

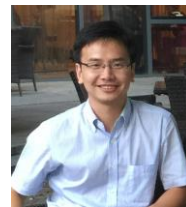
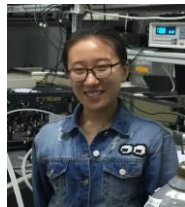
**ICQM**International Center
for Quantum Materials

Photo-induced nonequilibrium response in underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$

Nan-Lin Wang

*International Center for Quantum Materials (ICQM),
School of Physics, Peking University, China*

Collaborators: S. J. Zhang, Z. X. Wang, T. Dong, ... @ PKU



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Samples: H. Xiang, X Yao@SJTU

Outline

➤ Optical spectroscopy

Fourier transform optical spectroscopy

Time resolved Terahertz spectroscopy

.....

“photo-induced transient superconductivity”

➤ Recent results on YBCO $T_c=55K$ and $35K$ by NIR ($1.28\mu m$) and MIR ($15\mu m$) pumping

➤ Conclusions

Physical quantities probed by optical spectroscopy

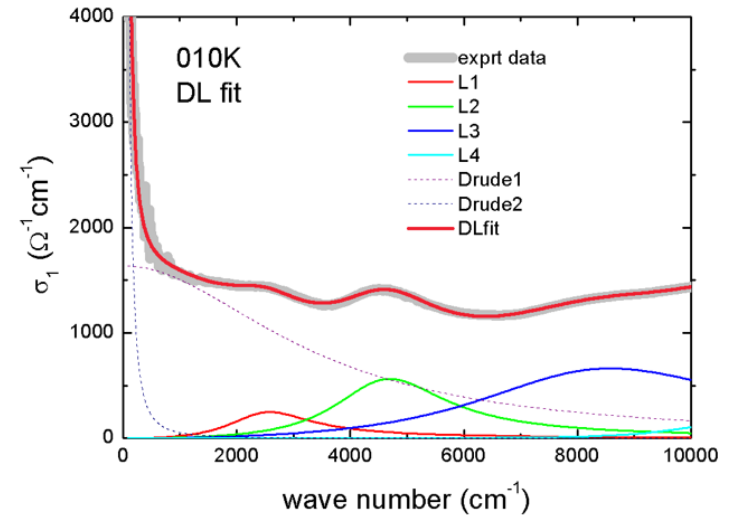
$$\begin{cases} n(\omega), K(\omega) \\ \epsilon_1(\omega), \epsilon_2(\omega) \\ \sigma_1(\omega), \sigma_2(\omega) \end{cases}$$

Dielectric constant $\hat{\epsilon}$		Conductivity $\hat{\sigma}$	Refractive index $\hat{N}$
$\hat{\epsilon}$	$\hat{\epsilon} = \epsilon_1 + i\epsilon_2$	$\epsilon_1 = 1 - \frac{4\pi\sigma_2}{\omega}$ $\epsilon_2 = \frac{4\pi\sigma_1}{\omega}$	$\epsilon_1 = \frac{n^2 - k^2}{\mu_1}$ $\epsilon_2 = \frac{2nk}{\mu_1}$
$\hat{\sigma}$	$\sigma_1 = \frac{\omega\epsilon_2}{4\pi}$ $\sigma_2 = (1 - \epsilon_1)\frac{\omega}{4\pi}$	$\hat{\sigma} = \sigma_1 + i\sigma_2$	$\sigma_1 = \frac{nk\omega}{2\pi\mu_1}$ $\sigma_2 = \left(1 - \frac{n^2 - k^2}{\mu_1}\right)\frac{\omega}{4\pi}$
$\hat{N}$	$n = \left\{ \frac{\mu_1}{2} [\epsilon_1^2 + \epsilon_2^2]^{1/2} + \frac{\epsilon_1\mu_1}{2} \right\}^{1/2}$ $k = \left\{ \frac{\mu_1}{2} [\epsilon_1^2 + \epsilon_2^2]^{1/2} - \frac{\epsilon_1\mu_1}{2} \right\}^{1/2}$	$n = \left\{ \frac{\mu_1}{2} \left[\left(1 - \frac{4\pi\sigma_2}{\omega}\right)^2 + \left(\frac{4\pi\sigma_1}{\omega}\right)^2 \right]^{1/2} + \frac{\mu_1}{2} - \frac{2\pi\mu_1\sigma_2}{\omega} \right\}^{1/2}$ $k = \left\{ \frac{\mu_1}{2} \left[\left(1 - \frac{4\pi\sigma_2}{\omega}\right)^2 + \left(\frac{4\pi\sigma_1}{\omega}\right)^2 \right]^{1/2} - \frac{\mu_1}{2} + \frac{2\pi\mu_1\sigma_2}{\omega} \right\}^{1/2}$	$\hat{N} = n + ik$

Optical constants

$$R(\omega) \xrightarrow{KK} \theta(\omega) \rightarrow \begin{cases} n(\omega), \kappa(\omega) \\ \varepsilon_1(\omega), \varepsilon_2(\omega) \\ \sigma_1(\omega), \sigma_2(\omega) \end{cases}$$

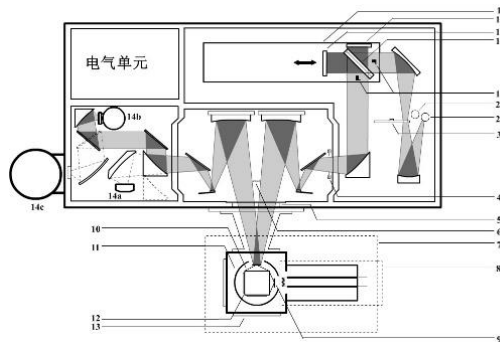
$$r(\omega) = \sqrt{R(\omega)} e^{i\theta} \Rightarrow \theta = \frac{\omega}{\pi} P \int_0^\infty \frac{\ln R(\omega')}{\omega^2 - \omega'^2} d\omega'$$



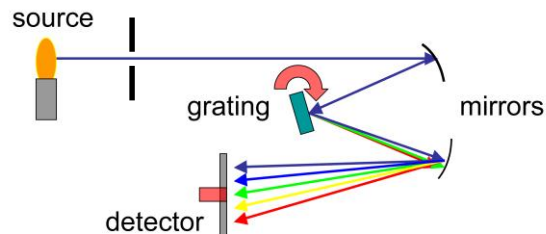
$$1 \text{ eV} = 10^3 \text{ meV} = 8065 \text{ cm}^{-1}$$

$$1 \text{ THz} = 33.3 \text{ cm}^{-1} = 4.2 \text{ meV}$$

✓ FTIR



✓ Grating spectrometer

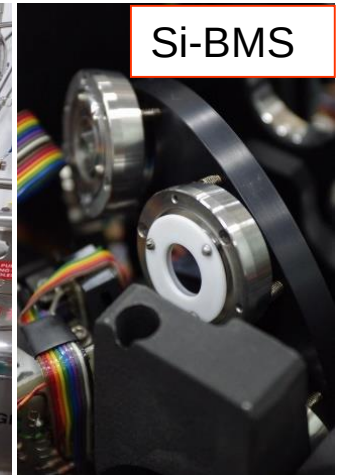
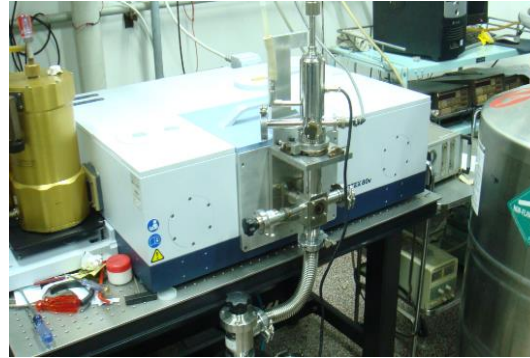


✓ Ellipsometry

✓ Time domain THz spectroscopy

Infrared and optical spectroscopy

- Bruker 80v: 15 - 50000 cm⁻¹
- Bruker 66v/s: 30 - 30000 cm⁻¹
- Bruker 113v: 17 – 8000 cm⁻¹
(thick Si-beam splitter with Si-bolometer)
- Grating type spectrometer:
10000-50000 cm⁻¹

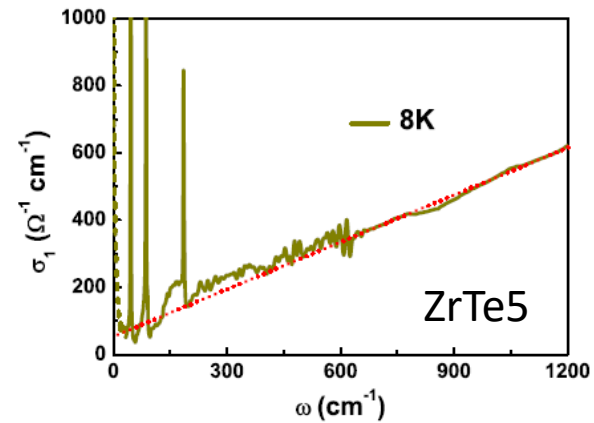


$$R(\omega) \xRightarrow{KK} \theta(\omega) \rightarrow \begin{cases} n(\omega), \kappa(\omega) \\ \varepsilon_1(\omega), \varepsilon_2(\omega) \\ \sigma_1(\omega), \sigma_2(\omega) \end{cases}$$

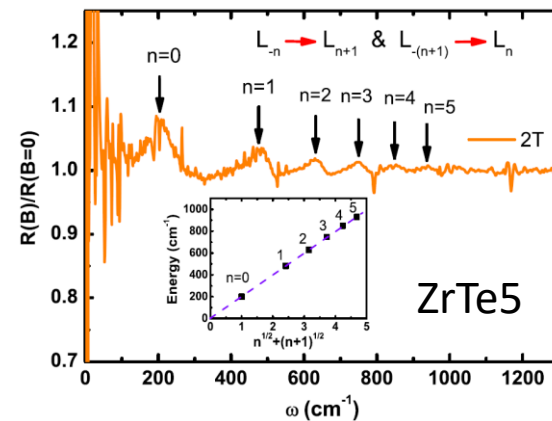
Optical measurement under magnetic field

Bruker 113v spectrometer

(10 Tesla split coils from Cryomagnetic Inc.)



RY Chen...NLW, PRB 2015



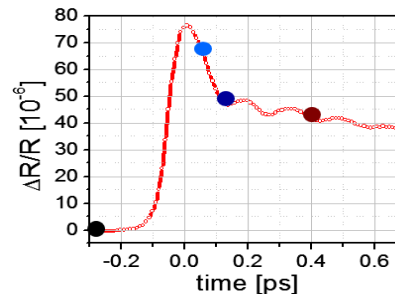
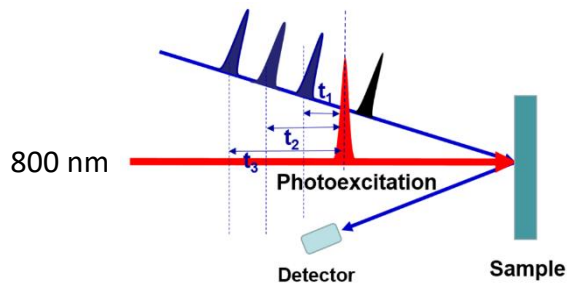
RY Chen...NLW, PRL 2015

Applications of femtosecond lasers in condensed matter

1、Pump-probe

Weak pulse: relaxation dynamics after quasiparticle excitations -- nonequilibrium process

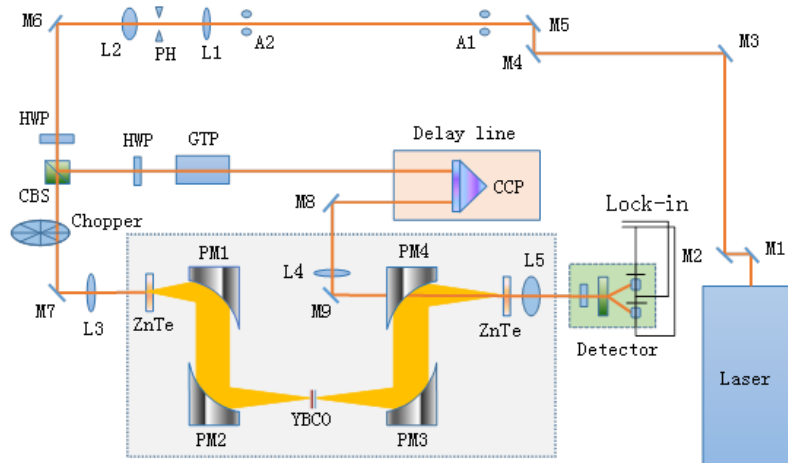
Intense pulse: drive a phase transition/create a new state



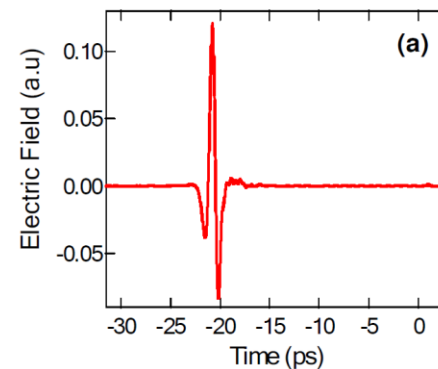
- Time resolved
- Roughly single frequency
- Resolution limited by pulse width

2、Time domain THz spectroscopy

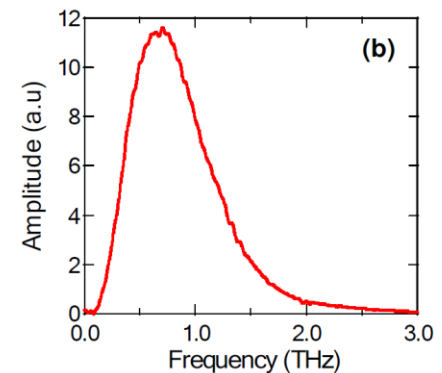
Spectroscopy probe in very far IR in equilibrium state



Time Domain



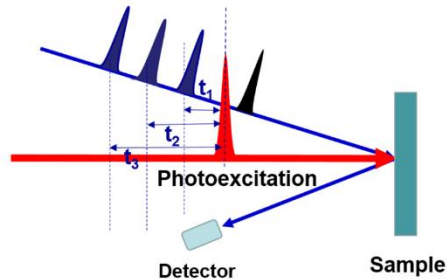
Frequency Domain



$$\tilde{E}(\omega) = A(\omega) \exp[-i\phi(\omega)] = \int E(t) \exp(-i\omega t) dt$$

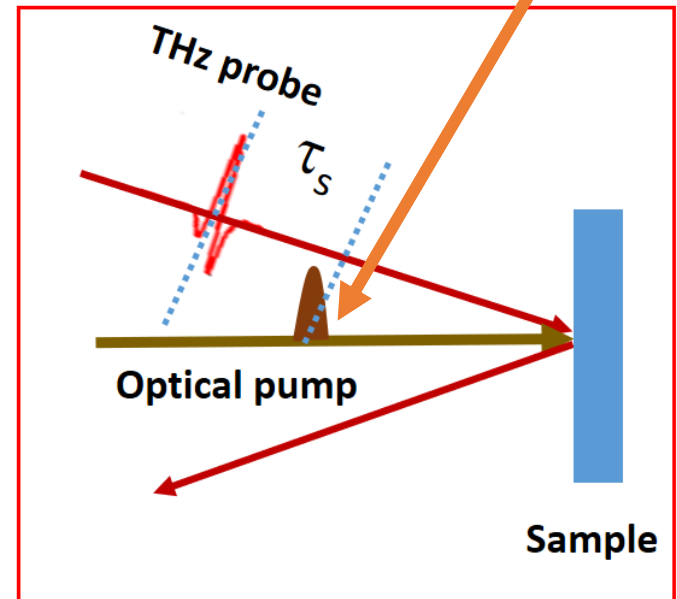
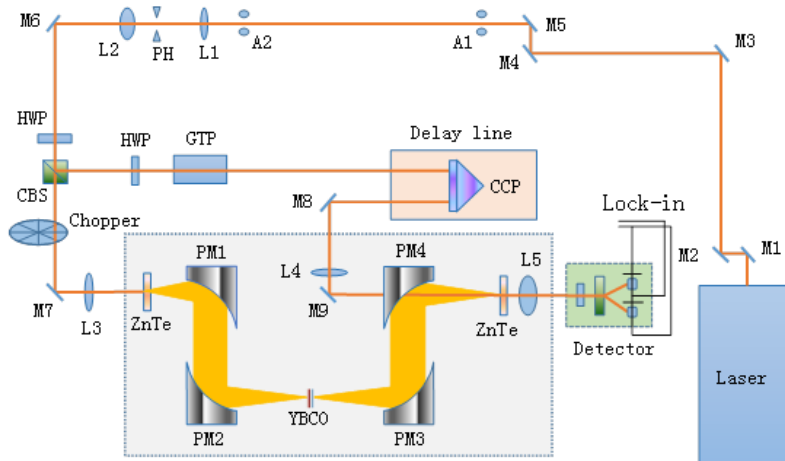
Tunable near- to mid-infrared pump- THz probe

1、 pump-probe



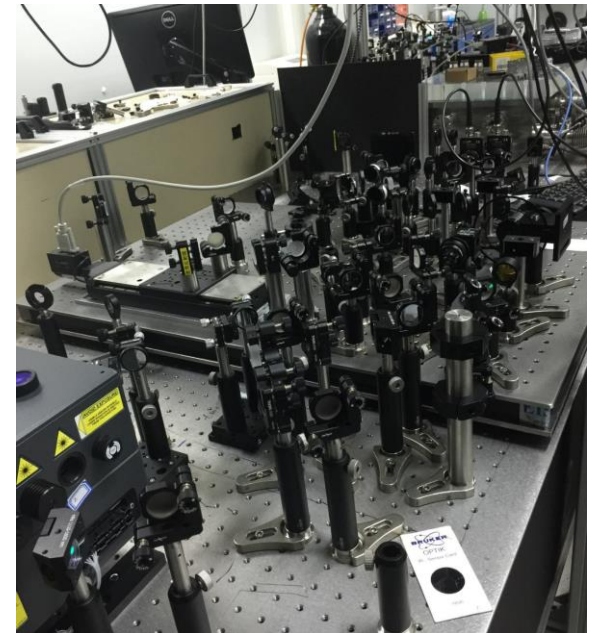
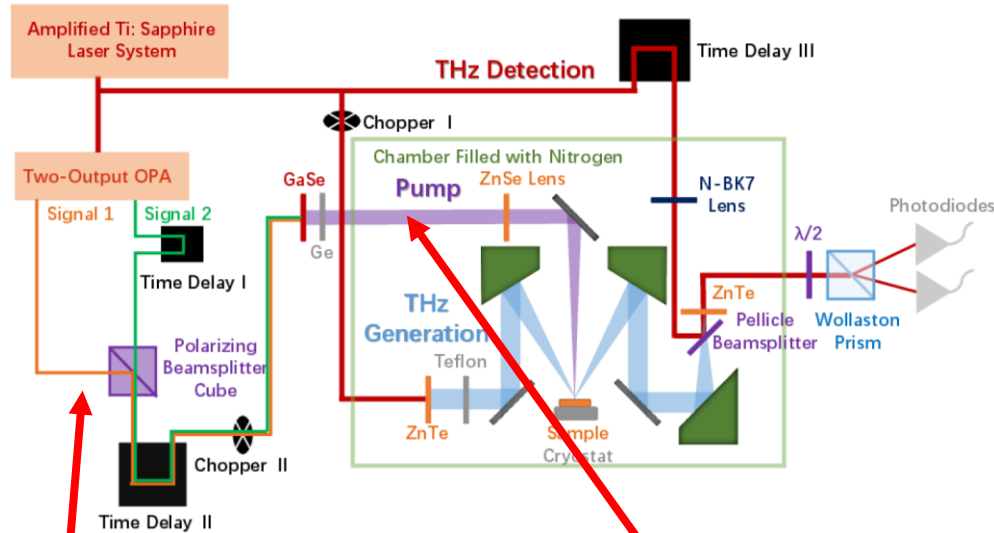
Tune the pump pulse energy from 1.55 eV (800 nm) to mid-infrared, e.g. 80 meV (20 THz or 15 μm).

