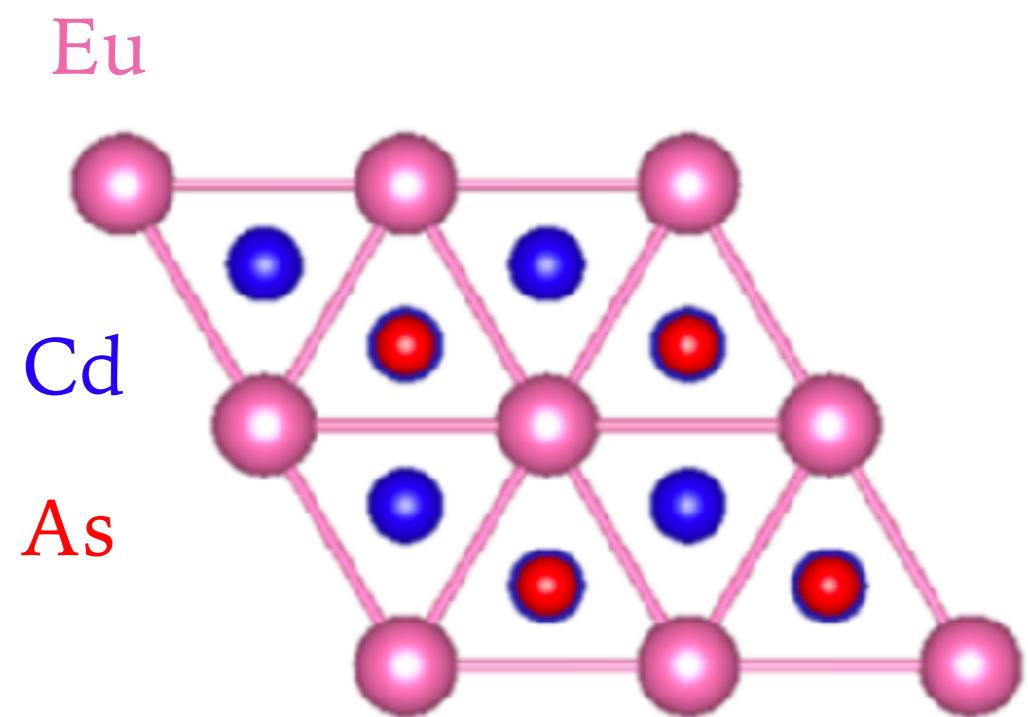
correlations

Topological semimetal EuCd_2As_2

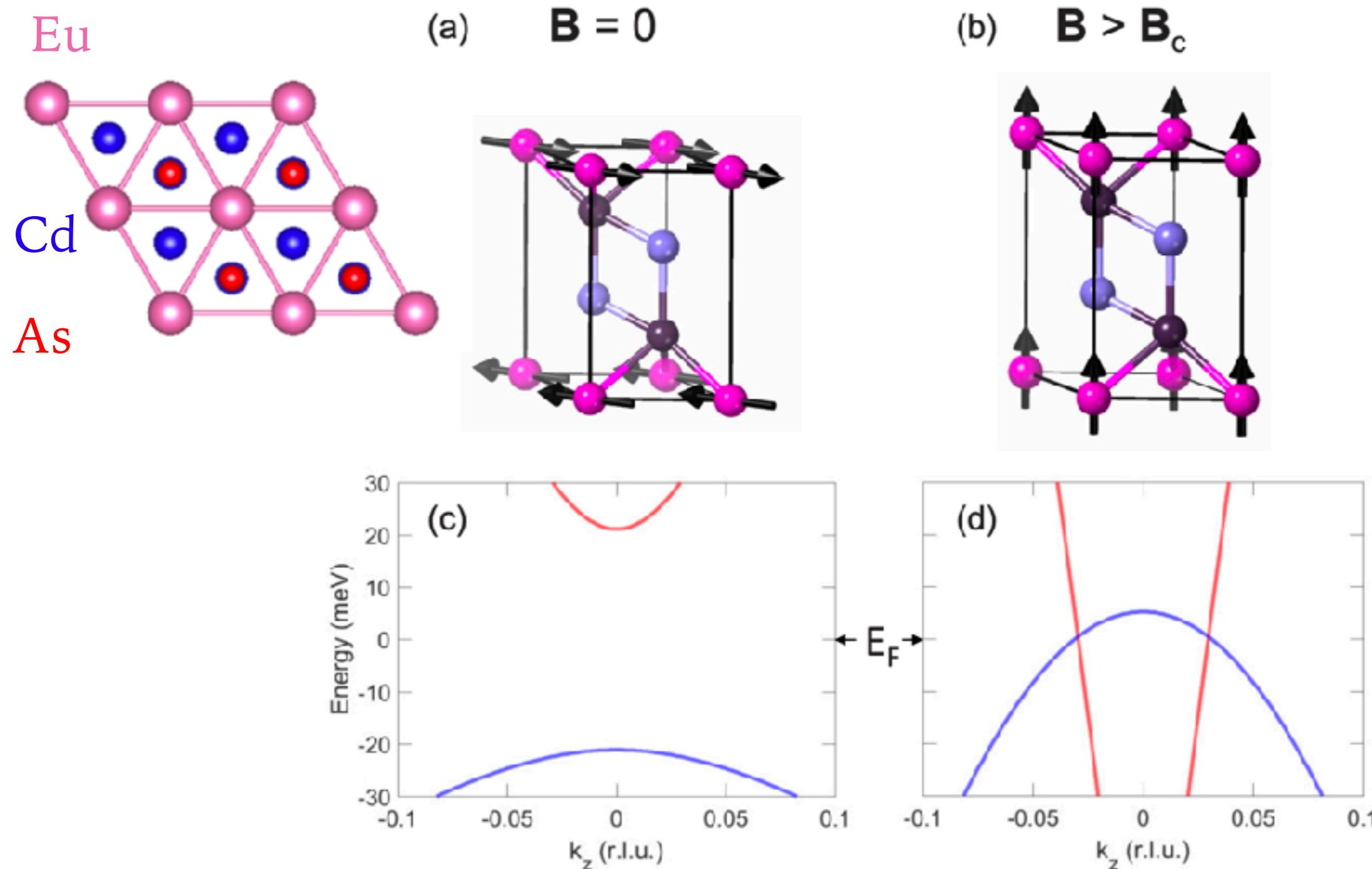


- Eu valence is $2+$
spin state $S=7/2$
- DFT says semimetal
($U = 5$ eV, by hand)

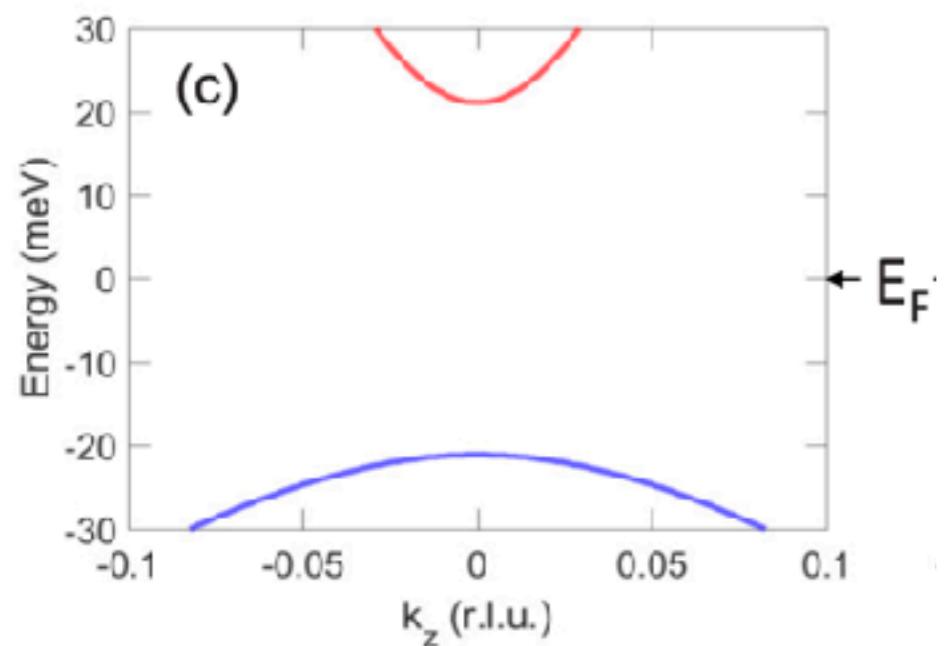
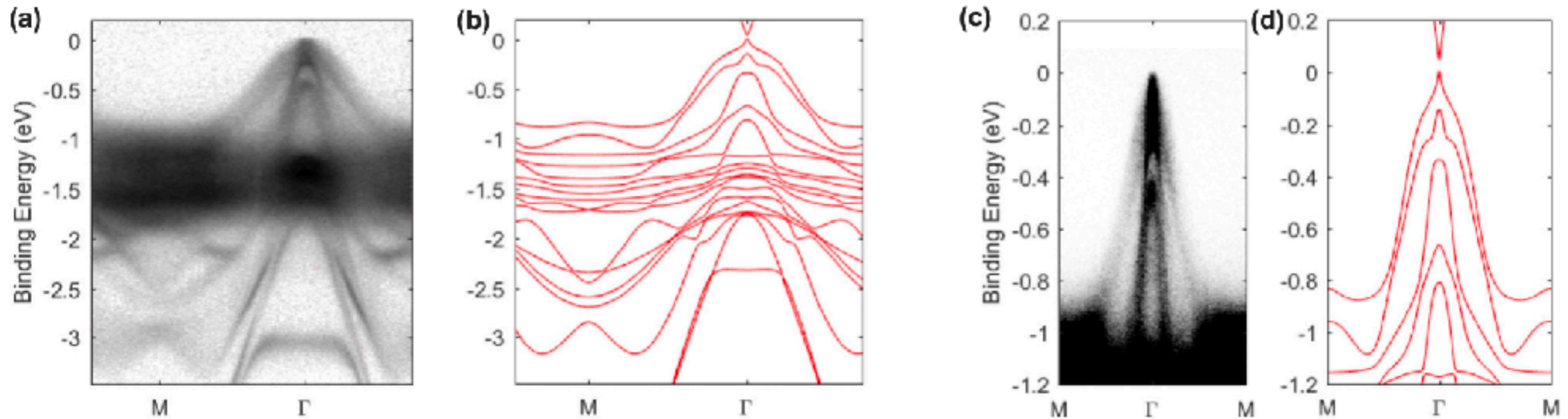
Topological semimetal EuCd_2As_2



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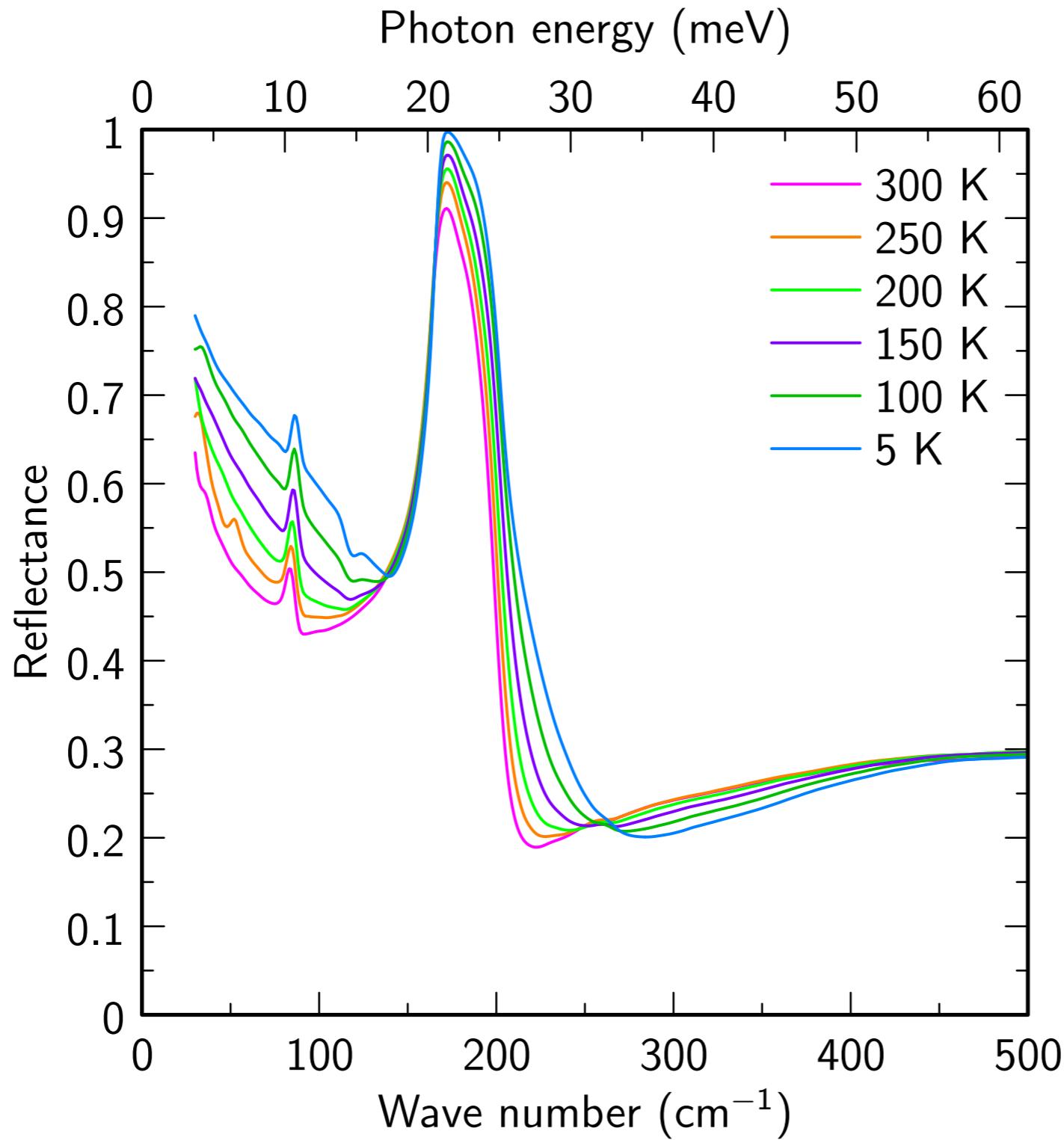


