

Time-resolved THz spectroscopy on BCS superconductors and ferromagnets

ACKNOWLEDGEMENTS:

M. Beck, I. Rousseau, M. Klammer, P. Leiderer (*Konstanz*)

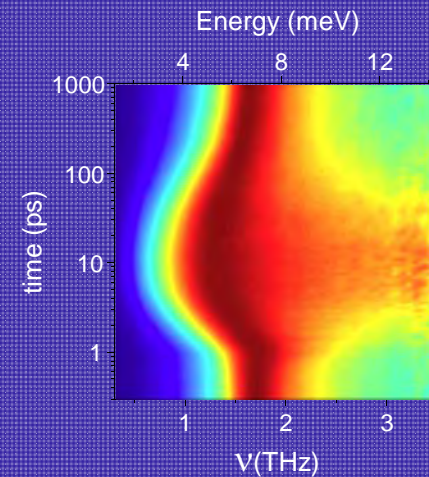
P. Leiprecht, J. Braun, M. Beyer, J. Braun, M. Obergfell,
H. Schäfer, P. Leiderer, S. Egle, T. Pietsch, E. Scheer (*Konstanz*)

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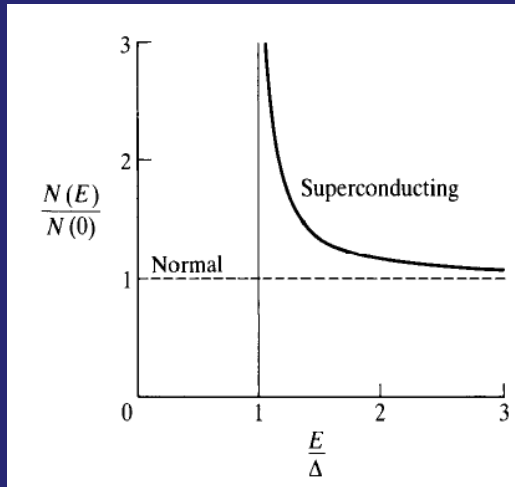


Outline

- Introduction – nonequilibrium superconductivity
- Transient enhancement of superconductivity in a BCS superconductor NbN by resonant THz pumping
- THz conductivity dynamics in SrRuO₃
- Future plans

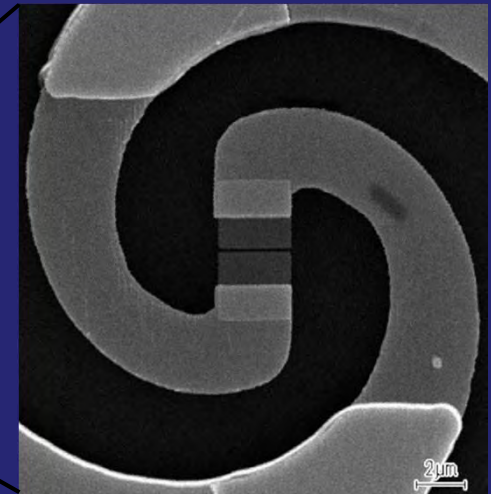
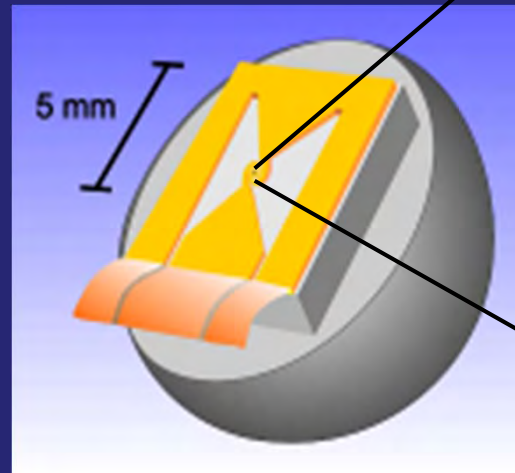
Non-equilibrium Superconductivity

as IR detectors (timescales?)

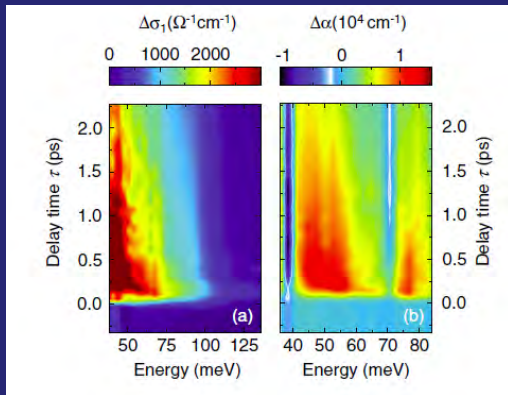
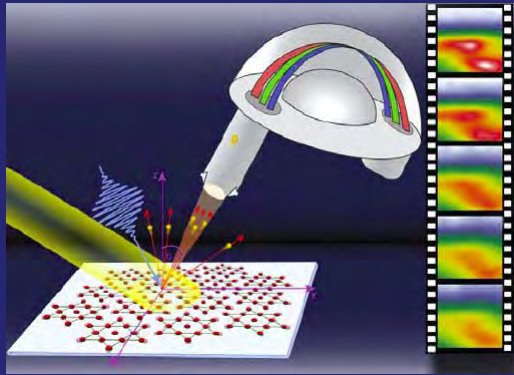


$\Delta \sim 1-10$'s THz

Hot electron bolometer



Real-time studies of Nonequilibrium phenomena in SC



- **Studies of e-ph(boson) coupling constants**

Perfetti, PRL 99, 197001 (2007); Mansart, PRB 82, 024513 (2010)

- **Nature of relaxation processes**

Han, PRL 65, 2708 (1990); Kabanov, PRB 59, 1497 (1999); Gedik, PRL 95, 117005 (2005); Kaindl, PRB 72, 060510 (2005); Cortes, 107, 097002 (2011)....

- **Manipulation of superconductivity**

Dienst, Nature Photon. 5, 485 (2011); Kaiser et al., arXiv:1205.4661, PRL 65, 2708 (1990);

- **Normal state pseudogaps**

Demsar, PRL 82, 4918 (1999); Kaindl, Science 287, 470 (2000). Kusar, PRB 72, 014544 (2005)

- **Theoretical models:**

Rothwarf & Taylor, PRL 19, 27 (1967); Owen & Scalapino, PRB 28, 1559 (1972); Parker, PRB 12, 3667 (1975); Kaplan et al., PRB 14, 4854 (1976); Nicol & Carbotte, PRB 67, 214506 (2003); Howell et al, PRL 92, 037003 (2004); Kabanov et al. PRL 95, 147002 (2005); Unterhinninghofen et al PRB 77, 180509 (2008); Papenkort et al, PRB 78, 132505 (2008)....

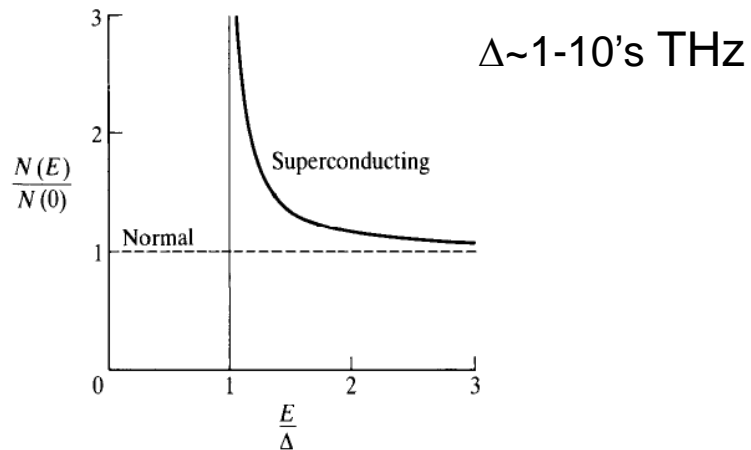
- **Conventional SC:**

Federici, PRB 46, 11153 (1992); Carr, PRL 85, 3001 (2000); Demsar, PRL 91, 267002 (2003); Lobo, PRB B 72, 024510 (2005).

Non-equilibrium Superconductivity

-Nonequilibrium superconductivity (from 1960's on)

as IR detectors (timescales?)



Amplification of SC

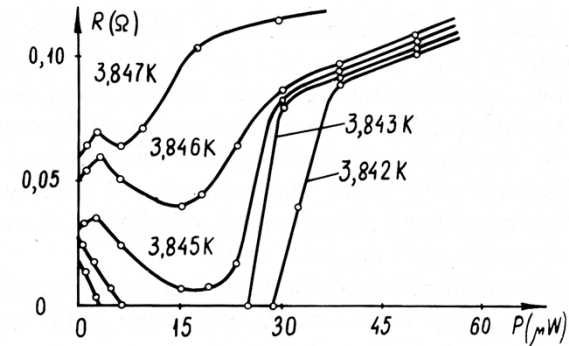


Fig. 2. Experimental dependence of Sn bridge resistance as a function of microwave power at different temperatures. Incident power frequency is ~ 10 GHz. $T_c = 3.84$ K.

- Dynamics in broken symmetry states of matter (superconductors, density waves..) **interaction strength between various degrees of freedom**
- From near-equilibrium to photoinduced SC suppression (NbN, YBCO, LSCO)

Amplification of SC by microwave or ultrasound absorption

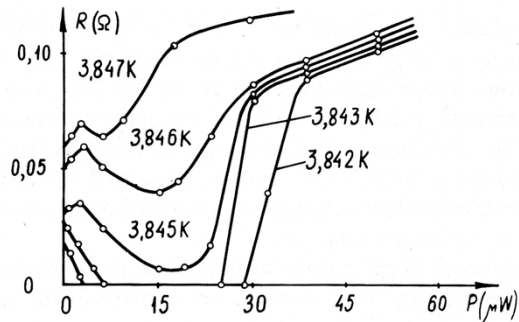


Fig. 2. Experimental dependence of Sn bridge resistance as a function of microwave power at different temperatures. Incident power frequency is ~ 10 GHz. $T_c = 3.84$ K.

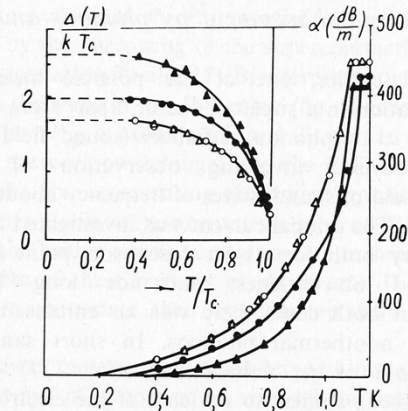


Fig. 14. Experimental temperature dependence of the attenuation of the longitudinal sound in superconducting gallium. \circ , Δ in the absence of a sound pumping field; \bullet pumping level μ_1 ; \blacktriangle pumping level $\mu_2 > \mu_1$. Signal and pumping frequencies are 54.2 MHz and 150 MHz respectively. The inset shows the temperature dependence of the nonequilibrium gap as calculated from attenuation curves by the BCS formula for the sound attenuation coefficient.

- 1970 Eliashberg:

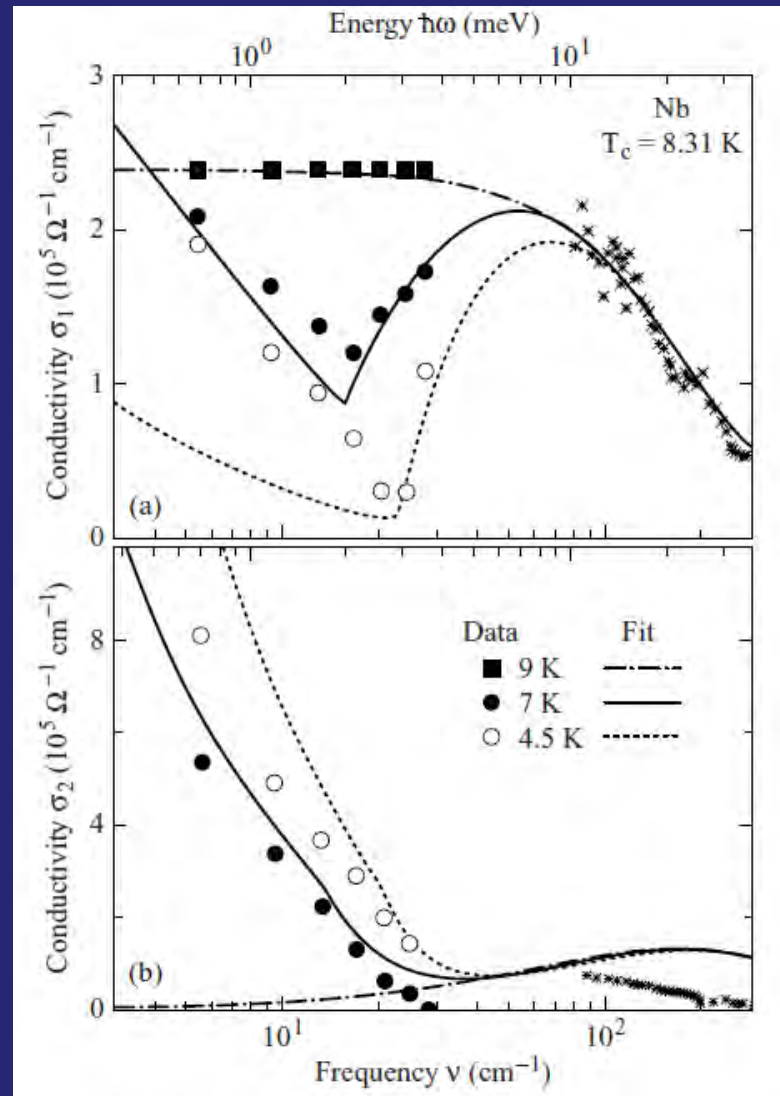
Kinetic equations for a superconductor in the external electromagnetic field, including the interaction between the field and QPs. Interaction changes the QP distribution function and can amplify pairing.