2、 Time domain THz



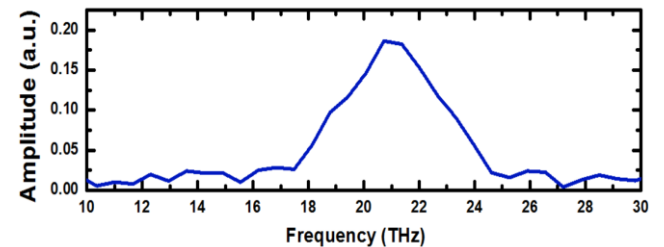
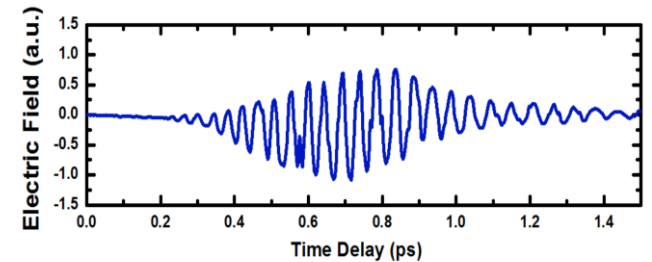
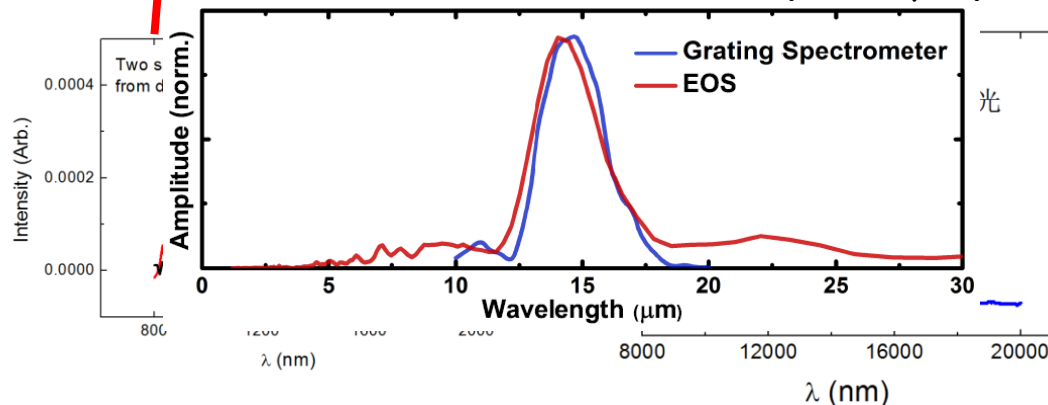
FTIR is unable to do time resolved measurement.

Tunable near- to mid-infrared pump-THz probe



The pump energy is reduced to (15 μm) 20 THz by DFG on GaSe from a dual OPA

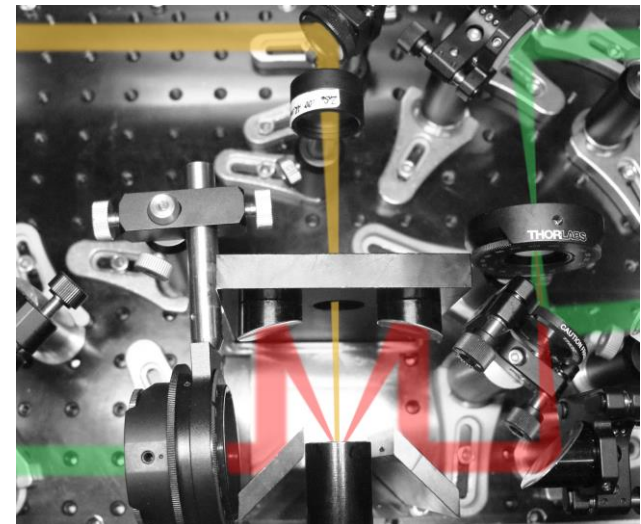
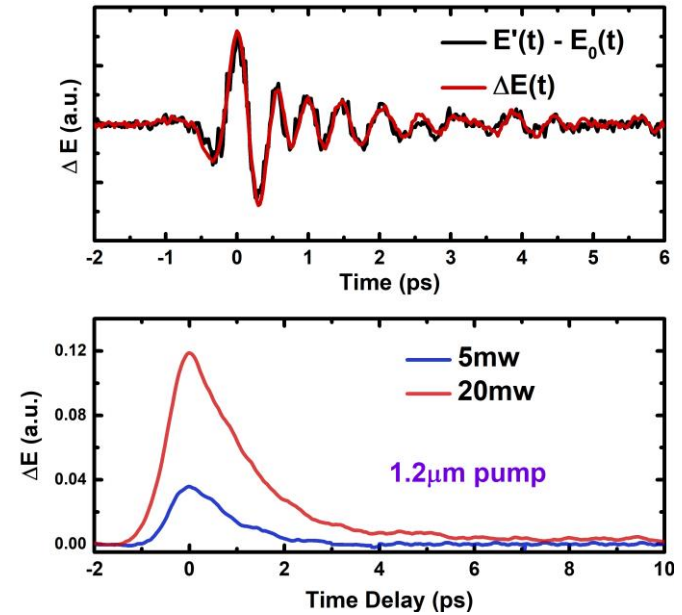
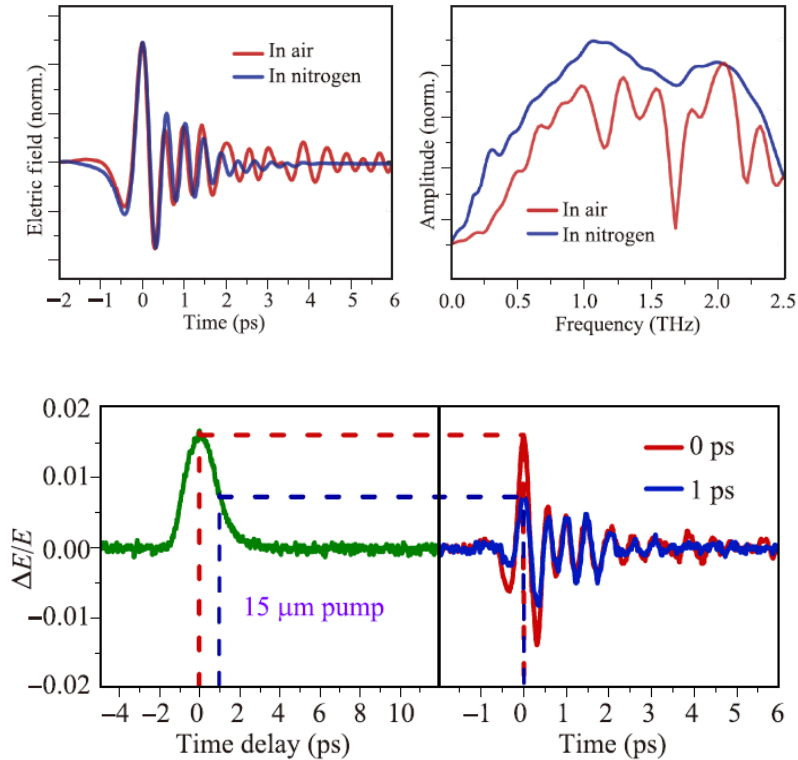
Up to 4 $\mu\text{J}/\text{pulse}$



S. J. Zhang et al. arXiv:1708.01991,
Front. of Phys. 12, 127802 (2017)

20 THz pump beam measured by
grating spectrometer and EOS

Apparatus test measurement on EuSbTe3 crystals

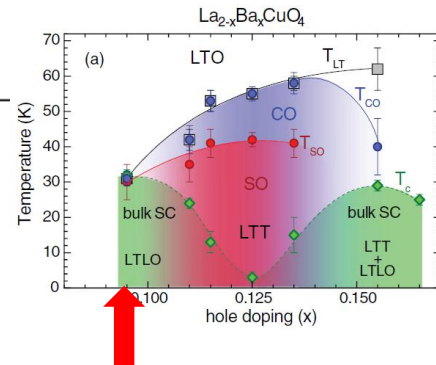


cryostat

S. J. Zhang et al. arXiv:1708.01991,
Front. of Phys. 12, 127802 (2017)

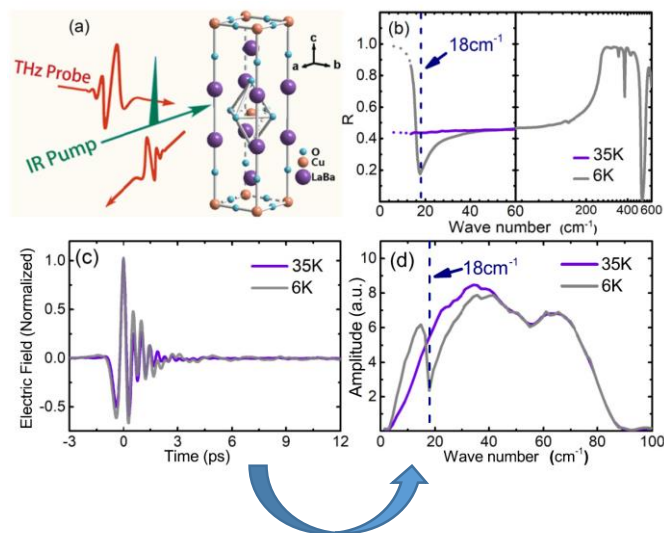
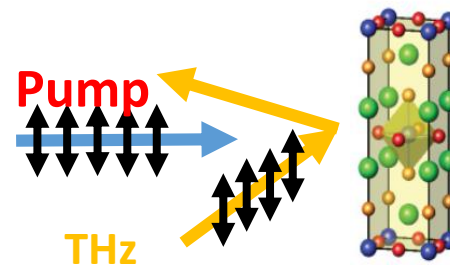
Light-induced new collective modes in the superconductor $\text{La}_{1.905}\text{Ba}_{0.095}\text{CuO}_4$

S. J. Zhang,¹ Z. X. Wang,¹ L. Y. Shi,¹ T. Lin,¹ M. Y. Zhang,¹ G. D. Gu,² T. Dong,^{1,*} and N. L. Wang^{1,3,†}

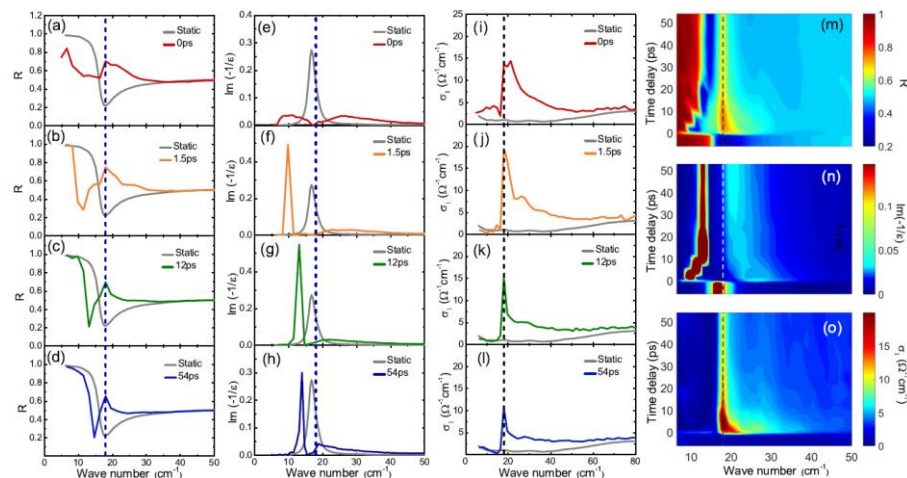


hole-doped $T_c=32\text{K}$

1.2 μm pump // c-axis,
Terahertz E//c-axis



FTIR



Two longitudinal plasmon modes

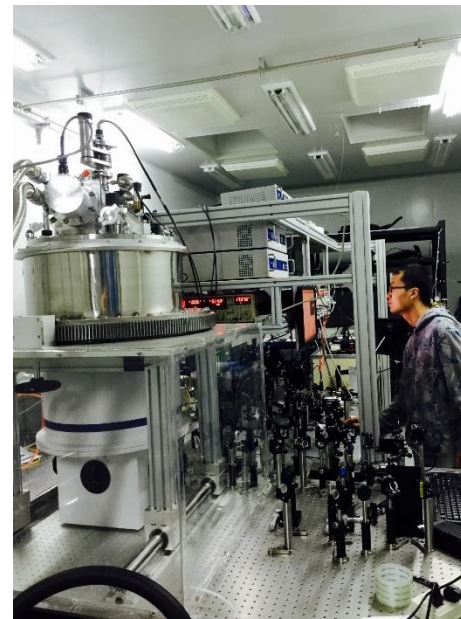
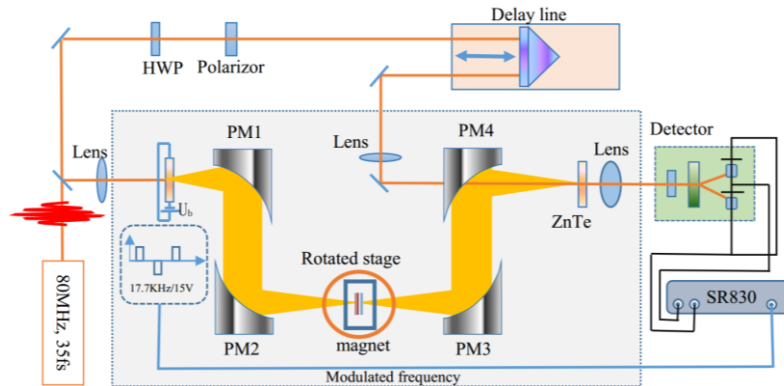
Transverse plasmon mode

We have been working and developing ultrafast time-resolved optical measurement systems

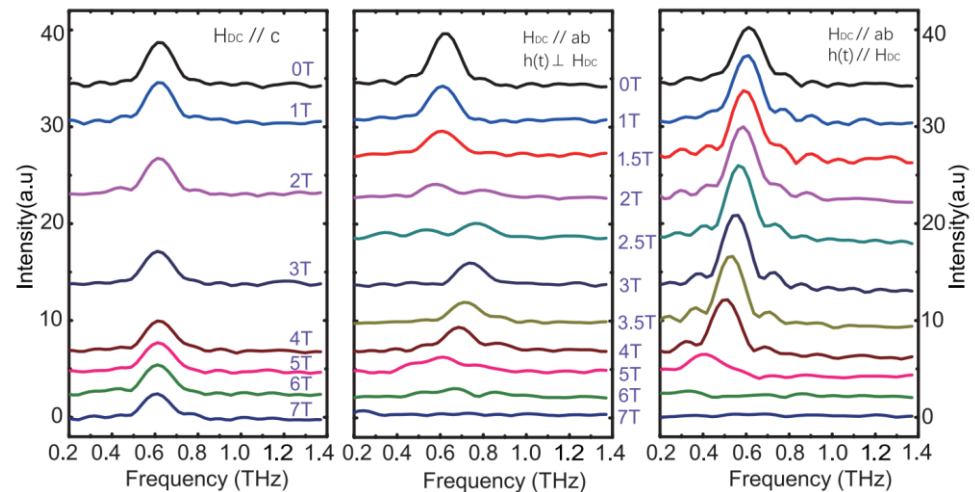
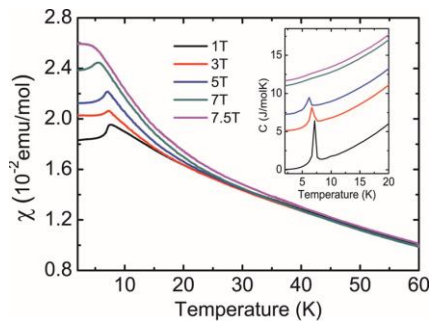
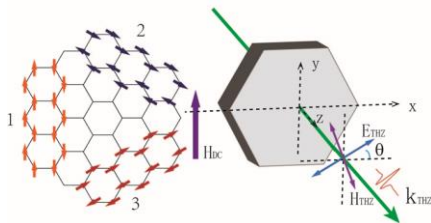


