Měření elektronových vlastností materiálů pomocí optické spektroskope (elipsometrie a magneto-transmise)

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Ústav fyziky kondenzovaných látek

- Spektroskopická elipsometrie a dielektrická funkce
- Základní optické modely: Lorentzův a Drudeův model
- Studium feromagnetického stavu $La_{0.7}Sr_3CoO_3$ pomocí elipsometrie
- Studium excitovaných stavů $LaCoO_3$ pomocí femtosekundové elipsometrie
- Magneto-optická spektroskopie Landauových přechodů v topologických izolátorech Bi_2Te_3

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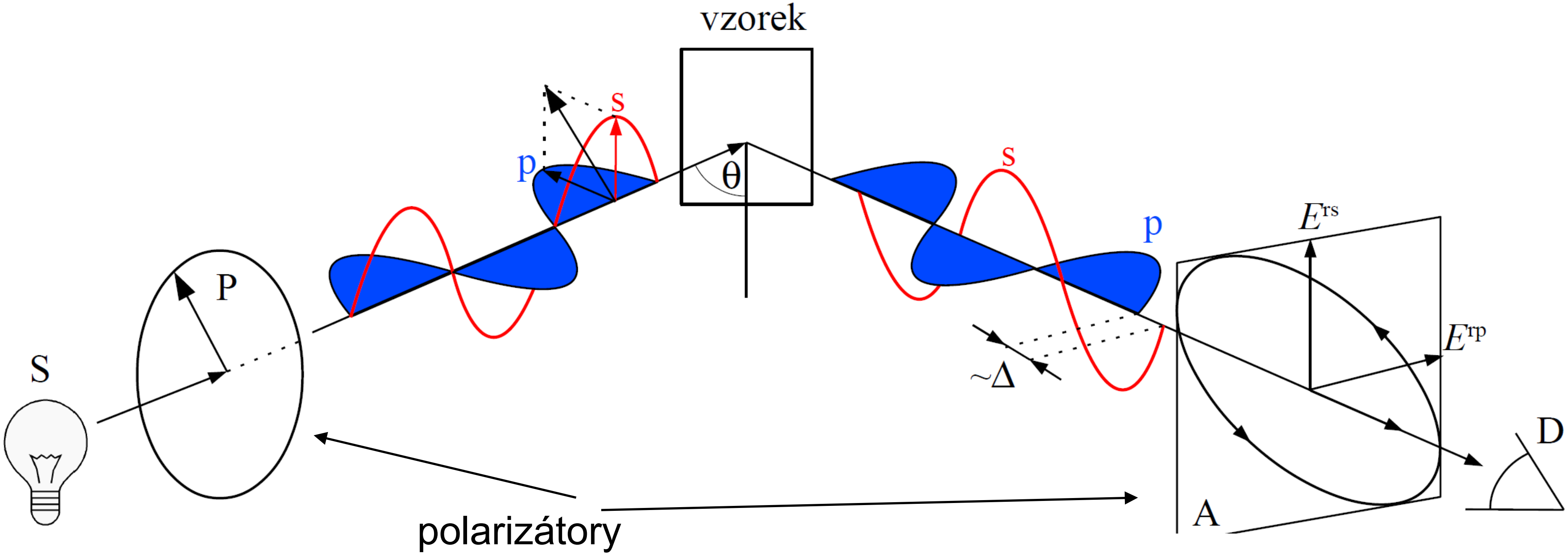
Princip elipsometrie



• Elipsometrie je de facto interferenční experiment s komponentou elektrického pole rovnoběžnou (p) a kolmou (s) k rovině dopadu.

Měřené veličiny v elipsometrii:

• úhel pootočení elipsy Ψ

• elipticita Δ

=> n,k nebo $\boldsymbol{\varepsilon_1, \varepsilon_2}$ bez dalších předpokladů

základní rovnice elipsometrie

Definice elipsometrických úhlů Ψ a Δ: $$\rho = \frac{r_\mathrm{p}}{r_\mathrm{s}} = \tan\Psi\, \mathrm{e}^{\mathrm{i}\Delta}$$

Fresnelovy koeficienty:

$$r_\mathrm{p} = \frac{N_2\cos\theta_1 - N_1\cos\theta_2}{N_1\cos\theta_2 + N_2\cos\theta_1} \qquad r_\mathrm{s} = \frac{N_1\cos\theta_1 - N_2\cos\theta_2}{N_1\cos\theta_1 + N_2\cos\theta_2}$$

Snellůlv zákon: $$N_1\sin\theta_1 = N_2\sin\theta_2$$

Index lomu okolí: $N_1 = \sqrt{\epsilon_\mathrm{a}}$ Index lomu vzorku: $N_2 = \sqrt{\epsilon_\mathrm{s}}$

Inverzí výše uvedených rovnic obdržíme v případě polonekonečného izotropního vzorku explicitní analytický výraz pro dielektrickou funkci (jak její reálnou tak i imaginární část):

$$\epsilon_\mathrm{s}(\Psi,\Delta) = \epsilon_\mathrm{a}\sin^2\theta_1\left(1 + \tan^2\theta_1\left(\frac{1-\rho(\Psi,\Delta)}{1+\rho(\Psi,\Delta)}\right)^2\right)$$

shrnuto: ze dvou měřených veličin Ψ a Δ určíme dvě veličiny ε_1 a ε_2

Absorpce- reálná část optické vodivosti

Optická vodivost se vztahuje k dielektrické funkci jako $\sigma(\omega) = -\mathrm{i}\omega\varepsilon_0\left(\varepsilon(\omega) - 1\right)$

Je to komplexní funkce: $\sigma(\omega) = \sigma_1(\omega) + \mathrm{i}\sigma_2(\omega)$

- Reálná část vodivosti $\sigma_1(\omega) = \omega\varepsilon_0\varepsilon_2(\omega)$, je úměrná absorpci elektromagnetické energie
- $\sigma_1(\omega=0) = \sigma_{DC}$

- σ_1 je vázaná sumačním pravidlem $\int_0^\infty \sigma_1(\omega)\mathrm{d}\omega = \frac{\pi n q^2}{2\varepsilon_0 m} = \mathrm{constant}$

  - Integrál z $\sigma_1(\omega)$ přes široký frekvenční interval je proporční náboji který záření absorbuje.

Lorentzův oscilátor

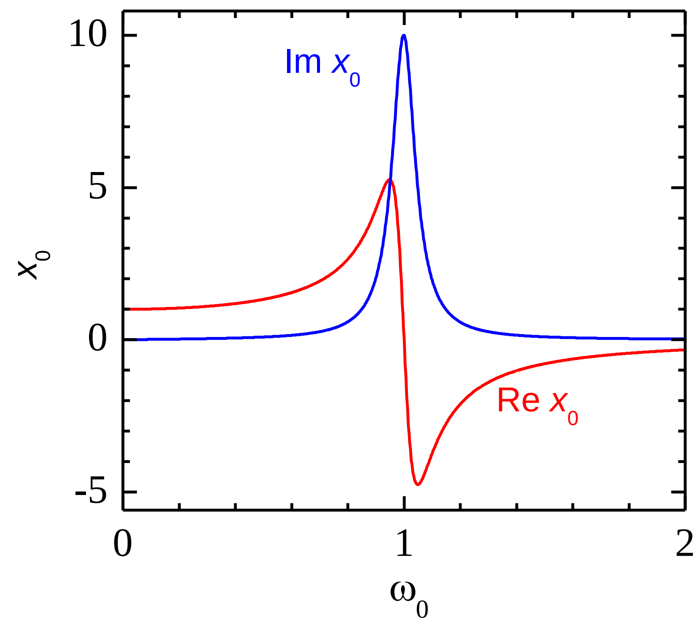
Newtonova rovnice harmonicky buzeného mechanického oscilátoru:

$$m\frac{\mathrm{d}^2x(t)}{\mathrm{d}t^2} = -k\,x(t) - m\gamma\frac{\mathrm{d}x(t)}{\mathrm{d}t} + qE_0\mathrm{e}^{-\mathrm{i}\omega t}$$

Řešení:

$$x_0(\omega) = \frac{F}{\omega_0^2 - \omega^2 - \mathrm{i}\omega\gamma}$$

$$\omega_0 = \sqrt{\frac{k}{m}}, \quad F = \frac{qE_0}{m}$$



polarizace je hustota dipólového momentu

$$P(\omega) = \sum_j nqx_{0,j}(\omega)$$

n: koncentrace

z definice dielektrické funkce:

$$\varepsilon(\omega) = 1 + \frac{P(\omega)}{\varepsilon_0 E(\omega)} = 1 + \sum_j \frac{\omega_{pl,j}^2}{\omega_{0,j}^2 - \omega^2 - \mathrm{i}\omega\gamma_j}$$

plasmová frekvence:

$$\omega_{\mathrm{pl},j} = \sqrt{\frac{q_j^2 n_j}{\epsilon_0 m_j}}$$

příspěvek vysokofrekvenčních přechodů lze nejhruběji aproximovat konstantou:

$$\epsilon(\omega) = \epsilon_\infty + \sum_j \frac{\omega_{\mathrm{pl},j}^2}{\omega_{0,j}^2 - \omega^2 - \mathrm{i}\omega\gamma_j}$$

- dielektrická fukce nezávislých Lorentzových oscilátorů. Typicky dobře funguje pro fonony. Drudeův model kovů dostaneme dosazením ω_0=0

Drudeova formule

• odezvu volných nosičů náboje získáme pro ω_0=0

$$\varepsilon(\omega) = \varepsilon_\infty - \frac{\omega_{pl}^2}{\omega(\omega + \mathrm{i}\gamma)}$$

plasmová frekvence $$\omega_{pl} = \sqrt{\frac{q^2 n}{\varepsilon_0 m^*}}$$

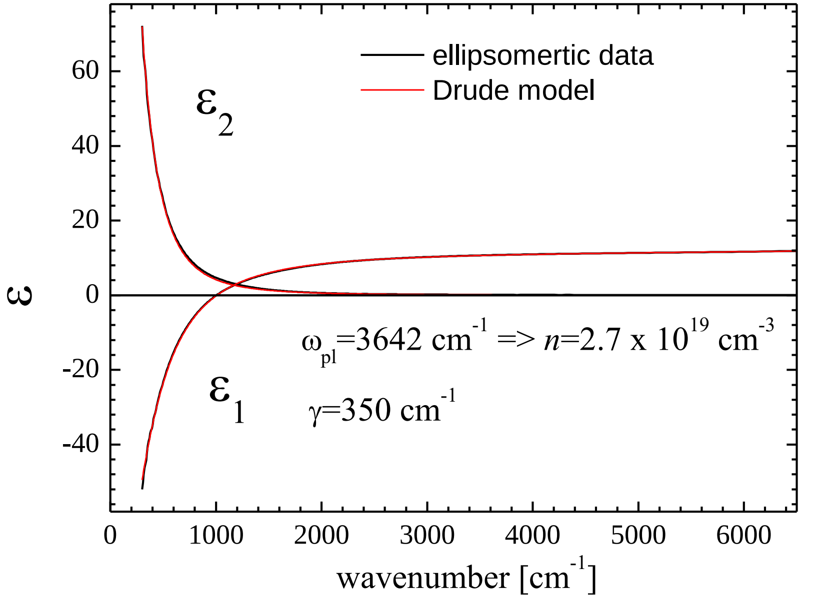
závisí na koncentraci nositelů n a na jejich efektivní hmotnosti m^*

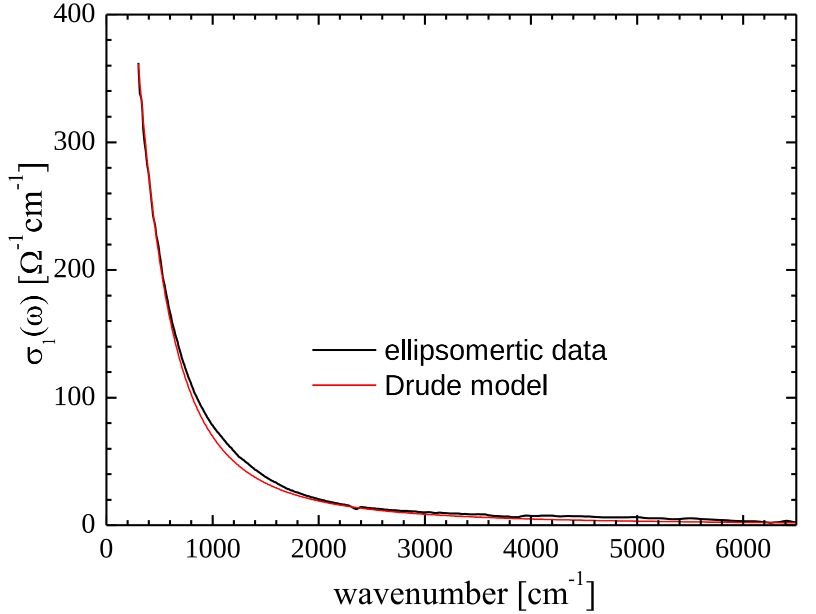
ε_1 prochází nulou (pro γ ~0) pro

$$\omega = \frac{\omega_{pl}}{\sqrt{\varepsilon_\infty}}$$

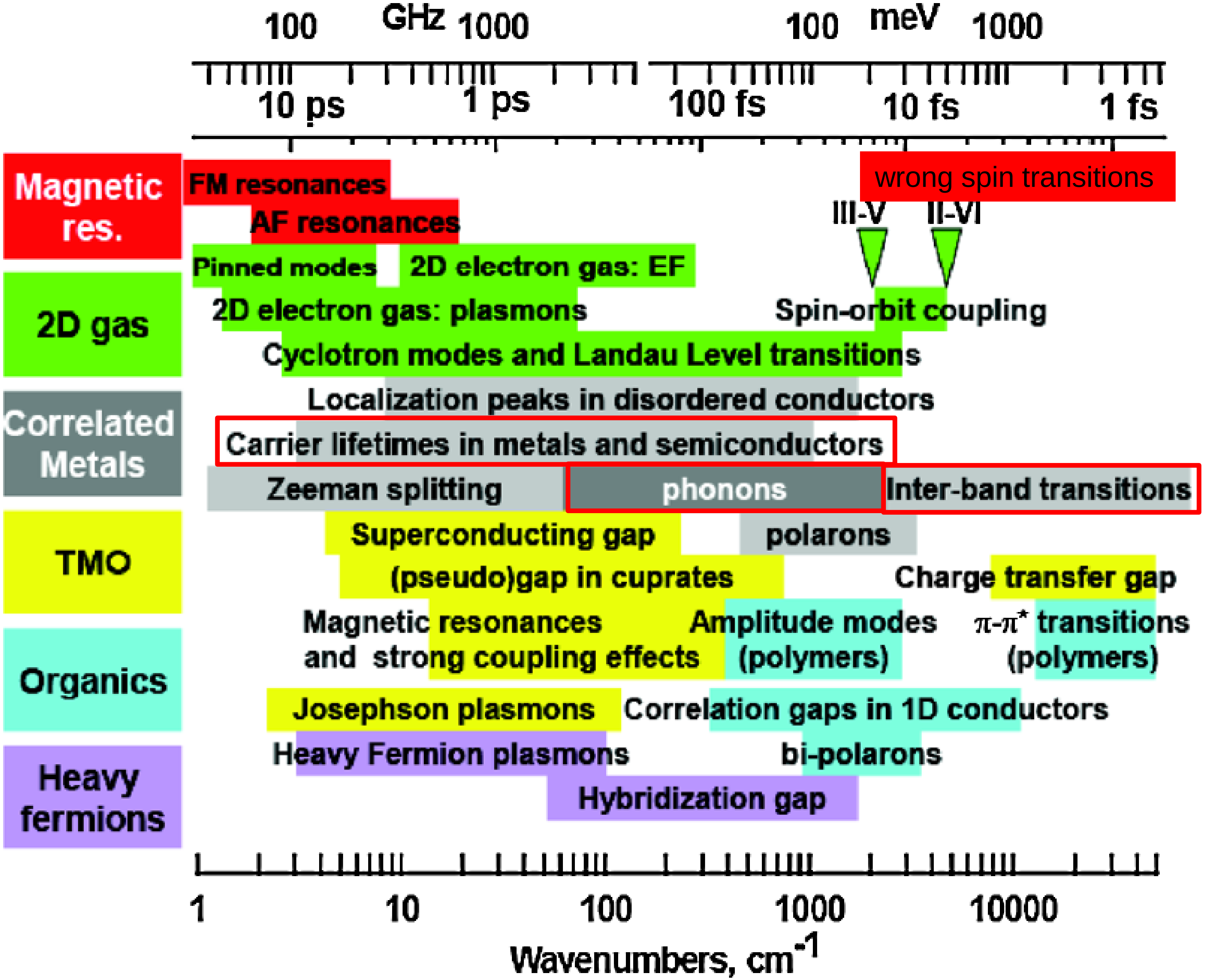
pro $\varepsilon_\infty = 1$ je to přímo ω_{pl}. Na této frekvenci se v látce propaguje longitudinální plasmon, proto se této frekvenci říká plasmová.

Ukázka dielektrické funkce n-dopovaného křemíku





Opticky aktivní excitace mezi THz a ultrafialovým oborem

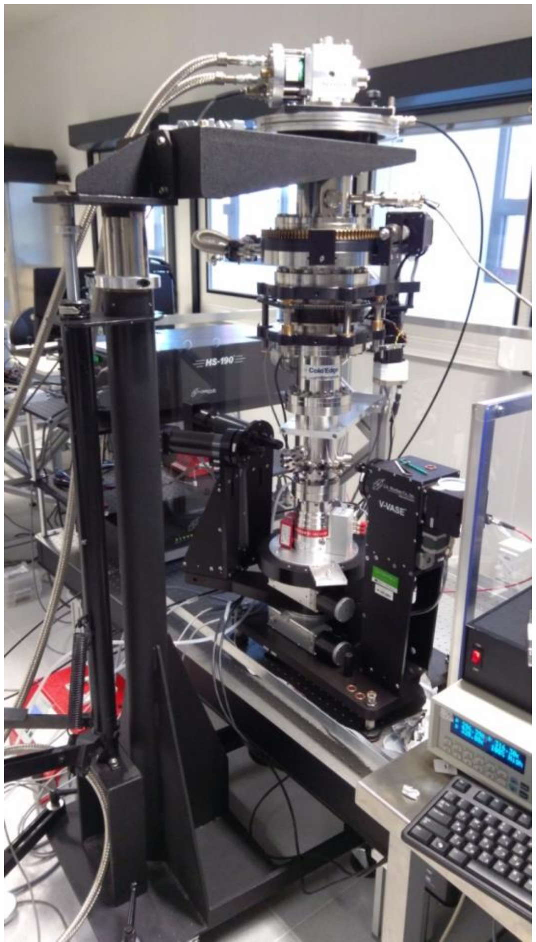


D. Basov *et al.*, Phys. Mod Rev. 2011

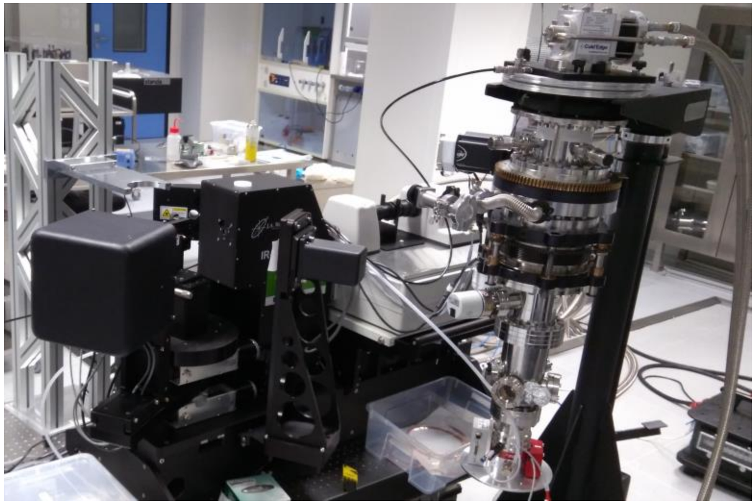
Equilibrium ellipsometry at CEITEC Nano

VYSOKÉ UČENÍ TECHNICKÉ V BRNĚ + MUNI SCI = CEITEC NANO in CZECH NANO LAB.