$$\frac{df_{\varepsilon}(r, k, t)}{dt} = \dots$$

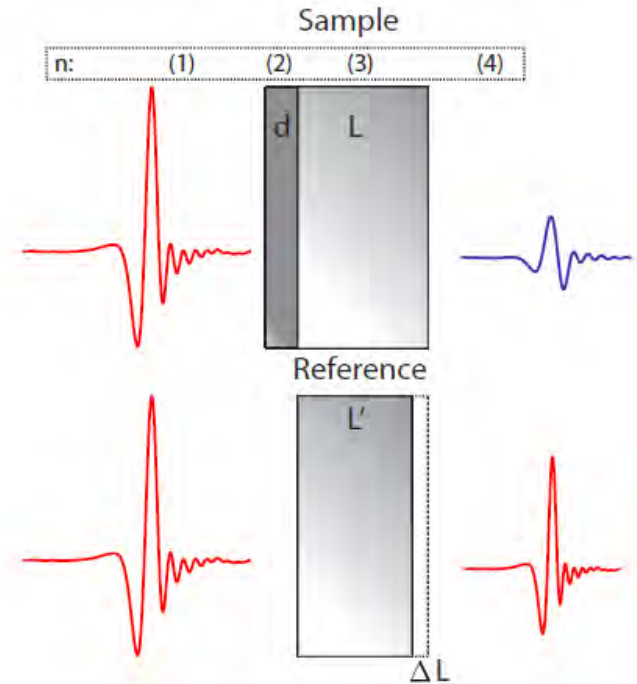
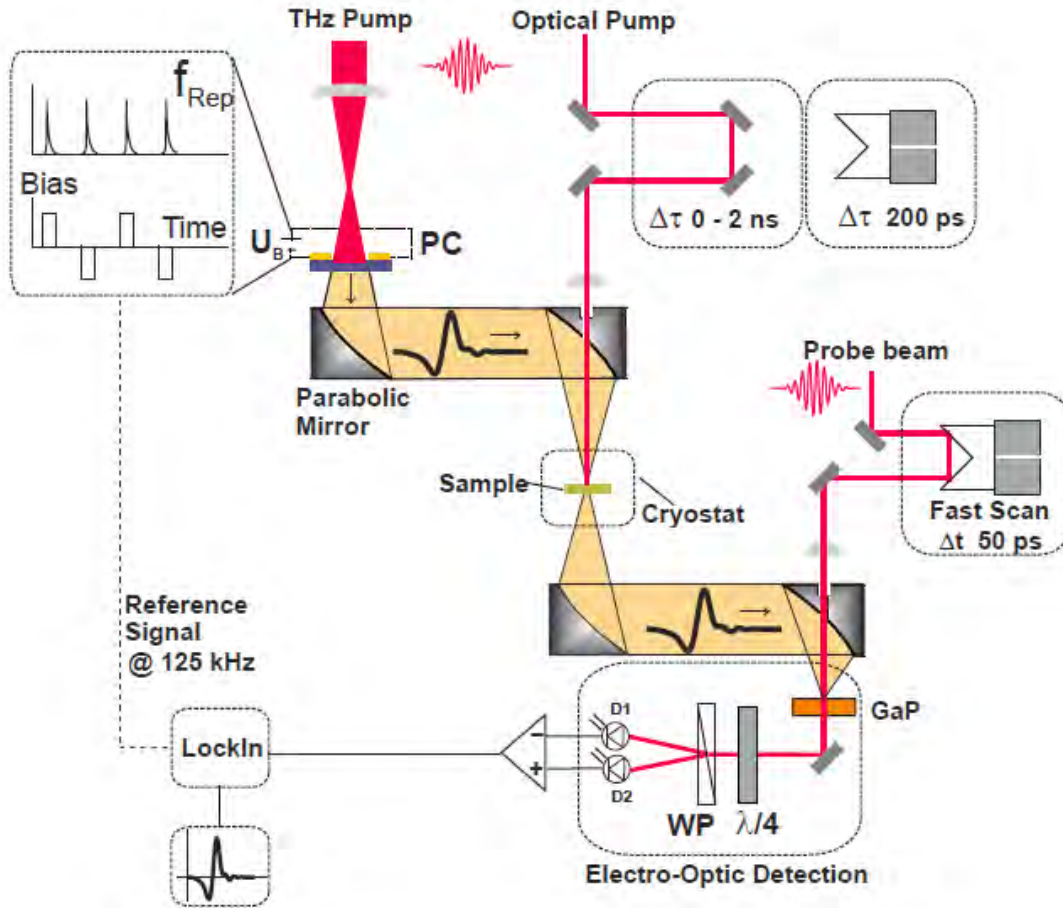
$$\frac{dn_{\varepsilon}(r, k, t)}{dt} = \dots$$

$$\Delta = V \sum_{k'} \Delta_{k'} \frac{(1 - 2f_{k'})}{\varepsilon_{k'}}$$

Time evolution of a BSC superconductor



Time-domain THz Spectroscopy



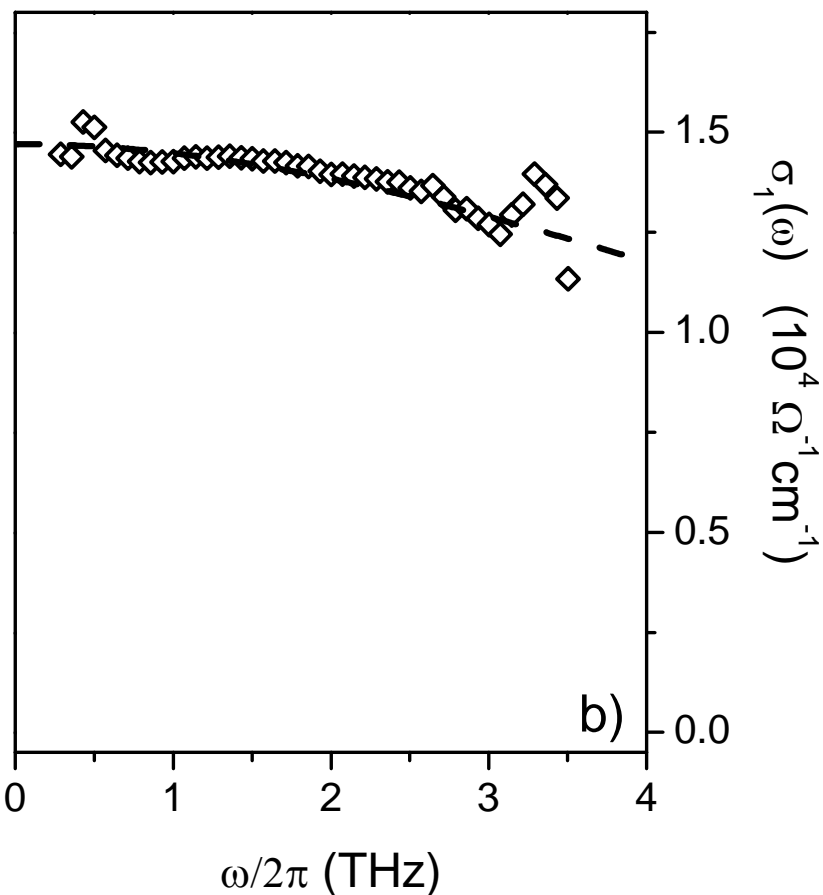
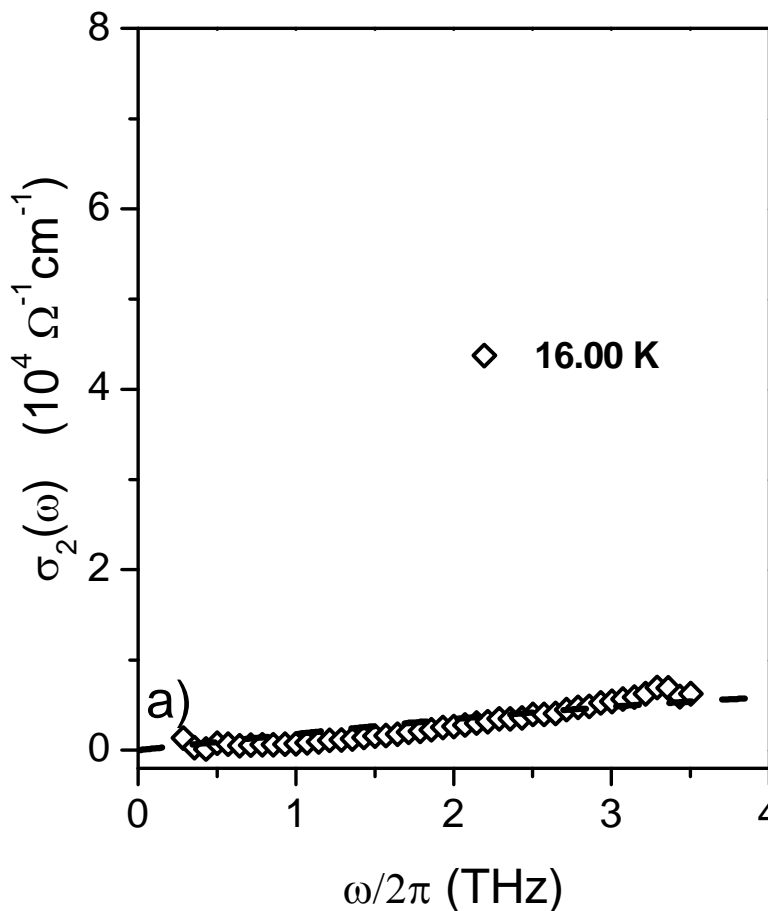
$$t(\omega) \equiv \frac{E_{OUT}(\omega)}{E_{IN}(\omega)} \approx \frac{2}{1 + n_S + d Z_0 \sigma(\omega)}$$



NbN ($T_c = 15.4$ K): Normal state optical conductivity

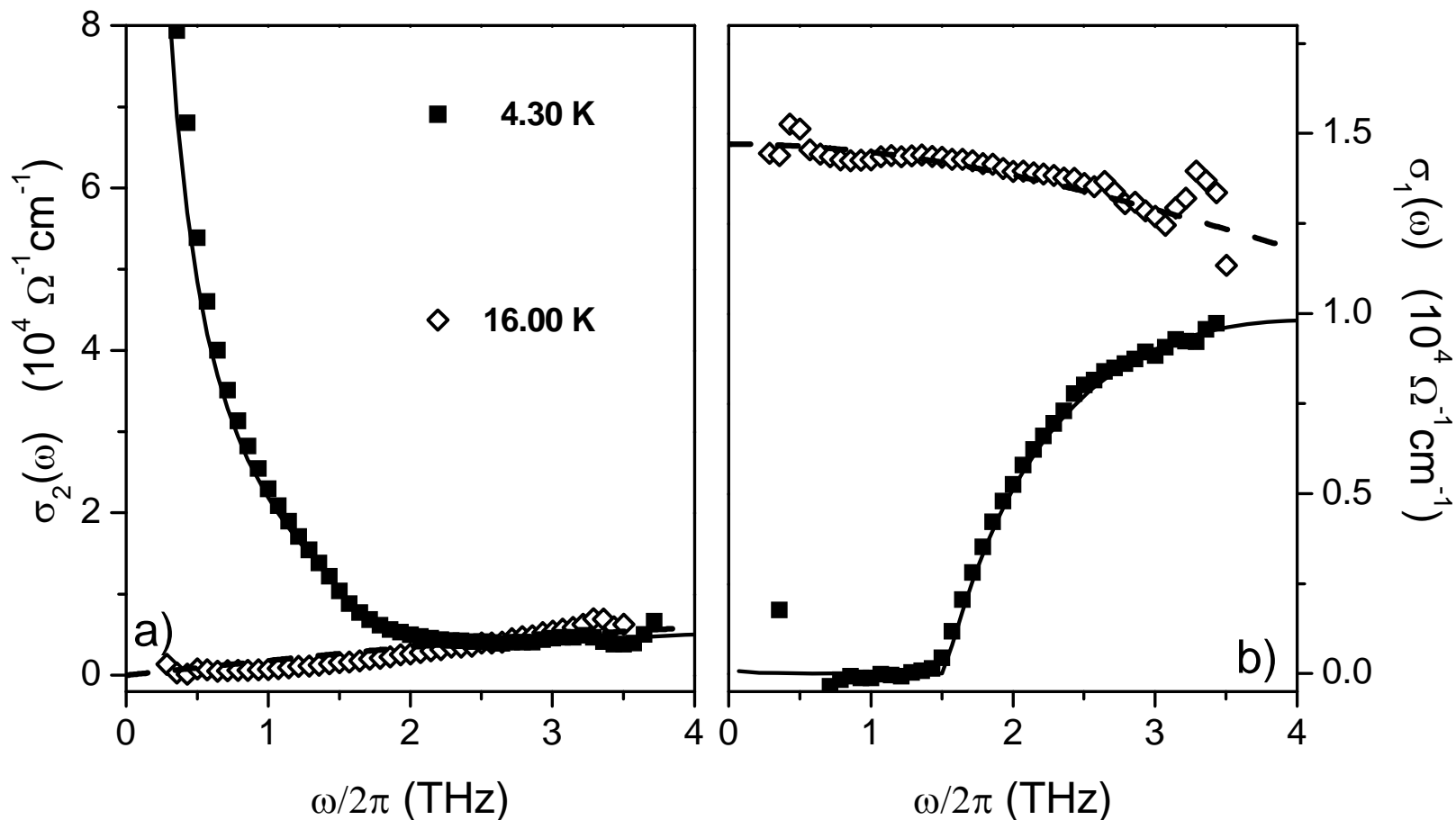
$$\nu_{pl} = 460 \text{ THz} \quad \& \quad \tau = 8 \text{ THz}$$

$$\sigma(\omega) = \frac{\sigma_0}{1 + i\omega\tau}$$



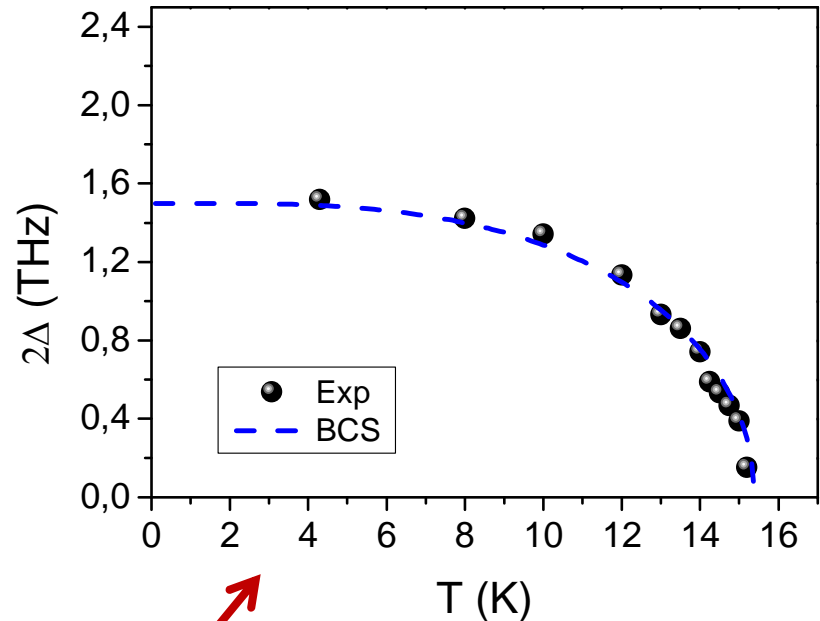
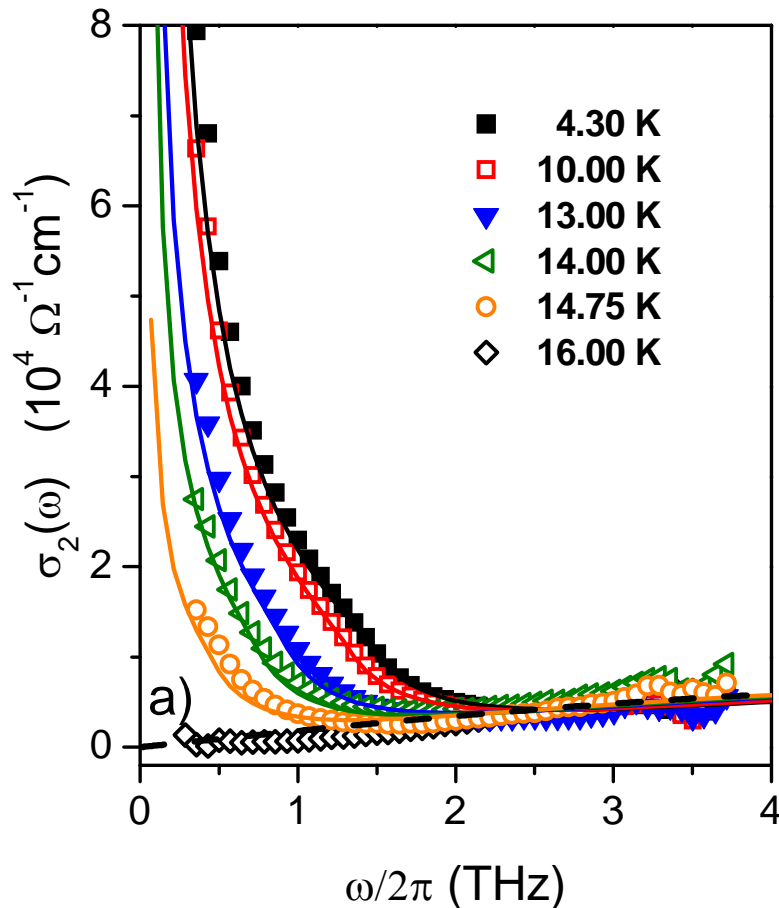
NbN ($T_c = 15.4$ K): SC signature in optical conductivity

Fits in the SC are BCS-calc of cond. in dirty limit
Zimmermann, *Physica* **183C**, 99 (1991).