- Optical pump-THz probe
- **MIR-to-NIR pump-THz probe**
- THz pump-THz probe
- Time-domain THz spectroscopy under magnetic field
- Optical pump SHG probe
- Optical pump dc transport
-

Time domain THz spectroscopy under magnetic field



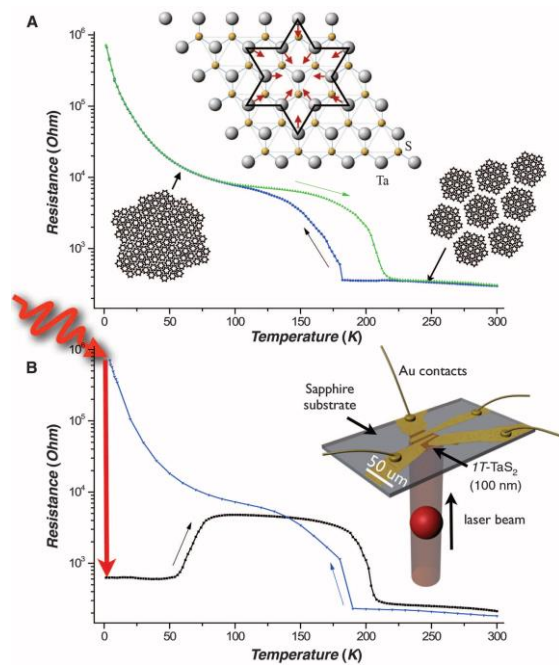
For example: $\alpha\text{-RuCl}_3$



L. Y. Shi...NLW, PRB 98, 094414 (2018)

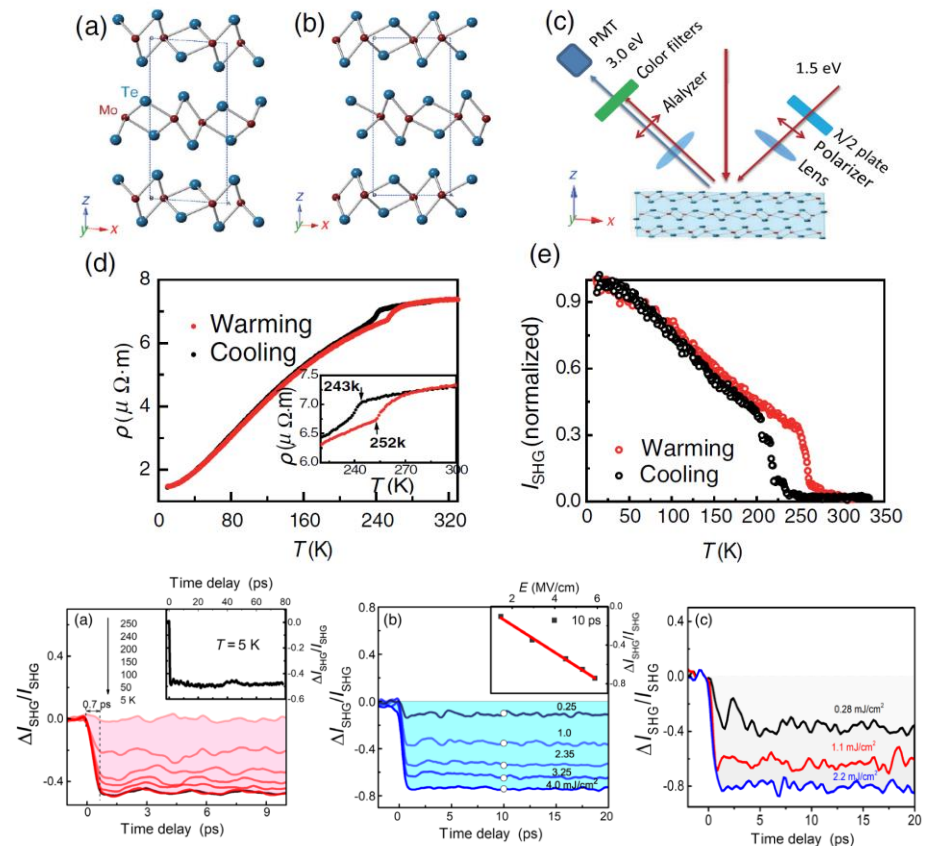
Intense ultrashort laser pulse can induce a phase transition or create new stable/metastable states in an ultrafast fashion

1T-TaS₂



L. Stojchevska et al. Science
344, 177 (2014)

T_d-MoTe₂ to T'-MoTe₂



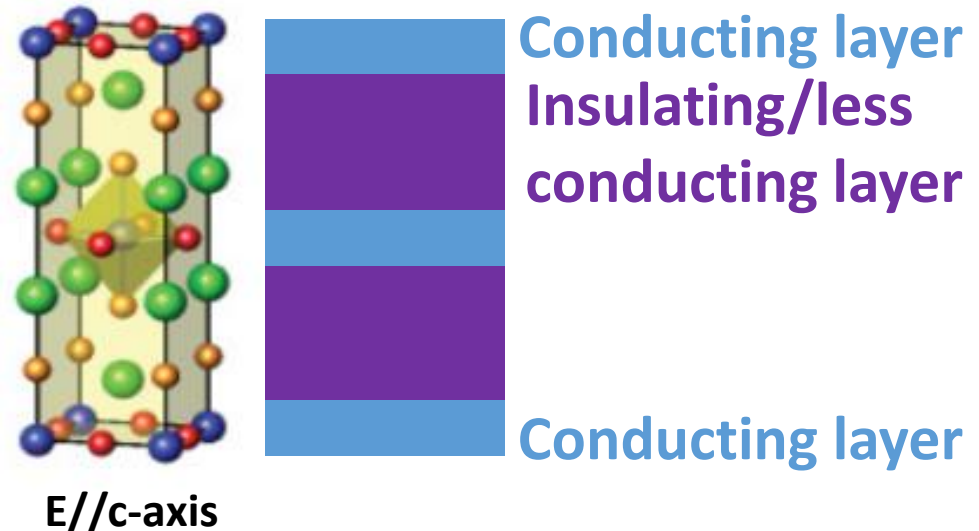
MY Zhang...NLW, PRX (2019)

Light-induced high temperature superconductivity

initiated by Andrea Cavalleri and collaborators

Cuprates:

- Insulating response along the c-axis above T_c
- Sharp **Josephson plasma edge (JPE)** develops below T_c

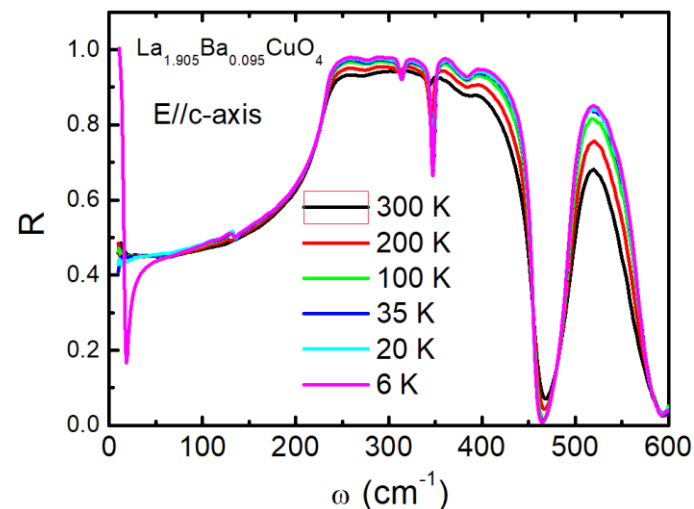


FTIR measurement

on **La_{1.905}Ba_{0.095}CuO₄**

$$\lambda_{\perp}^2 = \hbar c^2 / 8 \pi d e J_c$$

$$J_c = [\pi \Delta(T) / 2 e R_n] \tanh[\Delta(T) / 2 k T]$$



Electrodynamics of a superconductor

$$T=0 \text{ K} \quad \sigma = \frac{1}{8} \omega_{ps}^2 \delta(\omega) + i \omega_{ps}^2 / 4\pi\omega$$

$$\omega_{ps}^2 = 8 \int_0^{\omega_c} d\omega \left[\sigma_1(\omega, T > T_c) - \sigma_1(\omega, T < T_c) \right]$$

$$\lambda_L = c / \omega_{ps}$$

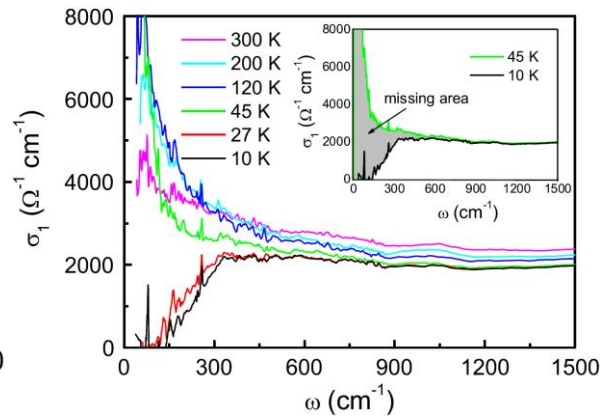
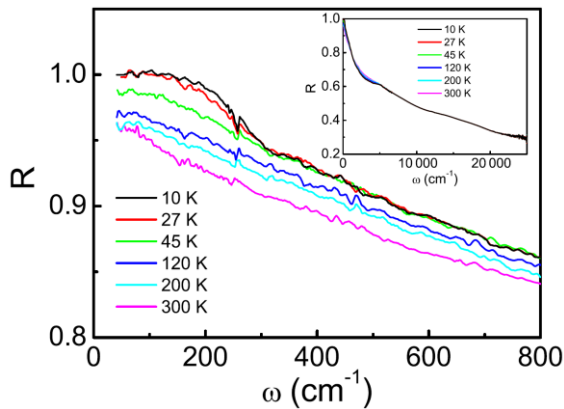
$$\omega \rightarrow 0$$

$$\frac{1}{\lambda_L^2} = \frac{\omega_{ps}^2}{c^2} = \frac{4\pi}{c^2} \omega \sigma_2(\omega)$$

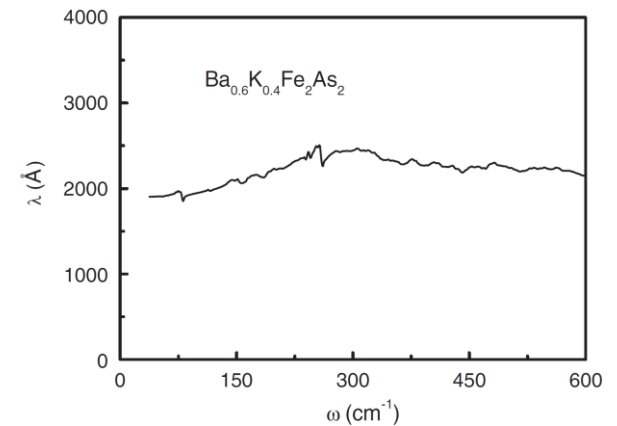
$$\lambda(\omega) = c / \omega_{ps} = c / \sqrt{4\pi\omega\sigma_2(\omega)}$$

For example: $\text{Ba}_{1.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$ $T_c=37 \text{ K}$

G. Li...NLW, PRL 101, 107004 (2008)

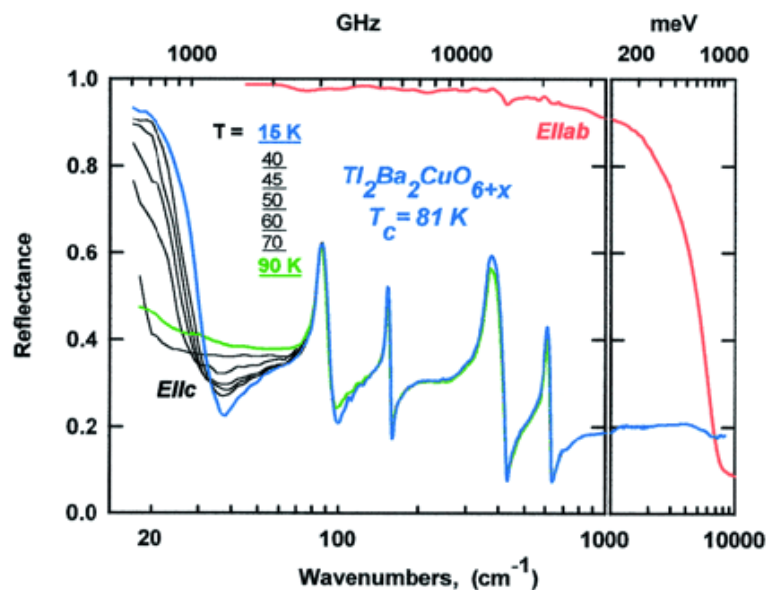


$$\lambda_L = 2080 \text{ \AA}$$



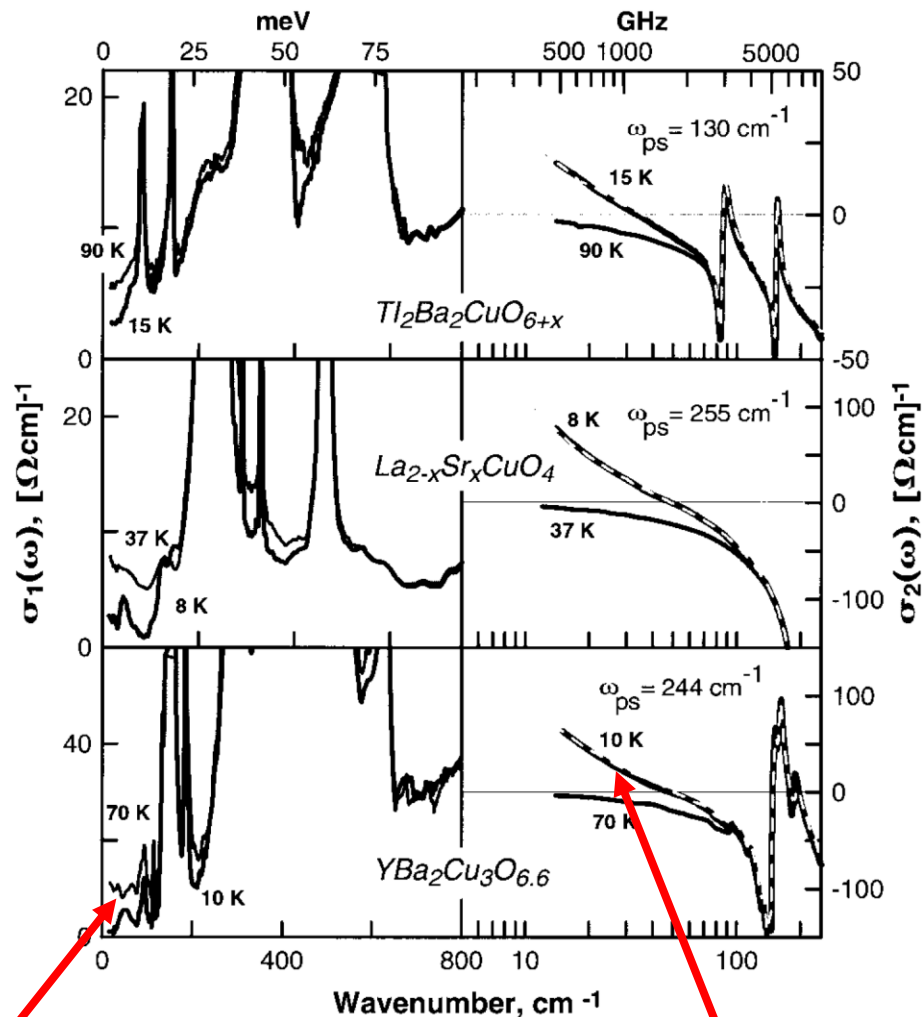
$$\lambda_L = 1950 \text{ \AA}$$

D. N. Basov et al., Science 283, 49 (1999)



$T=0 K$

$$\sigma = \frac{1}{8} \omega_{ps}^2 \delta(\omega) + i \omega_{ps}^2 / 4\pi\omega$$



$$\omega_{ps}^2 = 8 \int_0^{\omega_c} d\omega [\sigma_1(\omega, T > T_c) - \sigma_1(\omega, T < T_c)]$$

$$\omega \rightarrow 0$$

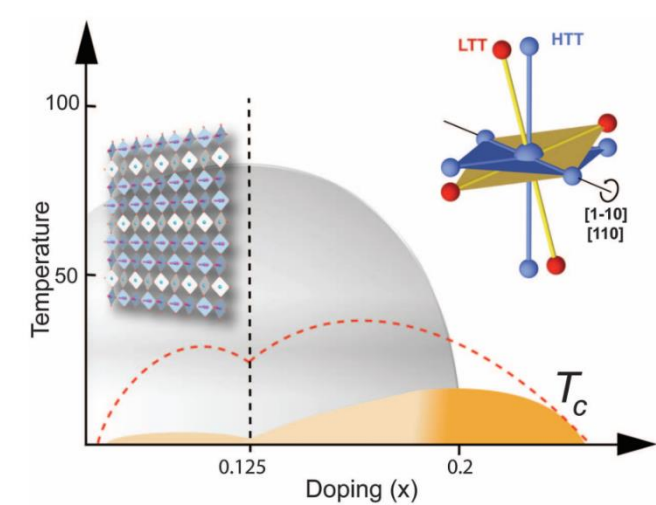
$$\frac{1}{\lambda_L^2} = \frac{\omega_{ps}^2}{c^2} = \frac{4\pi}{c^2} \omega \sigma_2(\omega)$$

