

***Emergent Phenomena in  
Driven Quantum Materials***

***Andrea Cavalleri***

*Max Planck Institute for the Structure and Dynamics of Matter*

*Department of Physics, University of Oxford*

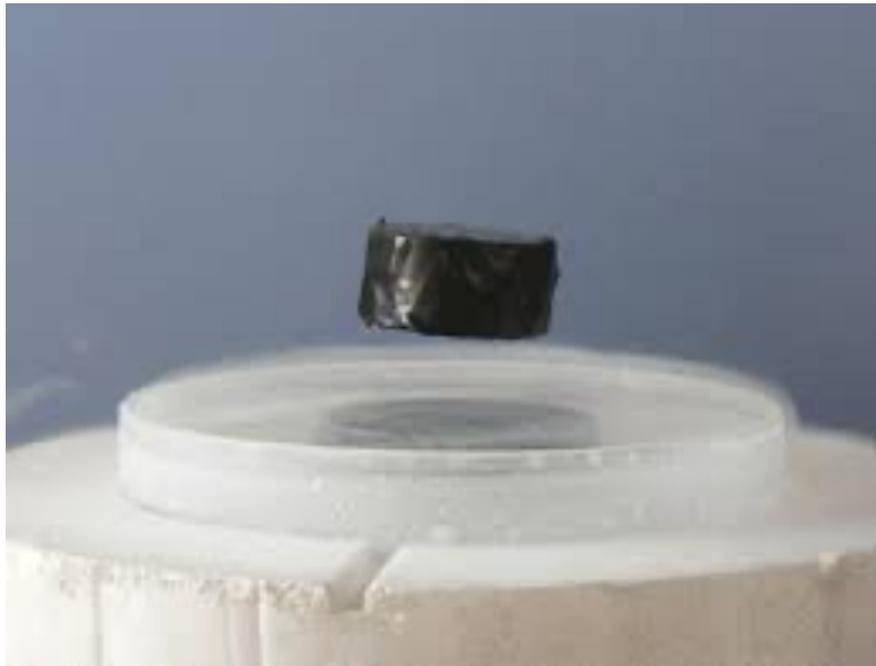
# Quantum Materials do “big things”

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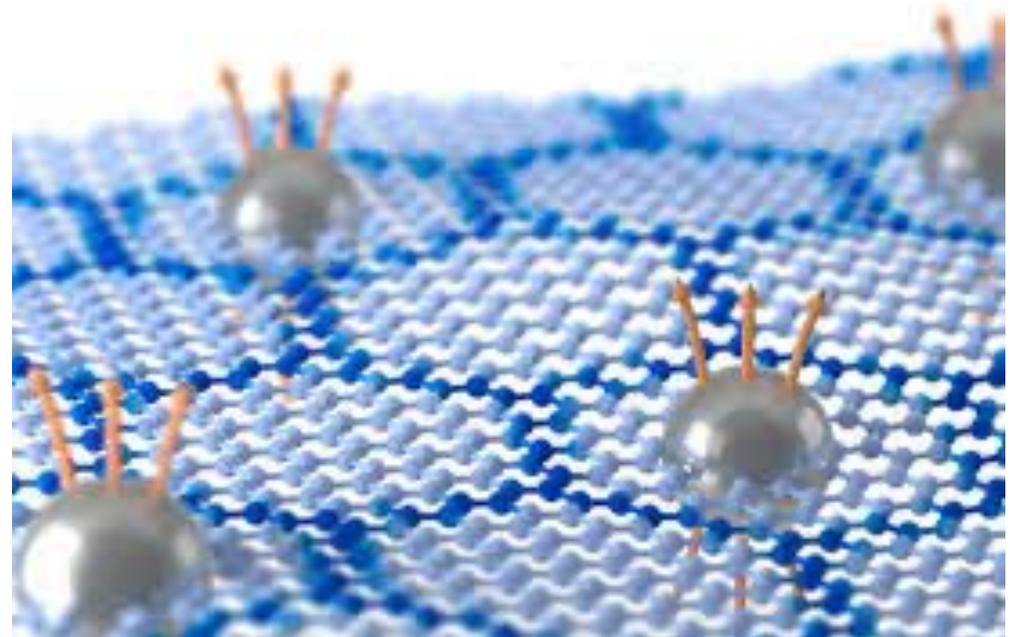
**Macroscopic properties determined by quantum mechanics**

**Exhibit large response functions and extreme sensitivity**

**High  $T_c$  Superconductivity**

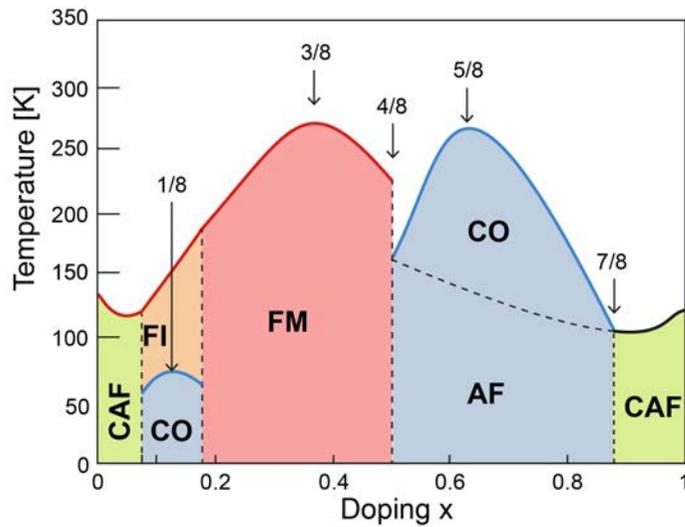


**Topologically Protected Transport**

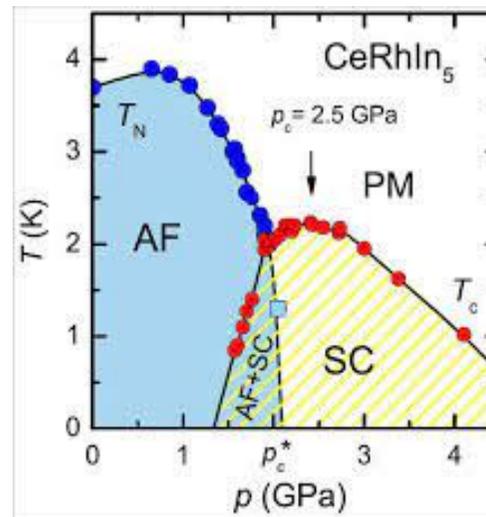


# Quantum Materials – difficult to optimize

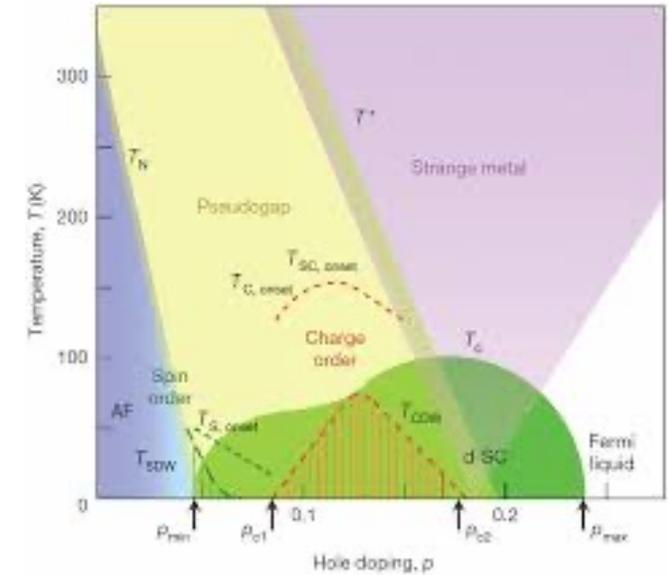
## Magnetoresistive Manganites



## Heavy Fermions



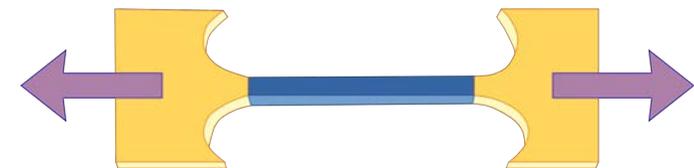
## High $T_c$ Superconductors



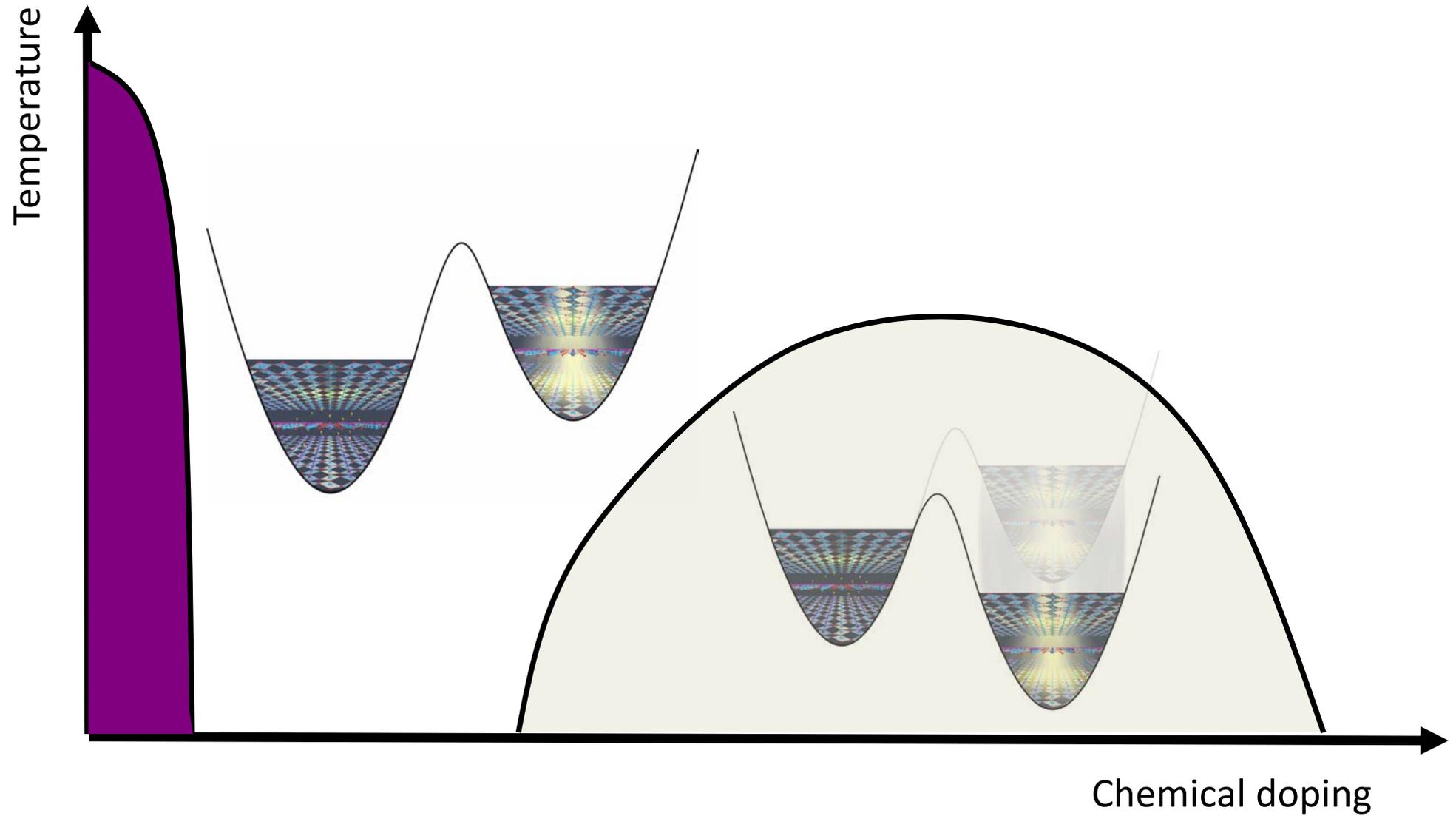
## Materials Growth



## Strain

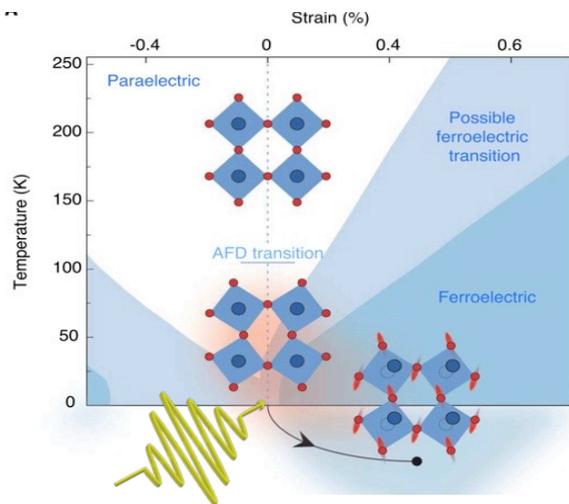


# Phase competition on similar energy scales

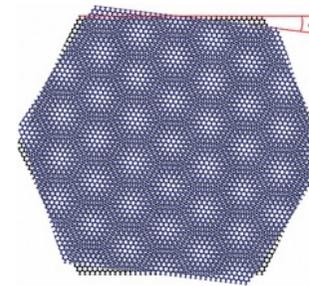


# Control of quantum materials by non-standard means

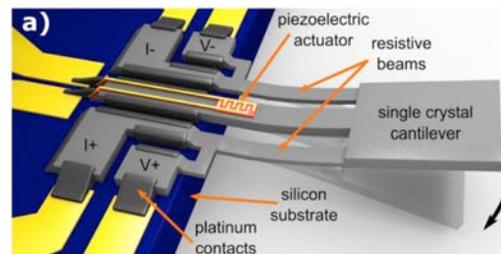
Important scientific advances and **new physical phenomena** are expected in settings in which quantum materials are exposed to **unconventional fields**