Topological semimetal EuCd_2As_2

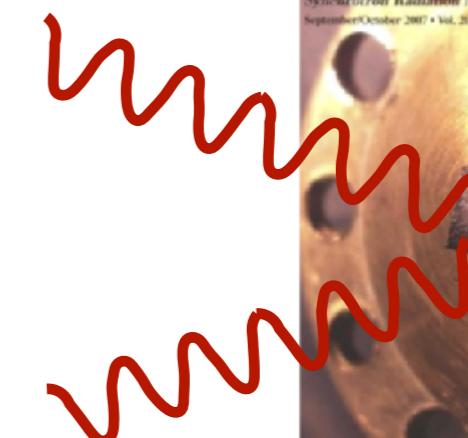




Surprise



$I(\omega)$



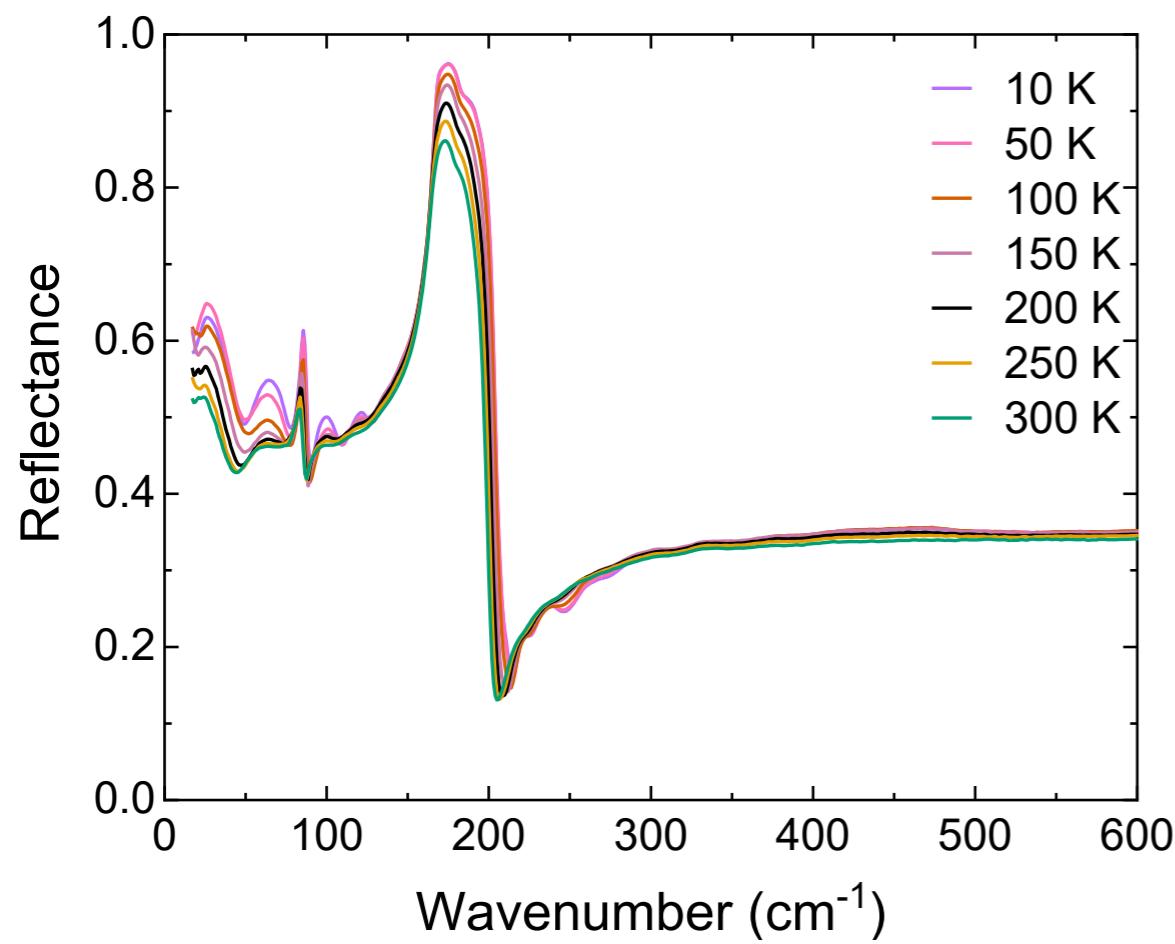
$R(\omega)$



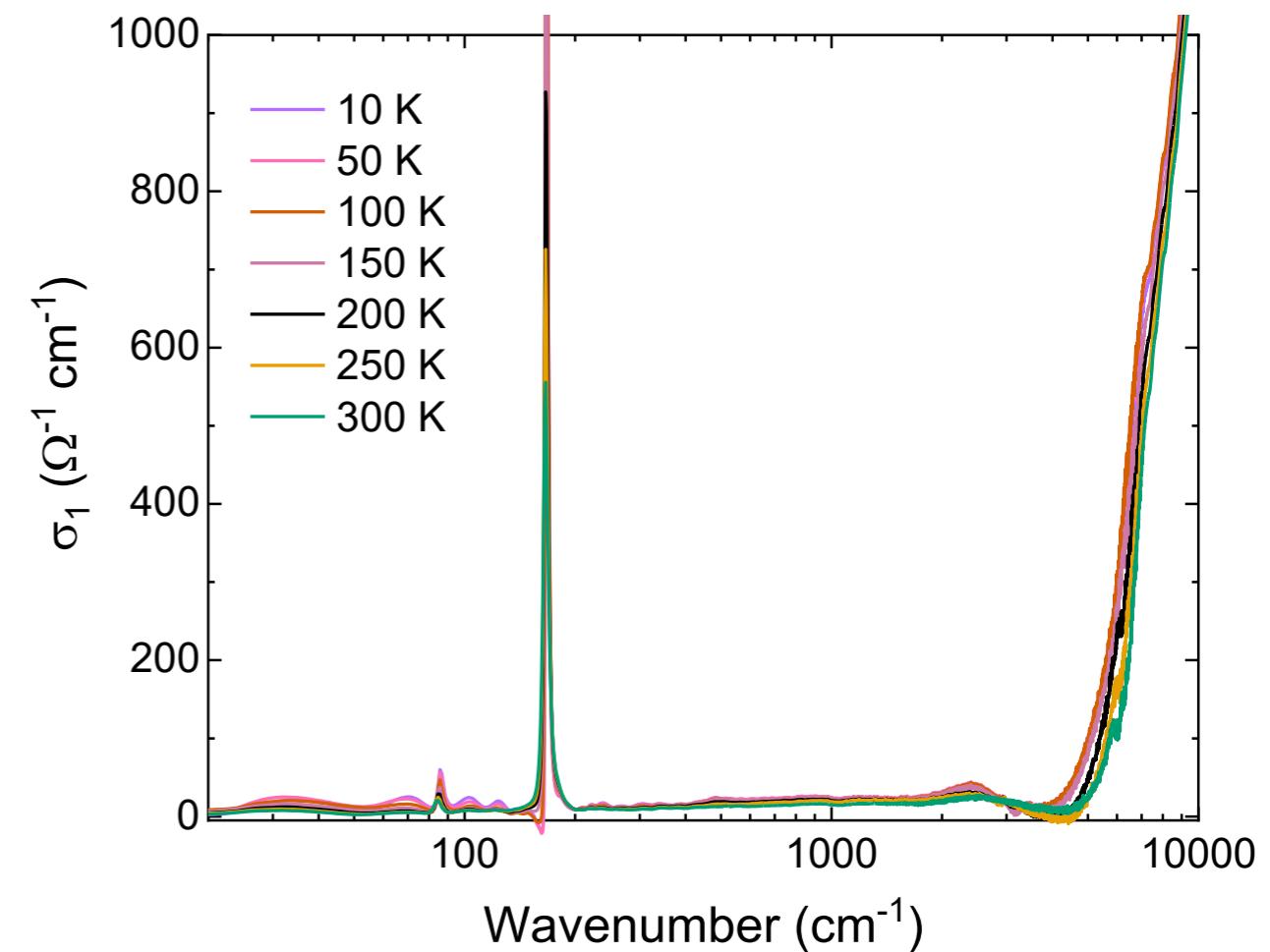
- weakly screened phonon modes
- low reflectance

Is this really a semimetal?

Reflectance

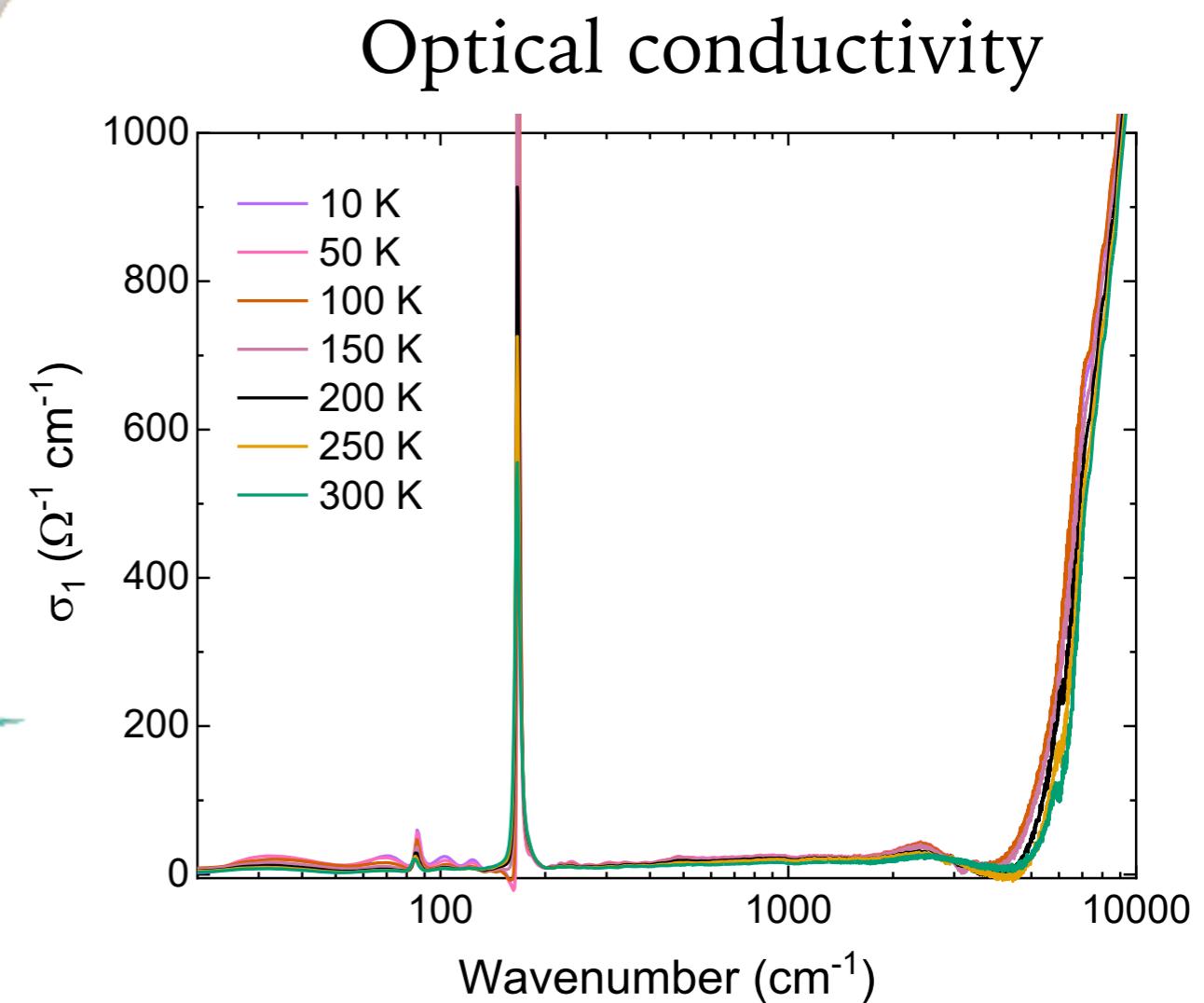
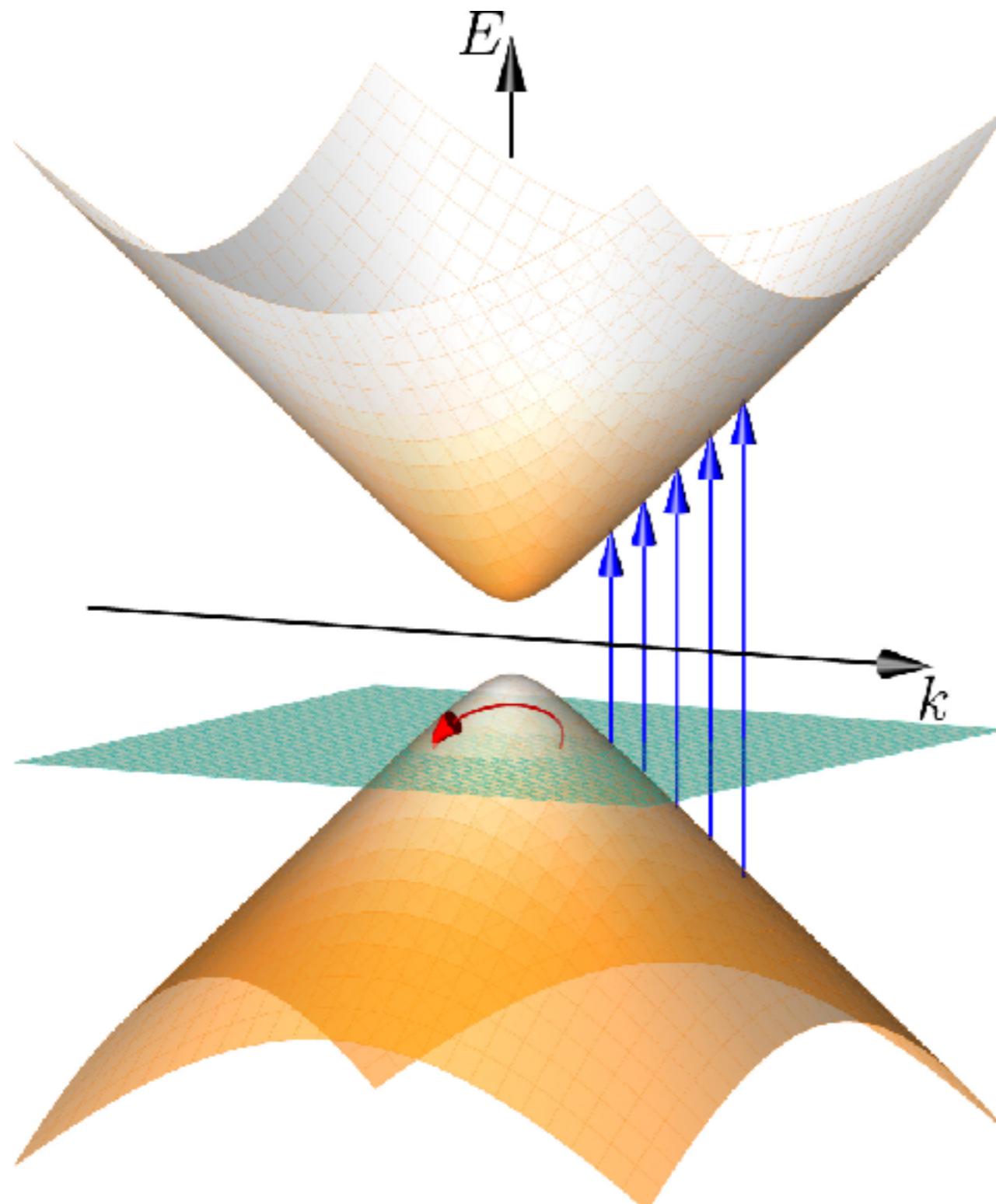


Optical conductivity



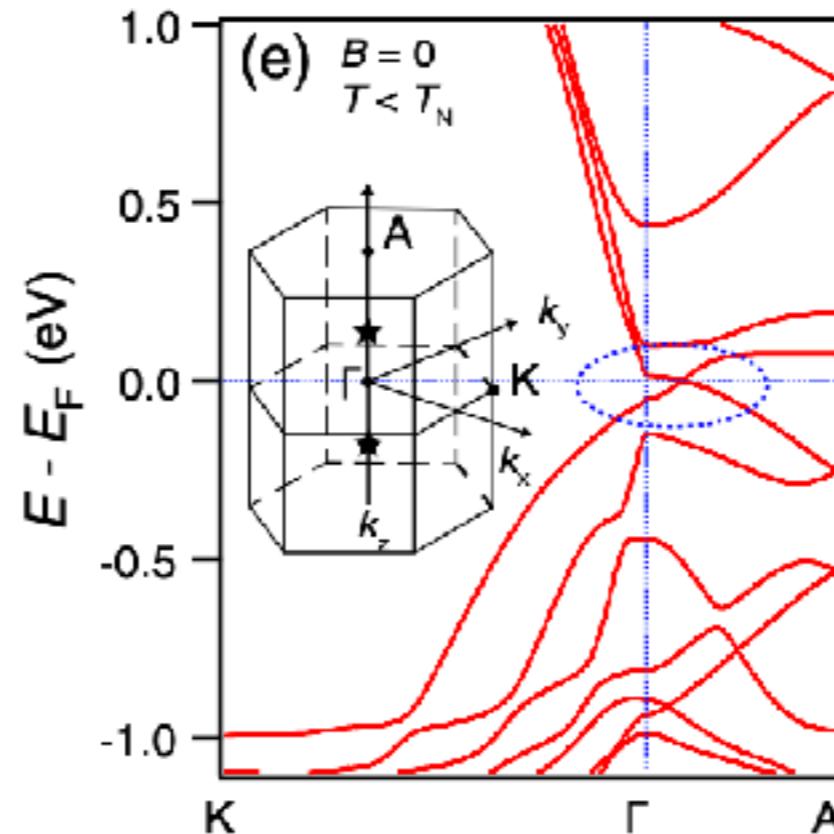
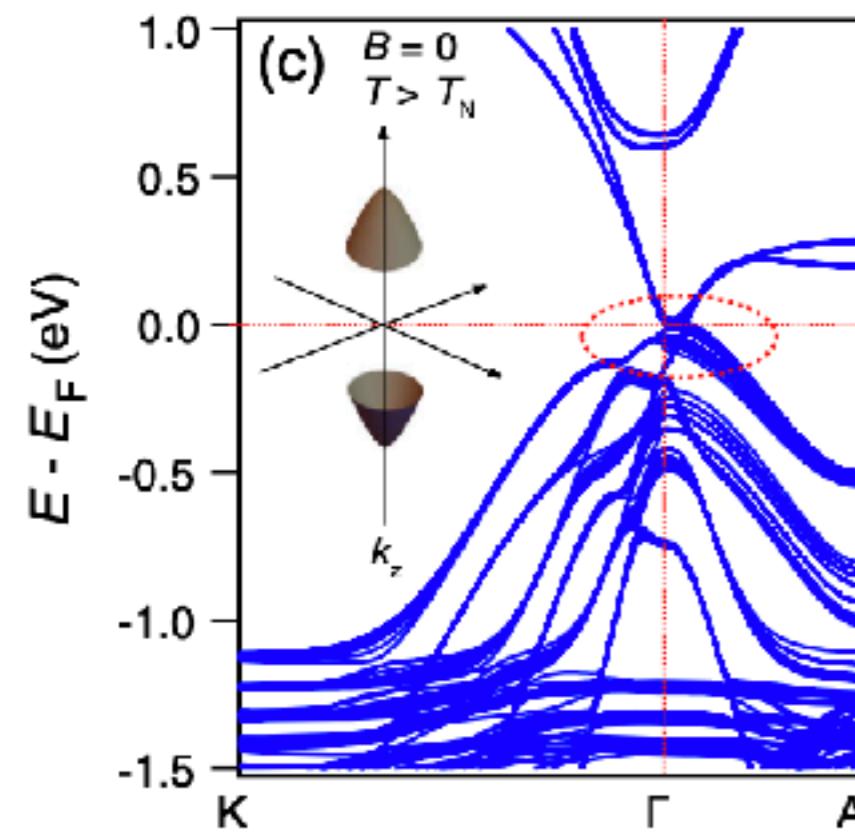
- weak temperature dependence of R
- weak Drude contribution
- gap-like feature above 0.5 eV

Is this really a semimetal?



