Locating the Missing Superconducting Electrons in the Overdoped Cuprates (and cyclotron resonance!)

N. Peter Armitage
The Johns Hopkins University
Canadian Institute for Advanced Research

Ivan Bozovic (BNL)

Xi He (BNL)

U.S. DEPARTMENT OF ENERGY

GORDON AND BETTY MOORE FOUNDATION

Kirk Post
(NHMFL)

Anaëlle Legros
(JHU)

Fahad Mahmood
(was JHU, now UIUC)

David Ingram
(Ohio University)

Jeffrey Clayhold
(a van, down by the river)

NSF

Scott Crooker
(NHMFL)

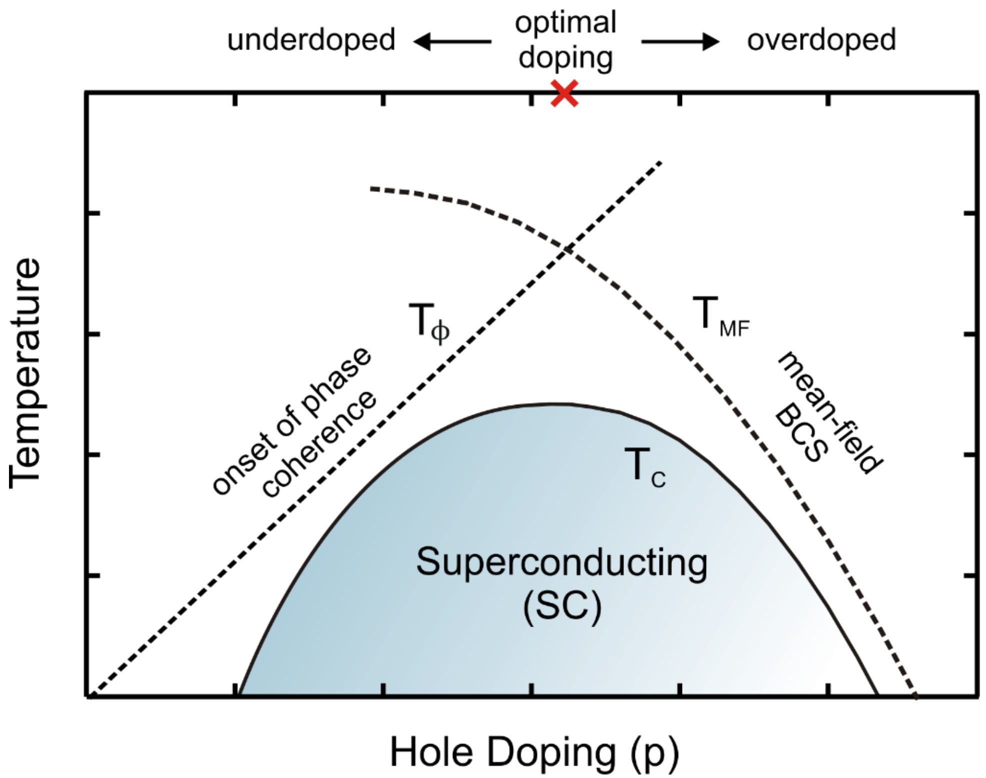
John Singleton
(NHMFL)

Ross McDonald
(NHMFL)

CIFAR

Overdoped Cuprates

High-T_c cuprates: Conventional Wisdom



e.g. Emery, V. & Kivelson, S. A.
Nature **374**, 434–437 (1994)

underdoped

- small n_s <--> small J_θ
- susceptible to phase fluctuations
- no mean-field description --> not BCS

overdoped

- large n_s <--> large J_θ
- rigid phase
- mean-field description --> BCS

Is n_s actually large for overdoped cuprates?

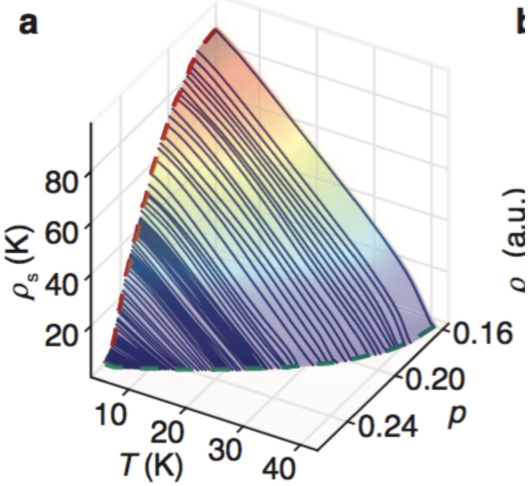
The puzzle of the missing superconducting electrons in overdoped cuprates

LETTER

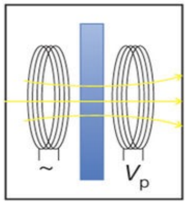
doi:10.1038/nature19061

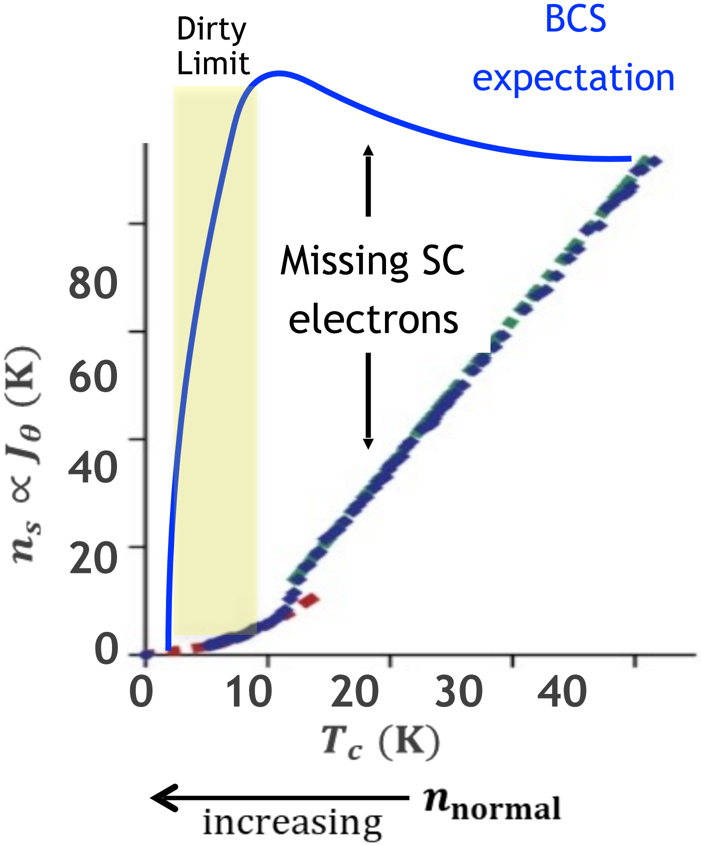
Dependence of the critical temperature in overdoped copper oxides on superfluid density

I. Božović[1,2], X. He[1,2], J. Wu[1] & A. T. Bollinger[1]



Two-coil mutual inductance; measure n_s in overdoped LSCO films



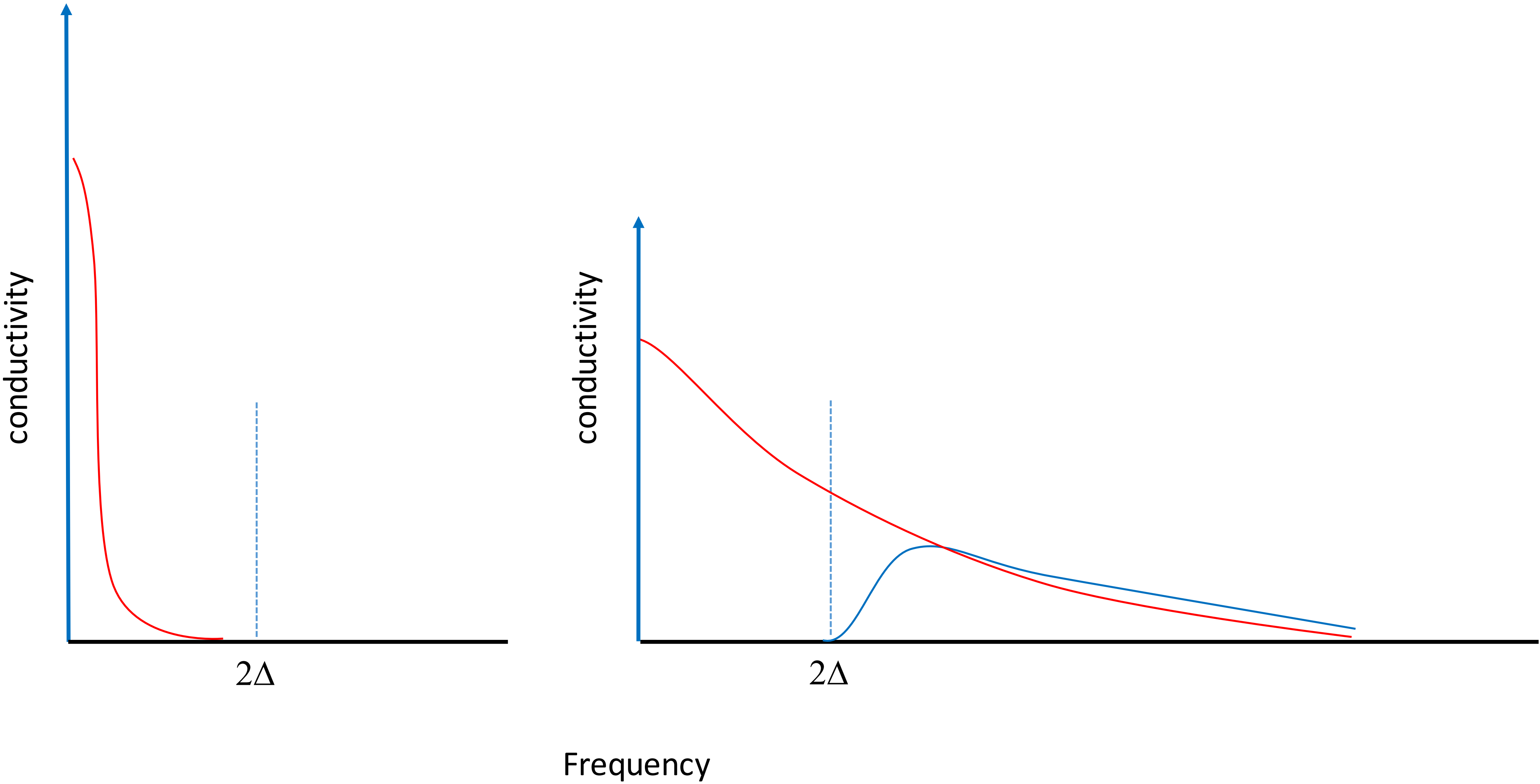


(1) Where are the missing SC electrons?

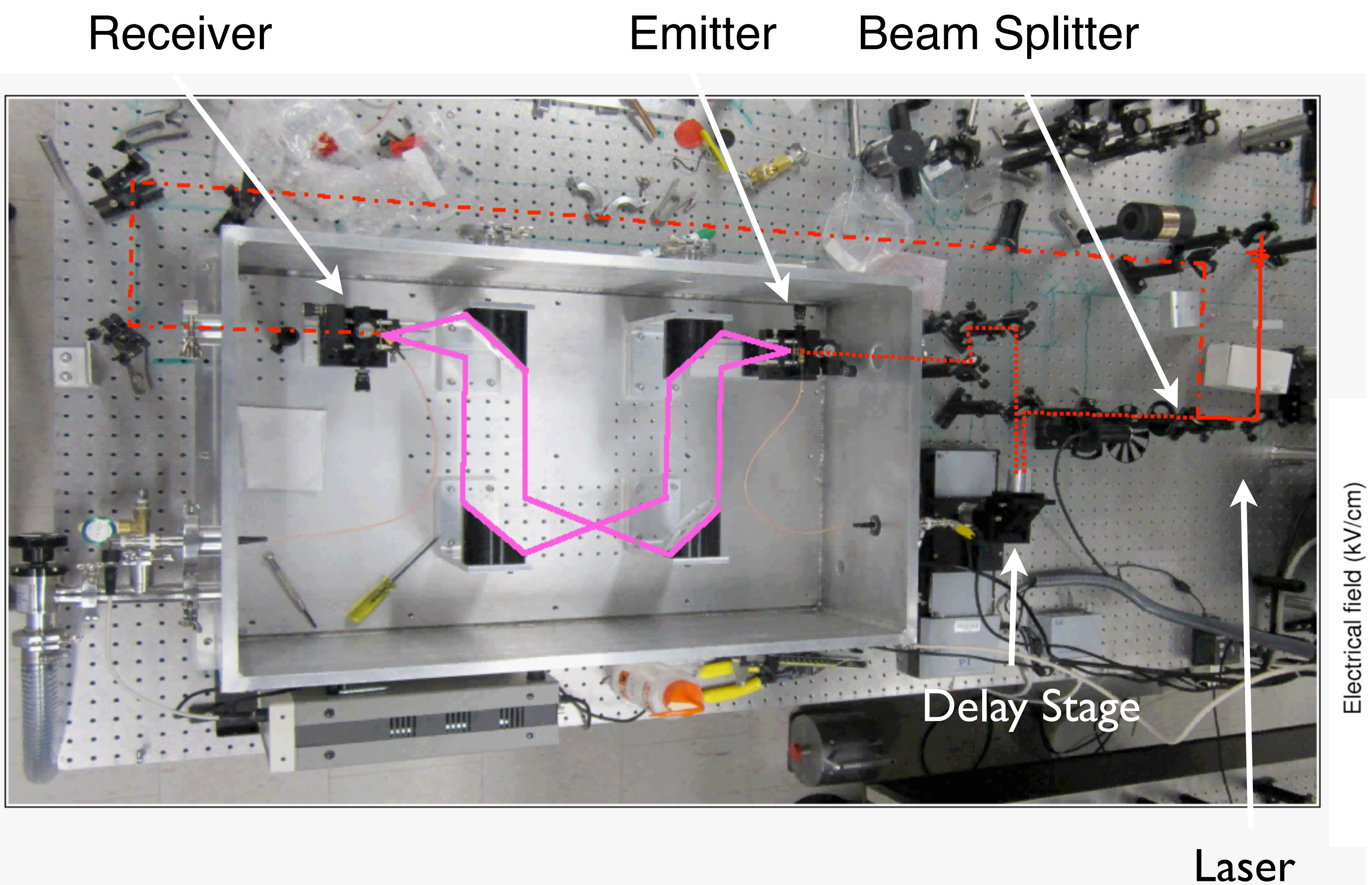
(2) Why do they fail to condense?