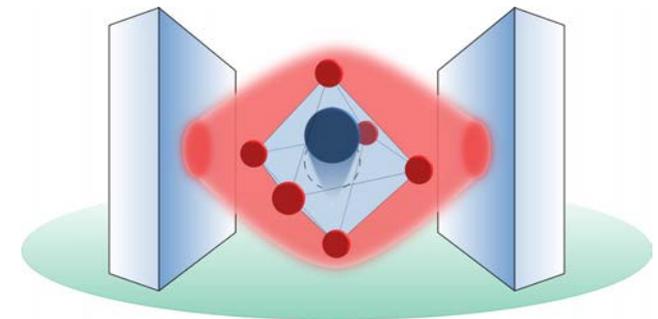
**Dynamically Driven Matter**



**Synthetic Quantum Materials**



**Extreme strain**



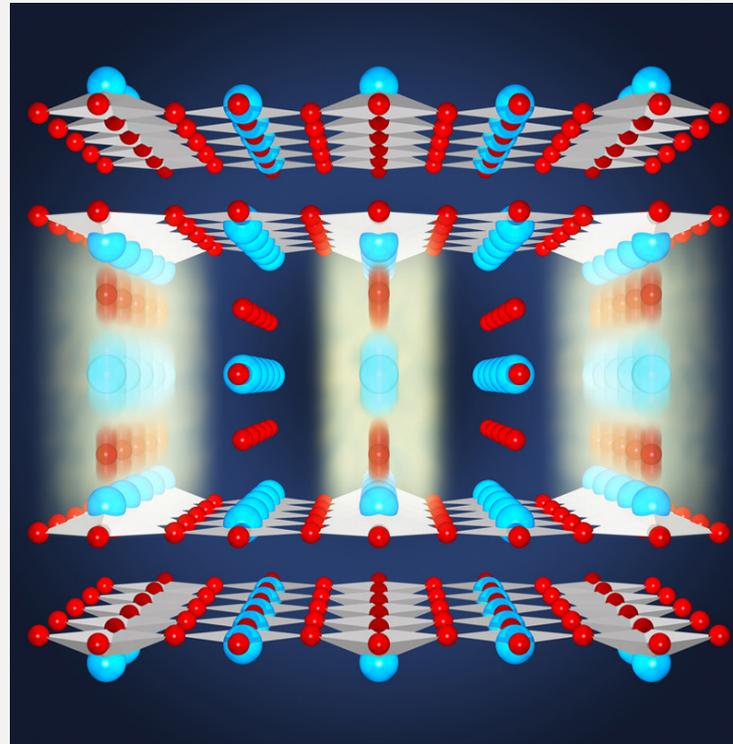
**Quantum Materials  
in Quantum Cavities**



# We look for new physics in driven quantum materials

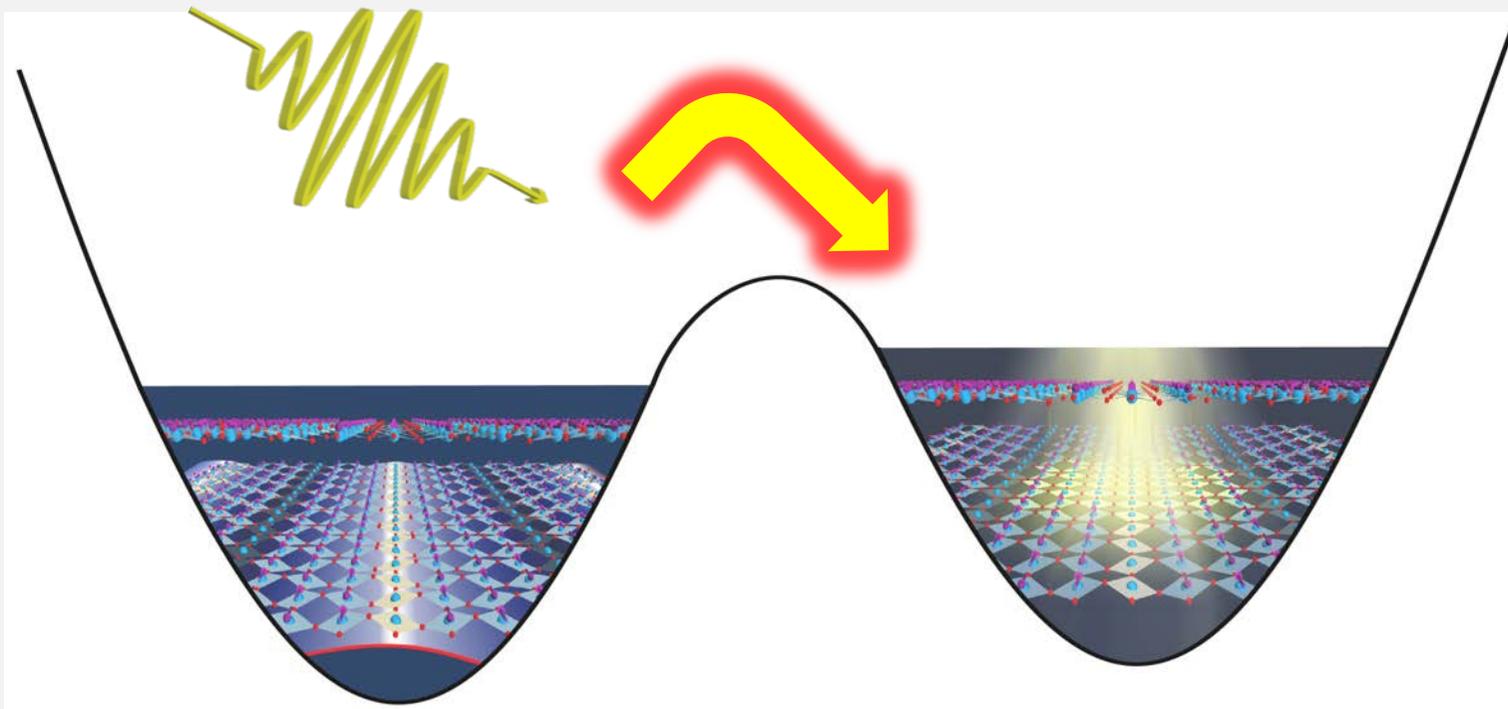
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**Dynamical modulation** can **manipulate** the functional properties of a solid



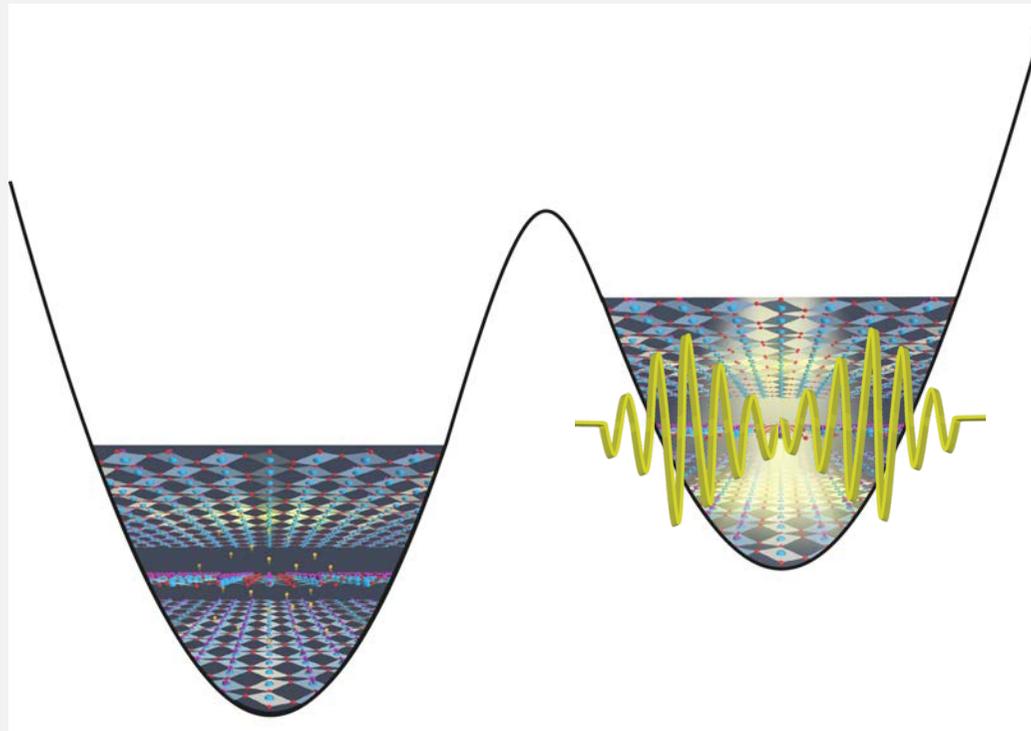
# Generating hidden phases

Dynamical modulation can create switch between metastable states



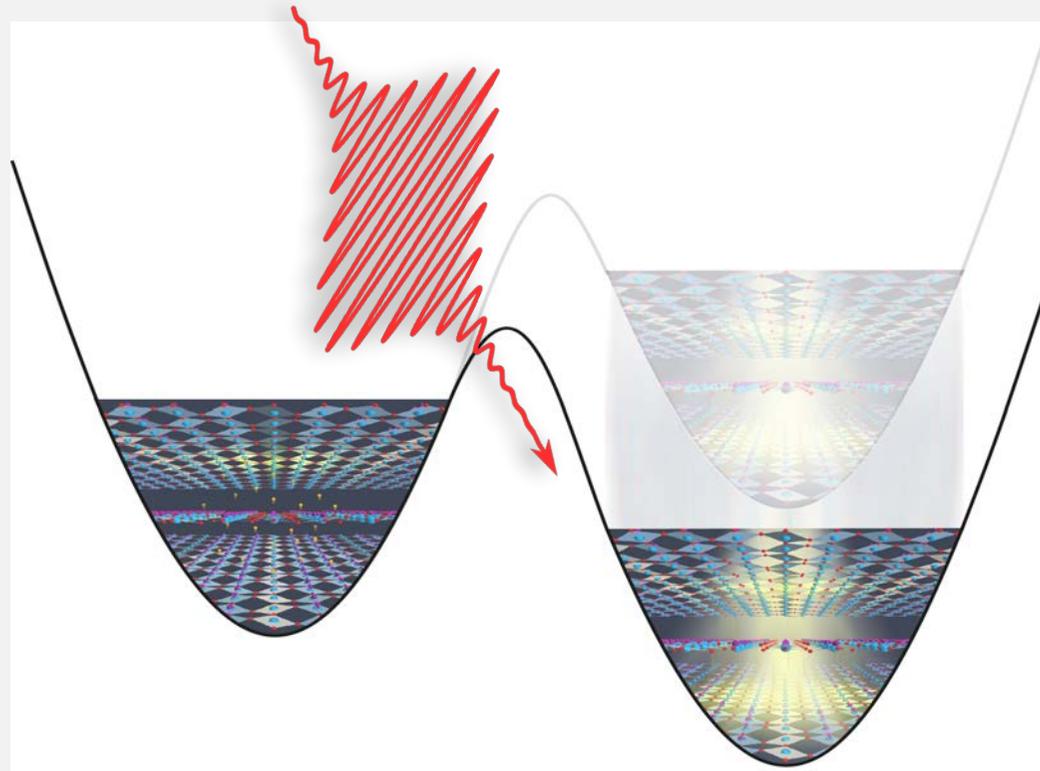
# Cooling fluctuations

Dynamical modulation can create cool fluctuations that destroy coherence

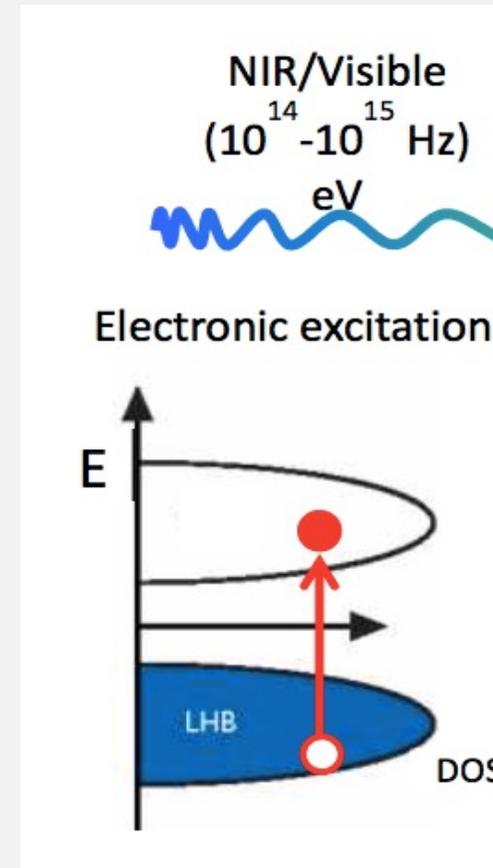
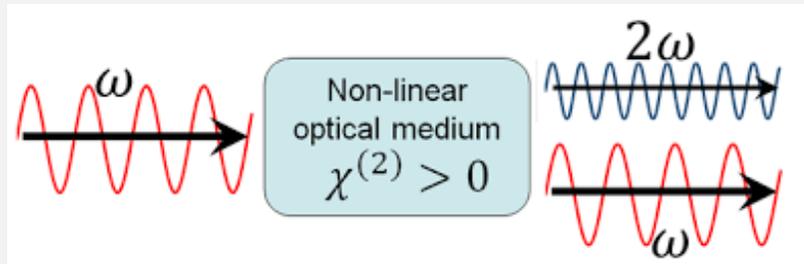