Woollam VASE, NIR-UV range
He closed-cycle cryostat 7-400 K



Woollam IR-VASE, mid infrared range

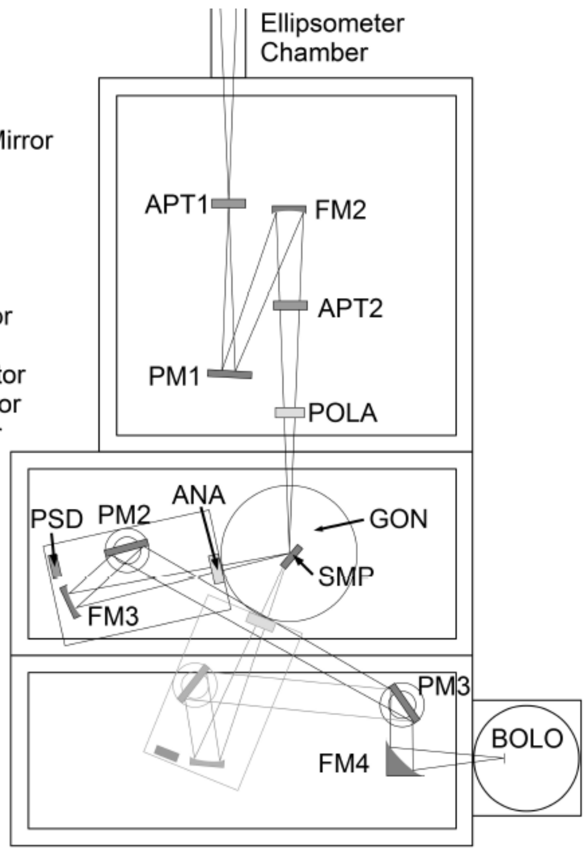


far-infrared (50-700 cm^{-1}) ellipsometer

Elipsometr pro vzdálenou infračervenou oblast v CEITECu

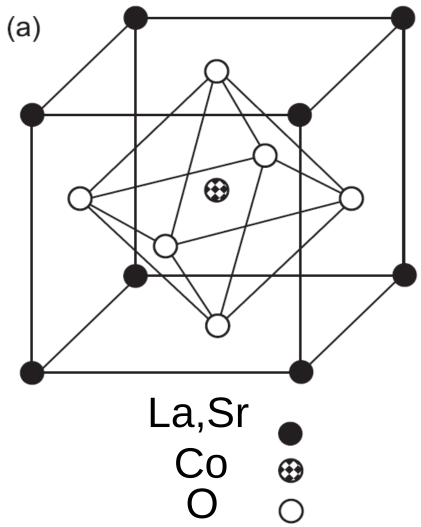
- jen asi 4 přístroje podobného typu na světě
- kryostat s uzavřeným cyklem helia 7-400 K
- rozhraní s ultra-nízkými vibracemi pro odsstranění vlivu vibrací
- motorizovaný goniometr s rozlišením 0.01 ^{o}
- automatizované měření ~15 teplot za 24 hodin
- detektor - 4.2K (a nově 1.6 K) bolometr

ANA - Analyzer
APT1,2 - Aperture
BMS - Beam Splitter
BOLO - Bolometer
FM1,3,4 - Parabolic Mirror
FM2 - Elliptical Mirror
GLB - Glow Bar
GON - Goniometer
HG - Mercury Lamp
LAS - Aligment Laser
PM1,2,3 - Plane Mirror
POLA - Polarizer
PSD - Position Detector
RM - Removable Mirror
SMP - Sample Holder
W - Tungsten Lamp



P. Friš a A. Dubroka, Appl. Surf. Science **421**, 430 (2017)

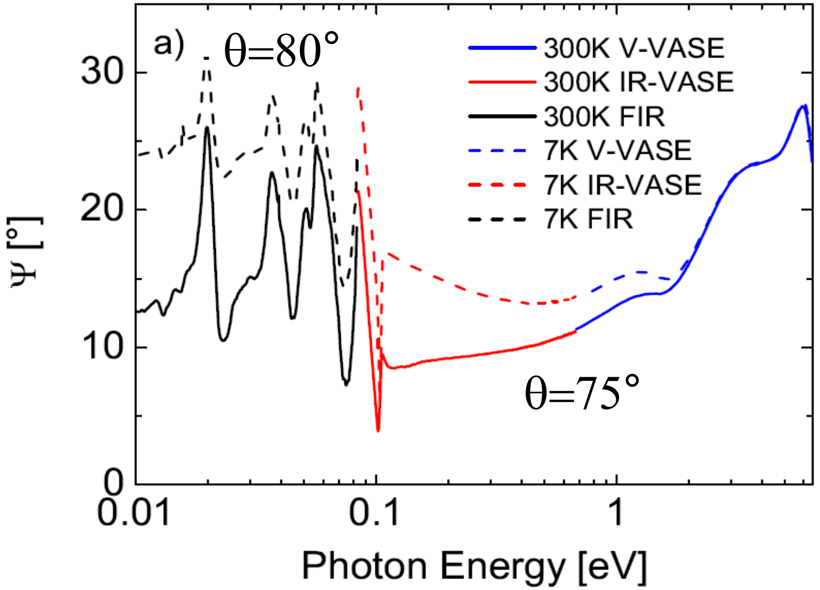
Optická odezva feromagnetických kobaltátů -hrubá data na 30nm vrstvách

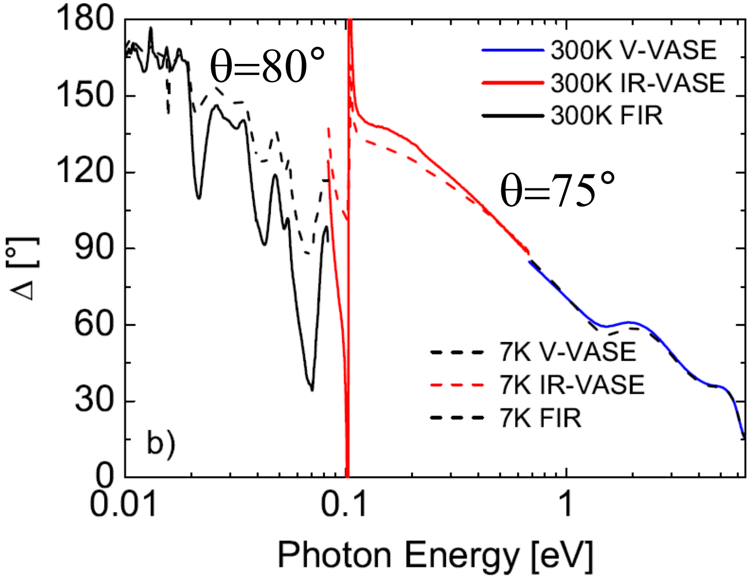


- tenké vrstvy (30 nm) feromagnetického $La_{0.7}Sr_{0.3}CoO_3$ vypěstované na substrátu LSAT pomocí pulsní laserové depozice (Alineason Materials Technology)

- Crurieova teplot T_c ~205 K

Hrubá data v podobě elipsometrických úhlů obsahují jak odezvu vrtsvy tak susbstrátu



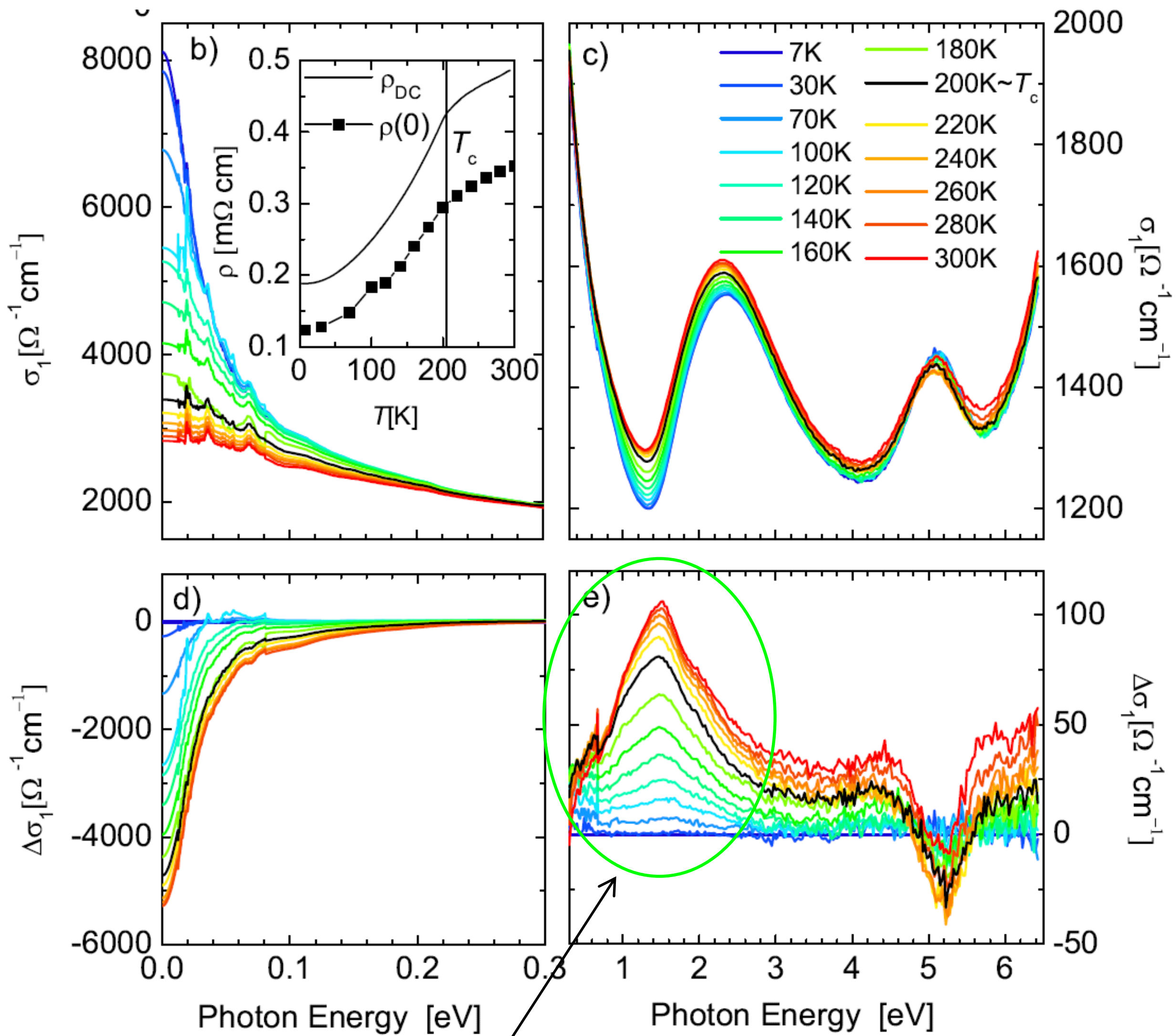


Optické projevy feromagnetického stavu

$La_{0.7}Sr_{0.3}CoO_3$,
T_c ~205 K

Absolutní optická
vodivost (absoprce)
$\sigma_1(\omega) = \omega\varepsilon_0\varepsilon_2(\omega)$

Relativní optická
vodivost
vzhledem k 7 K



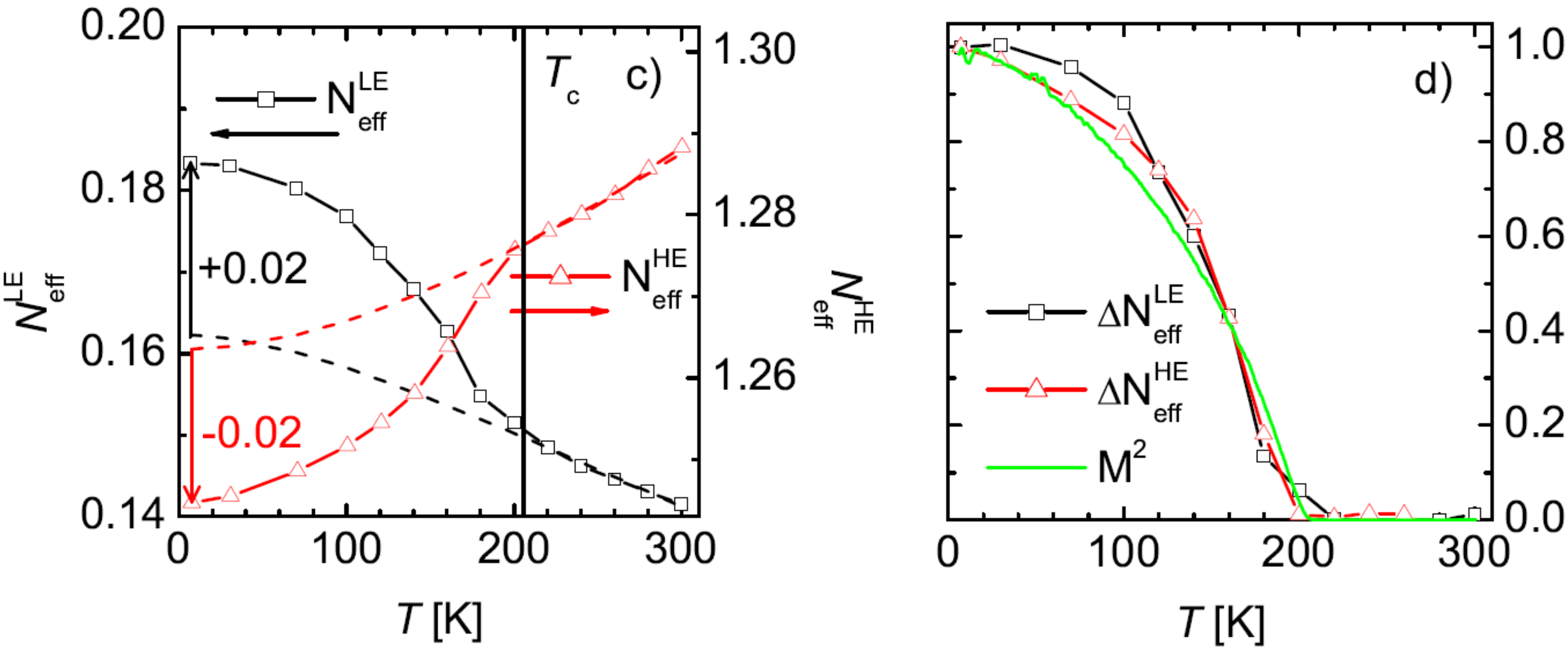
Kandidát pro přechod „špatného spinu“ na 1.5 eV

P. Friš et al, Phys. Rev. B **97,** 045137 (2017)

Optické projevy feromagnetického stavu

$La_{0.7}Sr_{0.3}CoO_3$, T_c ~205 K

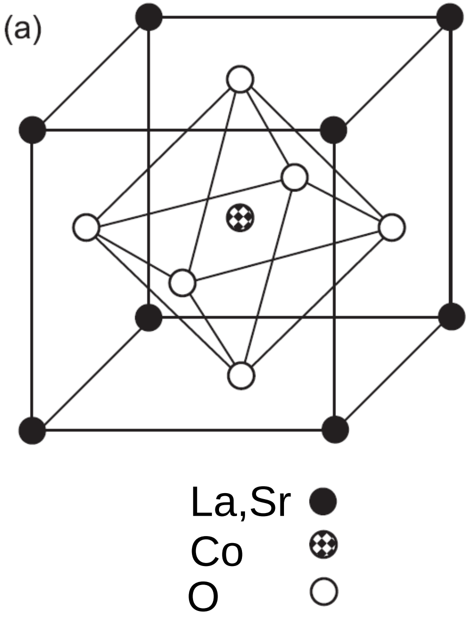
- spektrální váhy (integrál z σ_1) Drudeho píku a pásu na 1.5 eV sledují vývoj magnetizace

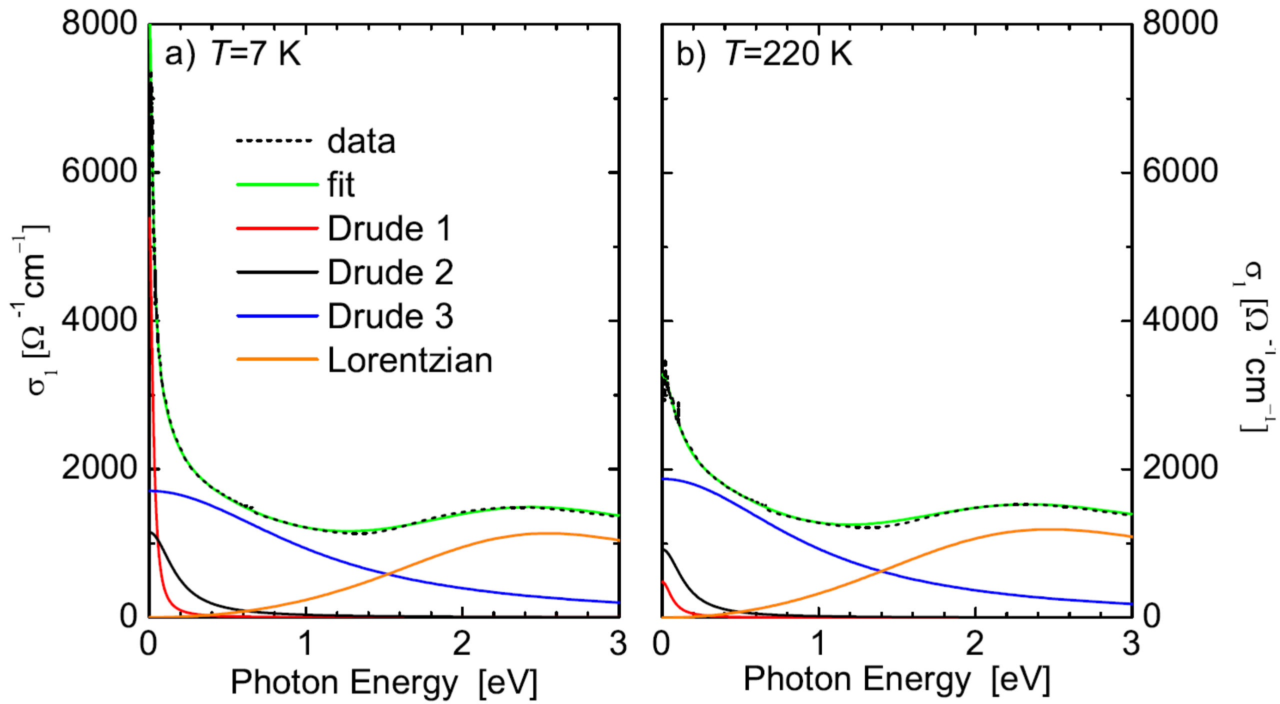


P. Friš et al, Phys. Rev. B **97,** 045137 (2017)

Modelování spekter pomocí Drudeovy-Lorentzovy formule

$La_{0.7}Sr_{0.3}CoO_3$, T_c ~205 K





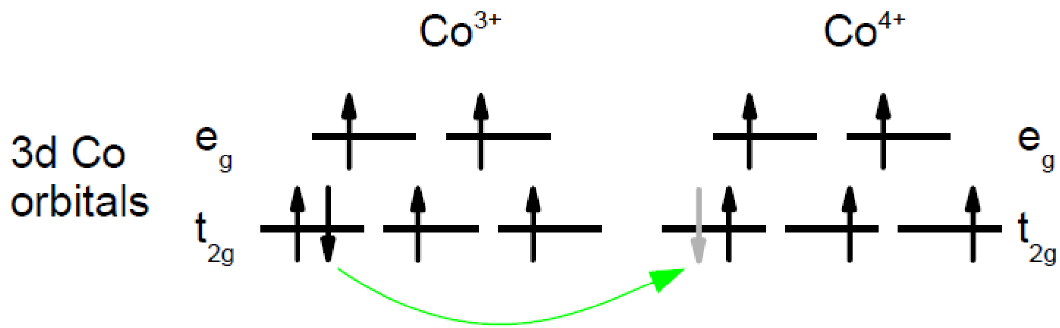
Modelování optických spekter pomocí Drudeovy-Lorentzovy formule

$$\epsilon(\omega) = 1 - \sum_j \frac{\omega_{D,j}^2}{\omega(\omega + \mathrm{i}\gamma_{D,j})} + \sum_k \frac{\omega_{L,k}^2}{\omega_{0,k}^2 - \omega^2 - \mathrm{i}\omega\gamma_{L,k}}$$

Vodivostní odezvu je nutné modelovat třemi Drudeho členy – typický znak interagujících vodivostních elektronů a/nebo přítomnosti několika vodivostních pásů

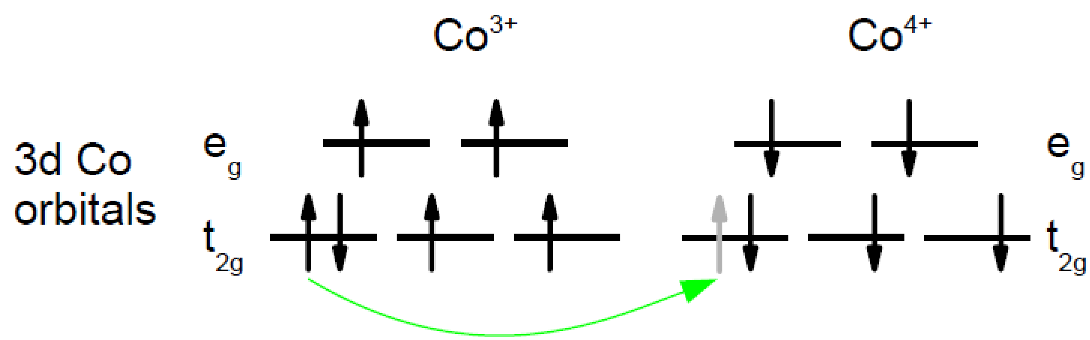
Přechody elektronů mezi ionty kobaltu

Feromagnetický stav



- dvojná výměnná interakce vedoucí k feromagnetismu
- delokalizace elektronů je hnací silou feromagnetického uspořádání
- vede k vodivým vlastnostem

Paramagnetický stav



- přechod mezi kobalty s antiparalelním uspořádáním spinů se nazývá přechod se „špatným spinem“ („wrong-spin-transition“)
- tento přechod porušuje Hundova pravidla, je na něho tedy potřeba určitou energii (~ 1.5 eV).