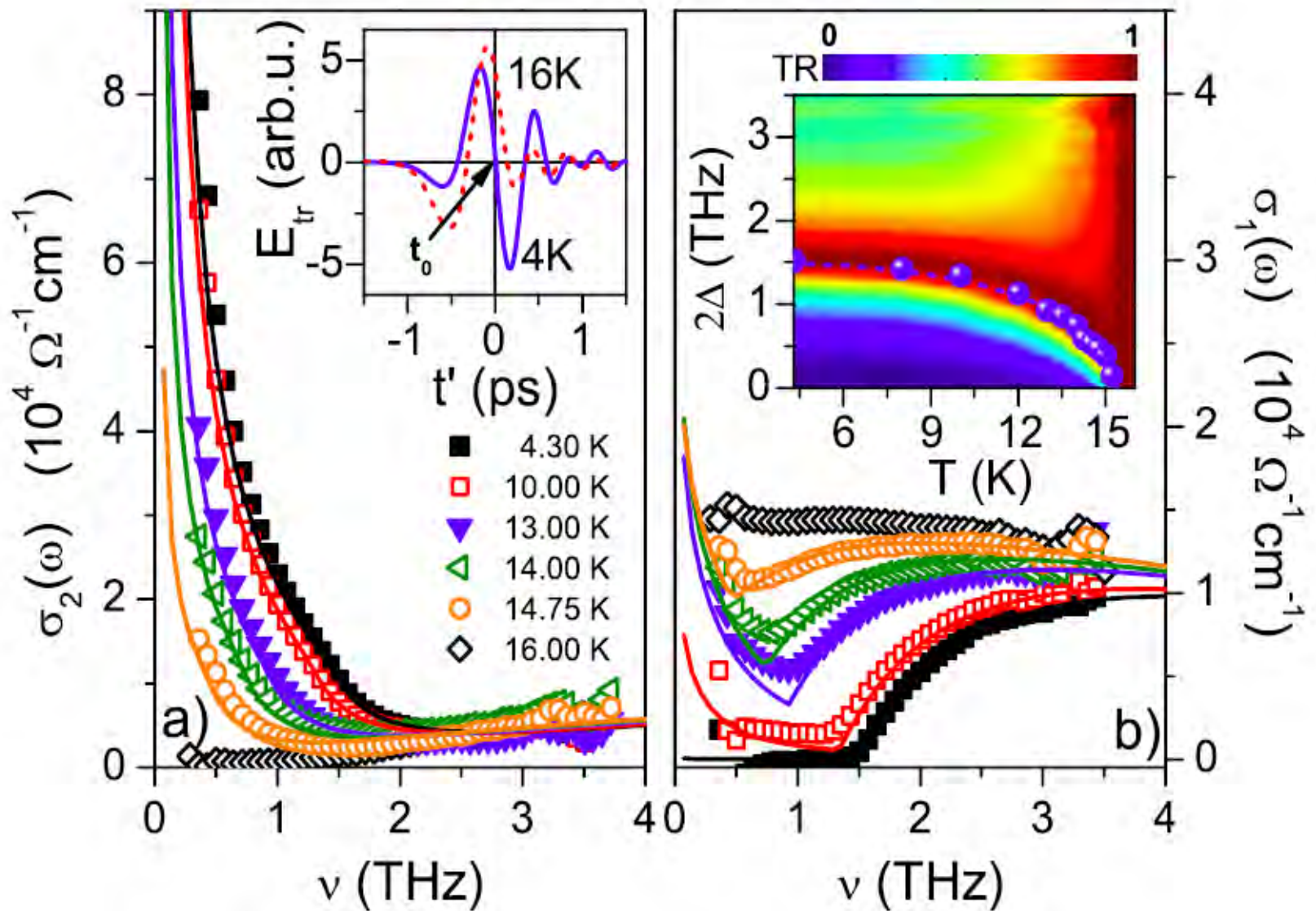
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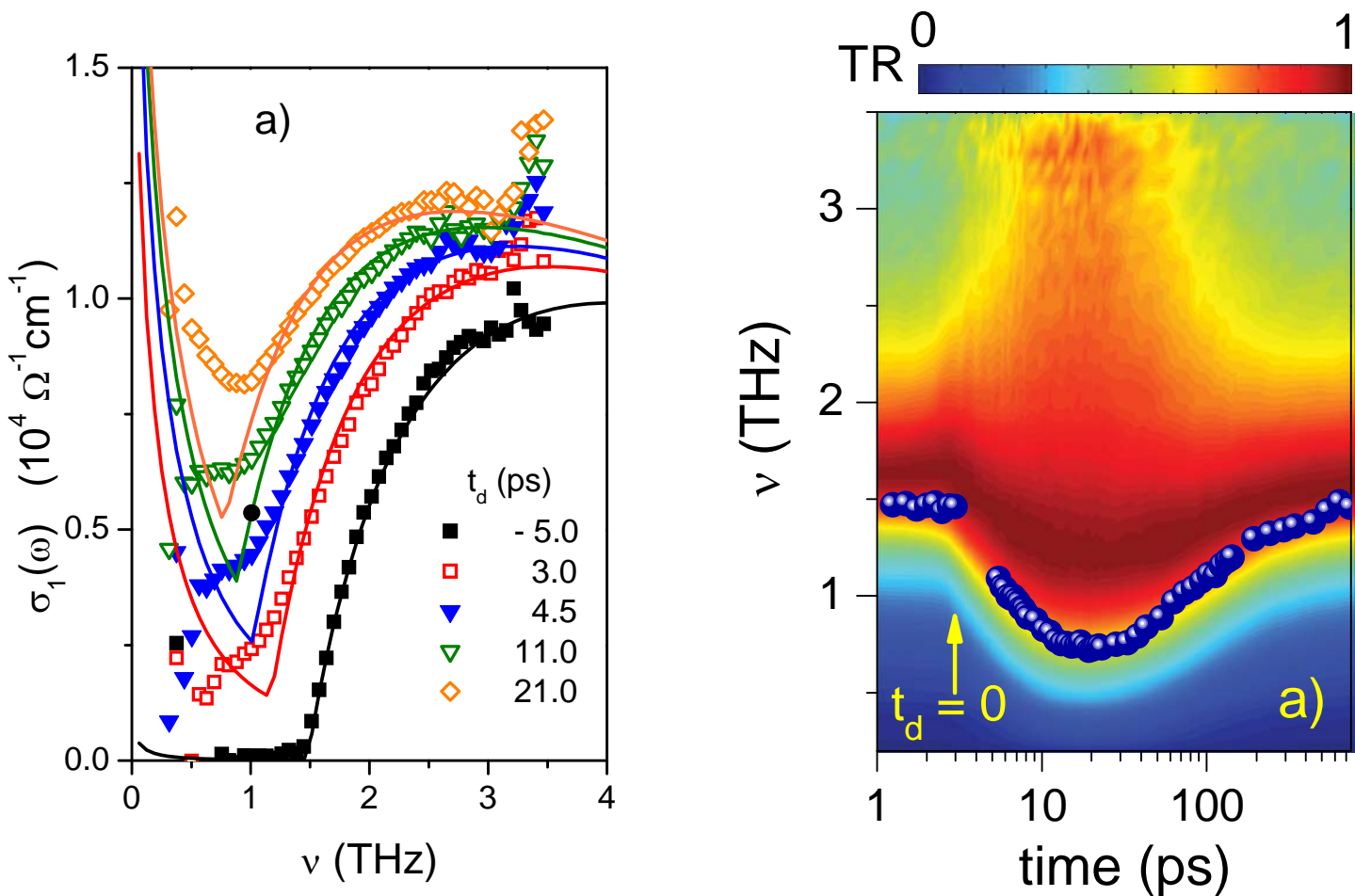


$2\Delta(T)$ follows BCS; $2\Delta(0) \sim 4.6 k_B T$: Strong coupled SC - dirty limit

