Temperature dependent optical properties of single-crystalline Ni (100)

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Ph.D. Defense

11/02/2020

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Curriculum Vitae

Education

2009	Bachelor in Physics, Shahrood University of Technology
2012	Master in Physics, Tehran University
2018	Master in Physics, New Mexico State University

Publications

- **F. Abadizaman** and S. Zollner, *Optical constants of single-crystalline Ni (100) from 77 K to 770 K from ellipsometry measurements* (in preparation).

- **F. Abadizaman** and S. Zollner, *Optical constants of polycrystalline Ni from 0.06 to 6.0 eV at 300 K*, J. Vac. Sci. Technol. B **37**, 062920 (2019).

- C. Emminger**, F. Abadizaman**, N. S. Samarasingha, T. E. Tiwald, and S. Zollner, *Temperature dependent dielectric function and direct bandgap of Ge*, J. Vac. Sci. Technol. B **38**, 012202 (2020).

- S. Zollner, P. Paradis, **F. Abadizaman**, and N. S. Samarasingha, *Drude and Kukharskii mobility of doped semiconductors extracted from Fourier-transform infrared ellipsometry spectra*, J. Vac. Sci. Technol. B **37**, 012904 (2019).

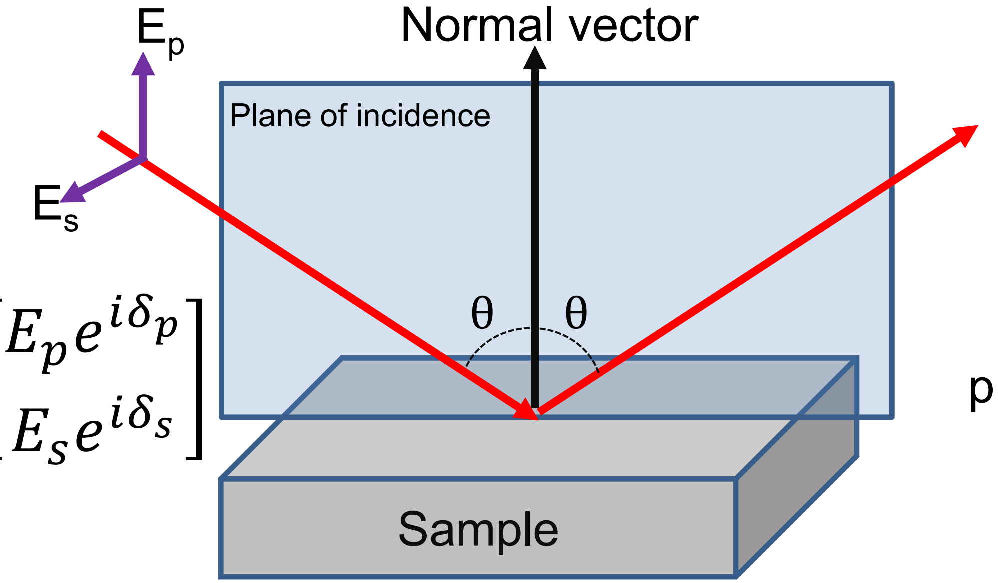
Conference presentations

- **F. Abadizaman**, S. Zollner, *Temperature dependent optical properties of single-crystalline Ni (100)*, ELI Beamlines User Conference, Prague, Czech Republic, October 2020. (Talk)
- **F. Abadizaman**, C. Emminger, S. Knight, M. Schubert, S. Zollner, *Optical Hall Effect in the Multivalley Semiconductor Te-doped GaSb*, AVS 66th International Symposium & Exhibition, Columbus, Ohio, USA, October 2019. (Talk)
- **F. Abadizaman**, S. Zollner, *Optical constants of polycrystalline Ni from 0.06 to 6.0 eV at 300 K*, AVS 66th International Symposium & Exhibition, Columbus, Ohio, USA, October 2019. (Talk)
- **F. Abadizaman**, S. Zollner, *Temperature Dependence of Critical points of Ni from 77-800 K*, AVS 65th International Symposium & Exhibition, Long Beach, Los Angeles, California, USA, October 2018. (Talk)
- **F. Abadizaman**, S. Zollner, *Anomaly in optical constants of Ni near the Curie temperature*, NMAVS Symposium, Albuquerque, NM, USA, May 2018. (Talk)
- **F. Abadizaman**, S. Zollner, *Temperature dependent Mueller matrix measurements of magnetized Ni near the Curie temperature*, APS March Meeting, Los Angeles, California, USA, March 2018. (Talk)
- **F. Abadizaman**, P. Paradis, S. Zollner, *Temperature Dependent Mueller Matrix Measurements of Magnetized Ni near the Curie Temperature*, AVS 64th International Symposium & Exhibition, Tampa, Florida, USA, October 2017. (Talk)
- **F. Abadizaman**, S. Zollner, *Temperature Dependent Mueller Matrix Measurements of Magnetized Ni near the Curie Temperature*, NMAVS Symposium, Albuquerque, NM, May 2017. (Talk)
- Six posters and contributions to many talks.

Outline

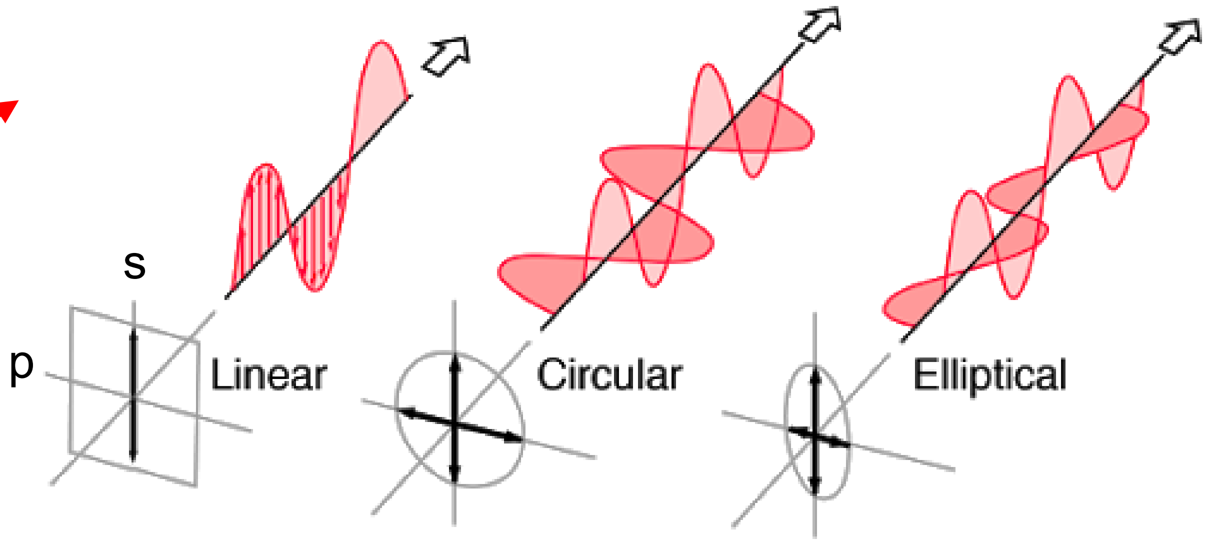
- Polarization of light
- Ellipsometry
- Experimental setup
- Anomaly in conductivity due to surface desorption
- Dielectric function model for Ni
- Optical conductivity
- Absorption peaks of single-crystalline Ni (100)
- Temperature dependent optical resistivity
- Summary

Polarization of light



$$\vec{E} = \begin{bmatrix} E_p e^{i\delta_p} \\ E_s e^{i\delta_s} \end{bmatrix}$$

http://hyperphysics.phy-astr.gsu.edu

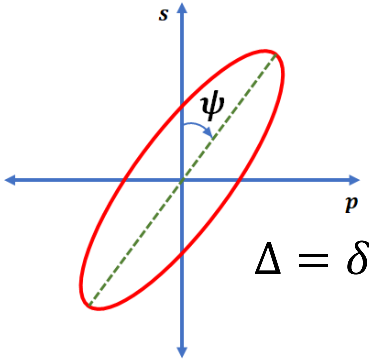


Jones matrix formalism

- Describes polarization state of light
- Cannot describe unpolarized light

$$\vec{E}_{linear,p} = \begin{bmatrix} 1 \\ 0 \end{bmatrix} \quad \vec{E}_{circular} = \frac{1}{\sqrt{2}} \begin{bmatrix} 1 \\ \pm i \end{bmatrix} \quad \vec{E}_{elliptical} = \begin{bmatrix} \sin\psi \exp(i\Delta) \\ \cos\Delta \end{bmatrix}$$

$$\vec{E}_{linear,s} = \begin{bmatrix} 0 \\ 1 \end{bmatrix}$$



$$\Delta = \delta_p - \delta_s$$

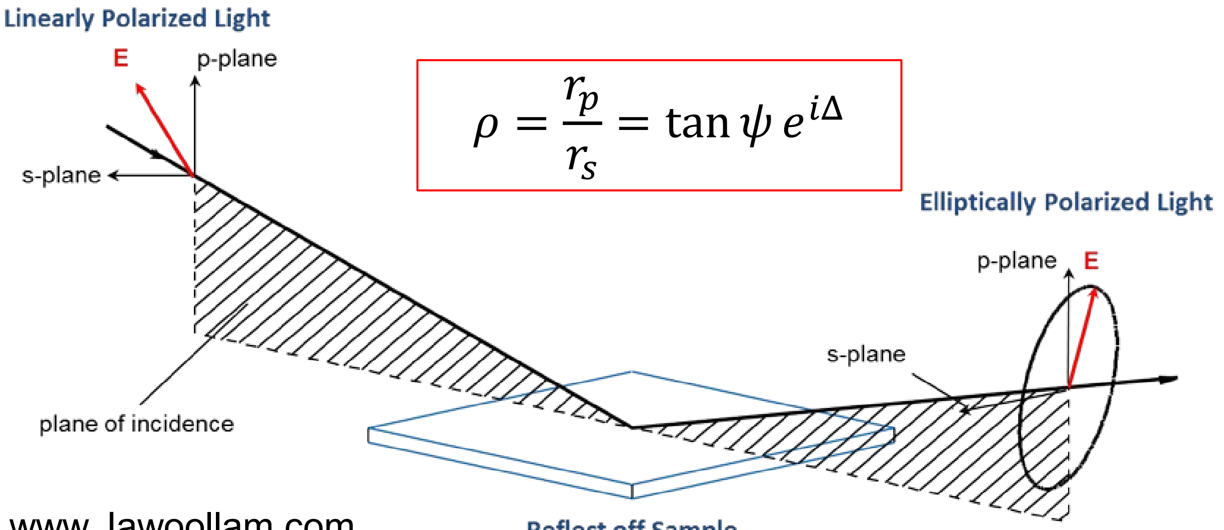
$$P(\phi) = \begin{bmatrix} \cos^2\phi & \sin\phi\cos\phi \\ \sin\phi\cos\phi & \sin^2\phi \end{bmatrix}$$

Polarizer

$$R_{qwp} = \begin{bmatrix} 1 & 0 \\ 0 & -i \end{bmatrix} \quad R_{hwp} = \begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix}$$

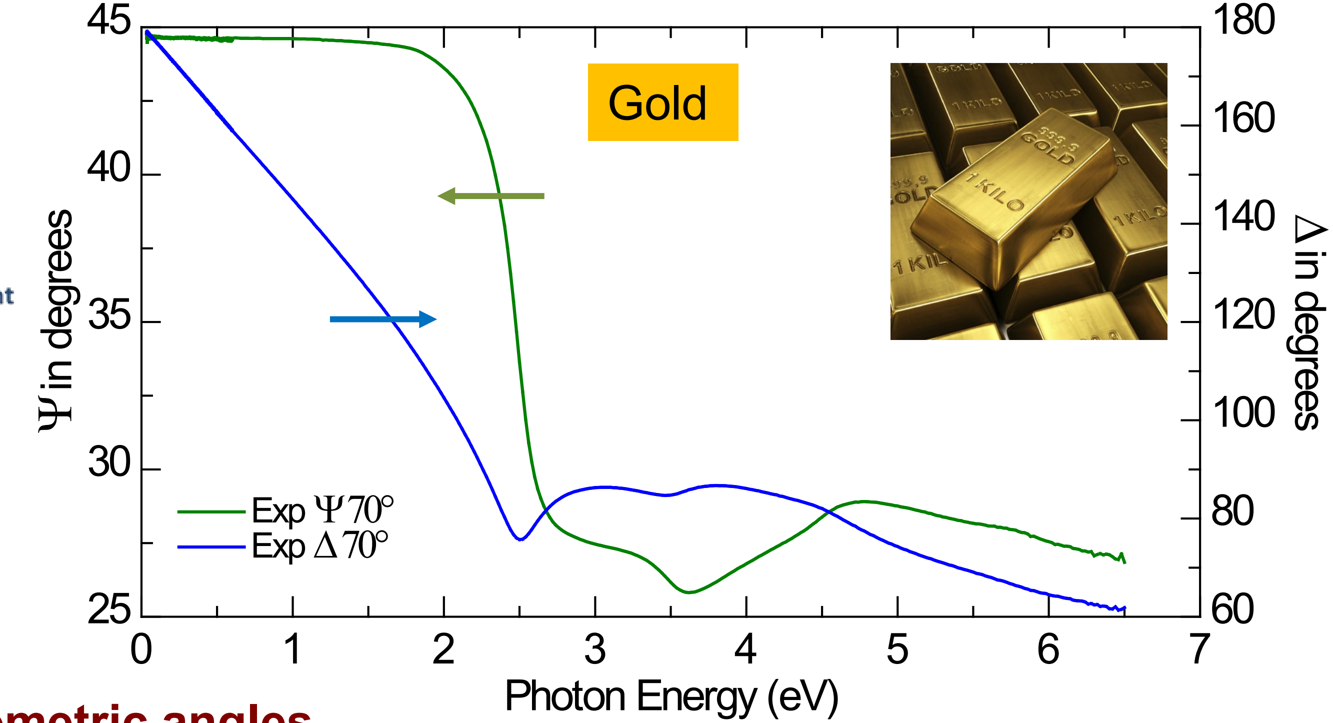
Retarder

Ellipsometry



$$\rho = \frac{r_p}{r_s} = \tan\psi\, e^{i\Delta}$$

www.Jawoollam.com



Fresnel equations

$$r_p = \left(\frac{E_{rp}}{E_{ip}}\right) = \frac{n_1\cos\theta_0 - n_0\cos\theta_1}{n_1\cos\theta_0 + n_0\cos\theta_1}$$

$$r_s = \left(\frac{E_{rs}}{E_{is}}\right) = \frac{n_0\cos\theta_0 - n_1\cos\theta_1}{n_0\cos\theta_0 + n_1\cos\theta_1}$$

Ellipsometric angles

$$\psi = \tan^{-1}\left(\frac{|r_p|}{|r_s|}\right) = \tan^{-1}\left(\frac{|r_p|}{|r_s|}\right) = \tan^{-1}\left[\left(\frac{R_p}{R_s}\right)^{\frac{1}{2}}\right]$$

Dielectric function

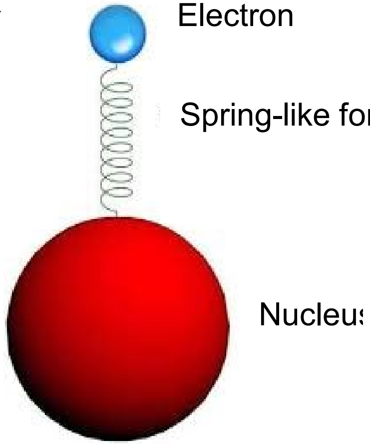
$$\langle\varepsilon\rangle = \sin^2\theta\left[1 + \tan^2\theta\left(\frac{1-\rho}{1+\rho}\right)^2\right]$$

$$n = \sqrt{\varepsilon}$$

Dielectric function model: Lorentz

Equation of motion:

$$m\frac{d^2\vec{x}}{dt^2} + m\gamma\frac{d\vec{x}}{dt} + m\omega_0^2\vec{x} = -e\vec{E}_0 e^{-i\omega t}$$



Solution:

$$\vec{x}(t) = \frac{-e\,\vec{E}_0/m}{\omega_o^2 - \omega^2 - i\gamma\omega} e^{-i\omega t}$$

Dipole moment:

$$\vec{P} = n\vec{p}, \qquad \vec{p} = -e\vec{x},$$

$$\vec{P} = \varepsilon_0\chi\vec{E}$$

Dielectric function:

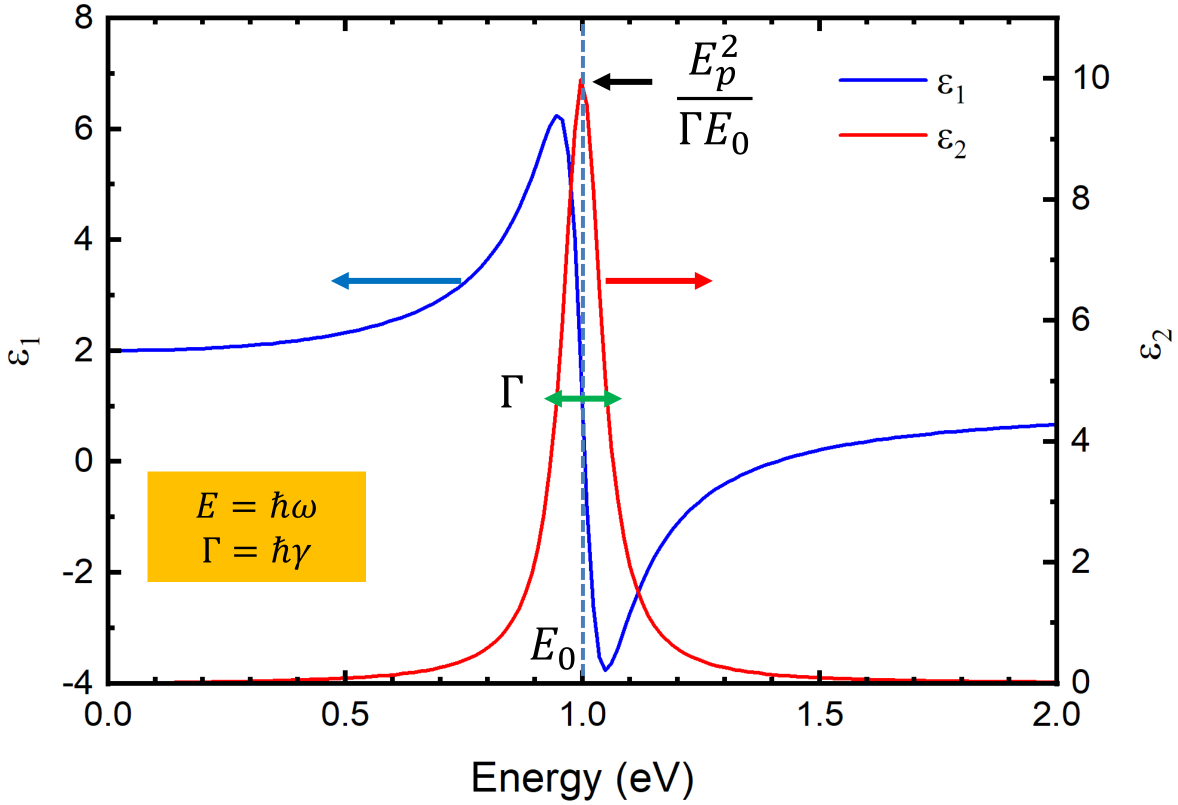
$$\omega_p^2 = \frac{ne^2}{\varepsilon_0 m_0}$$

$$\varepsilon(\omega) = 1 + \frac{\omega_p^2(\omega_0^2 - \omega^2)}{(\omega_o^2 - \omega^2)^2 + (\gamma\omega)^2} + i\frac{\gamma\omega\omega_p^2}{(\omega_o^2 - \omega^2)^2 + (\gamma\omega)^2}$$

$$\varepsilon = 1 + \chi$$

Applications:

- Electronic interband transitions
- Lattice vibrations

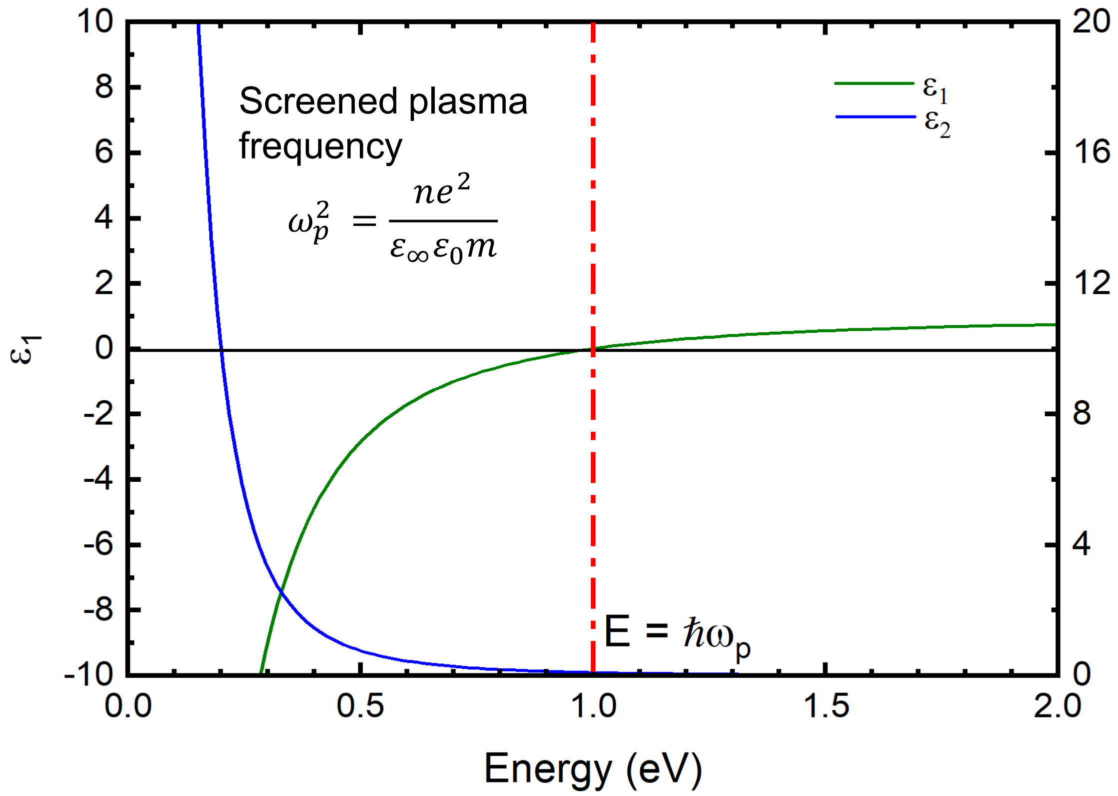


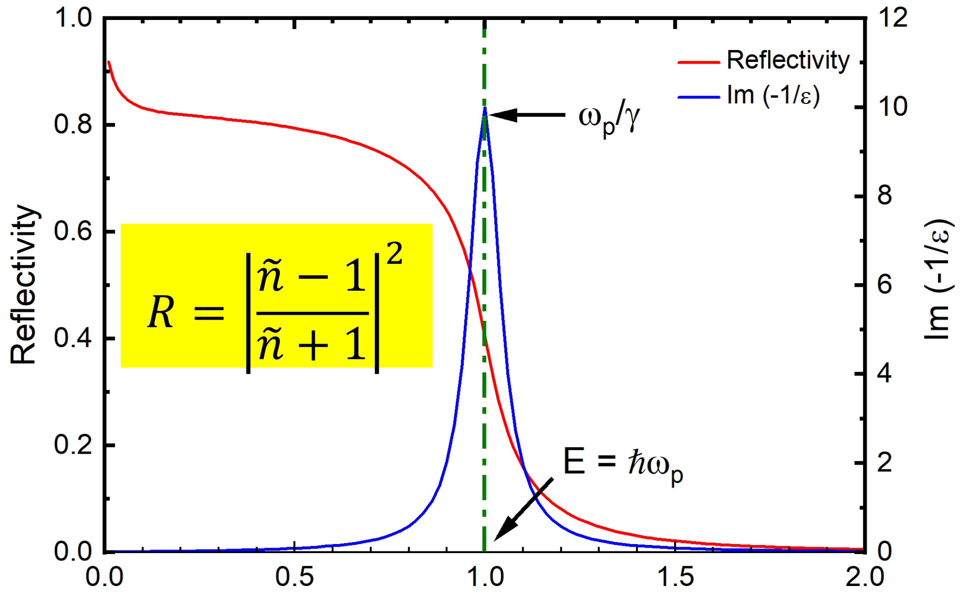
Dielectric function model: Drude

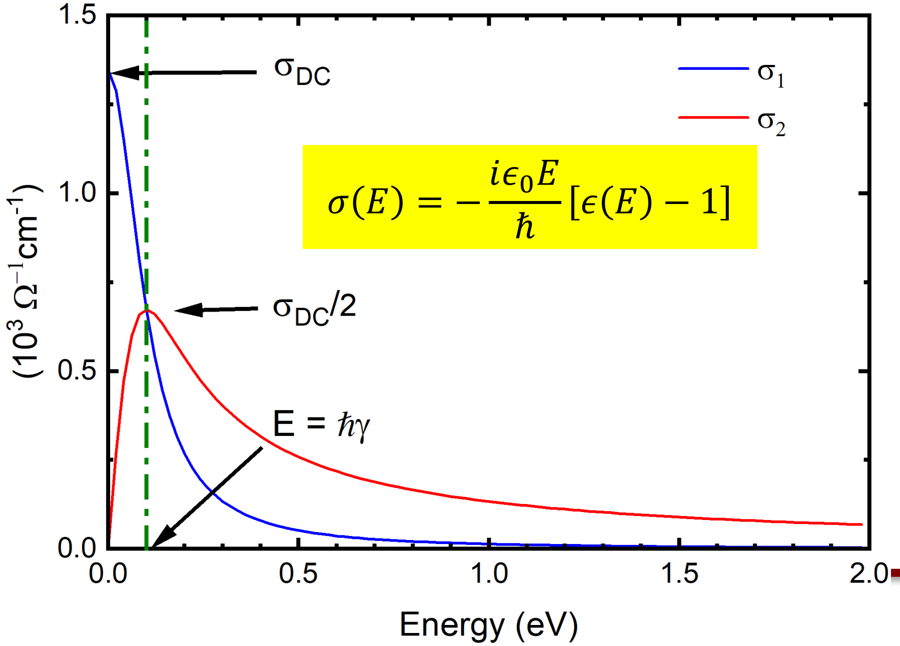
$$m\frac{d^2x}{dt^2} + m\gamma\frac{dx}{dt} = -eE_0e^{-i\omega t}$$

Application: Free carriers:
- Metals
- Doped semiconductors

$$\varepsilon(\omega) = 1 - \frac{\omega_p^2}{\omega^2 + i\gamma\omega} = 1 - \frac{\omega_p^2}{\omega^2 + \gamma^2} + i\frac{\omega_p^2\gamma}{\omega(\omega^2 + \gamma^2)}$$



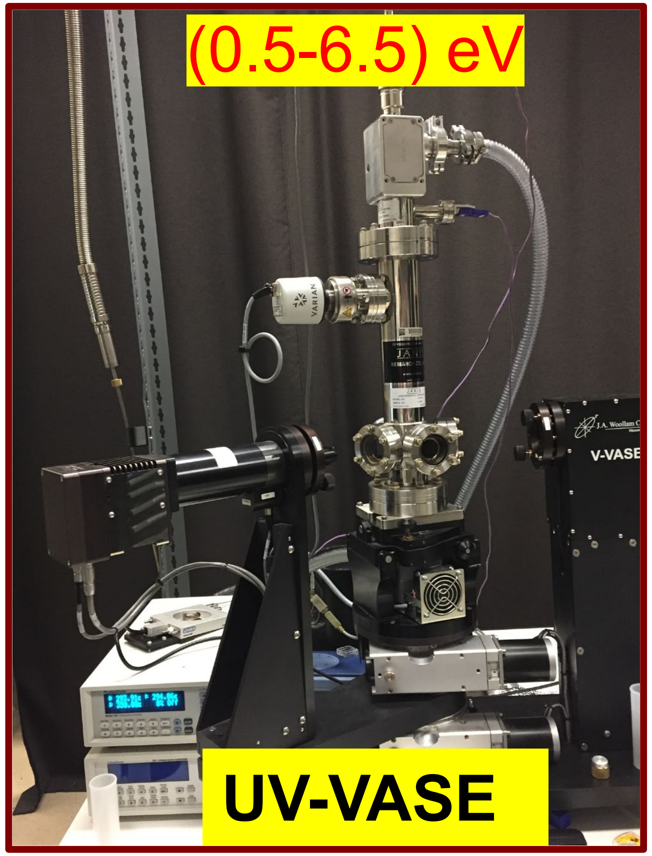


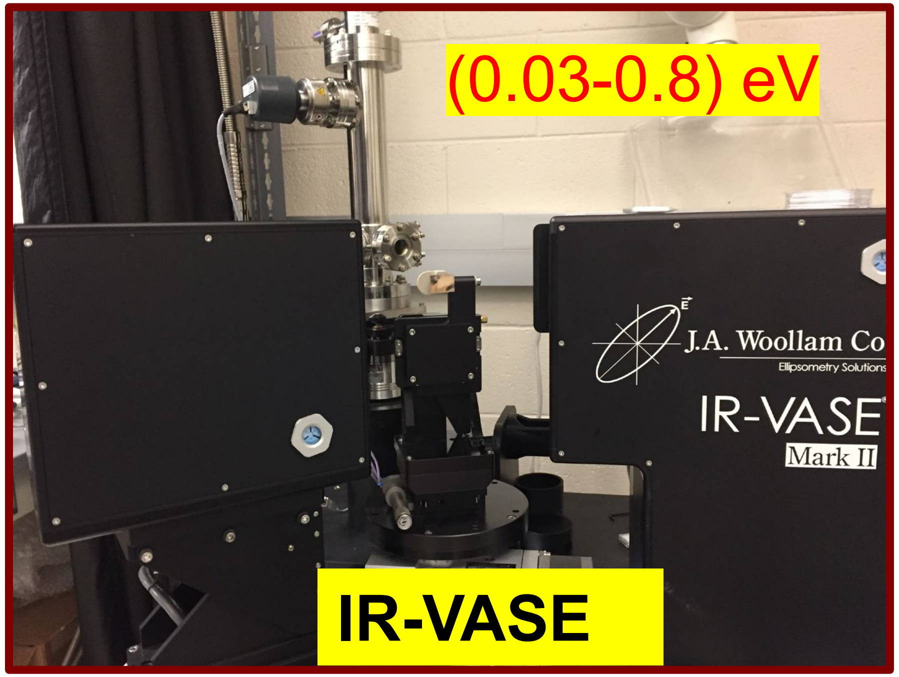


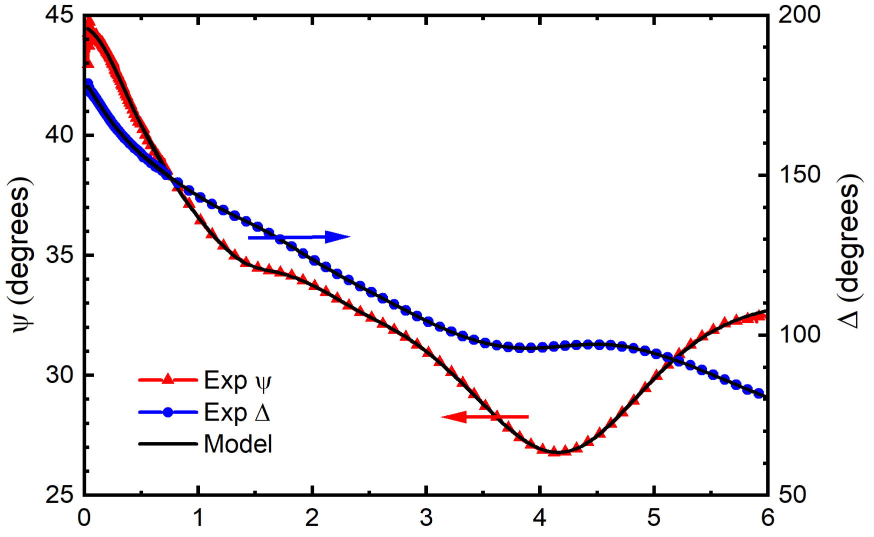
Experimental setup

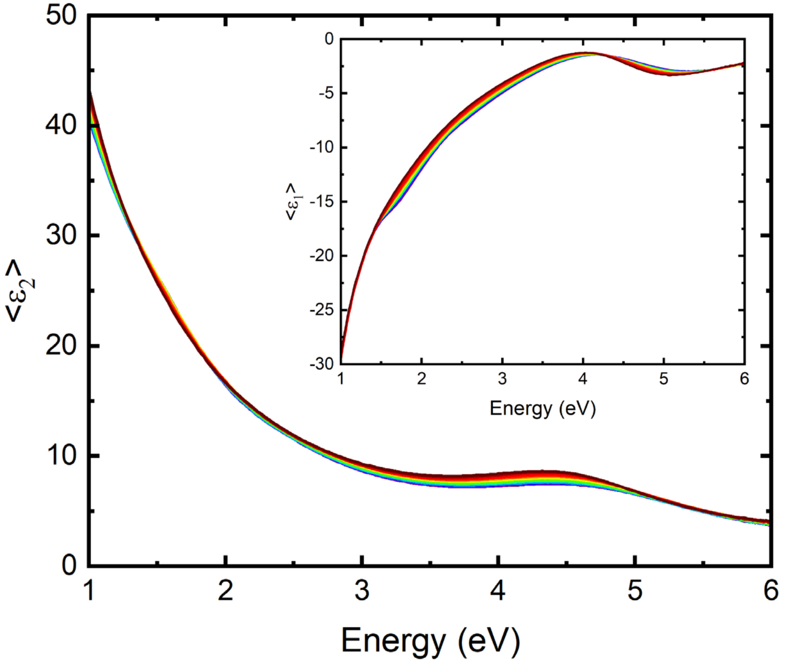
Sample:
Bulk single-crystalline Ni (100)
Measurement sequence