Light-Induced Superconductivity in a Stripe-Ordered Cuprate

Science 331, 189 (2011)

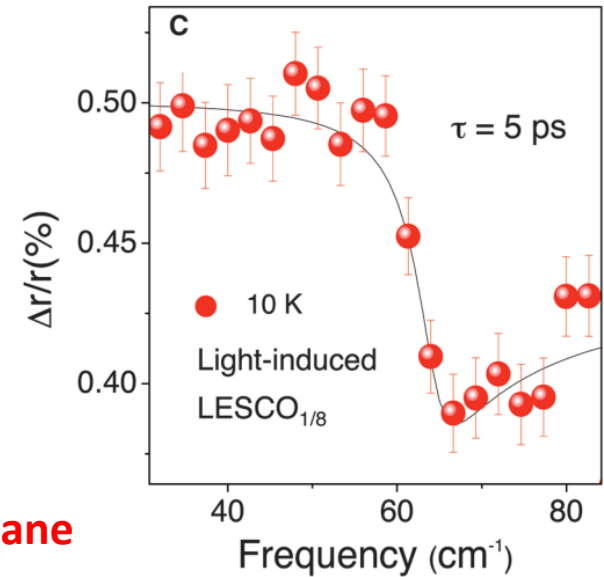
D. Fausti,^{1,2*}† R. I. Tobey,²§ N. Dean,^{1,2} S. Kaiser,¹ A. Dienst,² M. C. Hoffmann,¹ S. Pyon,³ T. Takayama,³ H. Takagi,^{3,4} A. Cavalleri^{1,2*}

Non-superconducting at 1.675 μm CuO_2 doping at 1/8



$E//ab$
Pump with $16\mu\text{m}$
MIR pulses

Resonant with in-plane
Cu-O phonons



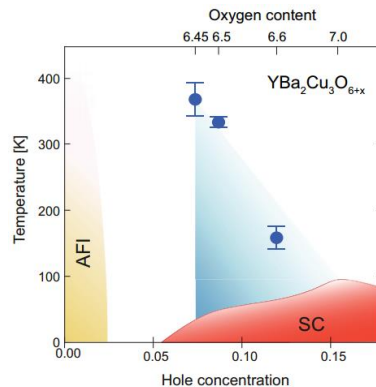
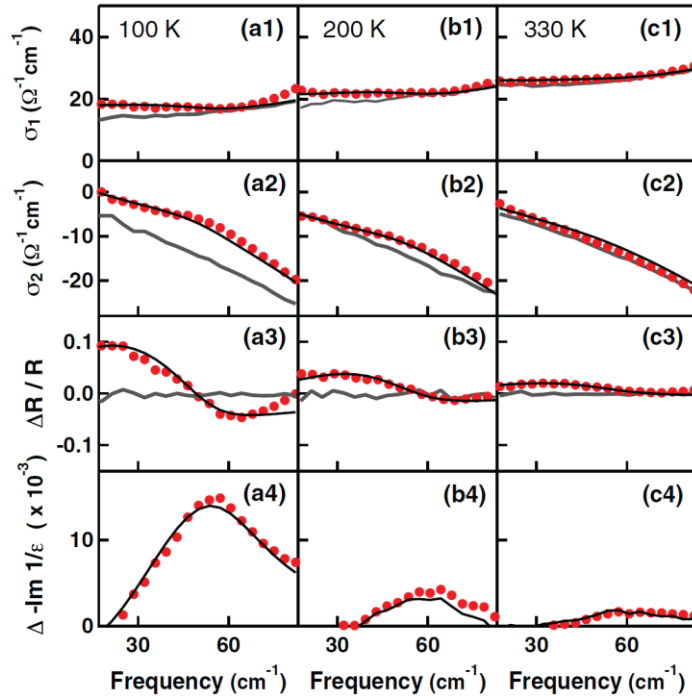
$$r = E_{\text{refl}}/E_{\text{inc}}$$

The small photo-induced change is attributed to the penetration depth mismatch (difference over 40 times) !

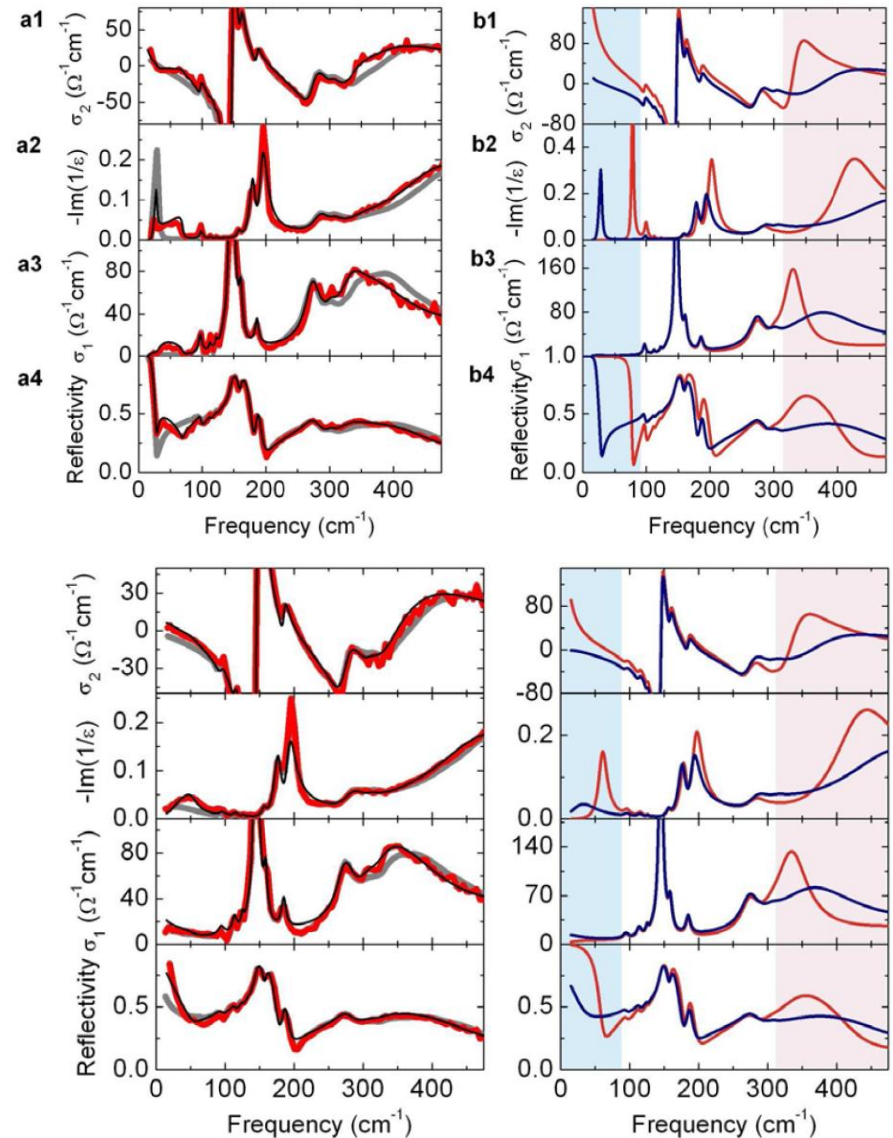
Light-induced transient high temperature superconductivity in YBCO

S. Kaiser et al. Phys. Rev. B 89, 184516 (2014); W. Hu et al. Nature materials 13, 705 (2014).

$E//c$, YBCO 6.5, $\tau=0.8$ ps



$E//c$, YBCO 6.5



To obtain the pump-induced optical constants, one has to have

- **capacity to measure static optical constants**
 - **capacity of obtaining mid-infrared pump beam by DFG (for mode-selective excitations)**
 - **capacity of measuring THz spectra in time domain with different time delay after pumping**
-
- **Complex data processing by considering the mismatch of penetration depths between pump and THz probe beams**

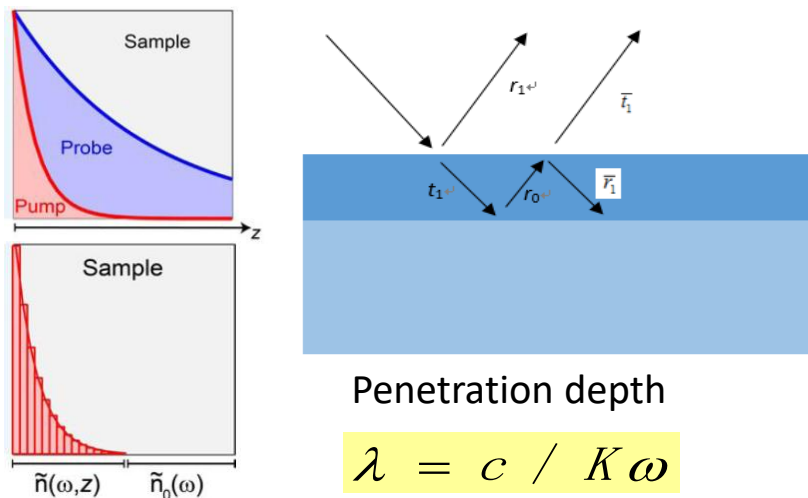
Measured quantities

$$\begin{array}{ccc}
 \left[\begin{array}{l} E_R(t) \quad \text{static} \\ \Delta E_R(t, \tau) = E_R^{\text{pumped}}(t, \tau) - E_R^{\text{unpumped}}(t, \tau) \end{array} \right. & \xrightarrow{\text{FFT}} & \left[\begin{array}{l} \tilde{E}_R(\omega) \\ \Delta \tilde{E}_R(\omega) = \tilde{E}_R^{\text{pumped}}(\omega) - \tilde{E}_R^{\text{unpumped}}(\omega) \end{array} \right. \\
 \text{Pump-induced change} & & \text{Frequency domain}
 \end{array}$$

The Fresnel reflection coefficient of photoexcited area $\tilde{r}'(\omega, \tau)$ can be obtained by following equation:

$$\frac{\Delta \tilde{E}_R(\omega, \tau)}{\tilde{E}_R(\omega)} = \frac{\tilde{r}'(\omega, \tau) - \tilde{r}(\omega)}{\tilde{r}(\omega)} \quad \text{Then,} \quad \frac{\Delta R}{R}(\omega, \tau) = (|\tilde{r}'(\omega, \tau)|^2 - |\tilde{r}(\omega)|^2) / |\tilde{r}(\omega)|^2$$

Consider penetration depth mismatch



$$\hat{r}_{123} = \frac{\hat{r}_{12} + \frac{\hat{r}_{13} - \hat{r}_{12}}{1 - \hat{r}_{13}\hat{r}_{12}} \exp\{2i\delta\}}{1 + \hat{r}_{12} \frac{\hat{r}_{13} - \hat{r}_{12}}{1 - \hat{r}_{13}\hat{r}_{12}} \exp\{2i\delta\}}$$

$$\delta = 2\pi d (n_1 + ik_1) / \lambda$$

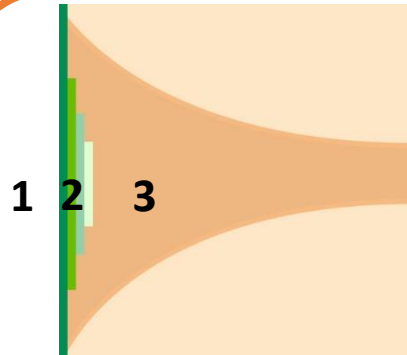
r_{123} is the measured complex reflection coefficient, r_{13} is the static reflection coefficient, r_{12} is the reflection coefficient in the pumped region (i.e. $\tilde{r}'$).

$$\Rightarrow \tilde{n}'(\omega, \tau) = n'(\omega, \tau) + i\kappa'(\omega, \tau) \Rightarrow R$$

Multilayer Model Used for Transient Optical Constants Calculation

Multilayer model:

- Pumped volume → A stack of thin layers
- The refractive index, $\tilde{n}$, of each layer evolves along the direction of propagation.



$$\begin{aligned}
 &\text{Each layer} \quad \mathbf{M}(z) = \begin{bmatrix} \cos(k_0 \tilde{n} z \cos \theta_0) & -\frac{i}{p} \sin(k_0 \tilde{n} z \cos \theta_0) \\ -i p \sin(k_0 \tilde{n} z \cos \theta_0) & \cos(k_0 \tilde{n} z \cos \theta_0) \end{bmatrix} \\
 &\text{Total pumped region} \quad \mathbf{M}(z_n) = \mathbf{M}_1(z_1) \mathbf{M}_2(z_2 - z_1) \cdots \mathbf{M}_n(z_n - z_{n-1}) \\
 &\text{Evolving form of } \tilde{n} \quad \tilde{n}(z) = \tilde{n}_0 + \Delta \tilde{n} \cdot e^{-z/l_p} \\
 &\mathbf{M} = \prod_{j=1}^N \mathbf{M}_j = \begin{bmatrix} 1 & -i k_0 \int dz \\ -i k_0 \int \tilde{n}(z)^2 \cos^2 \theta(z) dz & 1 \end{bmatrix}
 \end{aligned}$$

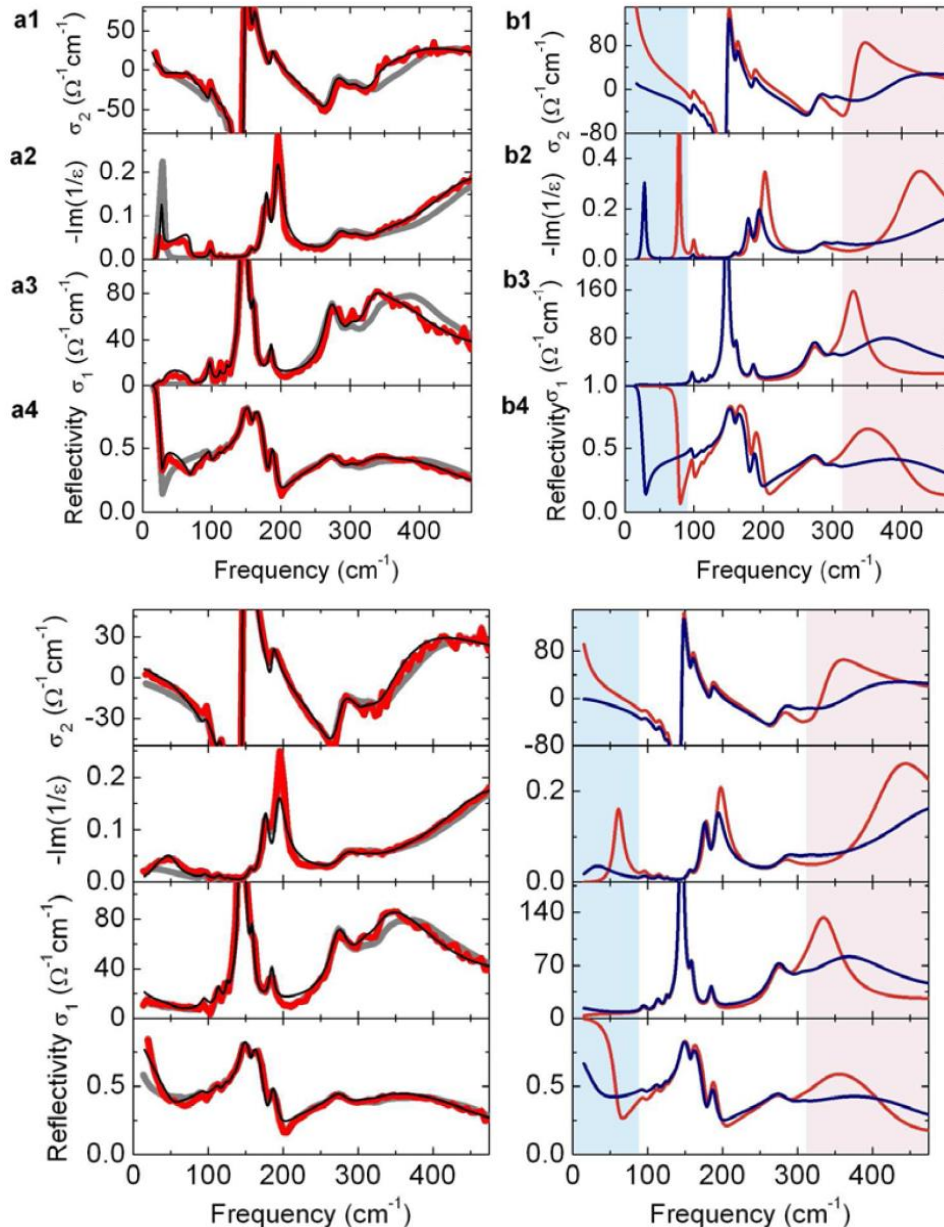
$$\tilde{r} = \frac{(m_{11} + m_{12} \tilde{p}_3) \tilde{p}_1 - (m_{21} + m_{22} \tilde{p}_3)}{(m_{11} + m_{12} \tilde{p}_3) \tilde{p}_1 + (m_{21} + m_{22} \tilde{p}_3)} \quad \tilde{p}_1 = \cos \theta_0 \text{ and } \tilde{p}_3 = \frac{1 - \tilde{r}_{13}}{1 + \tilde{r}_{13}} \text{ for TE}$$

$$\frac{1 - e^{-2}}{2} i k_0 l_p \cdot \Delta \tilde{n}^2 + 2(1 - e^{-1}) i k_0 l_p \tilde{n}_0 \cdot \Delta \tilde{n} + (1 - i k_0 l_p \tilde{p}_3) \tilde{p}_1 \frac{1 - \tilde{r}}{1 + \tilde{r}} + (i k_0 l_p \tilde{n}_0^2 - i k_0 l_p \sin^2 \theta_0 - \tilde{p}_3) = 0$$



$$\tilde{n}' = \tilde{n}_0 + \Delta \tilde{n}$$

Light-induced high temperature superconductivity in YBCO



W. Hu et al., Nature Materials (2014)

The pump-induced change is small even after considering the mismatch between penetration depths.