→ Combine THz optical conductivity with kHz mutual inductance

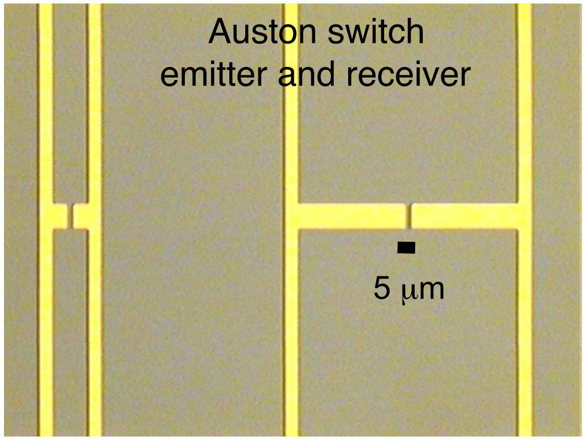
Expectation for clean and dirty BCS



Time Domain THz Spectroscopy



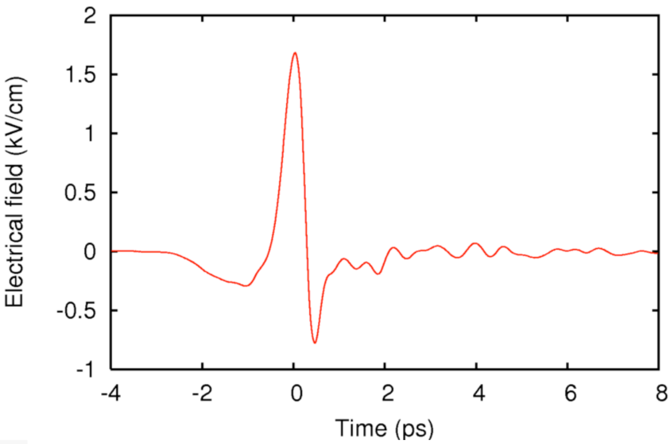
Laser
800nm
60fs

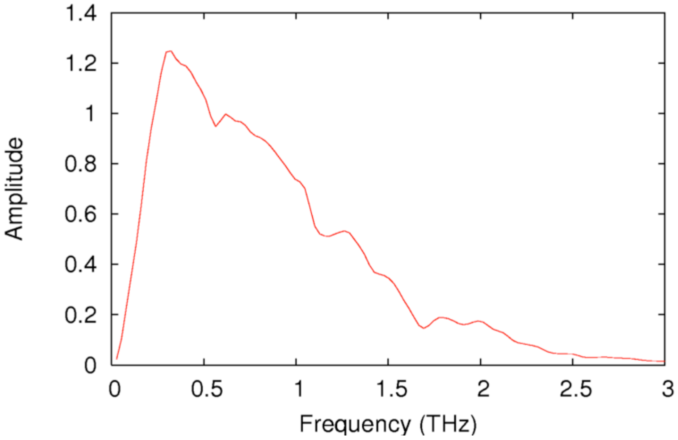


- fs laser excites photoconductive emitter and receiver. Coherent detection of field allows **complex** optical response functions to be measured

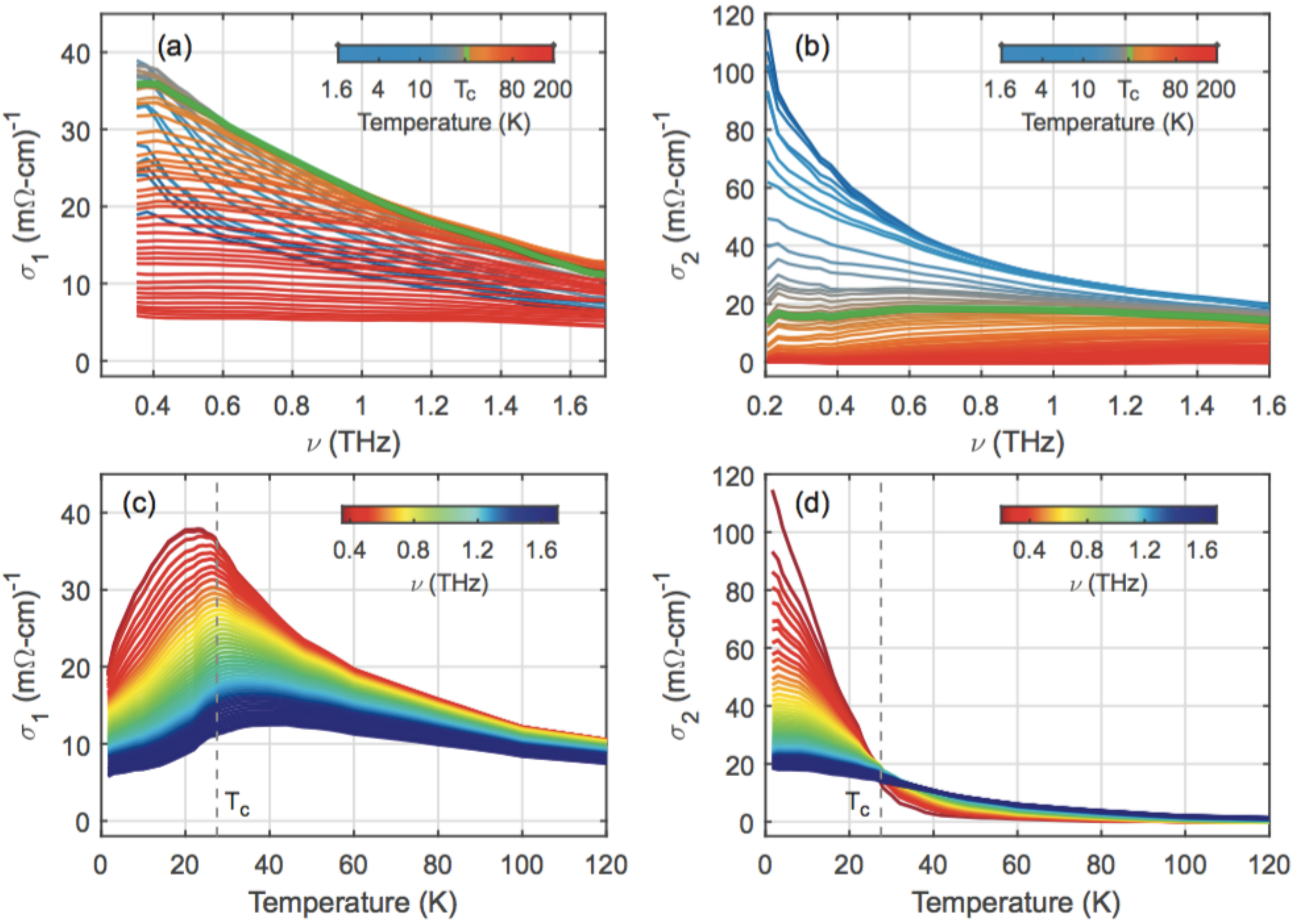
$$T(\omega) = \frac{4n}{n+1}\,\frac{e^{i\Phi_s}}{n+1+\sigma(\omega)dZ_0}$$

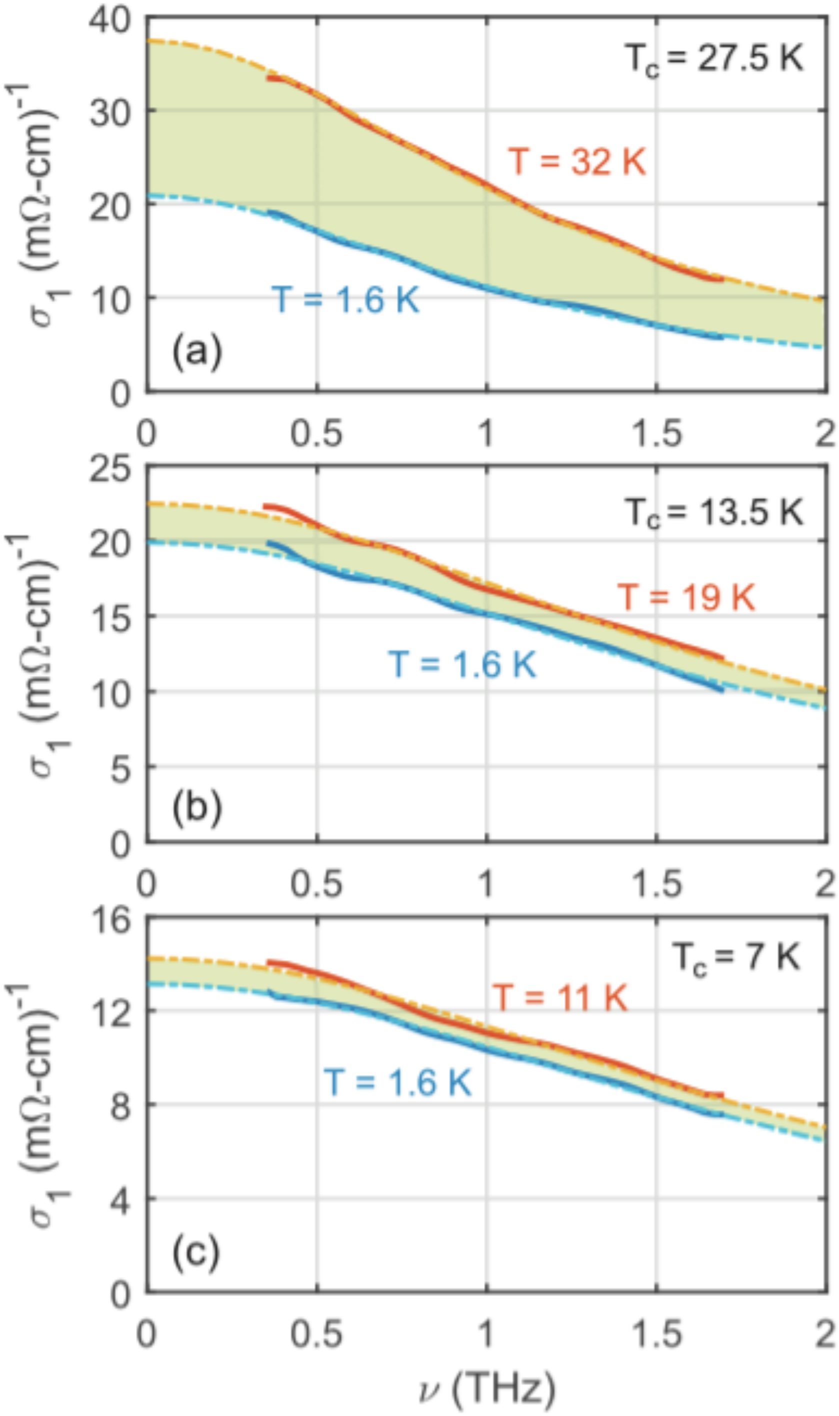
- 100 GHz - 3 THz (0.8 meV - 12 meV), @ 1.4K - 300K.





THz optical conductivity x = 0.23 $La_{2-x}Sr_xCuO_4$ thin film (T_c = 27.5 K).



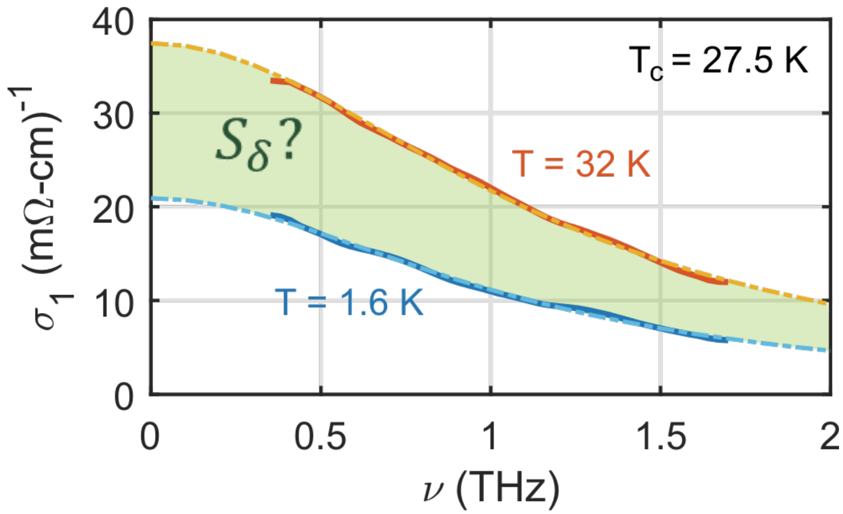


Significant residual THz conductivity in all overdoped $La_{2-x}Sr_xCuO_4$ samples

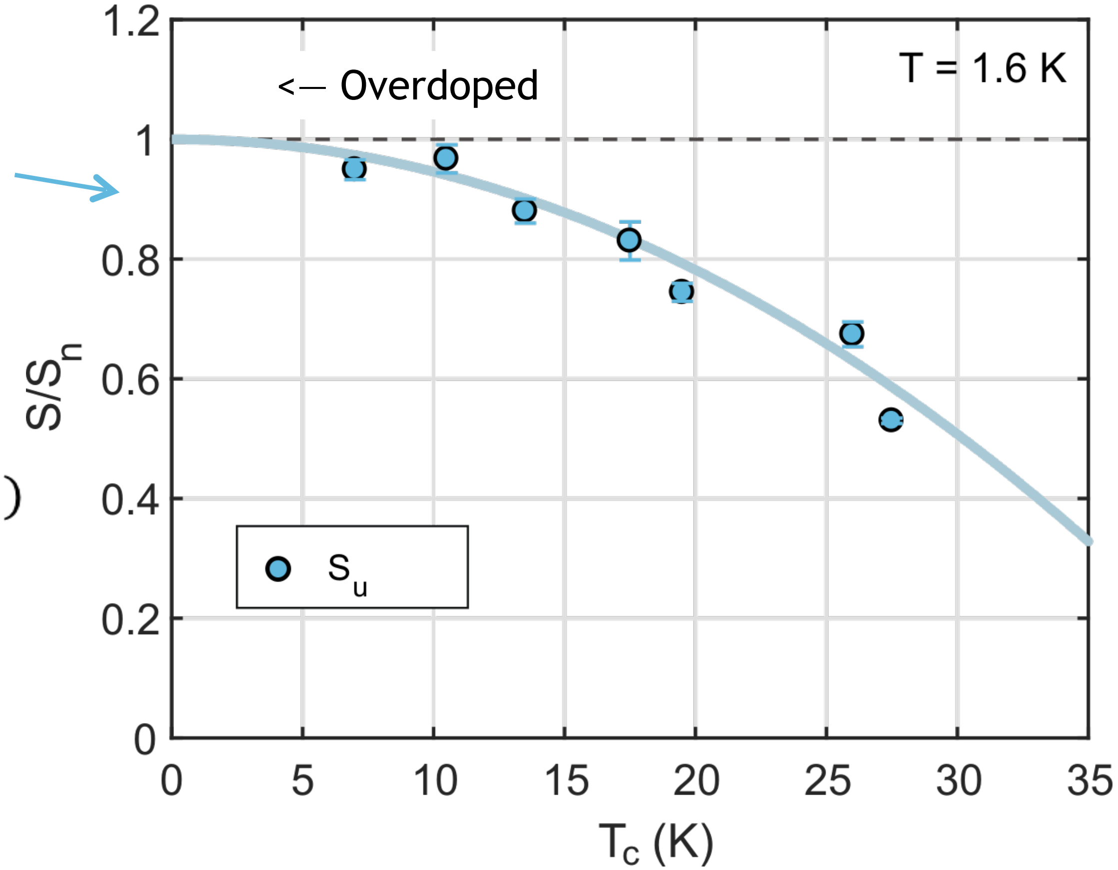
Larger residual for most overdoped

Residual and normal state real conductivity

Determine S_u and S_n using THz conductivity



Sum rule: $S_n - S_u = S_\delta$



Single Drude fit: $\sigma_1(\nu) = S\tau/(1+\nu^2\tau^2)$

$$\int_{0+}^{\infty} \sigma_1(\nu)d\nu = \frac{\pi}{2}S$$

--> normal state spectral weight (S_n)