# Renormalizing the energy landscape

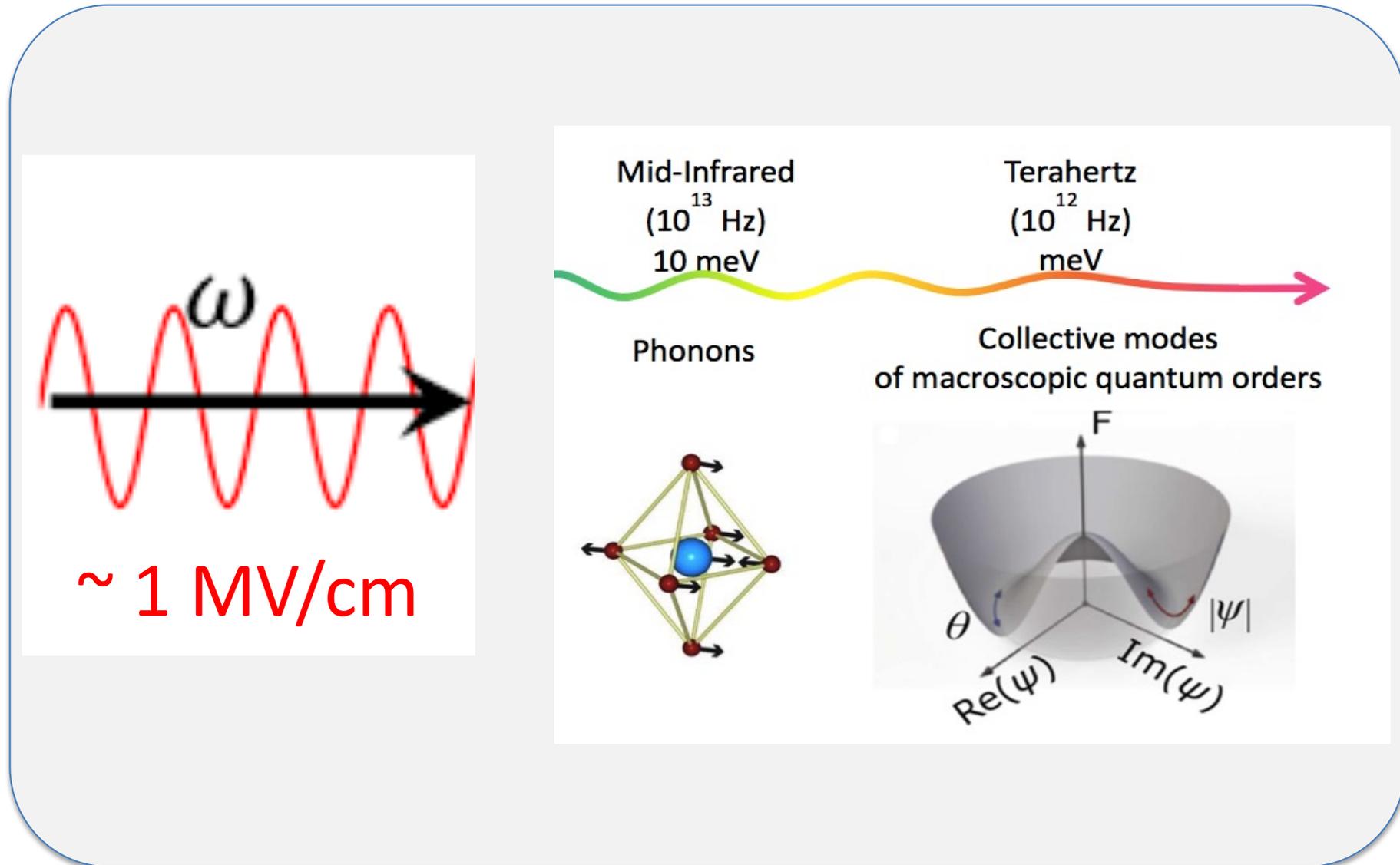
Dynamical modulation can create reshape the energy landscape



# 60 years of Nonlinear Optics in the visible



# Nonlinear Optics for quantum Materials – low frequencies

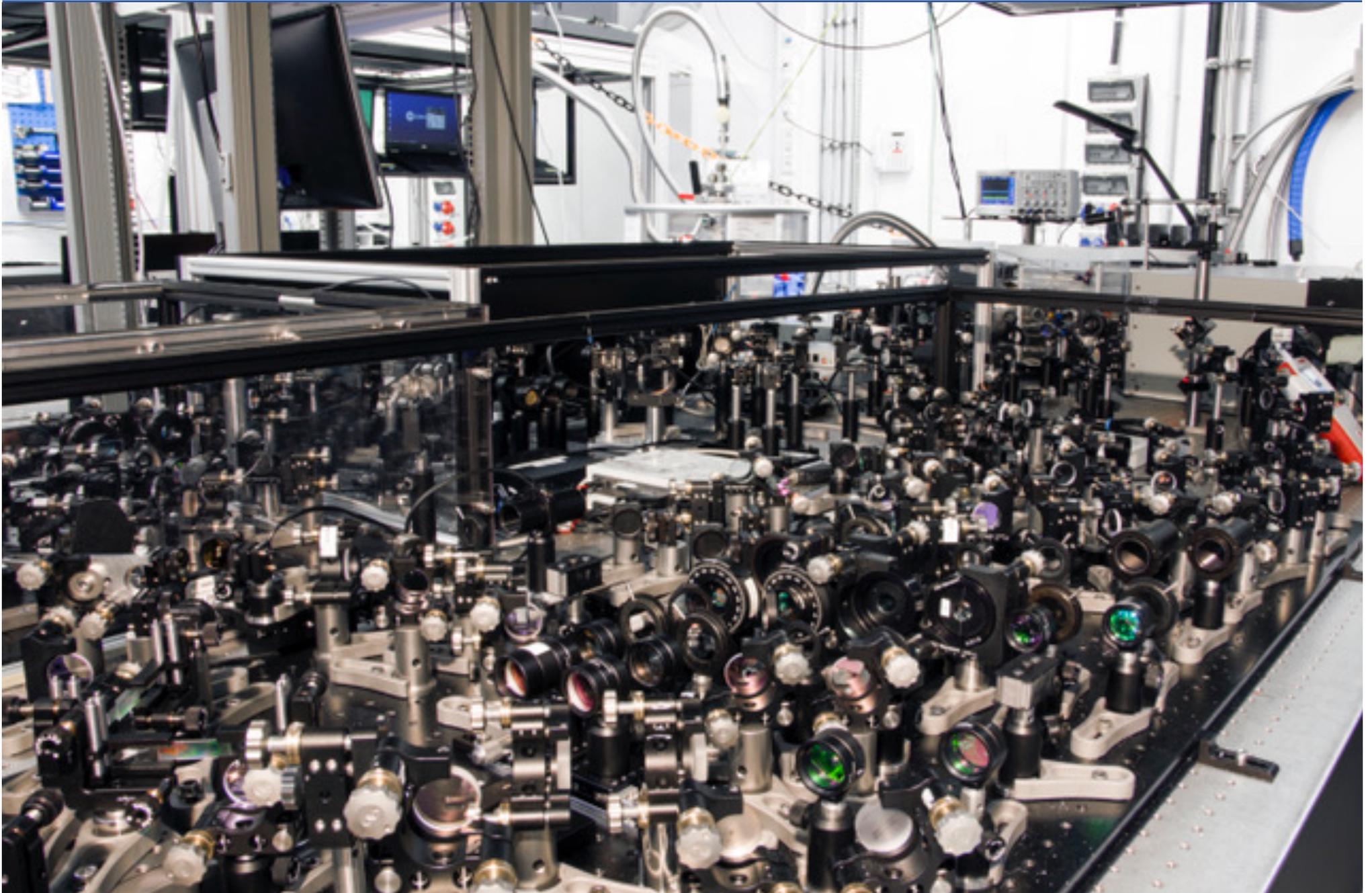


# Until recently only FELs could provide strong far IR

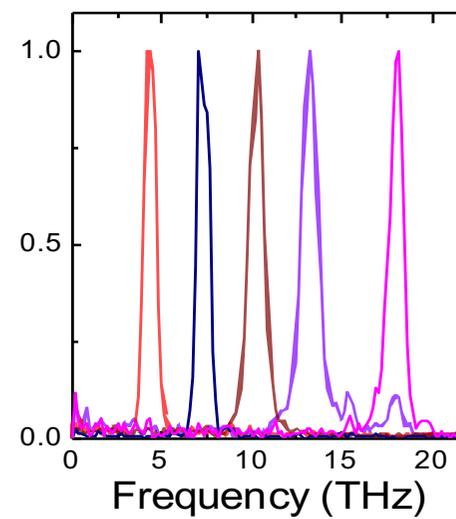
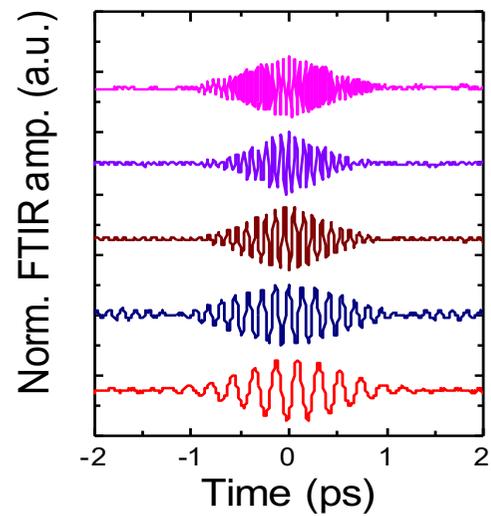
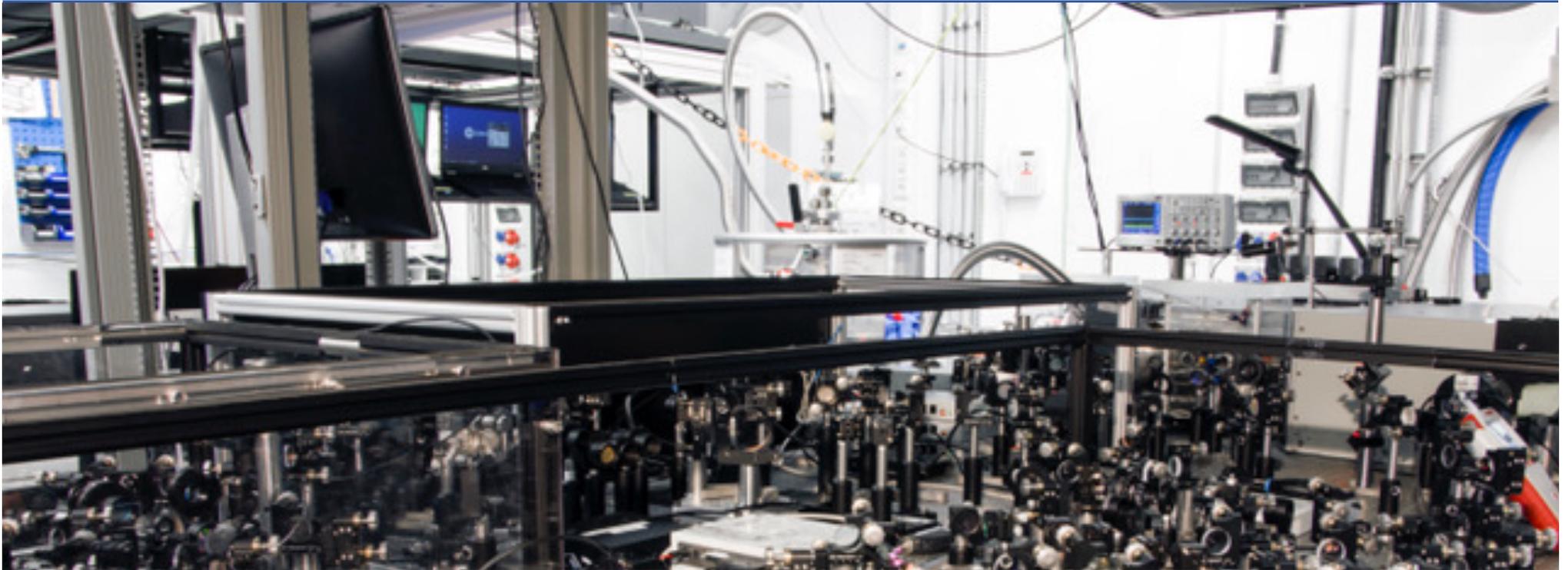
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## Modern Tabletop Optical Sources: strong fields across the spectrum



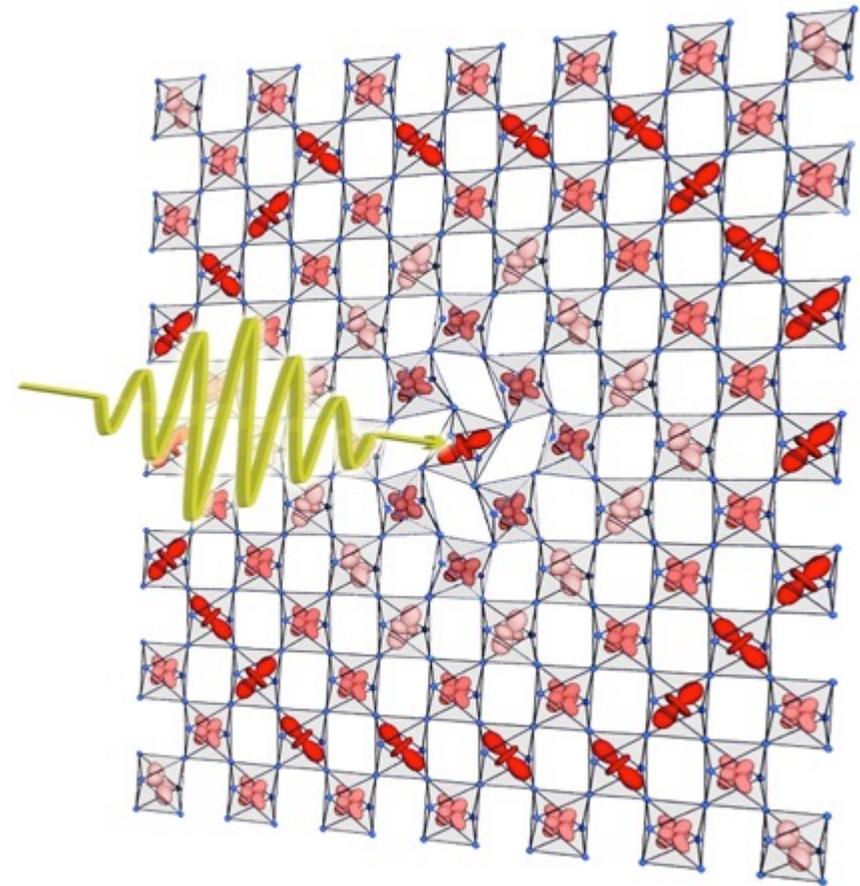
# Modern Tabletop Optical Sources: strong fields across the spectrum