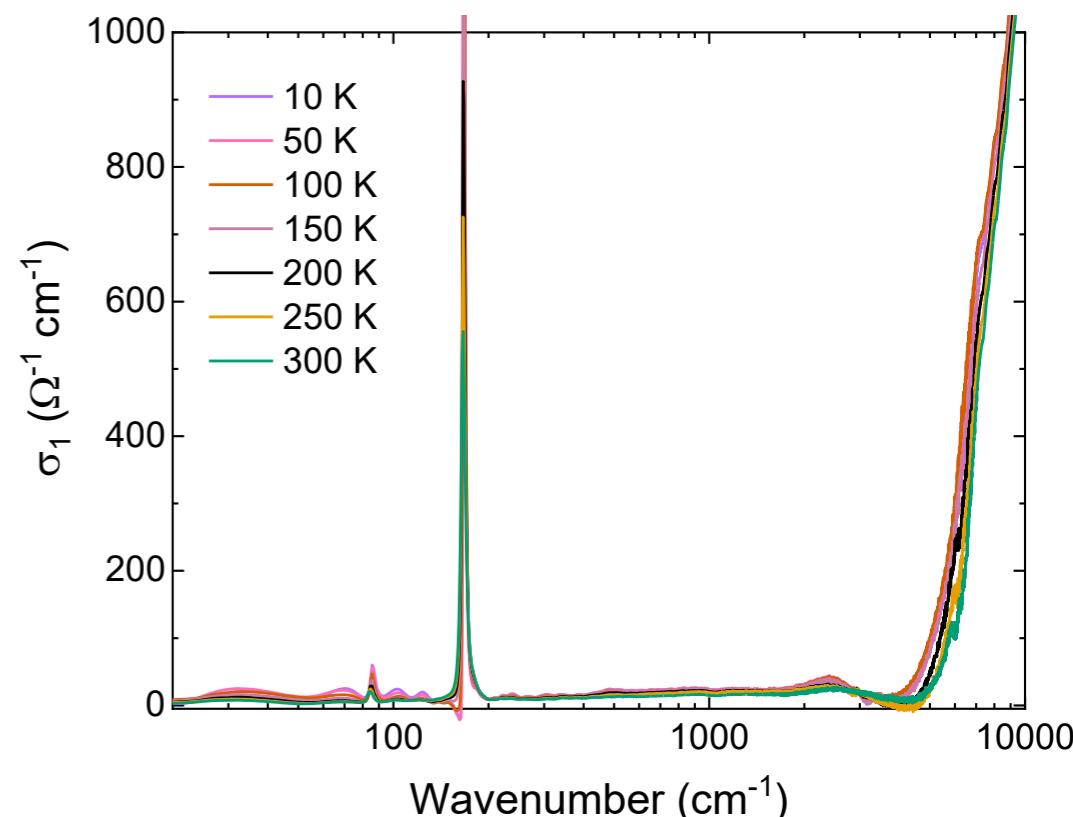
- weak Drude contribution
- gap-like feature above 0.5 eV

How to understand this?



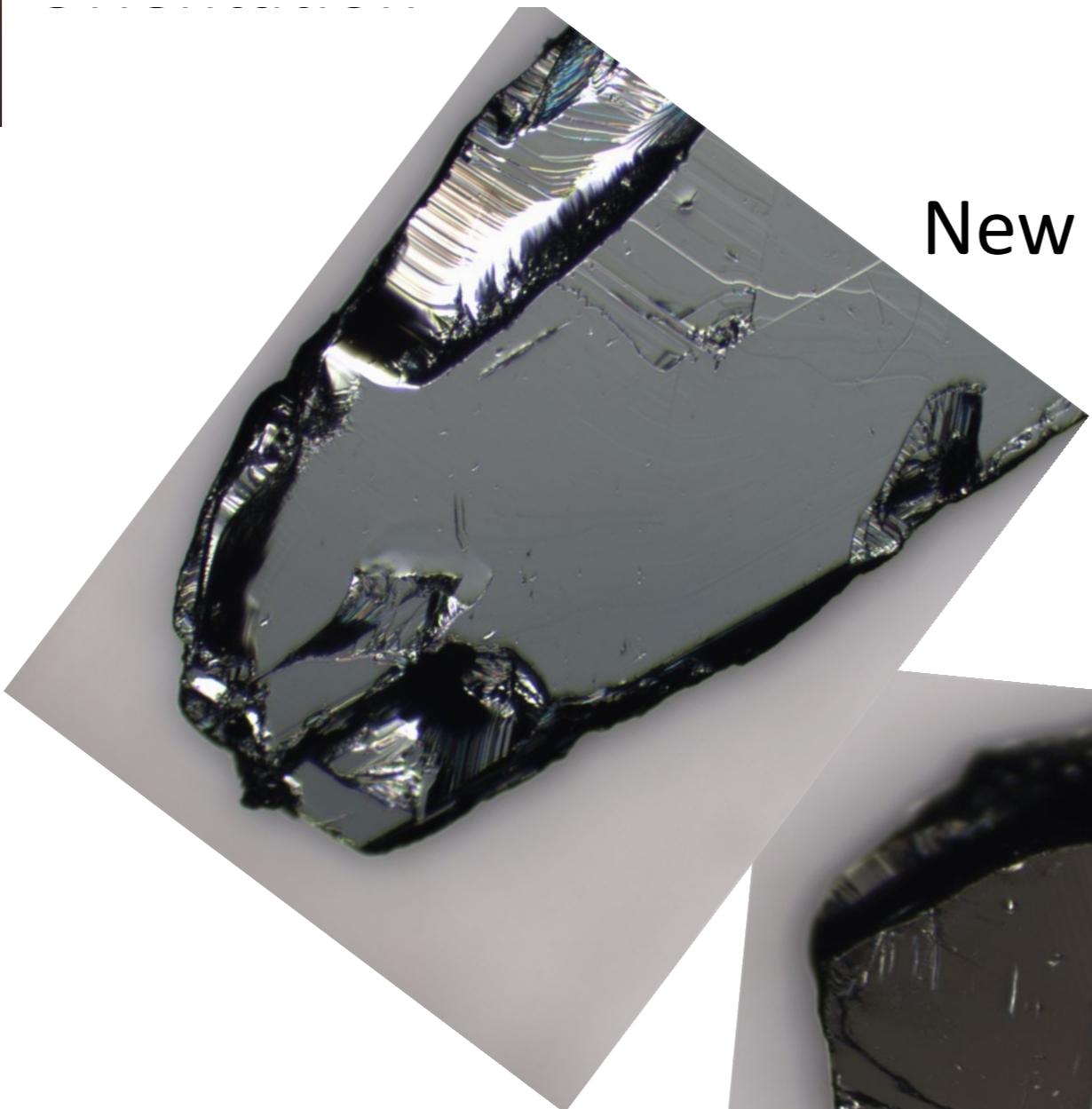
DFT:
Small or zero gap

Optical conductivity:
No interband transitions
below 0.5 eV
Weird selection rule?

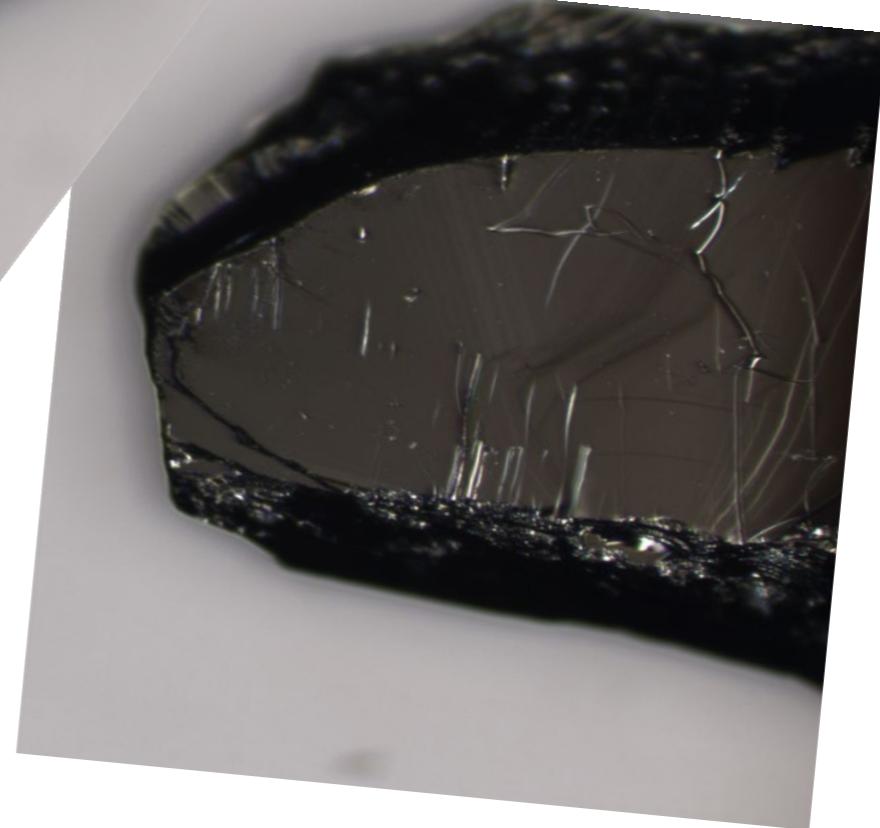




Better samples

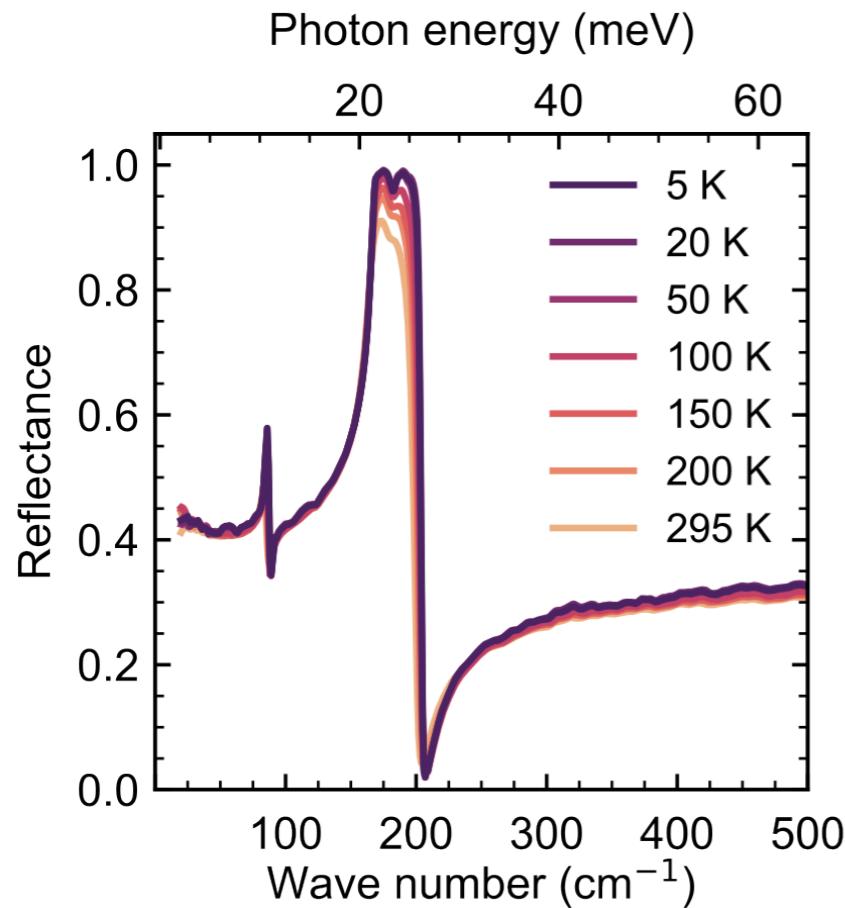


New batch



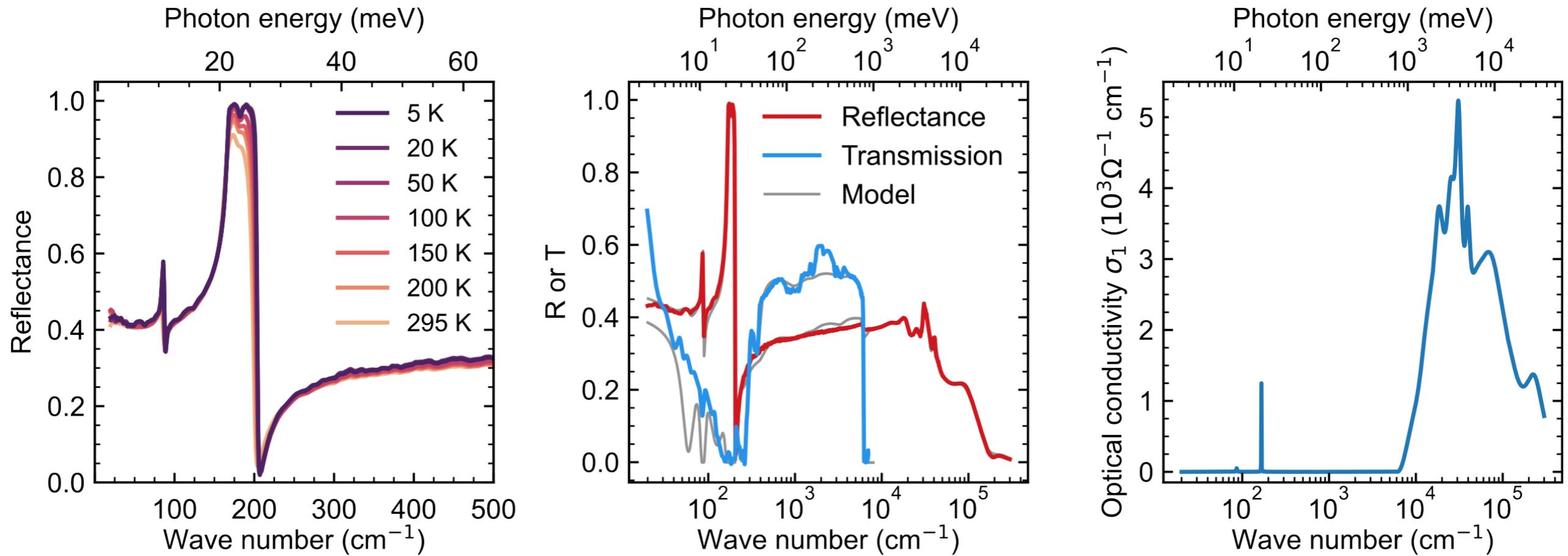
Old batch

Even fewer carriers



No Drude component.
Reststrahlen band between TO and LO frequencies

Even fewer carriers

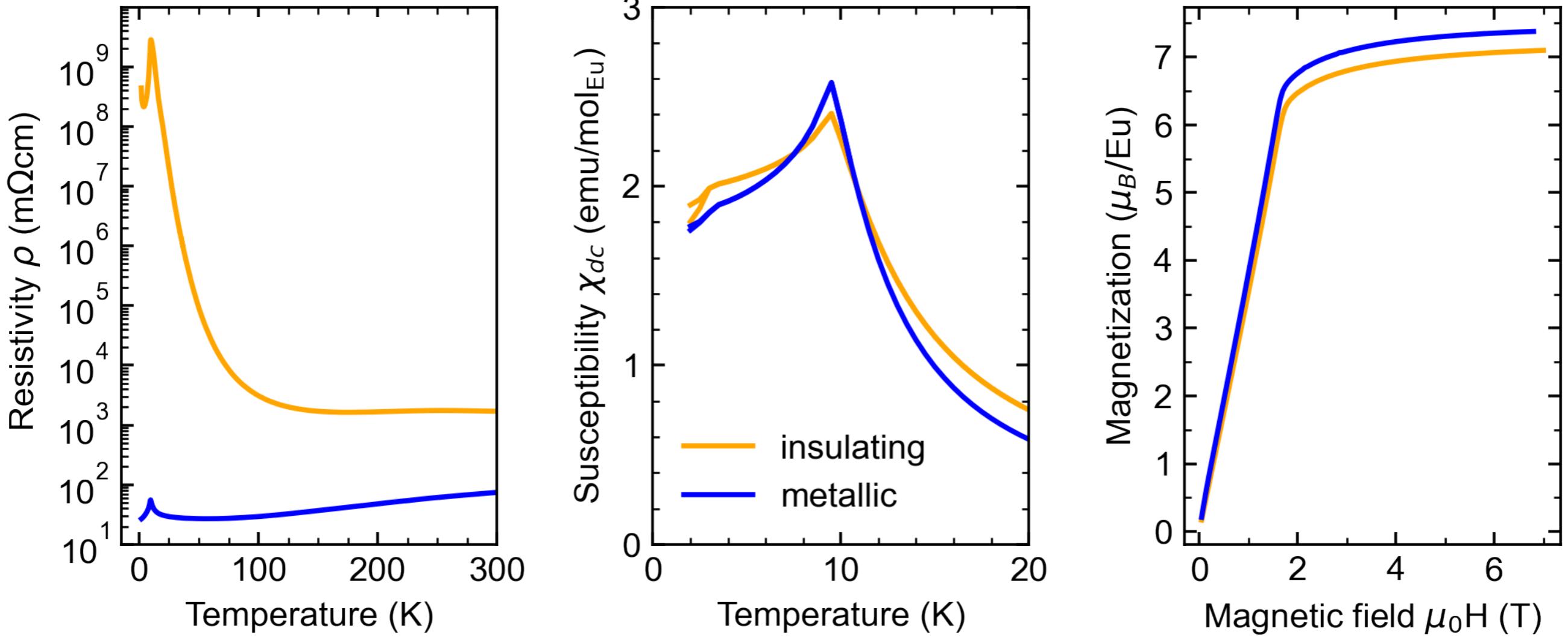


No Drude component.