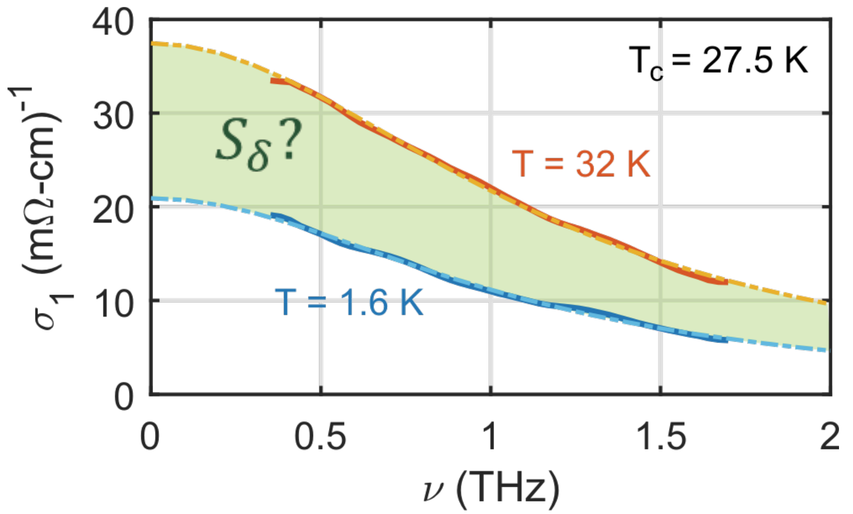
--> uncondensed spectral weight (S_u)

No sign of BCS d-wave gap, $2\Delta = 4.28k_BT_c$

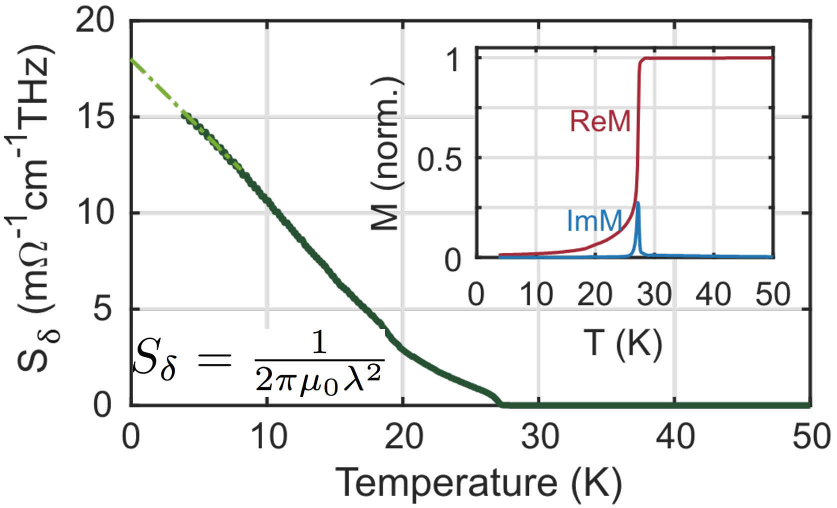
Proportion of uncondensed carriers increases as $T_c \rightarrow 0$

Residual and normal state real conductivity

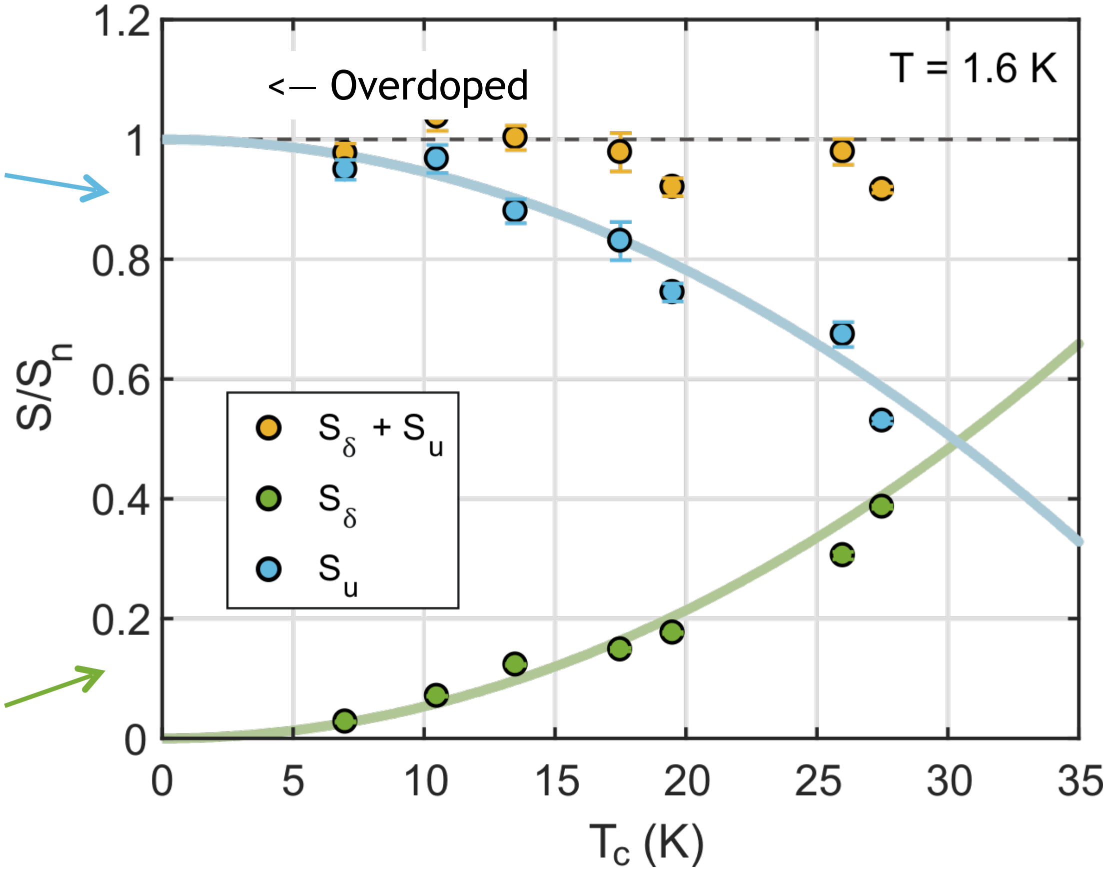
Determine S_u and S_n using THz conductivity



Determine S_δ using mutual inductance on the *same* films



Sum rule: $S_n - S_u = S_\delta$



Residual Drude peak obeys sum rule

Uncondensed superconducting electrons – why?

Possibilities….

1) Pair-breaking scattering due to impurities which smears out d-wave node (dirty d-wave)

2) Gross inhomogeneity e.g. macroscopic normal regions of the sample.

3) Other effects e.g. fluctuations of various kinds, inelastic scattering.

Uncondensed superconducting electrons – why?

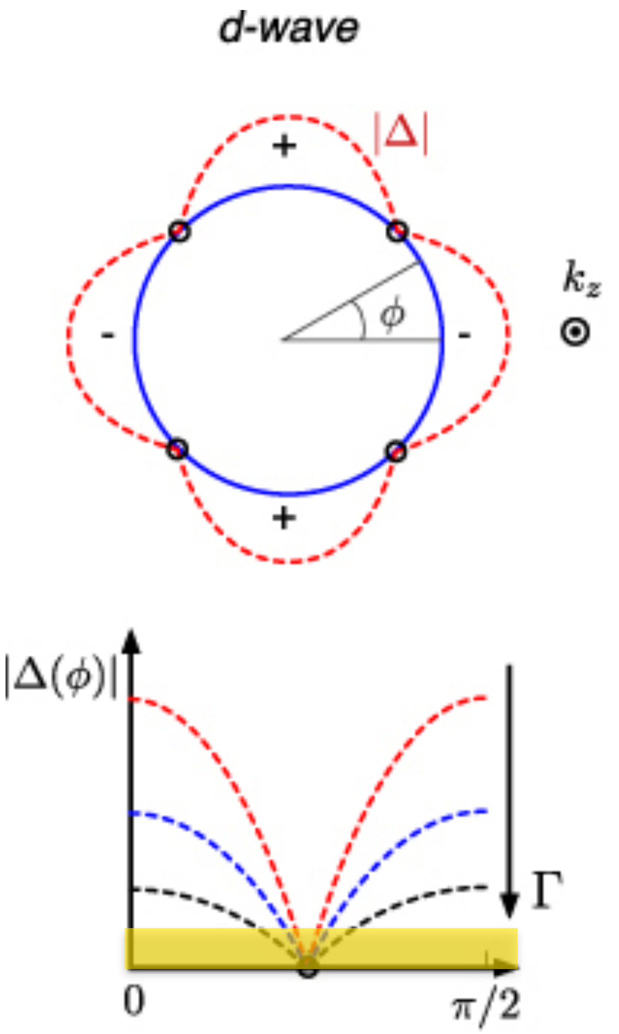
Possibilities….

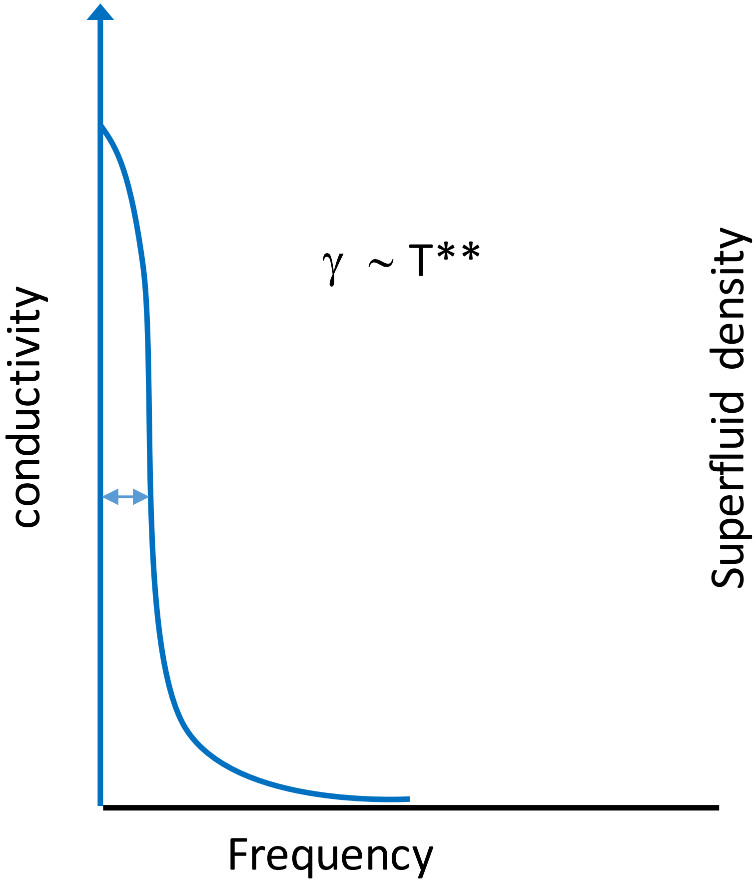
1) Pair-breaking scattering due to impurities which smears out d-wave node (dirty d-wave)

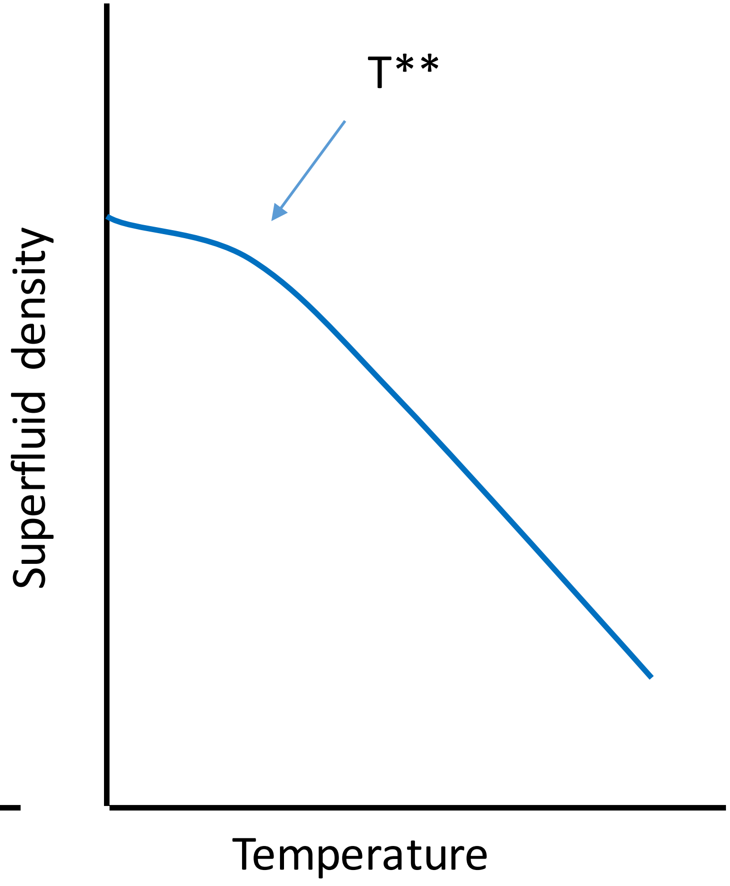
2) Gross inhomogeneity e.g. macroscopic normal regions of the sample.

3) Other effects e.g. fluctuations of various kinds, inelastic scattering.