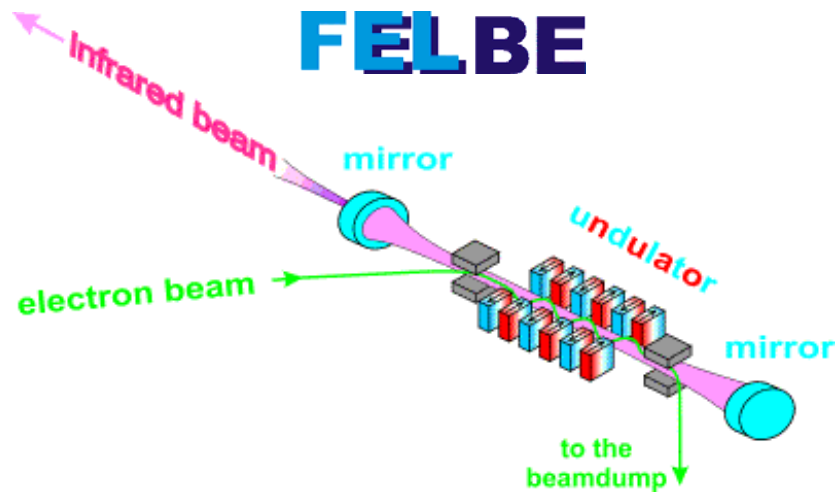
NbN ($T_c = 15.4$ K) in equilibrium



Optical pump – THz probe studies: direct access to $\Delta(t_d)$

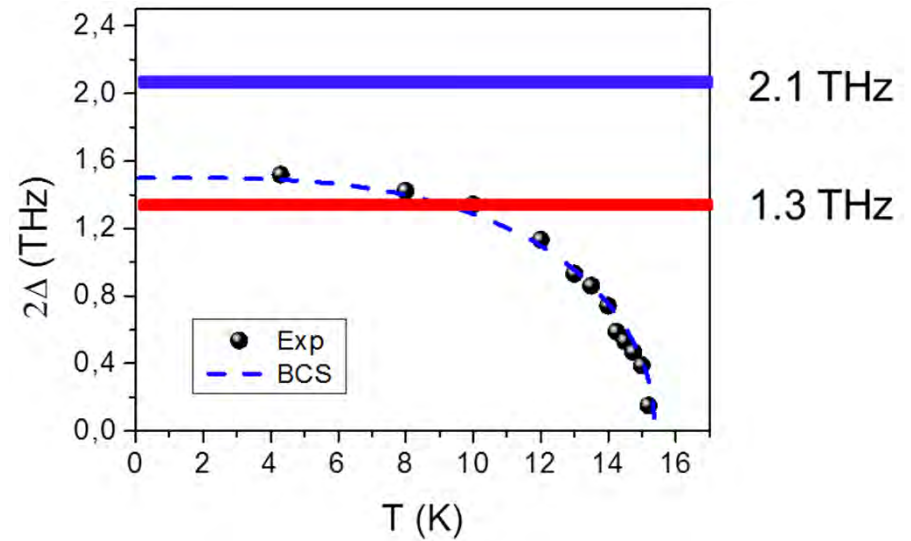


Resonant THz excitation of a BCS Superconductor

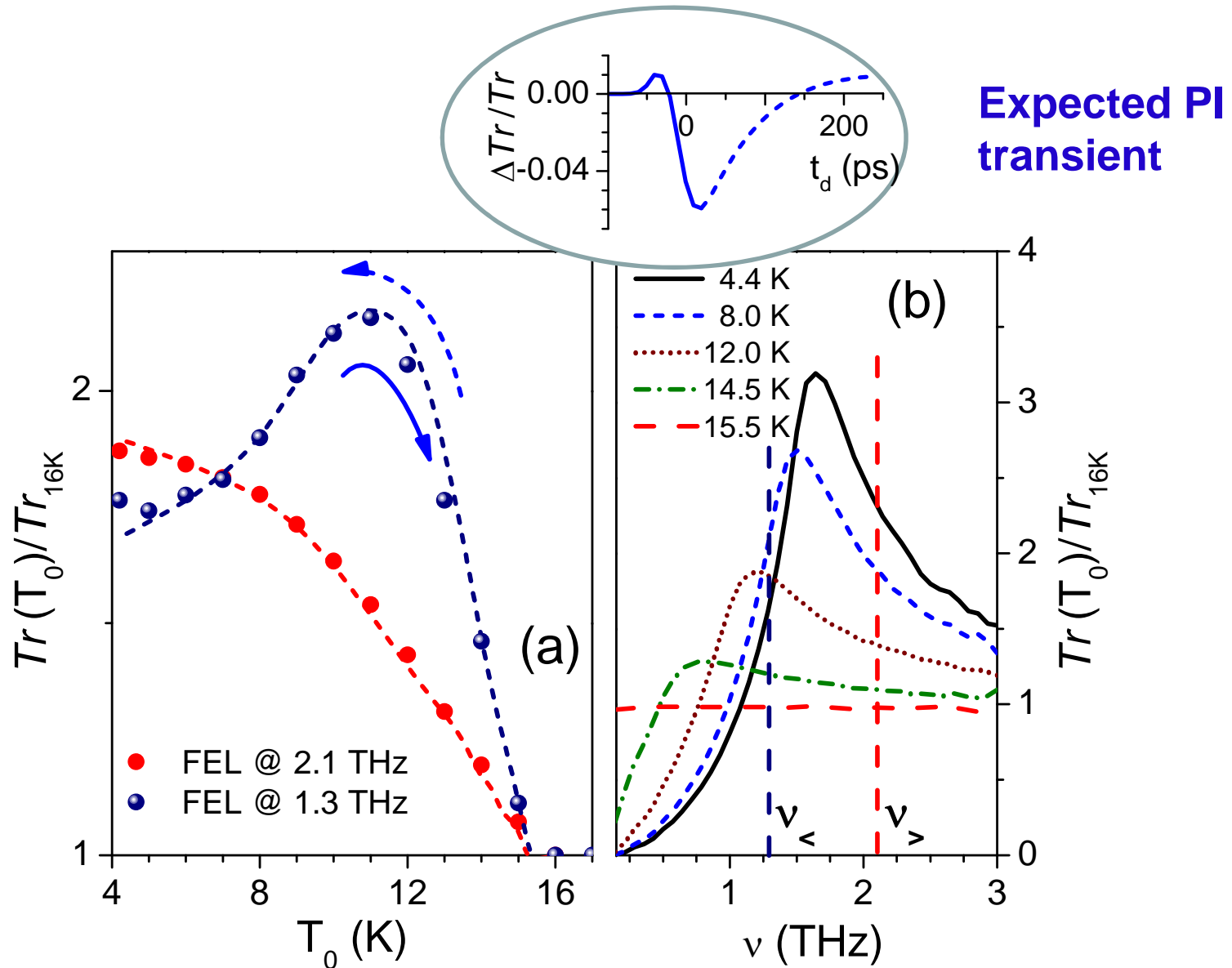


Radiation

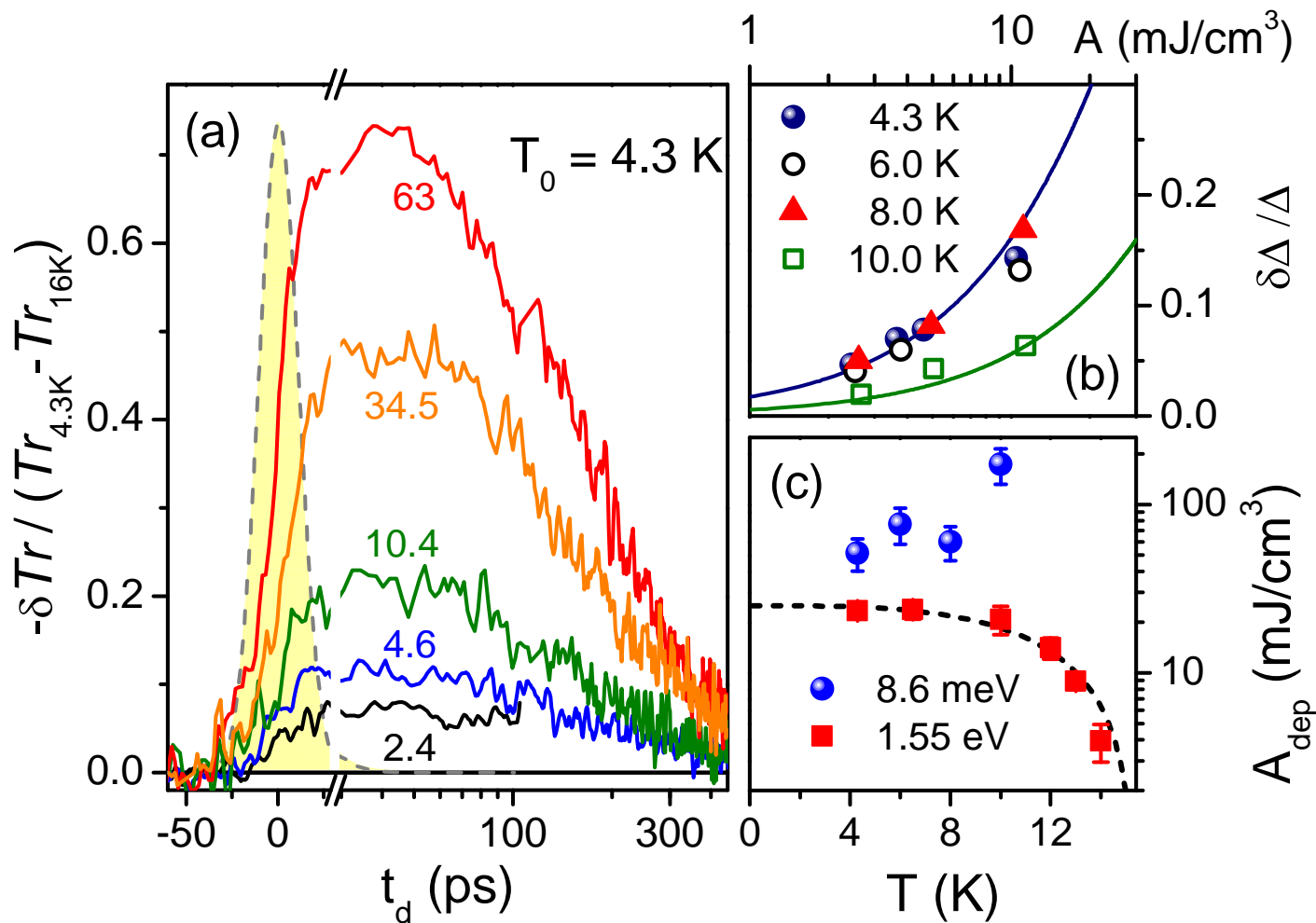
Undulator	U27	U100	
Wavelength [μm]	3.5 - 22	18 - 250	details
Average output power [W]	0.1 - 40	0.1 - 40	details
Pulse energy [μJ]	0.01 - 3	0.01 - 3	



T-dependence of THz transmission of ps pulses

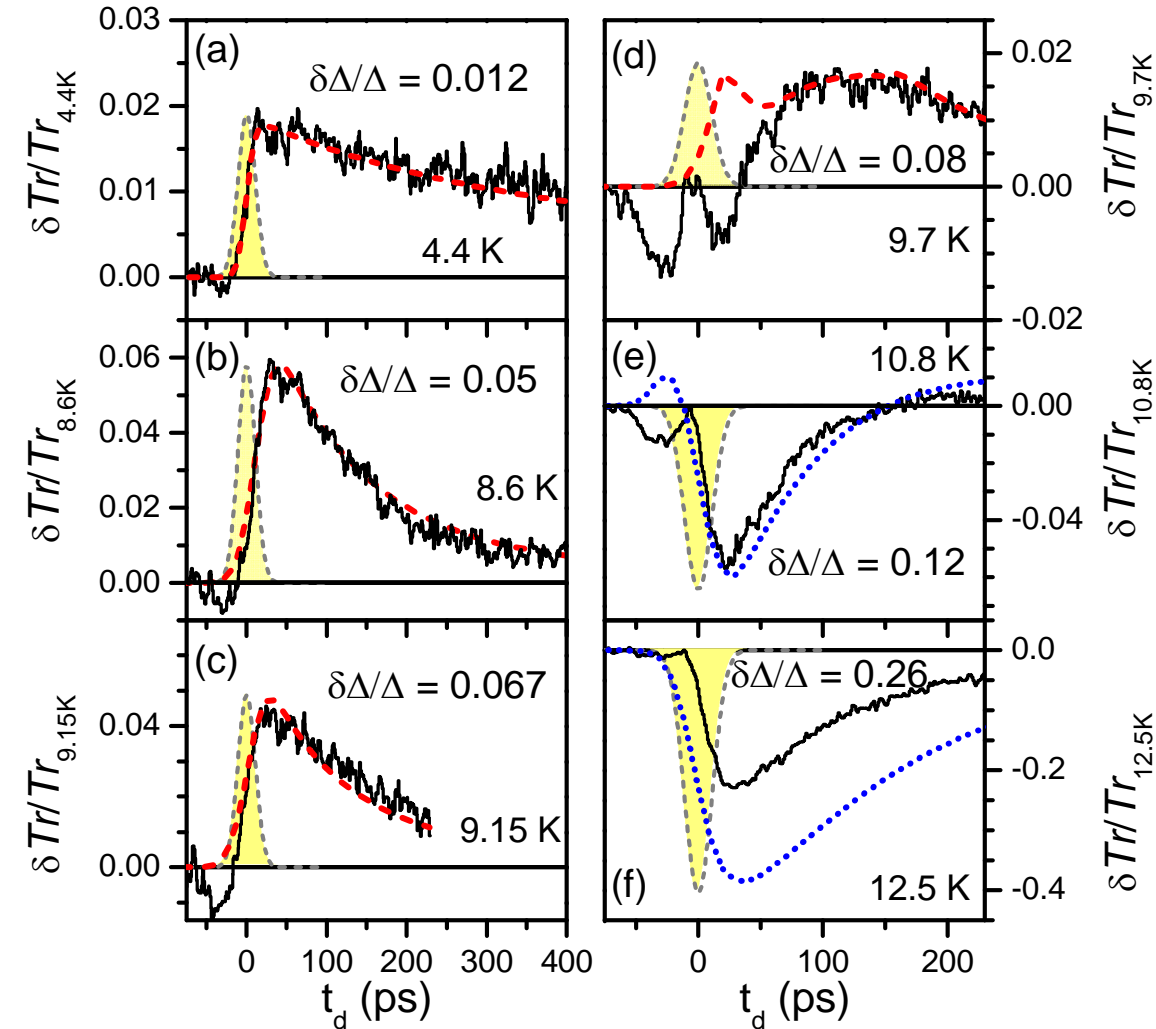


Melting of Superconductivity using above gap pumping



- The energy required to quench superconductivity increases with temperature!
- Competing SC enhancement effects by narrowband THz pumping!

SC state Dynamics following sub-gap pumping



i) Sub gap pumping, $T \ll T_c$
 - direct pair breaking not possible
 - (weak) suppression of SC via “dynamic pair-breaking”;
 maximum super-current close to de-pairing current.

ii) Above gap pumping
 - Similar to the case of 2.1 THz pumping (for higher temperatures induced changes smaller than expected)

iii) Resonant pumping
 - for early times the transmission changes opposite to the expected for the case of induced gap-suppression!

- $T \sim 10$ K, $\nu \sim 2\Delta/h$: gap enhancement at early time-delays !

Photo-enhancement of Superconductivity

Eliashberg (1970):

SC gap enhancement due to the induced changes in the electronic distribution function

$$\Delta = g \int_{\Delta}^{\hbar\omega_c} d\epsilon \frac{\Delta}{\sqrt{\epsilon^2 - \Delta^2}} [1 - 2f(\epsilon)]$$

Self-consistent (BCS) gap equation

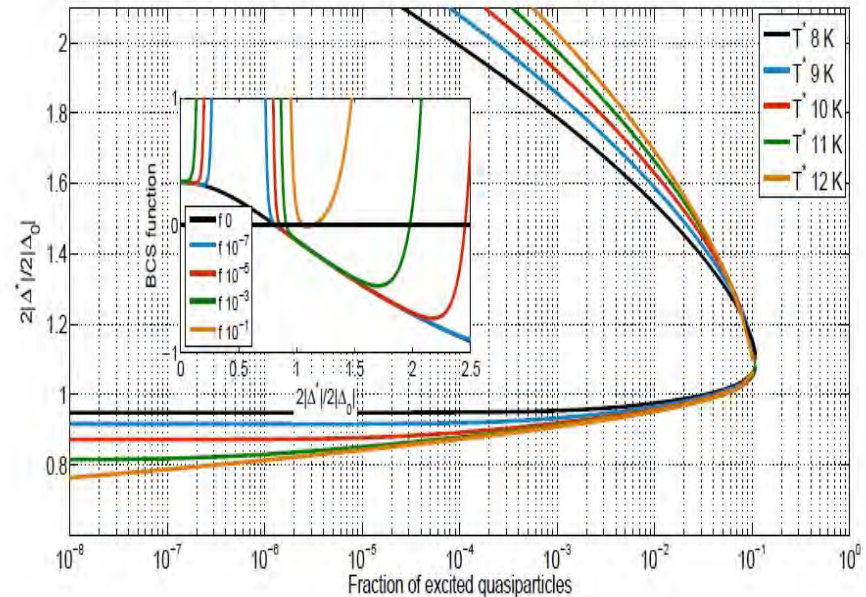
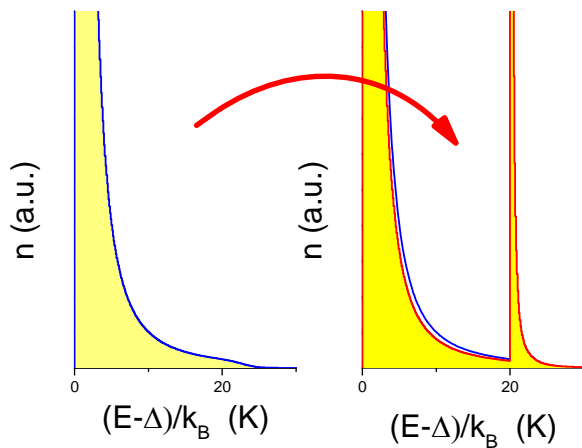
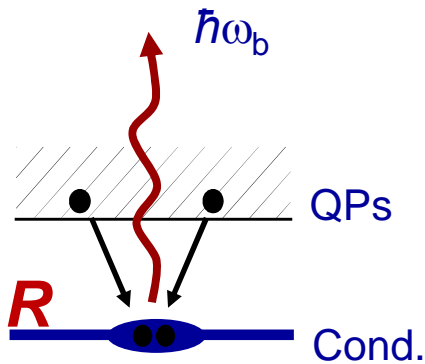


Photo-enhancement of Superconductivity

Scalapino (1977):

- Numerical studies of linearized coupled kinetic equations
- **Pair-recombination rate (R) depends on QP energy** ($\propto \omega^2$)
- Enhanced recombination gives rise to QP cooling (pair-density/gap further increases)



QP recombination via
 $\hbar\omega_b > 2\Delta$ boson emission

$$\frac{dn}{dt} = \beta N - Rn^2$$

$$\frac{dN}{dt} = +\frac{1}{2} [Rn^2 - \beta N] - \frac{(N - N_T)}{\tau_\gamma}$$

Characteristic timescale $\tau_c \sim Rn_T$
R (averaged) $\sim 160 \text{ ps}^{-1}$ unit cell
 $n_T(10 \text{ K}) \sim 2 \times 10^{-4}$ unit cell $^{-1}$

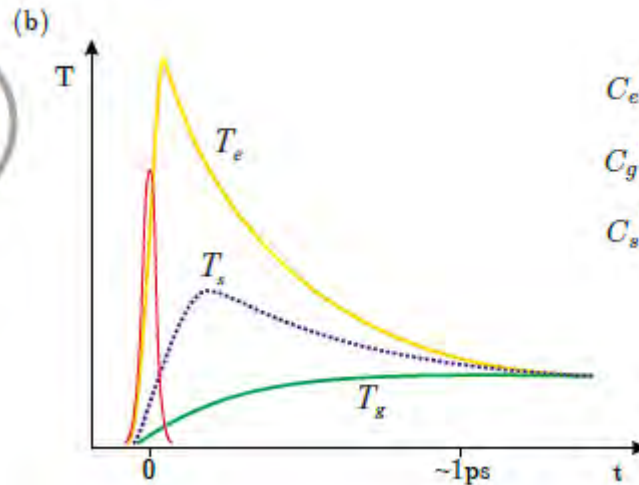
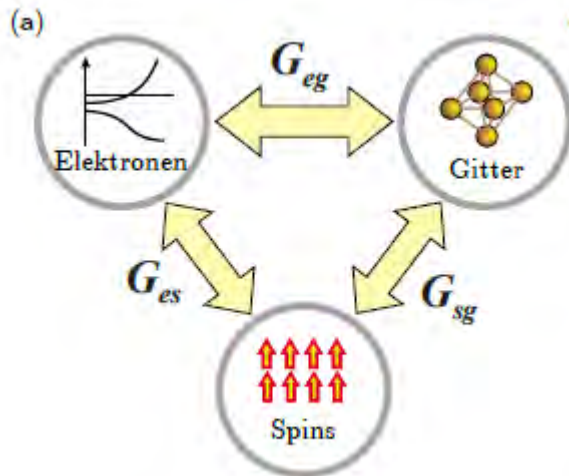
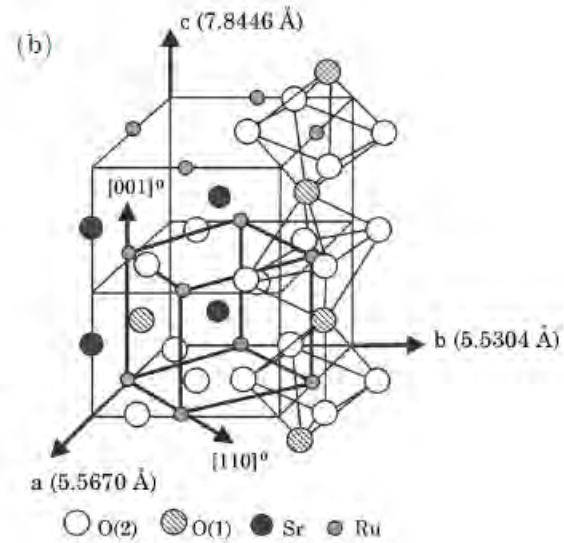
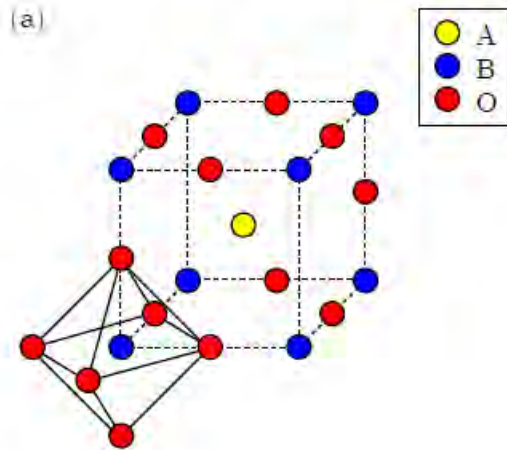
$$\tau_c \sim 20 \text{ ps}$$

Summary (I)

Time-resolved THz conductivity dynamics in Superconductors:

- **Determination of microscopic parameters**, e.g. pair-breaking rate by phonon absorption, e-ph coupling constant (PRL 91, 267002 (2003), PRL 95, 147002 (2005), PRL 107, 177007 (2011))
- Resonant excitation with narrow band pulses:
for temperatures close to T_c **enhancement of SC** (gap/condensate density increase) can be achieved due to PI changes in the distribution function (timescale governed by thermalization time) ?
- High- T_c superconducting cuprates: multi-component response, anomalous overdoped range, optical energy required to suppress SC an order of magnitude larger than condensation energy (PRL 101, 227001 (2008), PRL 105, 067001 (2010), PRB 83, 214515 (2011))
- THz pumping of PCCO: energy required to suppress SC equals condensation energy (with optical pumping the energy 6x larger)