Reststrahlen band between TO and LO frequencies.

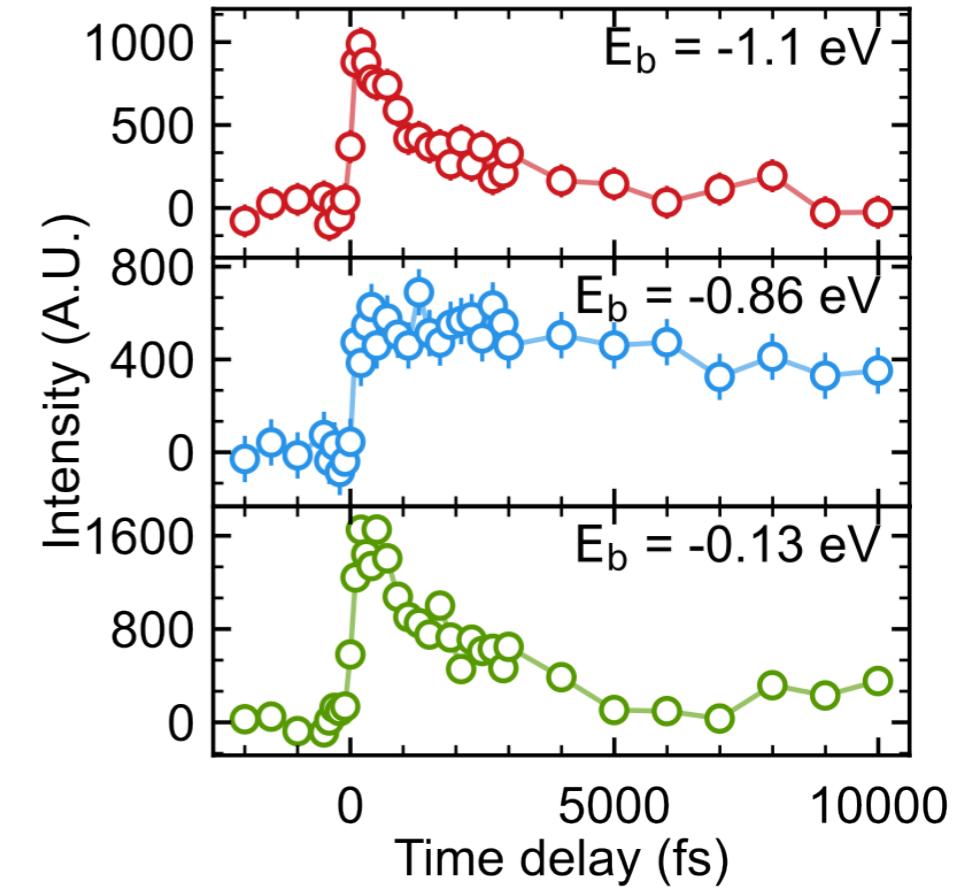
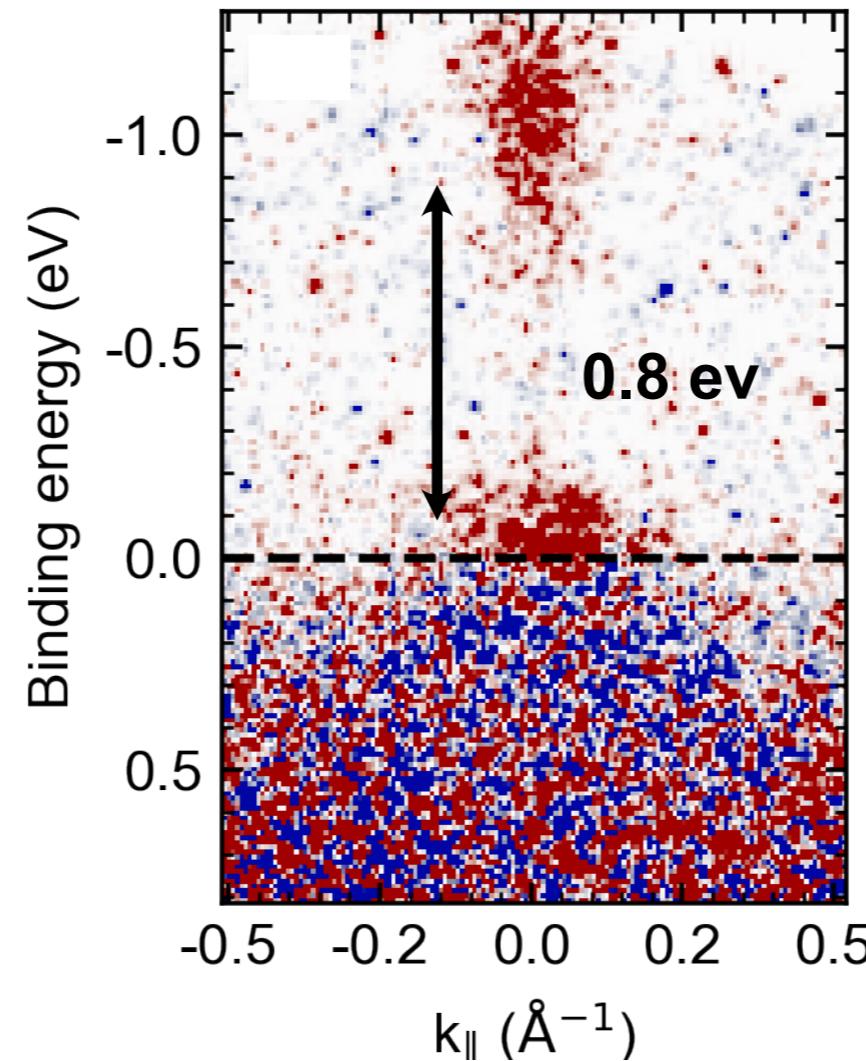
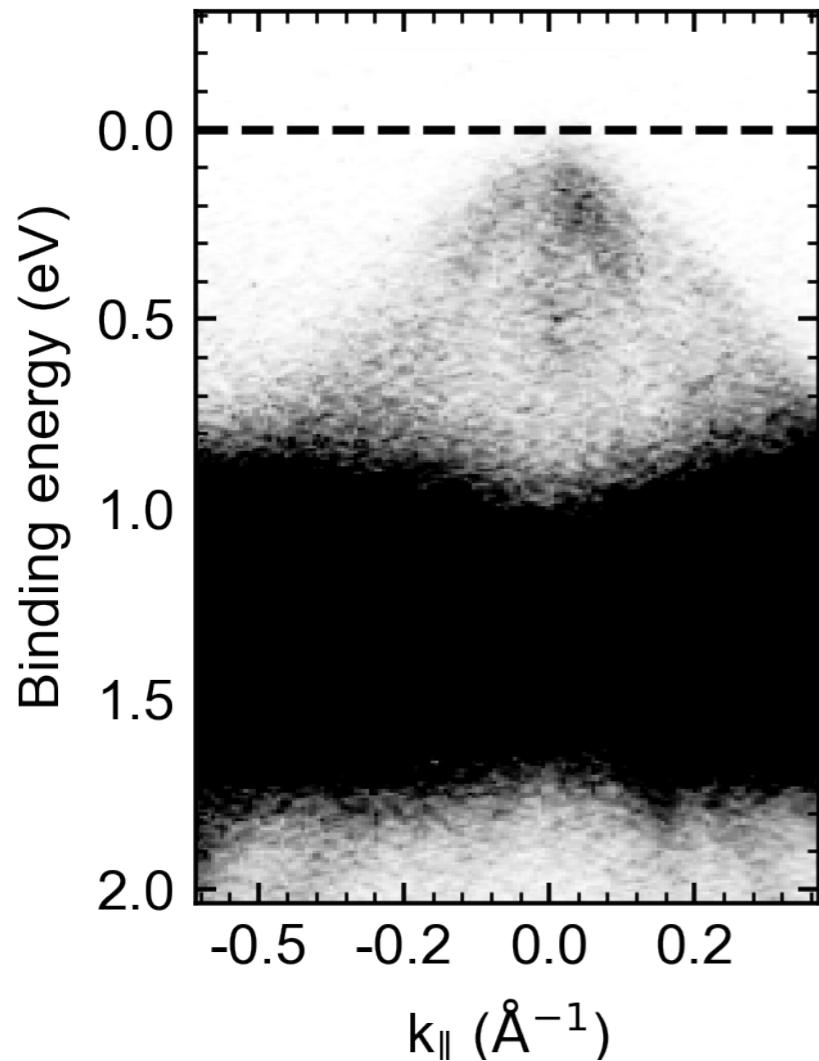
Can't use simple Kramers-Kronig analysis!

Not a semimetal...



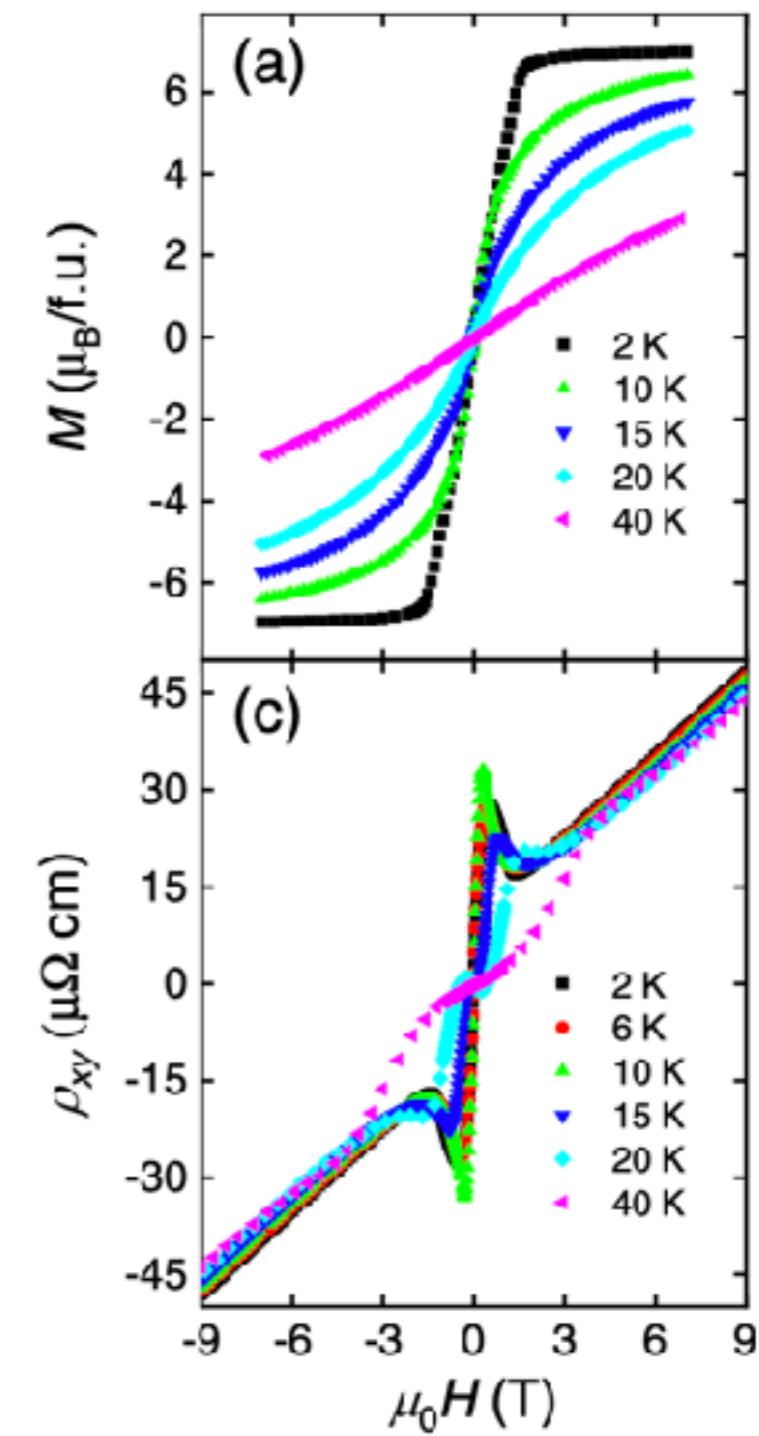
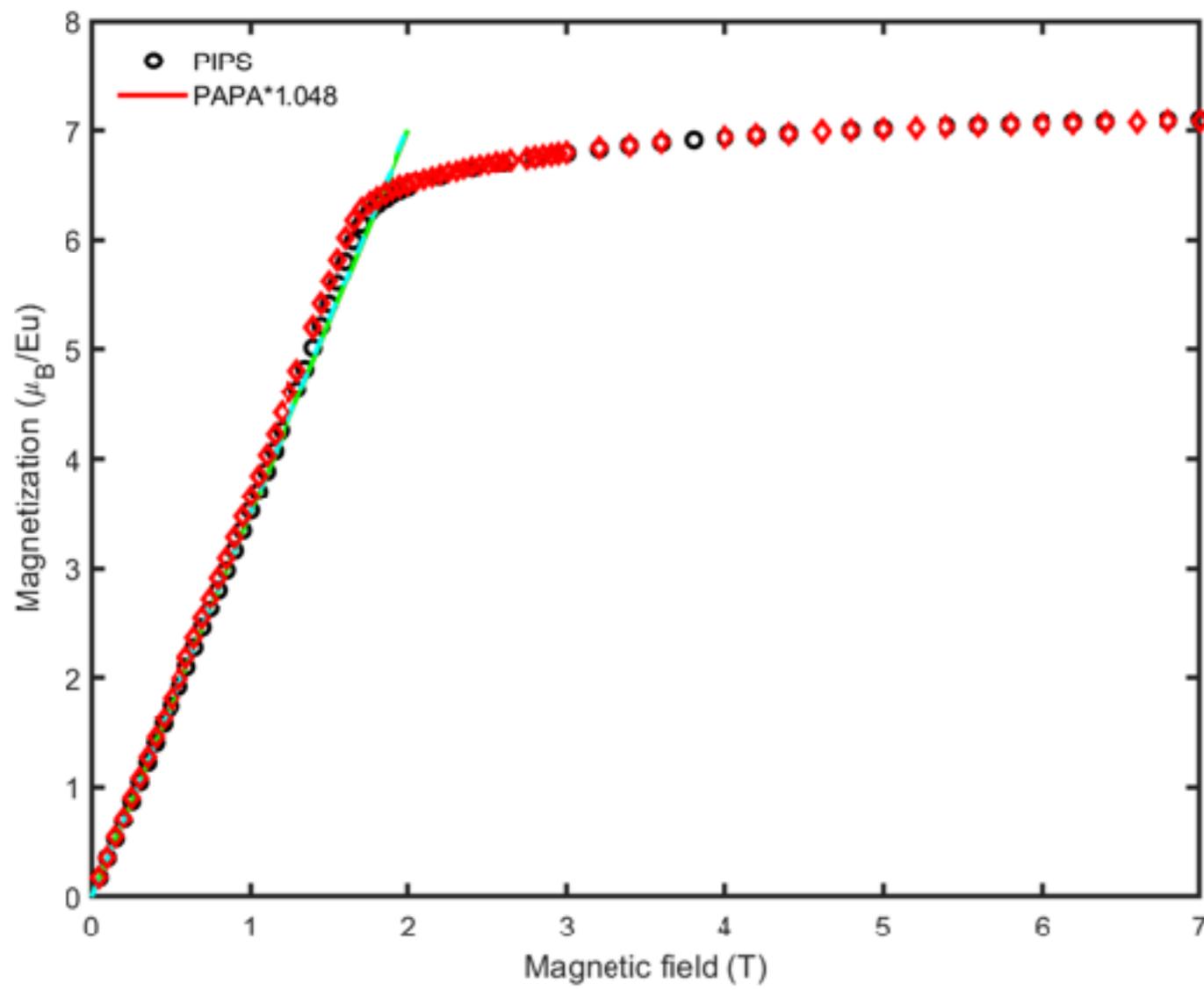
Drude doesn't know about topology!
This cannot be a semimetal.

Not a semimetal...