# Periodically driven lattices

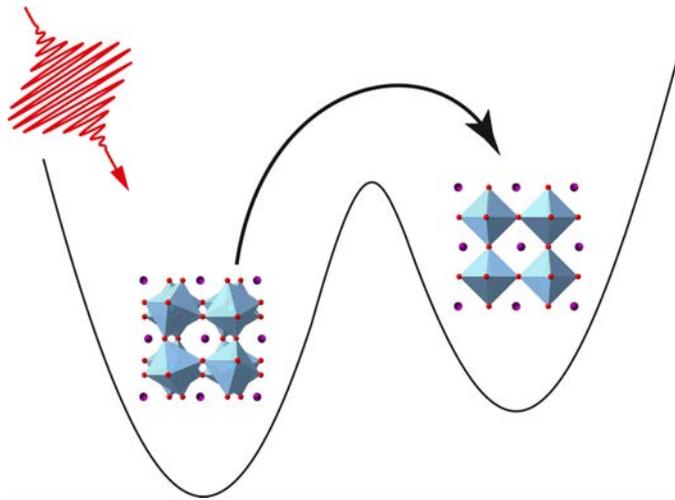
**$E \sim \text{MV/cm}$**

**Displacements  $\sim \%$**



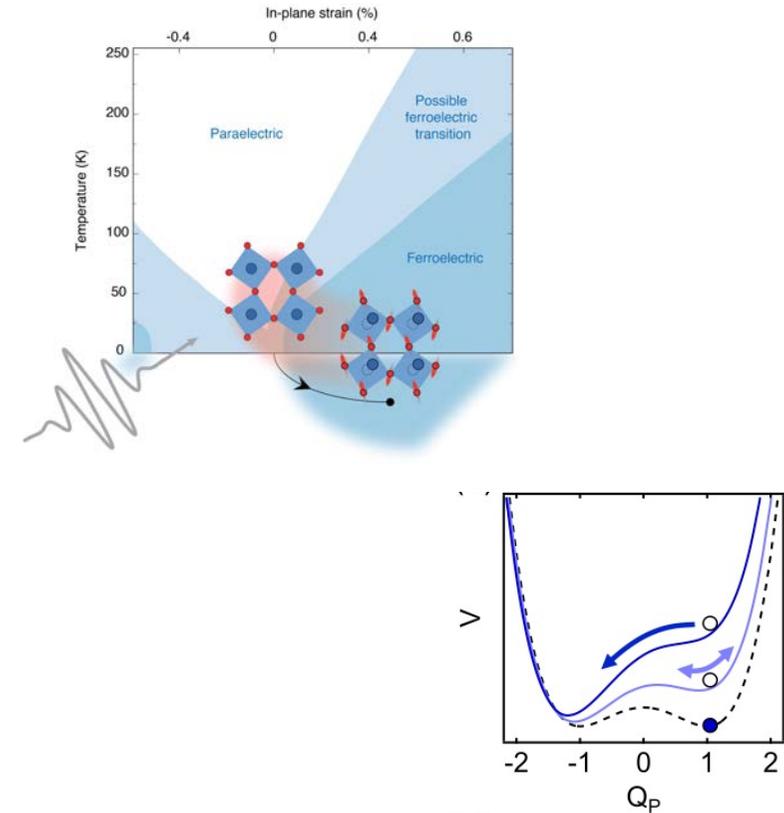
# Inducing new Crystal Structures with Light

## Hidden Phases



M. Rini et al., *Nature* 449, 72 (2007)

## Switching ferroelectricity



A. von Högen et al. *Nature* 555, 79 (2018)

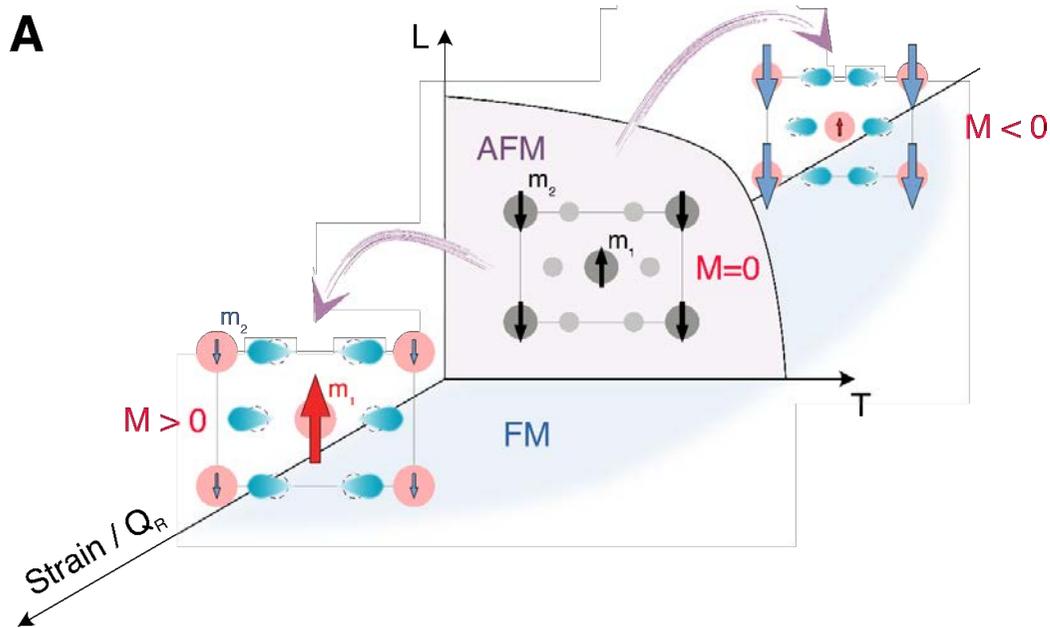
T.F. Nova et al. *Science* 364, 1075 (2019)

M. Henstridge et al. *Nature Physics* (2022)



# Controlling Magnetism and Topology

## Induced ferromagnetism

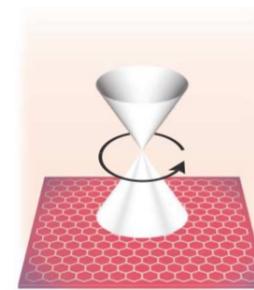


T. F. Nova et al., *Nature Physics* 13, 132 (2017)

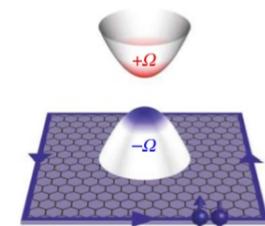
A. Disa et al., *Nature Physics* 16, 937 (2020)

A. Disa et al., *Nature* 617, 73 (2023)

## Induced Topology



Time-dependent picture



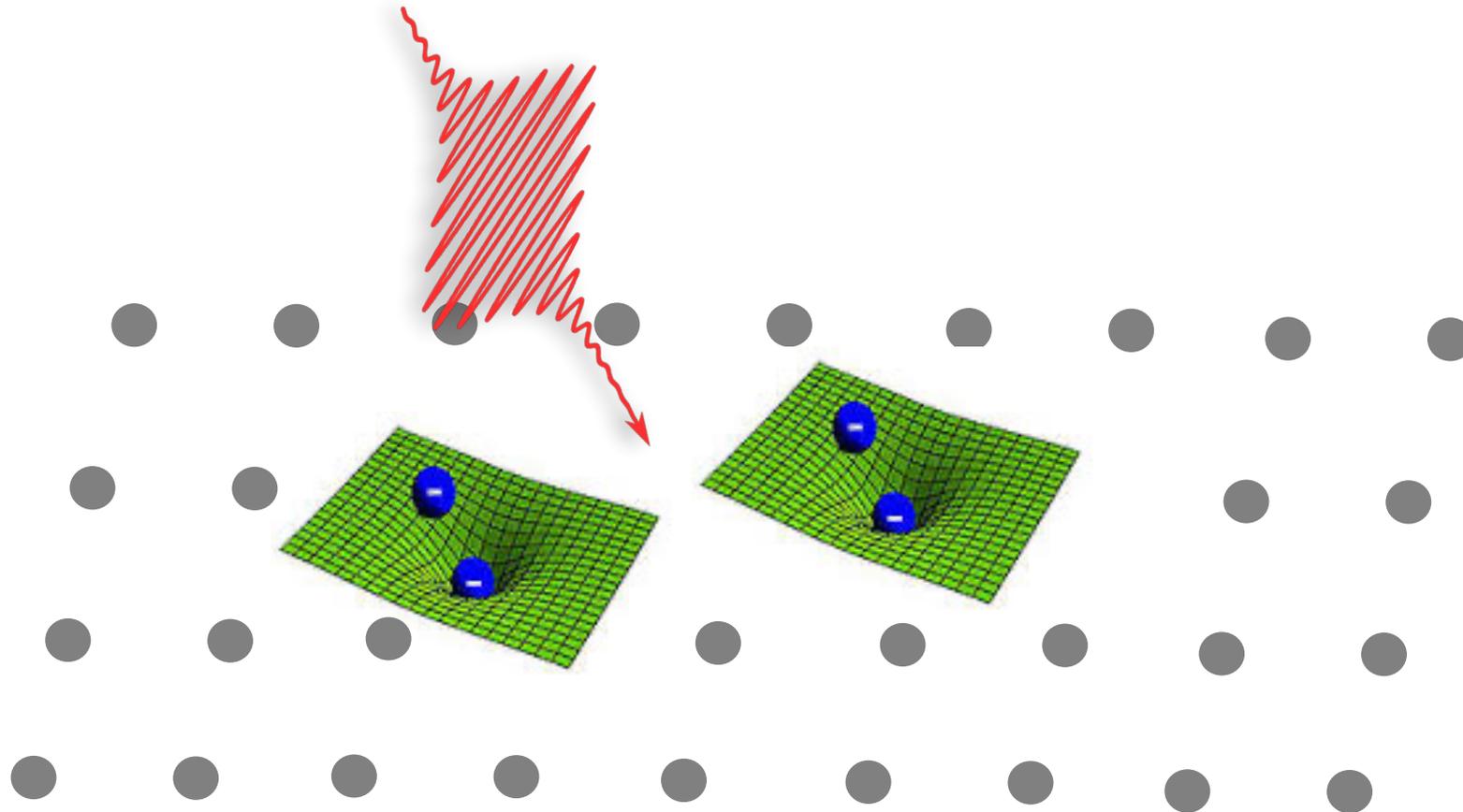
Effective Floquet picture

J. Mciver et al., *Nature Physics* 16, 38 (2020)



# Today's talk: Controlling Superconductivity

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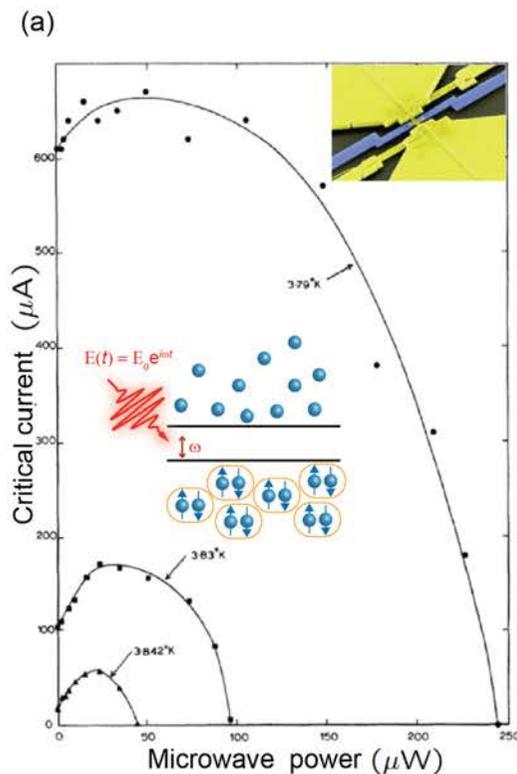
**(1) Control pairing fields**

**(2) Control “phase” coherence**

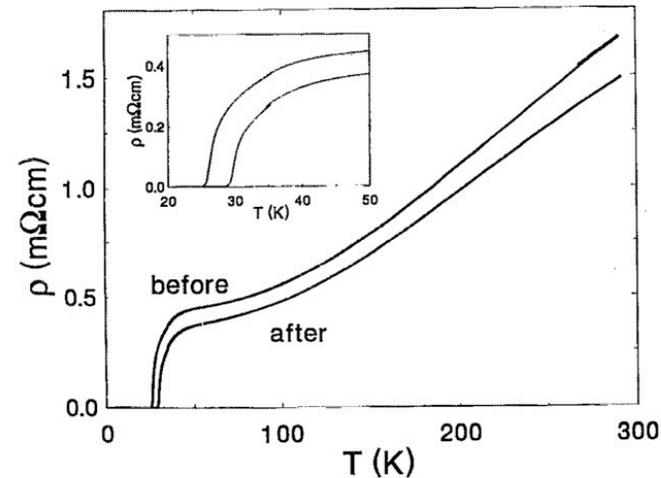


# Enhancing Superconductivity with radiation: history

## Microwave irradiation



## UV irradiation



Nieva, G. *et al. Applied Physics Letters* 60, 2159-2161, (1992).

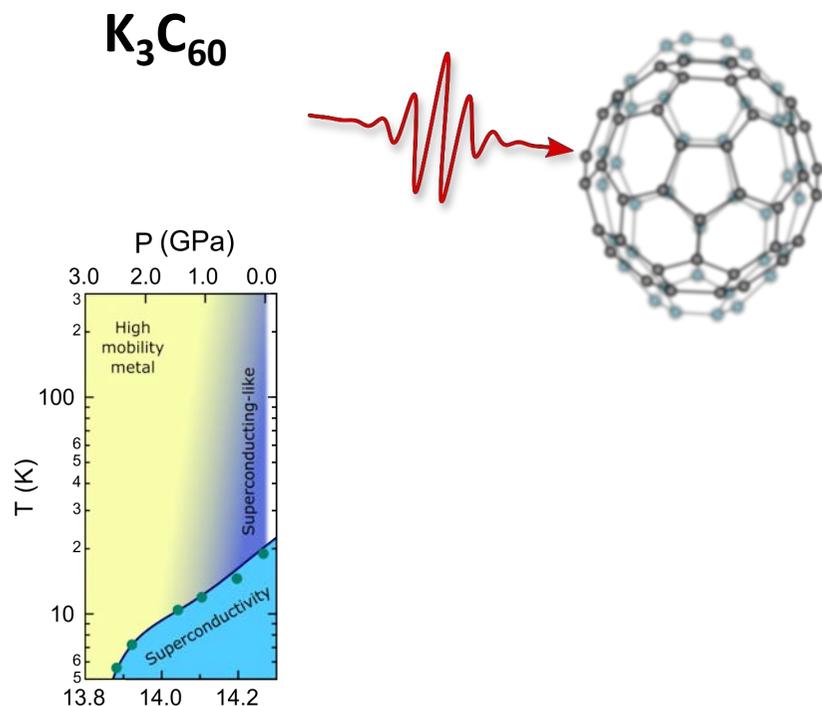
Yu, G. *et al. Physical Review B* 45, 4964-4977, (1992).