Photo-induced insulator-to-metal transition in $LaCoO_3$ explored by femtosecond pump-probe ellipsometry

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Pump-probe femtosecond ellipsometry in ELI beamlines, Dolní Břežany

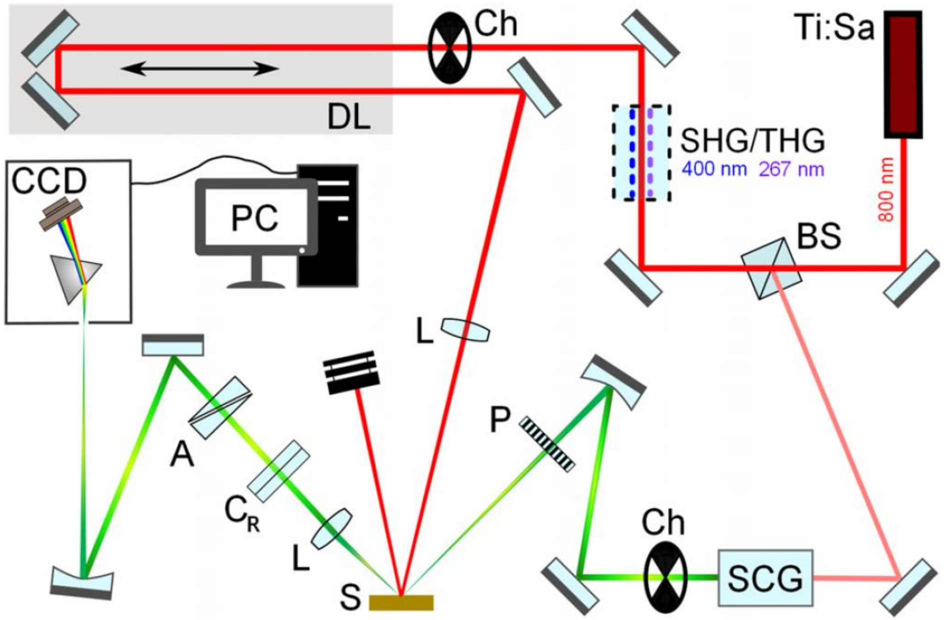
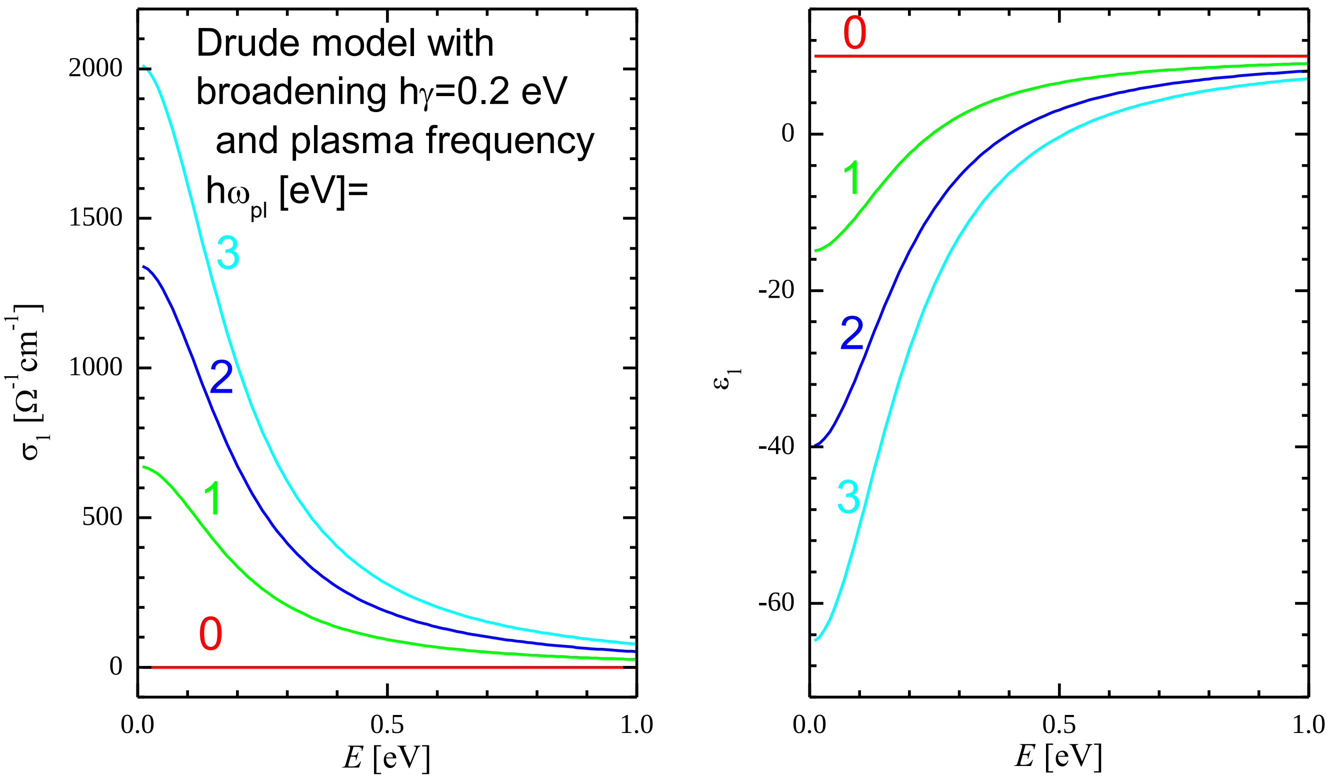


FIG. 2. Experimental setup of the femtosecond pump-probe spectroscopic rotating-compensator ellipsometer. Ch, chopper; A, analyzer; P, wire-grid polarizer; C_R, rotating compensator; L, lens; S, sample; DL, delay line; BS, beam splitter; SHG/THG, second/third harmonic generation (optional), SCG, super-continuum generation; and CCD, charge-coupled device detector. A photograph is shown in Fig. S1.

- Ti:Sapphire laser (Coherent Astrella)
- 35 fs pulses at 800 nm
- 1 kHz rep. rate with 6 mJ pulse en.
- 10 mJ for pump mean
- fluency ~ 10 mJ/cm^2
- Angle of incidence of probe 60 deg
- Angle of incidence of pump 55 deg
- Rotating compensator design
- measurement range: 1.6-3.4 eV

S. Espinoza et al., APL **115b** 052105 (2019)
S. Richter et al., Rev. Sci. Instrum. **92**, 033104 (2021)

Drude model



Drude model

$$\varepsilon(\omega) = \varepsilon_\infty - \frac{\omega_{pl}^2}{\omega(\omega + i\gamma)}$$

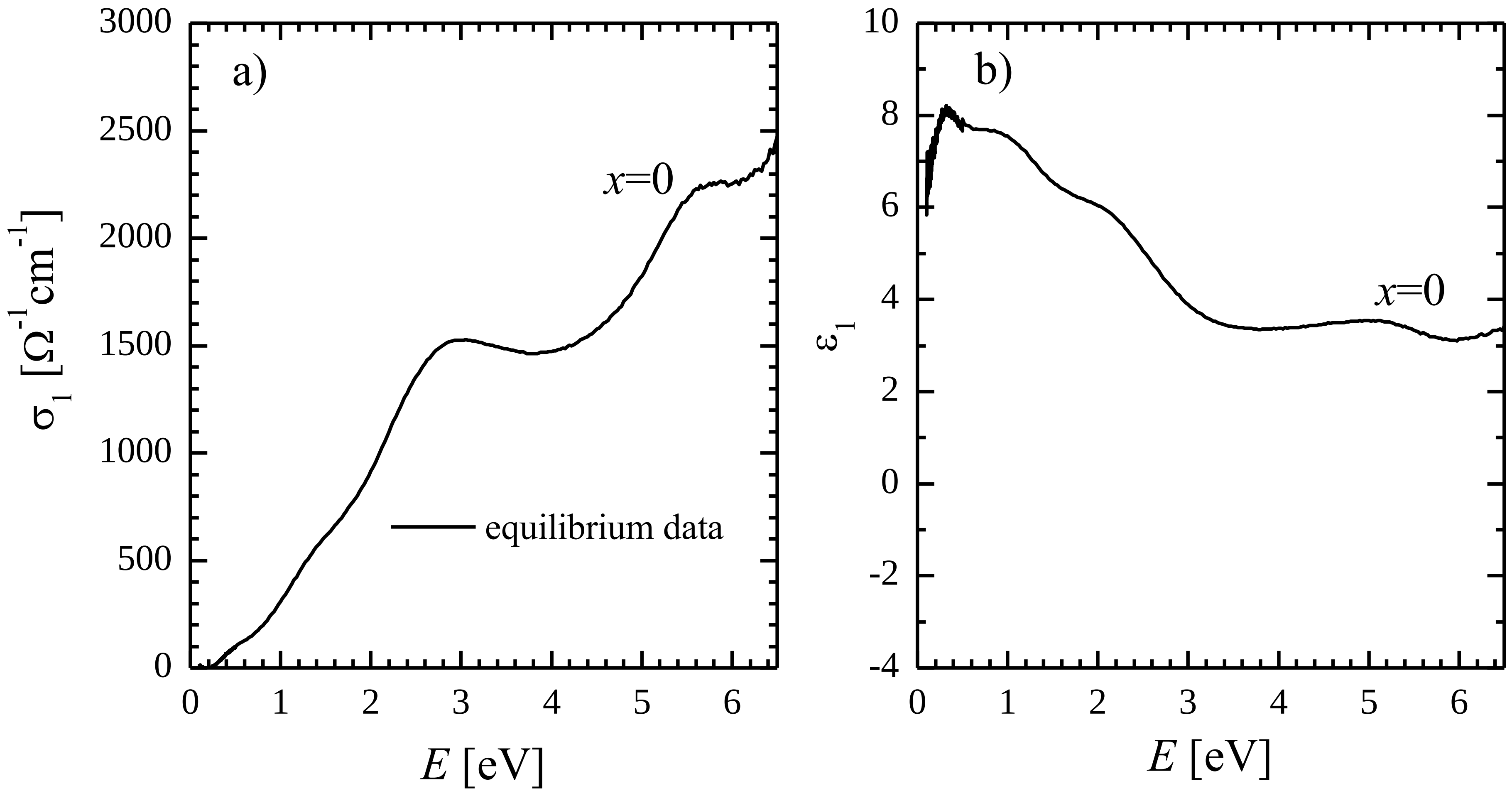
$$\omega_{pl} = \sqrt{\frac{q^2 n}{\varepsilon_0 m^*}}$$

Optical conductivity $\sigma(\omega) = -i\omega\varepsilon_0(\varepsilon(\omega) - 1)$

The real part of optical conductivity
is the absorption per unit of frequency $\sigma_1(\omega) \quad (= \omega\epsilon_0\epsilon_2(\omega))$

absorption sum rule: $\int_0^\infty \sigma_1(\omega)\,\mathrm{d}\omega = \frac{\pi}{2}\frac{nq^2}{\epsilon_0 m} = \mathrm{const.}$

Optical response of $La_{1-x}Sr_xCoO_3$



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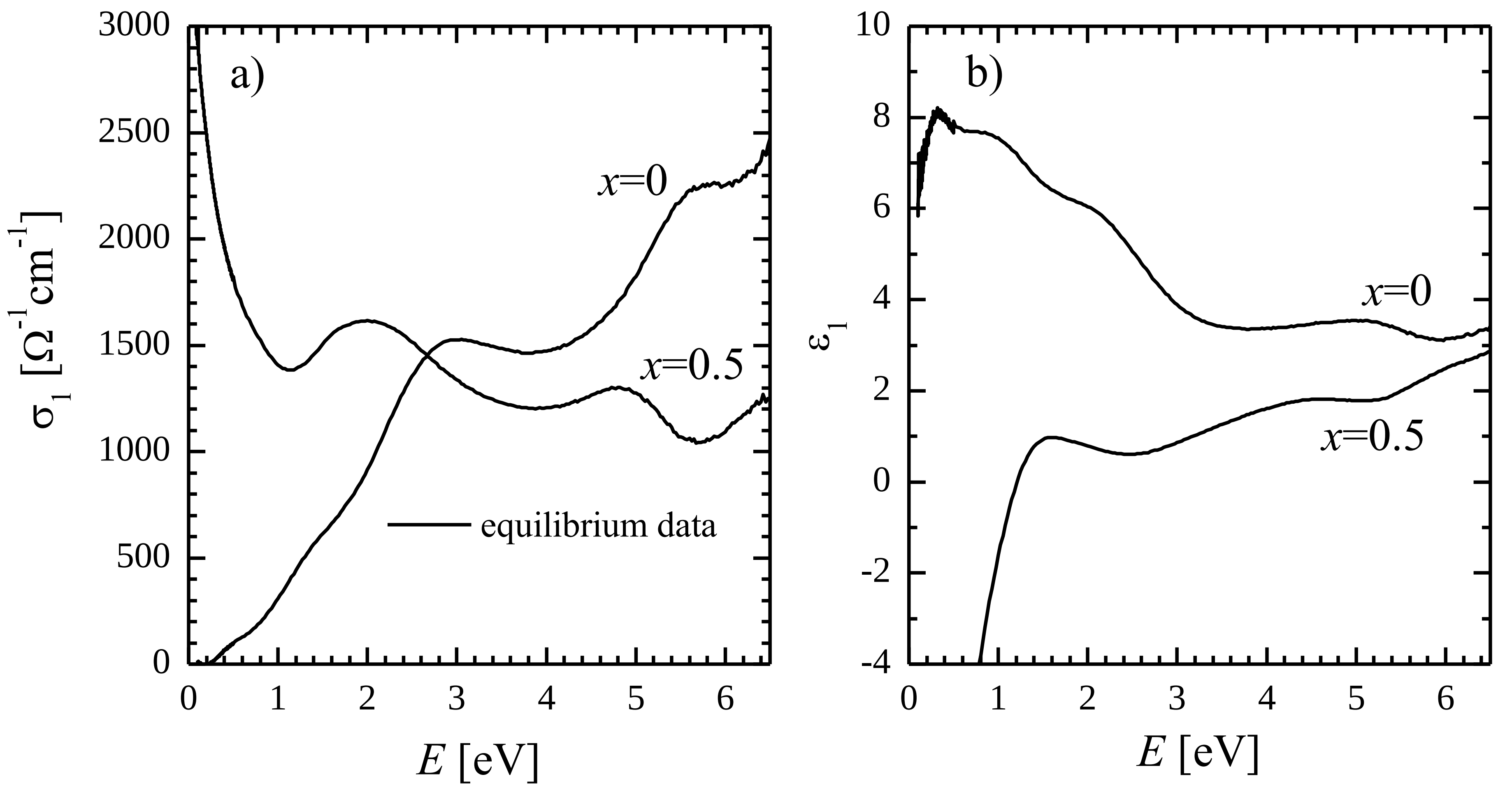
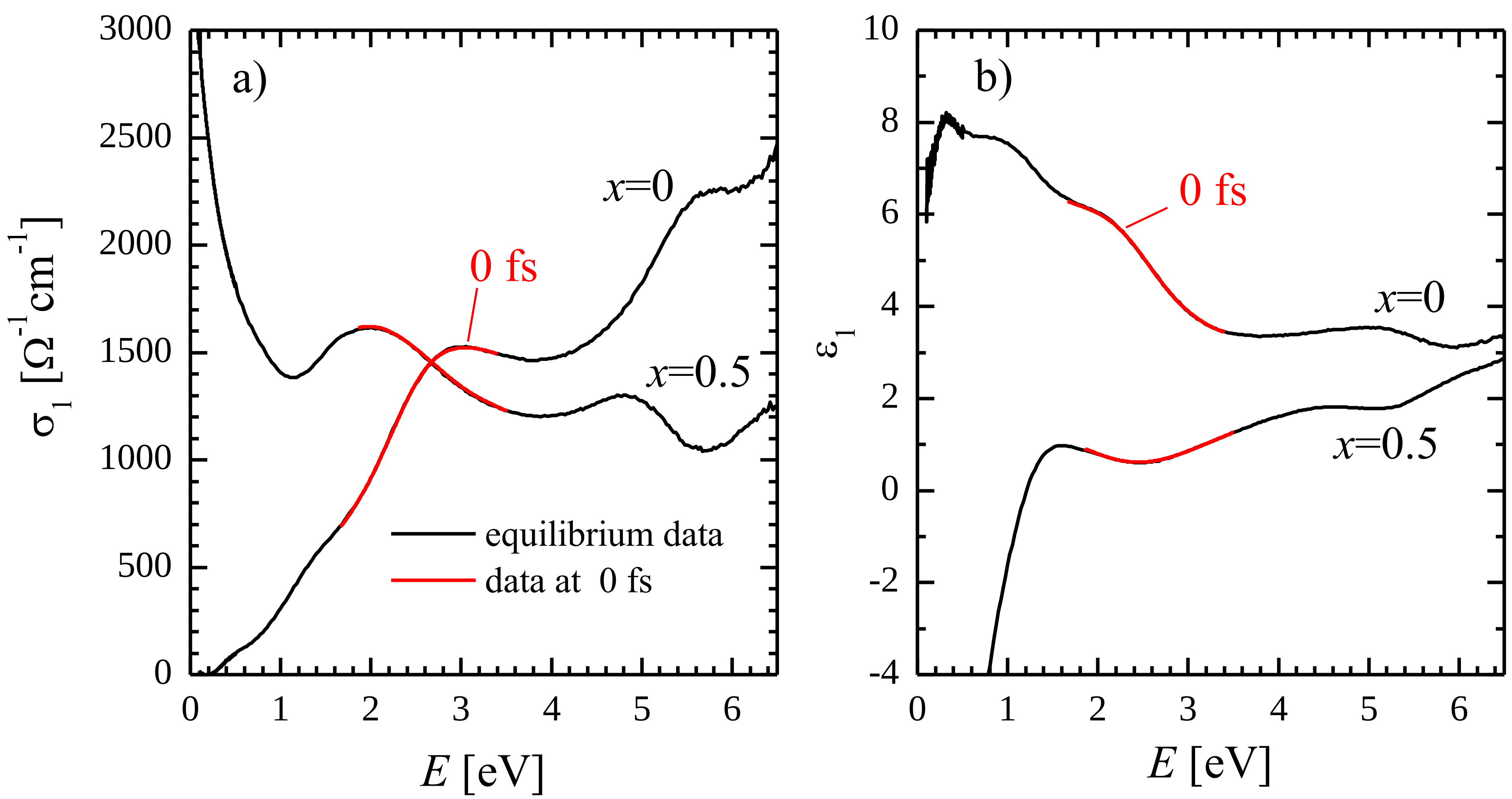
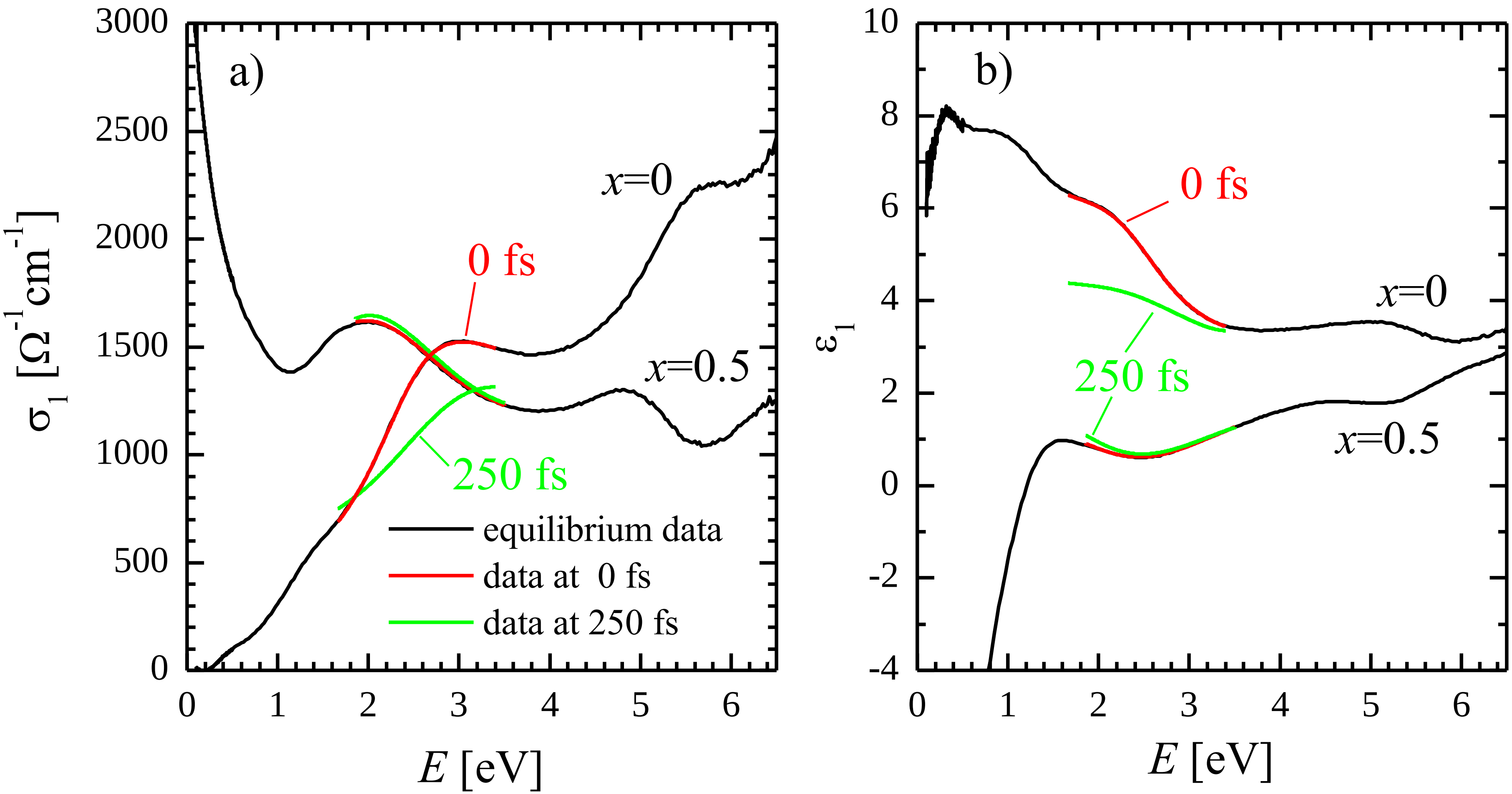