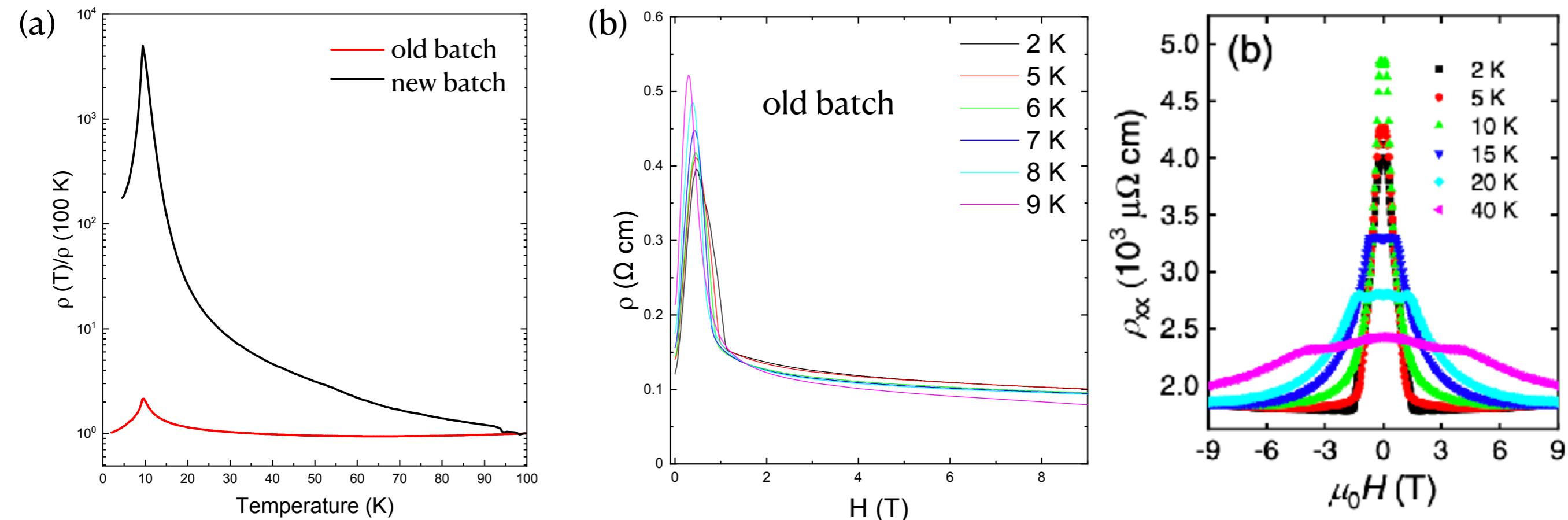


Hugo Dil's group, EPFL

Magnetic properties

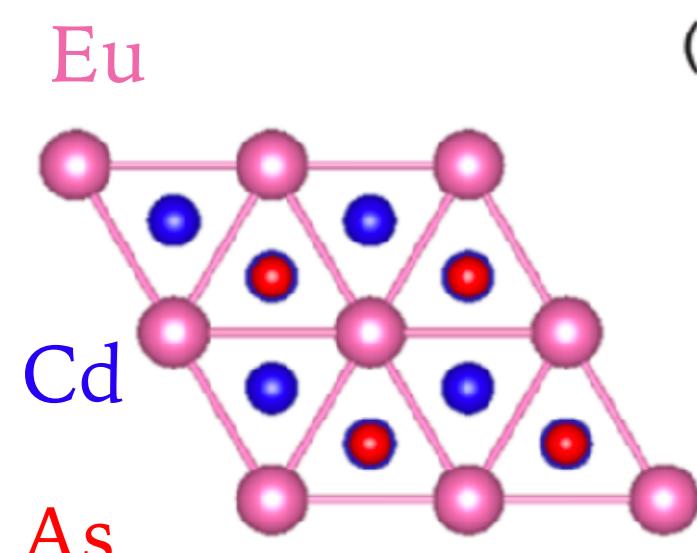


Resistivity

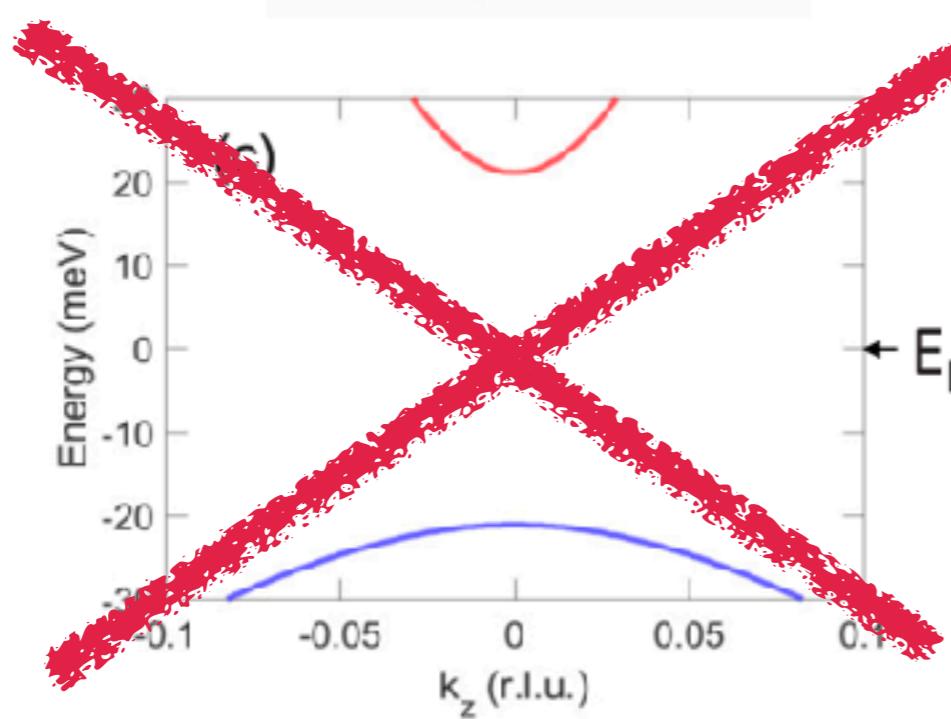
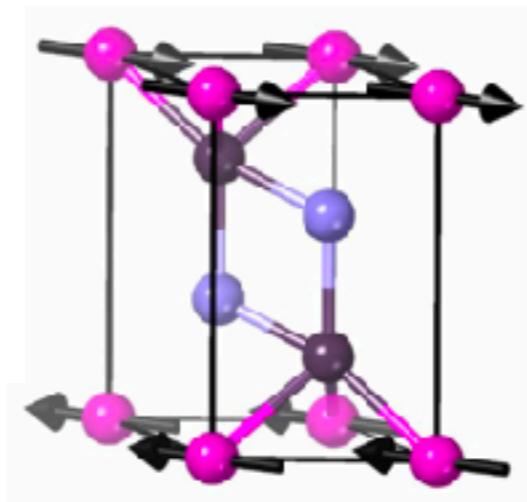


This cannot be a semimetal.

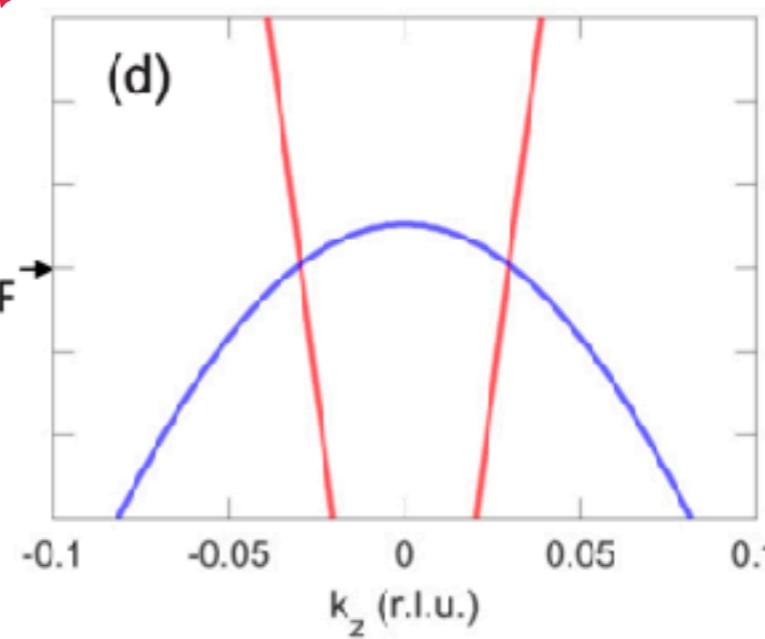
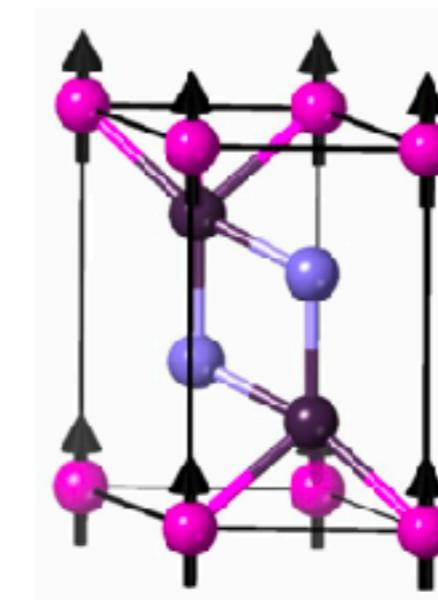
Topological semimetal EuCd_2As_2