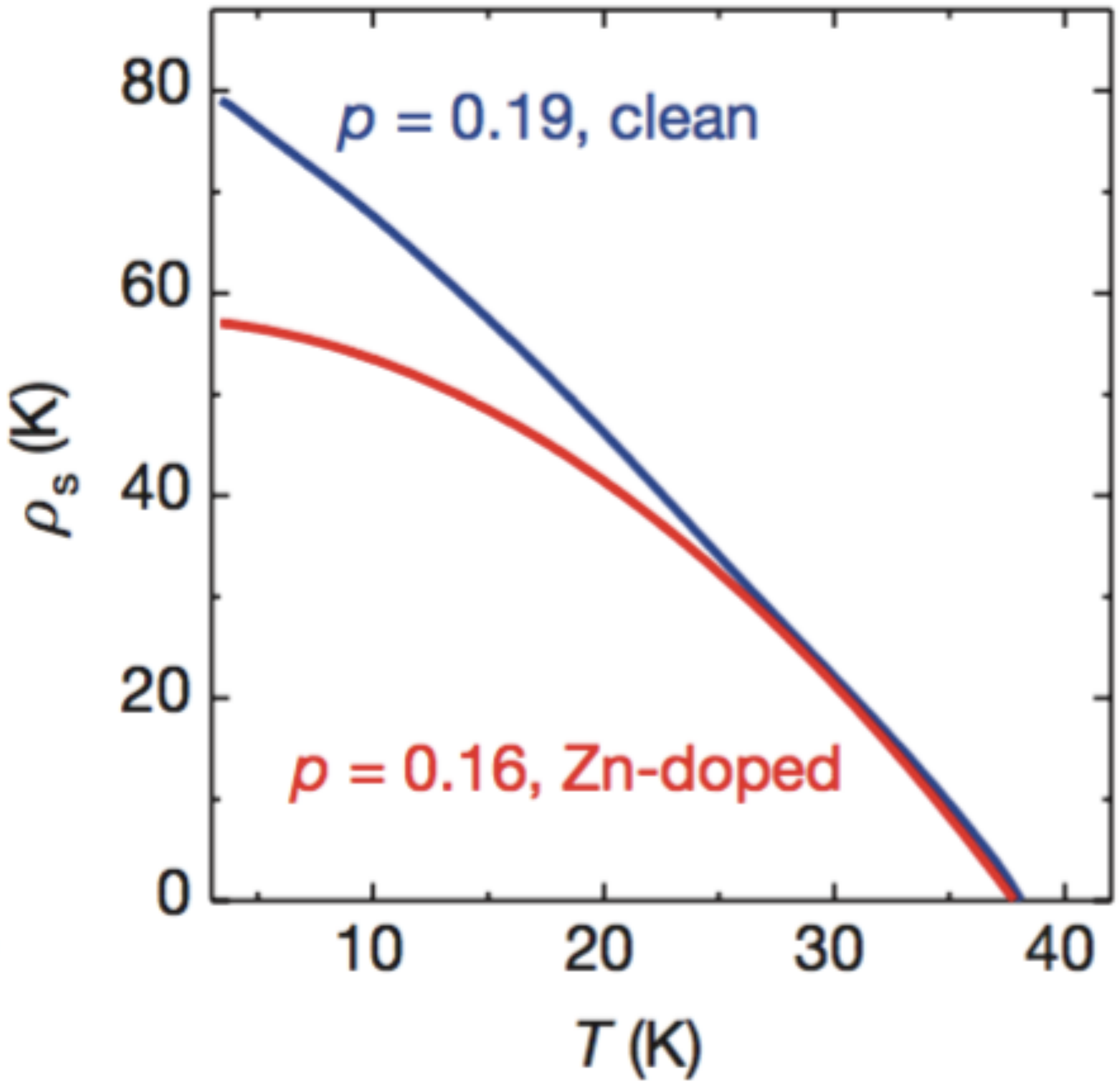
Expectations for dirty d-wave





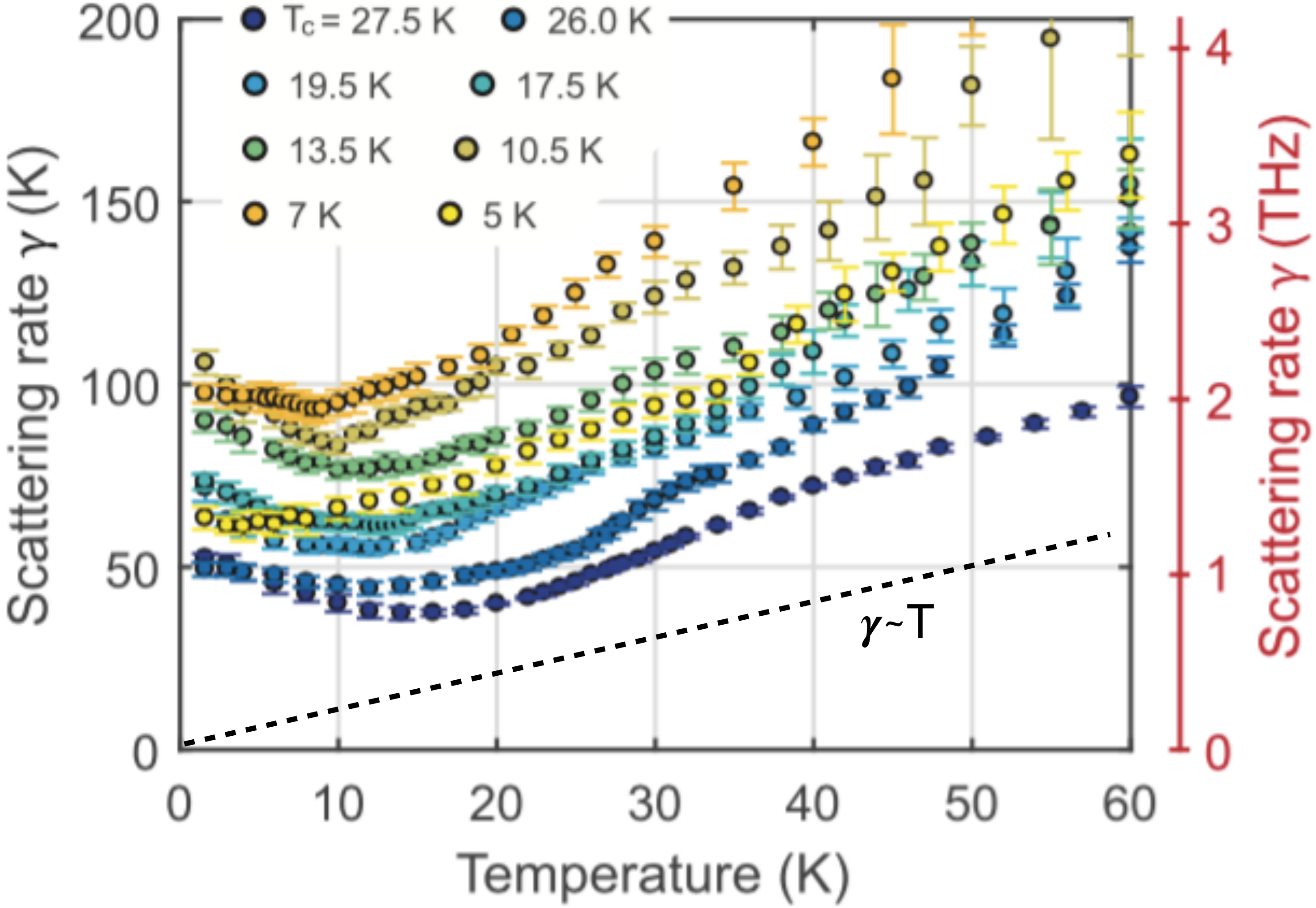


Expectations for dirty d-wave



Bozovic et al. 2016

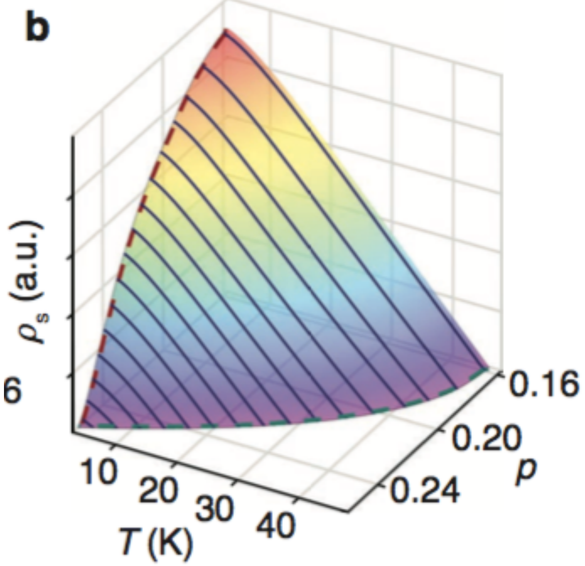
Drude width as a function of temperature

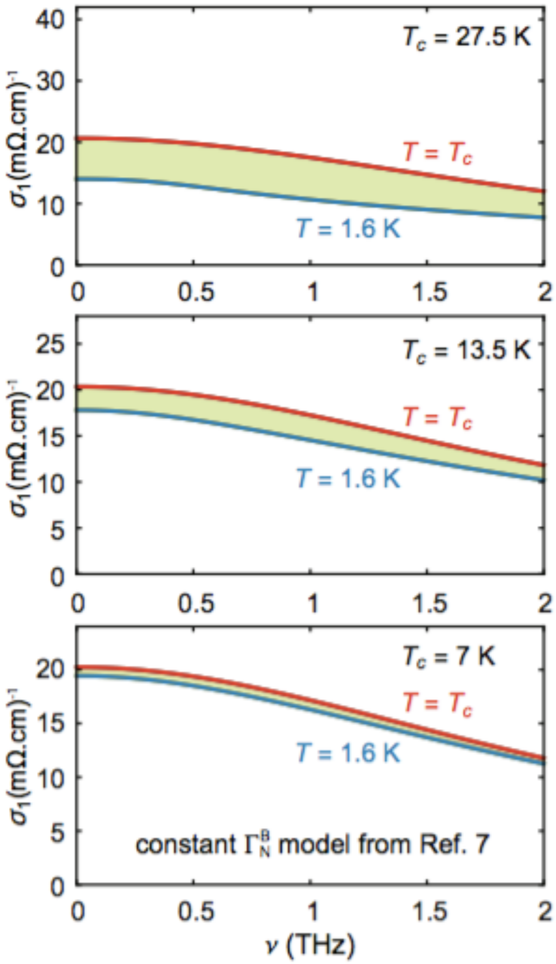


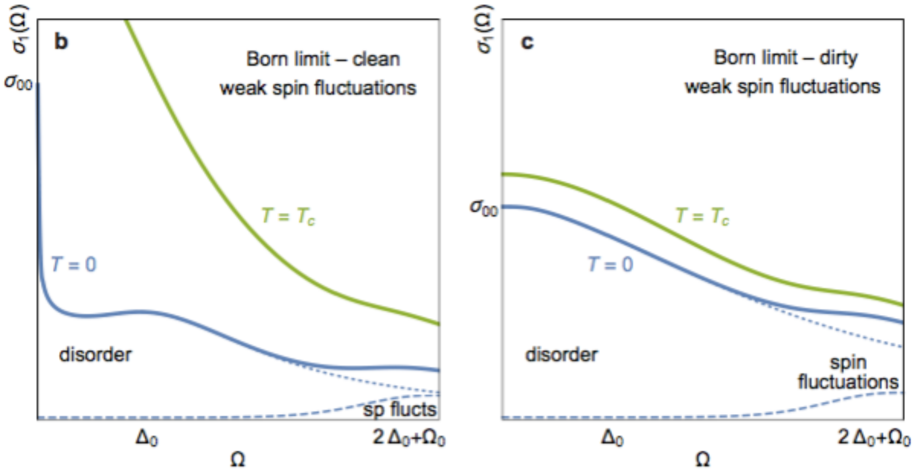
In unitary scattering limit γ>T means T** > T_c

Optical conductivity of overdoped cuprate superconductors: application to LSCO

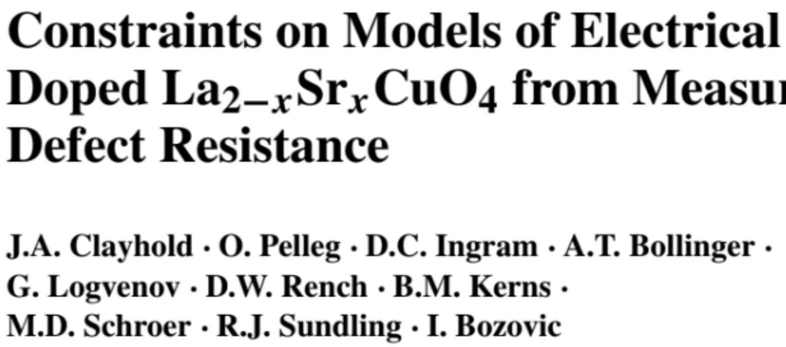
N. R. Lee-Hone,[1] V. Mishra,[2] D. M. Broun,[1,3] and P. J. Hirschfeld[4]
[1] *Department of Physics, Simon Fraser University, Burnaby, BC, V5A 1S6, Canada*
[2] *Computer Science and Mathematics Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA*
[3] *Canadian Institute for Advanced Research, Toronto, ON, MG5 1Z8, Canada*
[4] *Department of Physics, U. Florida, Gainesville FL 32611*







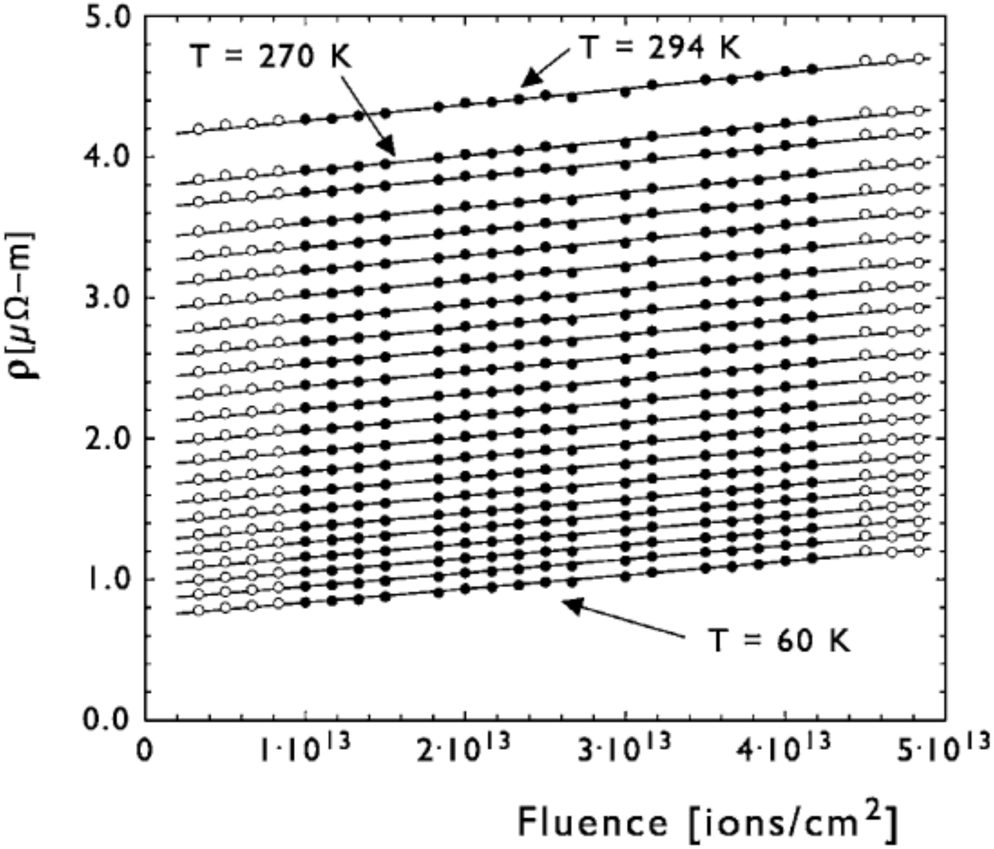
Radiation induced disorder/defects

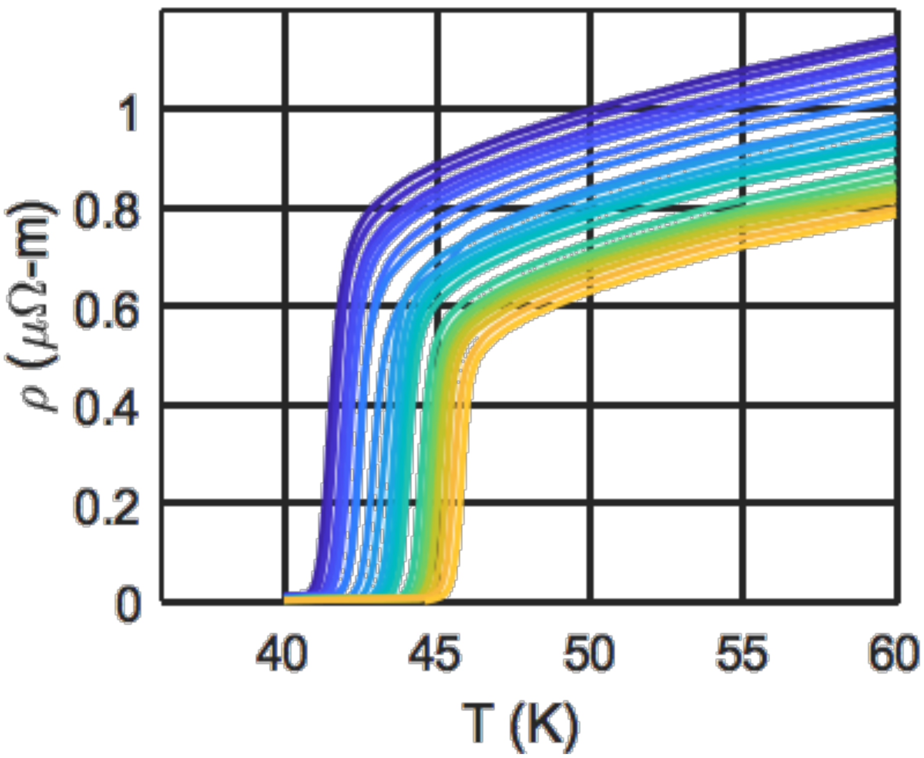


Constraints on Models of Electrical Transport in Optimally Doped $La_{2-x}Sr_xCuO_4$ from Measurements of Radiation-Induced Defect Resistance