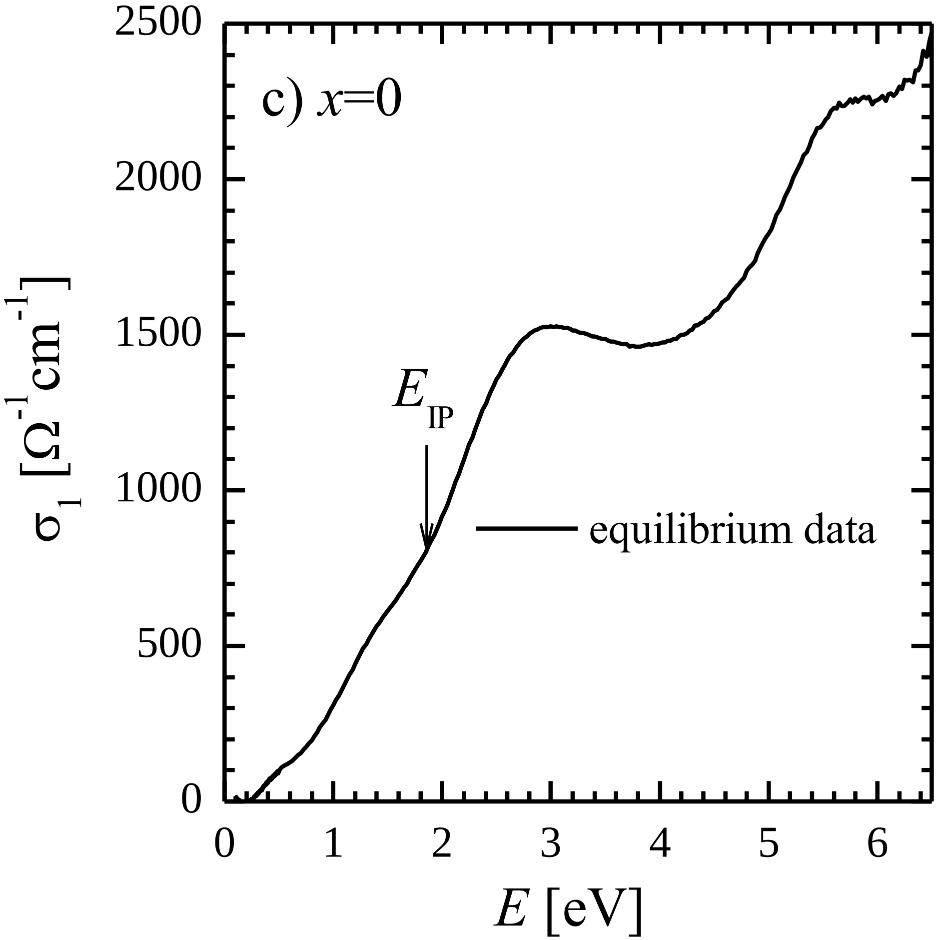
Optical response of $La_{1-x}Sr_xCoO_3$

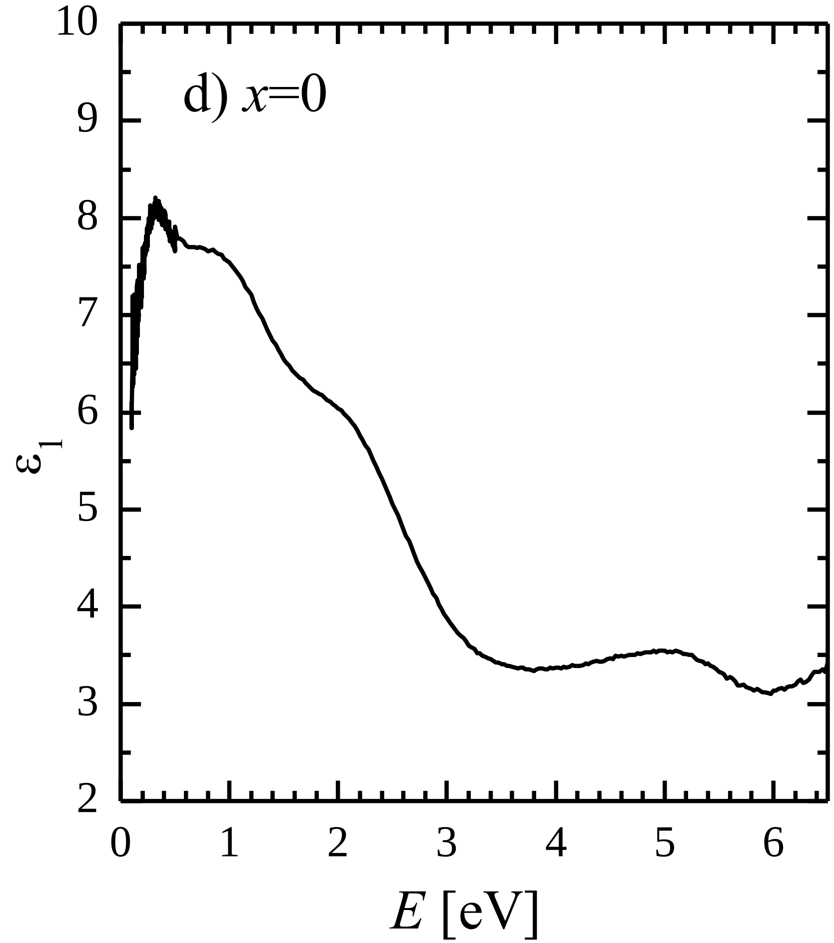


Optical response of $La_{1-x}Sr_xCoO_3$

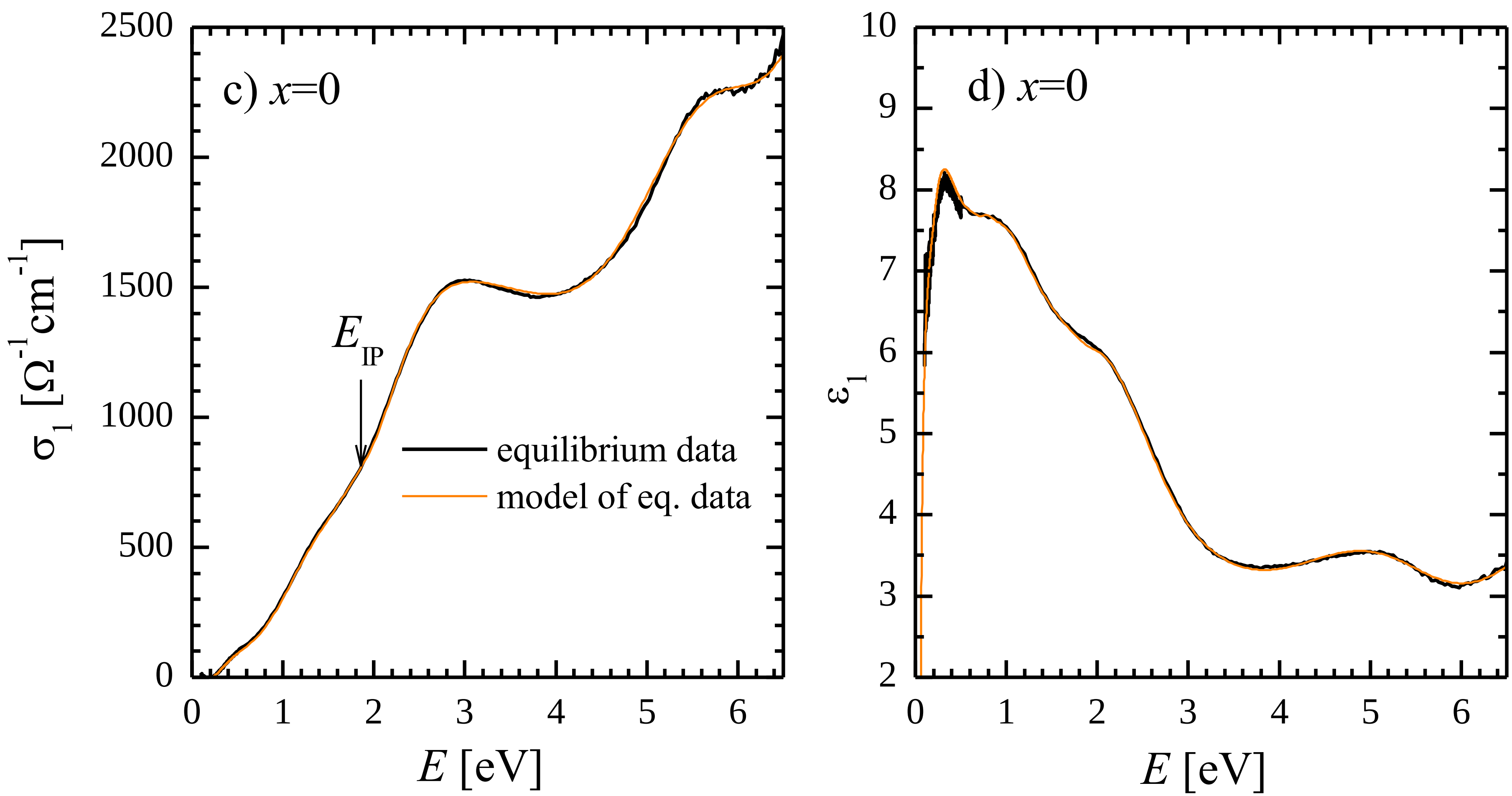


Kramers-Kronig modeling of $LaCoO_3$ at 250 fs



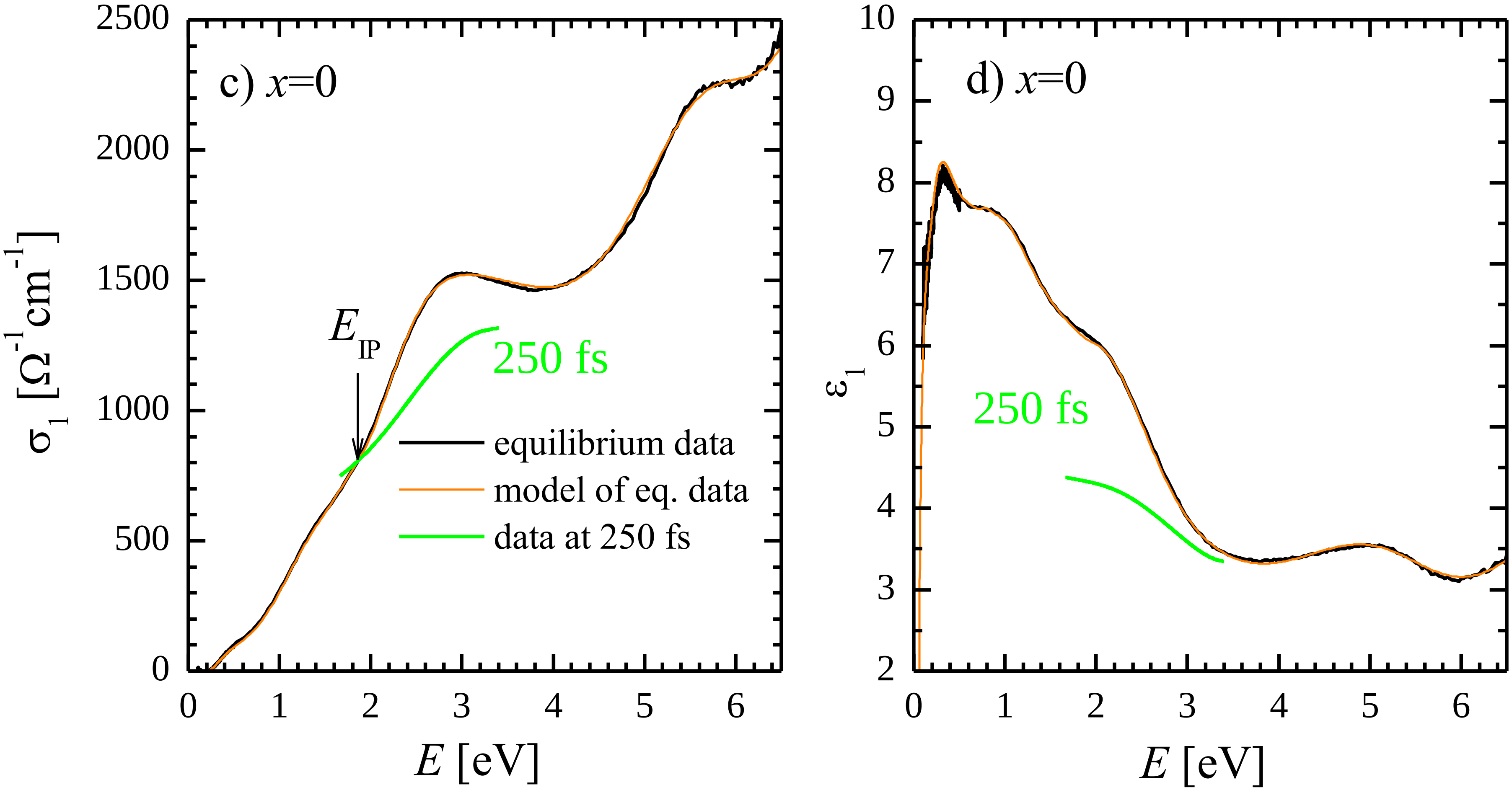


Kramers-Kronig modeling of $LaCoO_3$ at 250 fs

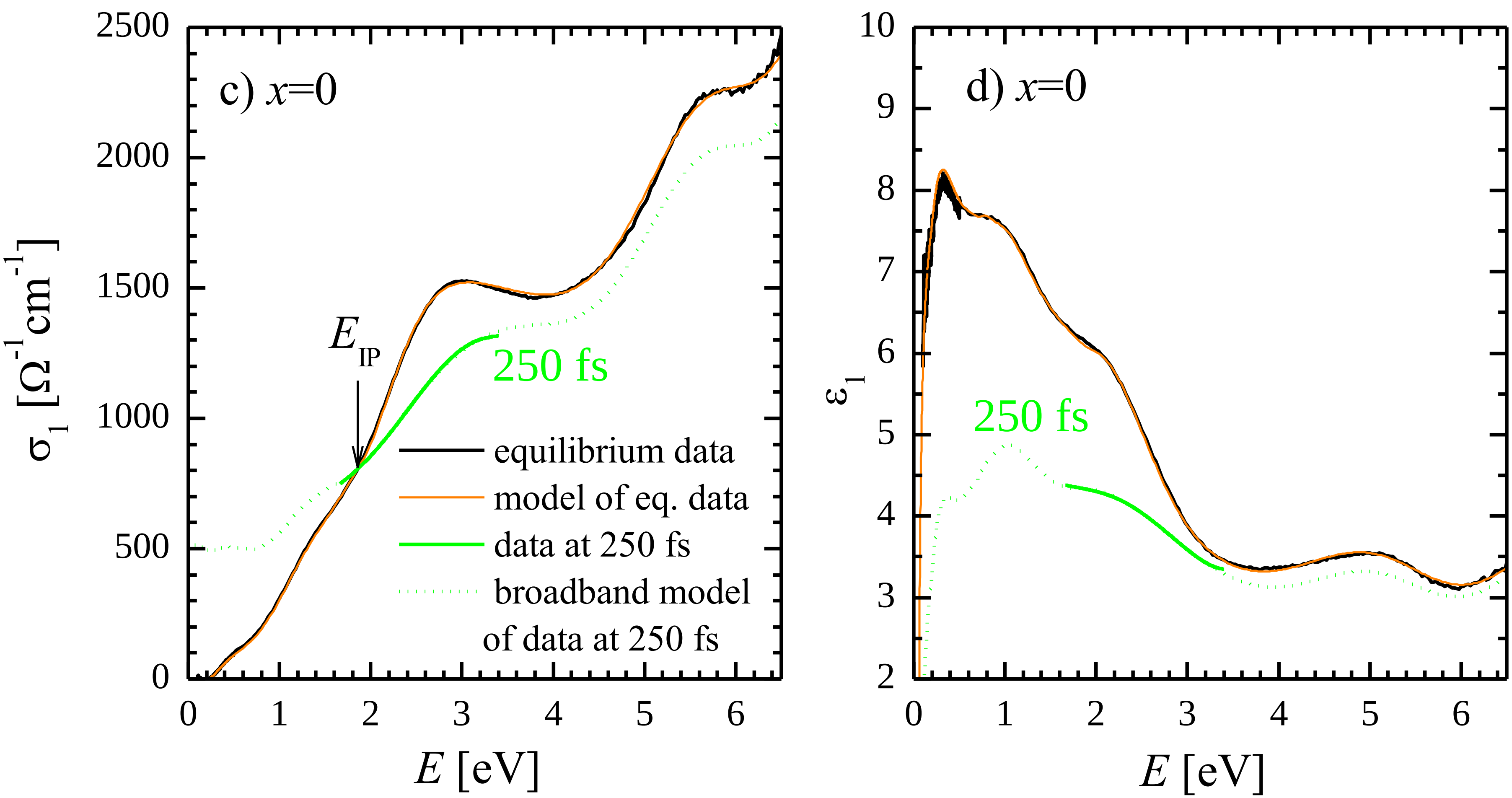


- Modeling the equilibrium data with a set of Kramers-Kronig consistent functions (Tauc-Lorentz+ Gaussian)

Kramers-Kronig modeling of $LaCoO_3$ at 250 fs

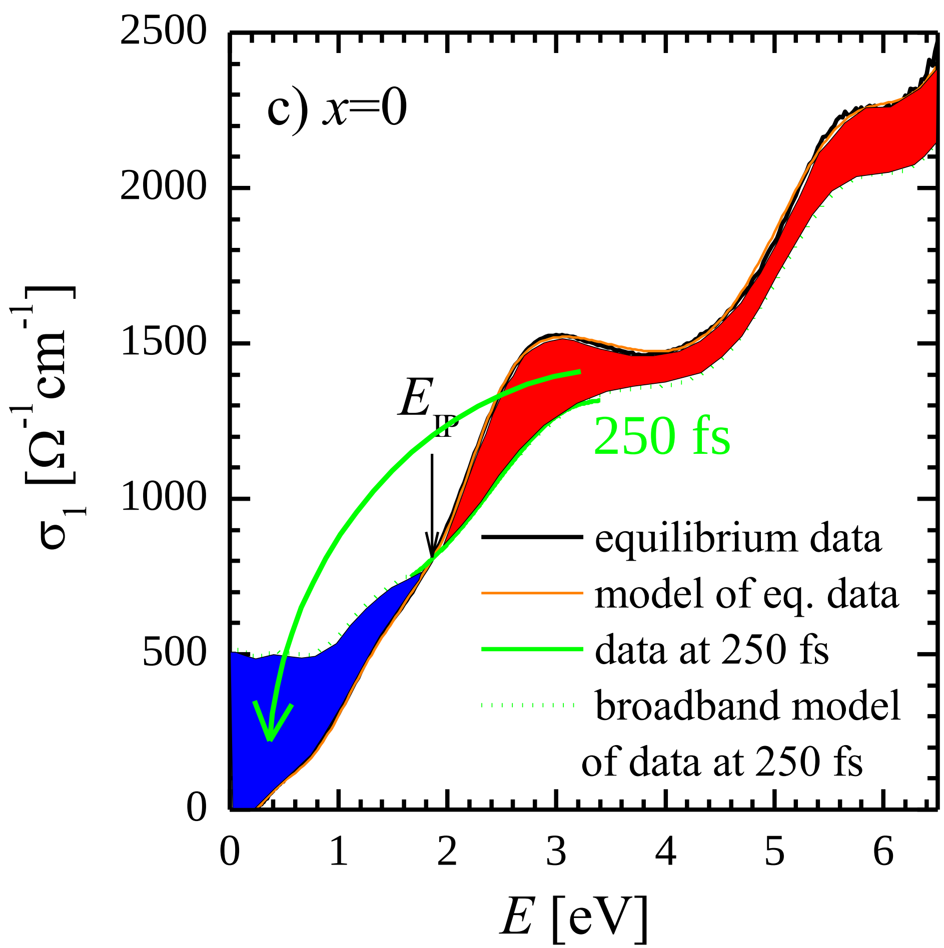


Kramers-Kronig modeling of $LaCoO_3$ at 250 fs

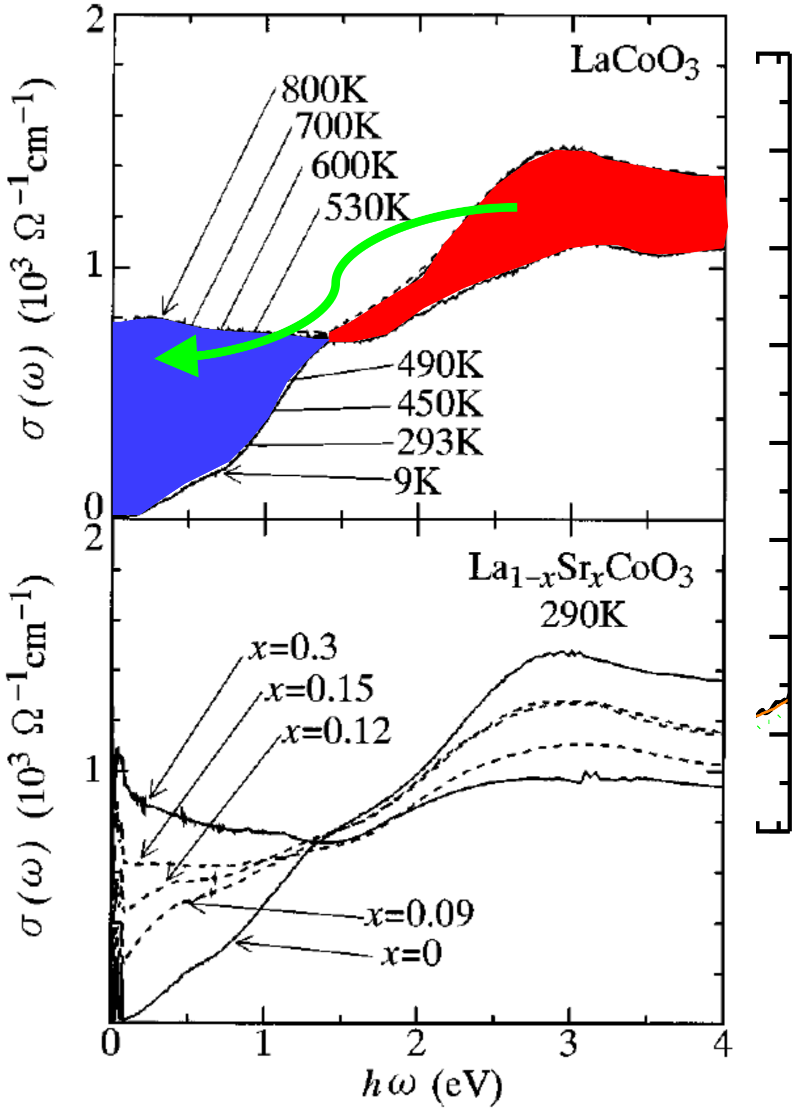


- Modeling 250 fs data with the same model function + Drude term $-\frac{\omega_{pl}^2}{\omega(\omega+i\gamma)}$ $\quad \omega_{pl} = \sqrt{\frac{q^2 n}{\varepsilon_0 m^*}}$
- Modeling yields ω^2_{pl}=3.8±0.1 eV^2 with γ fixed to 1 eV
- For charge per Co ion $N=n^*a^3$, we obtain N=0.15 with $m^*=m_e$
- The modelling strongly suggest that **pump-induced insulator-to-metal transition takes place**

Kramers-Kronig modeling of $LaCoO_3$ at 250 fs



Y. Tokura et al., PRB R1699 (1998)



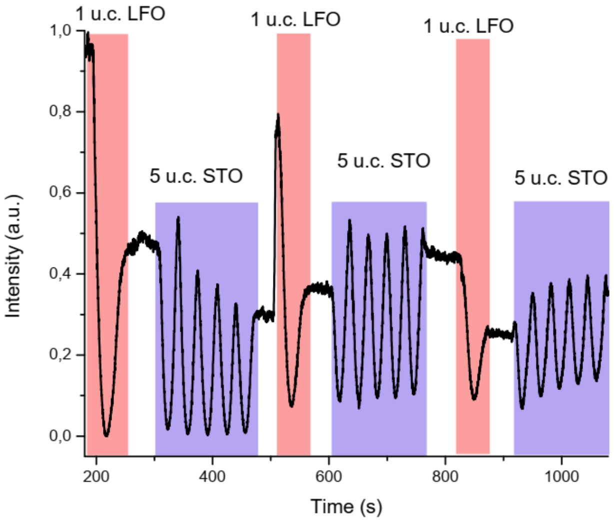
- pump induces shift of spectral weight to low frequencies just like the with temperature
- Observation of pump-induced insulator-to-metal transition

3D to 2D crossover in antiferromagnetic $LaFeO_3$/SrTiO3 superlattices

- $LaFeO_3$ – G-type antiferromagnet with T_N=740 K
- $SrTiO_3$ – nonmagnetic insulator (semiconductor)
- substrate - $SrTiO_3$

Series of superlattices:
$[(LaFeO_3)_N + (SrTiO_3)_5] \times 10$
with N = 3, 2, 1
Grown by M. Kiaba using
PLD with in-situ RHEED monitoring

RHEED pattern during growth



Atomically flat SrTiO3

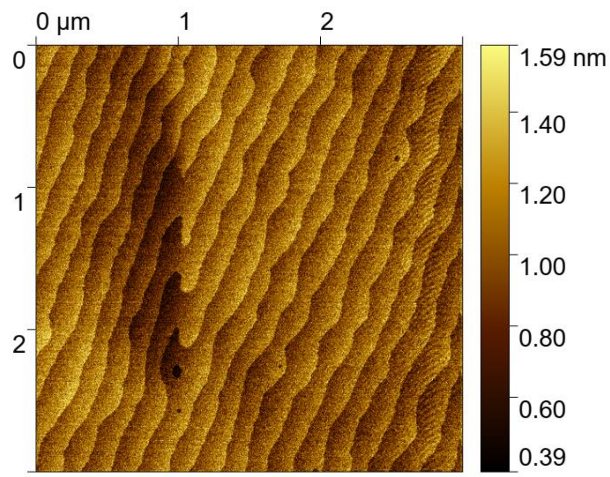


FIG. 5. Typical surface morphology of $SrTiO_3$ substrate made by AFM.