Below T_c , the low energy Josephson plasma edge shows a small knee;
Above T_c , $R(\omega)$ shows a small upturn, $\sigma_2(\omega)$ shows a $1/\omega$ -like enhancement.

A non-uniform excitation model is used:

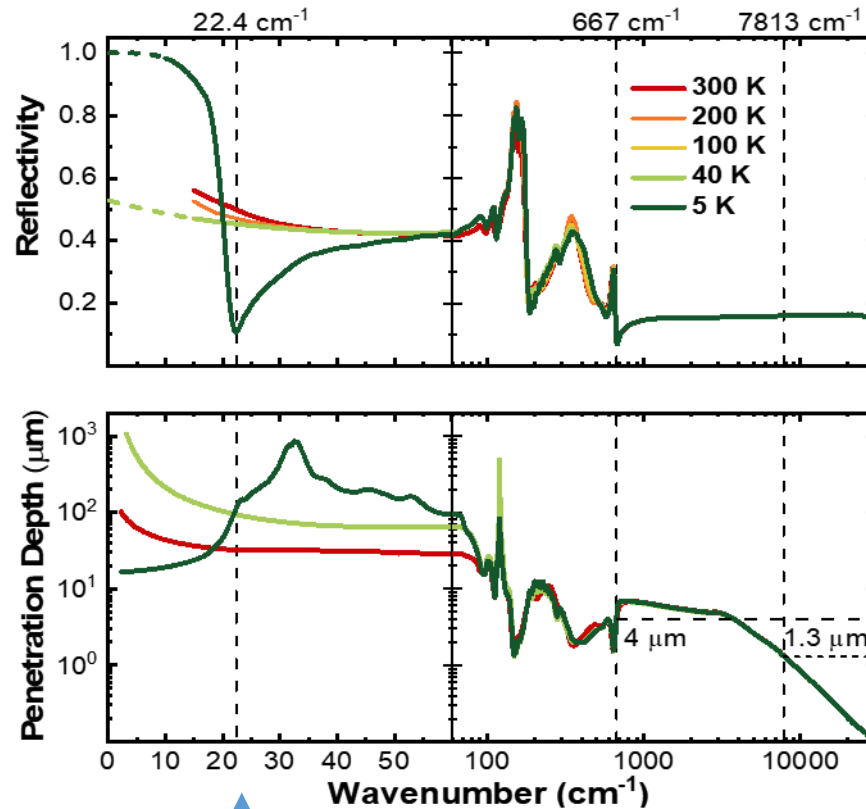
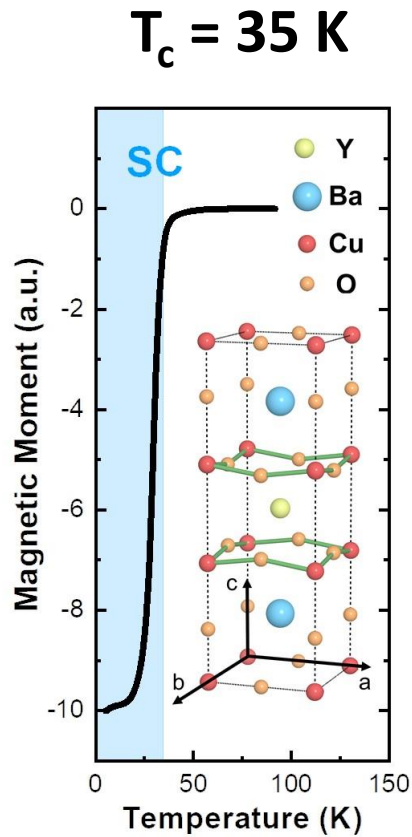
$$f \frac{\tilde{\epsilon}_T(\omega) - \tilde{\epsilon}_E(\omega)}{\tilde{\epsilon}_T(\omega) + 2\tilde{\epsilon}_E(\omega)} + (1 - f) \frac{\tilde{\epsilon}_{eq}(\omega) - \tilde{\epsilon}_E(\omega)}{\tilde{\epsilon}_{eq}(\omega) + 2\tilde{\epsilon}_E(\omega)} = 0$$

Bruggeman's effective medium model

~20% of excited volume becomes superconducting even after considering penetration depth mismatch!

Underdoped YBCO6.45 with $T_c=35$ K

No report on NIR pump THz probe on YBCO. We want to compare the photoinduced effect between NIR and MIR (phonon resonant) excitations.

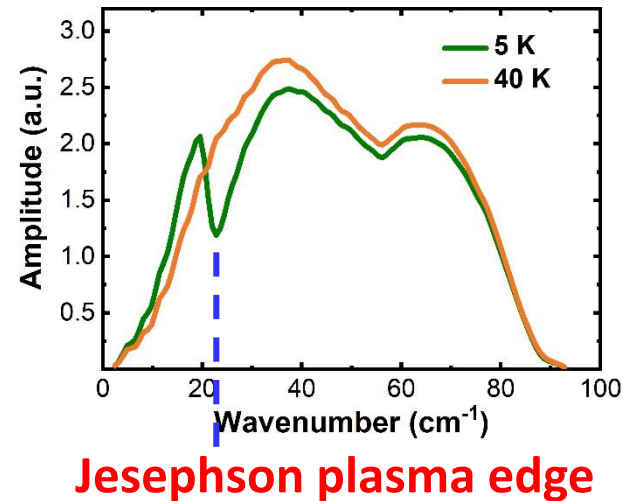
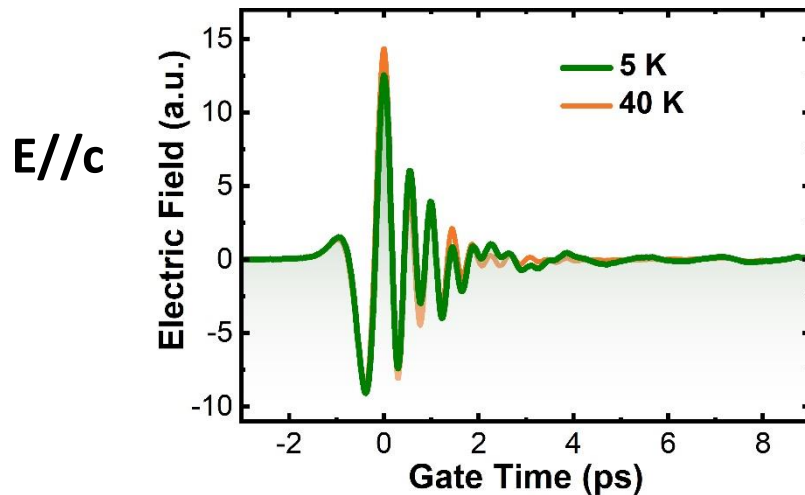


$$d(\omega) = c / \omega \cdot K$$

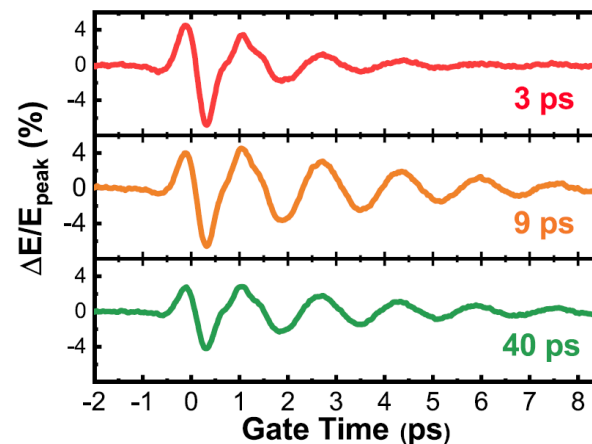
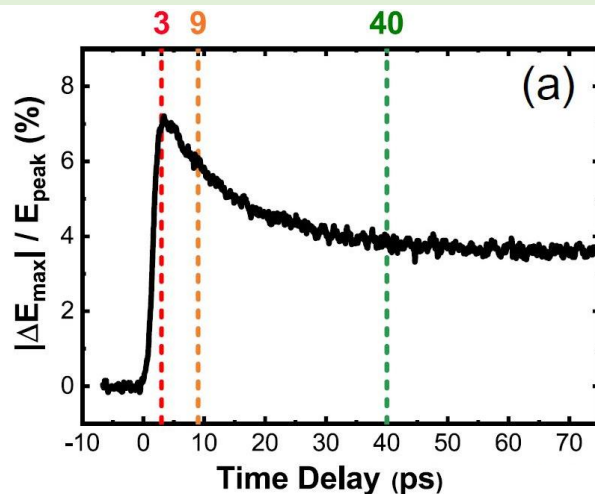
Josephson plasma edge

Excitation wavelengths

Static THz measurement in normal and SC state



NIR pump-induced change in SC state

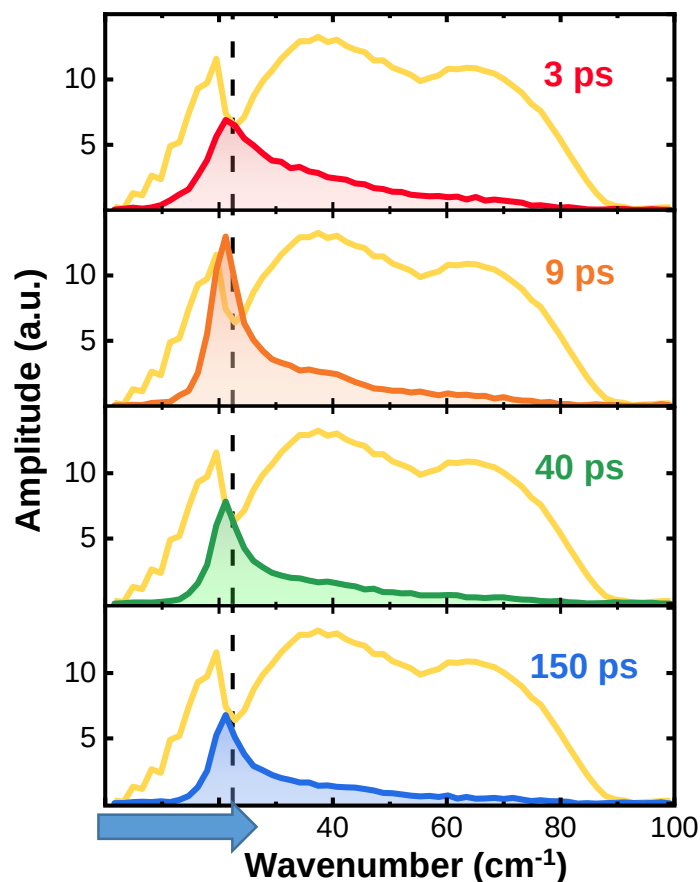


5K, 1mJ/cm²

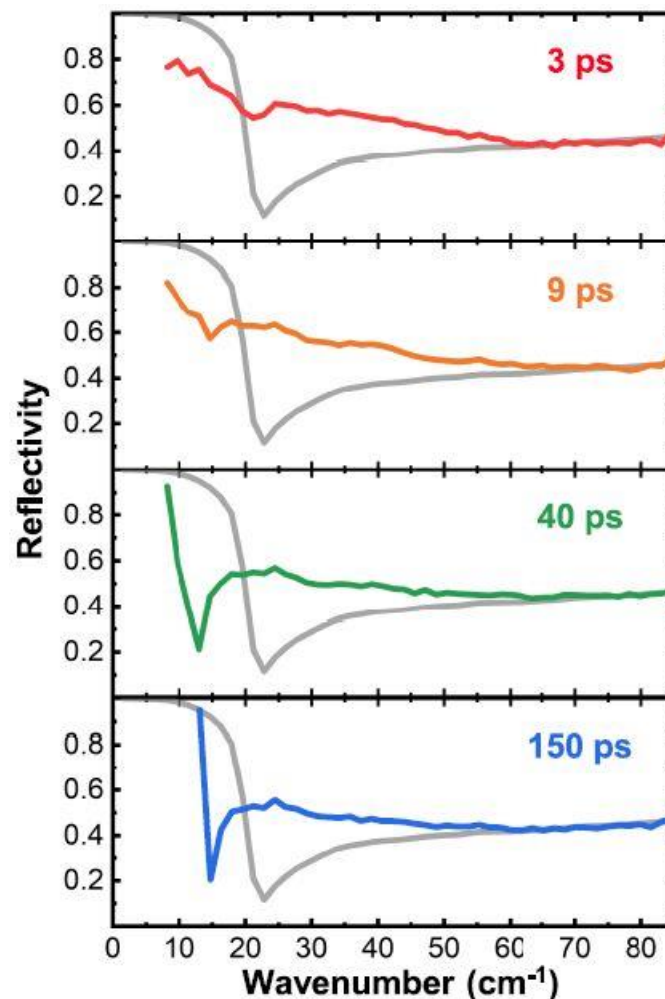
Pump wavelength at 1.28 μm

The oscillation gives a peak near JPE

NIR pump-induced change in SC state

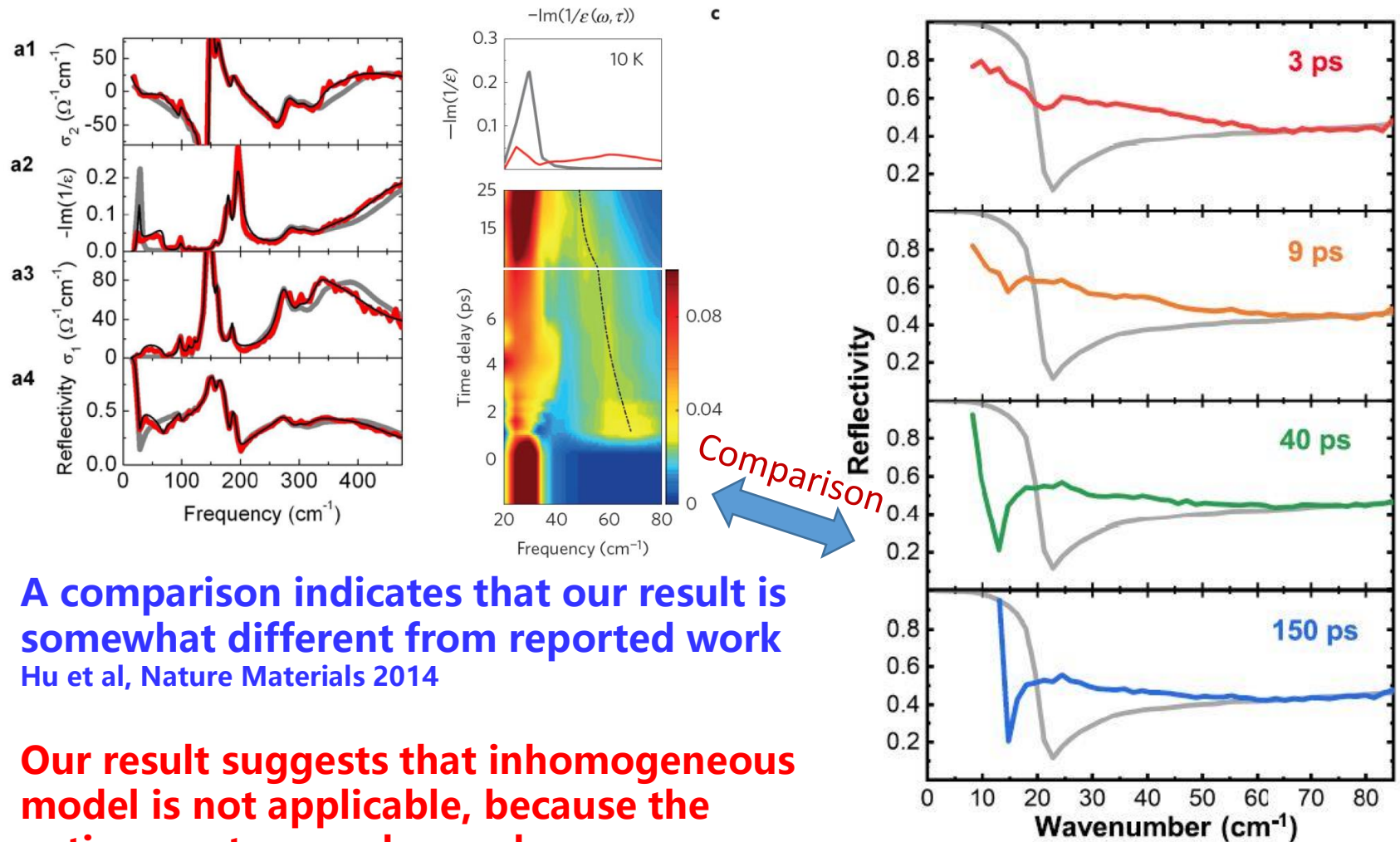


Multilayer model after considering mismatch of penetration depths



Quasiparticle (hot electron) excitations are present after initial pumping. With time delay a JPE develops at low energy and shifts gradually to higher energy.