- Cleaning (keeping at 770 K for 12 hours)
- VASE measurements from 770 to 77 K
- VASE measurements from 77 to 770 K
- Cleaning in FTIR-VASE
- Identical measurements with FTIR-VASE







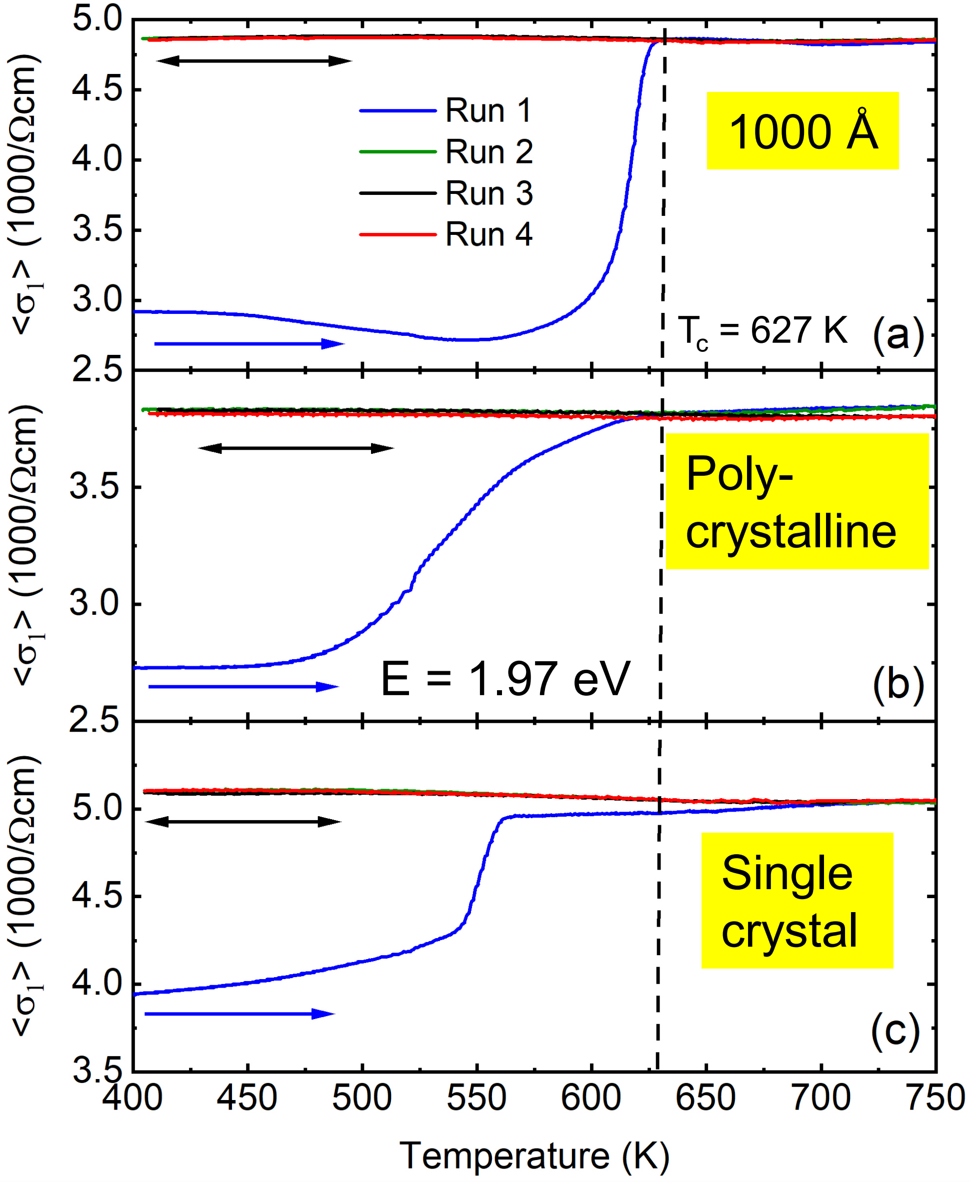


Anomaly in pseudo-conductivity

Anomaly occurs as the temperature rises.
Possible explanations:

- Magnetization
  - Reversible ❌
- Bulk crystal structure
  - Grain growth ❌
- Surface overlayers
  - Adsorbed overlayers ✔

Heating the sample at 770 K seems to be the best way of cleaning



Merging data

VASE windows: Quartz
FTIR windows: Diamond

- Windows change the polarization state of light under stress (retarder)
- Windows are calibrated by default for only the calibration sample SiO_2/Si.
- Windows should be calibrated for the desired sample.

$$\delta(\lambda) = \frac{a_1}{\lambda}\left(1 + \frac{a_2}{\lambda^2} + \frac{a_3}{\lambda^3}\right)$$

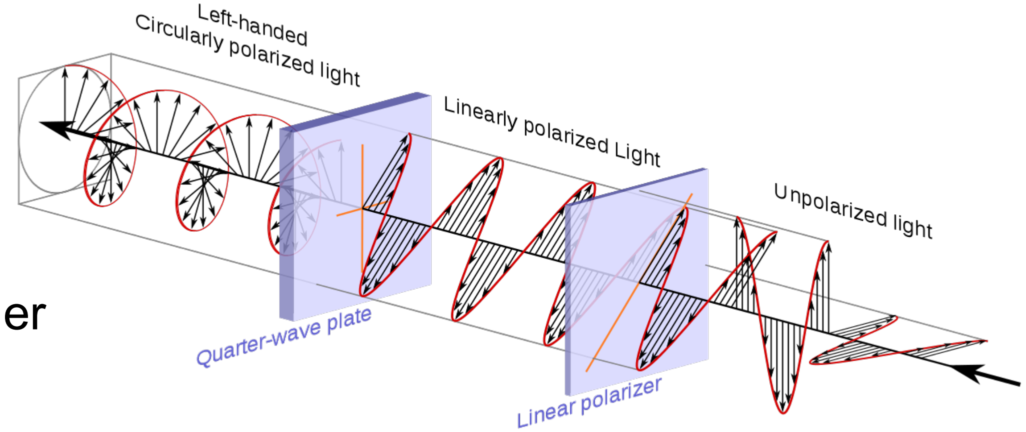
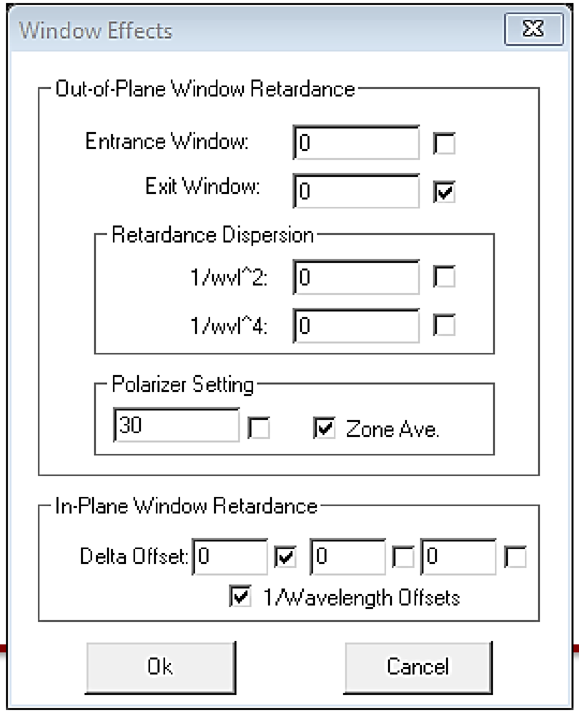
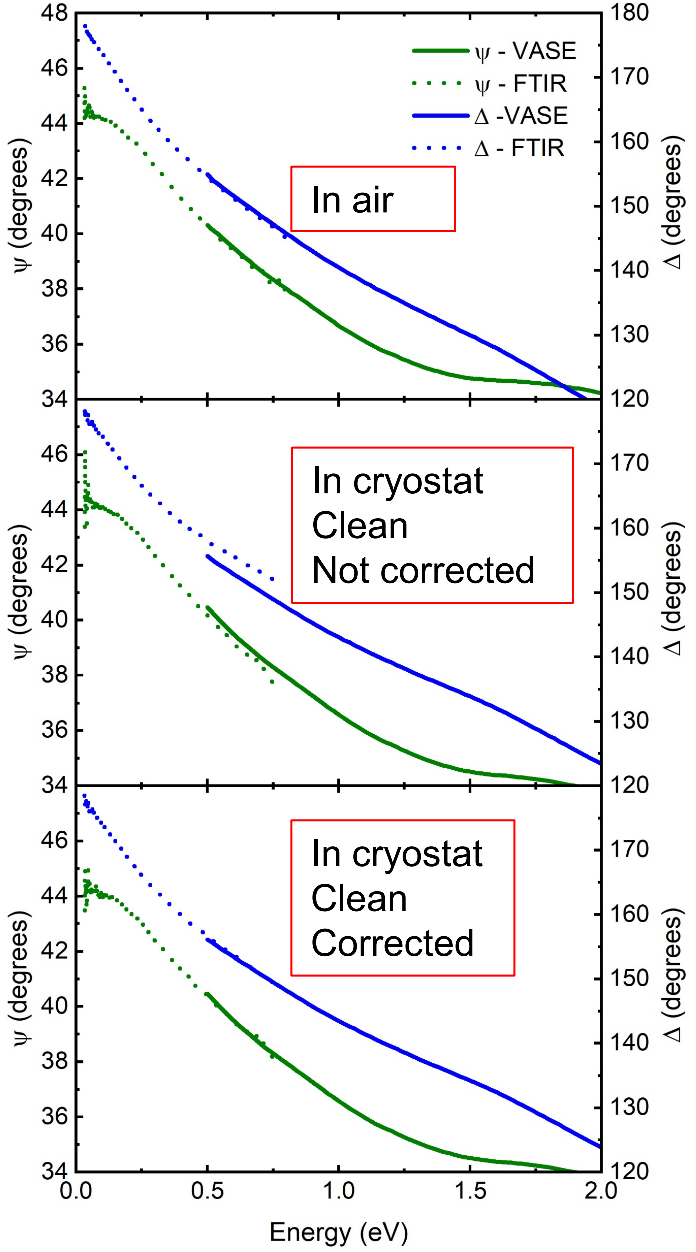


Figure from Wikipedia, public domain





Dielectric function

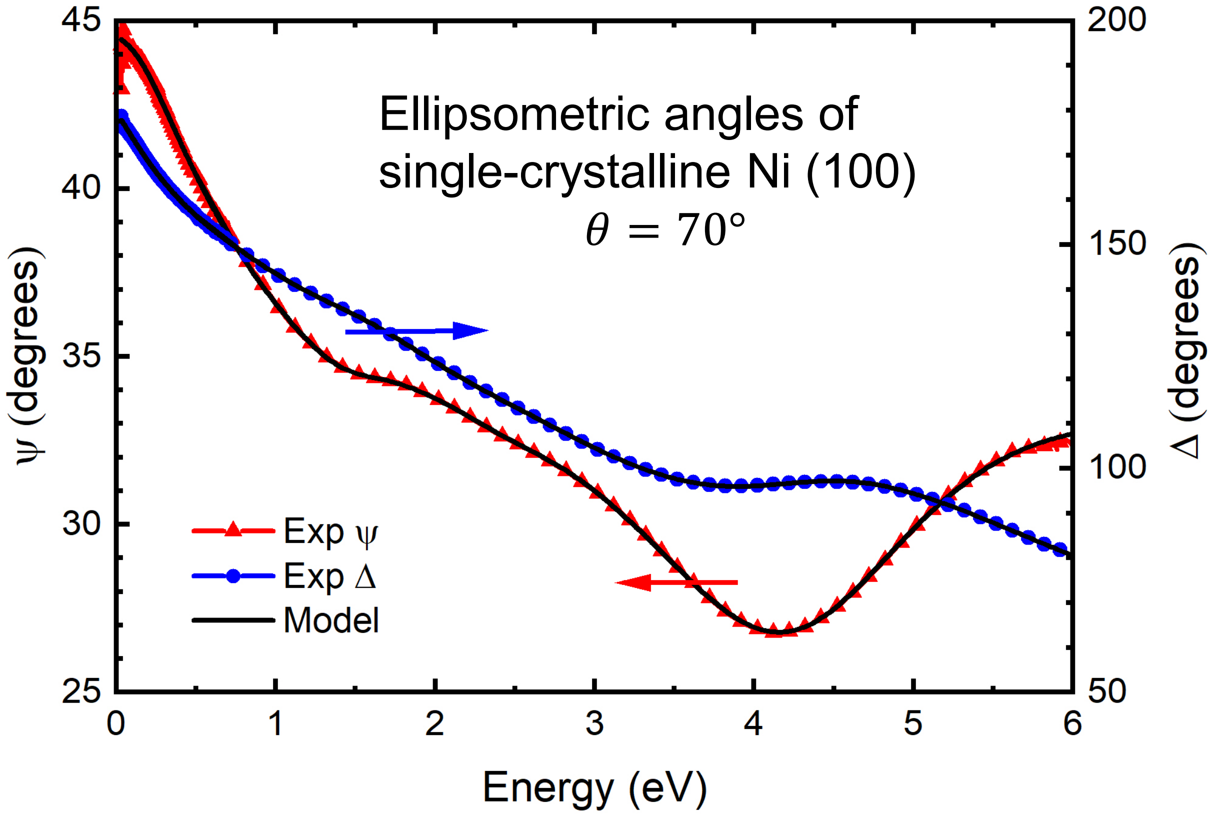
$$Ni^{28} = [Ar]3d^{9.4}\,4s^{0.6}$$

0.6 Electron
0.6 Hole

$$\epsilon(E) = 1 - \sum_{i=1}^{2} \frac{E_{p,i}^2}{E(E + i\Gamma_i)} + \sum_{j=1}^{4} \frac{A_j E_{o,j}^2}{E_{0,j}^2 - E^2 - i\Gamma_j E}$$

	A	E_p (eV)	E_0 (eV)	Γ (eV)	σ_0 ($1/\Omega$cm)
Drude 1 (d)		12.1		2.91	6,766
Drude 2 (s)		4.81		0.0403	77,200
Lorentz 1	1.83		1.57	0.847	
Lorentz 2	0.138		2.58	0.888	
Lorentz 3	2.42		4.77	2.08	
Lorentz 4	1.91		12.7	6.01	

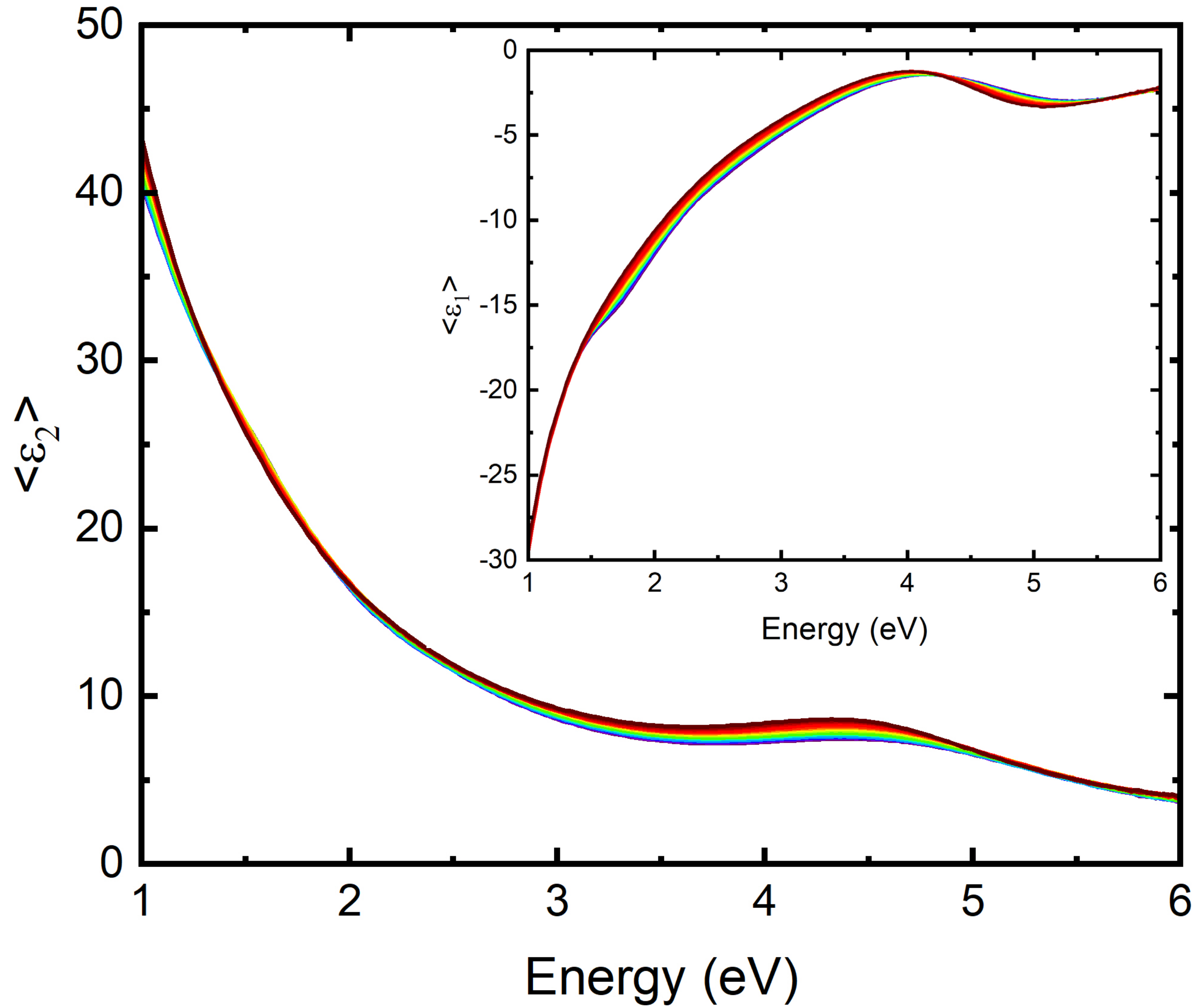
All data are corrected for 20 Å of surface roughness.