THz conductivity dynamics in SrRuO₃ and NiPd



$$C_e \frac{dT_e}{dt} = -G_{eg}(T_e - T_g) - G_{es}(T_e - T_s) + P(t)$$

$$C_g \frac{dT_g}{dt} = -G_{ge}(T_g - T_e) - G_{gs}(T_g - T_s)$$

$$C_s \frac{dT_s}{dt} = -G_{sg}(T_s - T_g) - G_{se}(T_s - T_e)$$

SrRuO₃ - Transport

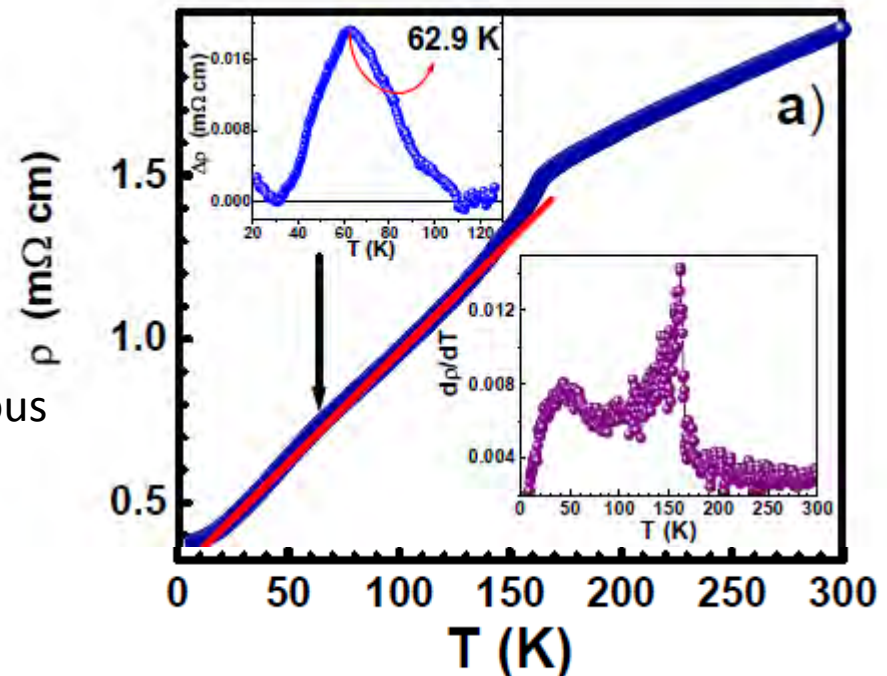
- Above T_c : $\rho \sim T$ up to 1000 K (! Mott-Ioffe-Regel Limit)
- $T < 40$ K: $\rho \sim T^2$ Fermi-liquid behaviour (deHaas v.A. Effekt)
- $60\text{K} < T < 130\text{K}$: $\rho \sim T^{3/2}$ (similar to ferromagnetic alloys)

-2 Anomalies

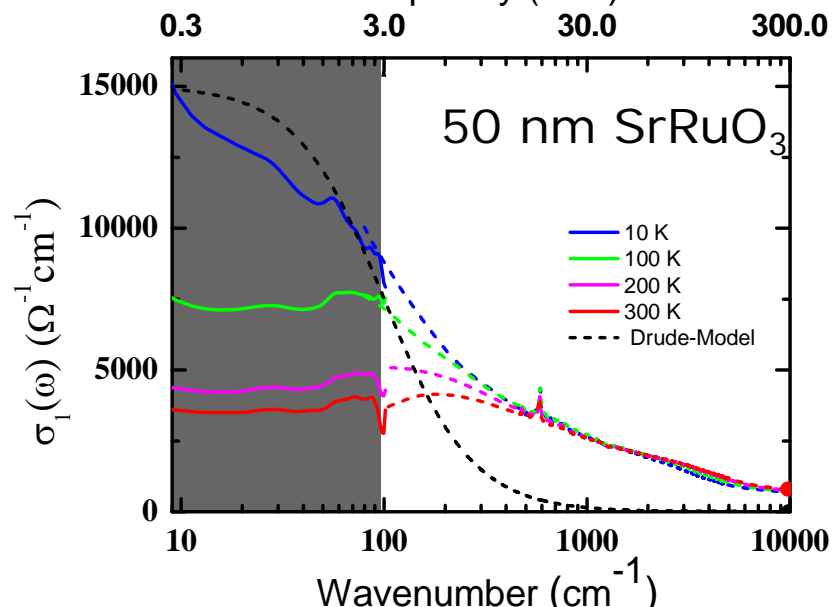
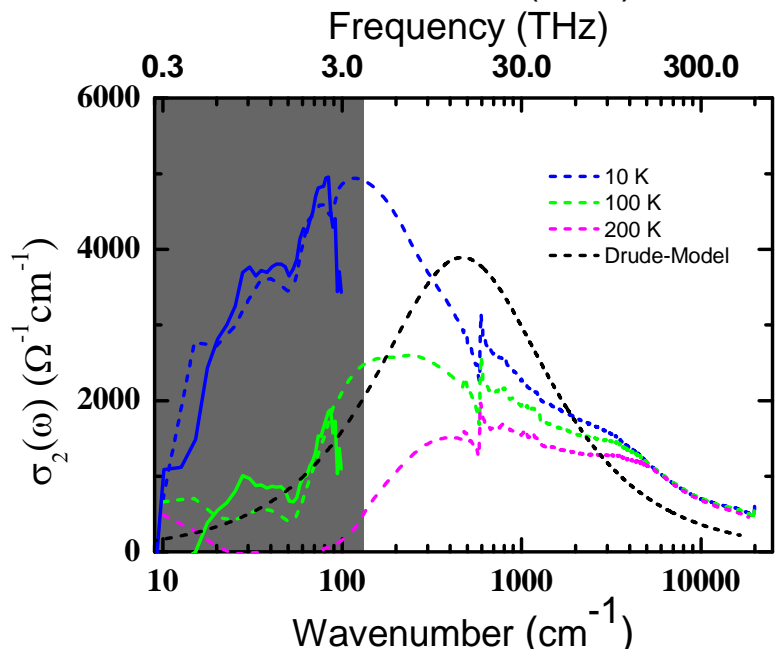
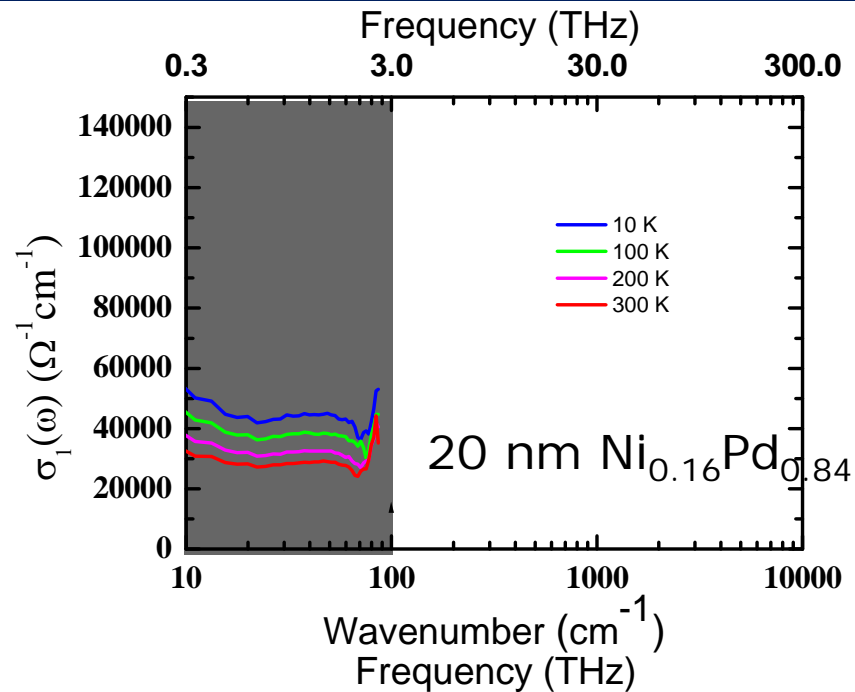
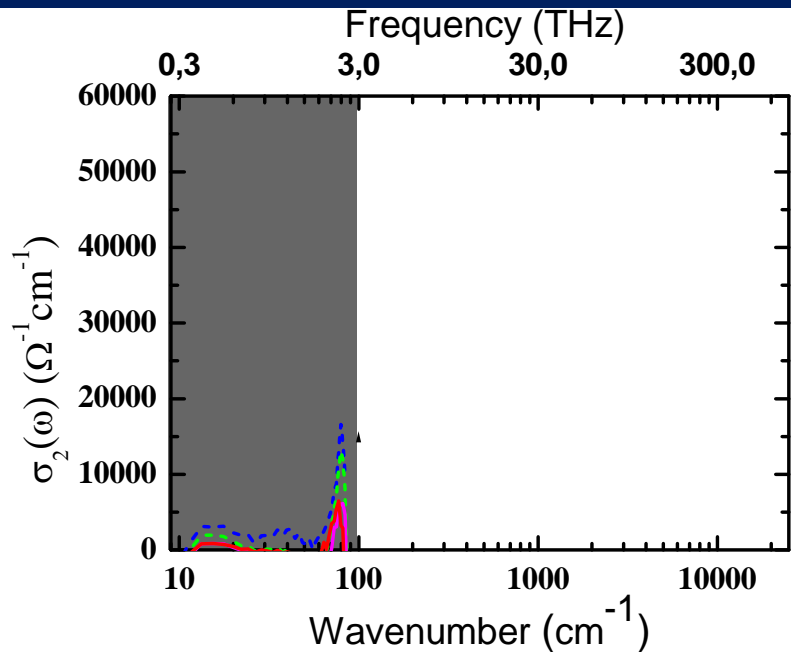
- $T = T_c$: Abrupt decrease (short range spin fluctuations -> Fisher-Langer Theory)
- $T = 60$ K: Peak in ρ -> Hidden Order (Spin Glass??)

-Critical behaviour different for C_v and M !
(different Ising/Heisenberg exponents)

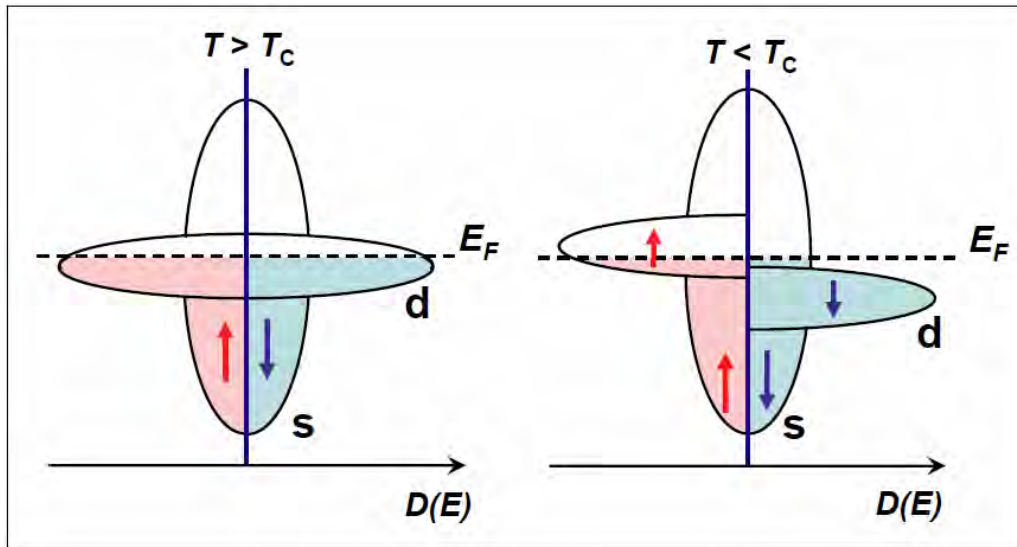
→ Short-range spin fluctuations anomalous
in SRO



Optical Conductivity of both materials

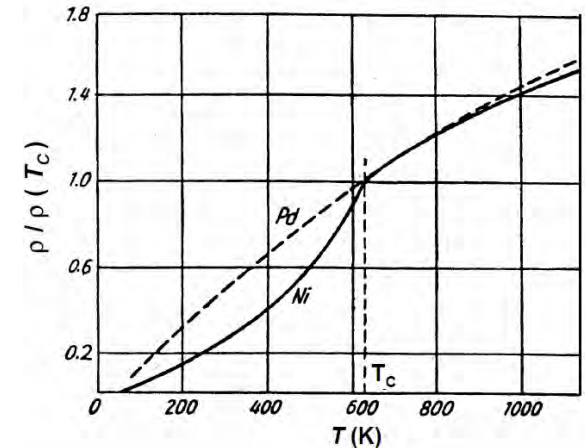


Magnetic Signatures in Transport

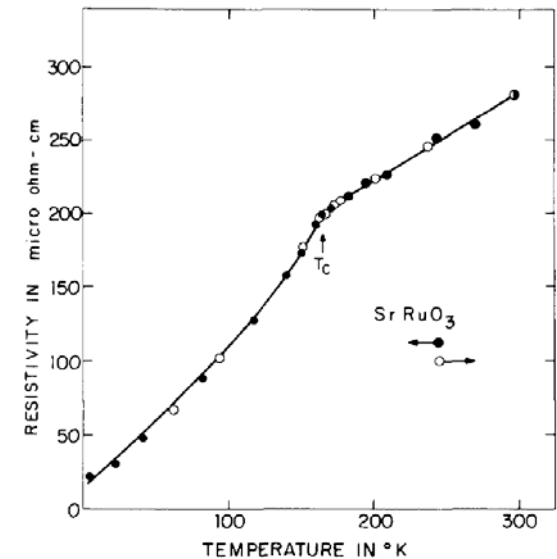


„Conductivity (Resistivity) as an indirect measure of the magnetic properties“

DC-resistivity measurements:



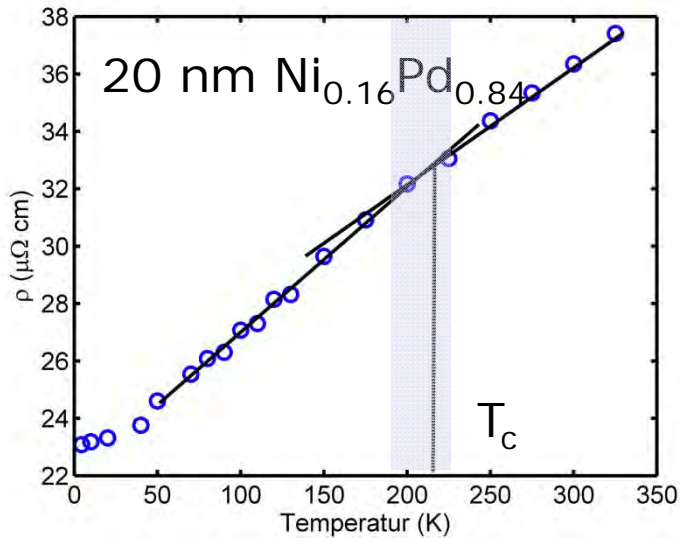
Coles *et al.*, *Adv. Phys.*, **7**: 40 (1958)



Bouchard *et al.*, *Mat. Res. Bull.* **7**, 873 (1972)