Wyatt, A. F. G., *Physical Review Letters* 16, 1166-1169, (1966).

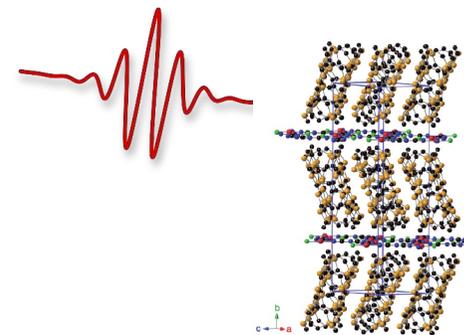
G. Eliashberg., M. *JETP Letters* 11, 114, (1970).



# Control of Superconductivity in Organics



$k\text{-ET}_2\text{Br}$

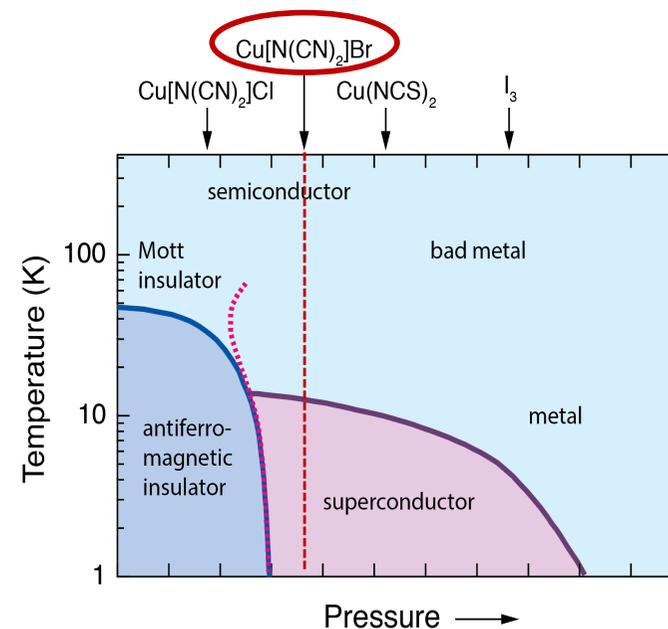


M. Mitrano et al., *Nature* 530, 461-464 (2016)

A. Cantaluppi et al., *Nature Physics* 14, 837 (2018)

M. Budden et al., *Nature Physics* 17 611 (2021)

E. Rowe et al., *Nature Physics* (2023)

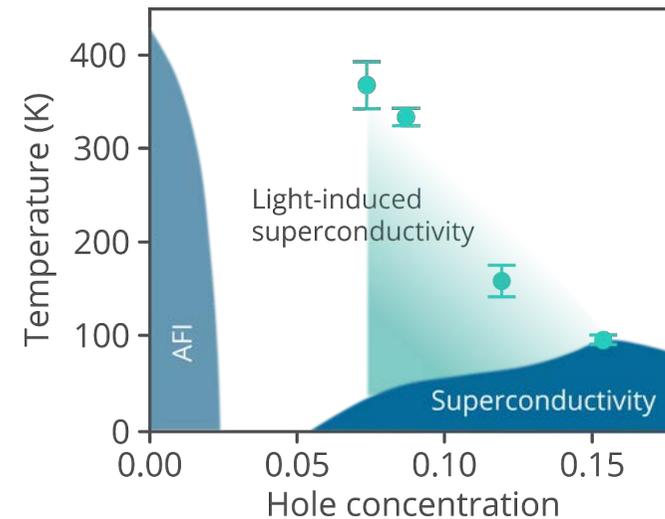
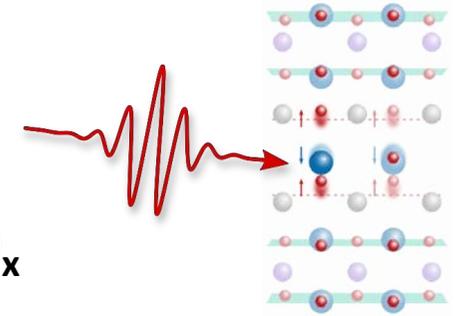
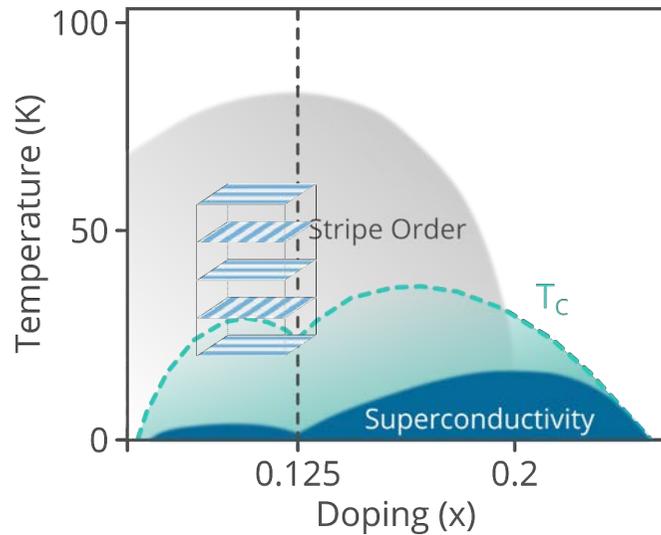
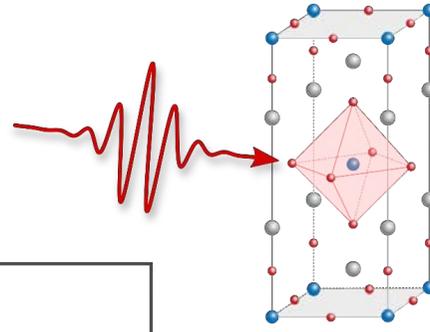


M. Buzzi et al, *Phys. Rev. X* 10, 031028 (2020)

M. Buzzi et al, *Phys. Rev. Lett* 127, 197002 (2021)



# Control of Superconductivity in Cuprates



D. Fausti et al, *Science* 331, 6014 (2011)

D. Nicoletti et al, *Phys Rev B* 90, 100503 (2014)

K. Cremin et al. *PNAS* 40, 19875 (2019)

M. Nishida et al. *ArXiv*2303.01961 (2023)

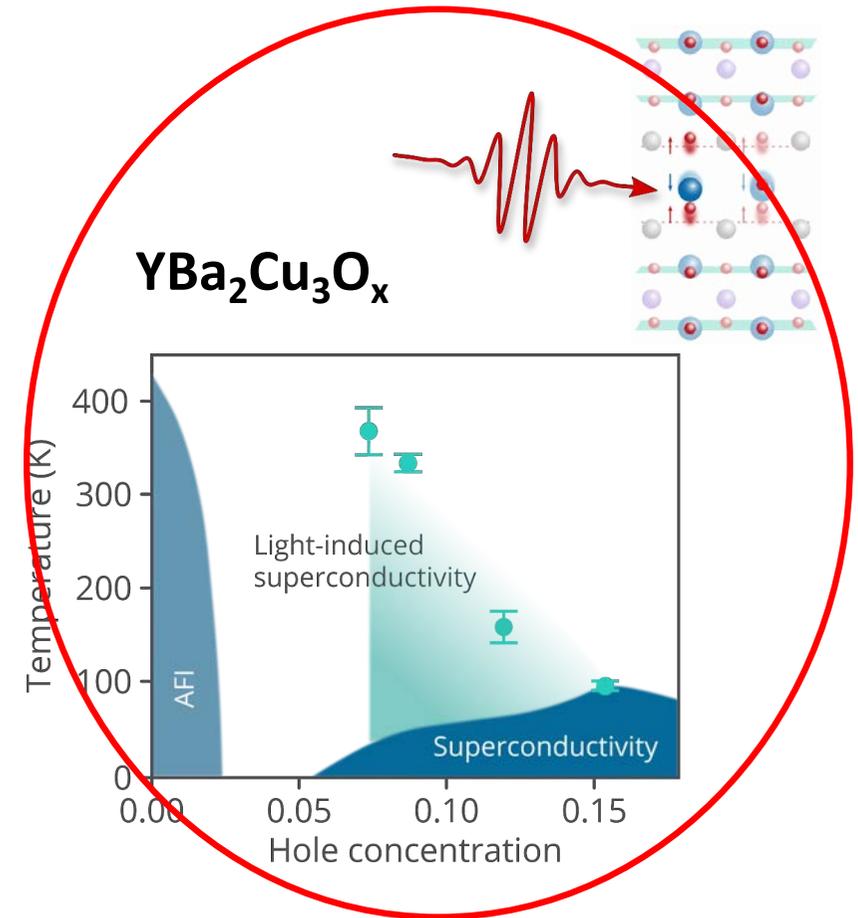
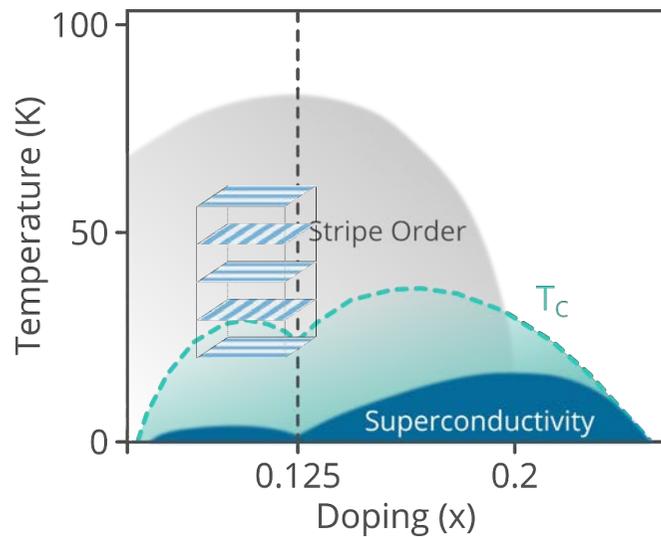
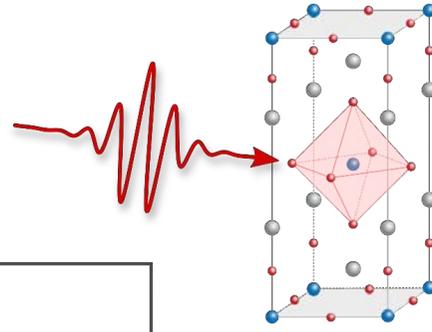
W. Hu et al, *Nature Materials* 13, 705 (2014)

B. Liu et al, *Phys. Rev. X* 10, 011053 (2020)

A. Von Hoegen et al. *Phys. Rev. X* 12, 031008 (2022)



# Control of Superconductivity in Cuprates



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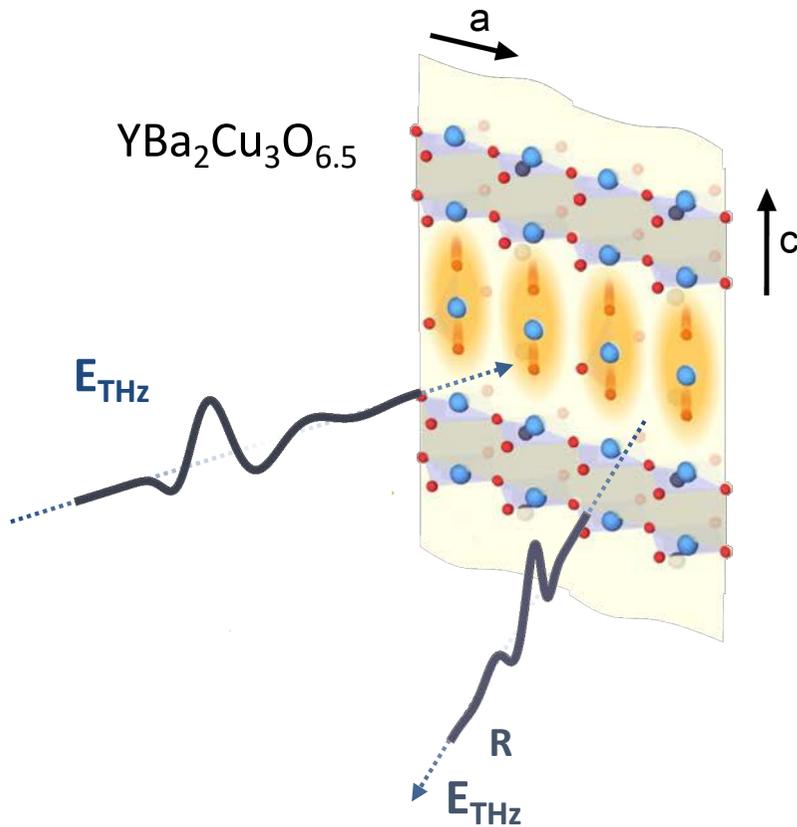
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B. Liu et al, *Phys. Rev. X* 10, 011053 (2020)