- Merging correction reduces the number of needed oscillators.
- Two Drude terms to account for s- and d- electrons.

Dielectric function at various temperatures

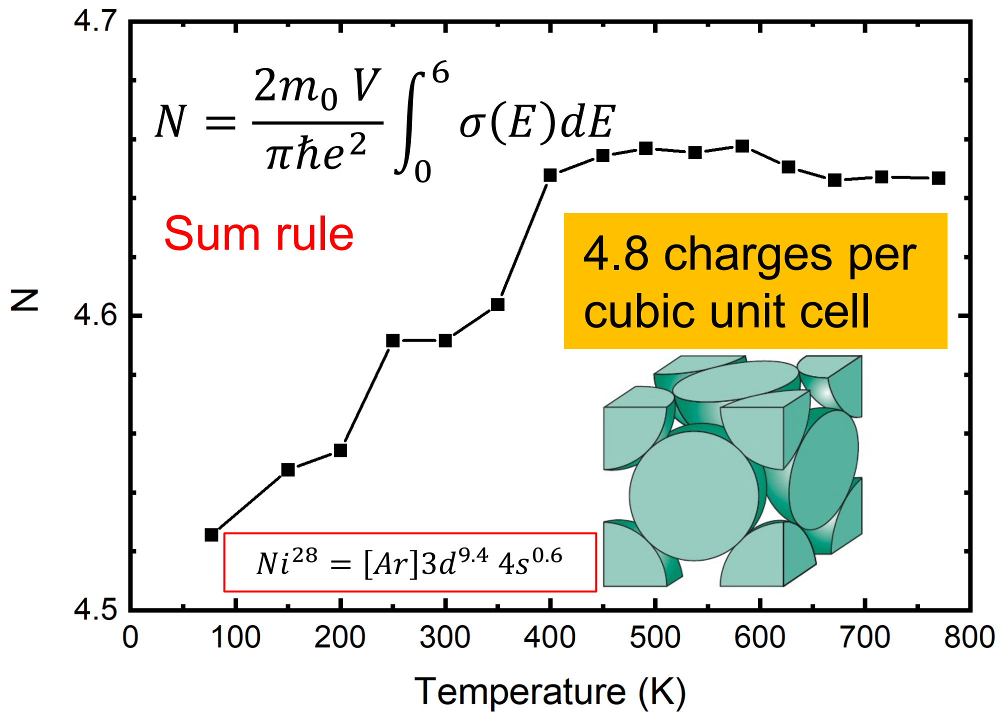
Due to the large free carrier absorption, temperature dependence of the peaks are not visible.

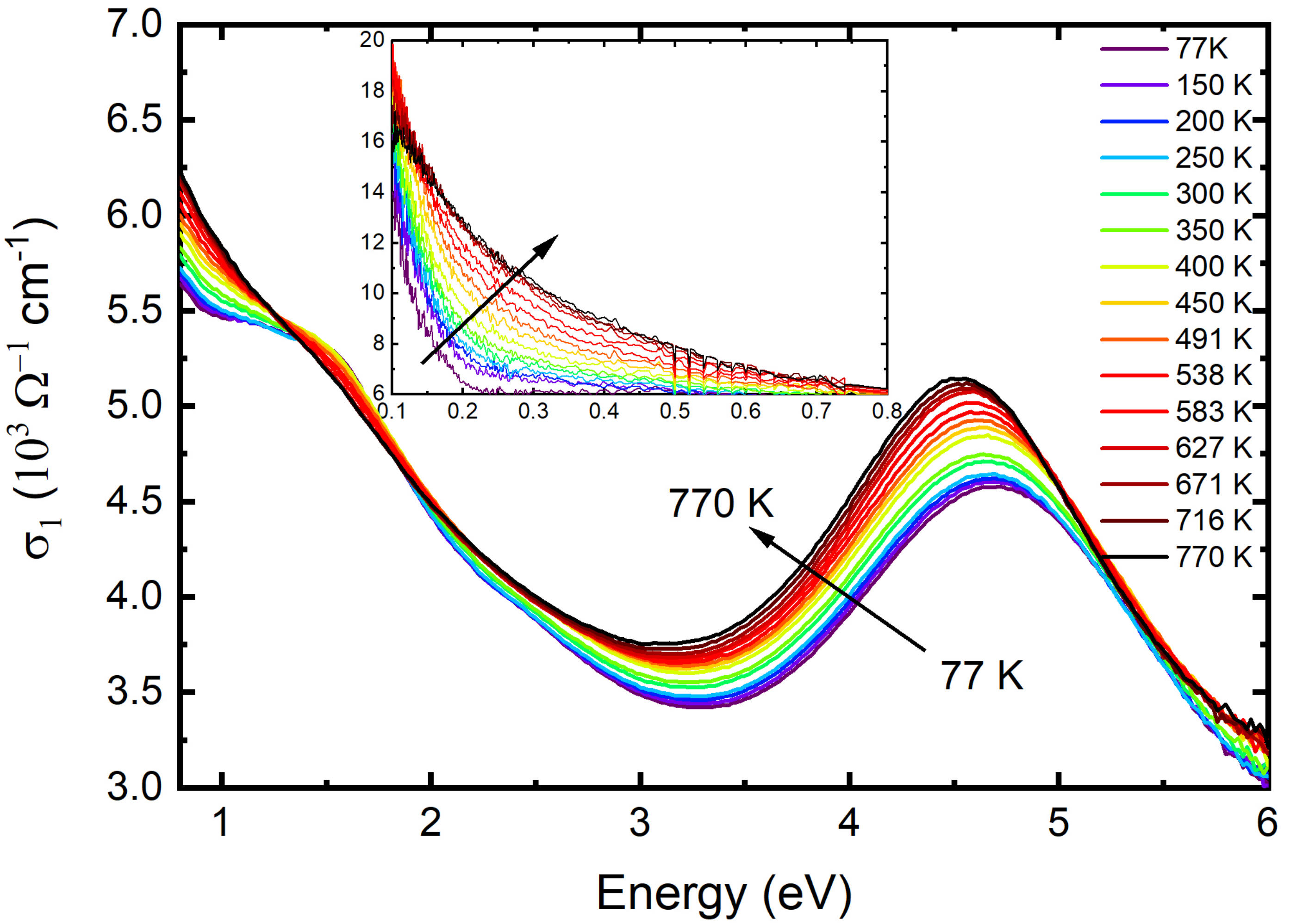


Optical conductivity at various temperatures

- There is a red shift in the main peak at 4.8 eV.
- The small peak at 1.5 eV changes with temperature.
- Drude broadening increases with temperature.

$$\sigma(E) = -\frac{i\epsilon_0 E}{\hbar}[\epsilon(E) - 1]$$





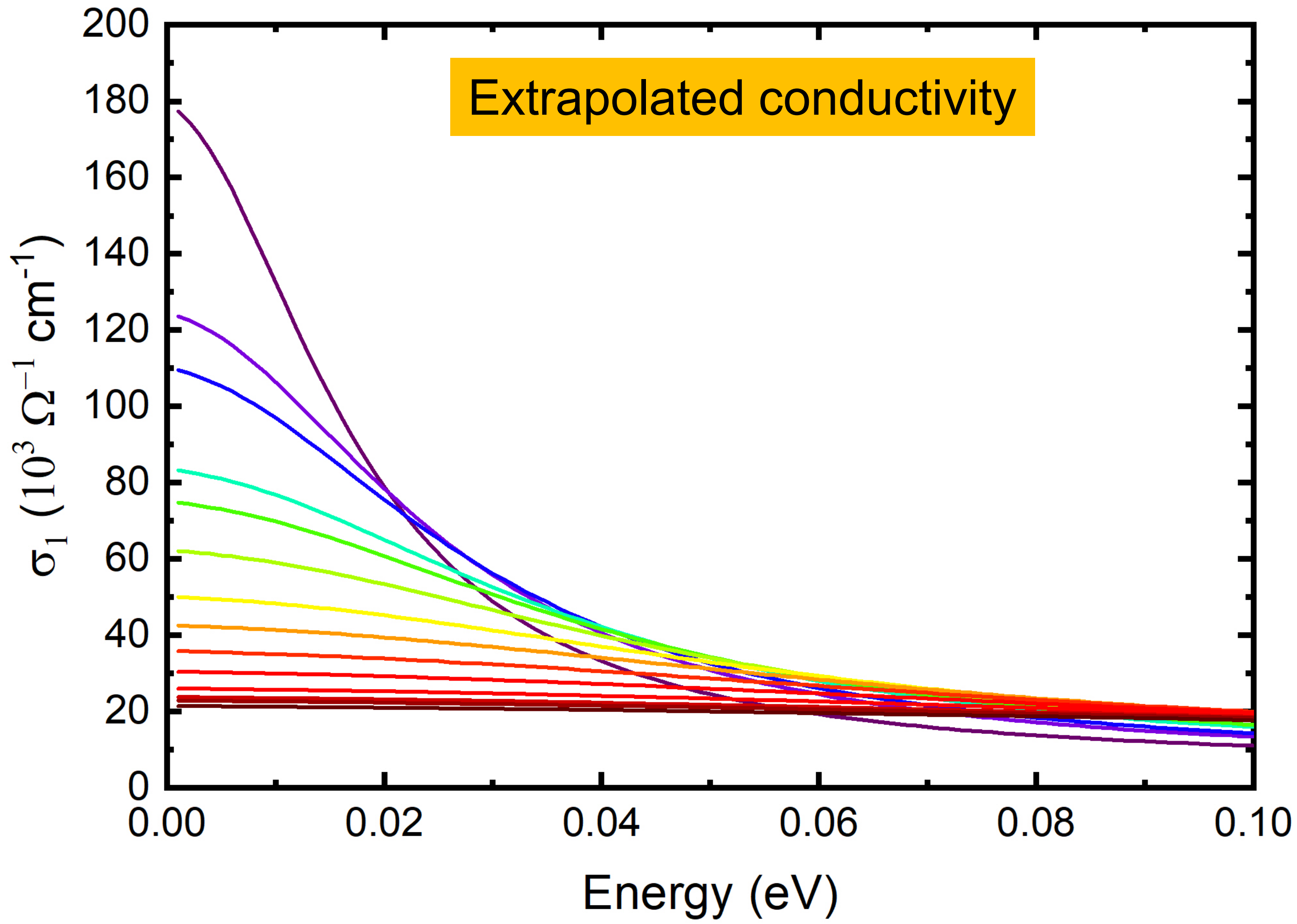
DC conductivity

$$\epsilon(E) = 1 - \sum_{i=1}^{2} \frac{E_{p,i}^2}{E(E + i\Gamma_i)} + \sum_{j=1}^{4} \frac{A_j E_{o,j}^2}{E_{0,j}^2 - E^2 - i\Gamma_j E}$$

$$\sigma_{DC} = \sigma(E = 0) = \frac{\epsilon_0}{\hbar}\left(\frac{E_p^2}{\Gamma}\right)$$

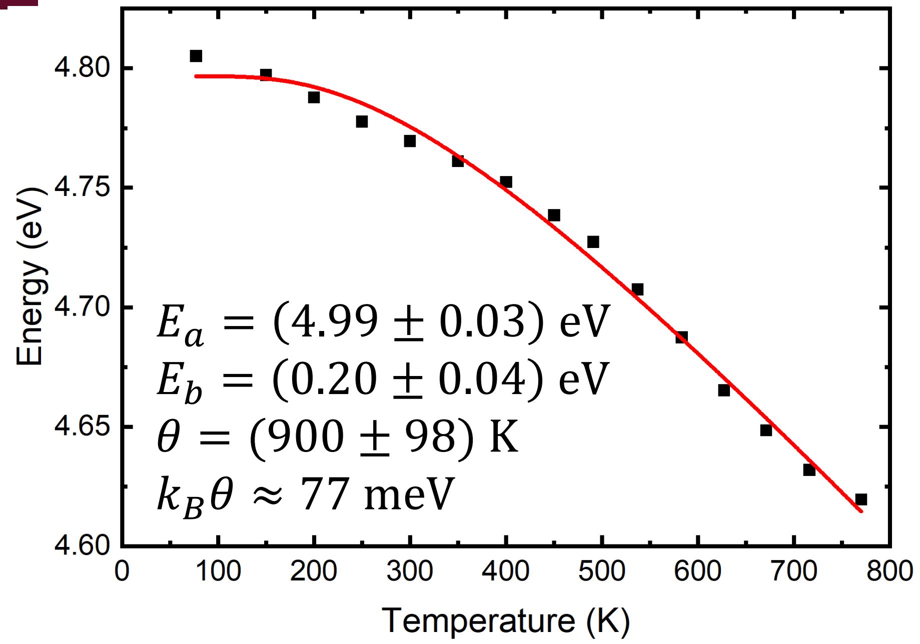
$$\sigma_{total} = \sigma_1 + \sigma_2$$

- Second Drude term behaves like a typical metal (thus s-electrons)
- No significant change with T in d-electrons
- IR ellipsometry measurements underestimates the DC conductivity.
- Far IR measurements might produce better results for DC conductivity.