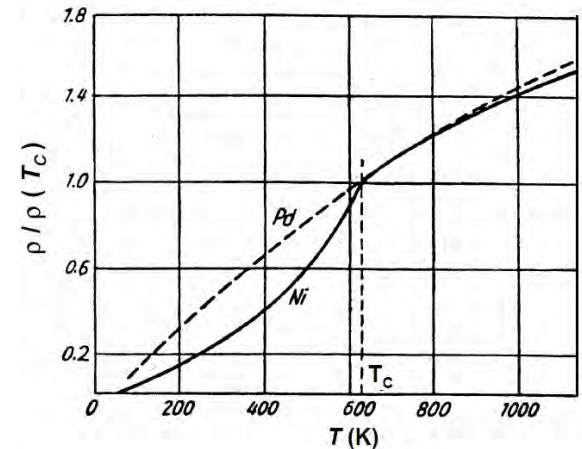
Temperature dependence of THz-Conductivity

AC-resistivity measurements:

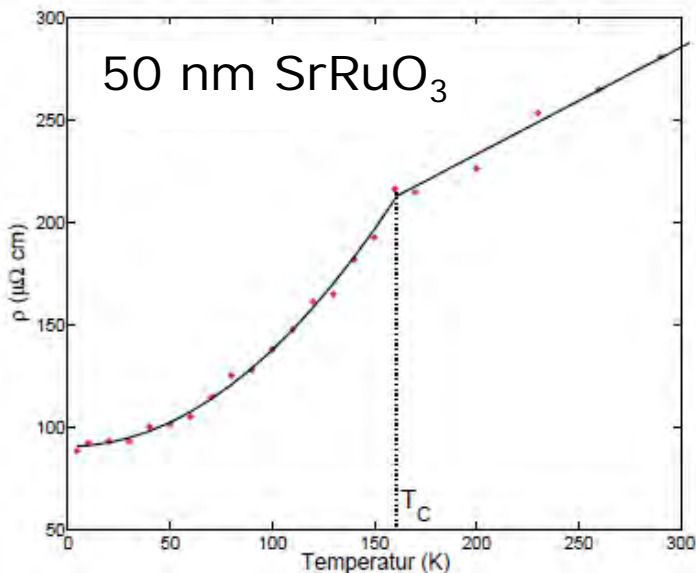


$$T_C = \sim 210 \text{ K}$$

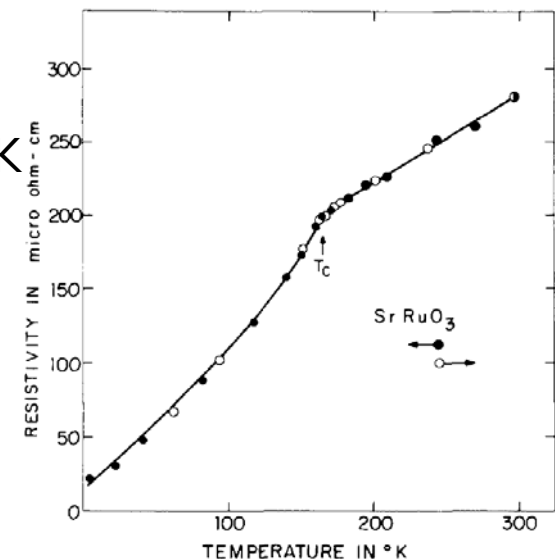
DC-resistivity measurements:



Coles *et al.*, *Adv. Phys.*, **7**: 40 (1958)

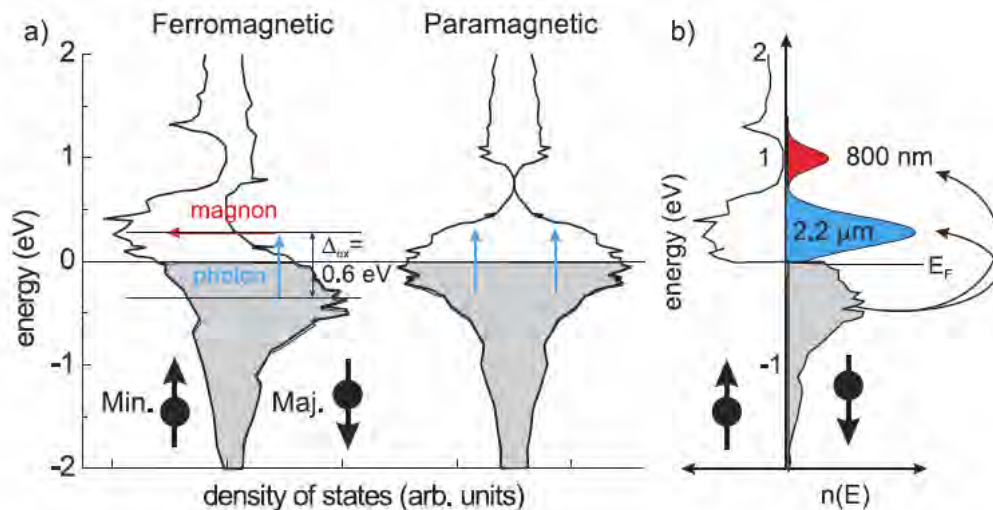


$$T_C = 150 - 160 \text{ K}$$

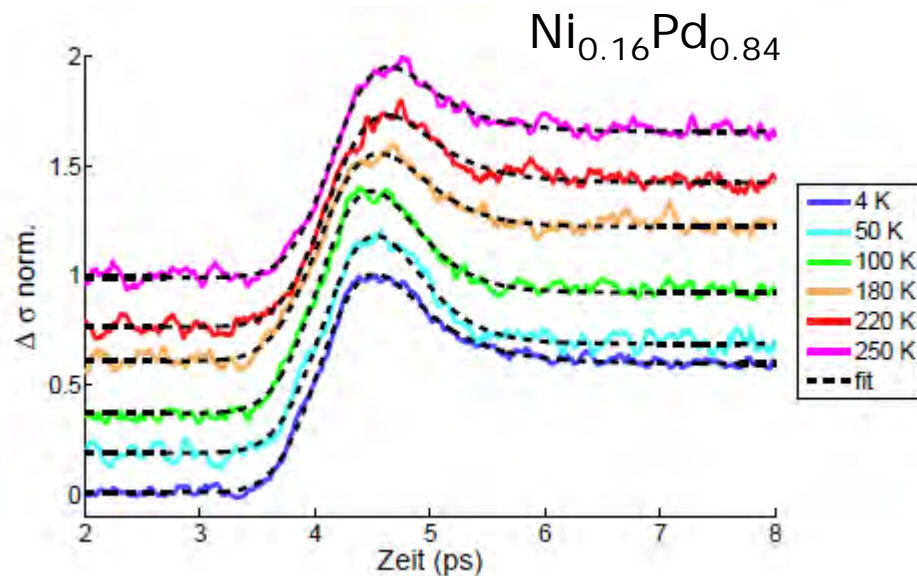
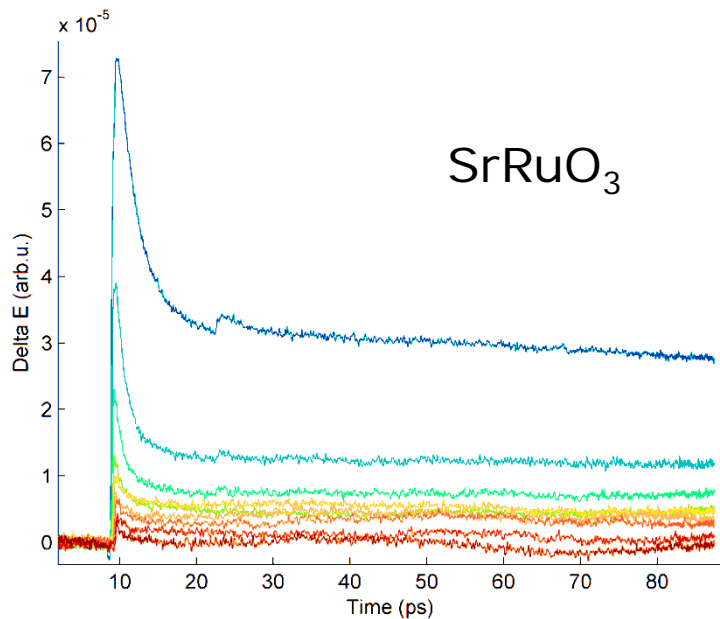


Bouchard *et al.*, *Mat. Res. Bull.* **7**, 873 (1972)

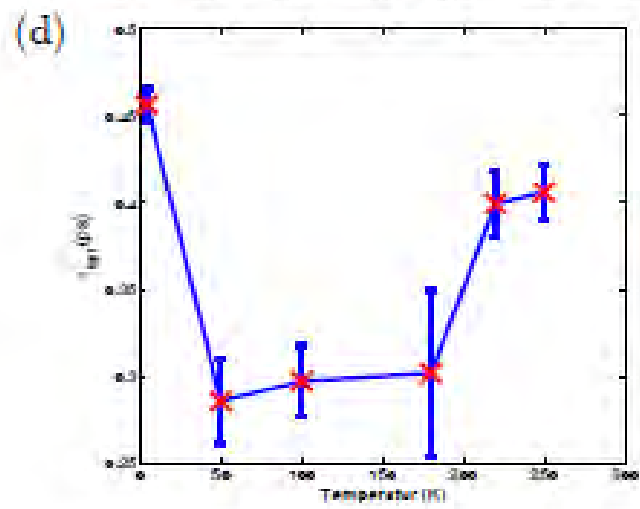
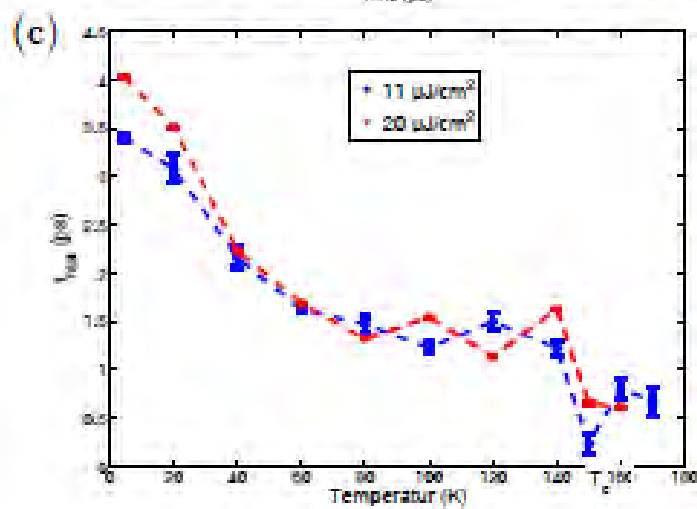
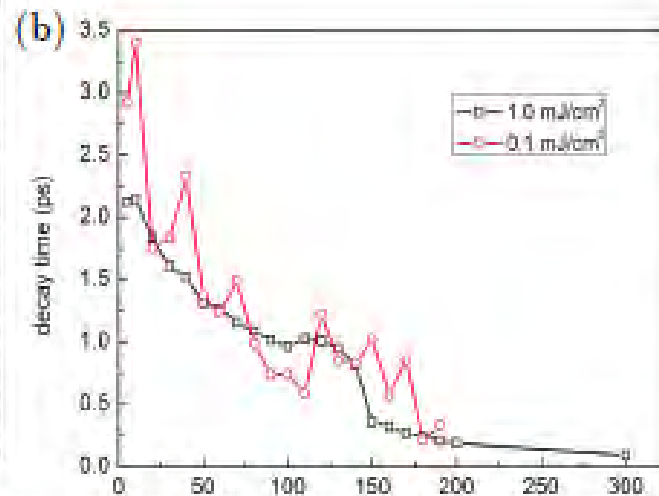
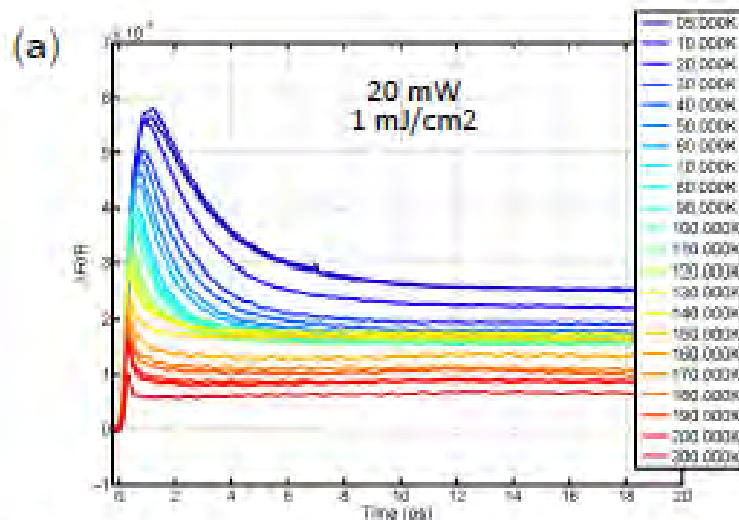
Conductivity Changes on Ultrafast Timescales



Woerner *et al.*, *Appl. Phys. A* **96**, 83 (2009)



SrRuO₃ – anomalous slowing down at T_c



Band Structure Calculations / DOS in the FM State

PHYSICAL REVIEW B

VOLUME 7, NUMBER 3

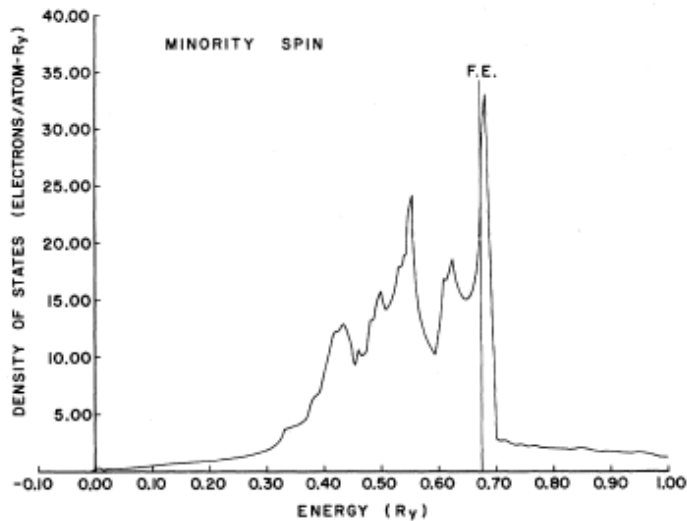
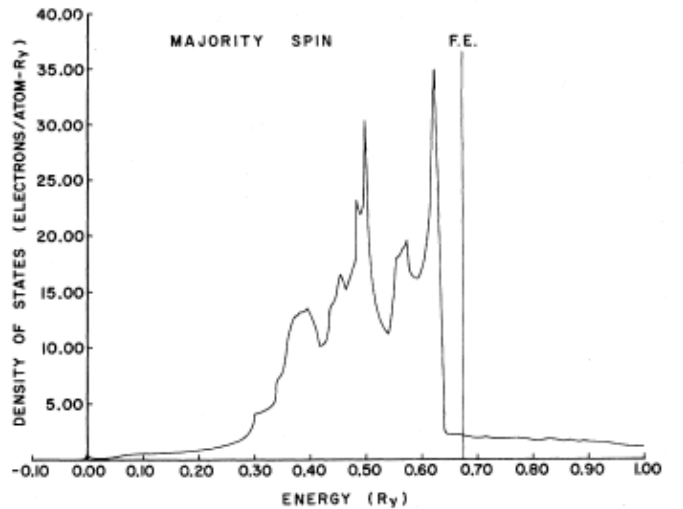
1 FEBRUARY 1973

Self-Consistent Calculation of Energy Bands in Ferromagnetic Nickel*

J. Callaway and C. S. Wang

Department of Physics, Louisiana State University, Baton Rouge, Louisiana 70803

(Received 19 June 1972)