J.A. Clayhold · O. Pelleg · D.C. Ingram · A.T. Bollinger · G. Logvenov · D.W. Rench · B.M. Kerns · M.D. Schroer · R.J. Sundling · I. Bozovic

Radiation is 1 MeV oxygen ions;
Columnar tracks through film

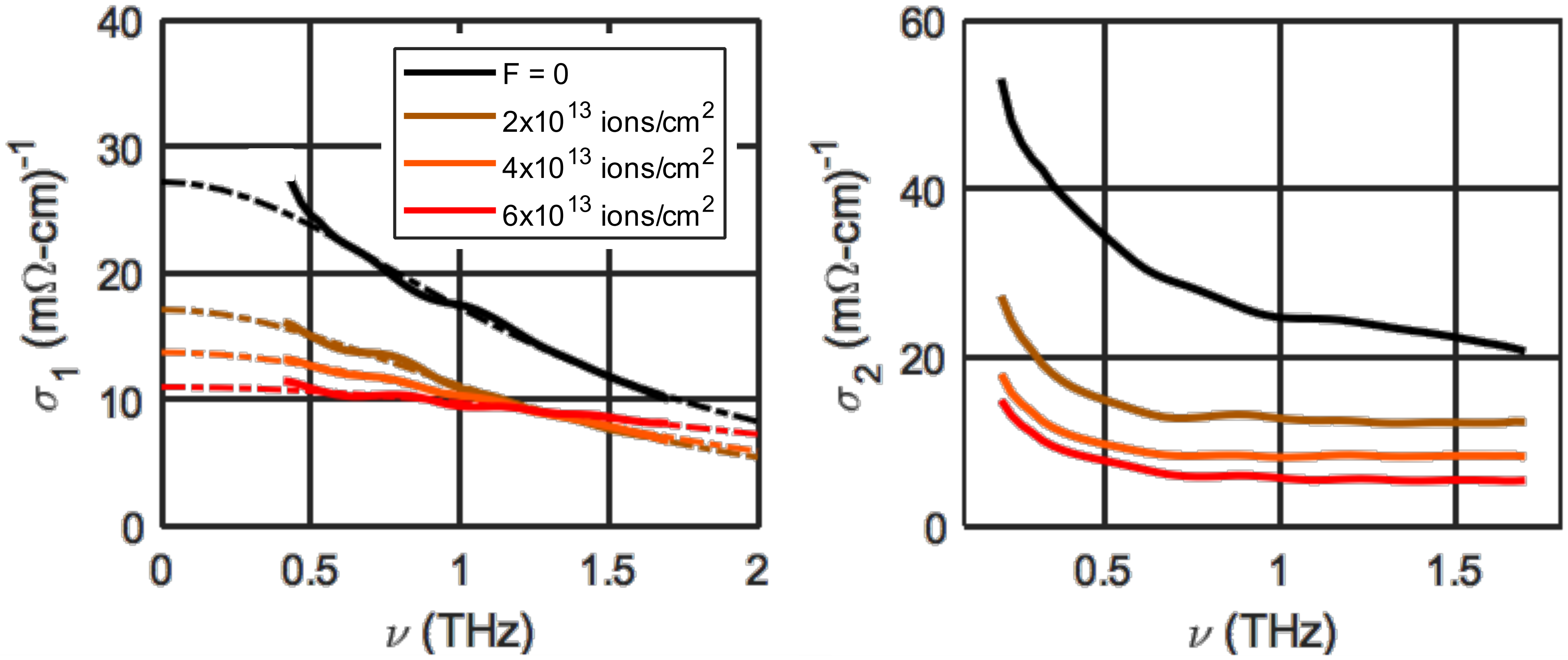




Mahmood et al. to be submitted PRB 2020

Low-T uncondensed carrier conductivity

Original T_c = 19.5 K, plots at T ~ 1.6 K

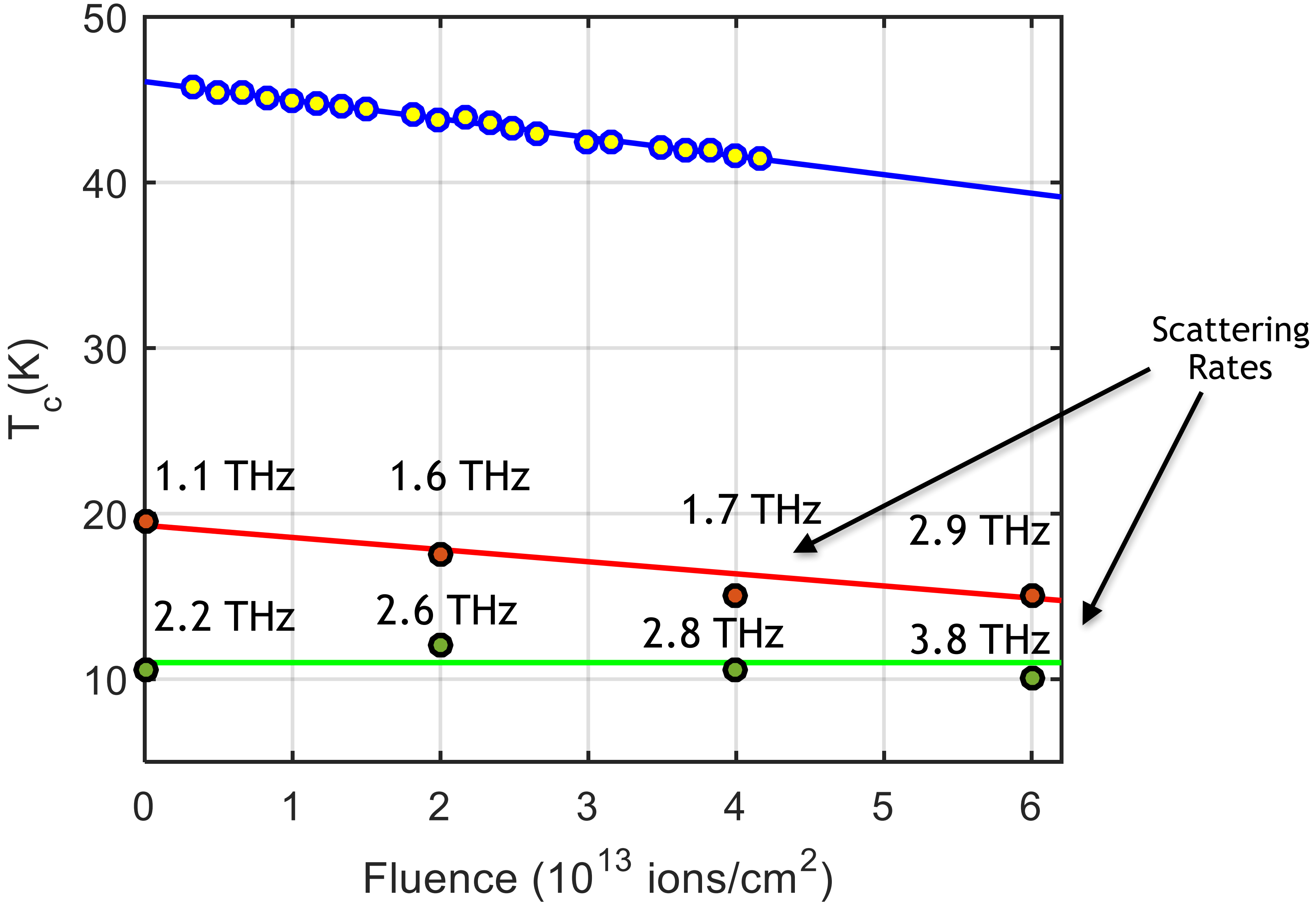


single Drude fit: $\sigma_1(\nu) = S\tau/(1+\nu^2\tau^2)$

Both normal state and low-T residual response remains single Drude

Mahmood et al. to be submitted PRB 2020

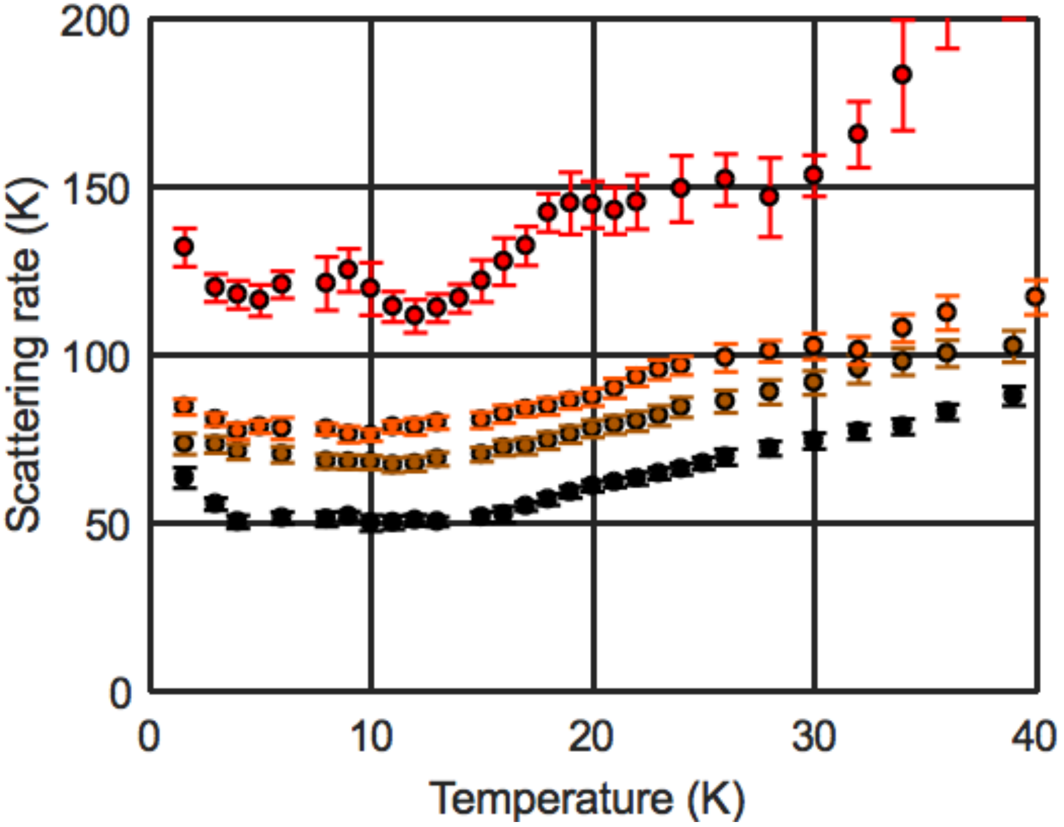
Effect of disorder on T_c

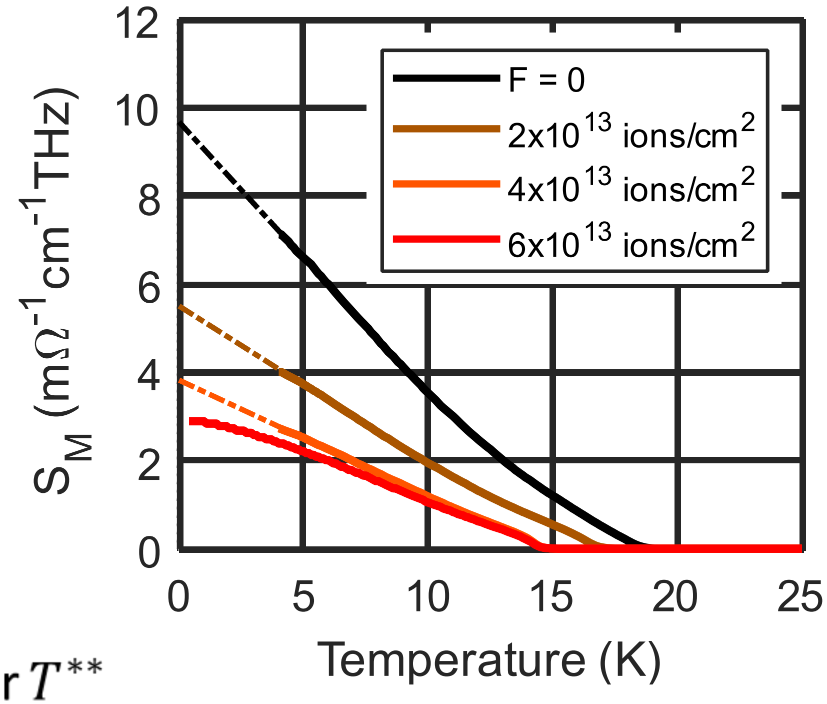


Mahmood et al. to be submitted PRB 2020

Scattering rate analysis

Original T_c = 19.5 K





For all films, $\gamma > T_c$, expect $\Delta n_s \sim T^2$ but…

…$n_s(T)$ remains linear as $T \to 0$, no crossover T^{**}

… maybe crossover for largest irradiation but T** still too small

Mahmood et al. to be submitted PRB 2020

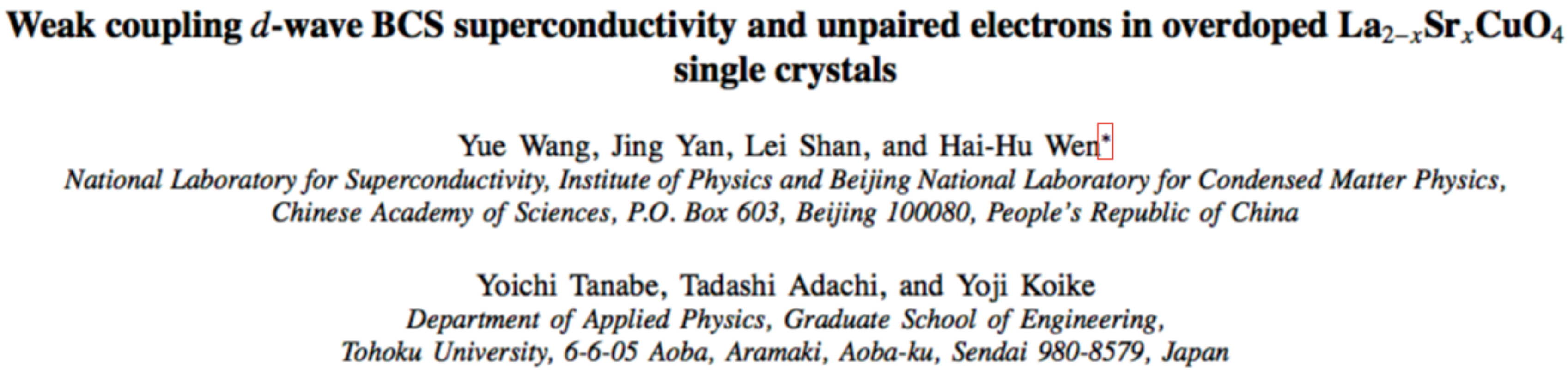
Uncondensed superconducting electrons – why?

Possibilities….

1) Pair-breaking scattering due to impurities which smears out d-wave node (dirty d-wave)

2) Gross inhomogeneity e.g. macroscopic normal regions of the sample.