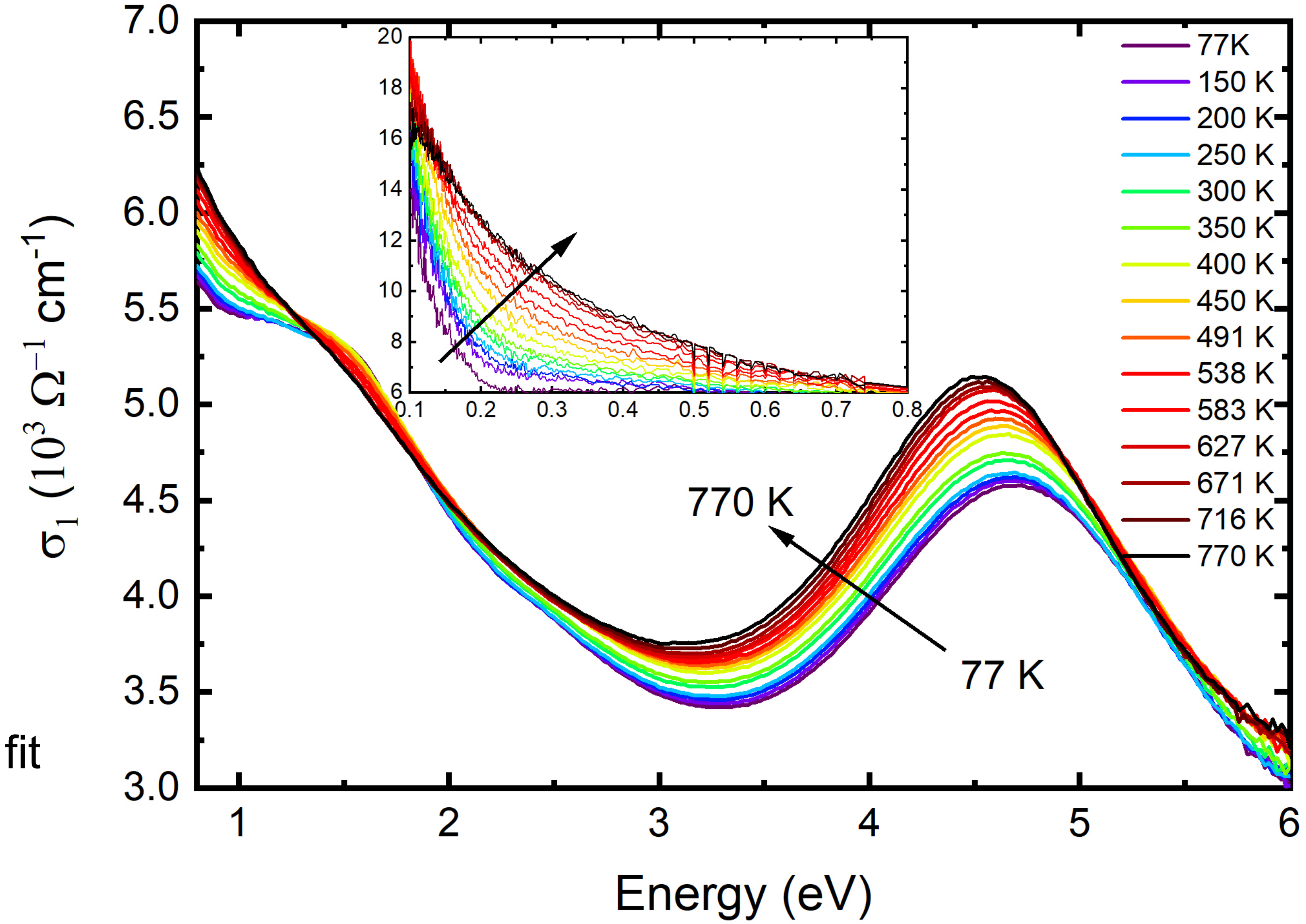
M. J. Laubitz, T. Matsumura, and P. J. Kelly, Can. J. Phys. **54**, 92 (1976).

Energy of the main peak



$$E(T) = E_a - E_b\left(1 + \frac{2}{e^{\frac{\theta}{T}} - 1}\right)$$

Bose-Einstein fit



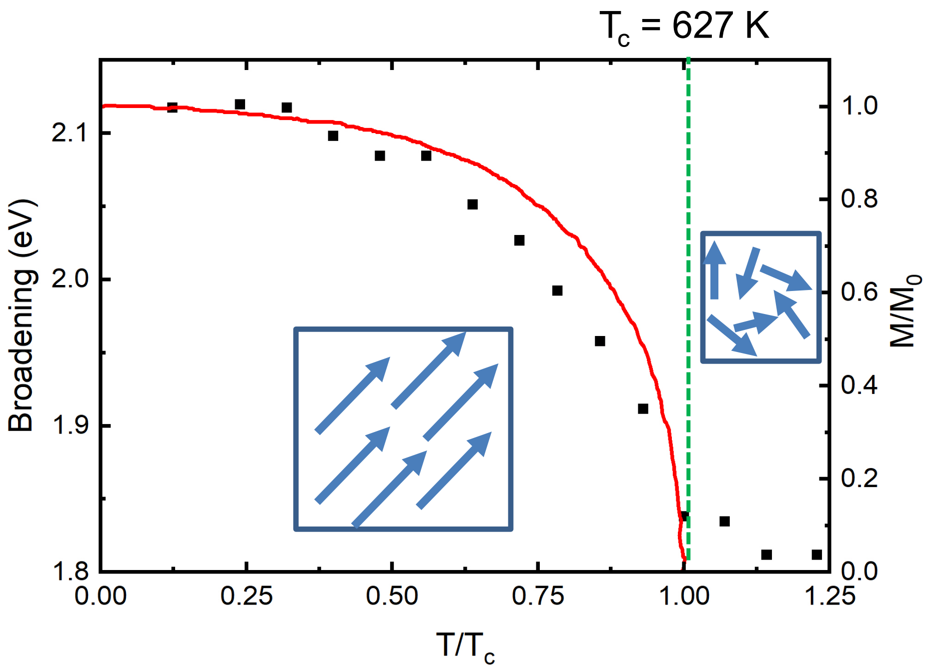
E_a: Unrenormalized energy

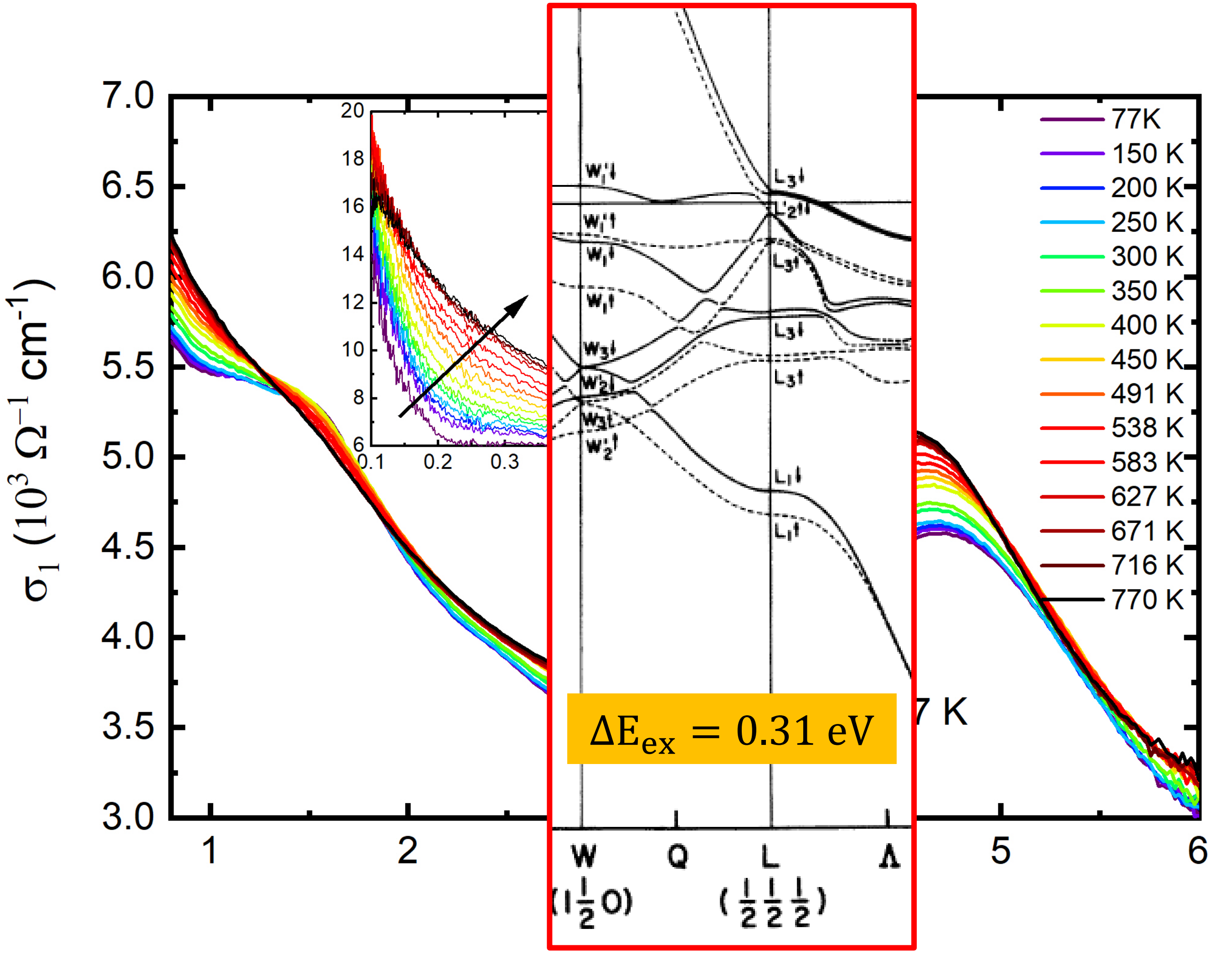
E_b: Coupling energy

$k_B\theta$: Effective energy

- The main peak is due to the transitions at the L-point of the BZ.
- The peak broadens by scattering with magnons.

Broadening of the main peak



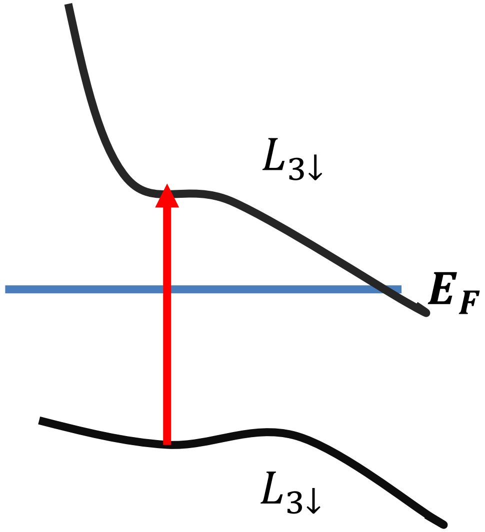


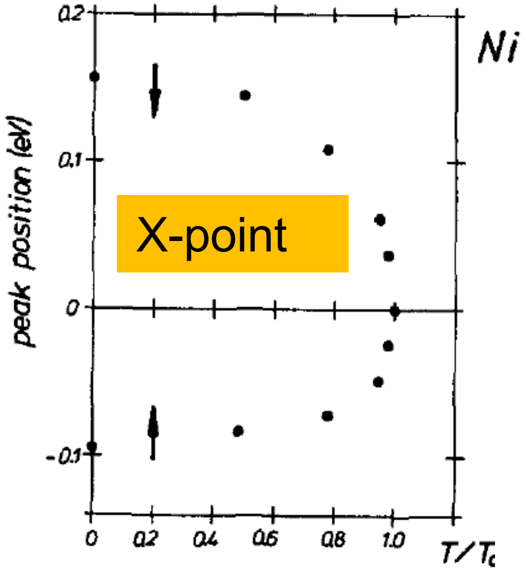
- Broadening and the spontaneous magnetization behave the same way.
- Reduction of the width of the peak is interpreted as the ΔE_{ex} of the d-band.

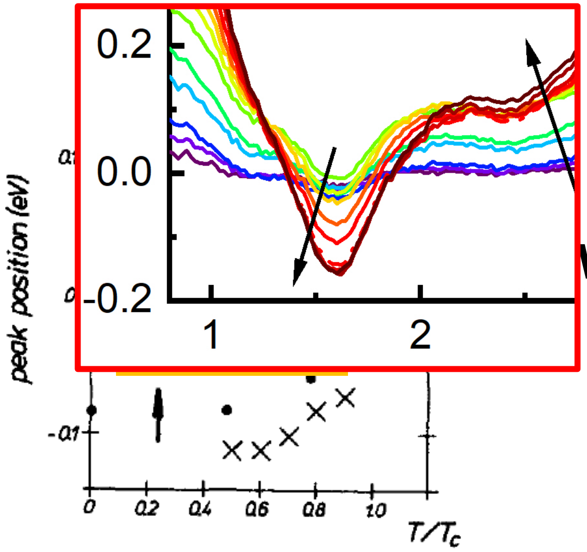
C. S. Wang and J. Callaway, Phys. Rev. B **9**, 4897 (1974)
M. Shiga and G. P. Pells, J. Phys. C. 2, 1847 (1969)
D. E. Eastman et al. Phys. Rev. Lett. **40**, 1514 (1978)

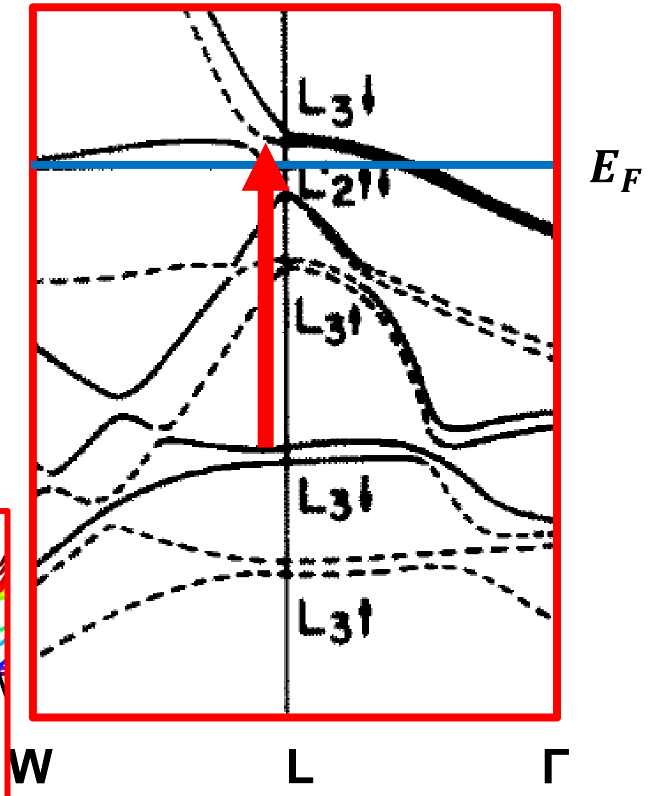
Small absorption peak

- Amplitude of the main peak increases.
- Amplitude of the small peak decreases and stays constant above T_c = 627 K.
- The energy of the small peak stays constant.
- Small peak $L_{3\downarrow} \rightarrow L_{3\downarrow}$









W. Borgiel, W. Nolting, and M. Donah, Solid. State. Commun. **72**, 825 (1989)
T. Greber, T. J. Kreutz, and J. Osterwalder, Phys. Rev. Lett., **79**, 4468 (1997)

Resistivity as a function of temperature

- Qualitative agreement between optical and electrical measurements.
- Scattering by magnons is dominant at high temperatures.
- Optical measurements are sensitive to surface conditions but not electrical measurements.

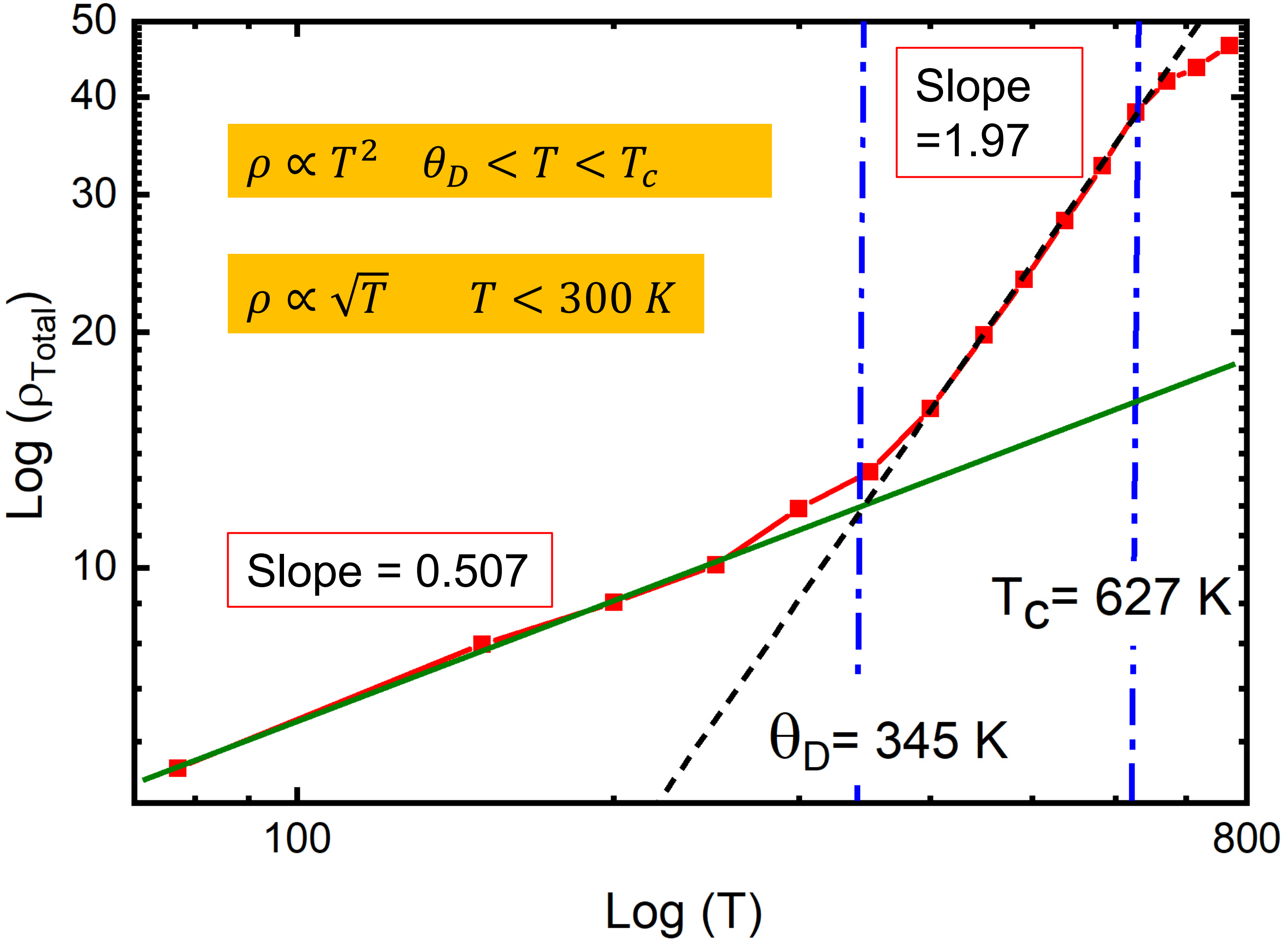
$$\rho_{total} = \rho_{imp} + \rho_{e-e} + \rho_{e-ph} + \rho_{e-mag}$$