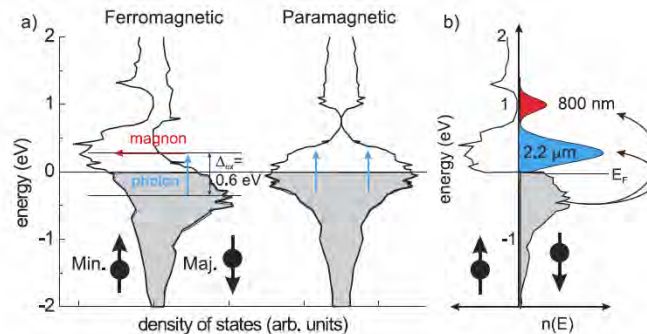
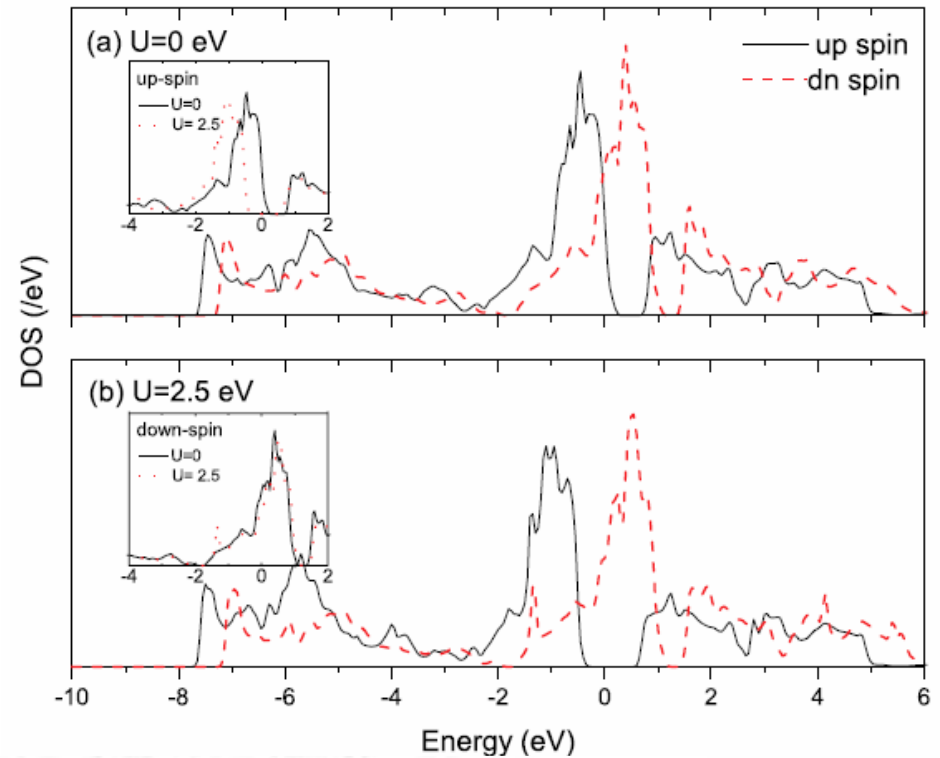


PHYSICAL REVIEW B 80, 035106 (2009)

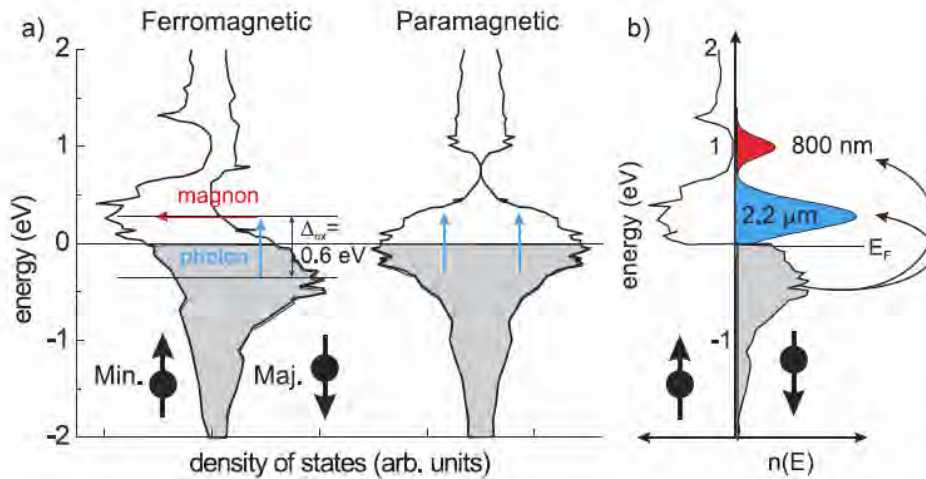
Evolution of the electronic structure of a ferromagnetic metal: Case of SrRuO₃

Priya Mahadevan,¹ E. Aryasetiawan,² A. Janotti,³ and T. Sasaki[†]

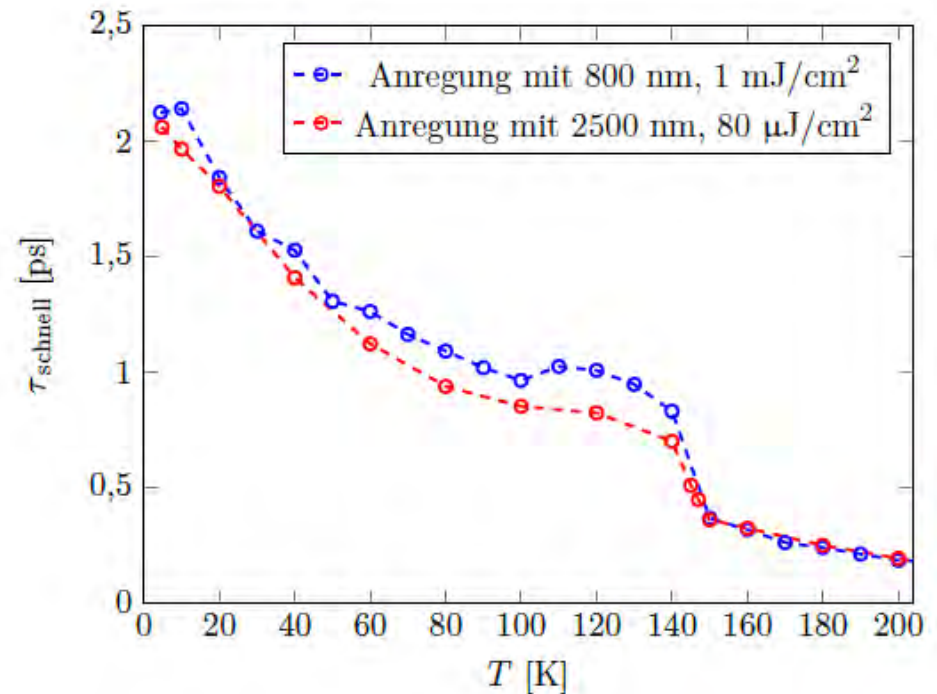
Ru *d* PDOS



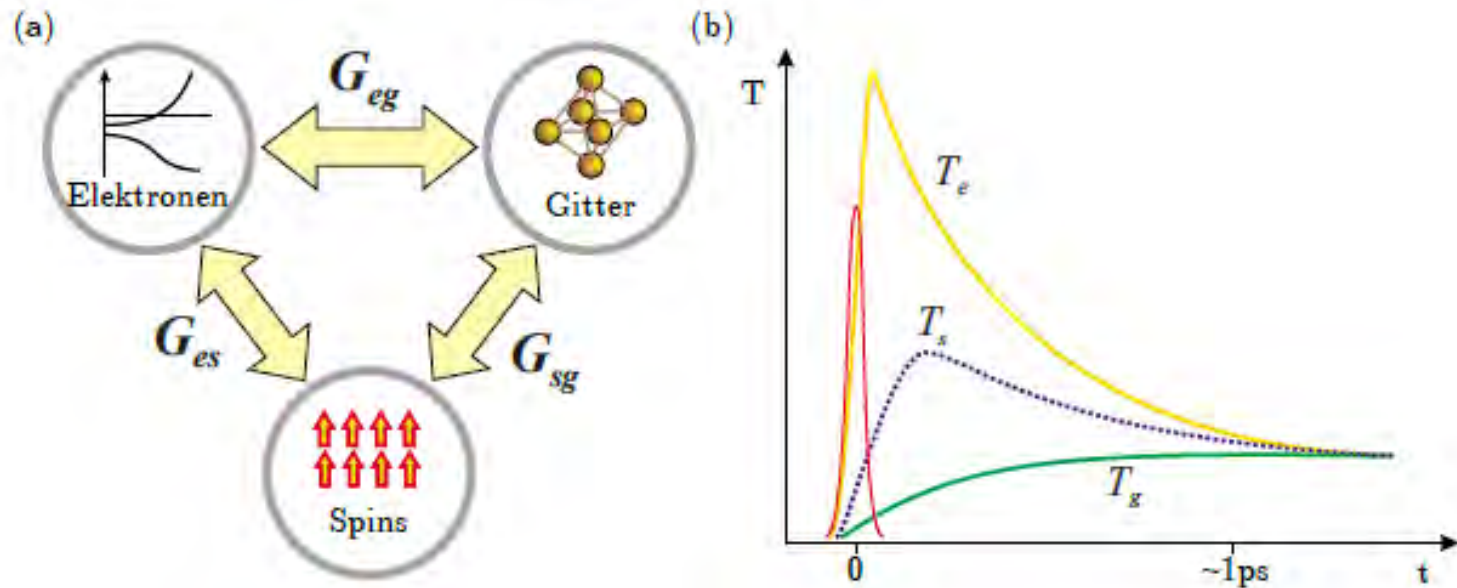
Excitation photon energy dependence



If the gap in the DOS for majority spin responsible for slowing down the gap is much smaller than the calculated 0.5 - 1 eV



What is governing (dynamic) changes in optical conductivity? Simple multi-temperature models

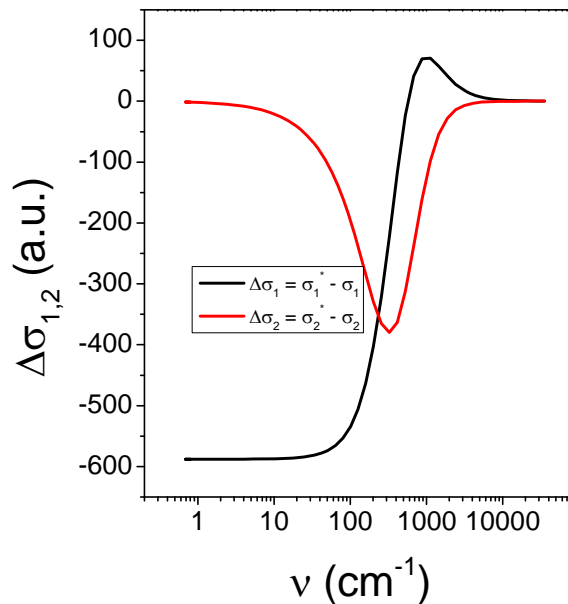
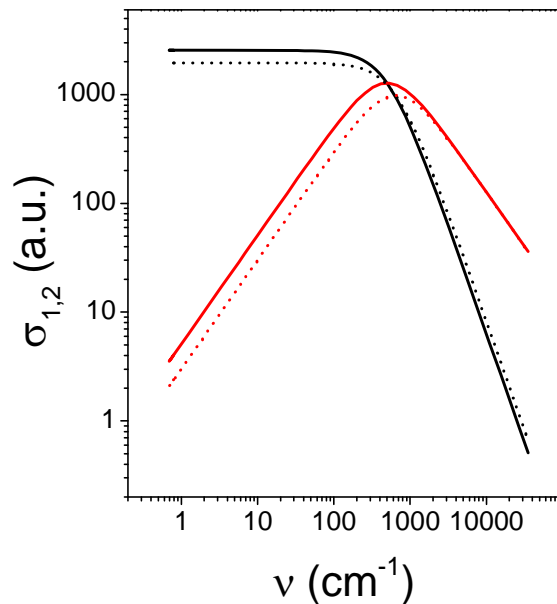


$$C_e \frac{dT_e}{dt} = -G_{eg}(T_e - T_g) - G_{es}(T_e - T_s) + P(t)$$

$$C_g \frac{dT_g}{dt} = -G_{ge}(T_g - T_e) - G_{gs}(T_g - T_s)$$

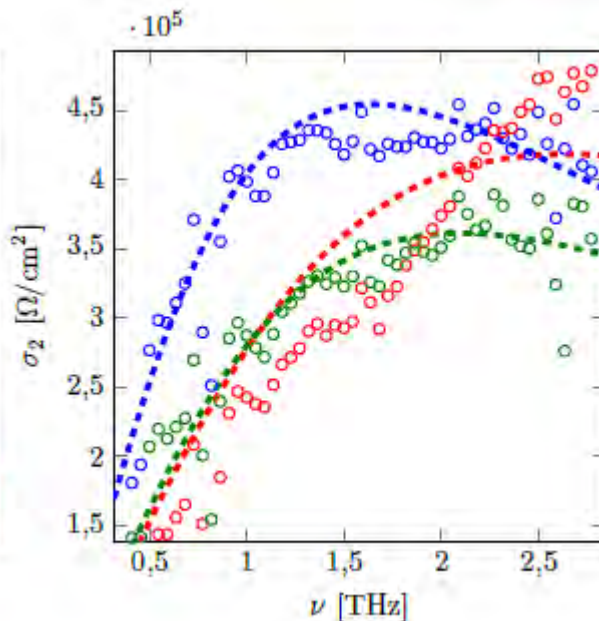
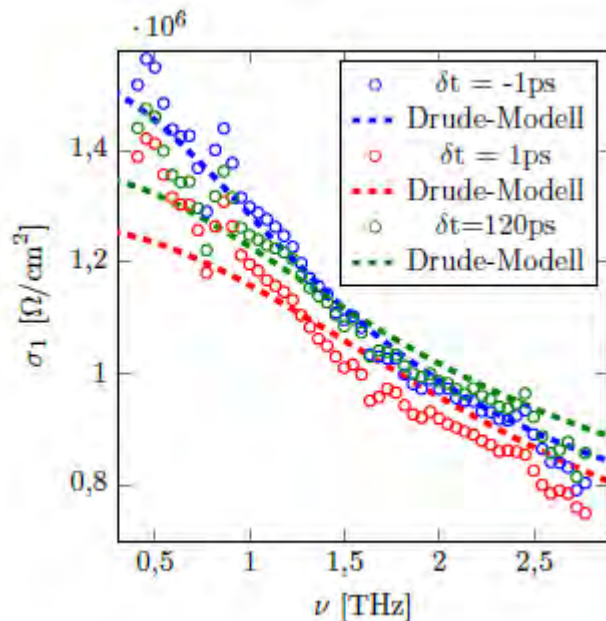
$$C_s \frac{dT_s}{dt} = -G_{sg}(T_s - T_g) - G_{se}(T_s - T_e)$$

Spectrally resolved THz conductivity dynamics



$\omega_p = 4000 \text{ cm}^{-1}$ $1/\tau = 500 \text{ cm}^{-1}$
 — σ_1 , — σ_2
 $\omega_p = 4000 \text{ cm}^{-1}$ $1/\tau = 650 \text{ cm}^{-1}$
 σ_1^* , σ_2^*
 (* excited - $1/\tau$ increases)

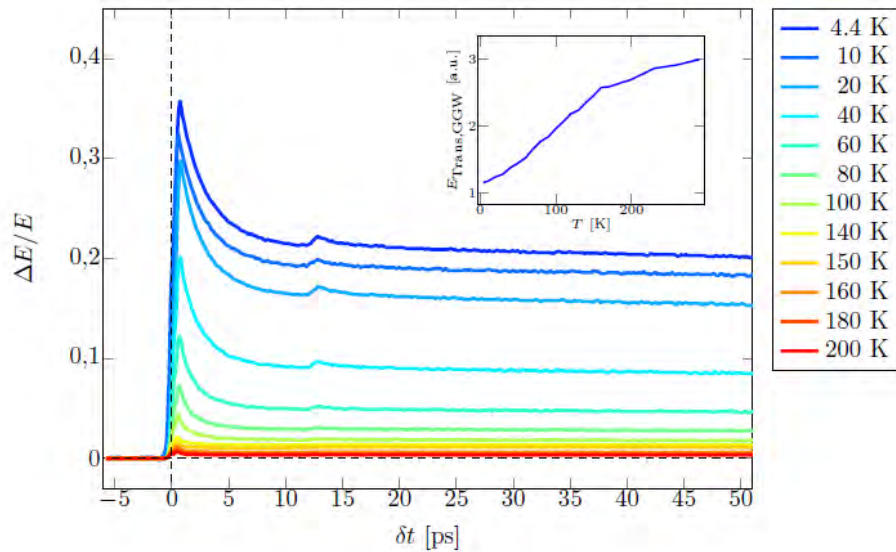
$$\hat{\sigma}(\omega) = \frac{Ne^2}{m} \frac{1}{1/\tau - i\omega}$$