Atomically flat superlattice

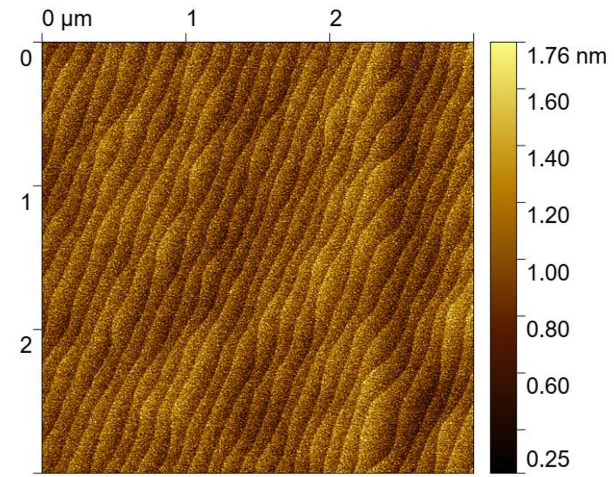
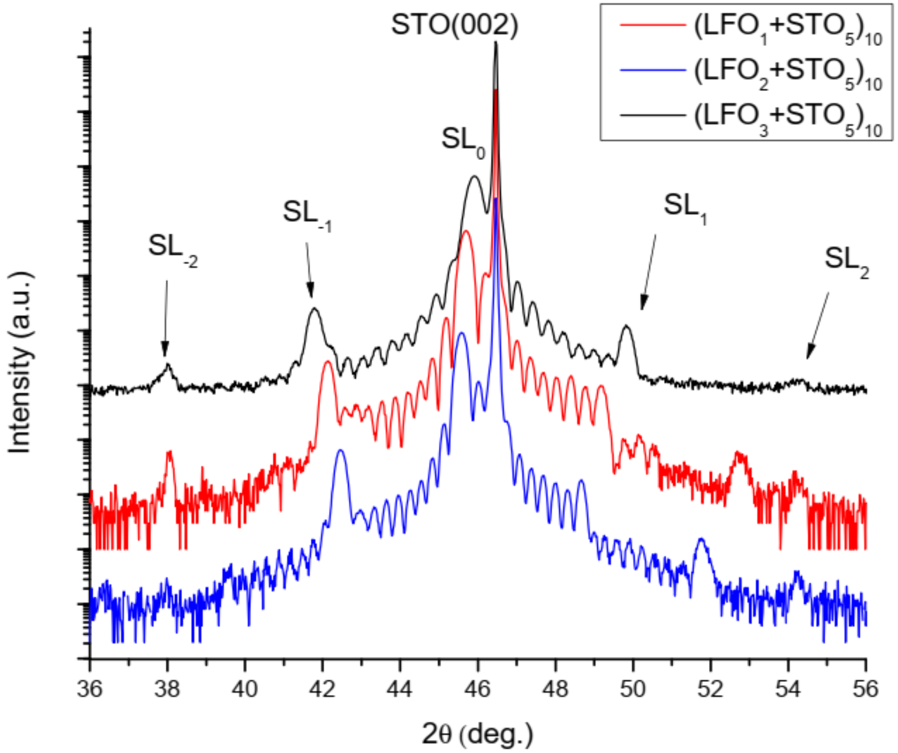


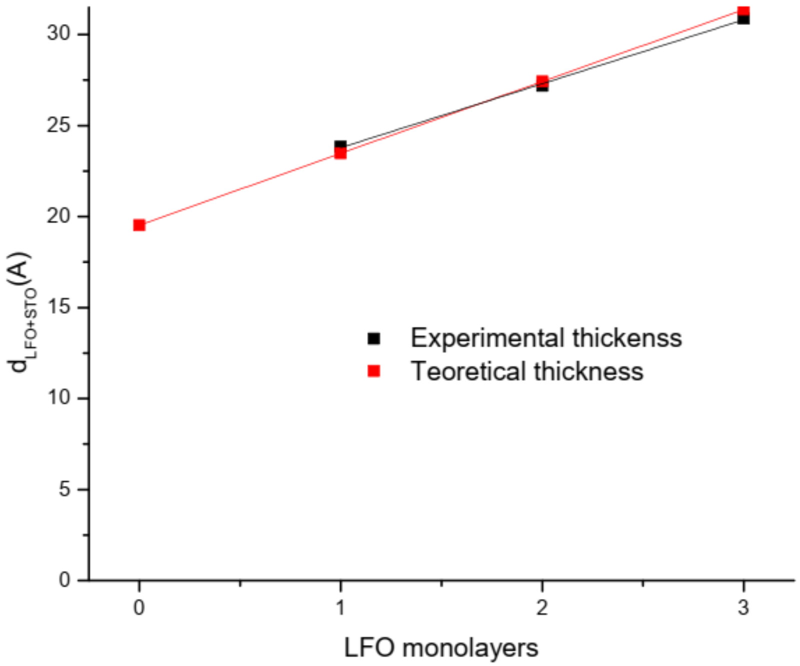
FIG. 4. Typical surface morphology of $(LFO_m/STO_5)_{10}$ made by AFM.

3D to 2D crossover in antiferromagnetic $LaFeO_3$/SrTiO3 superlattices

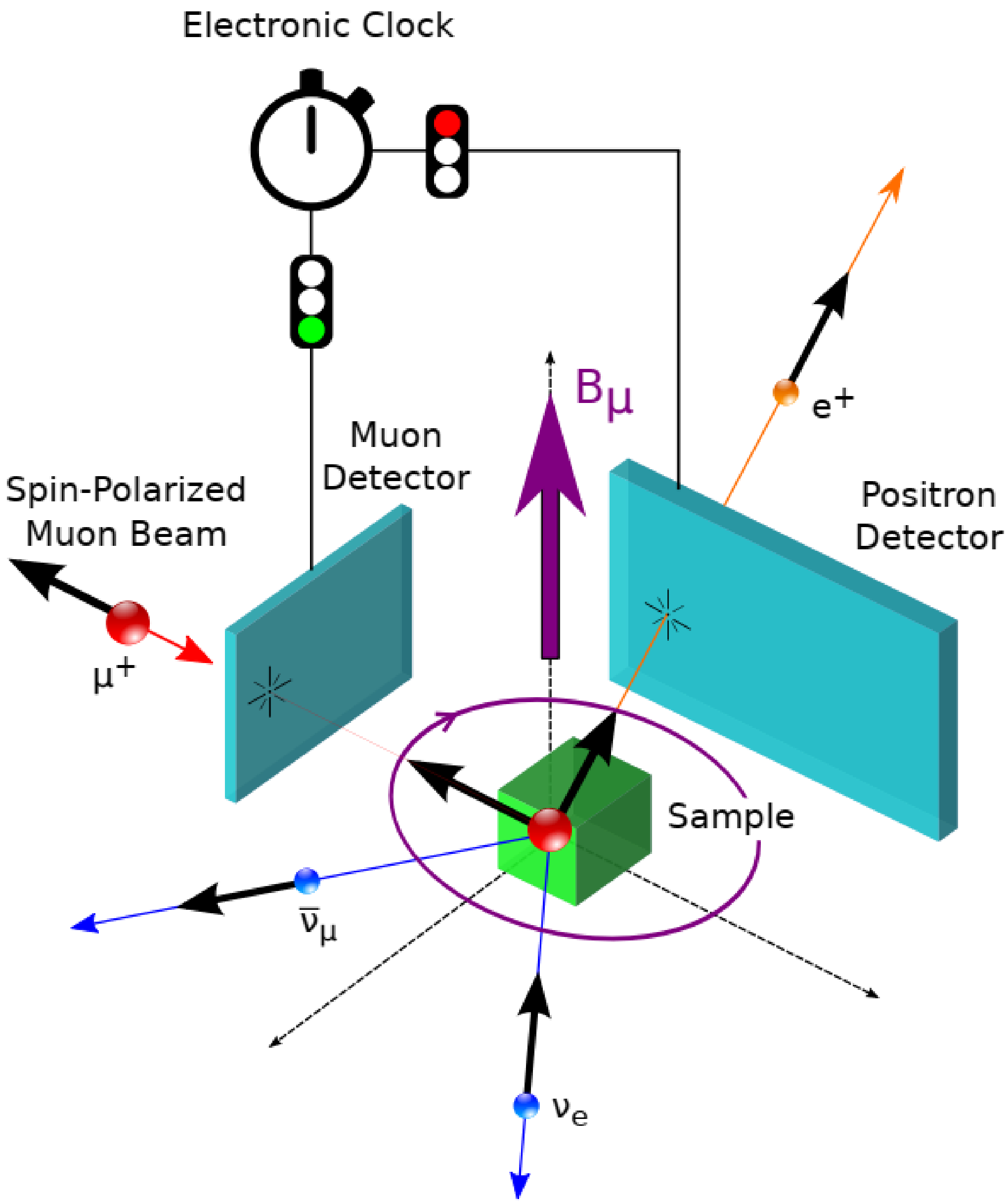
X-ray diffraction showing supelattice peaks and Kiessig fringes



Thickness of LFO+STO bilayer confirms order of suplattice on atomic level



Low-energy muon spin rotation (PSI, Villigen)

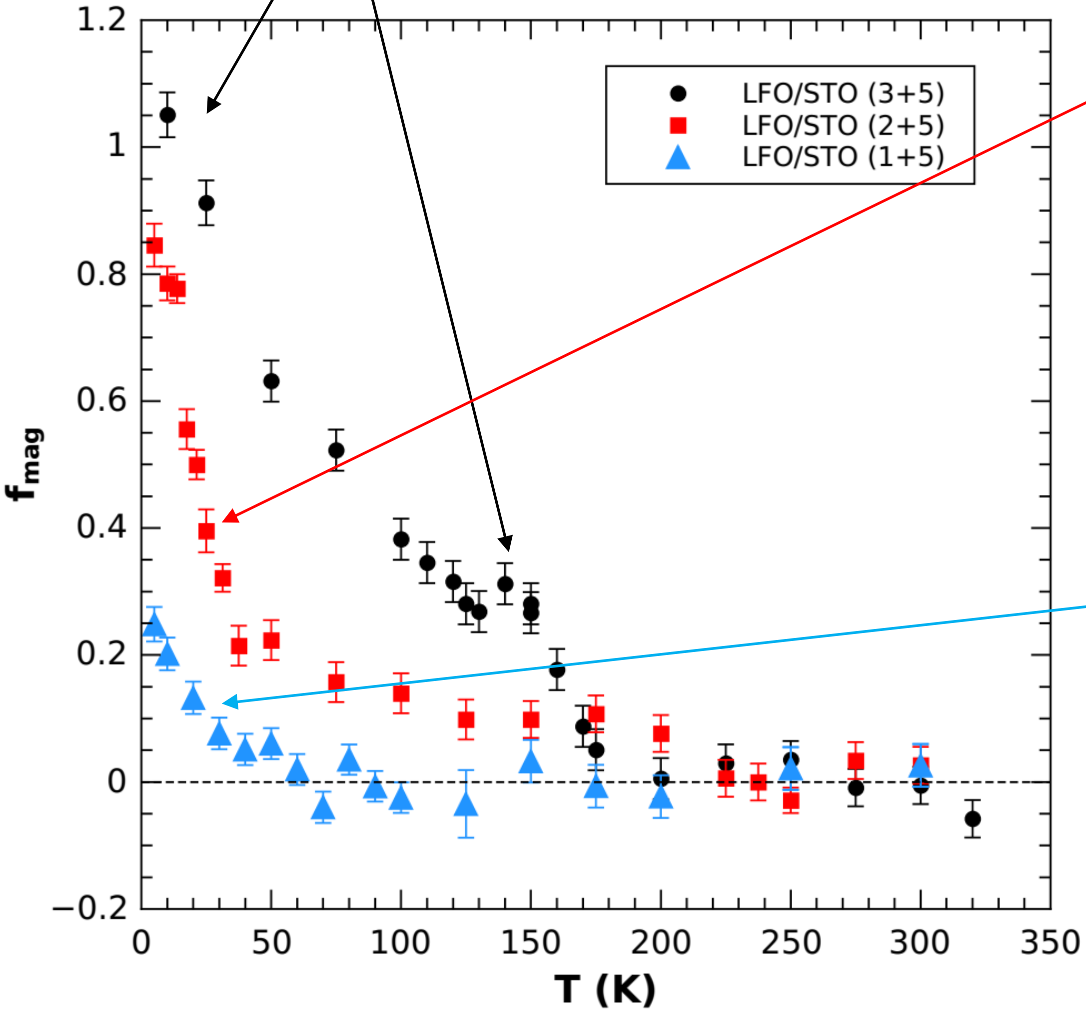


- Can determine magnetic volume fractions of antiferromagnetic order

Drawing A. Sutter, PSI

Low-energy muon spin rotation (PSI, Villigen)

Data on ***N*=3,** $(LaFeO_3)3+(SrTiO_3)5$
- 100% anftiferromagnetic volume fraction of LaFeO3 with T_N=160 K



Data on ***N*=2,** $(LaFeO_3)2+(SrTiO_3)5$
- an intermeditate 3D-2D crossover
- fluctuating AFM order above 50 K.

Data on ***N*=1**, $(LaFeO_3)1+(SrTiO_3)5$
- almost fully suppressed magnetic order down to 2 K
- Probably due to increased spin fluctuation due to Mermin-Wanger theorem

Magneto-spectroscopy on topological insulator Bi_2Te_3

optical spectroscopy: A. Dubroka (MU Brno)
M. Orlita (LNCMI Grenoble),
I. Mohelský (LNCMI Grenoble, BUT Brno)

sample growth: G. Springholz (Uni Linz)

MUNI

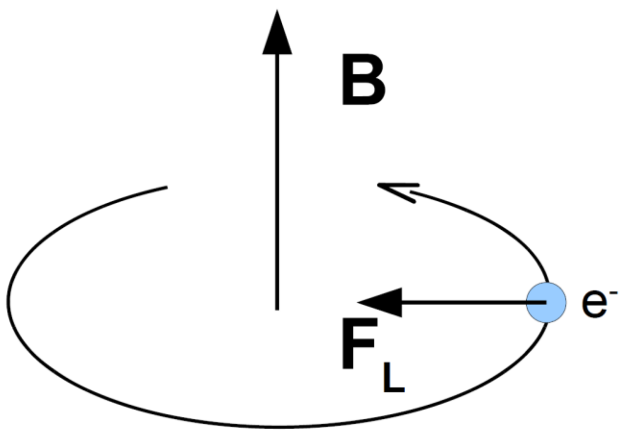
CEITEC
central european institute of technology
BRNO | CZECH REPUBLIC

LNCMI

JOHANNES KEPLER UNIVERSITY LINZ | JKU

Cyclotron frequency

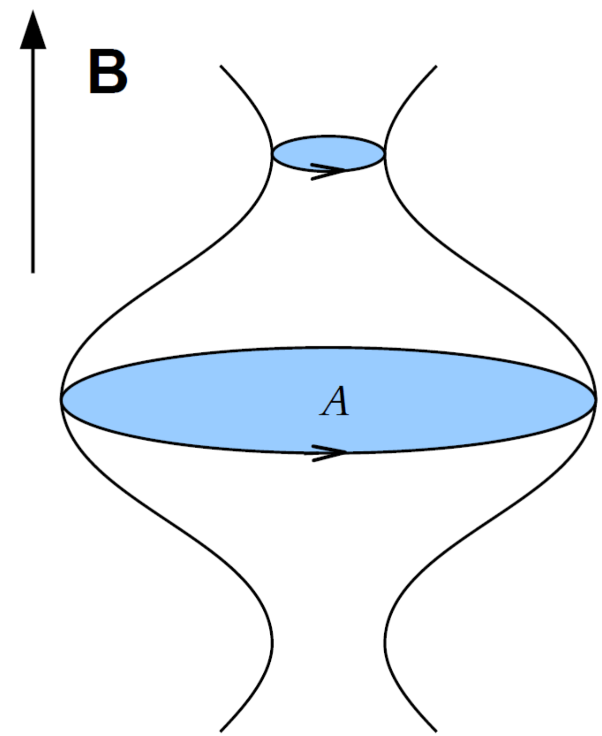
Classical free electron in magnetic field



$$\omega_c = \frac{eB}{m}$$

Cyclotron frequency

Electrons in a solid



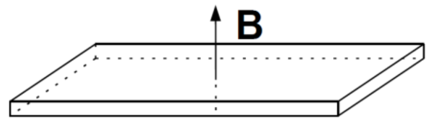
$$\omega_c = \frac{eB}{m_c}\,, \qquad m_c = \frac{\hbar^2}{2\pi}\frac{\partial A(E)}{\partial E}$$

A = surface of the orbit

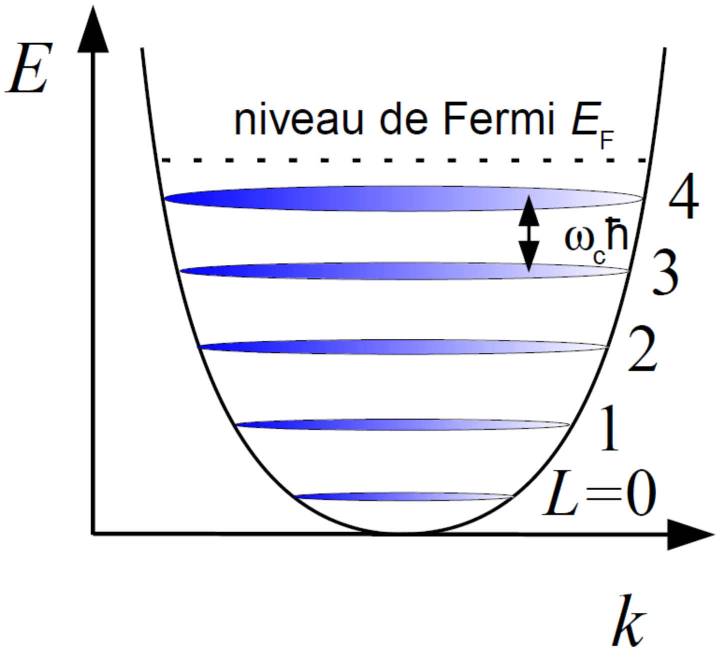
m_c = cyclotron mass

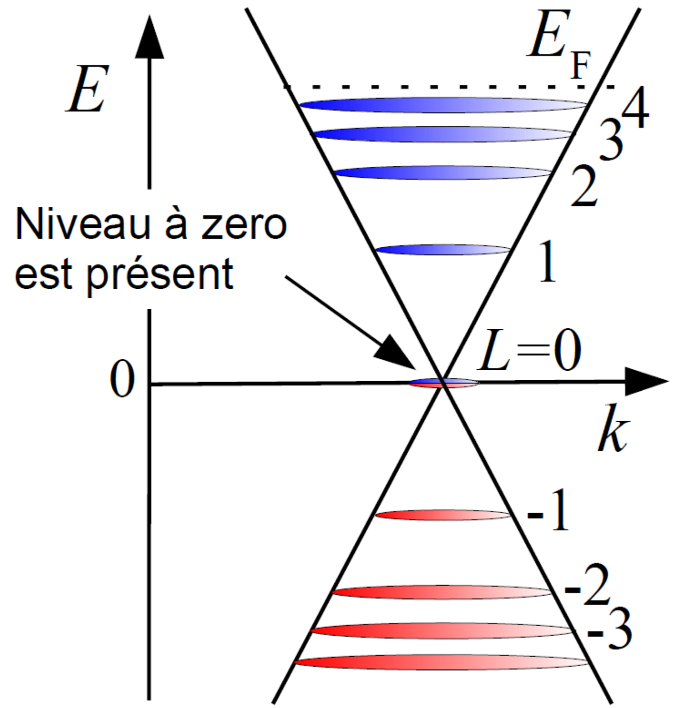
Landau levels in two dimmensions

Electron gas in 2D:
with parabolic dispersion



relativistic particles with zero mass:
linear dispersion





$$E(k) = \frac{\hbar^2 k^2}{2m} \longrightarrow$$

$$E_L = \hbar\omega_c(L + \frac{1}{2})$$

$$L = 0, 1, 2 \ldots \ , \quad \omega_c = \frac{eB}{m_c}$$

L.D. Landau, Z. Phys. 64, 629 (1930)