$\rho_{e-e} \propto T^2$ very low T

$\rho_{e-ph} \propto T^5$ $T \ll \theta_D$

$\rho_{e-ph} \propto T$ $T > \theta_D$

$\rho_{e-mag} \propto T^2$ $T \gg T_F$

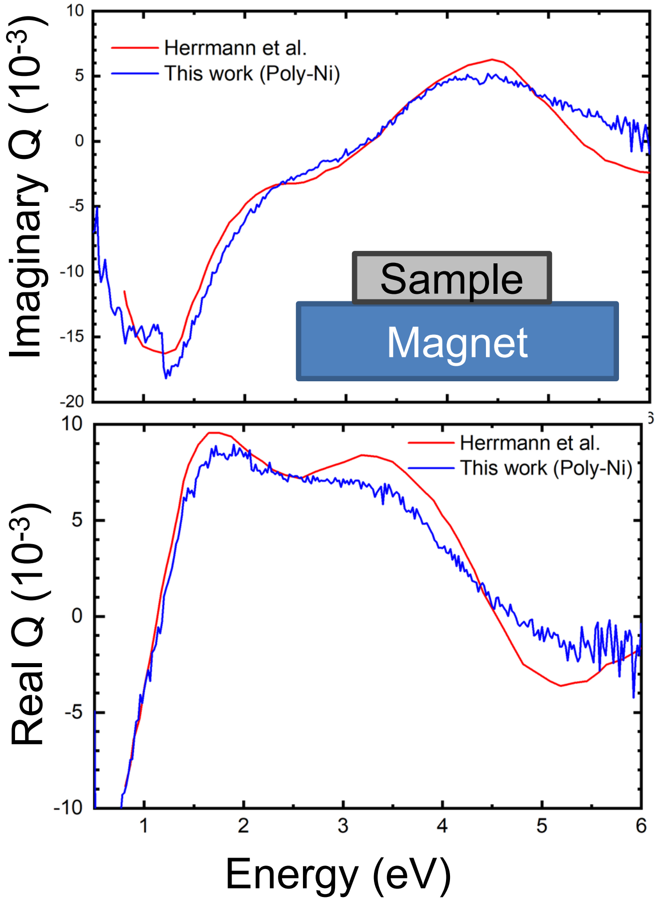


$T_F \approx 10 - 40\ K$

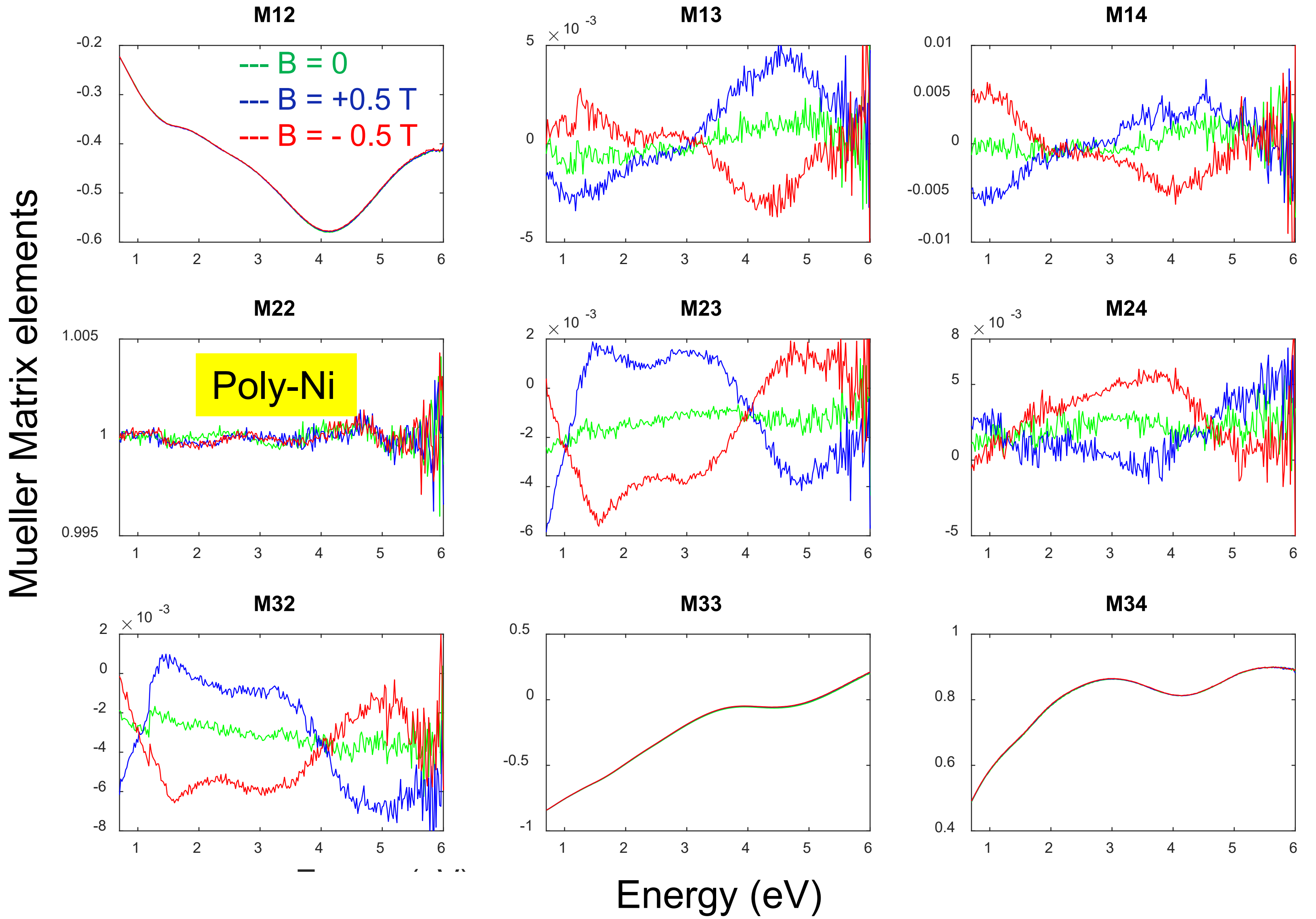
N. V. Volkenshtein, V. P. Dyakona, and V. E. Startsev, Phys. Stat. Sol. (b) **57**, 9 (1973)
B. Raquet, M. Viret, E. Sondergard, O. Cespedes, and R. Mamy Phys. Rev. B **66**, 024433 (2002).

Magneto-optical effects

Voigt parameter $Q = i\frac{\varepsilon_{xy}}{\varepsilon_{xx}}$



This research is still ongoing



Herrmann et l. Phys. Rev. B **73**,134408 (2006)

Summary

- Merging correction produces more realistic model with minimum oscillators.
- FTIR ellipsometry measurements underestimate the conductivity.
- Far IR measurements are required to model the Drude term of metals.
- Bose-Einstein fit to the red shift of the main peak near 4.8 eV indicates scattering by magnons.
- Reduction of the broadening of the main peak is interpreted as a measure of the exchange energy of the d-band at the L-point.
- The small peak is assigned to $L_{3\downarrow} \rightarrow L_{3\downarrow}$ transitions with simultaneous descent of the bands as the temperature rises.
- Temperature dependence of the resistivity also indicates scattering by magnons.

Outlook

- Temperature dependence of Ni thin films as a function of thickness.
- Research on NiPt and other alloys of Ni (tunable Curie temperature).
- Optical properties of single-crystalline Ni (111).
- Ellipsometry studies on Ni in a magnetic field.
- Optical properties of other metals for comparison.
- Comprehensive dielectric tensor to describe both on- and off-diagonal elements.
- Anomalous skin effect (scattering length > penetration depth).

Acknowledgements

Ph.D. committee members

Dr. Stefan Zollner

Dr. Heinrich Nakotte

Dr. Michael Engelhardt

Dr. Jason Jackiewicz

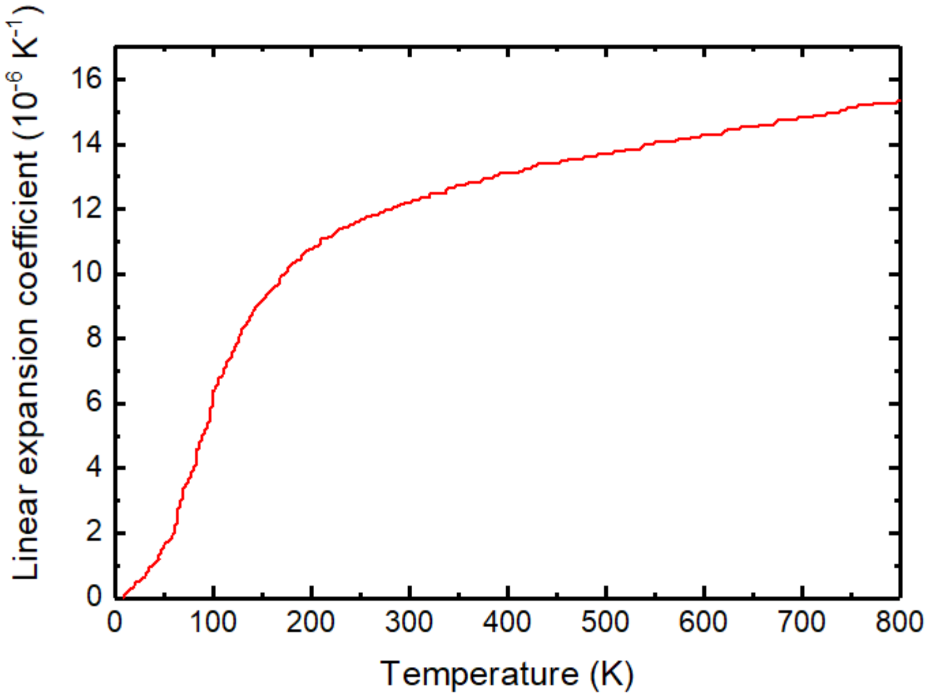
Colleagues

Carola Emminger

Nuwanjula Samarasingha

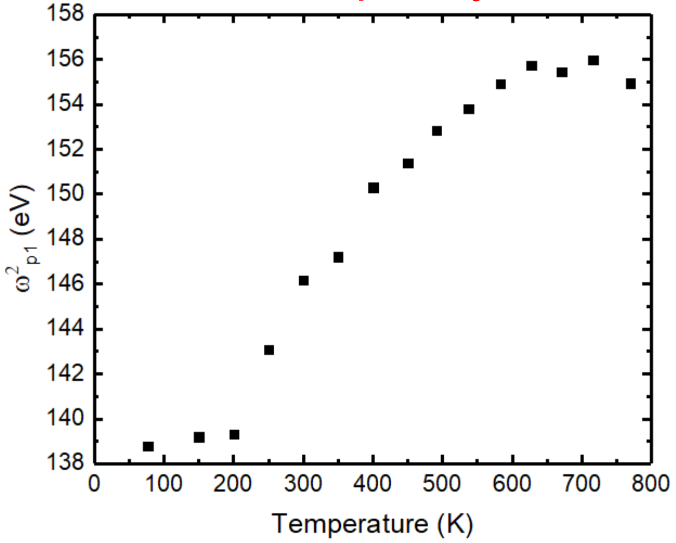
Rigo Carrasco

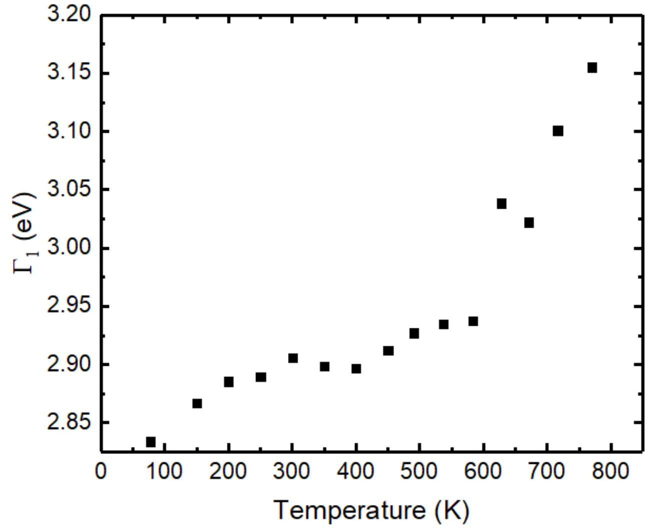
Physics faculty members

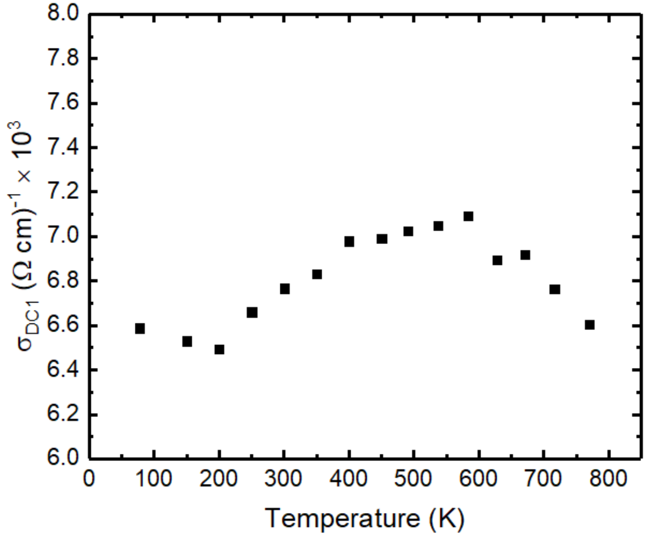

Linear expansion coefficient (10^-6 K^-1)
0
2
4
6
8
10
12
14
16
0
100
200
300
400
500
600
700
800
Temperature (K)