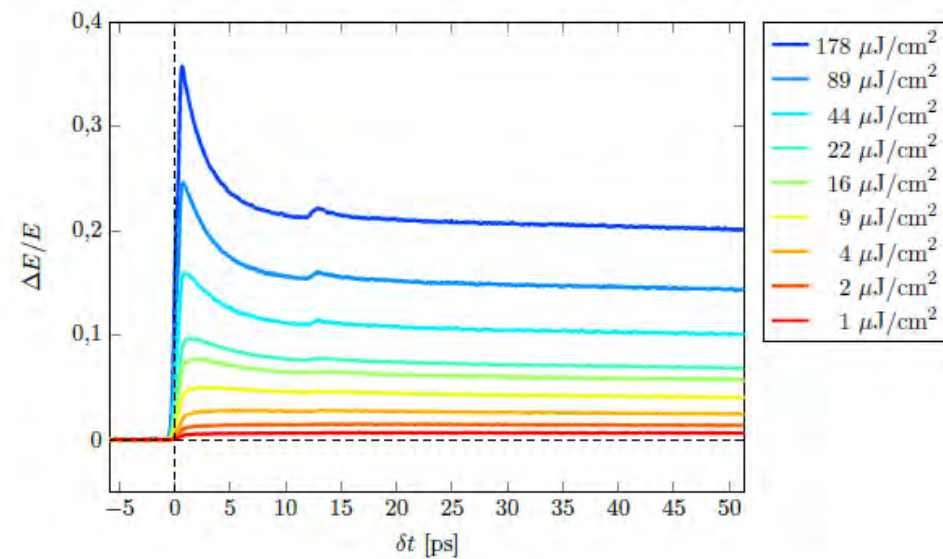
Data consistent with changes in the scattering rate!

SRO: 800 nm pump / THz probe dynamics

Temperature (T)-dependence



Power (P) -dependence

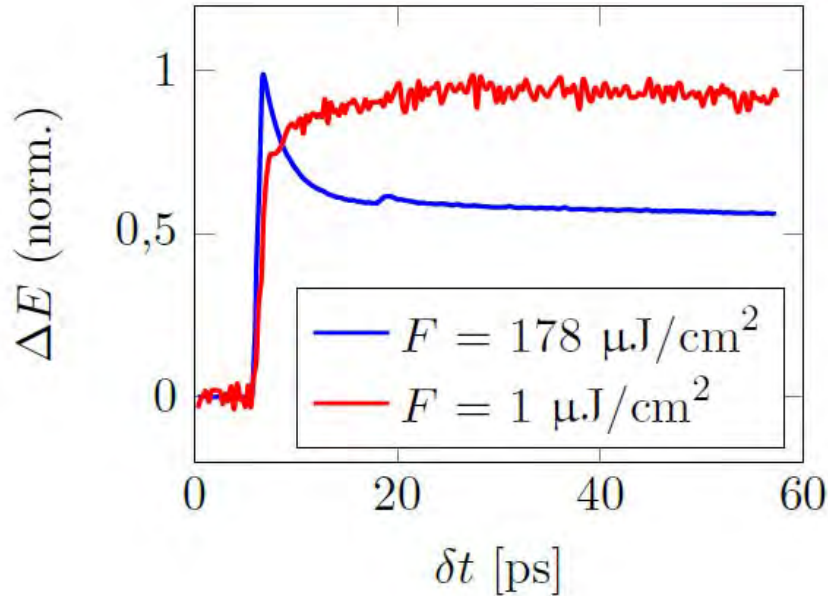


State 30 ps after photoexcitation:

from the T- and P-dependent amplitude of the induced change
+ the absorbed optical energy + specific heat

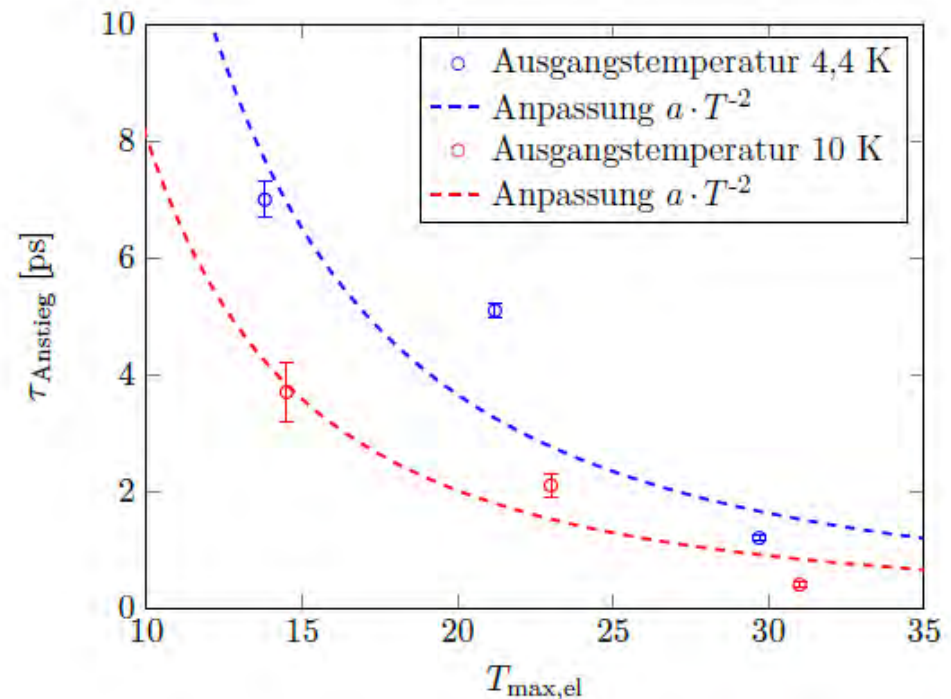
→ thermalized at temperature $T^* > T$

Dynamics at low-T and low - P

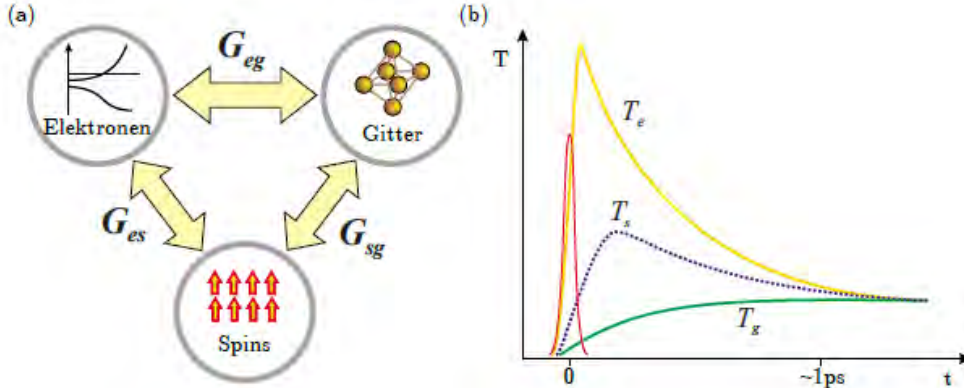


Observed only for very low P
and for $T < 10$ K

- at low-T: $\rho \sim T^2$ (Fermi liquid)
- e-e thermalization time (according to Fermi liquid) $\propto T^{-2}$
- **Observation of e-e thermalization**



What governs $\Delta\sigma(\omega)$ for $T > 10\text{-}20\text{ K}$?



$$C_e \frac{dT_e}{dt} = -G_{eg}(T_e - T_g) - G_{es}(T_e - T_s) + P(t)$$

$$C_g \frac{dT_g}{dt} = -G_{ge}(T_g - T_e) - G_{gs}(T_g - T_s)$$

$$C_s \frac{dT_s}{dt} = -G_{sg}(T_s - T_g) - G_{se}(T_s - T_e)$$

PHYSICAL REVIEW B 83, 134432 (2011)

Determination of the spin-flip time in ferromagnetic SrRuO₃ from time-resolved Kerr measurements

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A. J. H. M. Rijnders,⁴ R. Ramesh,^{1,3} and J. Orenstein^{1,2}

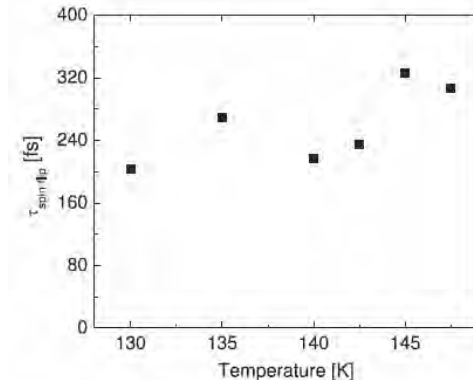
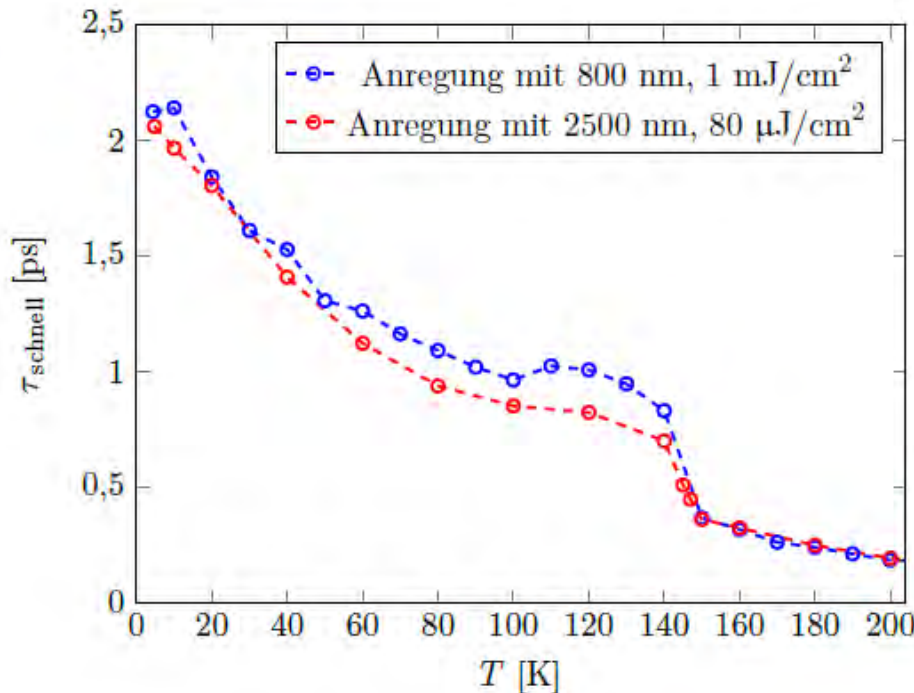


FIG. 10. Spin-flip time at high temperature.

For $T > 10\text{-}20\text{ K}$ relaxation governed by the spin-lattice relaxation

Time Resolved Kerr Studies of SrRuO₃

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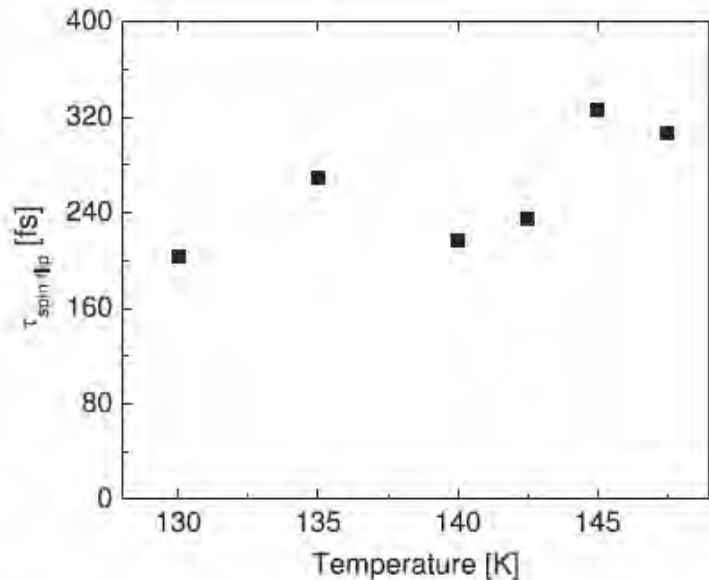
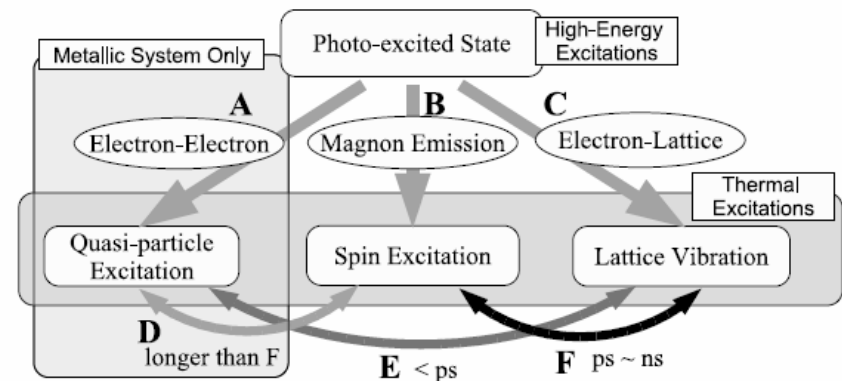
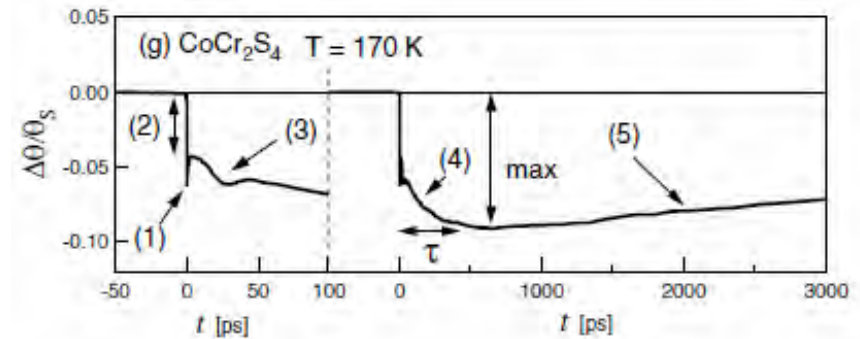


FIG. 10. Spin-flip time at high temperature.

General Features of Photoinduced Spin Dynamics in Ferromagnetic and Ferrimagnetic Compounds

T. Ogasawara,¹ K. Ohgushi,² Y. Tomioka,¹ K. S. Takahashi,² H. Okamoto,^{1,3} M. Kawasaki,^{1,4} and Y. Tokura^{1,2,5}



3-Temperature Model

Summary II (preliminary)

THz conductivity dynamics in SRO:

At low temperatures THz conductivity governed by the e-e thermalization time (slow)

Over most of the temperature range THz conductivity governed by scattering on magnons/para-magnons

Spin-flip time on the 100 fs timescale, recovery governed by the spin-lattice relaxation time.

Why anomaly at T_c remains unclear...

Future plans

- LCO/ LSCO photodoping (collaboration with Koren/Keren)
(some additional measurements / publication)
- SRO/NiPd (collaboration with Koren, Scheer, Leiderer)
(theory input ?, publication in preparation)
- SRO/LSCO multi-layers
(THz conductivity in eq. + dynamics, MO/characterization)