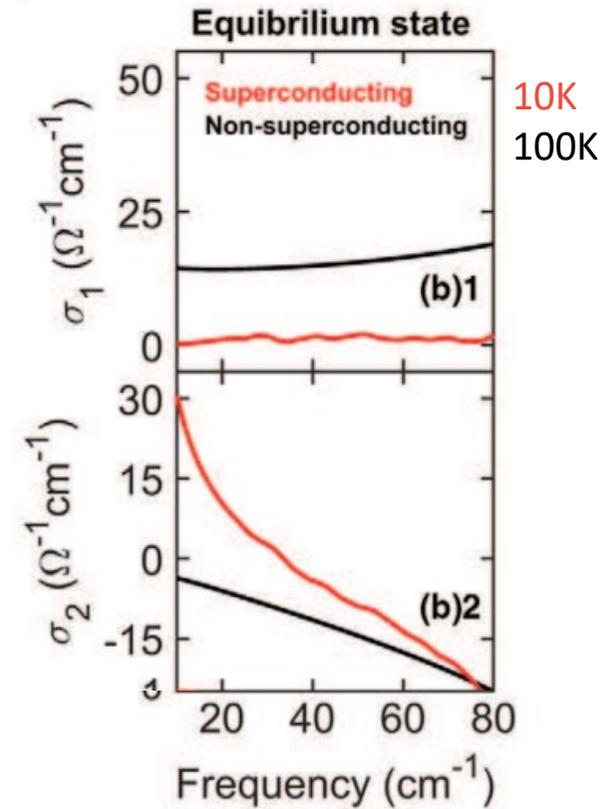
A. Von Hoegen et al. *Phys. Rev. X* 12, 031008 (2022)



# YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub>: signatures of equilibrium superconductivity

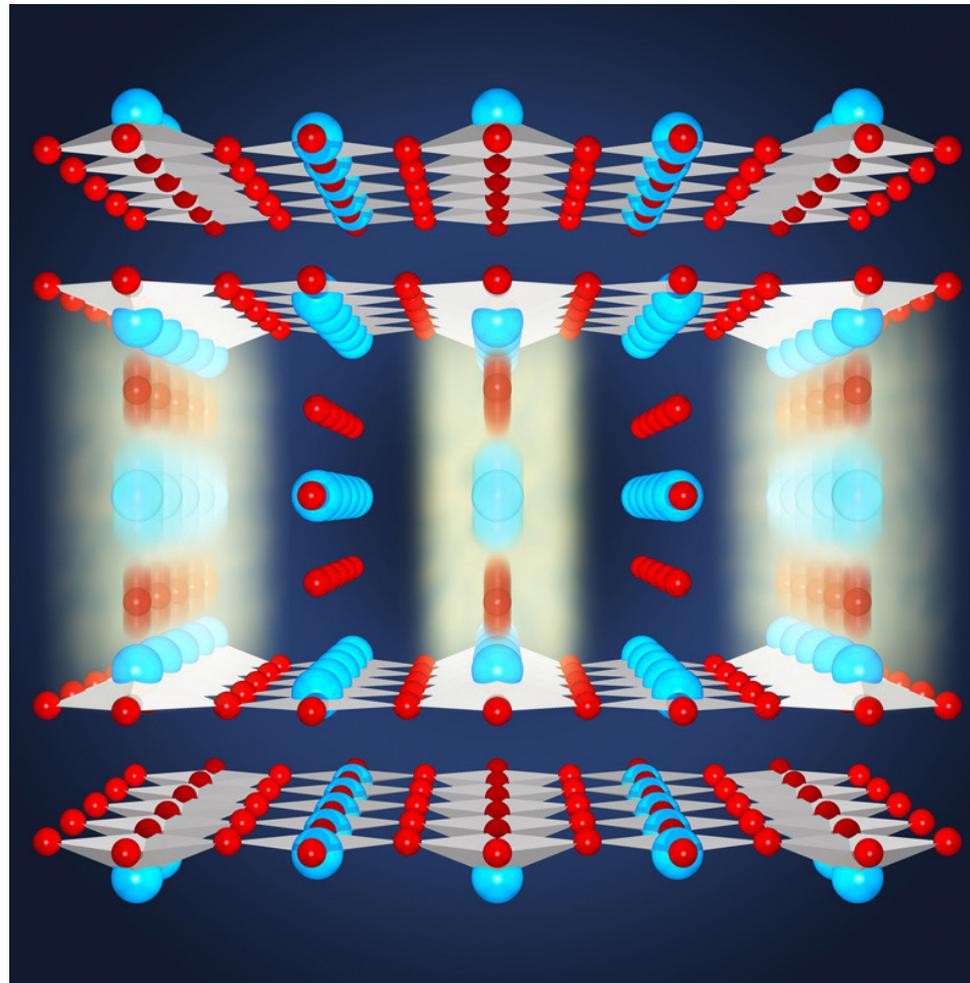


B. LIU et al. PHYS. REV. X 10, 011053 (2020)



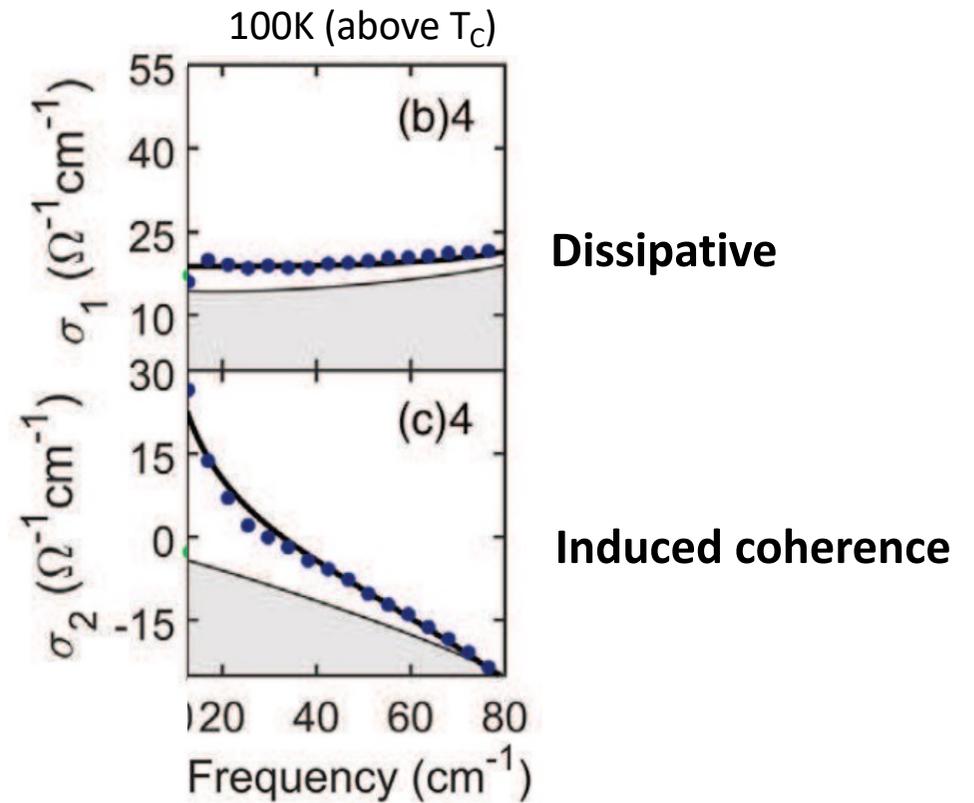
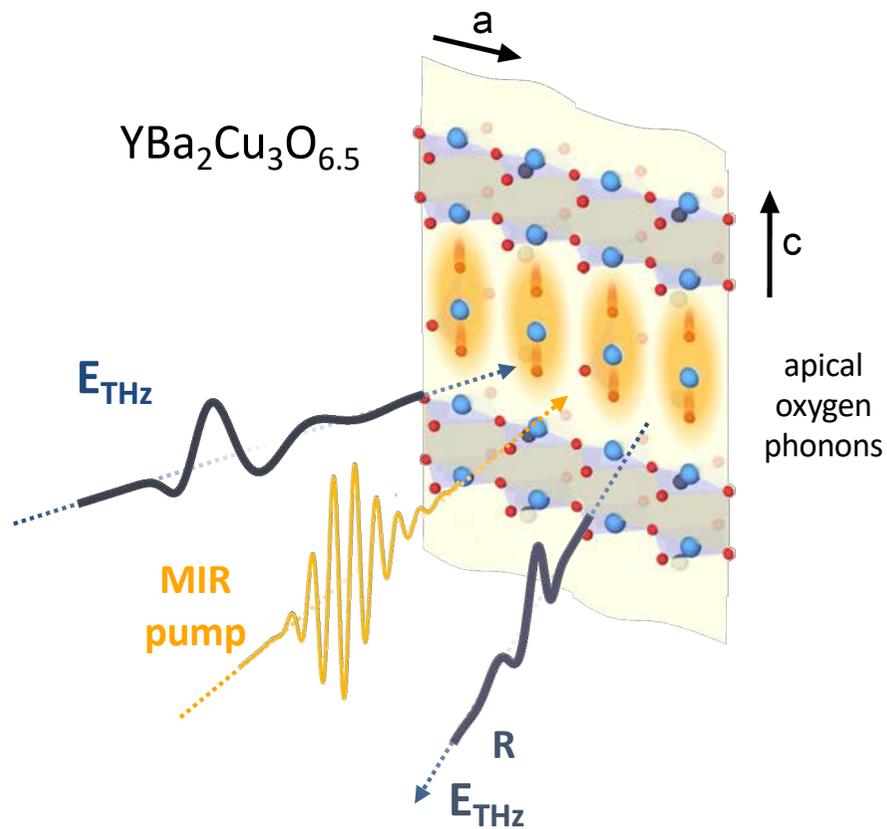
# Driven $\text{YBa}_2\text{Cu}_3\text{O}_x$

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# YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub> : signatures of induced coherent transport

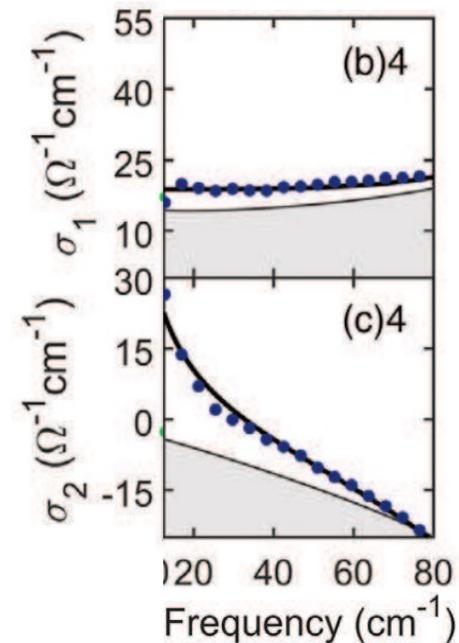
B. LIU et al. PHYS. REV. X 10, 011053 (2020)



# YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub>: signatures of induced coherent transport

$$\sigma_1(\omega) + i\sigma_2(\omega) = \frac{\sigma_0}{1 - i\omega\tau}$$

**Figure of merit –  
Extrapolated DC resistivity**



**Dissipative**

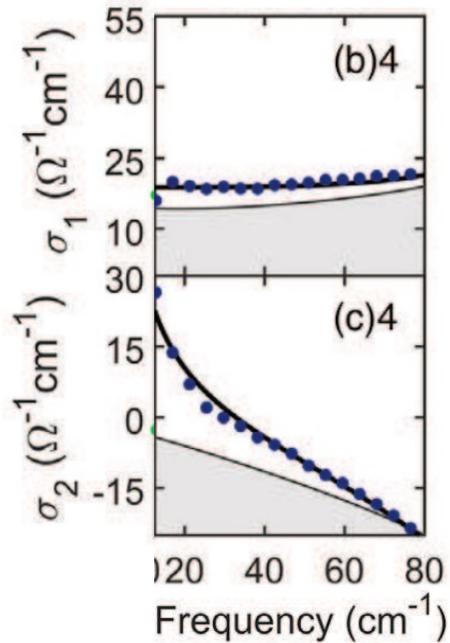
**Induced coherence**

$$\frac{1}{\rho_0} = \lim_{\omega \rightarrow 0} \sigma_1(\omega)$$



# YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub>: signatures of induced coherent transport

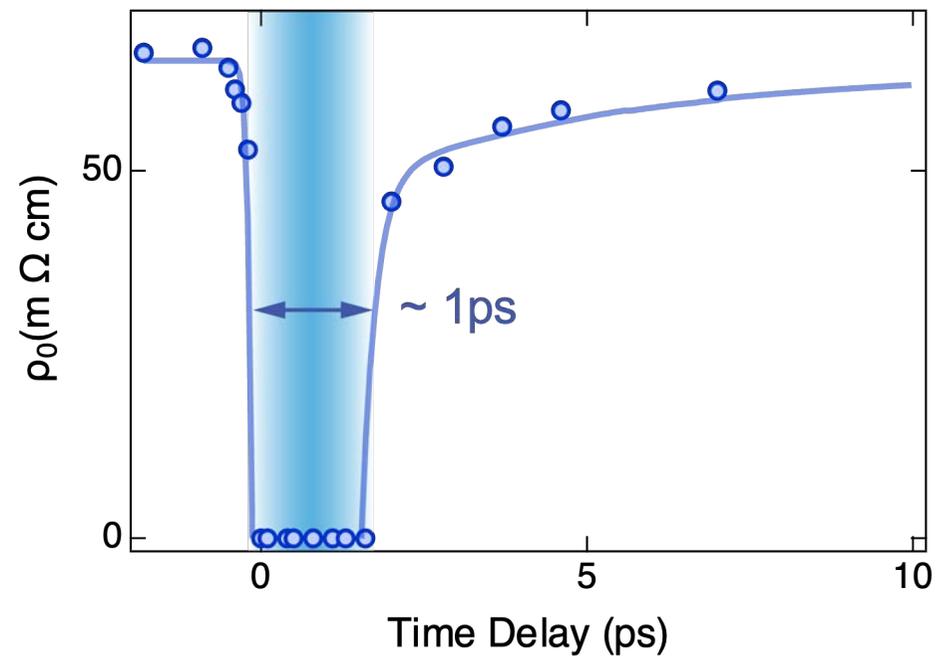
$$\sigma_1(\omega) + i\sigma_2(\omega) = \frac{\sigma_0}{1 - i\omega\tau} \quad \frac{1}{\rho_0} = \lim_{\omega \rightarrow 0} \sigma_1(\omega)$$