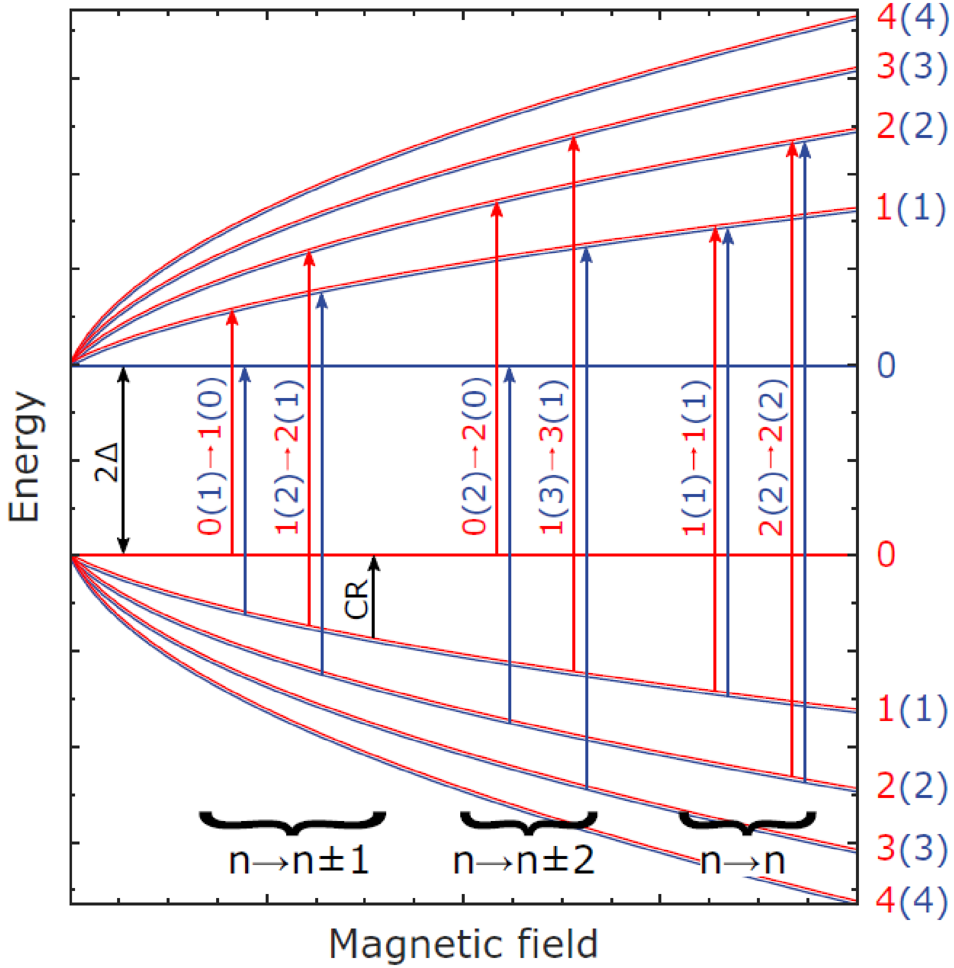
$$E = \pm v_F \hbar k \longrightarrow$$

$$E_L = \mathrm{sgn}(L)\sqrt{2e\hbar v_F^2 |LB|}$$

$$L = 0, \pm 1, \pm 2, \ldots$$

I.I. Rabi, Z. Phys. 49, 507 (1928)

Landau levels of two band model of Dirac Fermions



Two band model:

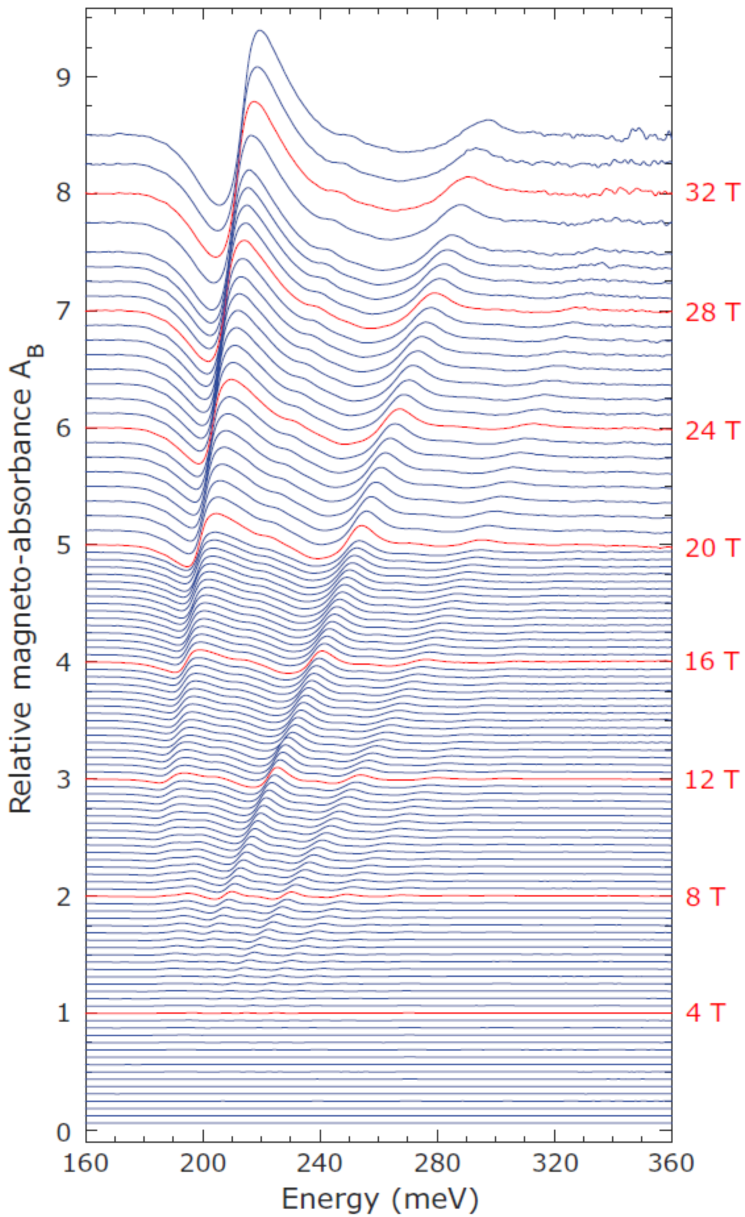
$$E(k) = \pm\sqrt{\Delta^2 + \hbar^2 v_D^2 k^2}$$

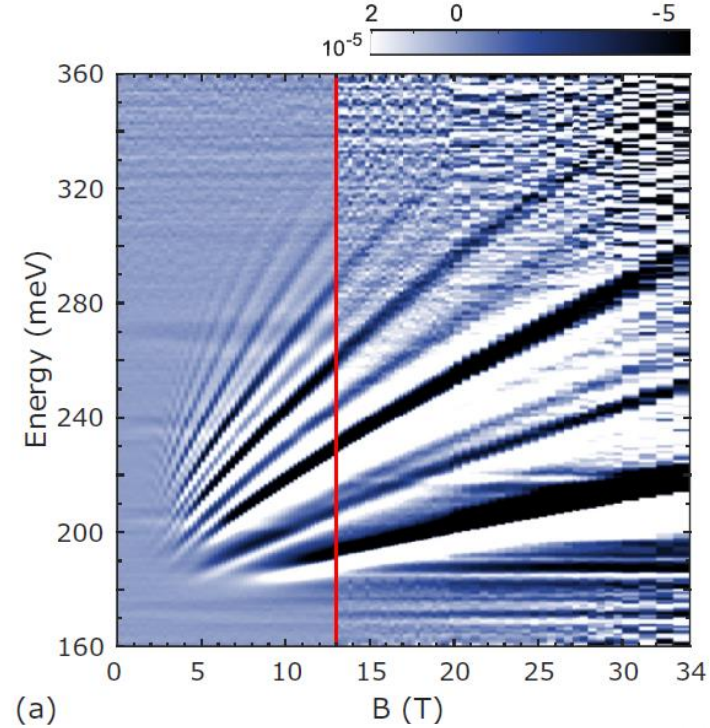
Landau level spectrum

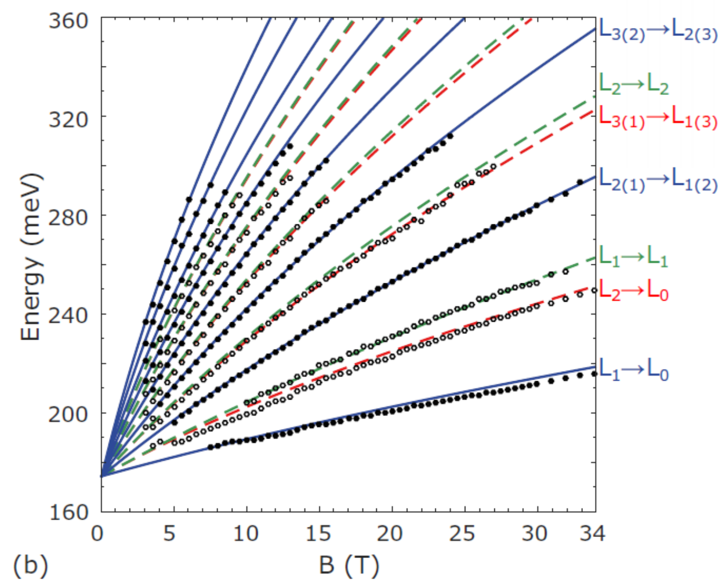
$$E_n = \pm\sqrt{v_D^2 2e\hbar B n + \Delta^2}, \text{ where } n > 0$$

Selection rule $n \to n \pm 1$

Magneto-transmission in high magnetic fields (Grenoble)

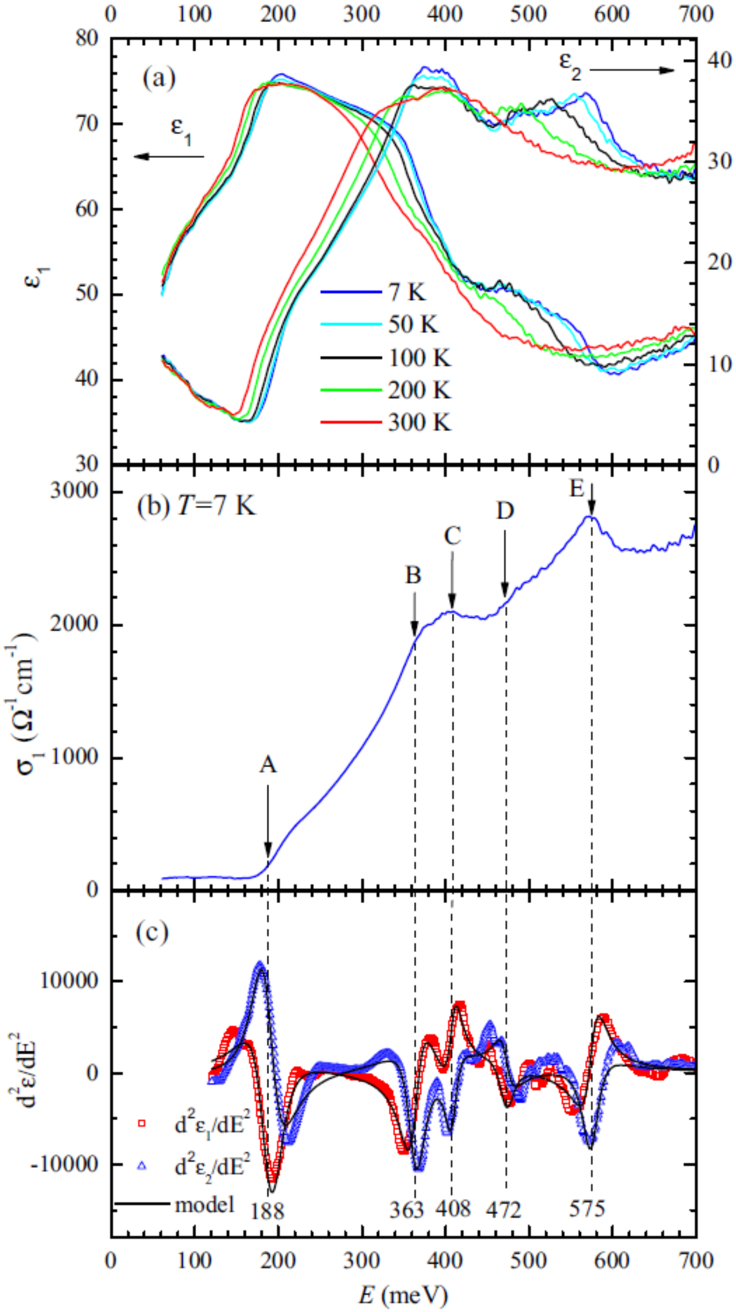


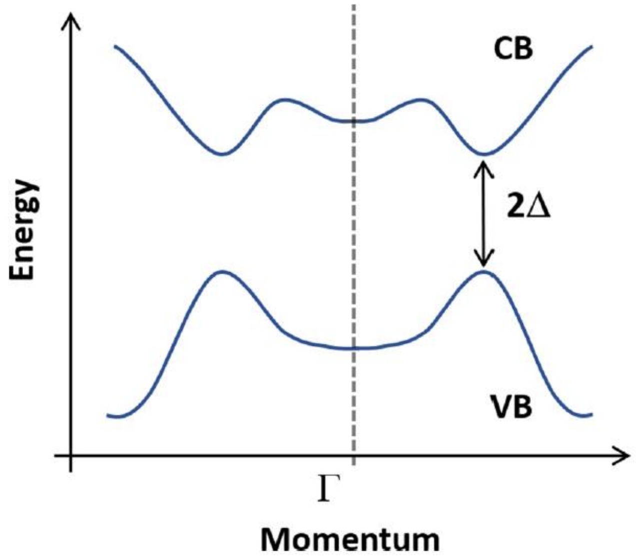




I. Mohelský etal., Physical Review B **102**, 085201 (2020).

Analysis of critical points of bandstructure of Bi_2Te_3





$$\frac{d^2\varepsilon}{dE^2} = Ae^{i\phi}(E - E_{\mathrm{CP}} + i\zeta)^{n-2}$$

TABLE I. The values of the amplitude A, energy E_{CP}, broadening ζ, and phase ϕ obtained from the fit of the CP model to the data shown in Fig. 10(c).

Label	A	E_{CP} (meV)	ζ (meV)	ϕ (deg)	Line shape
A	7.8	188	24	−29	2D
B	21 $\mathrm{eV}^{-1/2}$	363	16	23	3D
C	8 $\mathrm{eV}^{-1/2}$	408	11	76	3D
D	6 $\mathrm{eV}^{-1/2}$	472	13	300	3D
E	16 $\mathrm{eV}^{-1/2}$	575	15	60	3D

I. Mohelský etal., Physical Review B **102**, 085201 (2020).

Děkuji za pozornost

Historie elipsometrie

Paul Karl Ludwig Drude
1863-1906

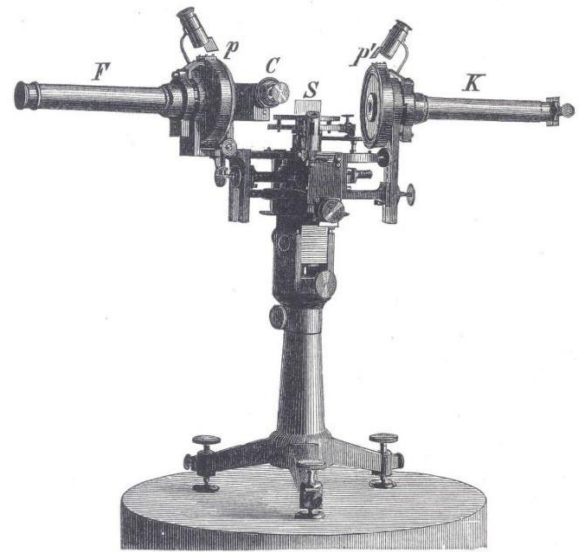
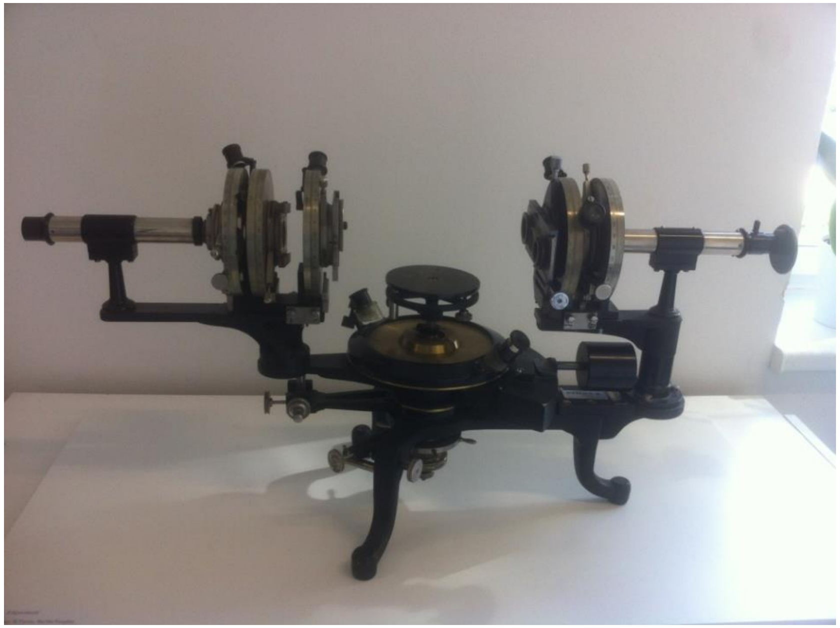


Fig. 81.