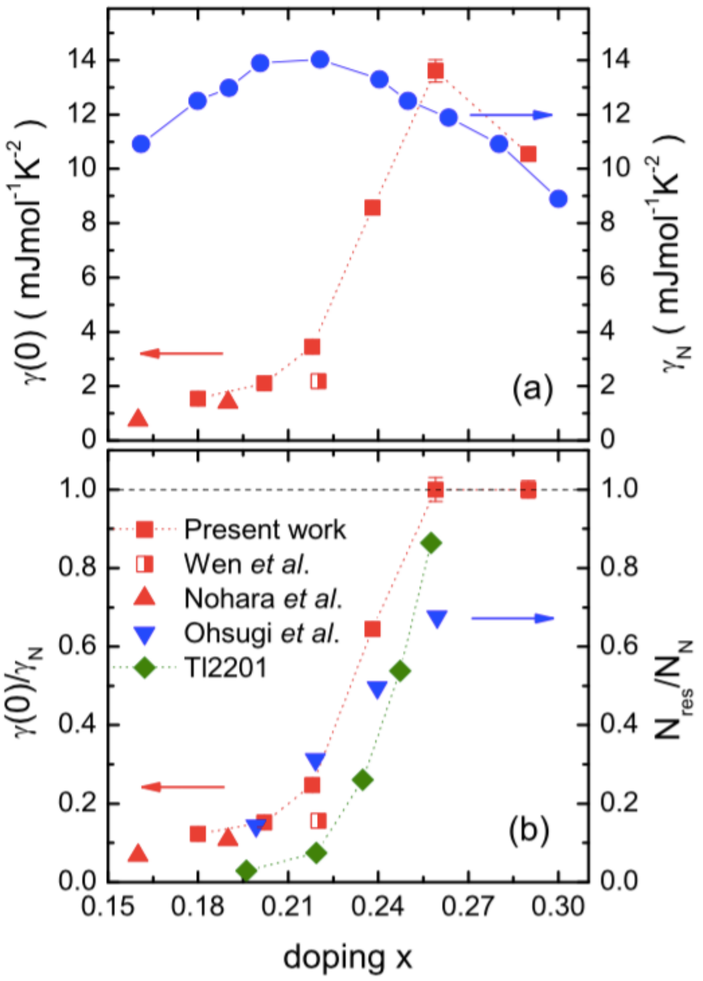
3) Other effects e.g. fluctuations of various kinds, inelastic scattering.



Weak coupling *d*-wave BCS superconductivity and unpaired electrons in overdoped $La_{2-x}Sr_xCuO_4$ single crystals

Yue Wang, Jing Yan, Lei Shan, and Hai-Hu Wen[*]
National Laboratory for Superconductivity, Institute of Physics and Beijing National Laboratory for Condensed Matter Physics,
Chinese Academy of Sciences, P.O. Box 603, Beijing 100080, People's Republic of China

Yoichi Tanabe, Tadashi Adachi, and Yoji Koike
Department of Applied Physics, Graduate School of Engineering,
Tohoku University, 6-6-05 Aoba, Aramaki, Aoba-ku, Sendai 980-8579, Japan



Very large residual fermonic heat capacity in overdoped LSCO.

For overdoped samples with $T_c \sim 20$ K the heat capacity coefficient was roughly 70% of the normal state and reached essentially 100 % by Tc ~ 7 K.

Interpreted in terms of large scale inhomogeneity

See also work by Barisic and Greven (dc, magnetization) that interprets data in terms of percolative transition.

Uncondensed superconducting electrons – why?

Possibilities….

1) Pair-breaking scattering due to impurities which smears out d-wave node (dirty d-wave)

2) Gross inhomogeneity e.g. macroscopic normal regions of the sample.

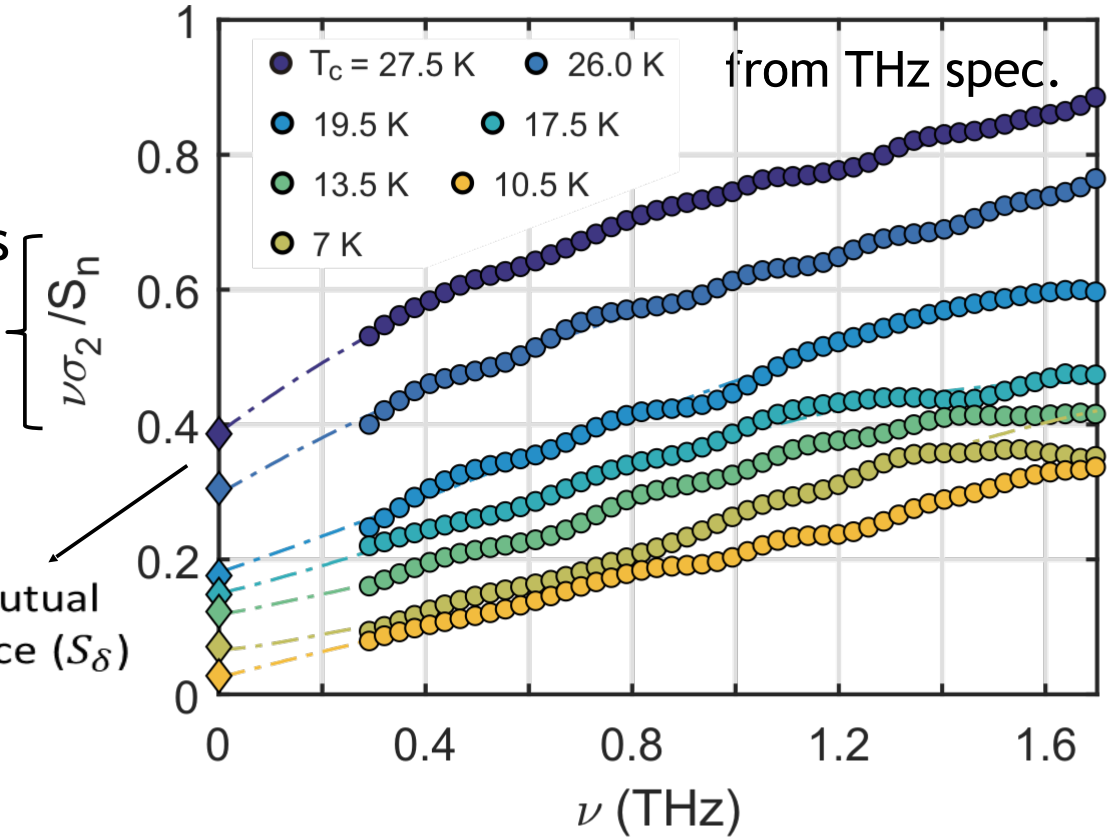
3) Other effects e.g. fluctuations of various kinds, inelastic scattering.

Superfluid phase stiffness

$\nu\sigma_2$ --> measure of J_θ over different length/time scales

renormalized phase stiffness

$$\frac{J_\theta(\nu)}{J_0}$$



from mutual inductance (S_δ)

$J_\theta(\nu)$ increases with probing frequency --> system appears ‘stiffer’

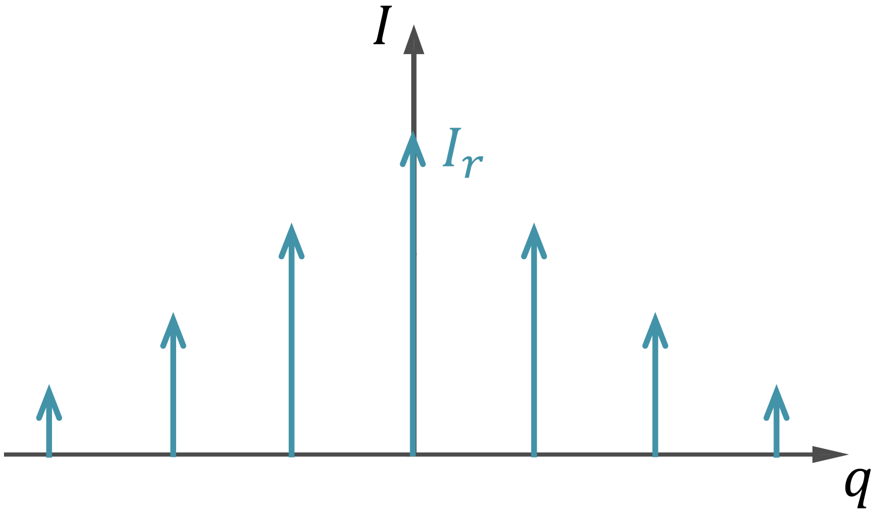
--> phase fluctuations degrade J_θ at longer length/time scales

Uncondensed superconducting electrons – why?

Quantum phase fluctuations - Debye-Waller factor

Bragg scattering

thermal/quantum vibration of atoms, u

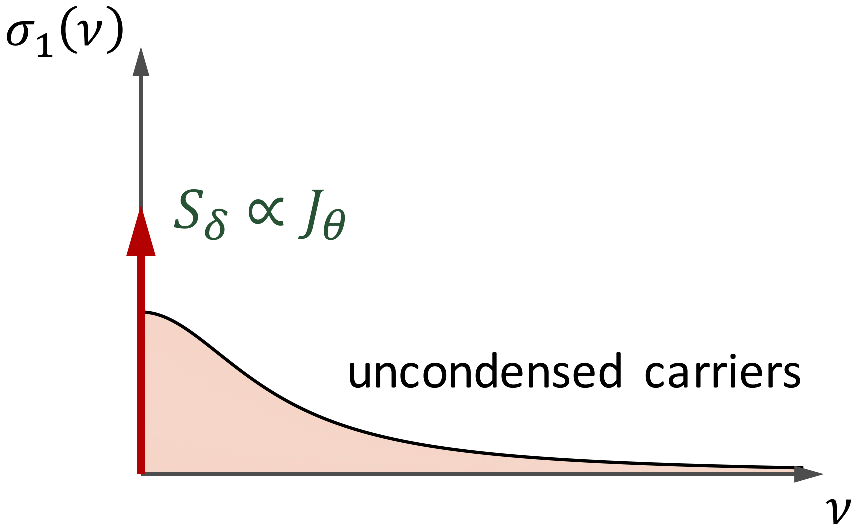


$$\frac{I_r}{I_0} = \exp(-q^2\langle u^2\rangle/2)$$

Suppression set by $\langle u^2\rangle$

SC complex conductivity

quantum phase fluctuations, $\delta\theta$



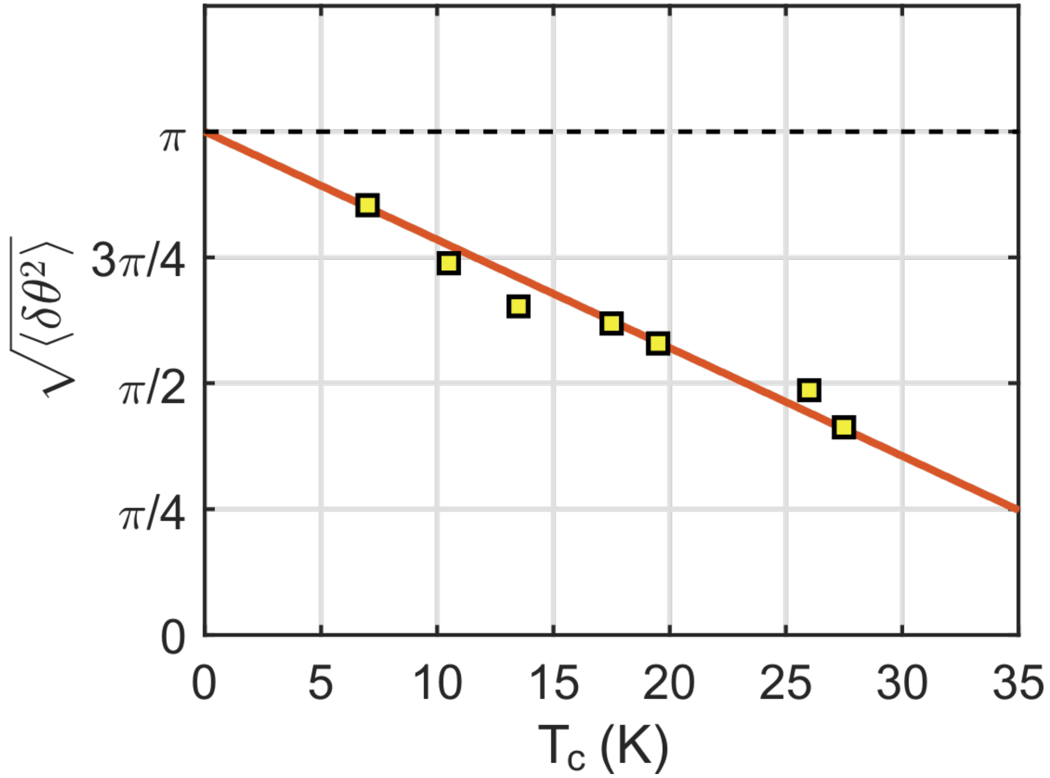
$$\frac{J_\theta}{J_0} = \frac{S_\delta}{S_n} = \exp(-\langle\delta\theta^2\rangle/2)$$

Within **self-consistent harmonic approximation** suppression set by $\langle\delta\theta^2\rangle$

Uncondensed superconducting electrons – why?

Quantum phase fluctuations

Quantum Debye-Waller factor



Renormalization of the SC phase stiffness

$$\frac{J_\theta}{J_0} = \frac{S_\delta}{S_n} = \exp(-\langle\delta\theta^2\rangle/2)$$

Max. T_c: $\langle\delta\theta\rangle_{rms} \to 0$

$T_c \to 0$: $\langle\delta\theta\rangle_{rms} \to \pi$

Time-domain THz spectroscopy --> superfluid phase stiffness

Prominent role of quantum phase fluctuations for overdoped $La_{2-x}Sr_xCuO_4$

Locating the missing superconducting electrons in overdoped cuprates

Wanted to explain the small overdoped superfluid density of Bozovic et al. (missing electrons)

We find large residual Drude deep into the superconducting state, proportion of uncondensed electrons increases with over doping.

Large width, much in excess of T_c, but with linear in T superfluid density

A number of explanations are possible:

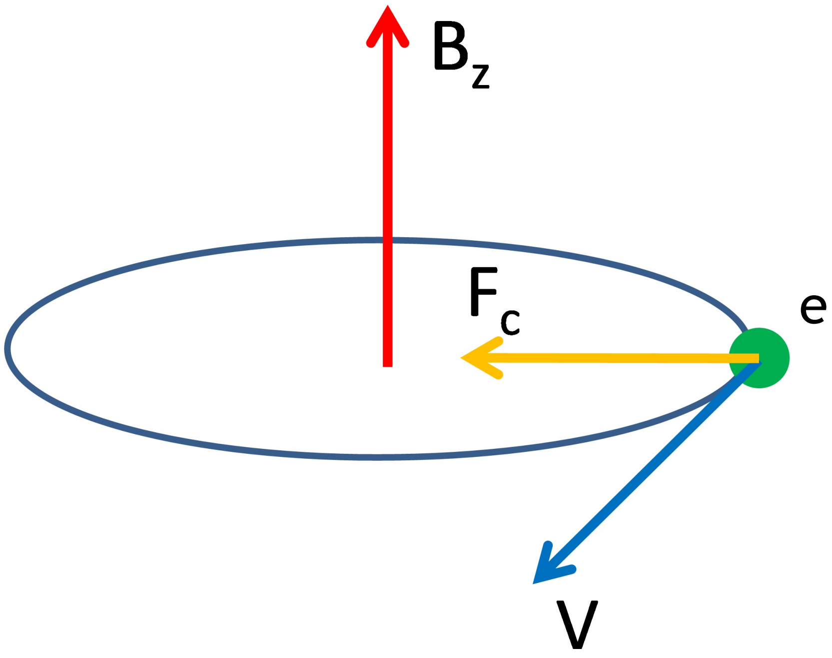
Open questions whether or not the “dirty d-wave” (with Born scattering) is in the physical limit

Irradiation studies bring open questions about whether Born scattering model is consistent with totality of data.