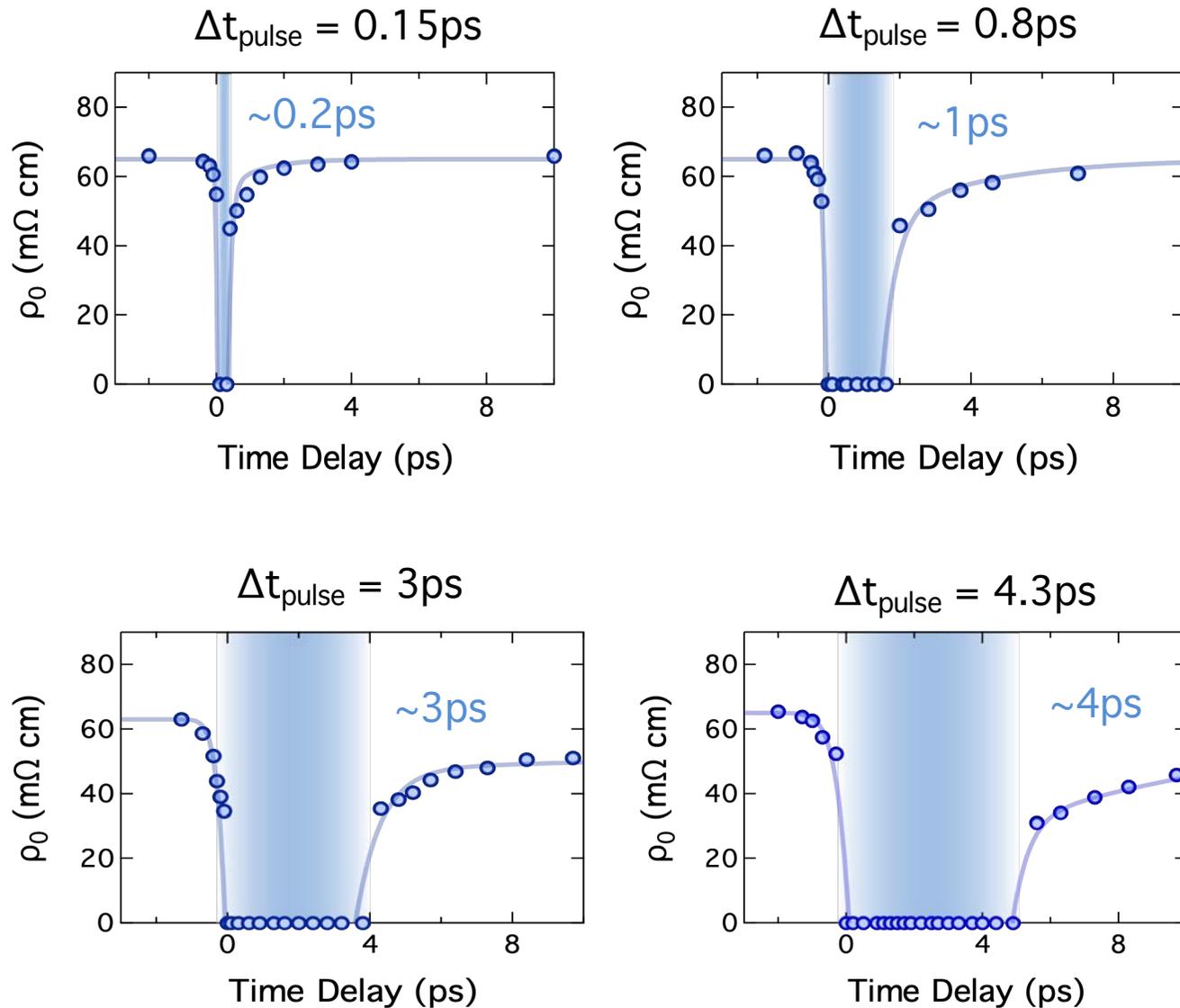
**Dissipative**

**Induced coherence**

**Extrapolated DC resistivity**



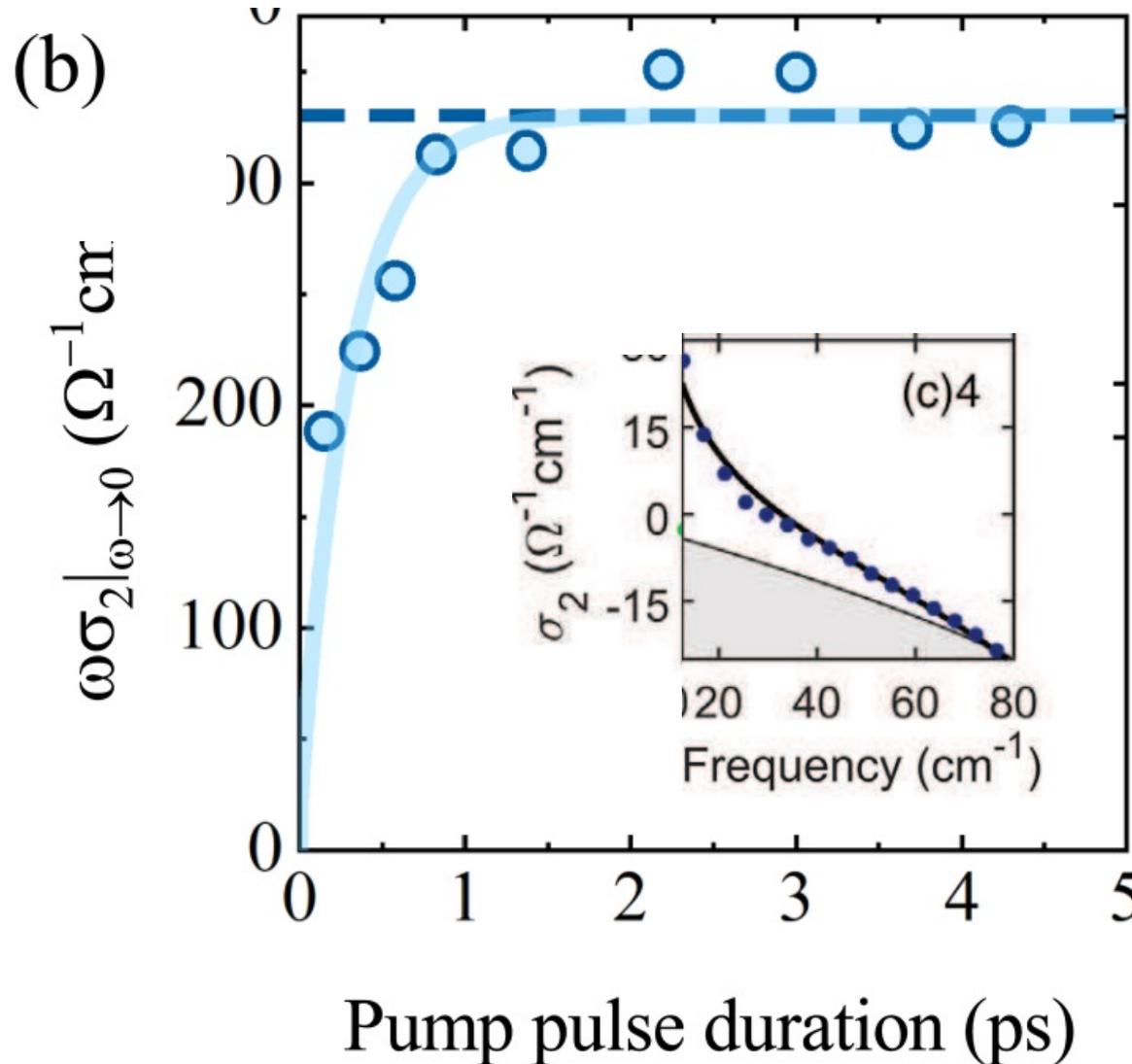
# $\rho_0$ vs. time for four different pulse durations



A. Ribak et al. *Phys. Rev. B* 107, 104508 (2023)

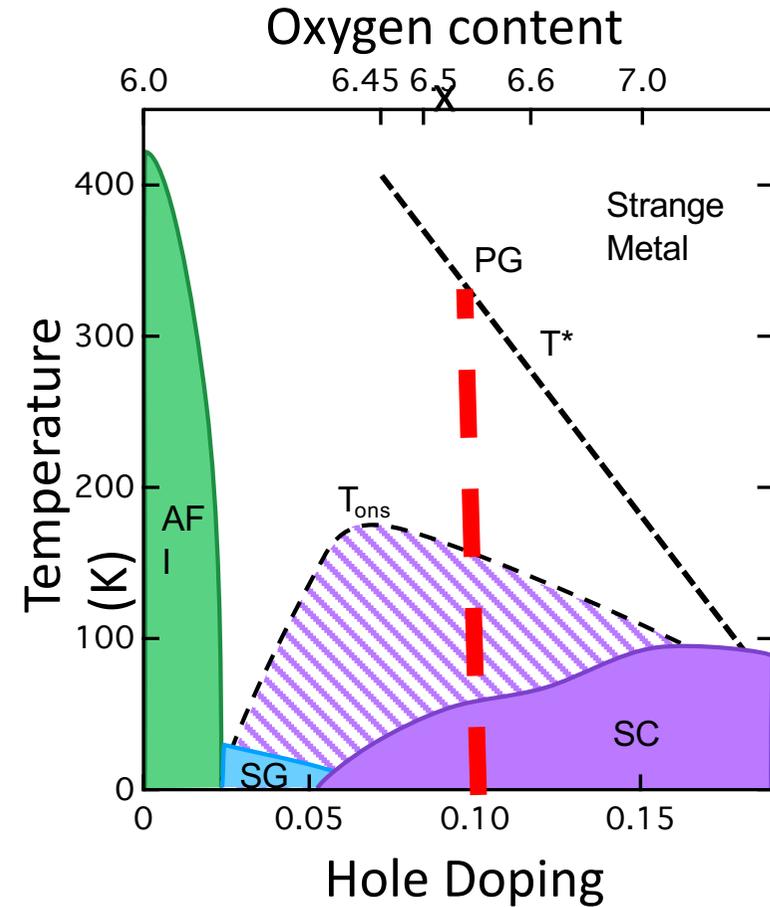
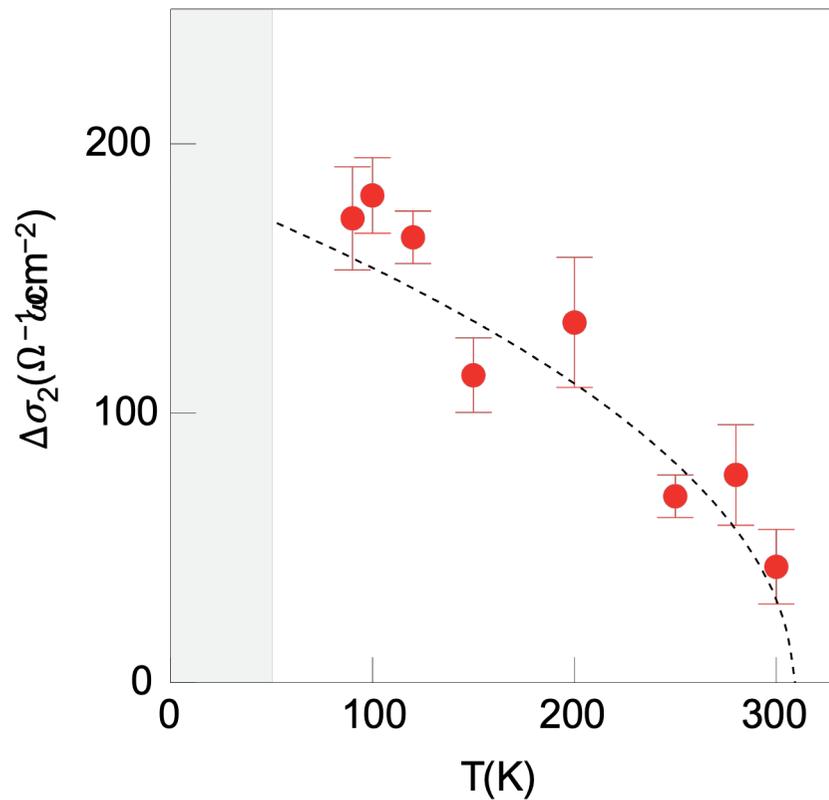


# YBa<sub>2</sub>Cu<sub>3</sub>O<sub>x</sub>: density of Cooper pairs



Zero temperature  
superfluid density

# Up to very high temperatures: pseudogap scale



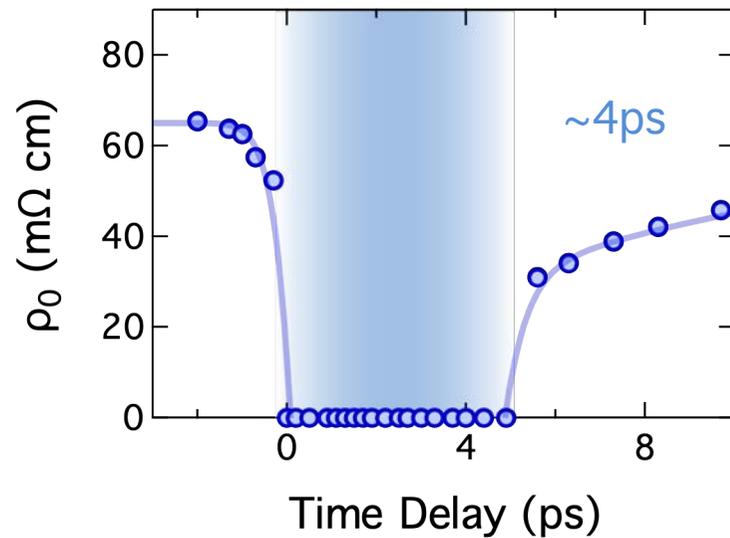
W. Hu et al., Nat. Mater. 13, 705 (2014)