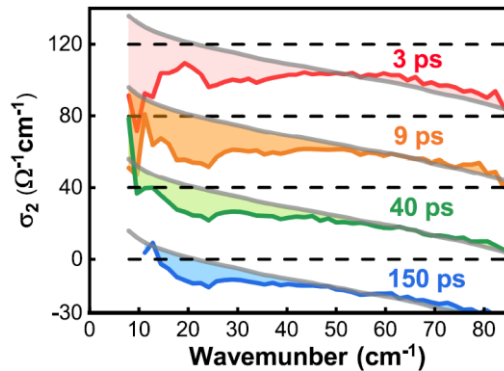
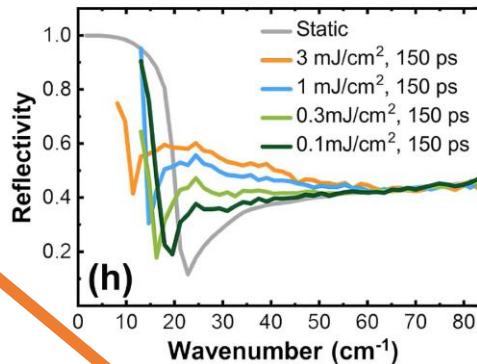
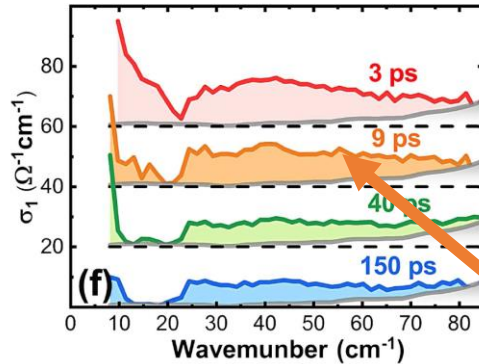
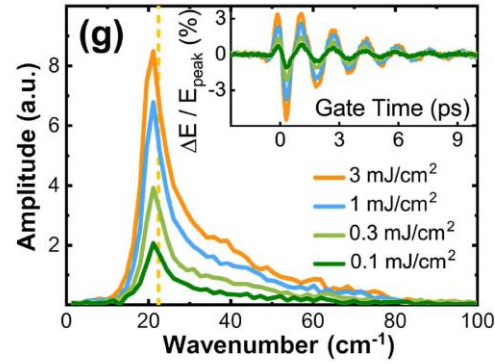
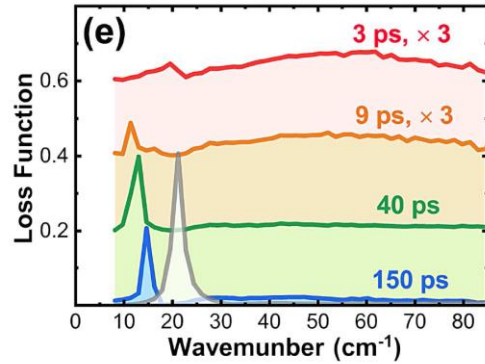
NIR pump-induced change in SC state



A comparison indicates that our result is somewhat different from reported work
Hu et al, Nature Materials 2014

Our result suggests that inhomogeneous model is not applicable, because the entire spectra are changed.

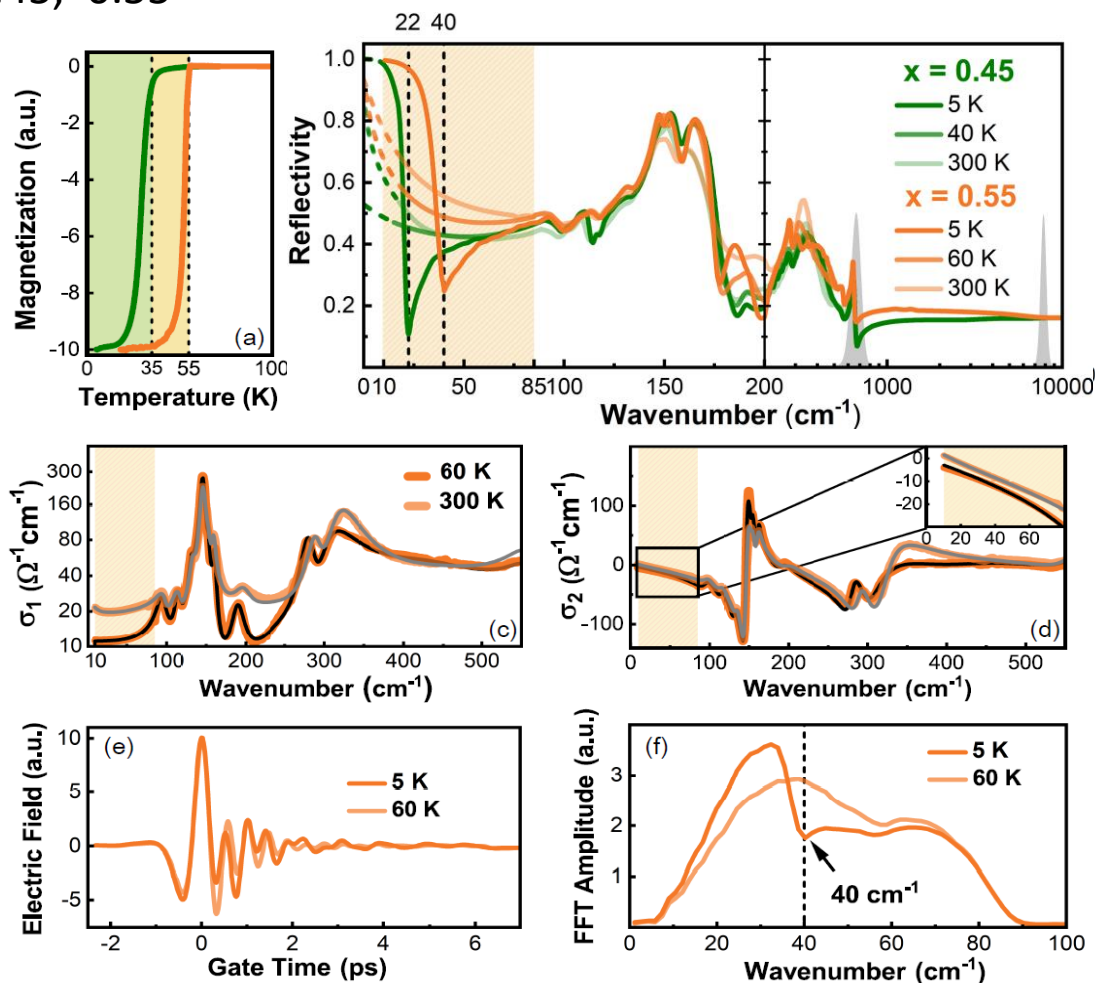
Quasiparticle (hot electron) excitations are present after initial pumping. With time delay a JPE develops at low energy and shifts gradually to higher energy.



Incoherent quasiparticles are present above JPE. Its spectral weight becomes smaller with time delay but always present.

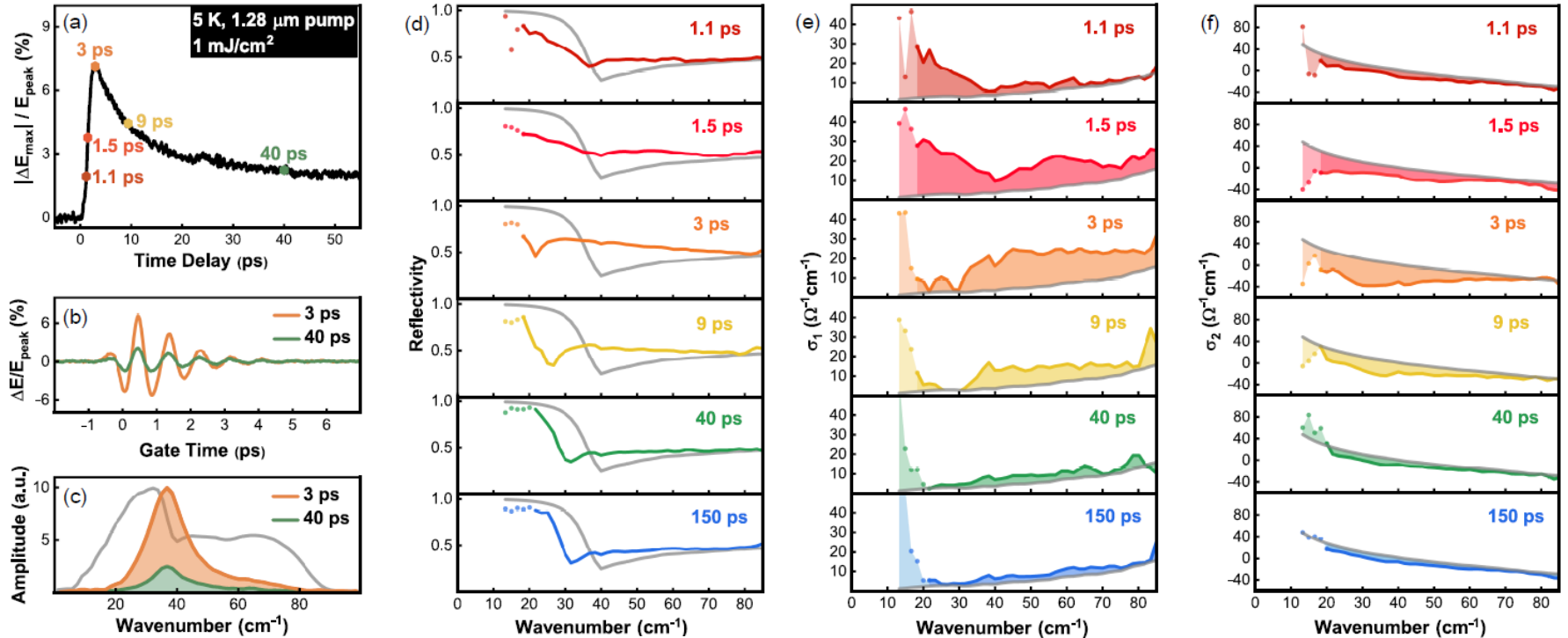
Underdoped YBCO ($x=6.45, 6.55$) with $T_c=35$ K, 55 K

$X=0.45, 0.55$



NIR pump-induced change in the superconducting state

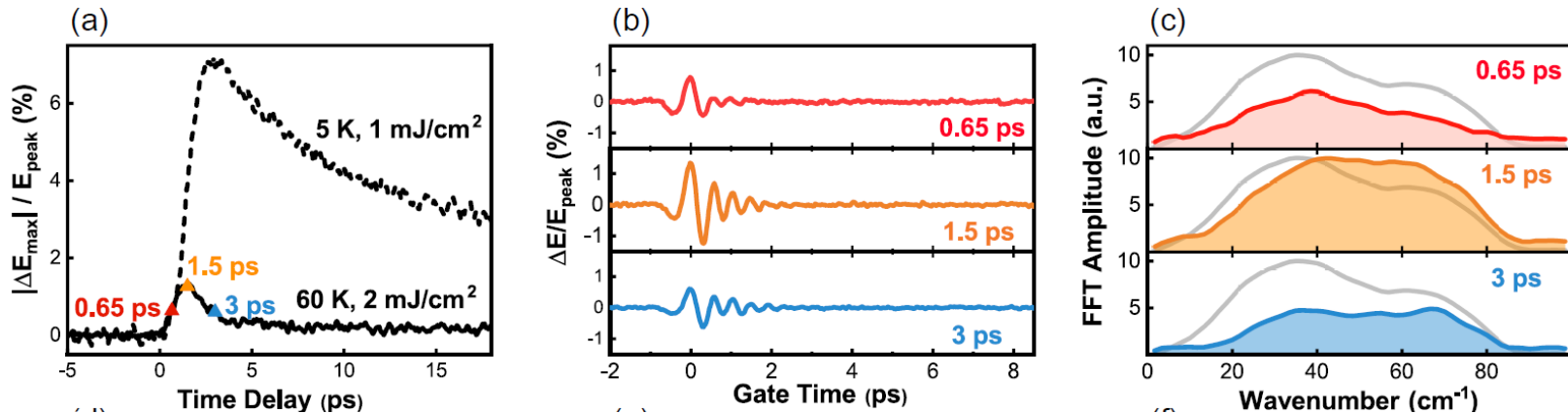
YBCO6.55 $T_c=55$ K



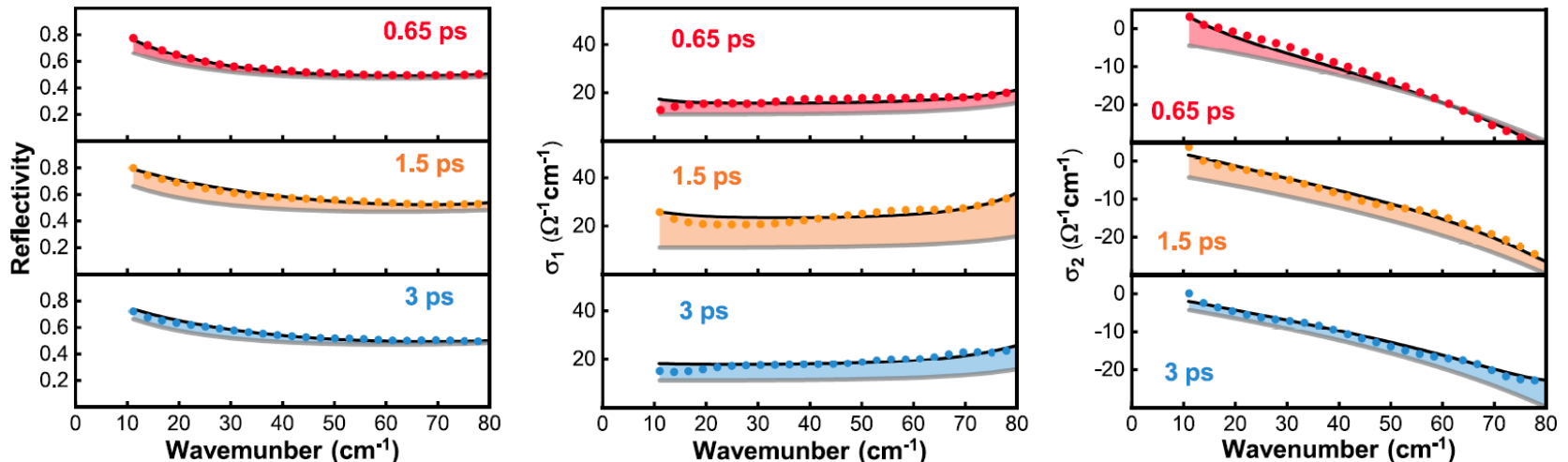
Quasiparticle (hot electron) excitations are present after initial pumping. With time delay a JPE develops at low energy and shifts gradually to higher energy.

NIR pump-induced change in the normal state

YBCO6.55 $T_c=55$ K



Multilayer model after considering mismatch of penetration depths

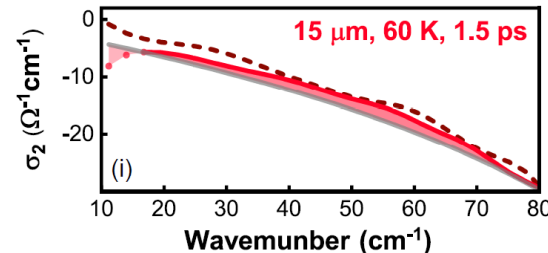
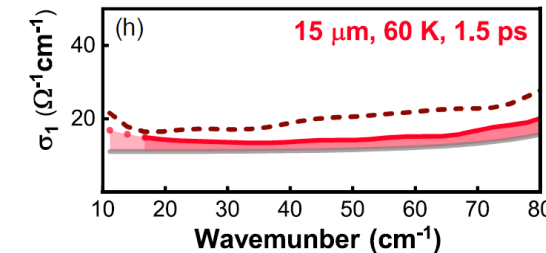
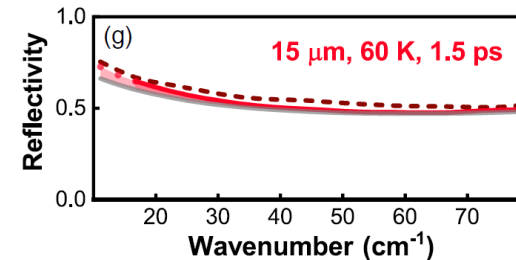
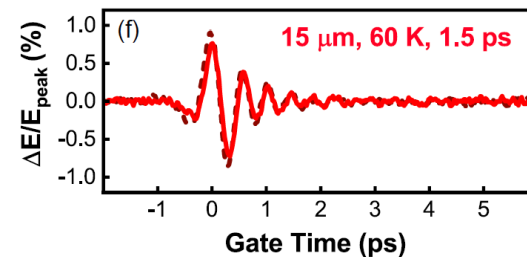
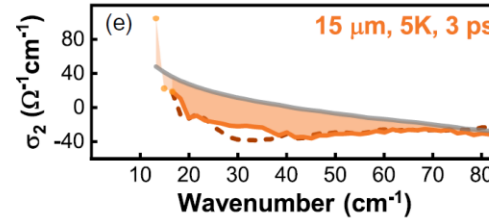
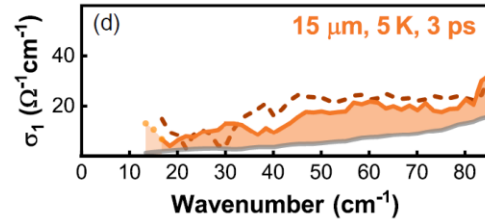
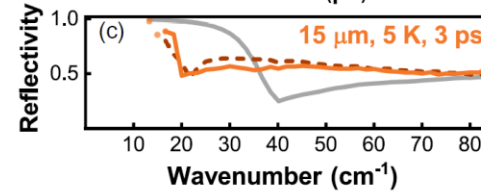
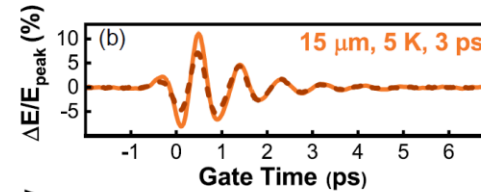
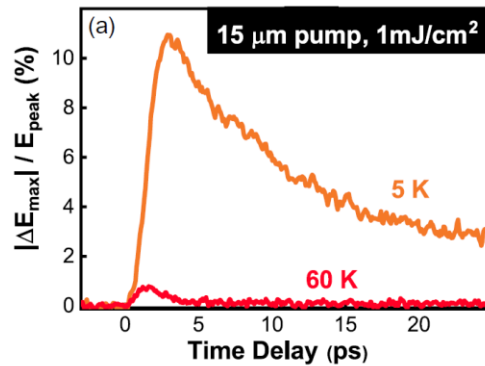


Comparison between NIR and MIR pumping

YBCO6.55

SC state

60K,
Normal state



Solid curves:
15 μm ;

Dashed curves:
1.2 μm

We observe enhancement in R (or edge-like shape in $\Delta R/R$), and increase/enhancement in both $\sigma_1(\omega)$, $\sigma_2(\omega)$ in the normal state!