Interesting correlations found in the generalized sf density demonstrating the possible relevance of phase fluctuations on the approach to the OD critical point.

Cyclotron Resonance!

Cyclotron resonance



Semiclassical Lorentz force

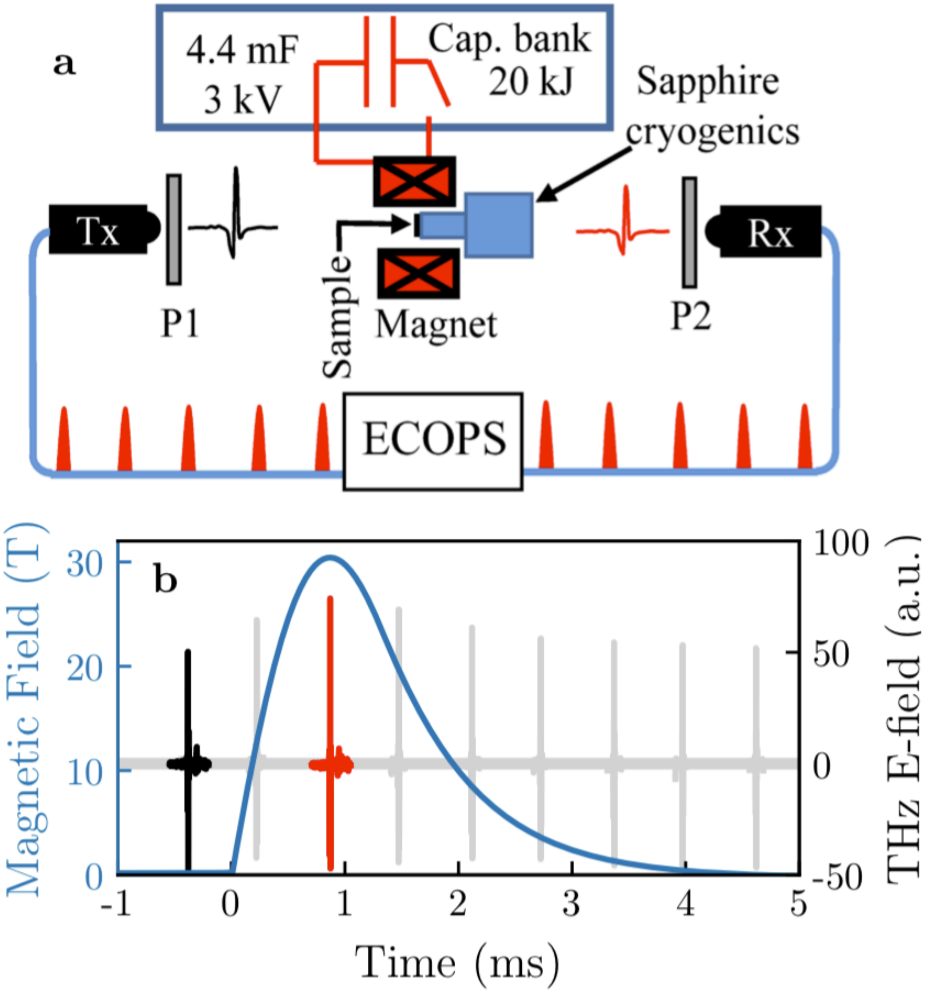
$F_c = mv^2/r = evB_z$

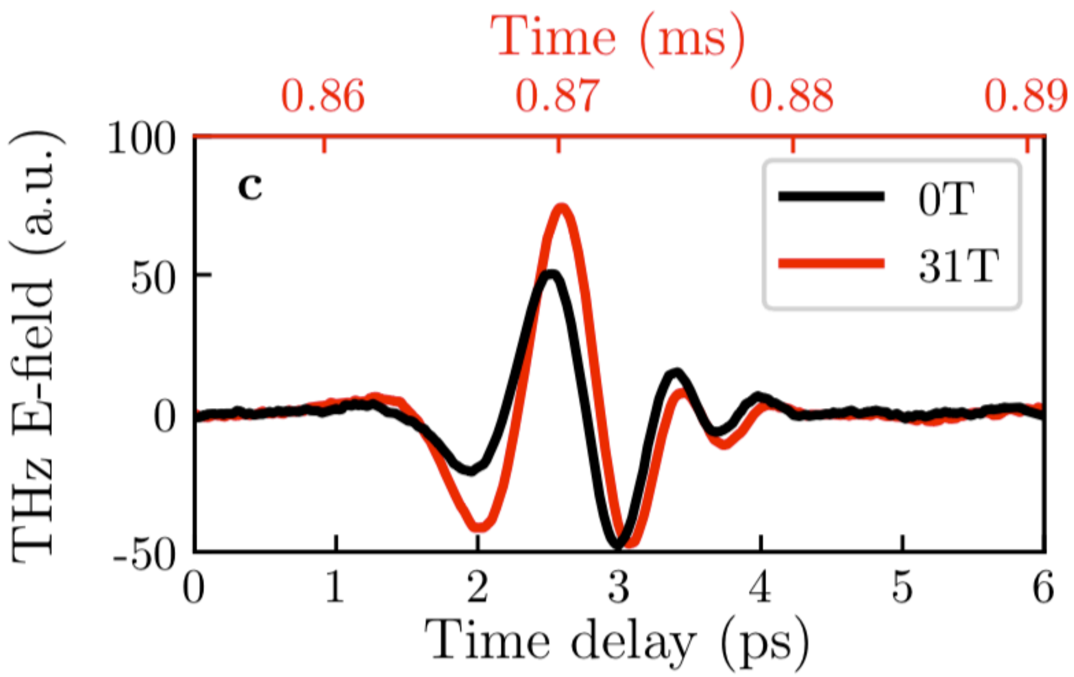
$\boldsymbol{\omega = eB_z/m}$

Optical conductivity →

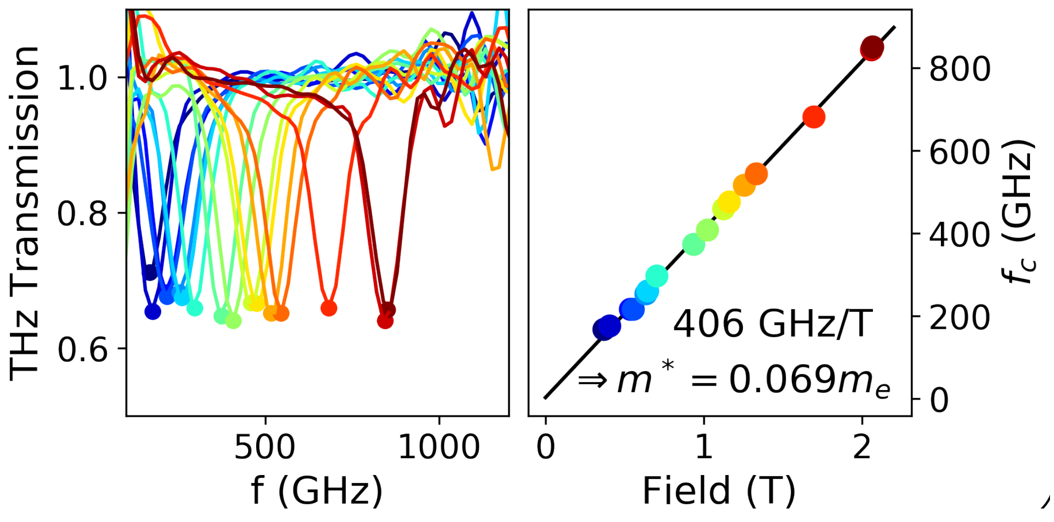
$$\sigma_{l,r} = i\epsilon_0\left(\frac{\omega_p^2}{\omega \pm \omega_c + i\Gamma}\right)$$

Time domain spectroscopy in large pulsed magnetic field



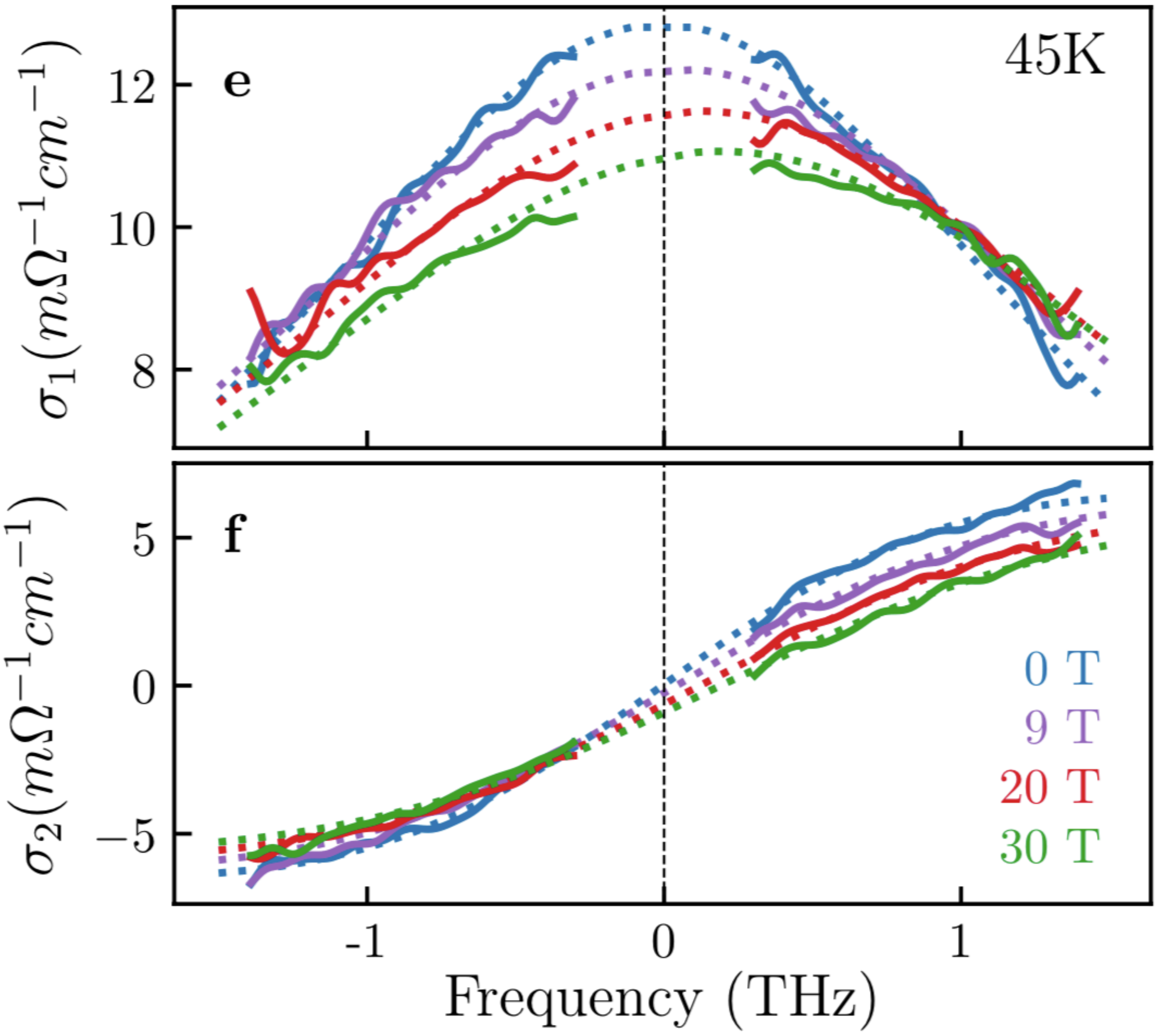


Cyclotron Resonance GaAs Quantum Well



$$\sigma_{R,L} = \sigma_{xx} \pm i\sigma_{xy}$$ <— Overdoped

Time domain spectroscopy in large pulsed magnetic field



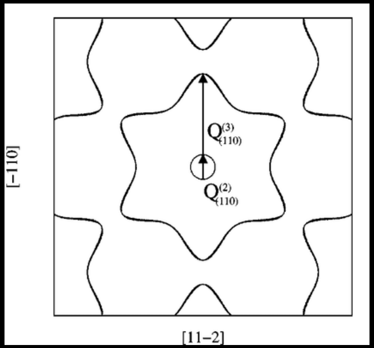
What do we know about cyclotron resonance?

Inverse time to complete a Fermi surface orbit → parameterize as $\omega = eB/m$; but what m?

- Galilean invariant system → mass independent of interactions (Kohn 1961)

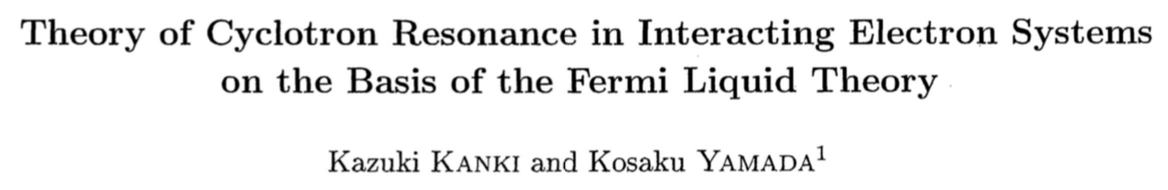
- Non-interacting system →

$$m_{cr} = \frac{\hbar^2}{2\pi} \frac{\partial A}{\partial E}\Big|_{E_F}$$



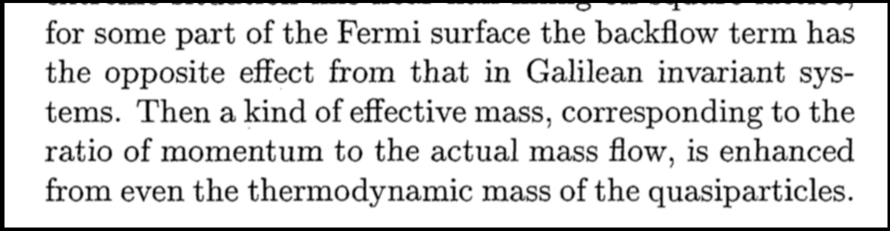
- Effective Galilean invariance; low density systems e.g. 2DEG $\lambda_F >> a$ → $m_{cr} = m_b$

- Deviations from Galilean invariance (disorder, nonparabolicity, Umklapp scattering etc.) cause e-e and e-p interactions to manifest in m_{cr} (Kallin and Halperin, MacDonald and Kallin, Kanki and Yamada) → interactions manifest differently than in other masses!