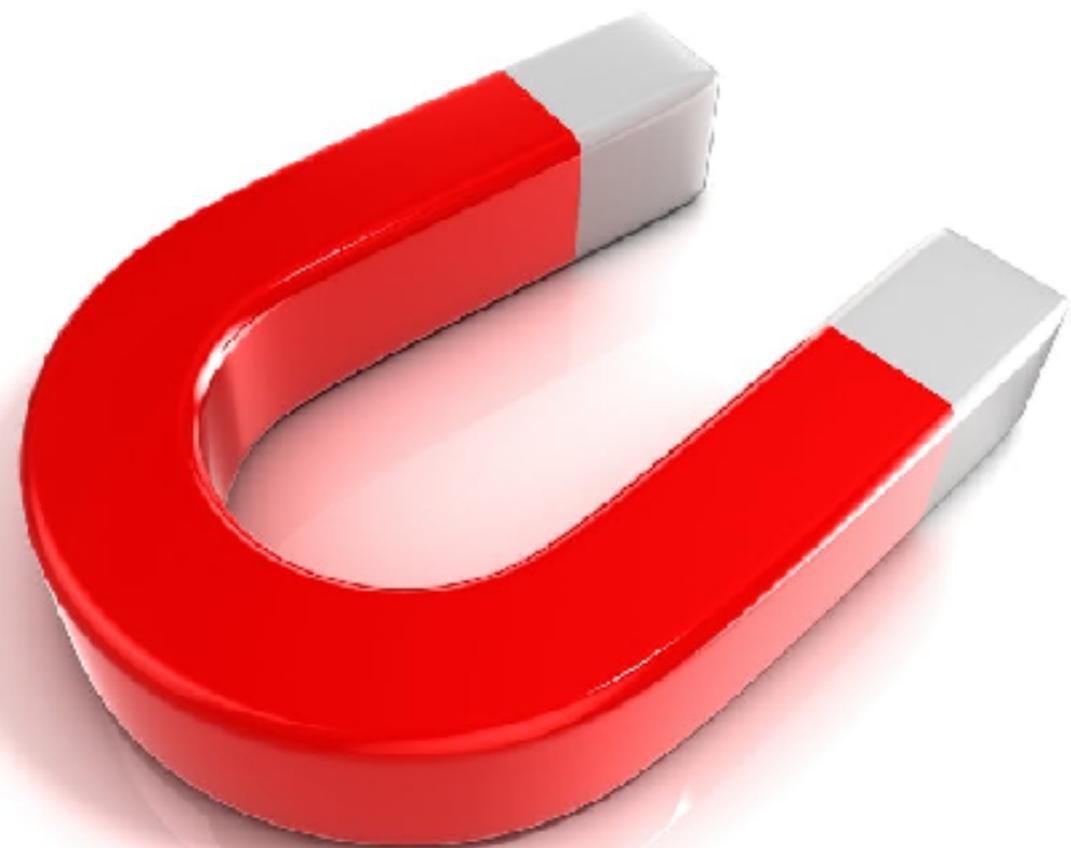
(a) $\mathbf{B} = 0$



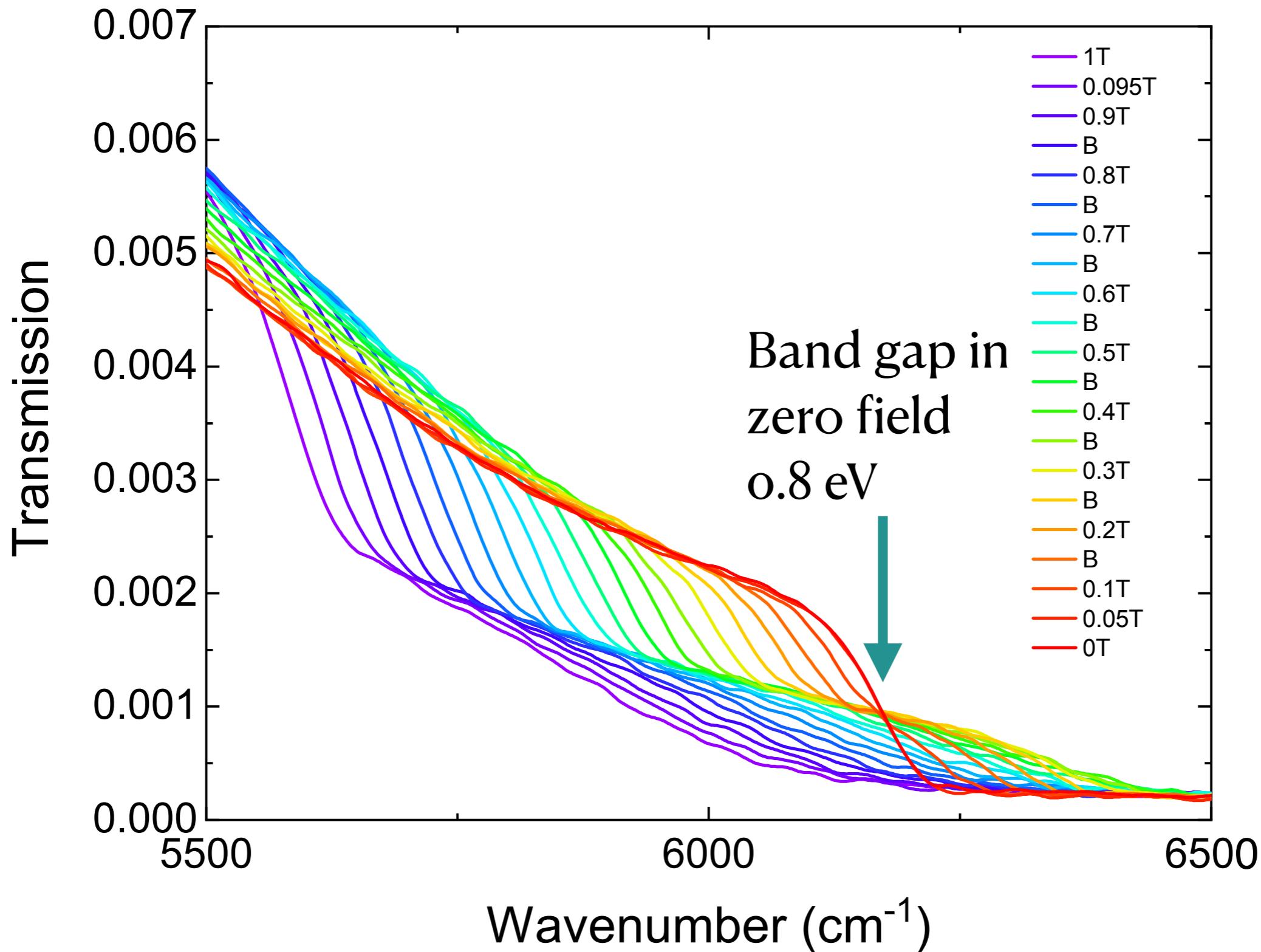
(b) $B > B_c$



Magneto-transmission at 4 K

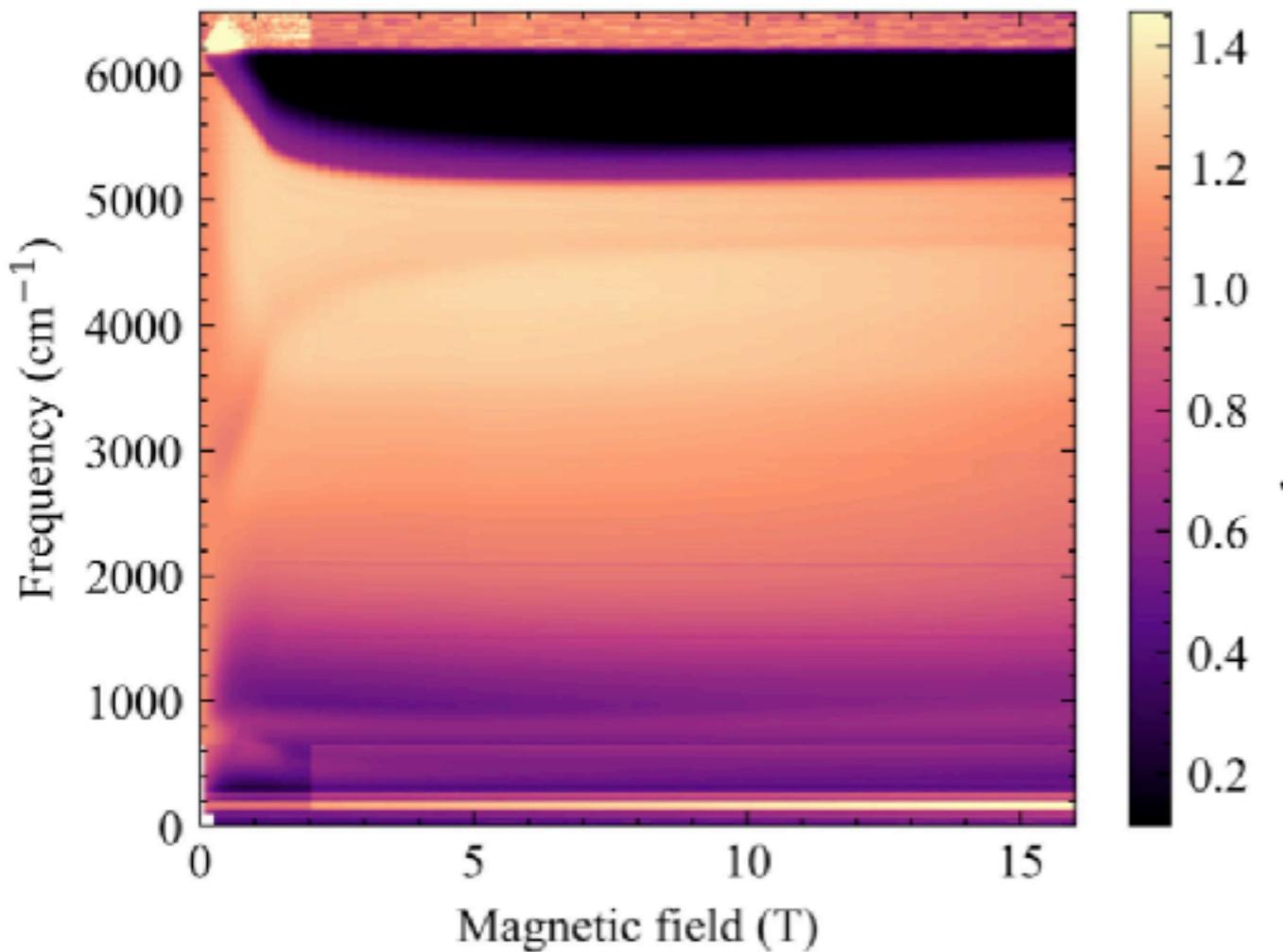


Magneto-transmission below 1 T

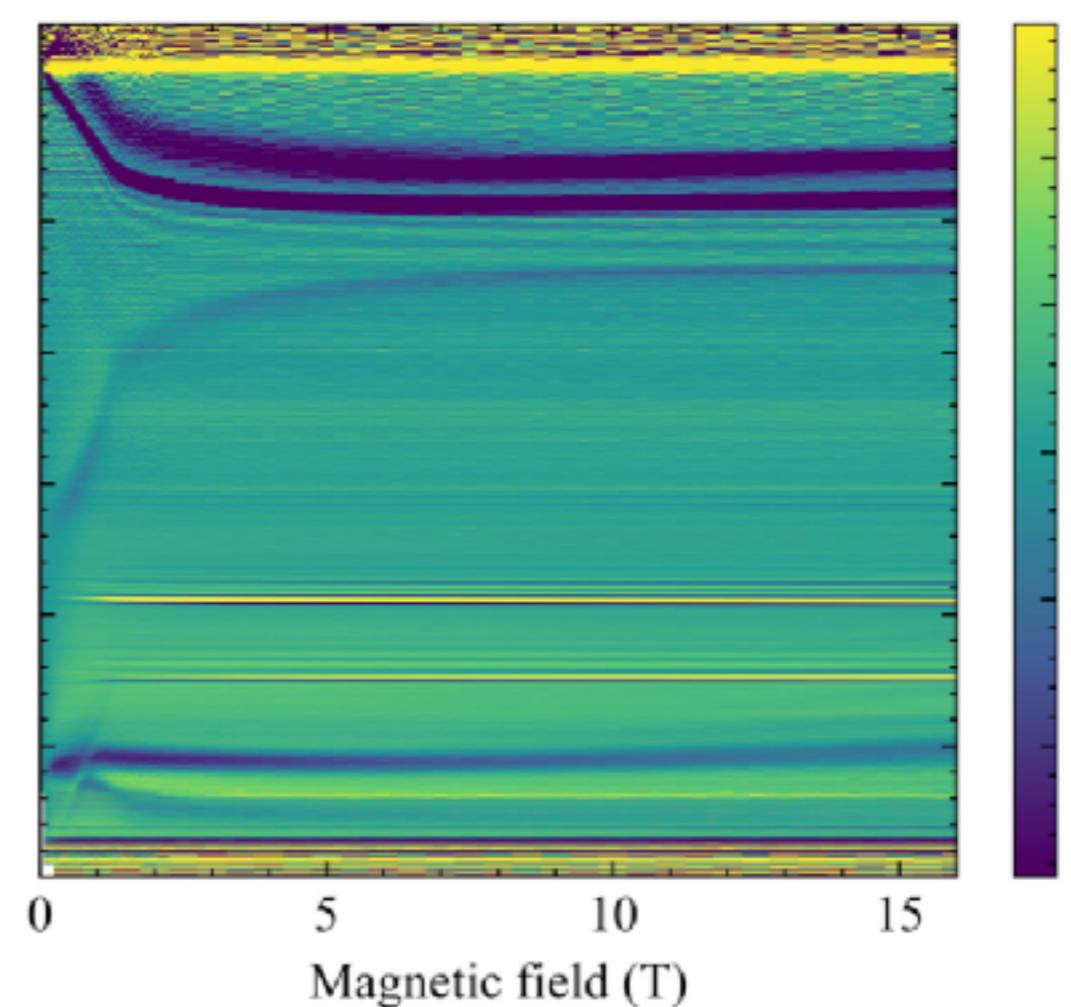


T_B/T_o in the mid infrared

Relative transmission



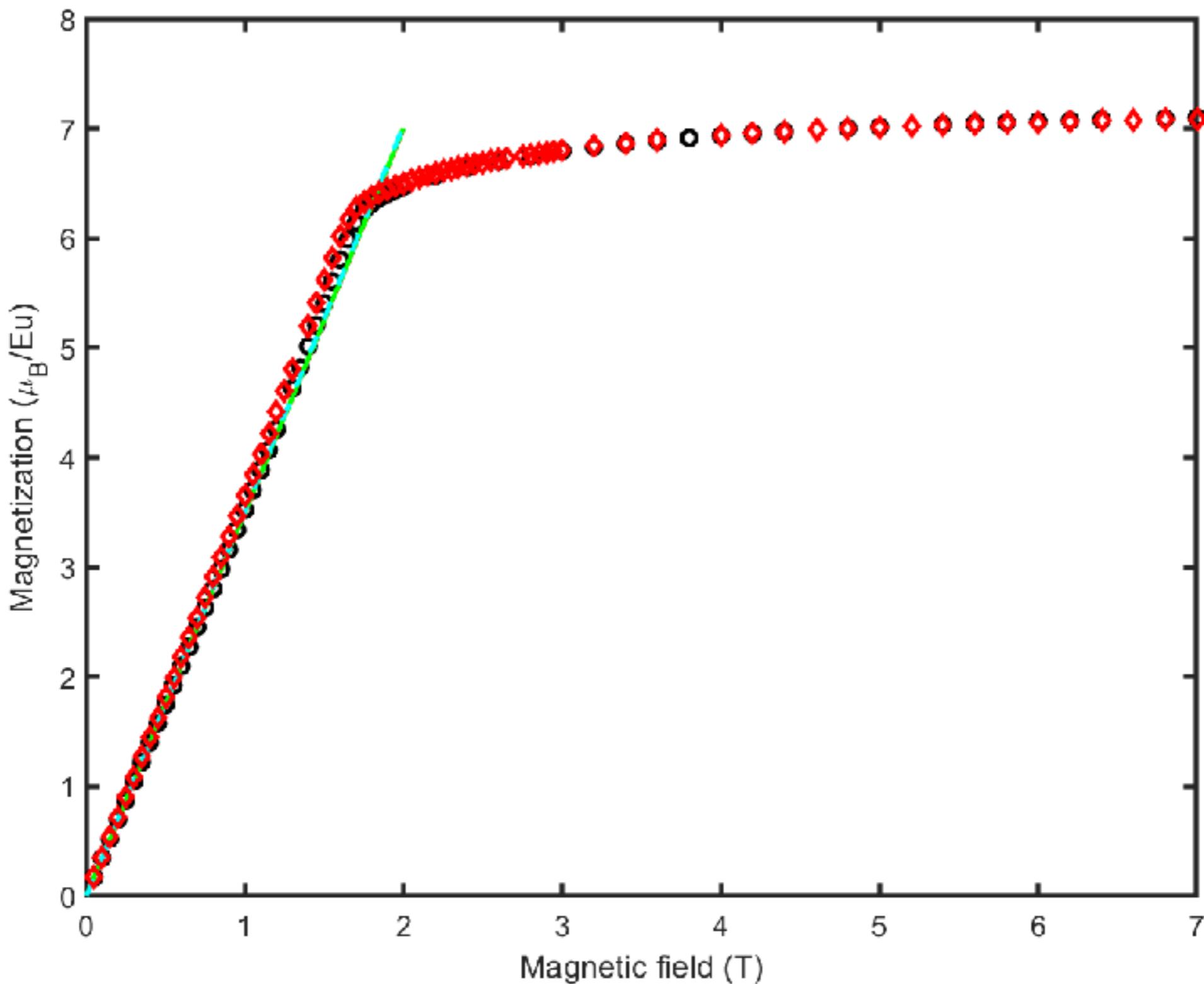
Derivative of relative transmission



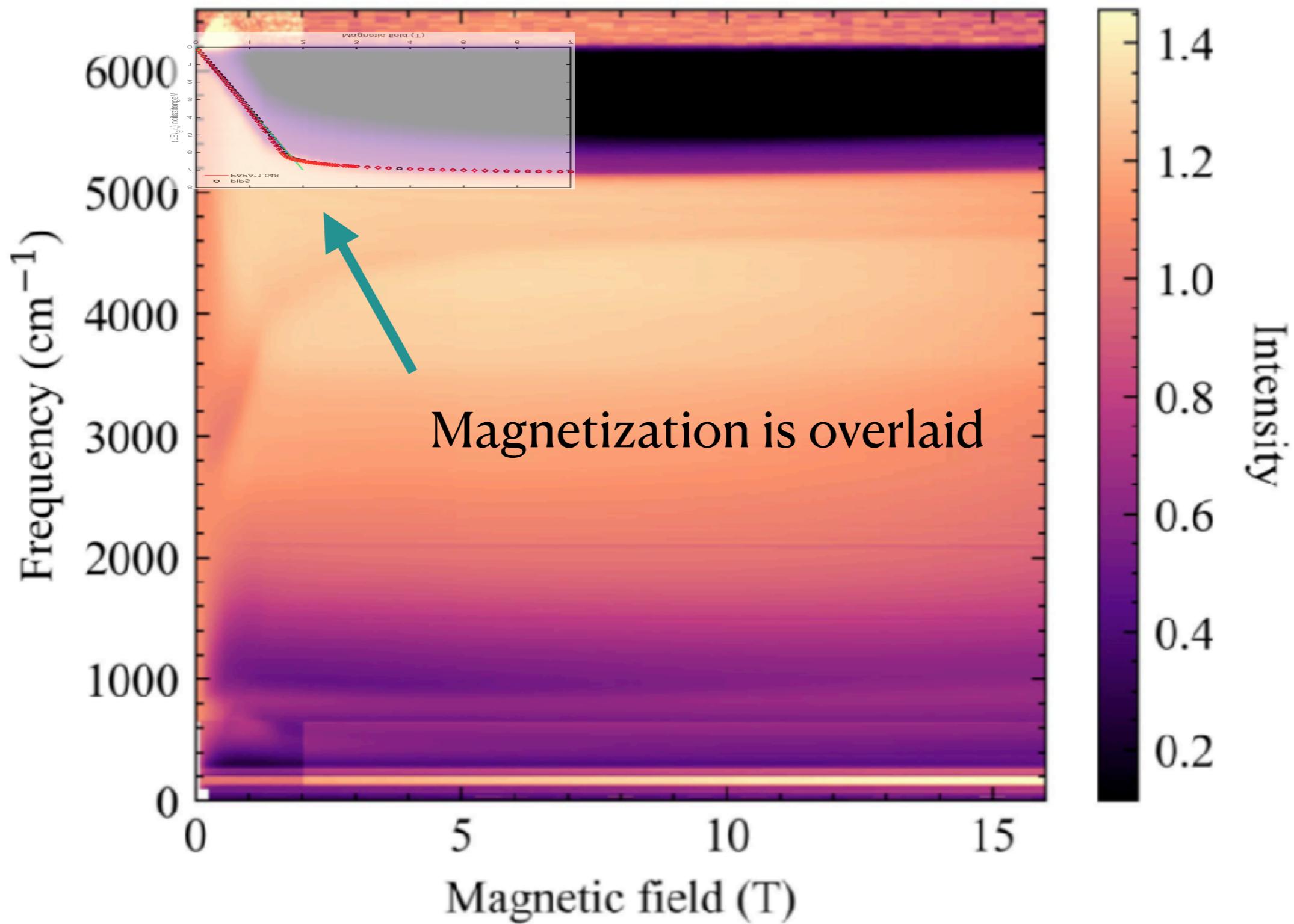
Gap of 0.8 eV decreases by 125 meV, under applied ~4 T.

Spin polarized bands. In-gap “stuff”.