Plasma frequency | Scattering rate | DC Conductivity

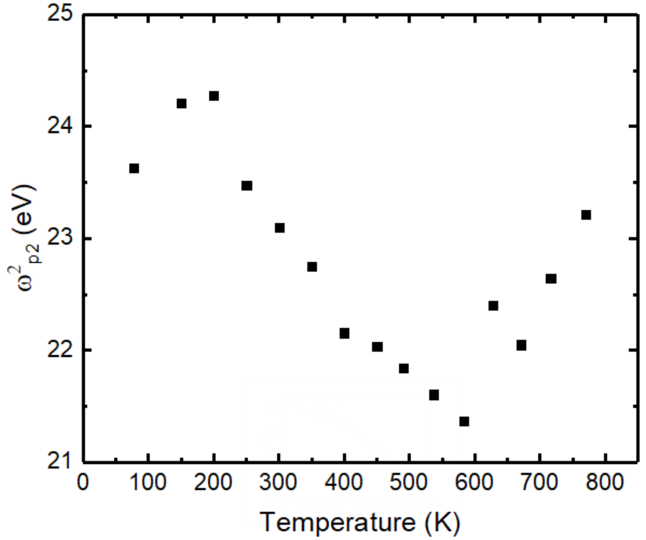
Drude 1

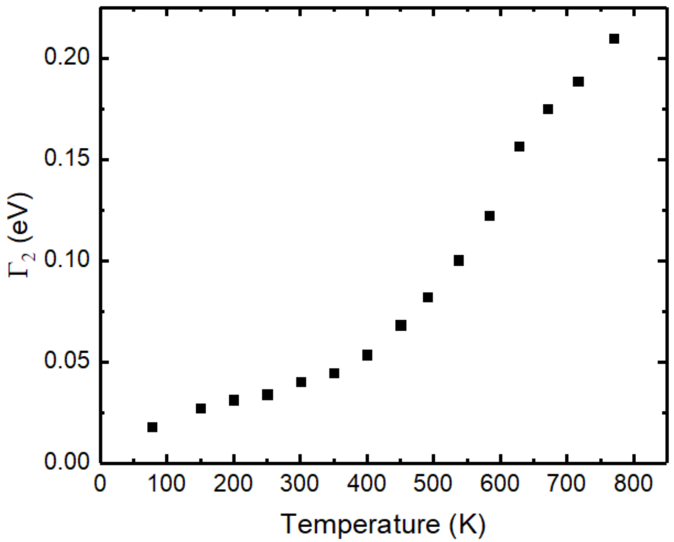


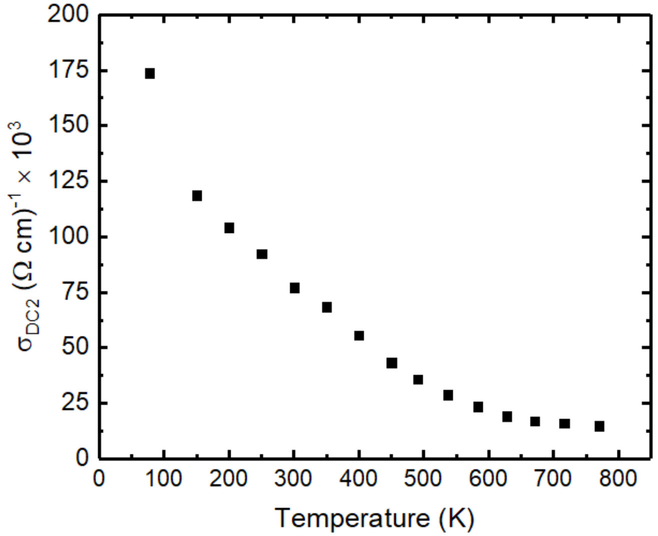




Drude 2





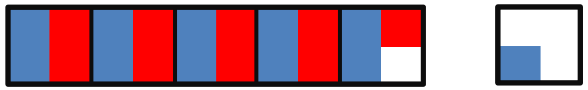


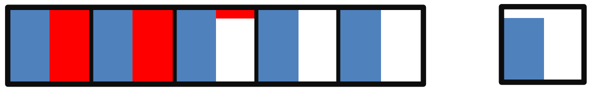
Electronic configuration of Ni

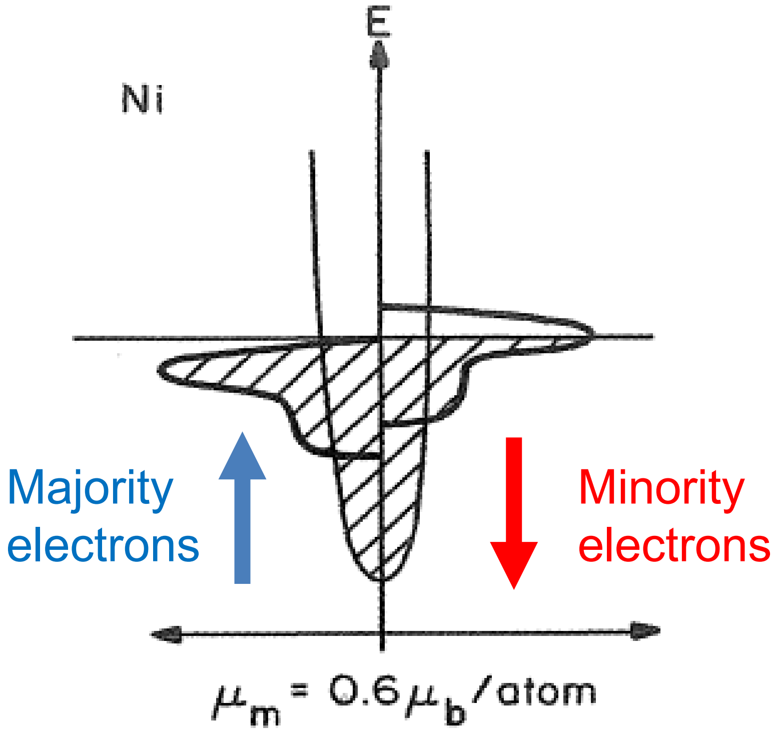
$Ni^{28} = [Ar]3d^{9.4}\ 4s^{0.6}$

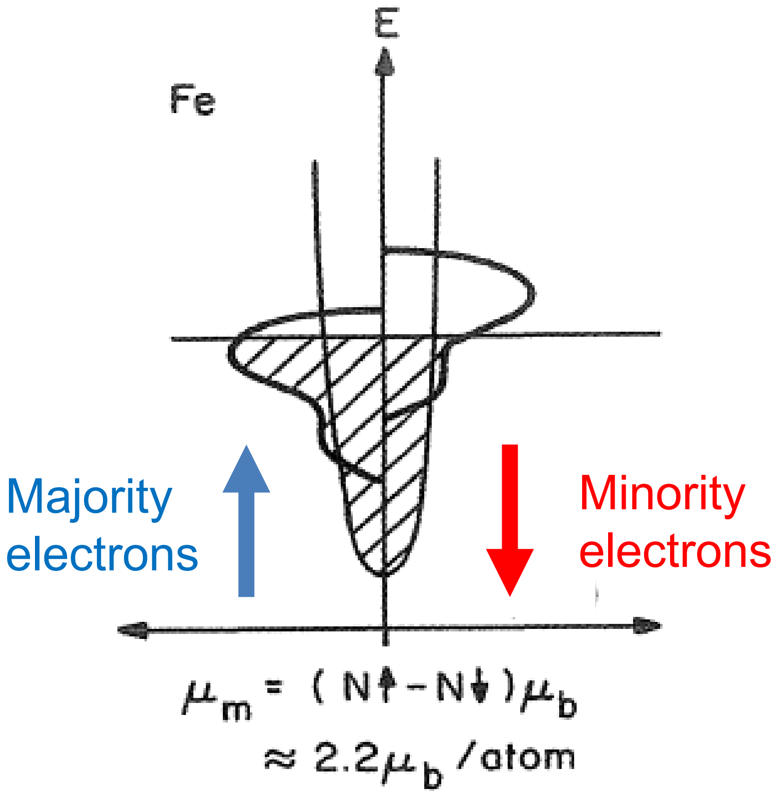
$Fe^{26} = [Ar]3d^{7.05}\ 4s^{0.95}$

3d 4s

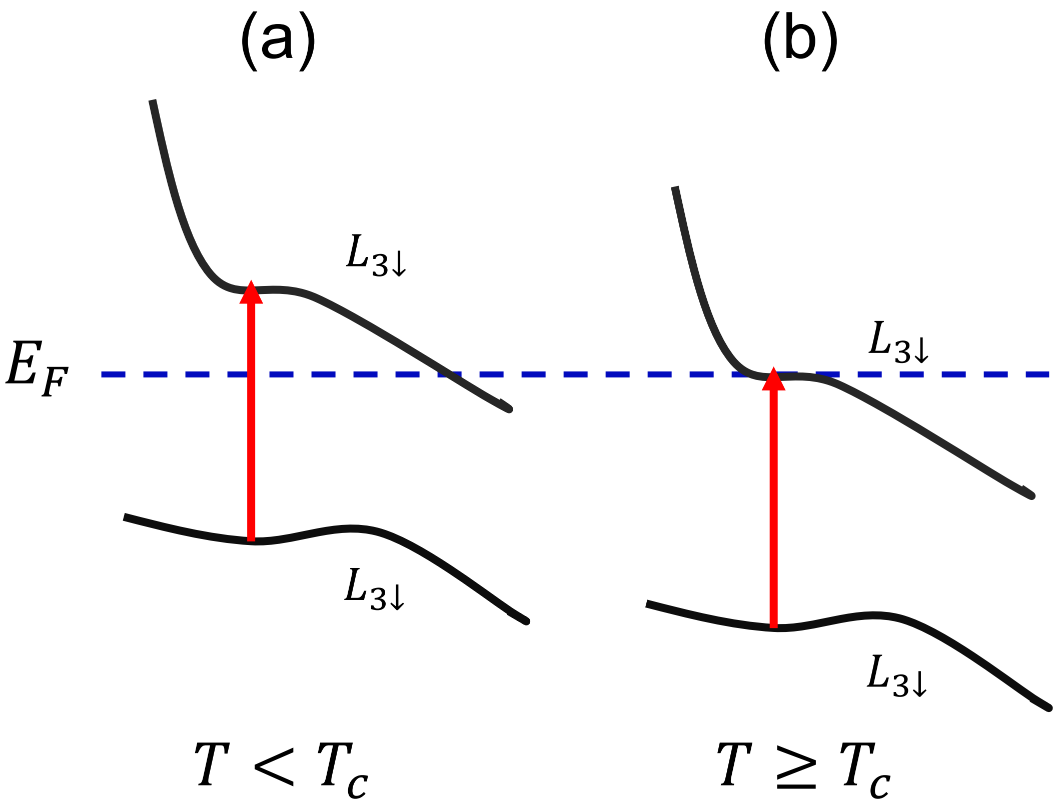




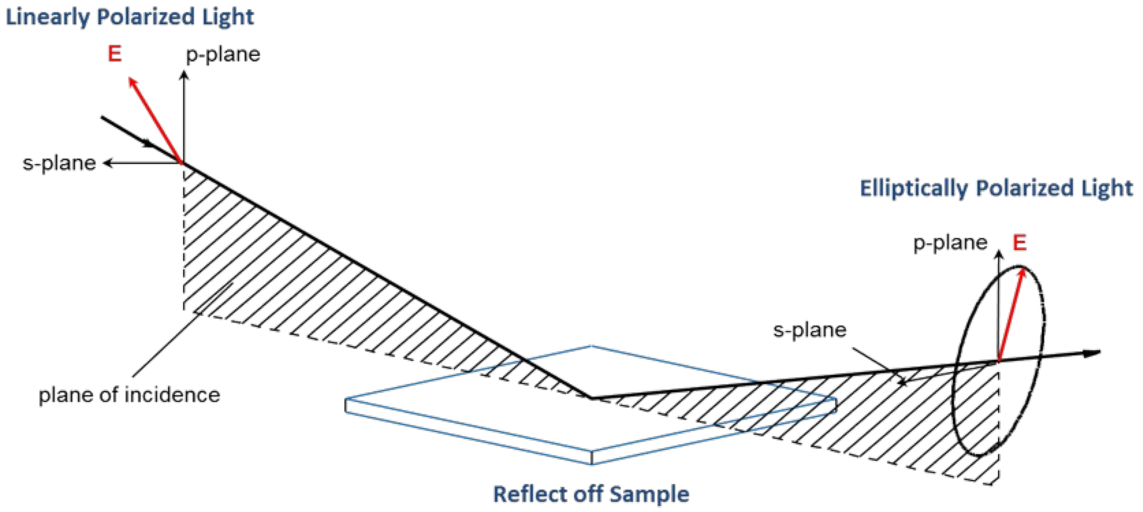


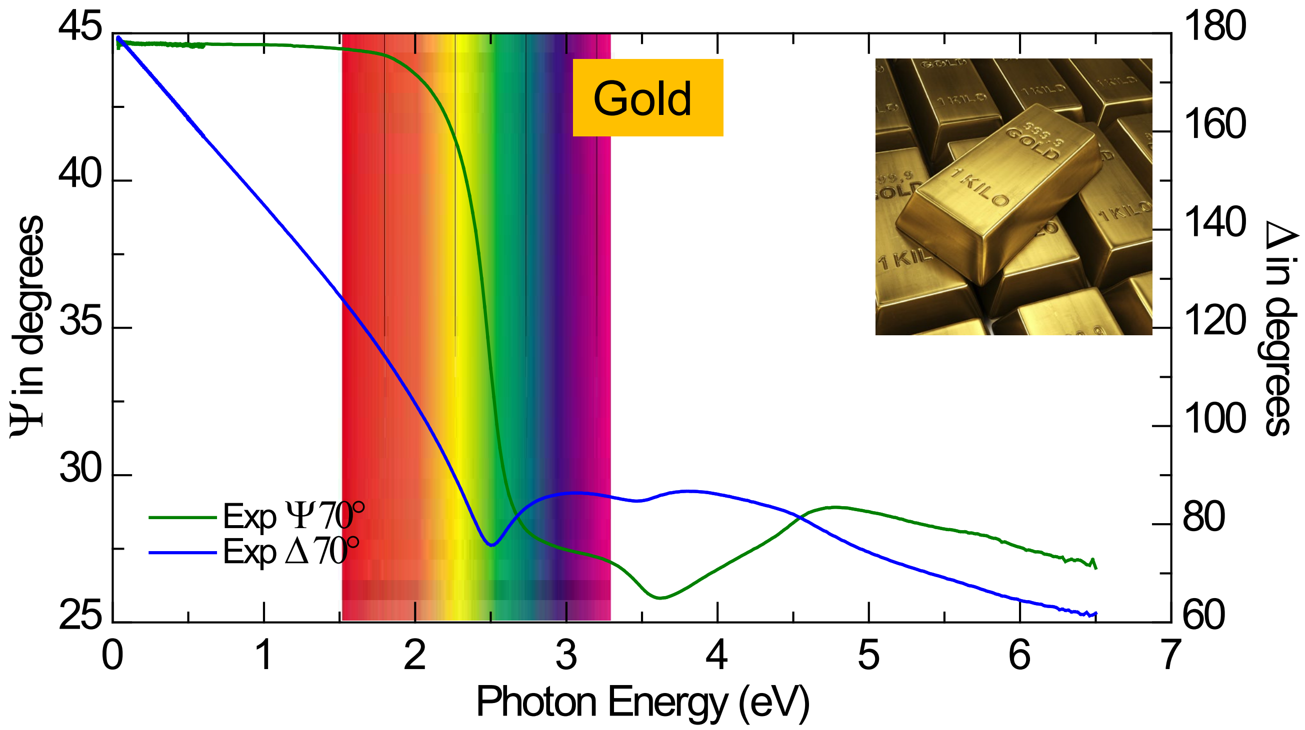


R. C. O'Handley, *Modern Magnetic Materials: principles and applications*, (Wiley, 2000)


(a)
(b)
$L_{3\downarrow}$
E_F
$L_{3\downarrow}$
$L_{3\downarrow}$
$L_{3\downarrow}$
$T < T_c$
$T \geq T_c$