There are two possible explanations:

1. light-induced transient superconductivity

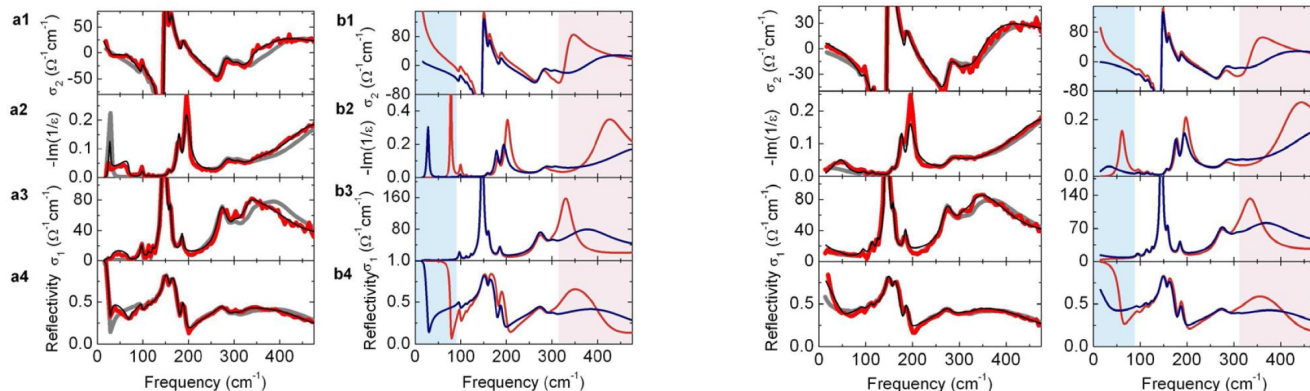
Superconducting condensate is taken into consideration

$$\sigma_2(\omega) = \omega_{ps}^2 / 4\pi\omega$$

$$\varepsilon(\omega) = \varepsilon_{\infty} - \frac{\omega_{ps}^2}{\omega^2} - \frac{\omega_p^2}{\omega^2 + i\omega\Gamma} + \text{Lorentz}$$

$$\begin{matrix} n(\omega), k(\omega) \\ \sigma_1(\omega), \sigma_2(\omega) \end{matrix} \rightarrow R(\omega)$$

A nonhomogeneous model was used to explain the small change in early reported work. However, **our measurement suggest that the nonhomogeneous model is not applicable**, because the entire spectra change.



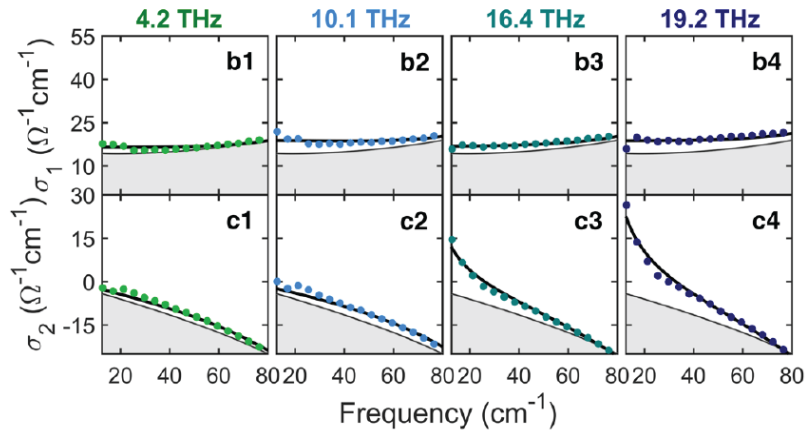
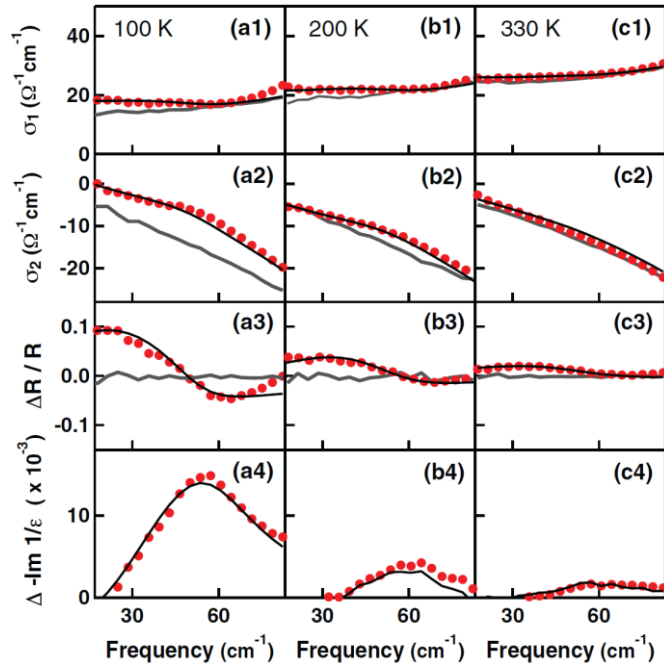
YBCO

S. Kaiser et al. PRB 2014

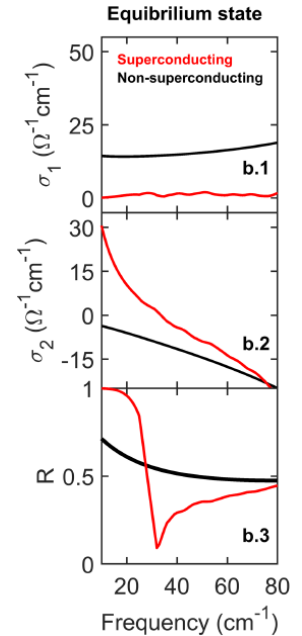
B. Liu et al. arXiv1905.08356

YBCO6.5

YBCO6.5 @100 K



More recent measurement at two phonon resonant frequencies indicates homogenous excitations.



Still unlikely to be SC. Missing spectral weight not observed in $\sigma_1(\omega)$.

$$\sigma = \frac{1}{8} \omega_{ps}^2 \delta(\omega) + i \omega_{ps}^2 / 4\pi\omega$$

$$\omega_{ps}^2 = 8 \int_0^{\omega_c} d\omega [\sigma_1(\omega, T > T_c) - \sigma_1(\omega, T < T_c)]$$

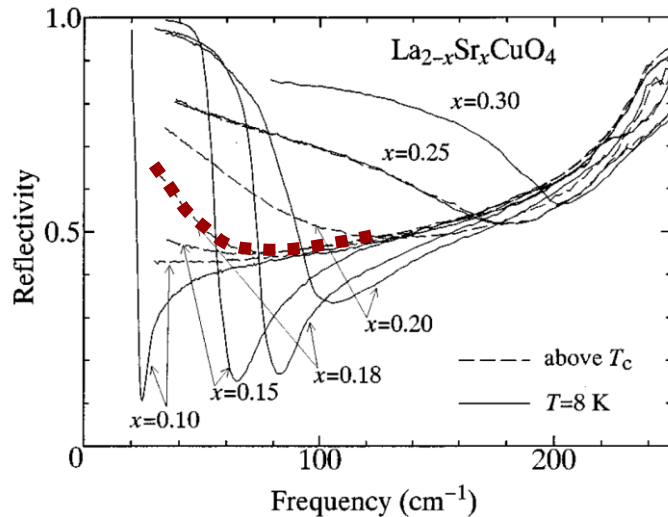
$$\lambda_L = c/\omega_{ps}$$

$$\omega \rightarrow 0$$

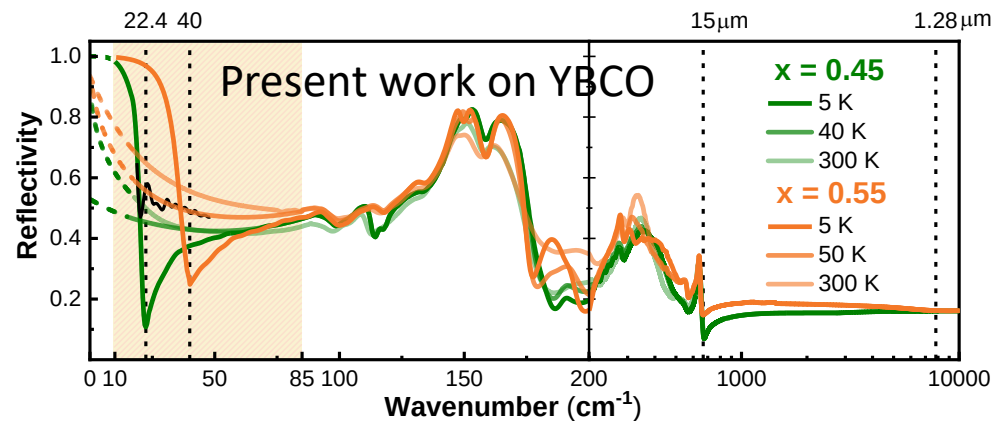
$$\frac{1}{\lambda_L^2} = \frac{\omega_{ps}^2}{c^2} = \frac{4\pi}{c^2} \omega \sigma_2(\omega)$$

2. Quasiparticle (hot electron) excitations

S. Uchida, K. Tamasaku, S. Tajima, PRB 53, 14558 (1996)

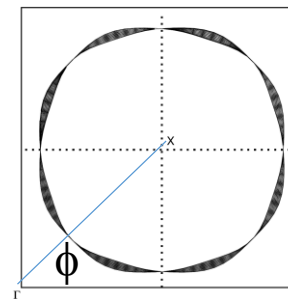


Photoexcited quasiparticles contribute to $R(\omega)$ in a way similar to the increase of doping levels.



C-axis conduction has complex contributions:

- Via O 2p and virtual Cu 4s orbital;
- **Hopping integral $t_{\perp}(\phi)$ strongly ϕ -dependent, vanishing at nodal direction;**
- **Carrier scattering rates along c-axis are also strongly ϕ -dependent;**
- Presence of pseudogap near antinodal regions;
- Presence of Cu-O chains.....



4 nondispersive nodes for Y-, Hg- cuprates;

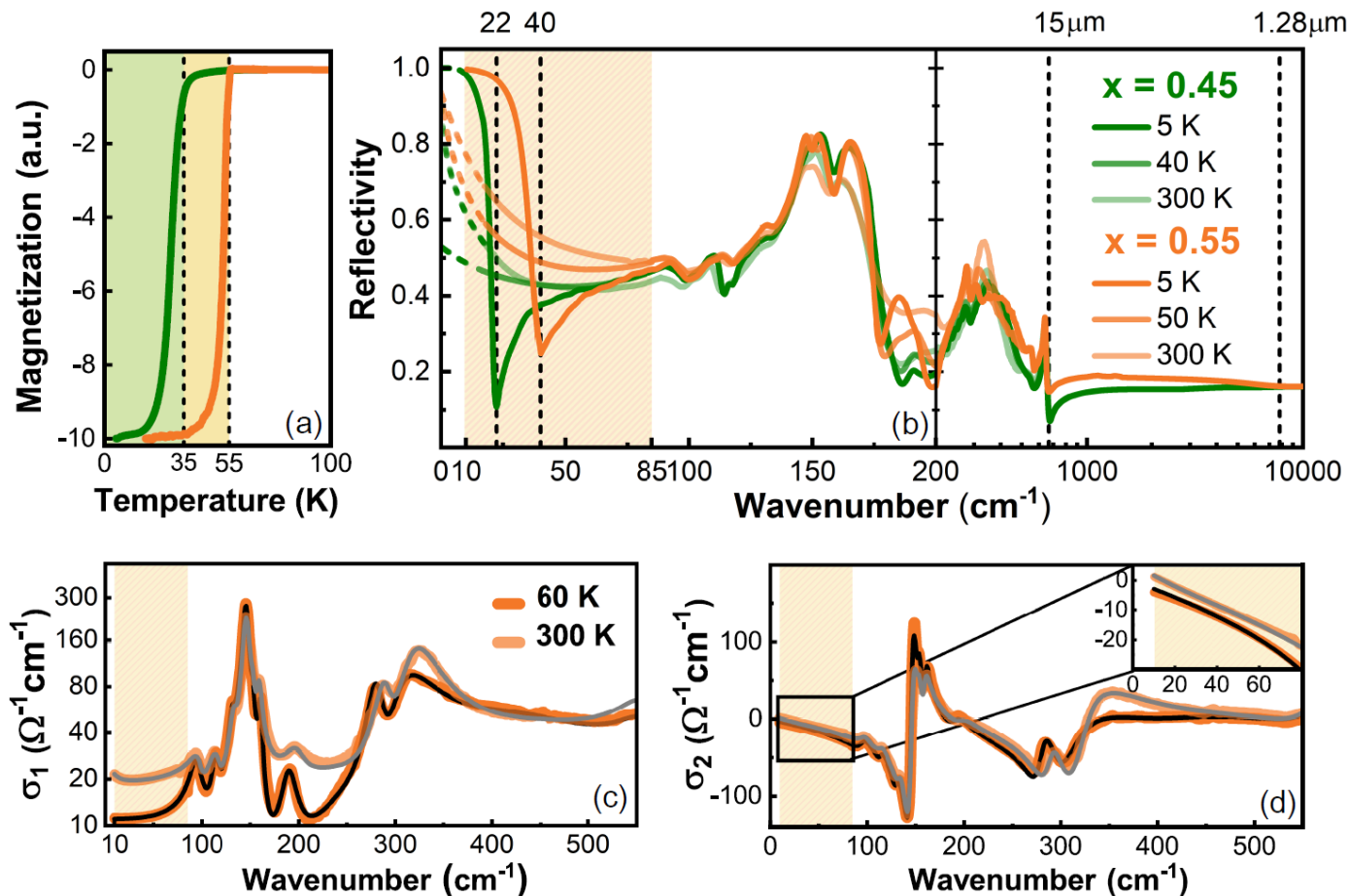
8 nondispersive nodes for La-, Bi-, Tl- cuprates

Simple Drude model is not expected to reproduce $\sigma_1(\omega)$, $\sigma_2(\omega)$, $R(\omega)$...