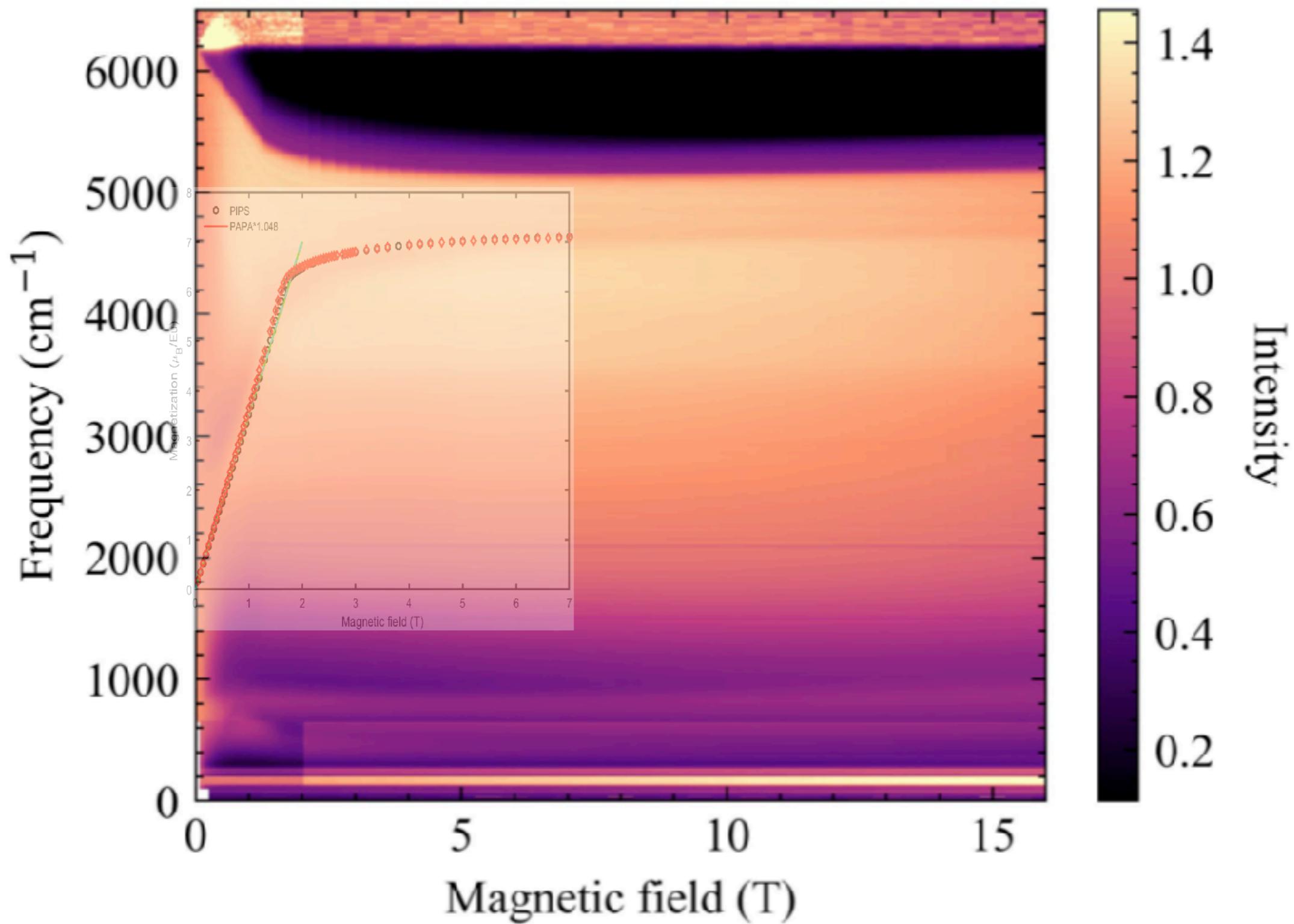
Magnetization



T_B/T_o in the mid infrared



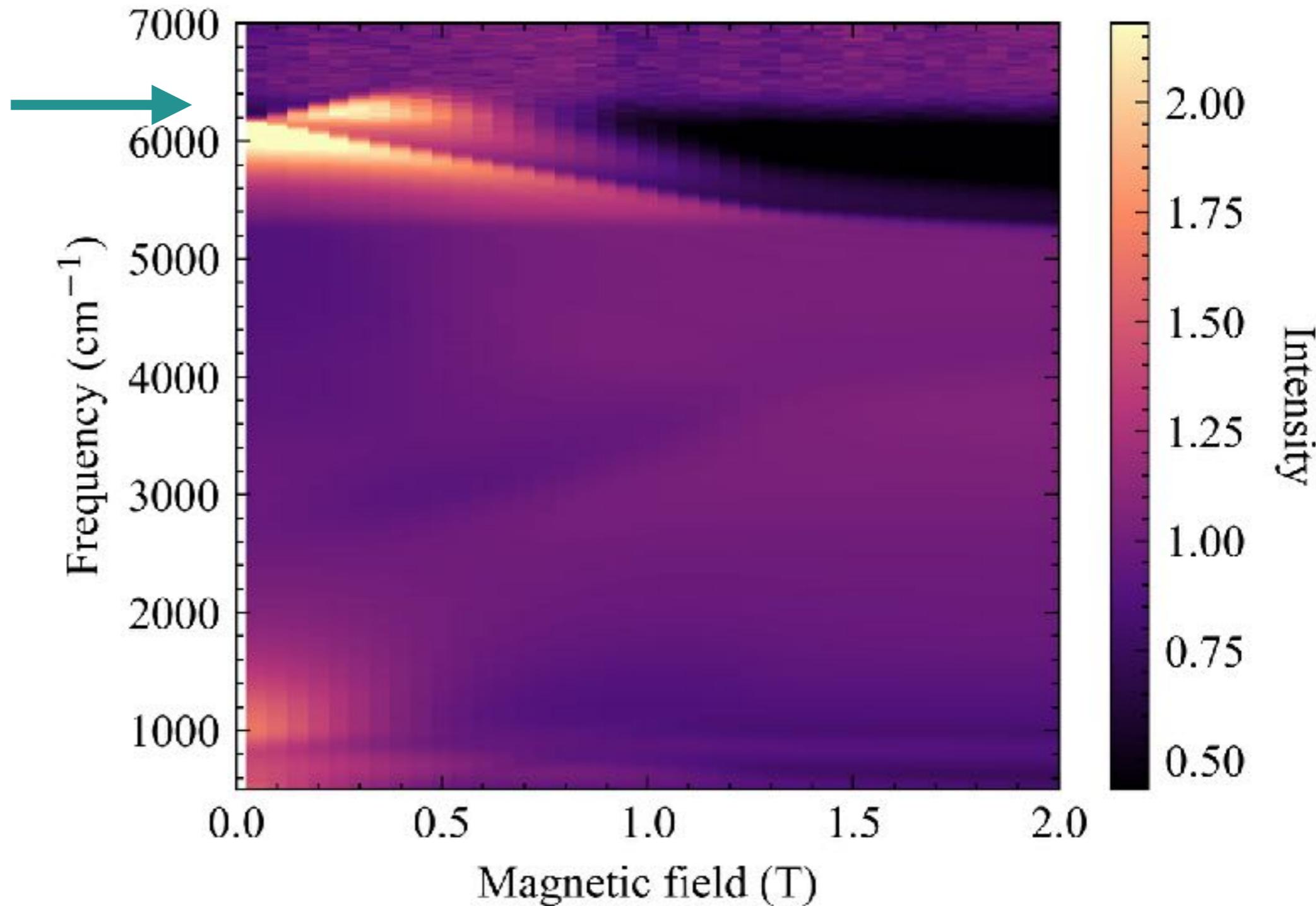
T_B/T_o in the mid infrared



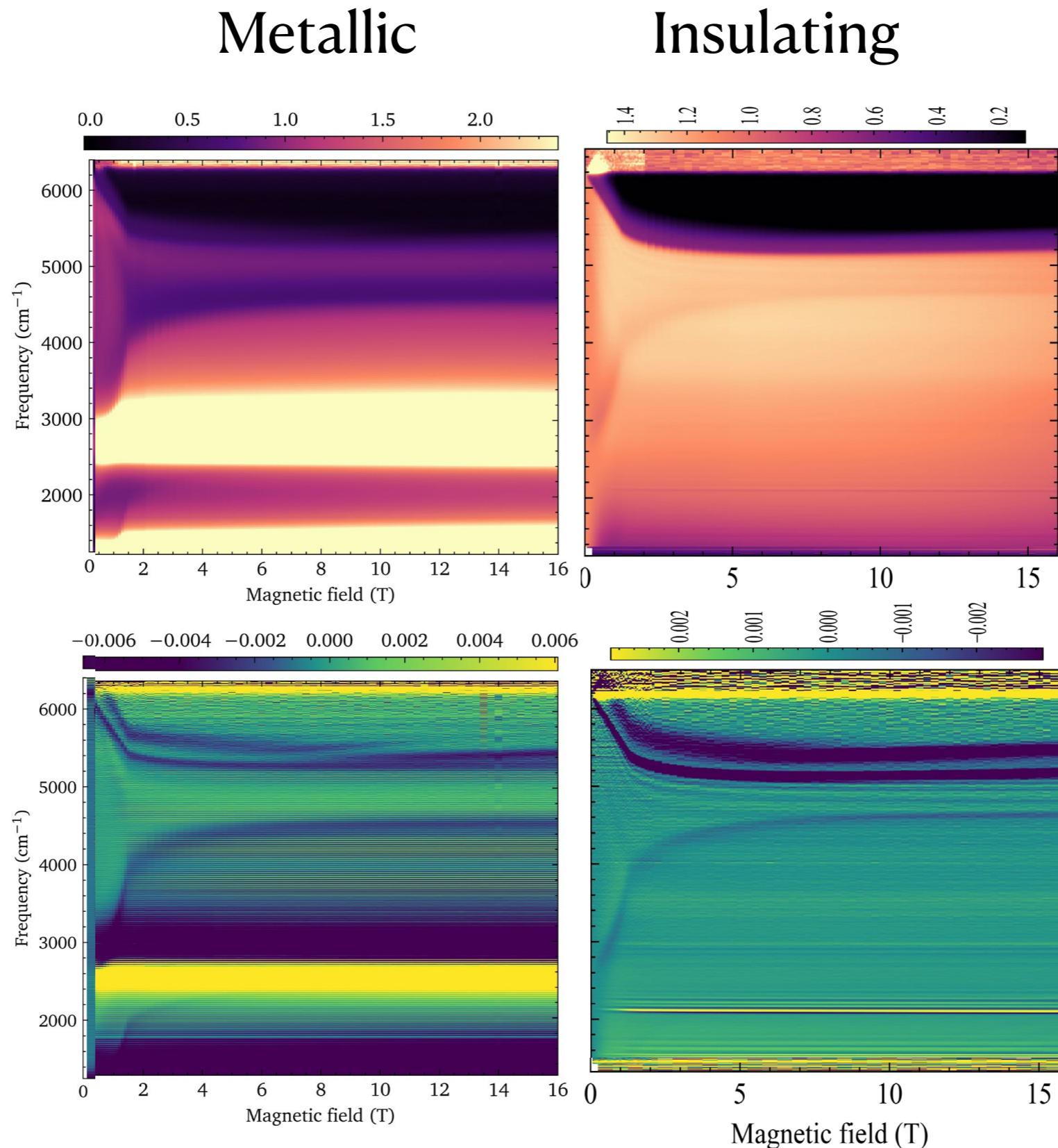
T_B/T_{avg}

The effective g factor is above 1500.

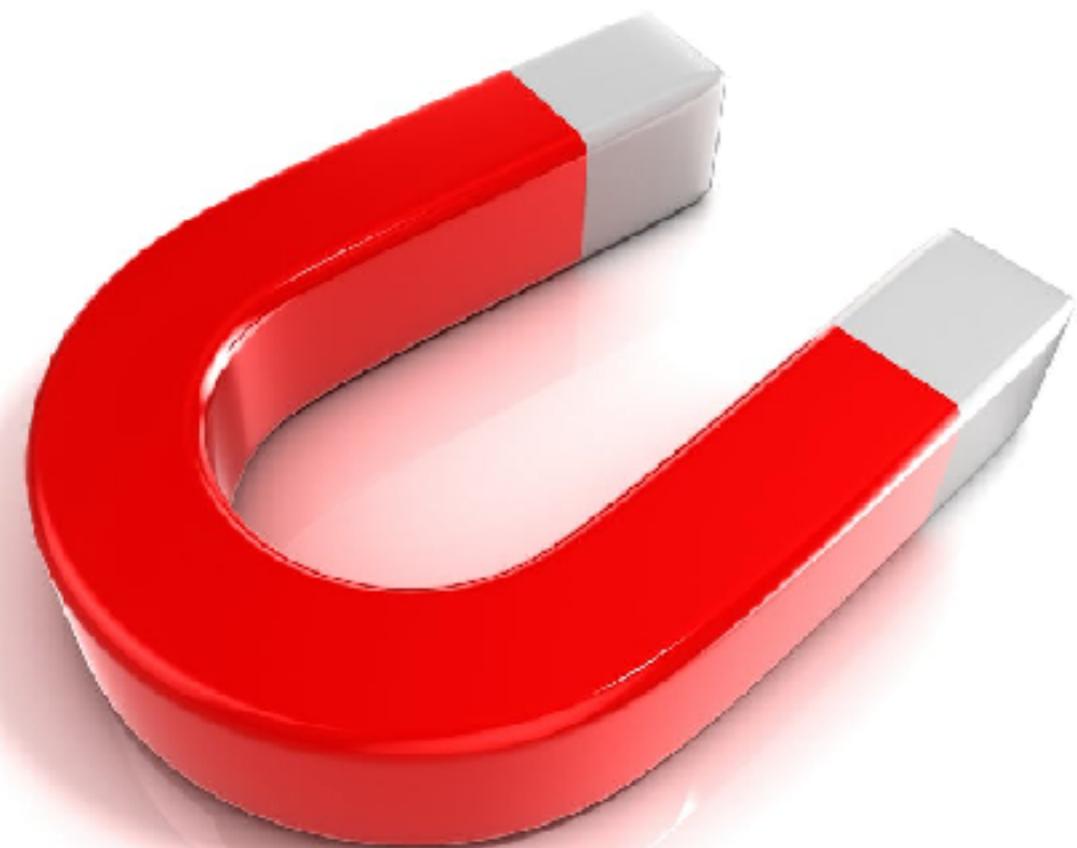
$$g \propto \frac{\Delta E}{\Delta B}$$



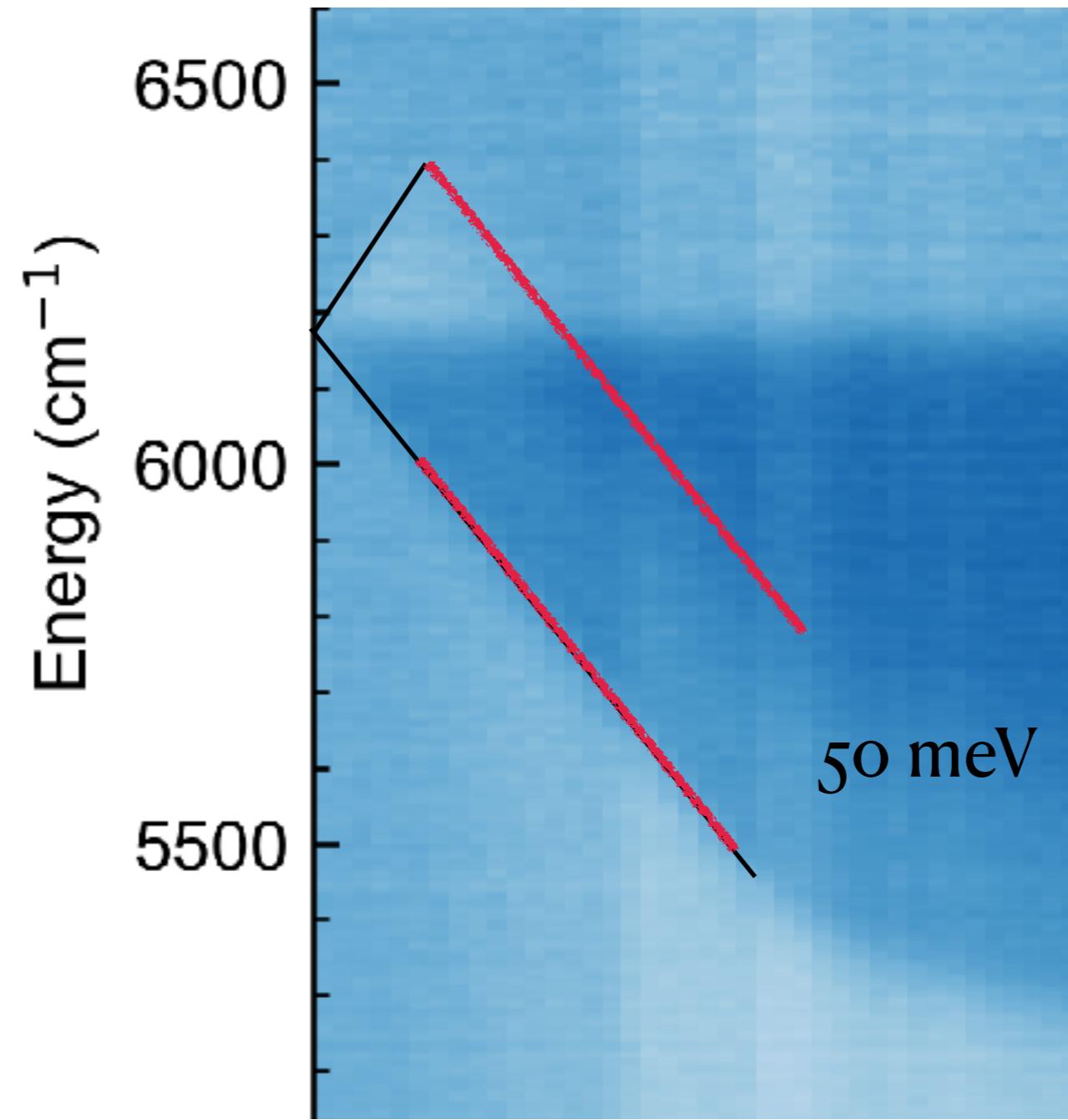
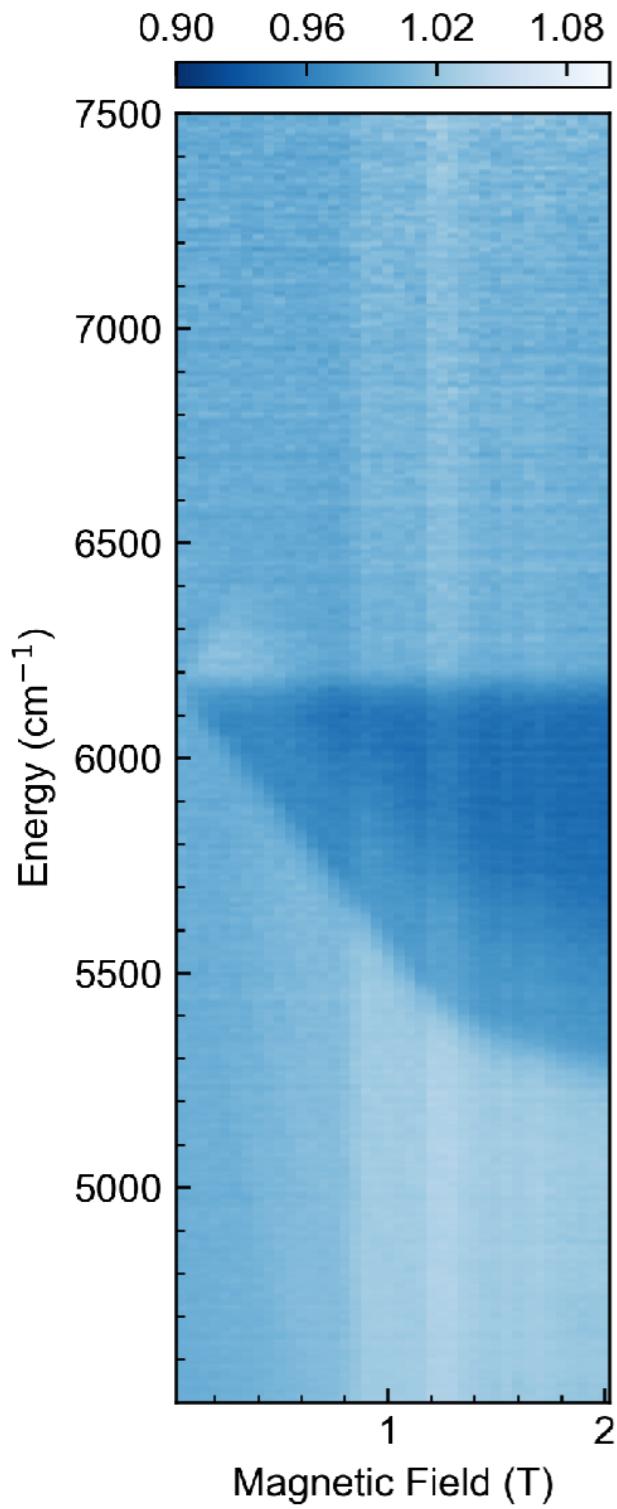
Metallic vs insulating samples