Ellipsometry





$$r_p = \left(\frac{E_{rp}}{E_{ip}}\right) = \frac{n_1 \cos\theta_0 - n_0 \cos\theta_1}{n_1 \cos\theta_0 + n_0 \cos\theta_1}$$

$$r_s = \left(\frac{E_{rs}}{E_{is}}\right) = \frac{n_0 \cos\theta_0 - n_1 \cos\theta_1}{n_0 \cos\theta_0 + n_1 \cos\theta_1}$$

$$\rho = \frac{r_p}{r_s} = \tan\psi\, e^{i\Delta}$$

$$\psi = \tan^{-1}\left(\frac{|r_p|}{|r_s|}\right) = \tan^{-1}\left(\frac{|r_p|}{|r_s|}\right) = \tan^{-1}\left[\left(\frac{R_p}{R_s}\right)^{\frac{1}{2}}\right]$$

$$\Delta = \delta_{rp} - \delta_{rs}$$

Depolarization

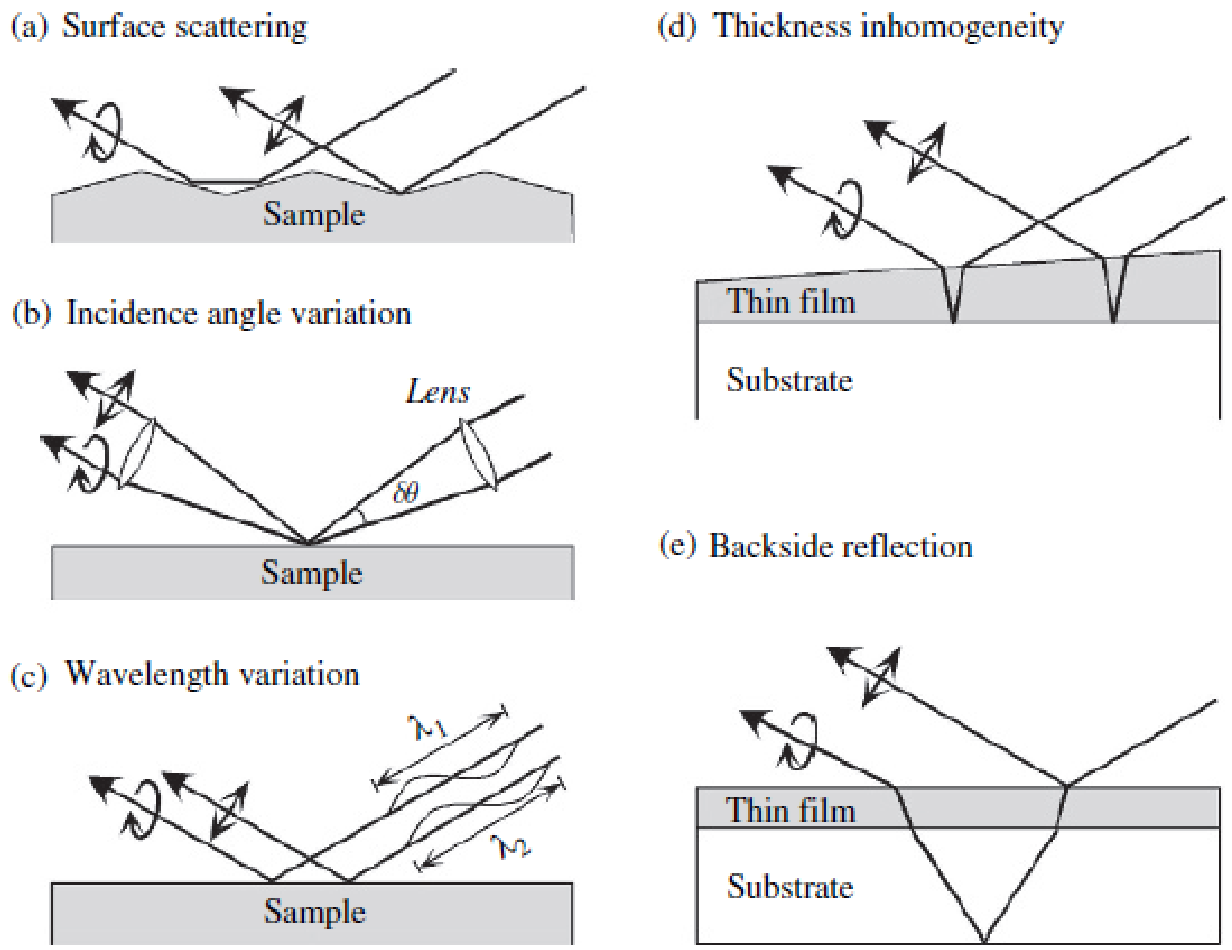
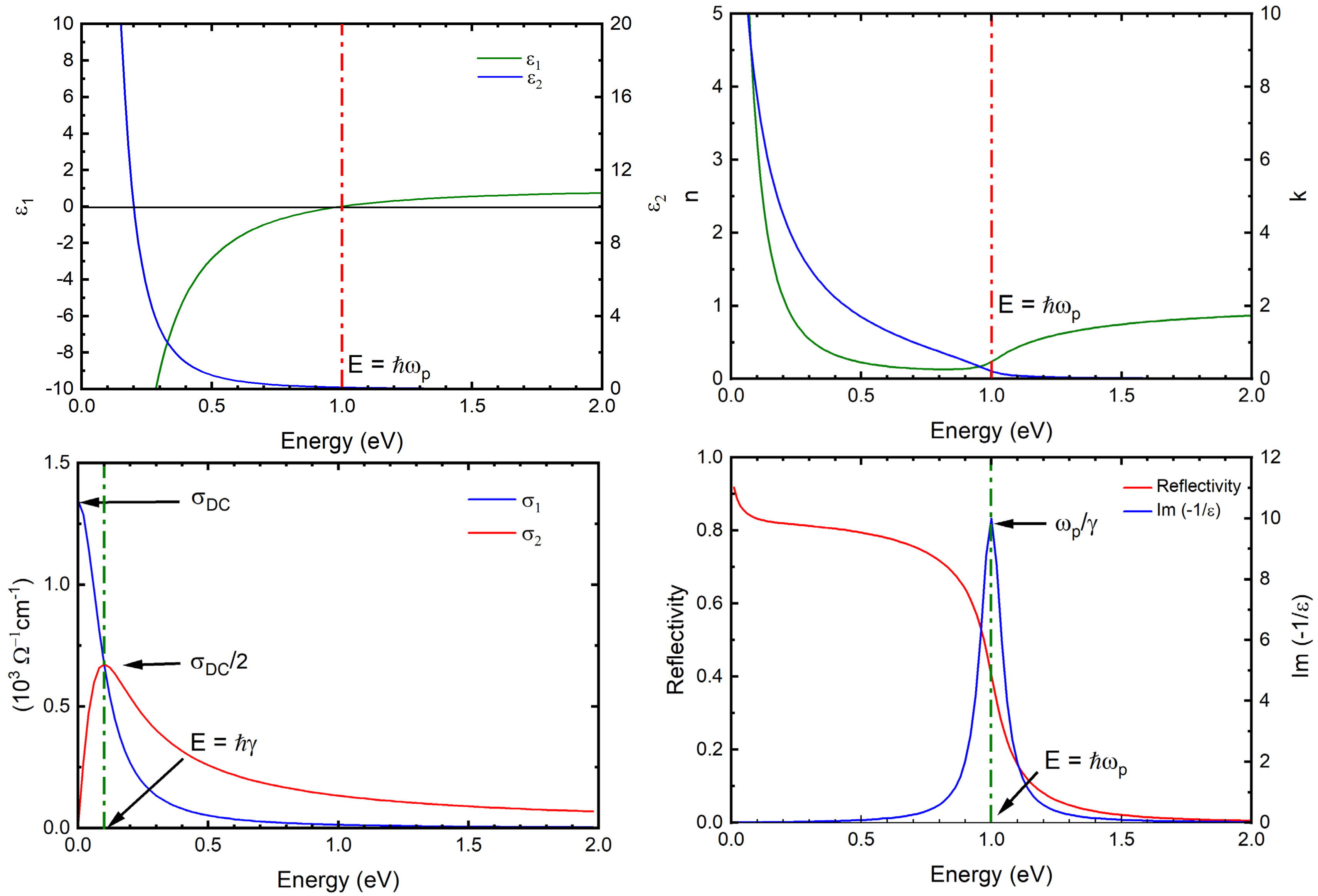


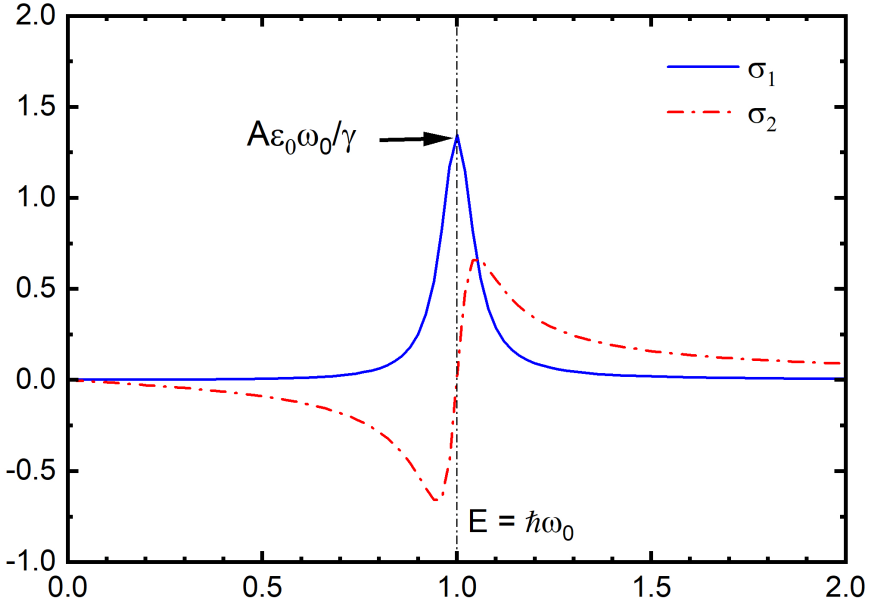
Figure 4.30 Depolarization of incident light by (a) surface scattering, (b) incidence angle variation, (c) wavelength variation, (d) thickness inhomogeneity, and (e) backside reflection.

H. Fujiwara, *Spectroscopic ellipsometry: principles and applications* (John Wiley & Sons, West Sussex, England, (2007).

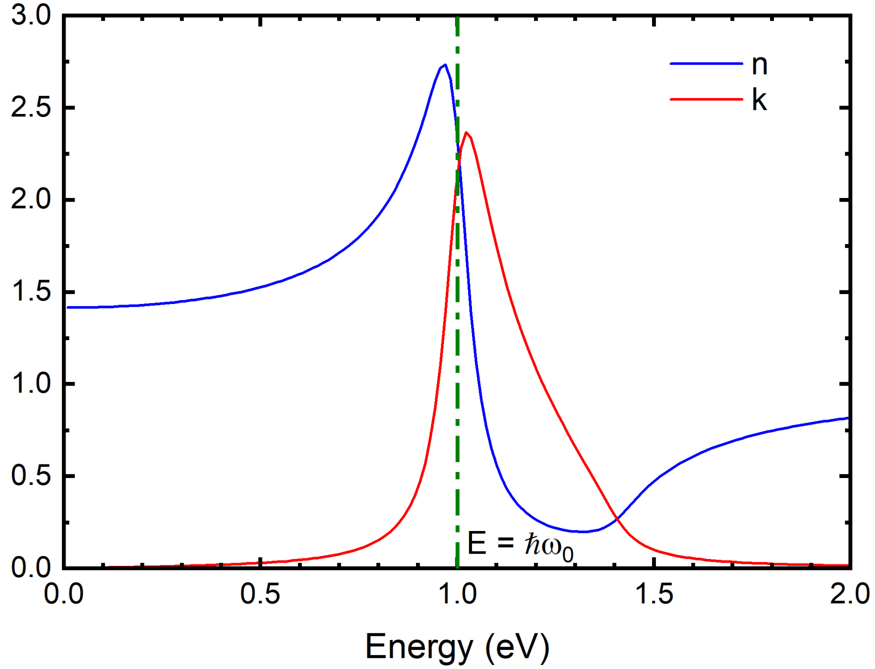
Drude term



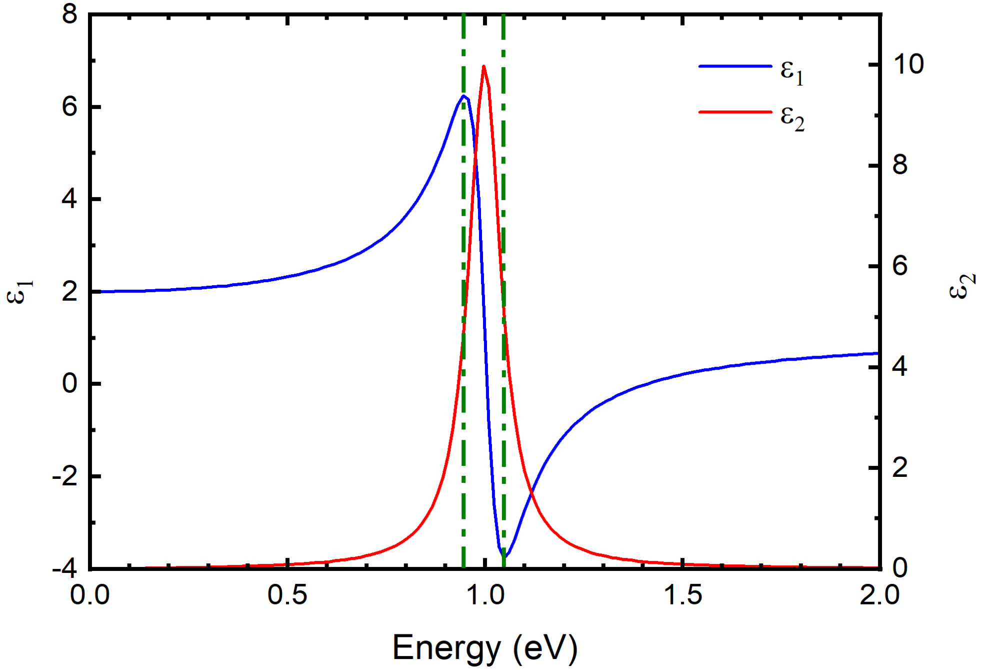
ε1
ε2
E = ℏωp
Energy (eV)
n
k
E = ℏωp
Energy (eV)
σDC
σDC/2
E = ℏγ
σ1
σ2
(10³ Ω⁻¹cm⁻¹)
Energy (eV)
Reflectivity
Im (-1/ε)
ωp/γ
E = ℏωp
Energy (eV)



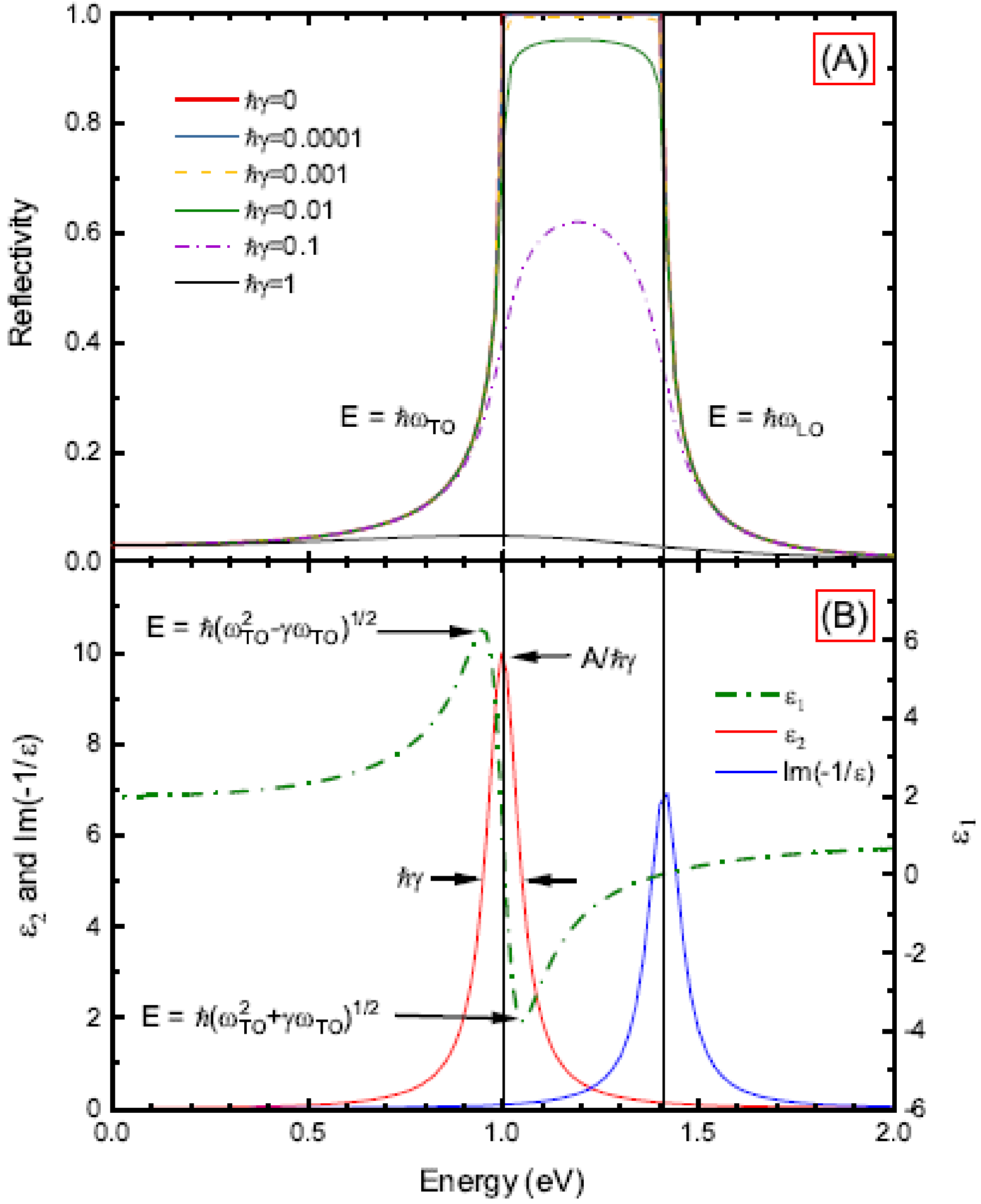
σ1
σ2
Aε0ω0/γ
E = ħω0



n
k
E = ħω0
Energy (eV)



ε1
ε2
Energy (eV)



(A)
ħγ=0
ħγ=0.0001
ħγ=0.001
ħγ=0.01
ħγ=0.1
ħγ=1
Reflectivity
E = ħω_TO
E = ħω_LO
(B)
E = ħ(ω²_TO-γω_TO)^1/2
A/ħγ
ħγ
E = ħ(ω²_TO+γω_TO)^1/2
ε₁
ε₂
Im(-1/ε)
ε₂ and Im(-1/ε)
ε₁
Energy (eV)