~ 1900

Prof. Antonín Vašíček
1903-1966



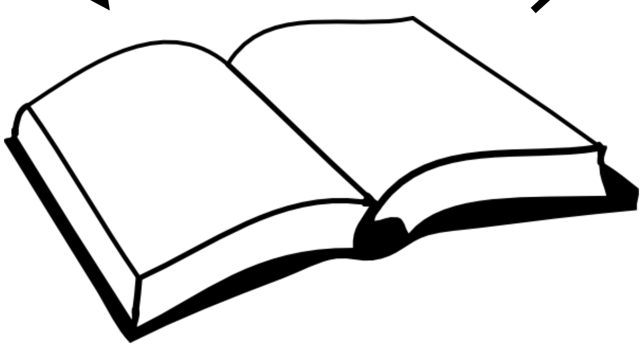
~ 1938

Spektroskopie: Studium interakce mezi látkou a sondou s určitou *energií*

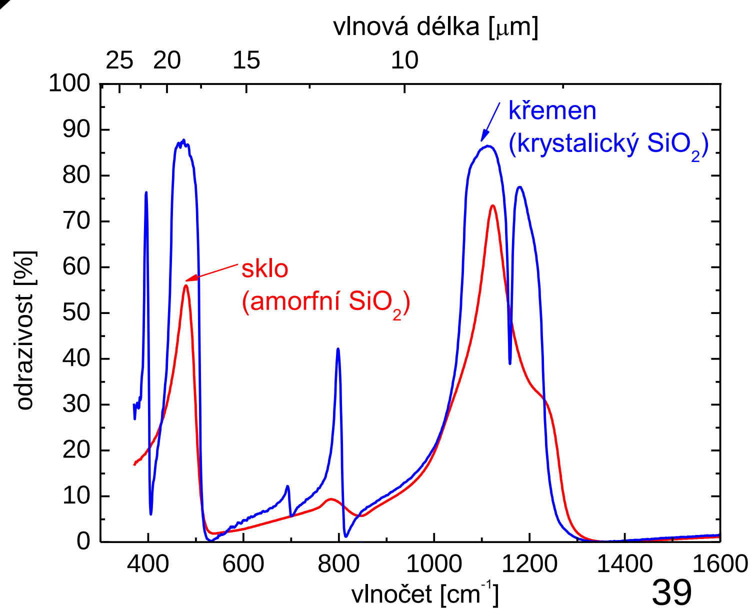
zdroj fotonů,
elektronů,
neutronů,
atomů

částice
po interakci
s objektem

detektor
a **spektrometr**



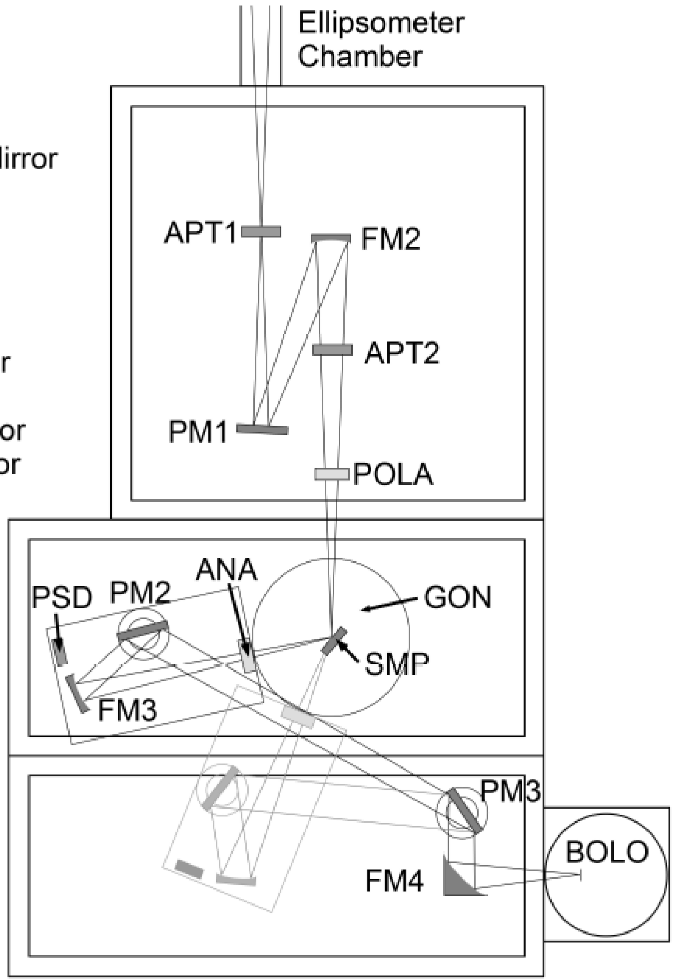
studovaný objekt

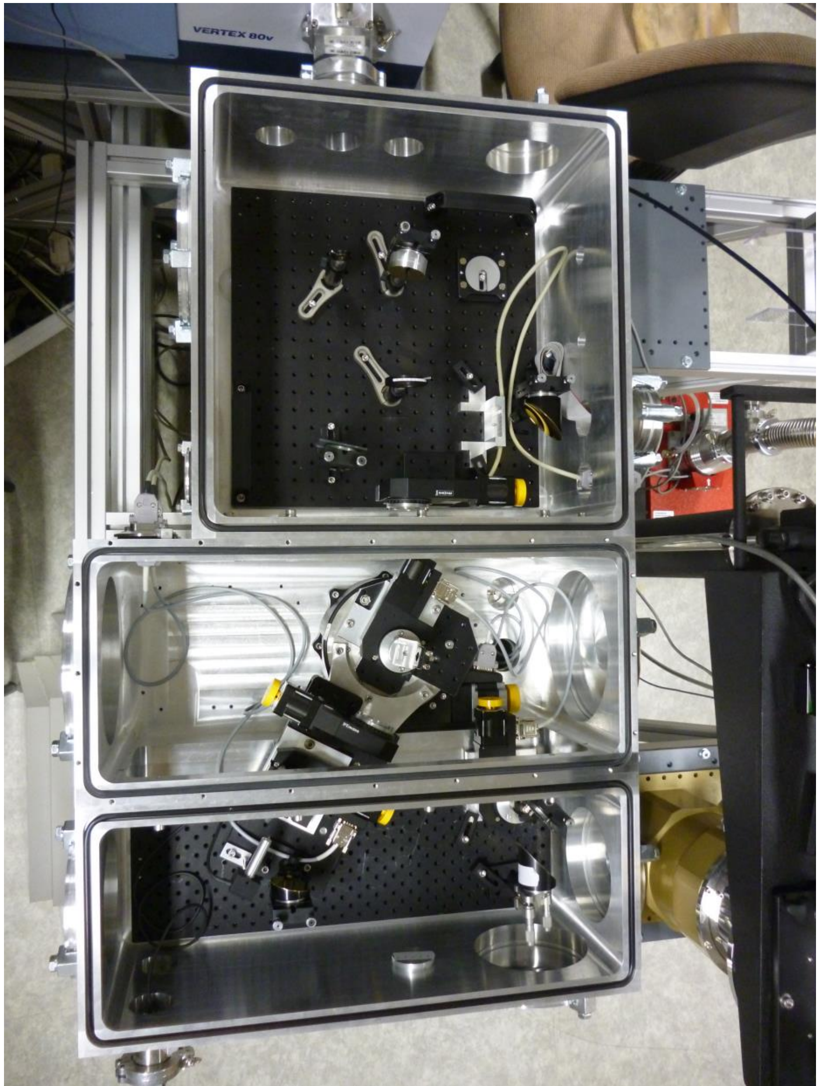


design of far-infrared ellipsometer at CEITEC

alternated Max-Planc design
- three chambers to support top loaded cryostat
- detection arm on a goniometer for reproducible exchange of angle of incidence

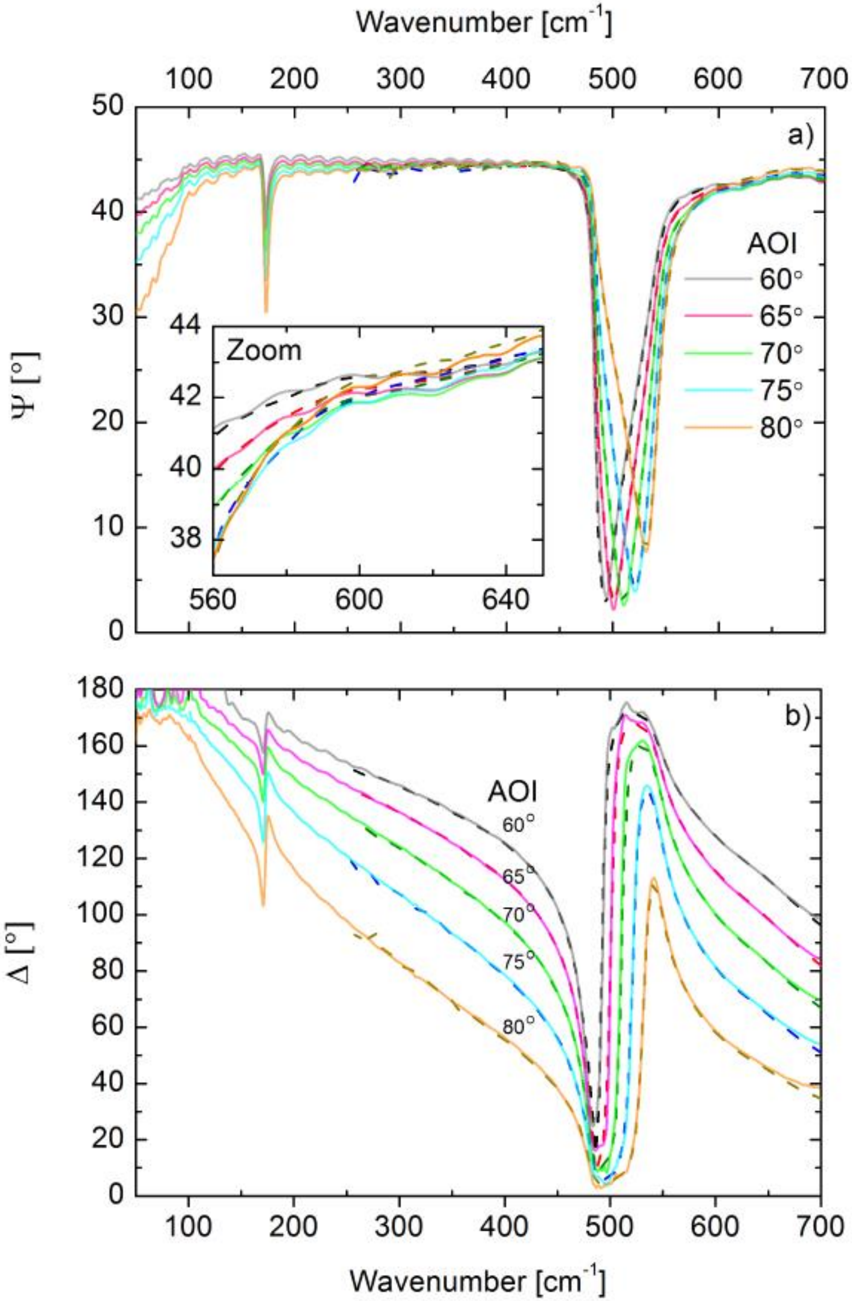
ANA - Analyzer
APT1,2 - Aperture
BMS - Beam Splitter
BOLO - Bolometer
FM1,3,4 - Parabolic Mirror
FM2 - Elliptical Mirror
GLB - Glow Bar
GON - Goniometer
HG - Mercury Lamp
LAS - Aligment Laser
PM1,2,3 - Plane Mirror
POLA - Polarizer
PSD - Position Detector
RM - Removable Mirror
SMP - Sample Holder
W - Tungsten Lamp





P. Friš a A. Dubroka, Appl. Surf. Science **421**, 430 (2017)

Testing performance on $SrTiO_3$ single crystal



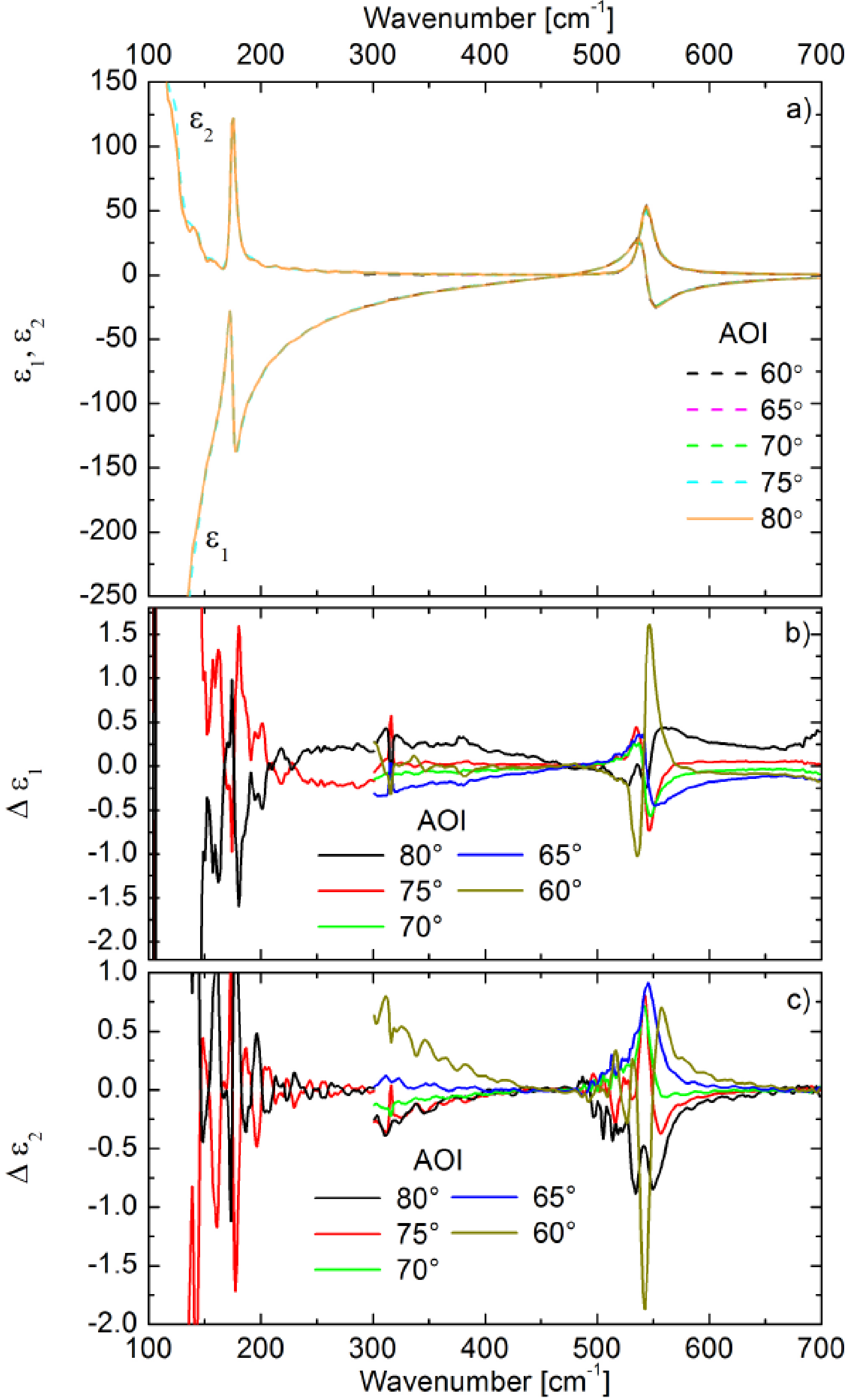
- Measured Ψ and Δ in comparison with those measured with Woollam IR-VASE.

- The difference is less than 0.2° in Ψ and less than 1° in Δ.

- Reproducibility in Ψ is better than 0.1° and better than 0.3° in Δ.

P. Fřiš a A. D., Appl. Surf. Sci. 421 (2017) 430

dielectric function of SrTiO3



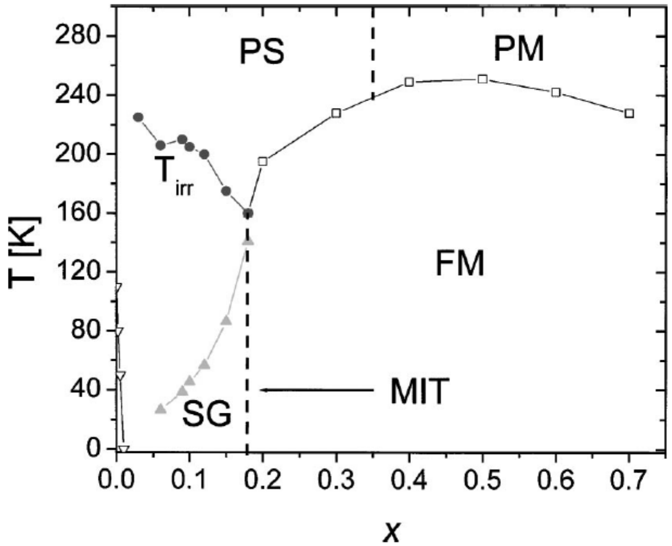
- obtained dielectric function of SrTiO3 from different angles of incidence
- agreement down to factor about 1:100

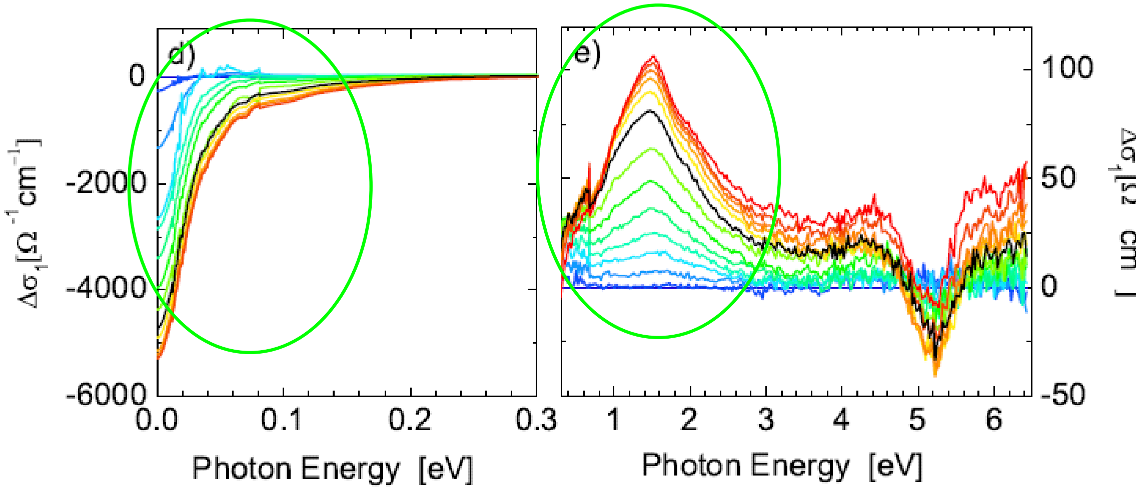
P. Fřiš a A. D., Appl. Surf. Sci. 421 (2017) 430

Jak vypadá přechod se „špatným spinem“ v $La_{1-x}Sr_xCoO_3$ pro další úrovně koncentrací stroncia?

• elektronová struktura (Drude a “wrong spin transition”) feromagnetického stavu v $La_{1-x}Sr_xCoO_3$ v závislosti na koncentraci děr x. Evoluce z izolujícího stavu (x=0) do feromagnetu (x>0)