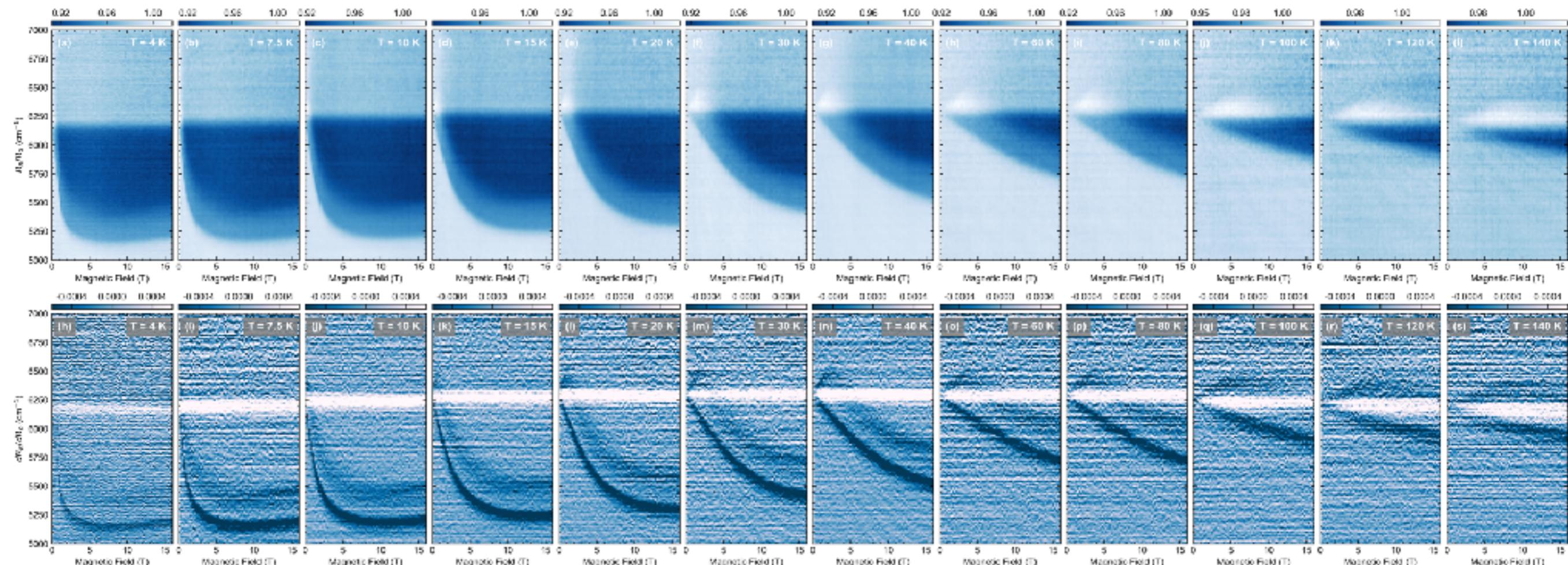
Magneto-reflection from 4 K to 140 K



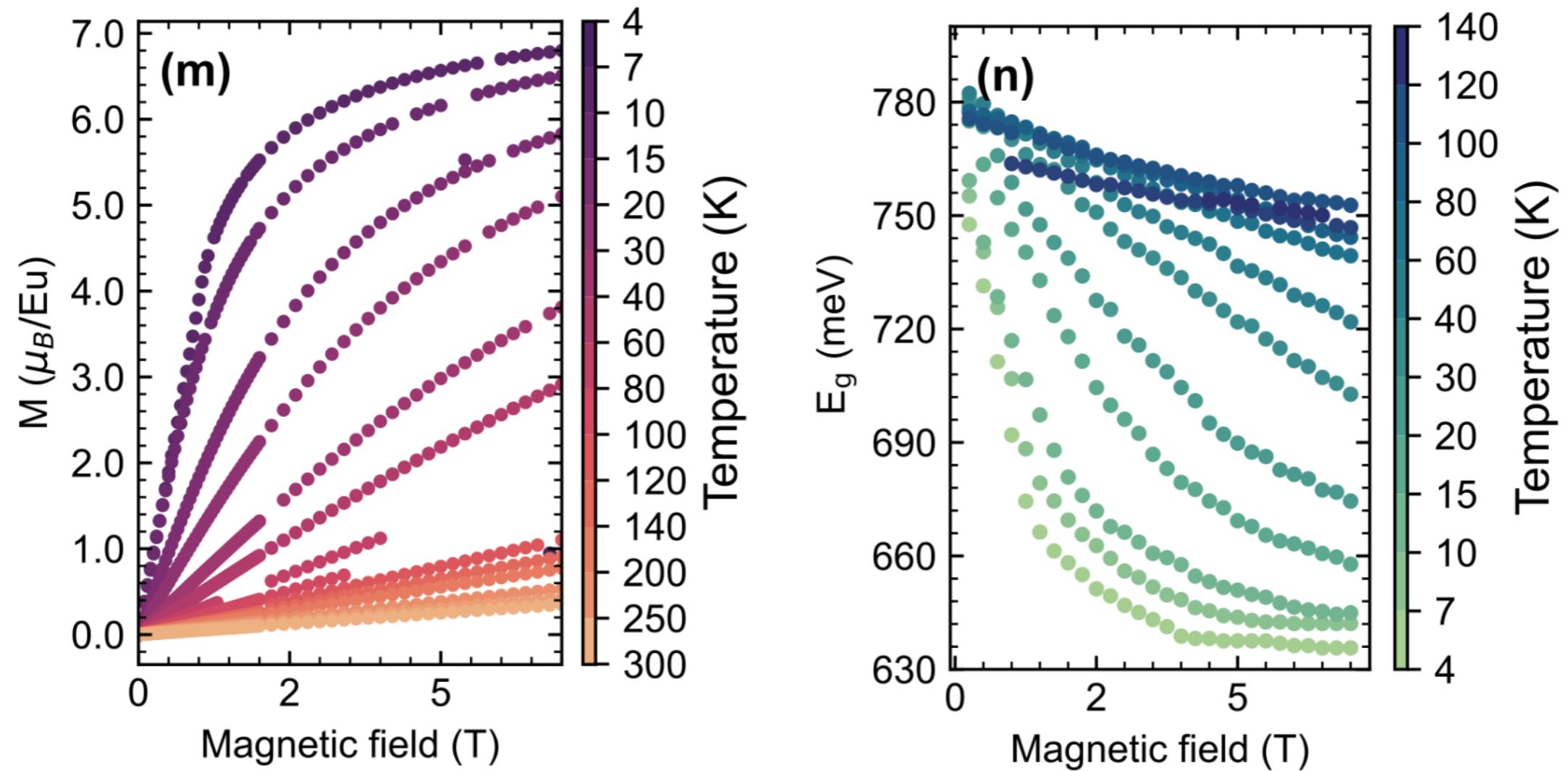
Gap splitting and decrease in field: 4 K



R_B/R_o up to 140 K - high field



Gap decrease mimics magnetization



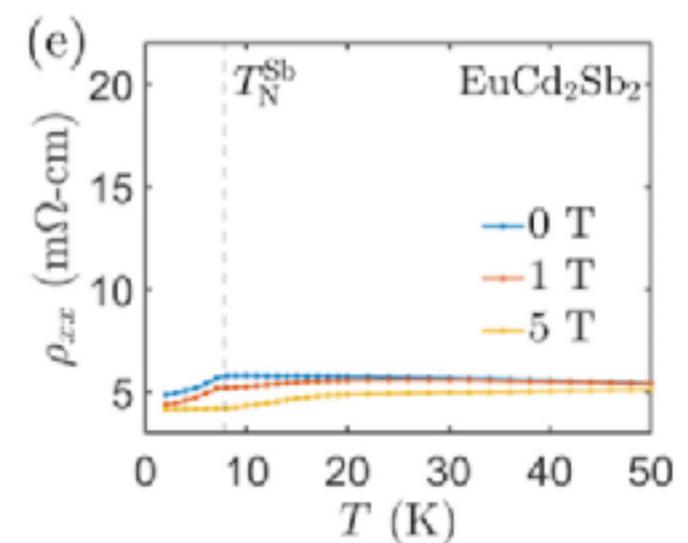
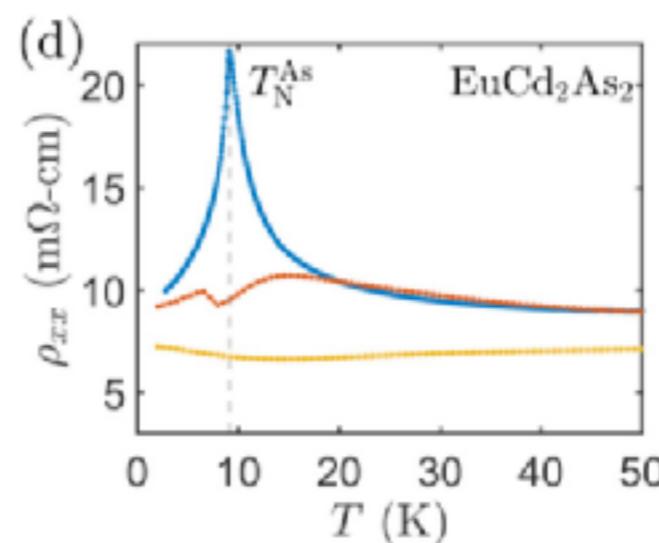
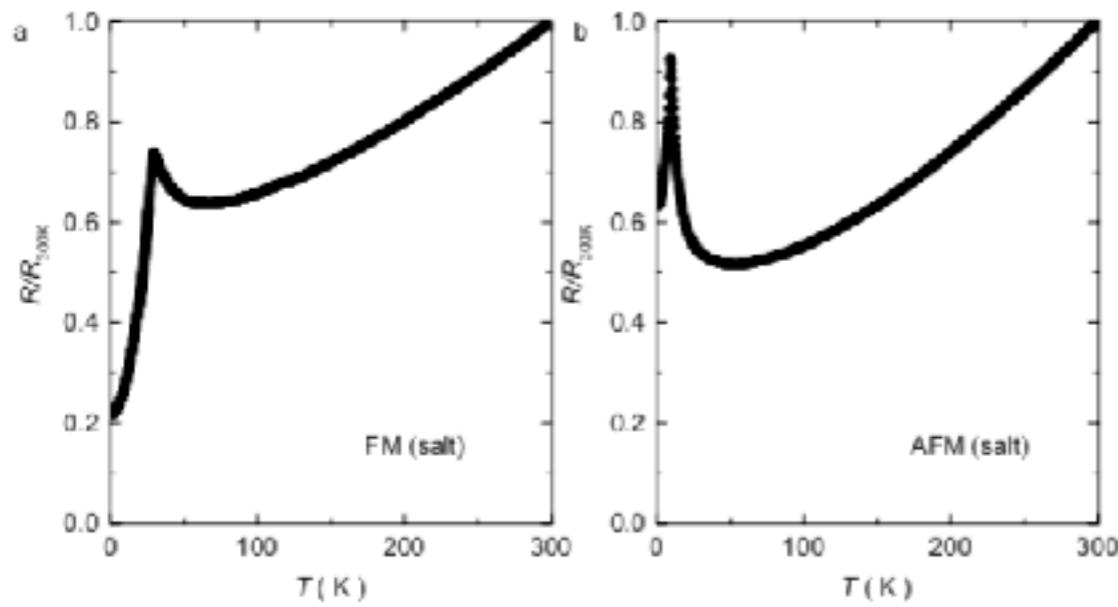
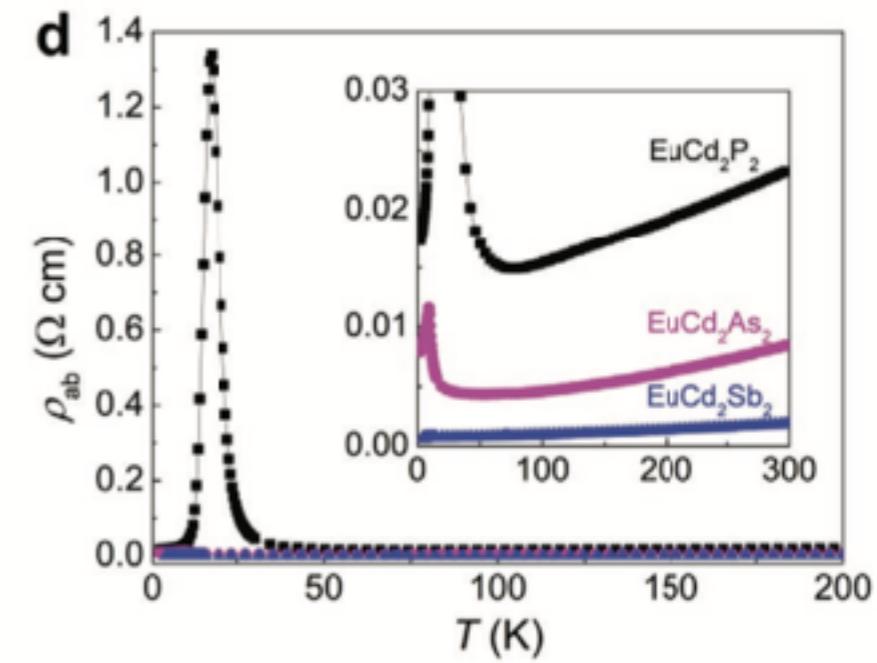
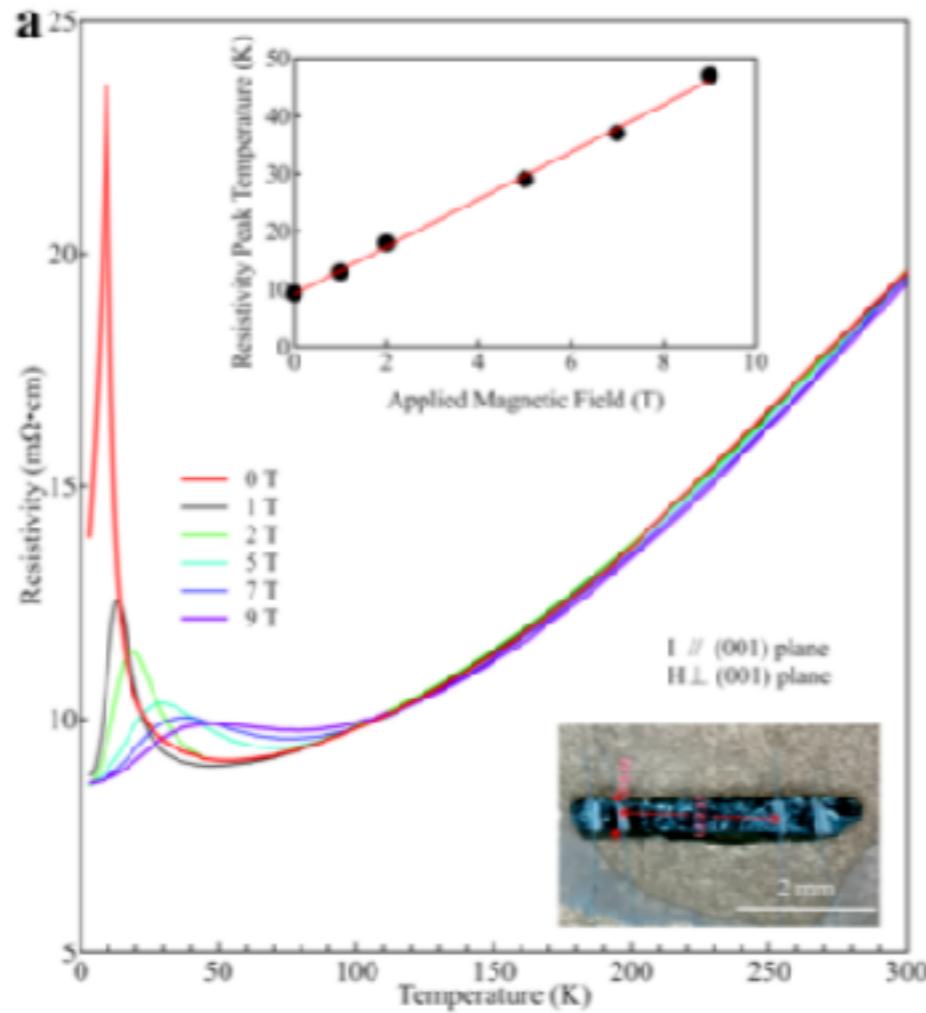
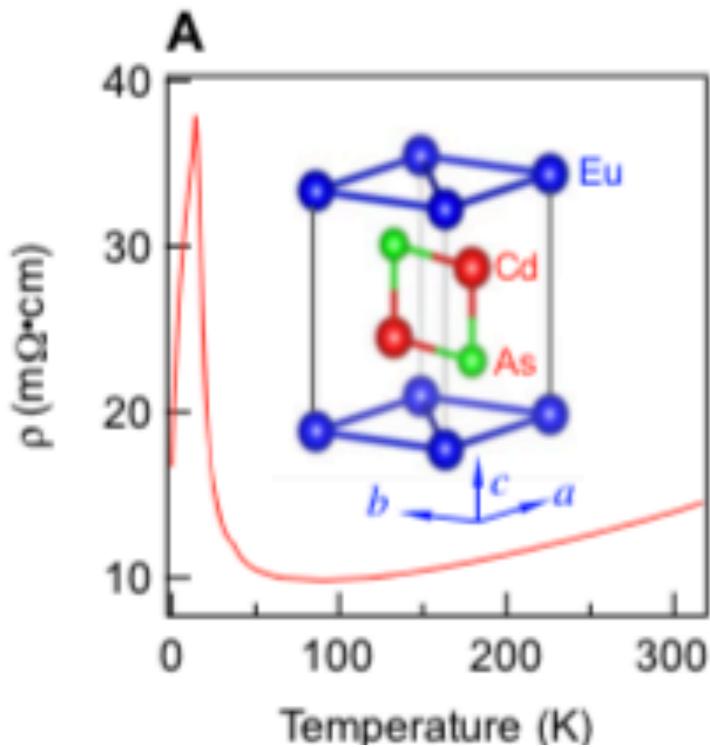
$$\Delta E_g = -\frac{1}{2} \mathcal{J}_{\text{eff}} S M(T, H) / M_S$$

$$\mathcal{J}_{\text{eff}} = 80 \text{ meV}$$

Exchange coupling dictates band structure

Local Eu magnetism strongly influences the band gap.

What about transport?



Conclusions

EuCd_2As_2 is a semiconductor with a 0.8 eV gap.

Clean samples are insulating.

The band structure is remarkably sensitive to magnetic fields.

Probably no topology involved.