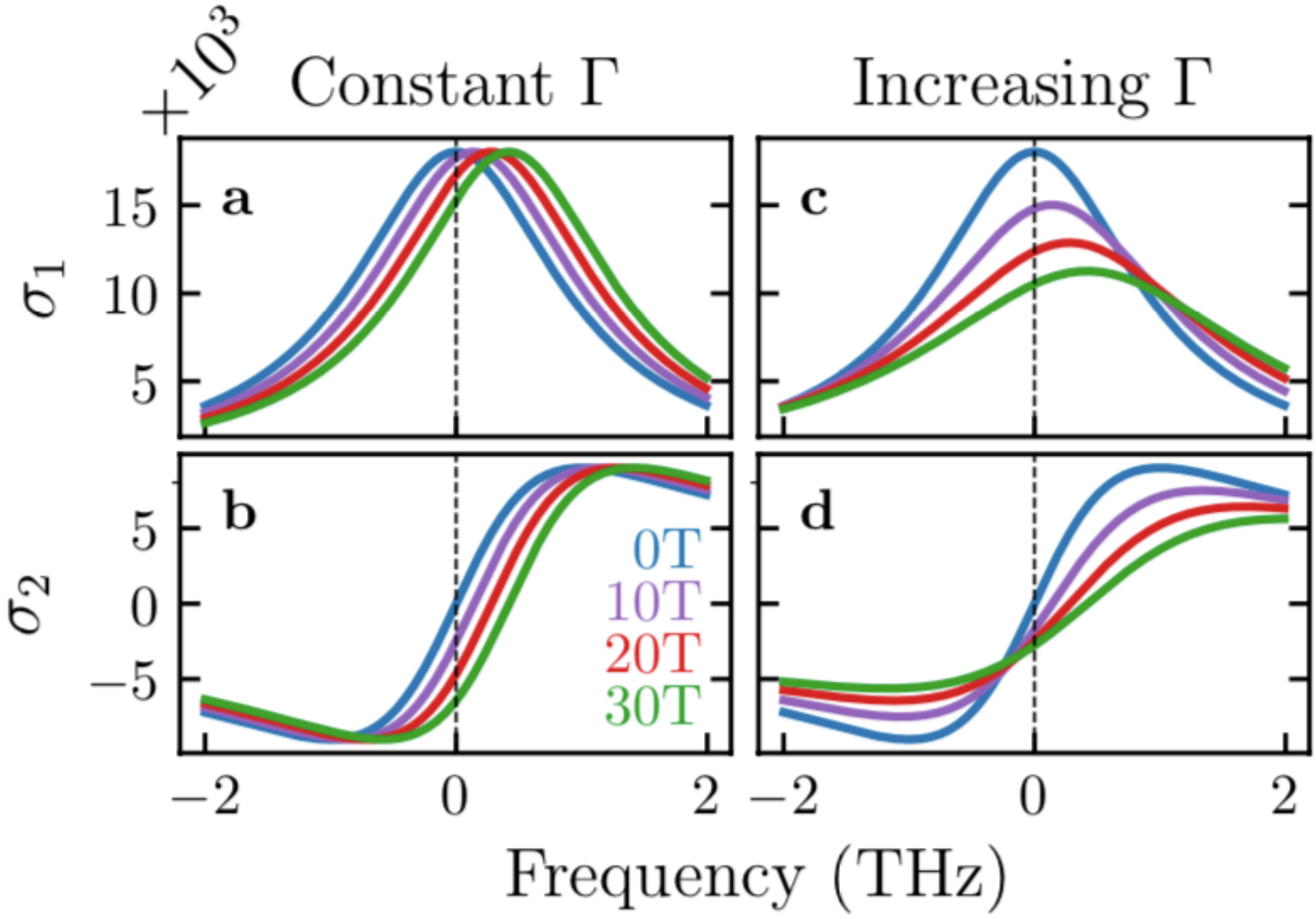
Theory of Cyclotron Resonance in Interacting Electron Systems on the Basis of the Fermi Liquid Theory

Kazuki KANKI and Kosaku YAMADA[1]



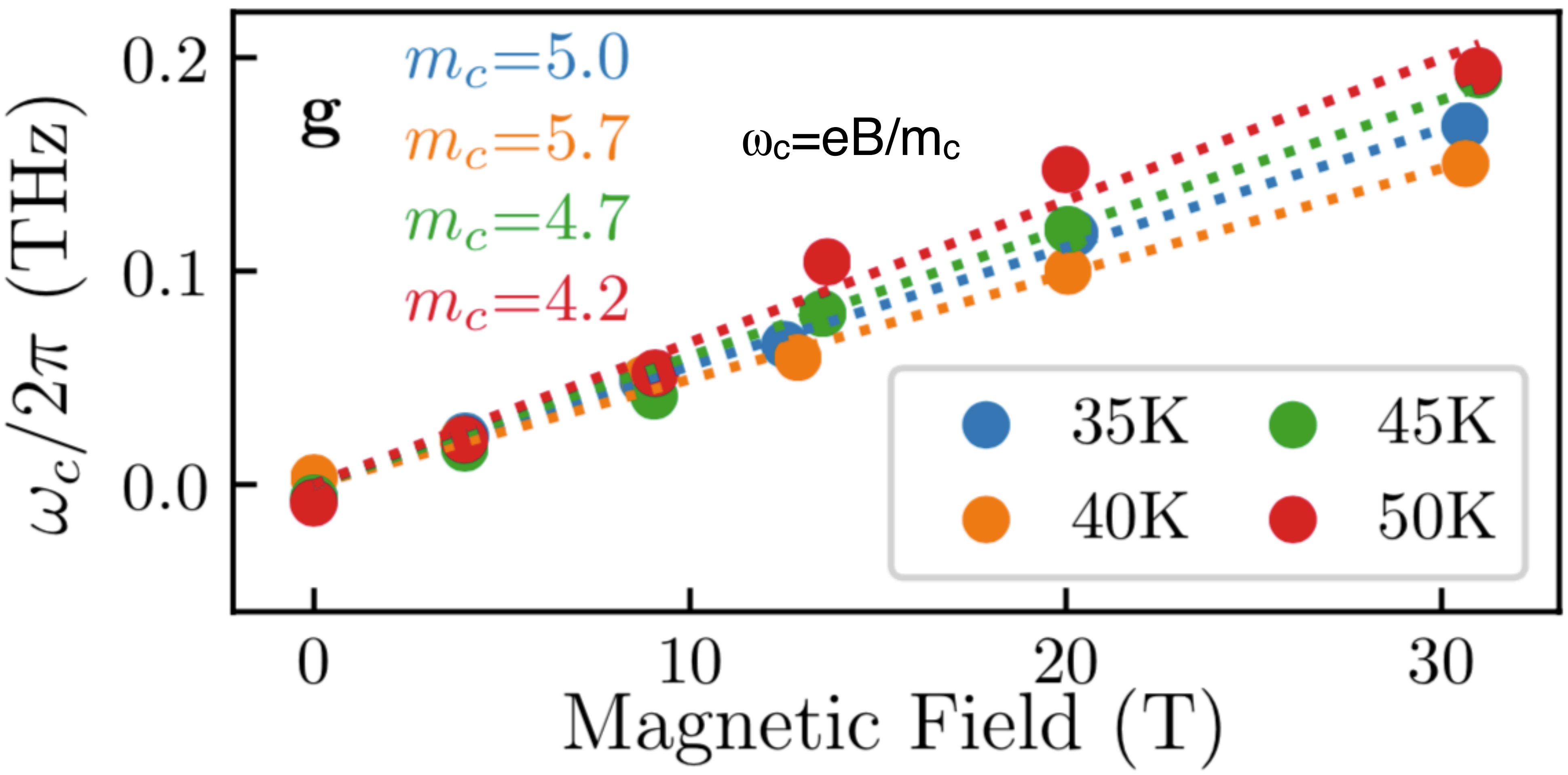
for some part of the Fermi surface the backflow term has the opposite effect from that in Galilean invariant systems. Then a kind of effective mass, corresponding to the ratio of momentum to the actual mass flow, is enhanced from even the thermodynamic mass of the quasiparticles.

Simple models for cyclotron resonance



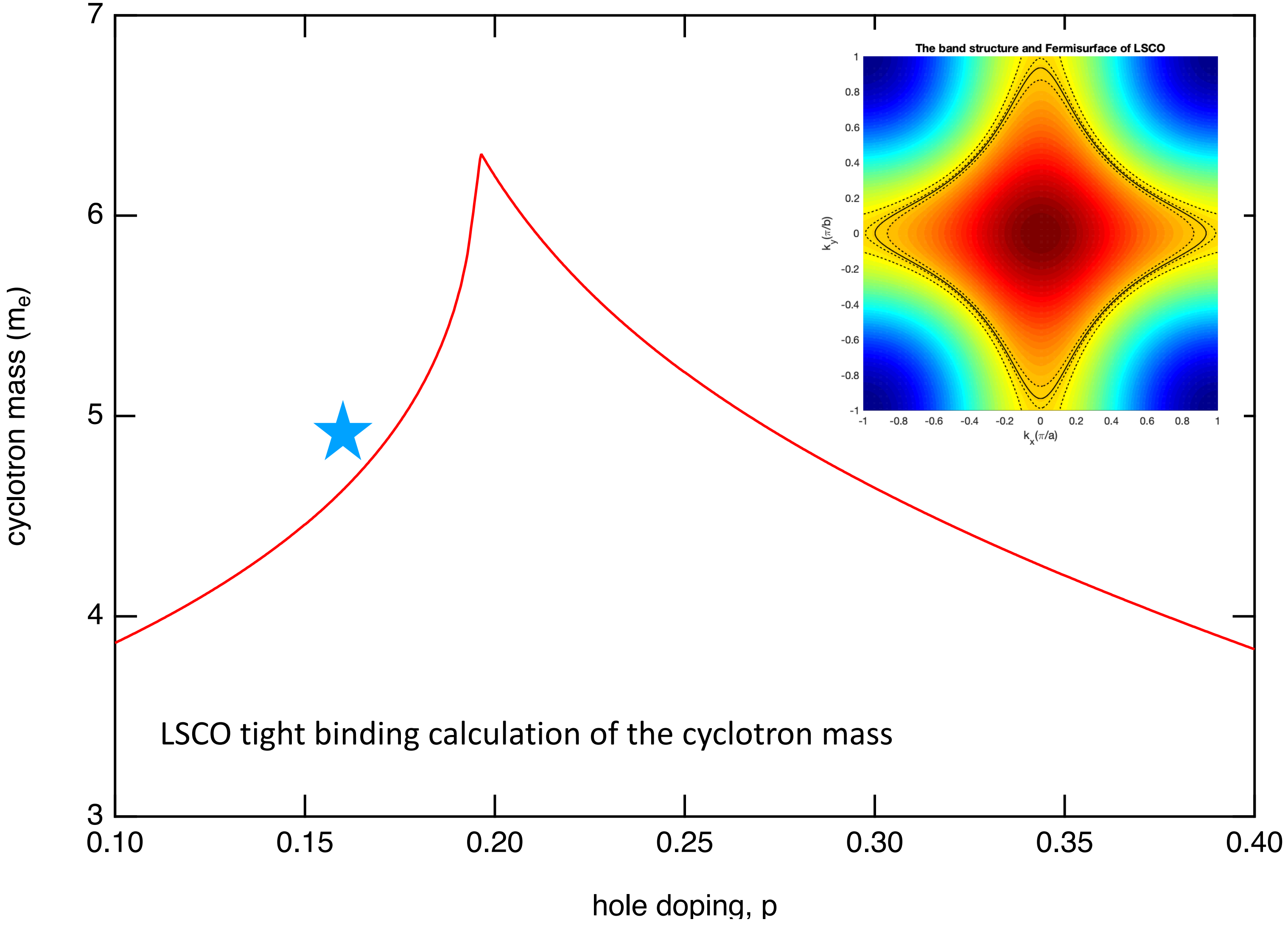
$$\sigma_{l,r} = i\epsilon_0 \left(\frac{\omega_p^2}{\omega \pm \omega_c + i\Gamma} \right)$$

Cyclotron resonance as a function of field



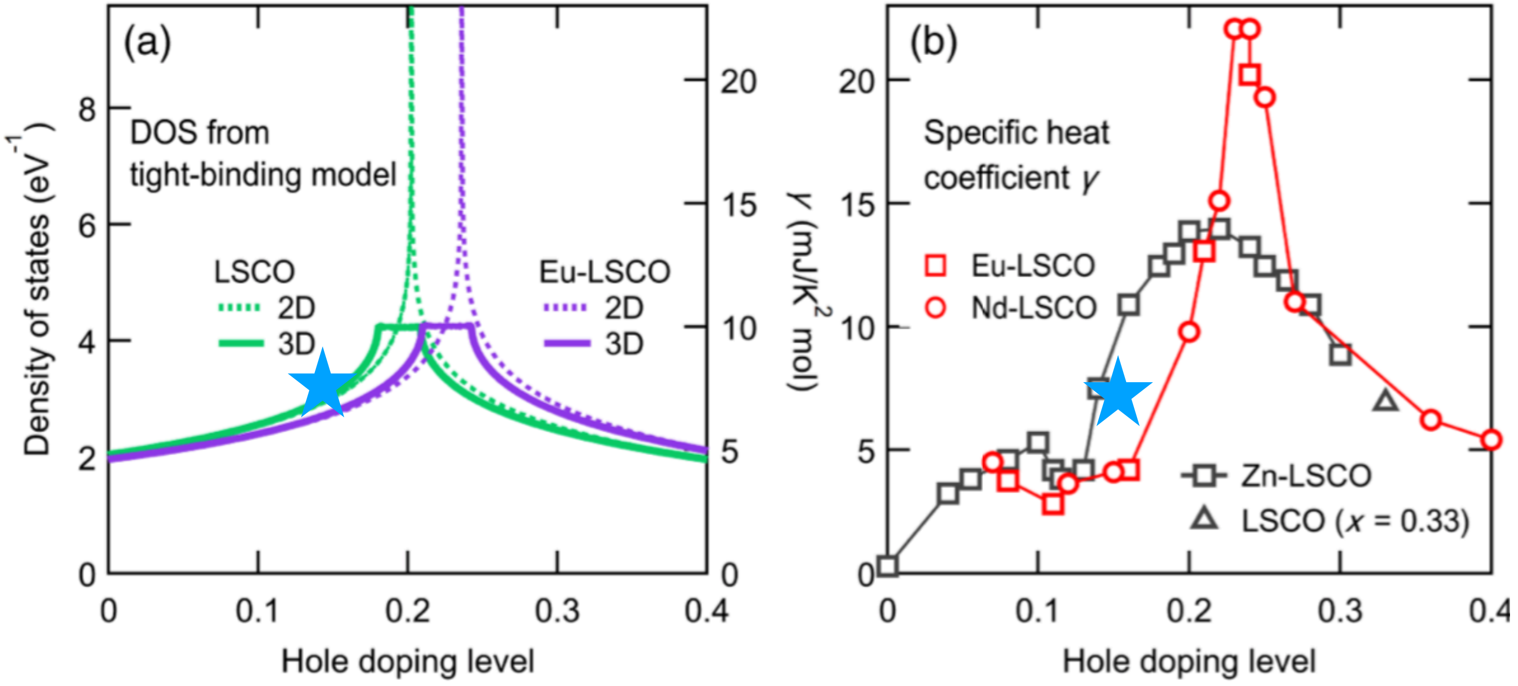
$\omega_c = eB/m_c$

How does cyclotron resonance mass compare with ARPES?



R. McDonald parameterization of Horio et al. PRL 2018 ARPES data

Doping dependence will be interesting



Horio et al. PRL 2018

Conclusions

- Large pulsed magnetic field coupled to time-domain THz spectroscopy → many opportunities for charge and spin systems
- Cyclotron resonance observed despite broad line shape
- Measured mass $m_{cr} \sim 4.9\ m_e$
- Similar to values from ARPES and heat capacity (at this doping)
- No signs of field driven Fermi surface reconstructions