2. Quasiparticle (hot electron) excitations

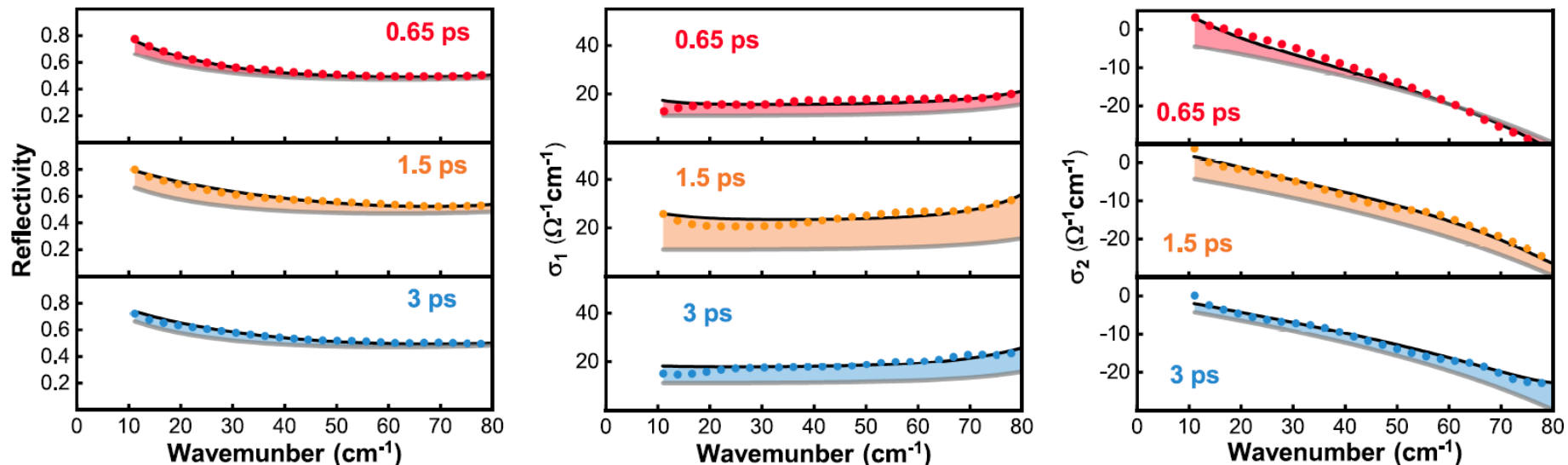
Just using two Drude components
+ Lorentz (for phonons)

$$\epsilon(\omega) = \epsilon_{\infty} - \sum_{i=1} \frac{\omega_{pi}^2}{\omega^2 + i\omega/\tau_i} + \sum_{j=1} \frac{S_j^2 e^{i\theta_j}}{\omega_j^2 - \omega^2 - i\omega/\tau_j} \quad \Rightarrow \quad \begin{matrix} \sigma_1(\omega) \\ \sigma_2(\omega) \end{matrix}$$



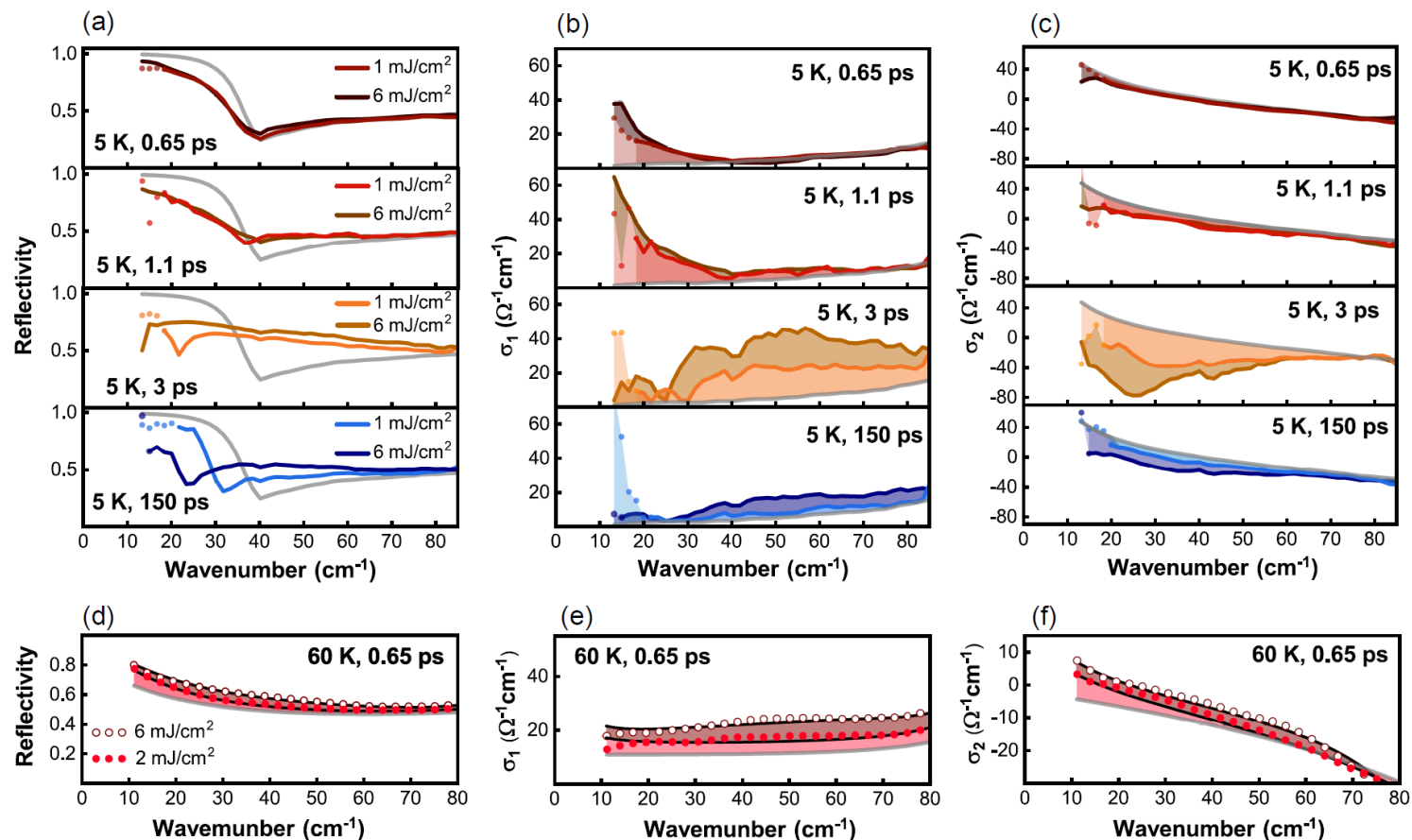
2. Quasiparticle (hot electron) excitations

$$\epsilon(\omega) = \epsilon_{\infty} - \sum_{i=1} \frac{\omega_{pi}^2}{\omega^2 + i\omega/\tau_i} + \sum_{j=1} \frac{S_j^2 e^{i\theta_j}}{\omega_j^2 - \omega^2 - i\omega/\tau_j} \quad \Rightarrow \quad \begin{matrix} n(\omega), k(\omega) \\ \sigma_1(\omega), \sigma_2(\omega) \end{matrix} \quad \Rightarrow \quad R(\omega)$$



1/ ω -like dependence of σ_2 in a very limited THz frequency range can not be uniquely attributed to transient superconductivity.

Pump at higher fluence/peak field in SC and normal states:



Our laser system: 800 nm, 35 fs, 1 kHz. After TOPAS OPA, pulse duration is about 50 fs for NIR 1.28 μm, 350 fs for MIR 15 μm by DFG on GaSe. At 6 mJ/cm², the peak electric field is about 9.5 MV/cm.

TABLE I. Fitting parameters of equilibrium state

Temperature (K)	$\omega_{p1}(cm^{-1})$	$1/\tau_1 (cm^{-1})$	$\omega_{p2}(cm^{-1})$	$1/\tau_2 (cm^{-1})$
60	56	60	420	270
300	65	8	550	270

TABLE II. Fitting parameters of non-equilibrium state at 60 K

Pump Fluence (mJ/cm ²)	Time Delay (ps)	$\omega_{P1}(cm^{-1})$	$\Gamma_1 (cm^{-1})$	$\omega_{P2}(cm^{-1})$	$\Gamma_2 (cm^{-1})$
2	0.65	70	4	490	270
2	1.5	70	10	600	270
2	3	30	10	530	270
6	0.65	85	4	520	270

Very small scattering rate arising from the sharp upturn in $R(\omega)$ was also observed in equilibrium state (e.g. $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ at higher doping)

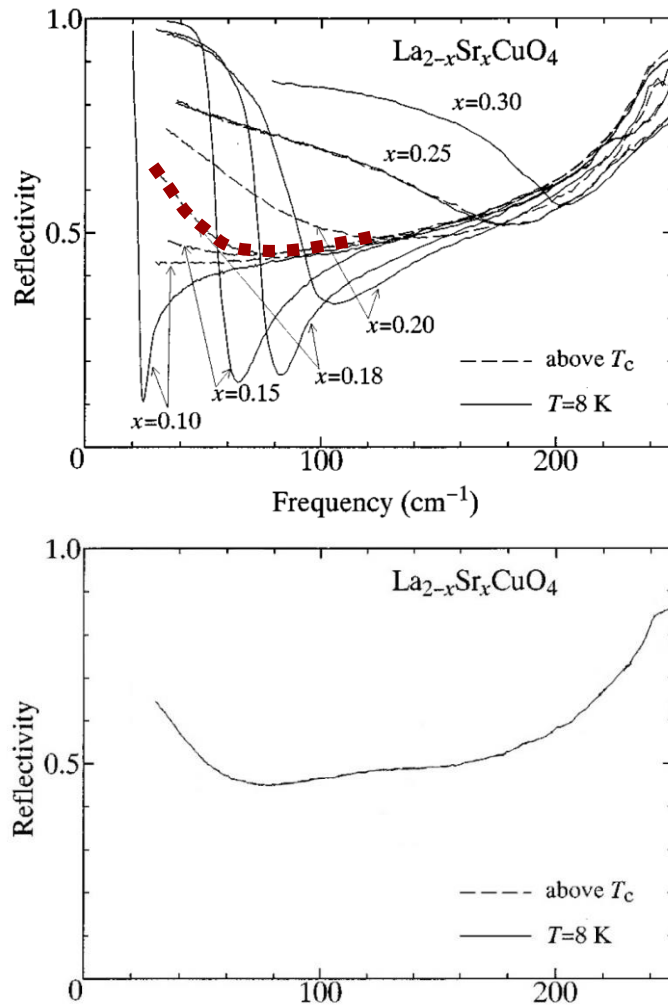


FIG. 14. $x = 0.18$, $\sim 40\text{ K}$

S. Uchida et al.
Phys. Rev. B 53, 14558 (1996)

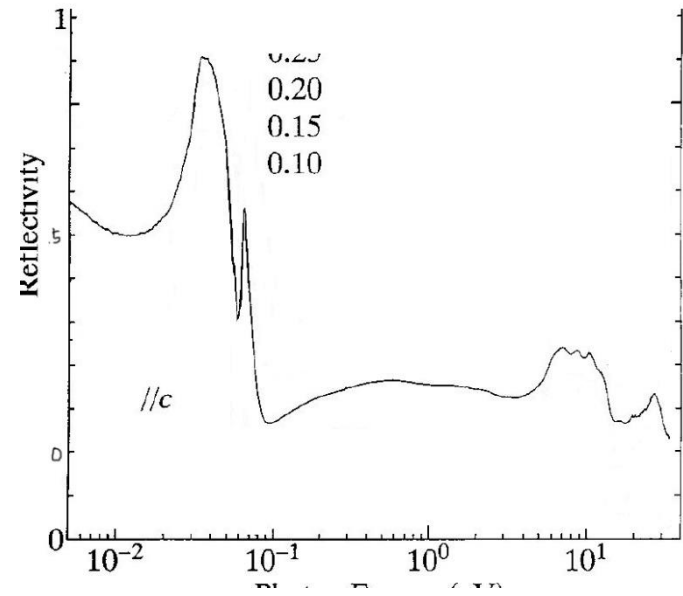
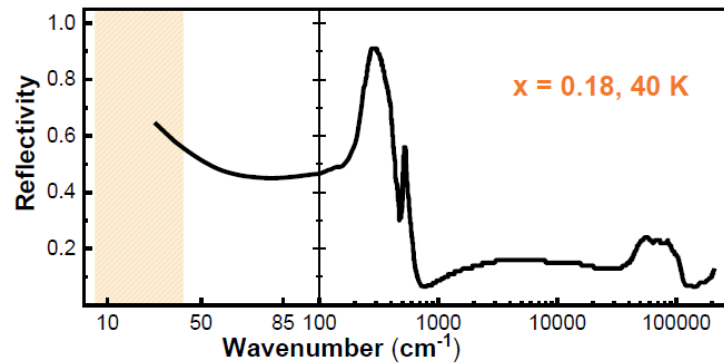
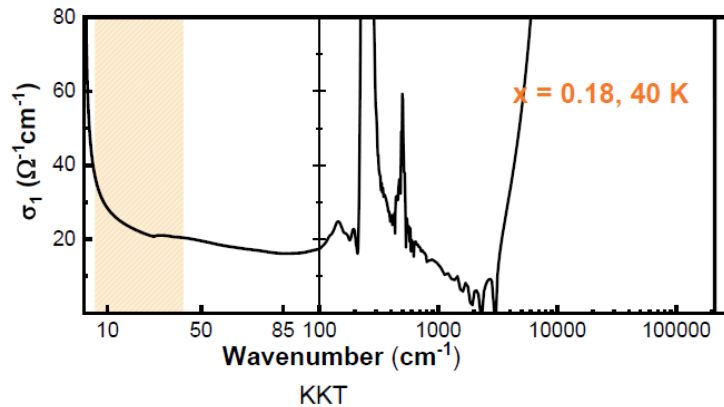


FIG. 3. $x = 0.2$, 300 K

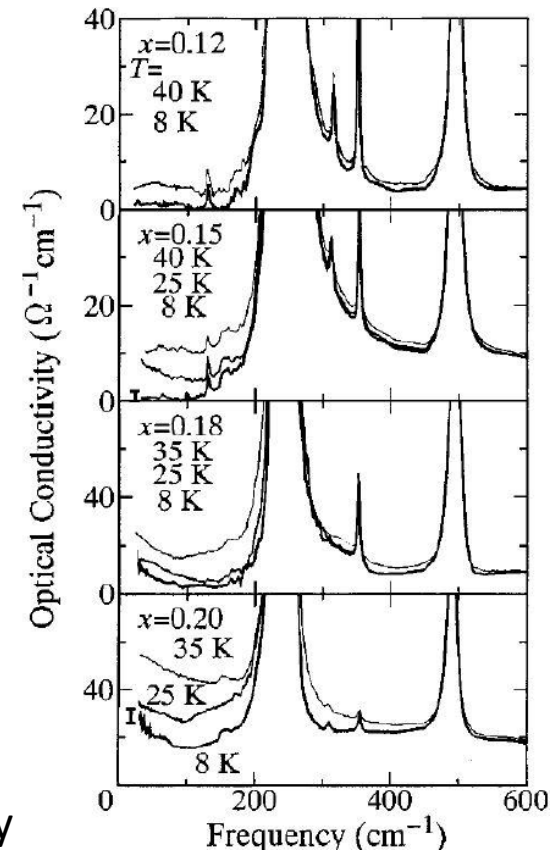
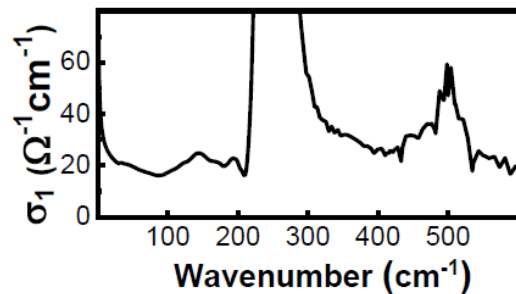
Data thief



KKT

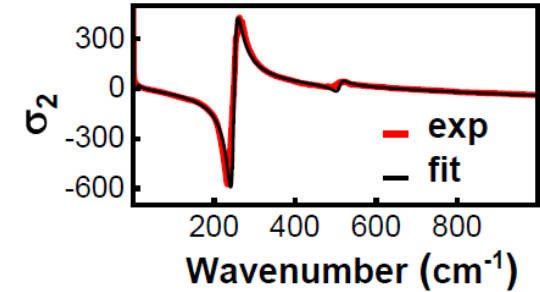
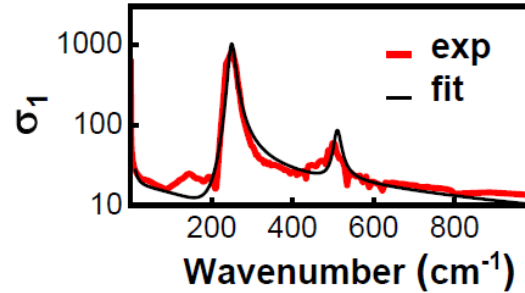
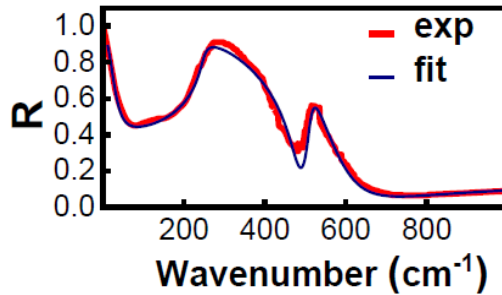


KKT



We digitized the reported reflectance spectrum, performed KK transformation to get optical conductivity and made similar Drude+Lorentz analysis...

Drude	ω_{p1}	$1/\tau_1$	ω_{p2}	$1/\tau_2$
	900	740	150	4



In early studies, the quasiparticle scattering rate along the c-axis was believed to be very large, for the conductivity is very small.

Since clear Drude-like increase and/or upturn of $R(\omega)$ at very low frequency are observed in the equilibrium state, it turns out that the normally assumed very large scattering rate may not be true. This issue should be further studied.

Summary:

- The pump-induced change occurs mainly at the Josephson plasma edge (JPE) position below T_c .
- Below T_c , intense laser pulses destroy the JPE and excite quasiparticles (hot electrons). With time delay a JPE re-develops at lower energy and shifts gradually to original JPE at higher energy.
- The excitation is homogeneous. Inhomogeneous model is not suitable.
- Above T_c , we observe a small increase in $R(\omega)$ (or edge-like shape in $\Delta R(\omega)/R$), together with enhanced spectral weight in $\sigma_1(\omega)$ and $\sigma_2(\omega)$, upon photoexcitation.
- Those effects can be qualitatively explained by photoexcited quasiparticles. Unlikely to be “a transient superconductivity”.
- NIR and MIR (phonon resonant) excitations have very similar effect.

How to solve the controversy (or to prove transient superconductivity) :

One possibility is to do measurement on very thin samples (thin films with c-axis within the plane) in which one does not need to consider the penetration depth mismatch. (This is because the values chosen for penetration depth have rather dramatic effect.)

Thank You for your attention!