• dopování x=0.3

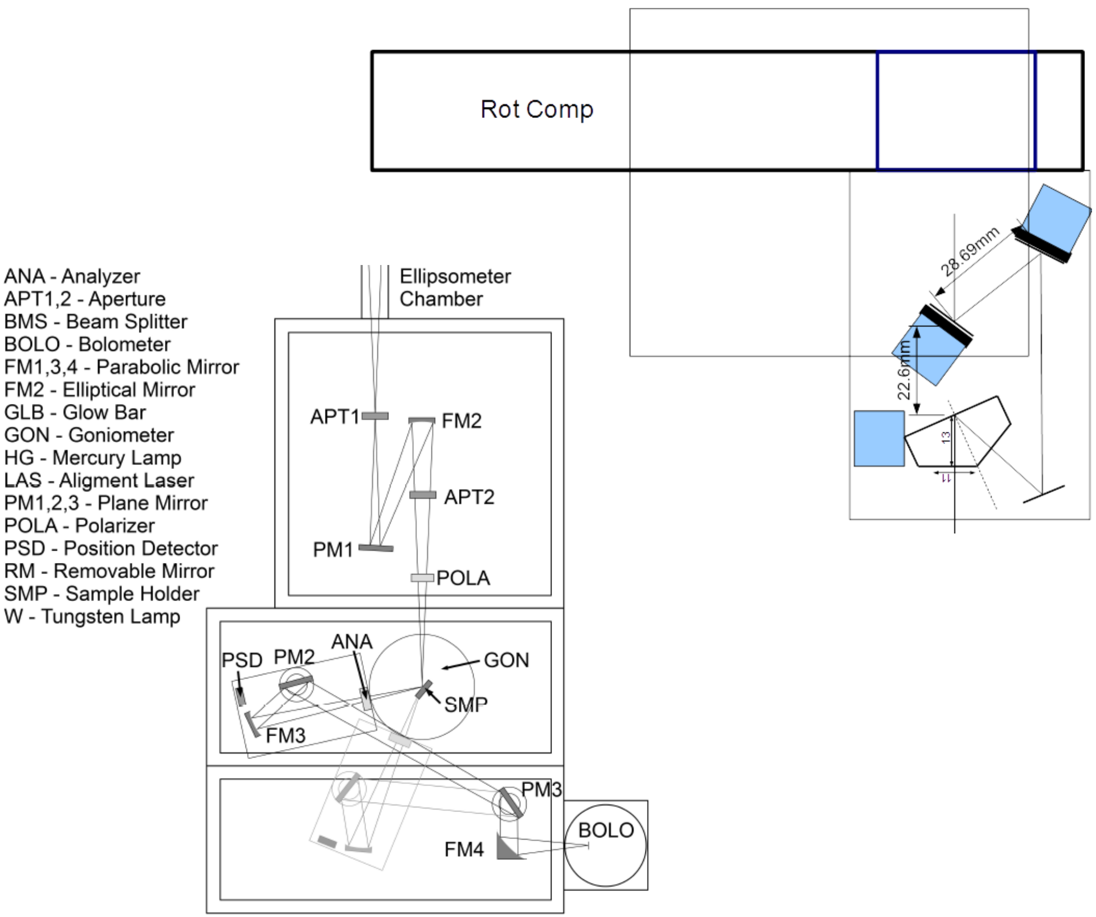


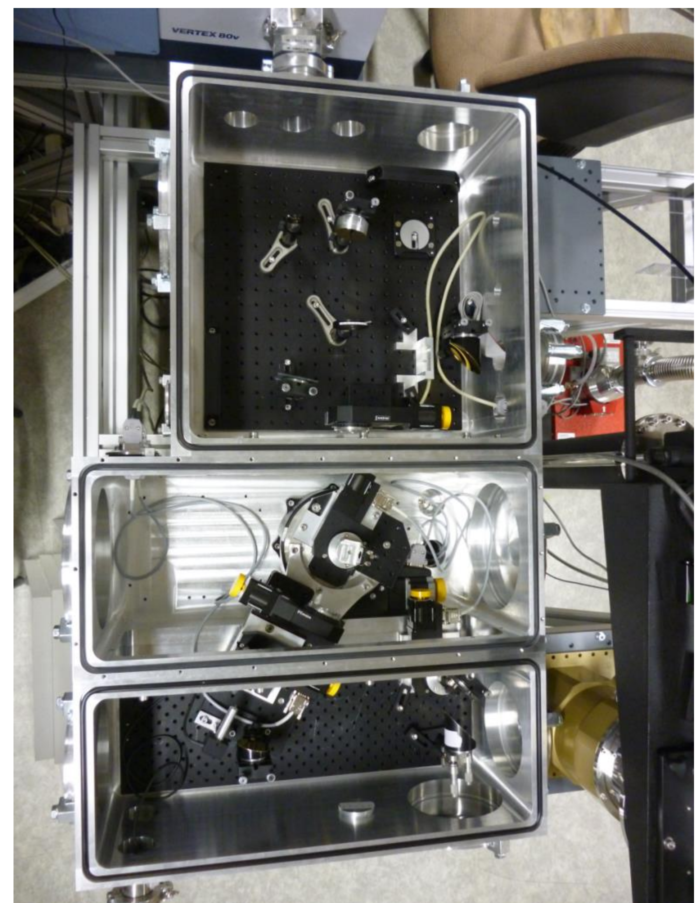


• dopování x=0.5, 0.7, a 0.2

Rotační kompenzátor (čtvrtvlnová destička) pro FIRový elipsometr

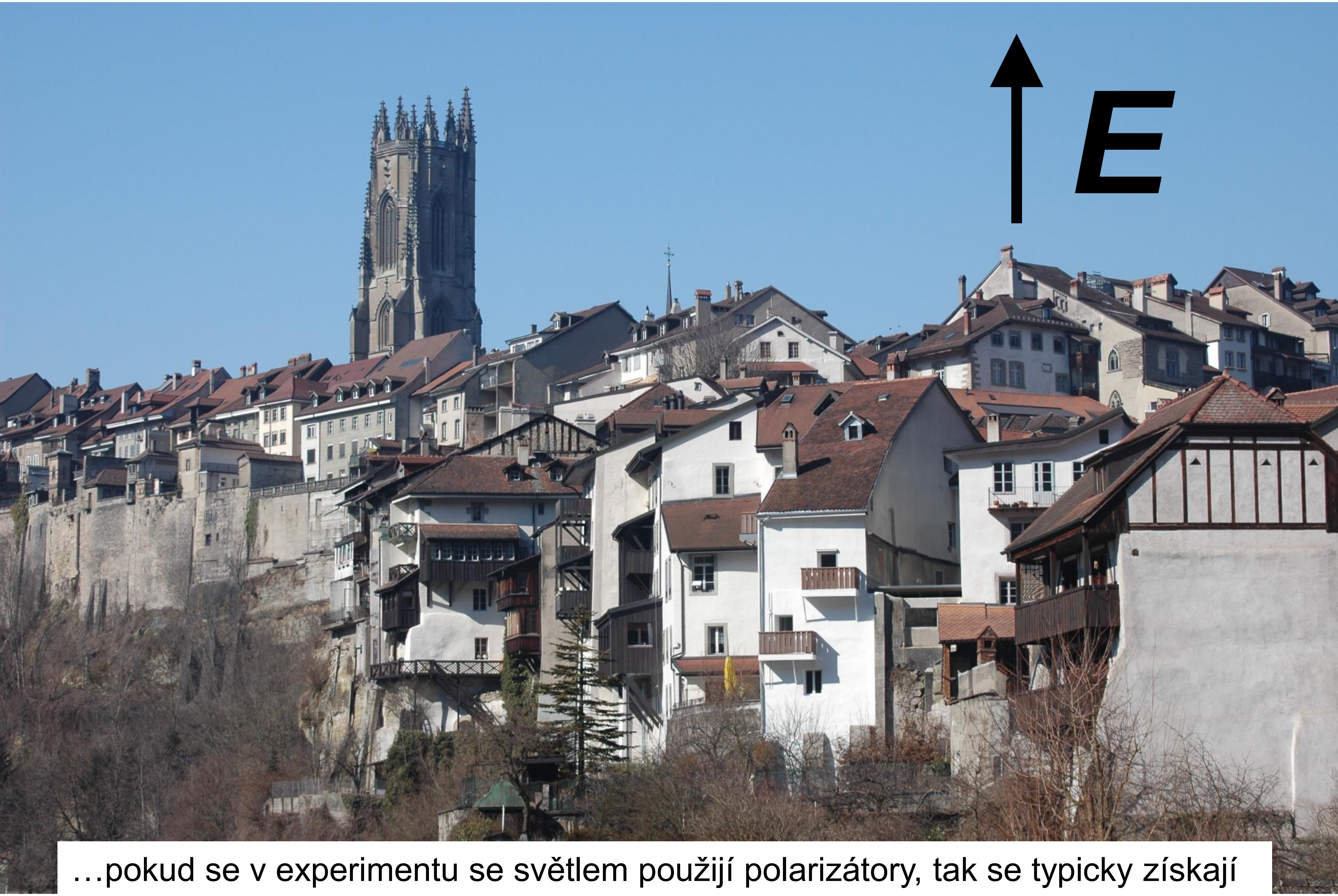
• kompenzátor (čtvrtvlnová „destička“) pro FIRový elipsometr
• umožňuje provádět elipsometrii s „rotačním kompenzátorem“, která umožňuje měřit depolarizaci a lépe měřit Δ v celém oboru
• pouze jediný další takový elipsometr na světě (Brookhaven, USA)





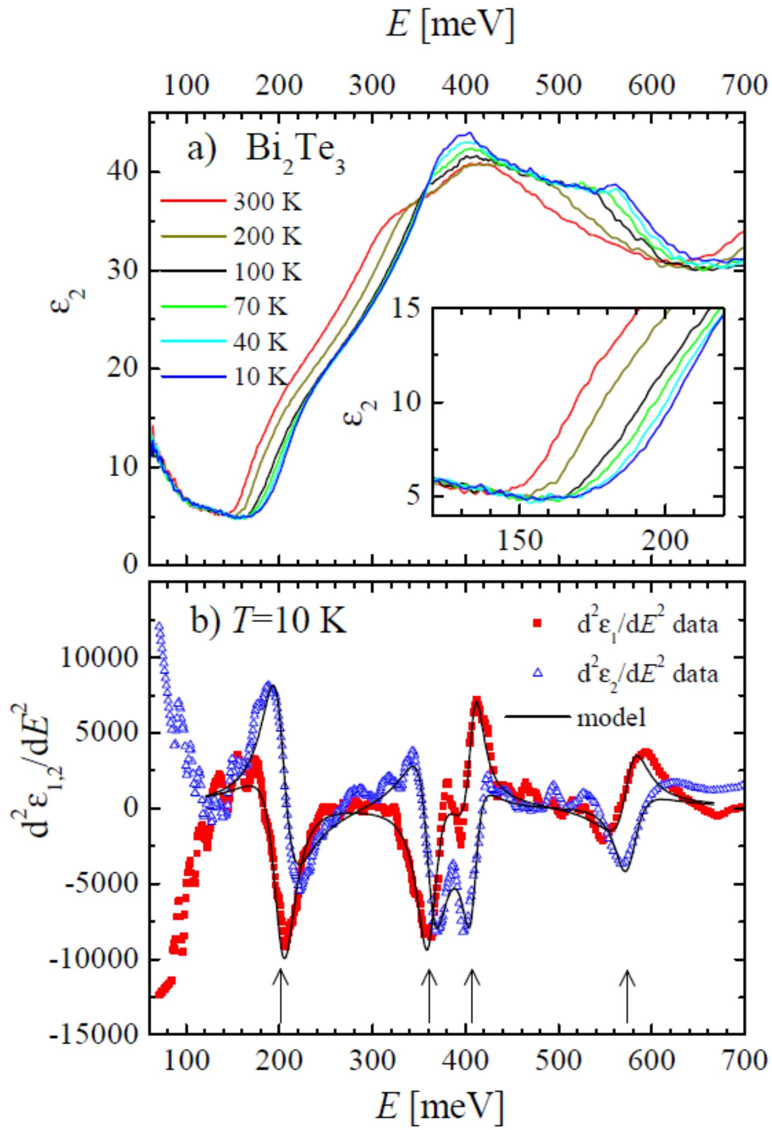
dodatky

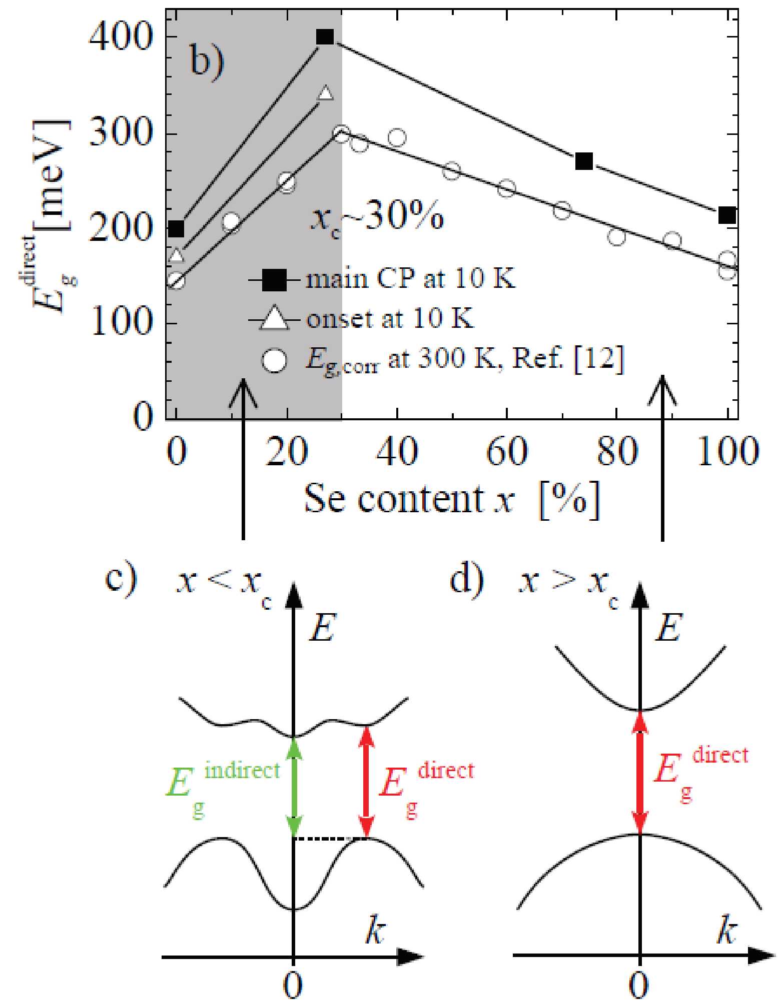

E



…pokud se v experimentu se světlem použijí polarizátory, tak se typicky získají nové informace

Absorpční hrana v topologických izolátorech $Bi_2Se_xTe_{1-x}$





Druhé derivace kritických bodů modelovány funkcí

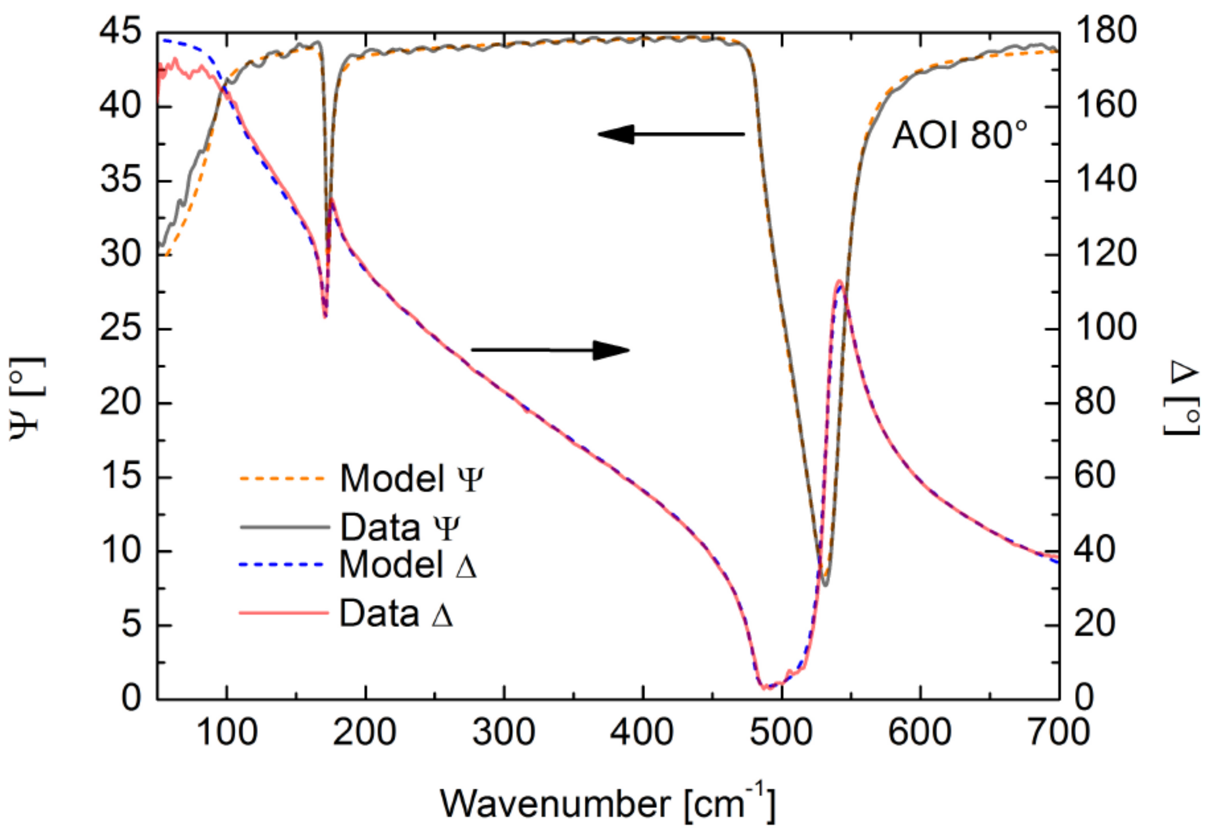
$$\frac{d^j \hat{\varepsilon}(E)}{dE^j} = Ae^{i\phi}(E - E_{\mathrm{CP}} + i\Gamma)^{-n-j},$$

A. D. et al, Phys. Rev. B, 96 235202 (2017)

automated variable angle far-infrared ellipsometer

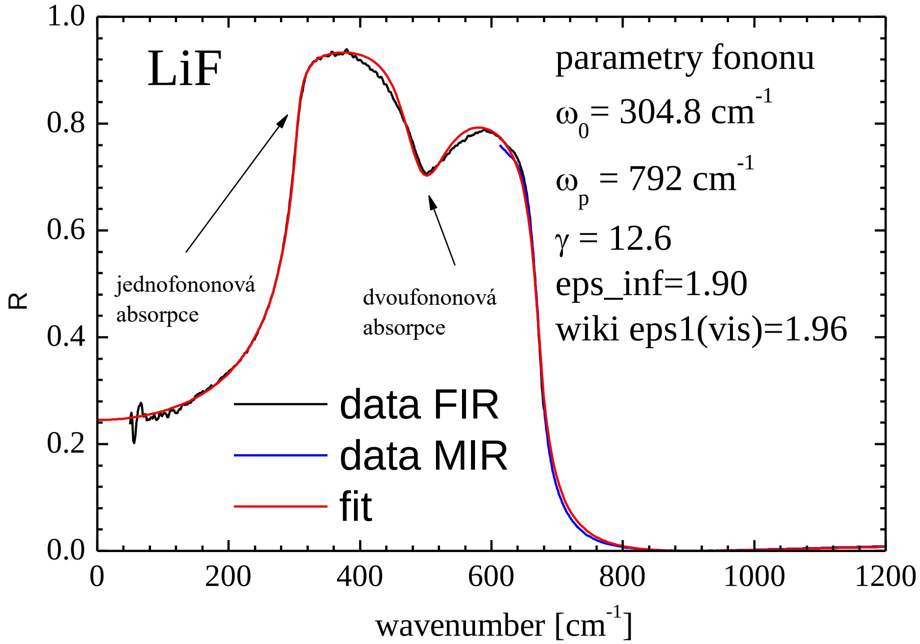
- Fitting of data at AOI = 80º with complex lorentzians demonstrates Kramers-Kronig consistency of our data.
- Effect in Δ below 100 cm-1 is likely an onset of diffraction effects

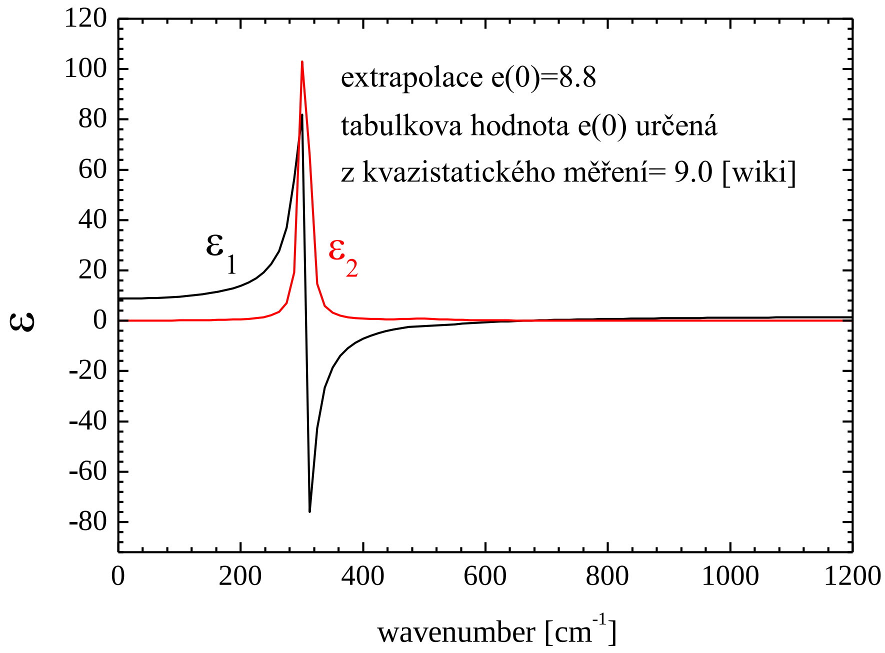
$$\varepsilon(\omega) = \varepsilon_\infty + \sum \frac{\omega_{pl}^2 + \mathrm{i}\omega_c\omega}{\omega_0^2 - \omega^2 - \mathrm{i}\omega\gamma}$$



P. Fřiš a A. D., Appl. Surf. Sci. 421 (2017) 430

ukázka: IČ Reflektivita LiF





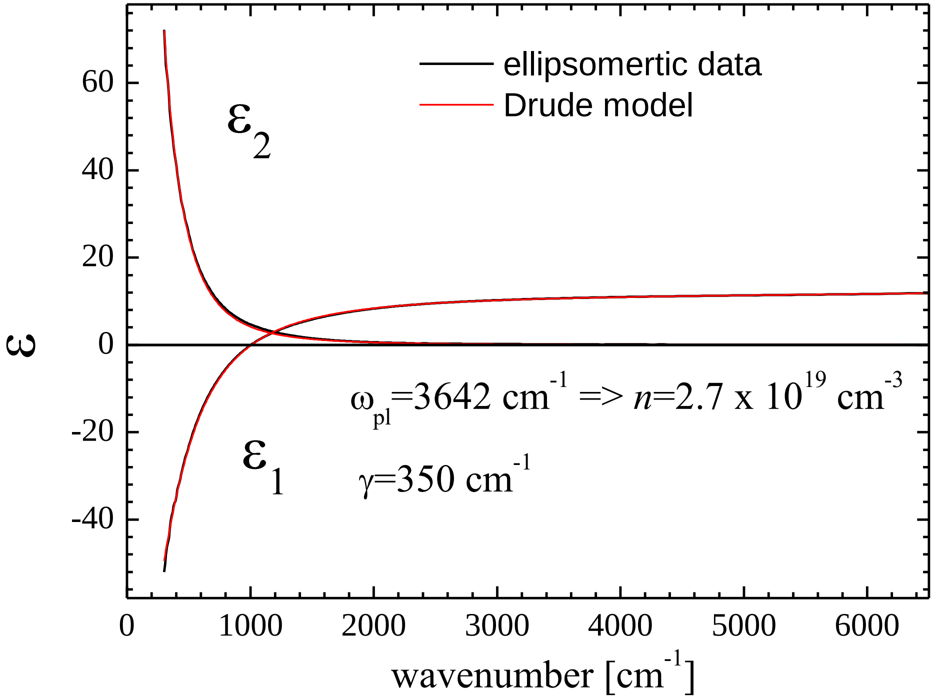
Drude model

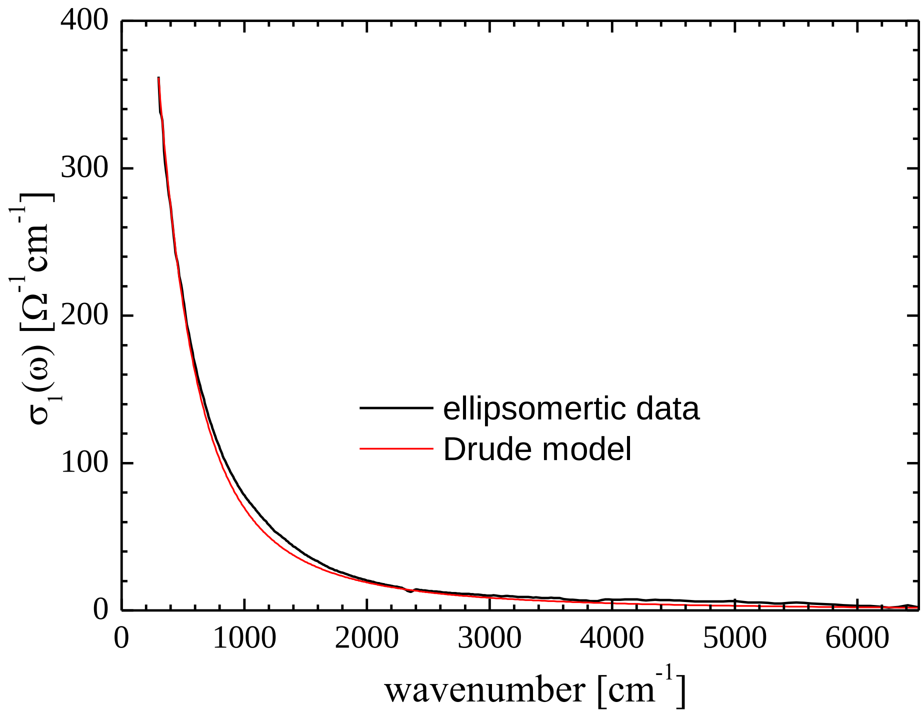
A classical model of dielectric response of free and ***mutually non-interacting*** charge carriers

$$\varepsilon(\omega) = \varepsilon_\infty - \frac{\omega_{\mathrm{pl}}^2}{\omega(\omega + i\gamma)}$$

where ω_{pl} is the plasma frequency $\omega_{\mathrm{pl}} = \sqrt{\dfrac{q^2 n}{\varepsilon_0 m^*}}$

Example on n-doped silicon:





Optical signatures of ferromagnetic state

$La_{0.7}Sr_{0.3}CoO_3$,
T_c ~205 K