S. Kaiser et al., Phys. Rev. B 89, 184516 (2014)

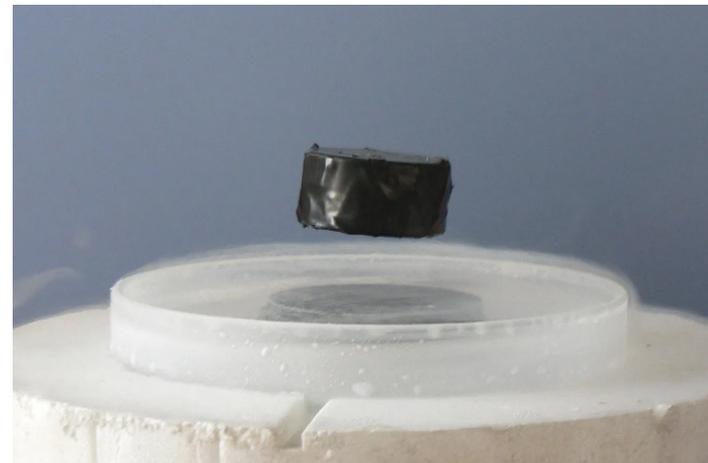


# Does this state also expel a magnetic field ?

Dissipation-less transport

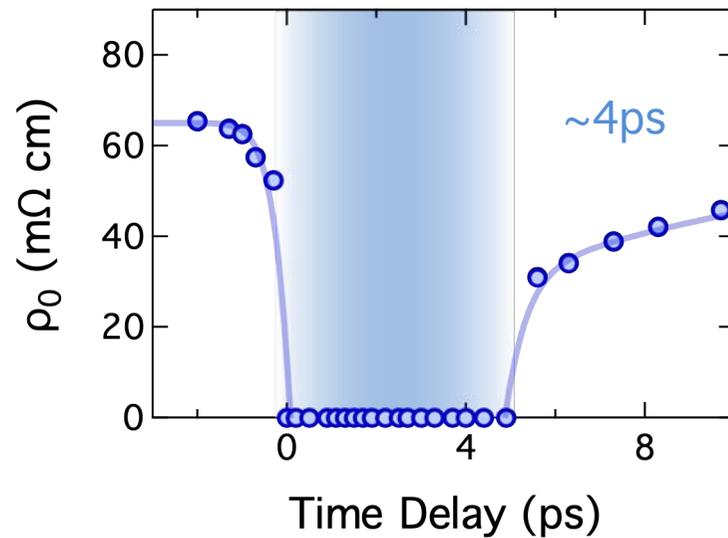


Meissner effect ?

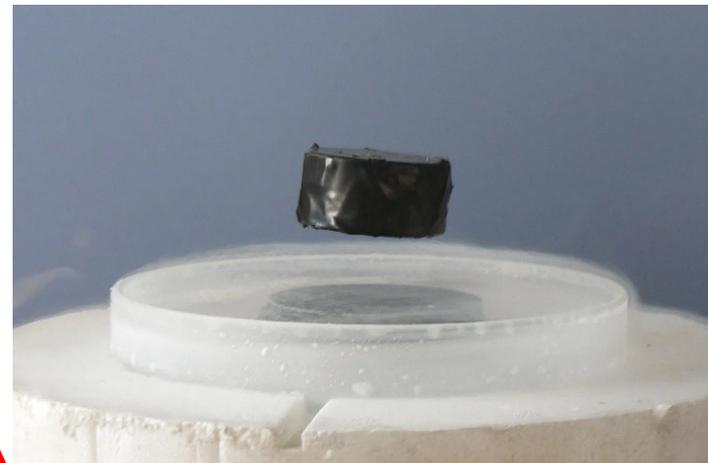


# Does this state also expel a magnetic field ?

Dissipation-less transport



Meissner effect ?



# People

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Giovanni De Vecchi



Sebastian Fava



Michele Buzzi



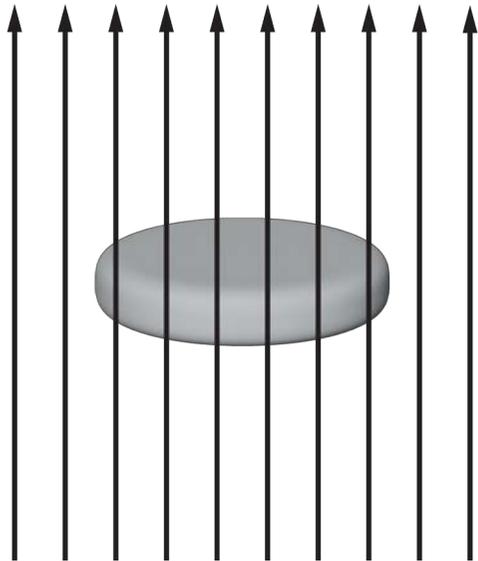
Gregor Jotzu

YBCO samples:  
**Bernhard Keimer &  
group**

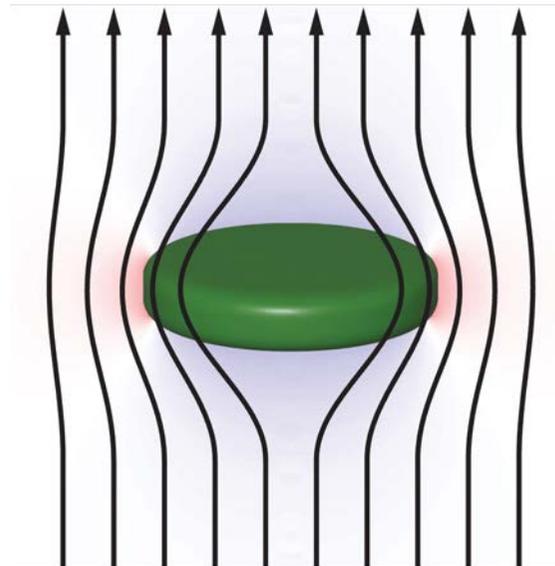


# Meissner Effect

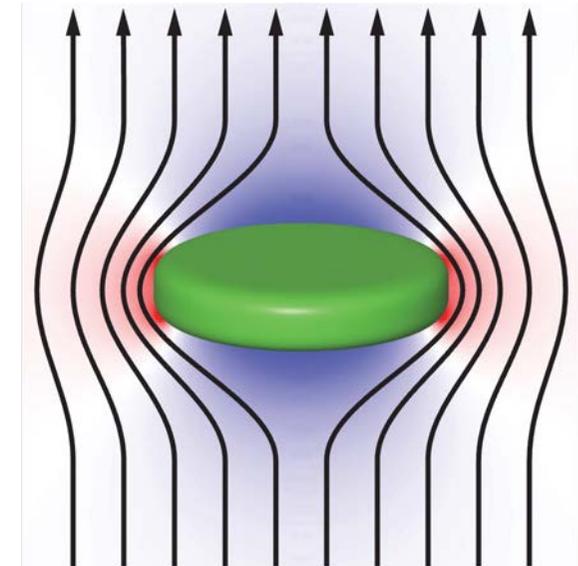
Initial Metallic State



Transient State

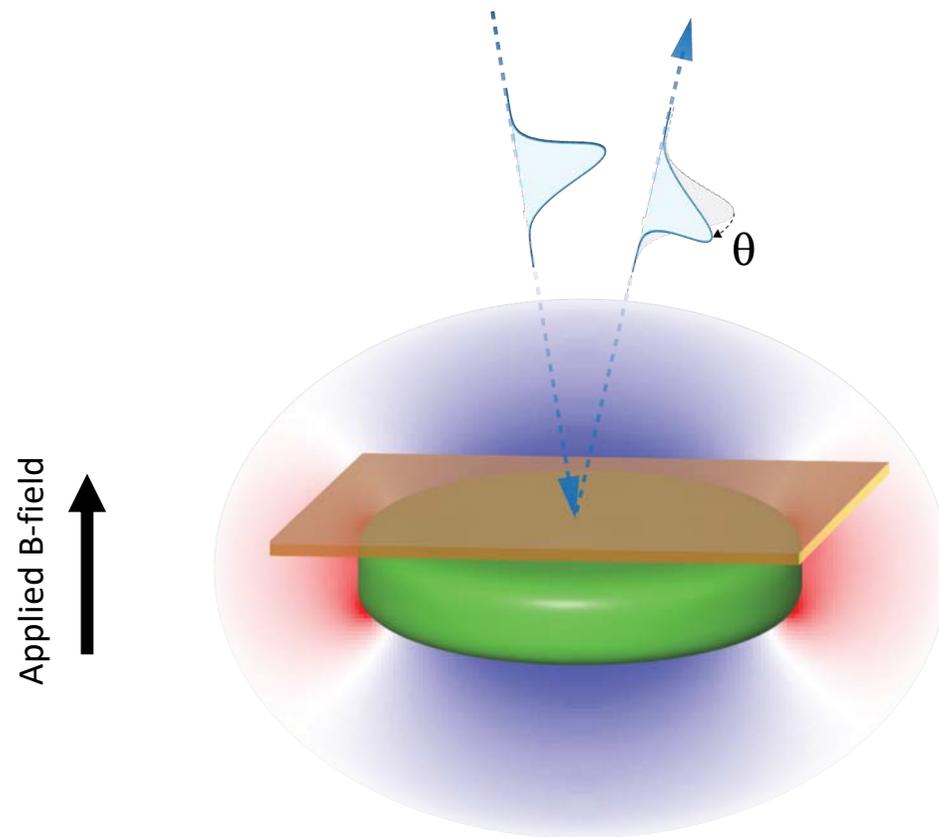


Superconducting State

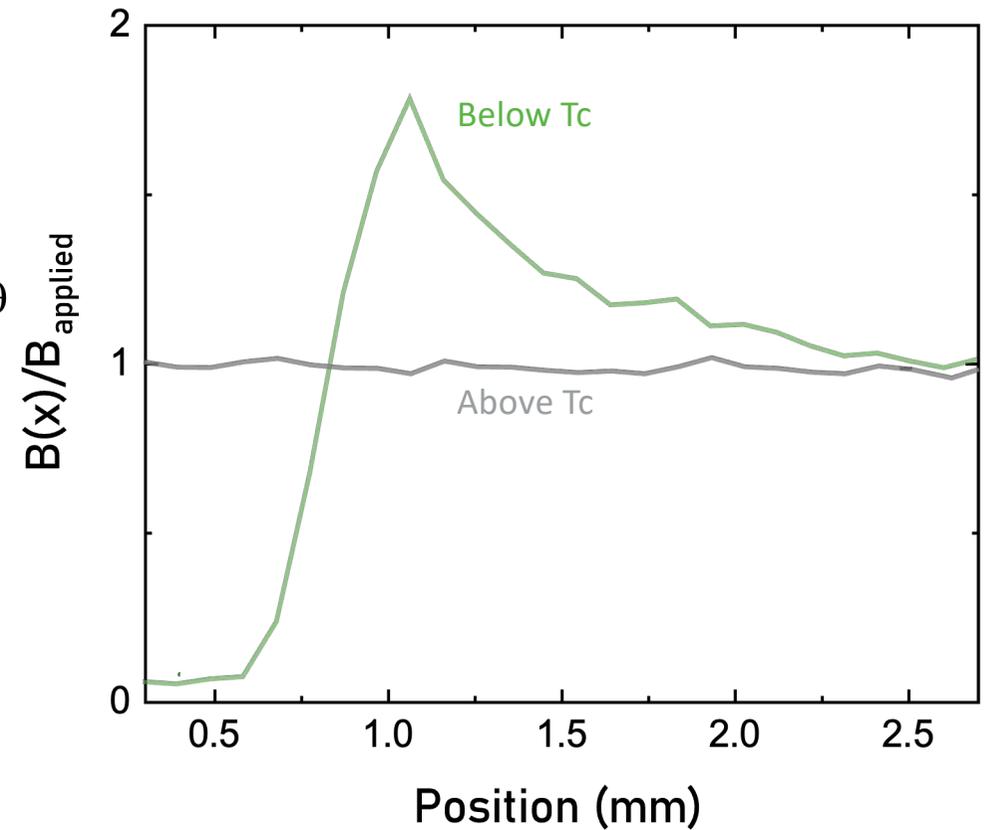
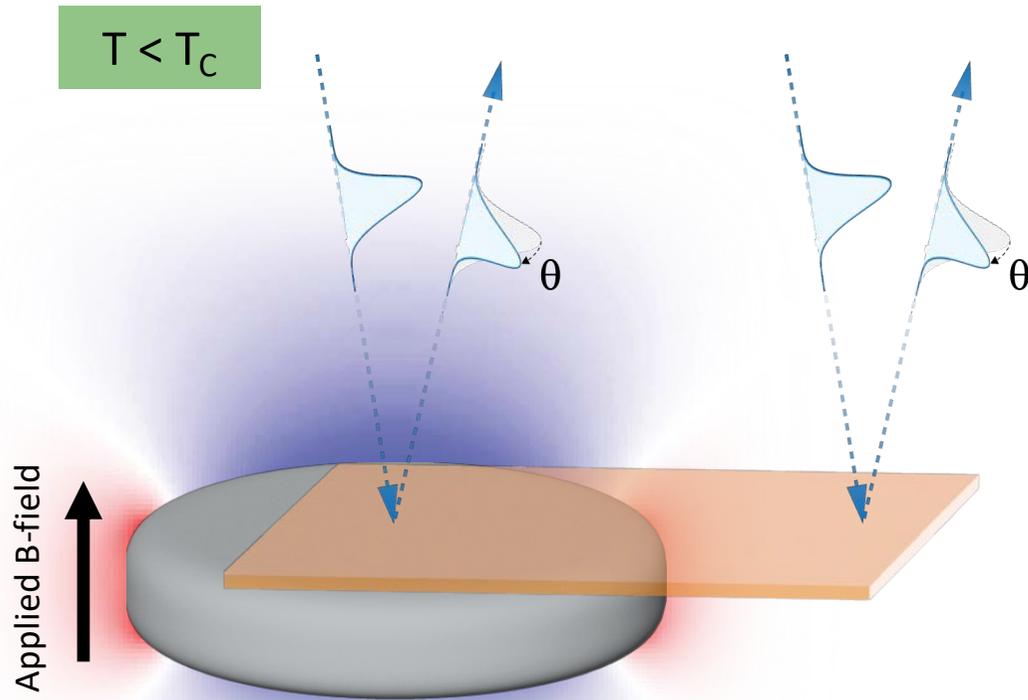


# Ultrafast Faraday Magnetometry

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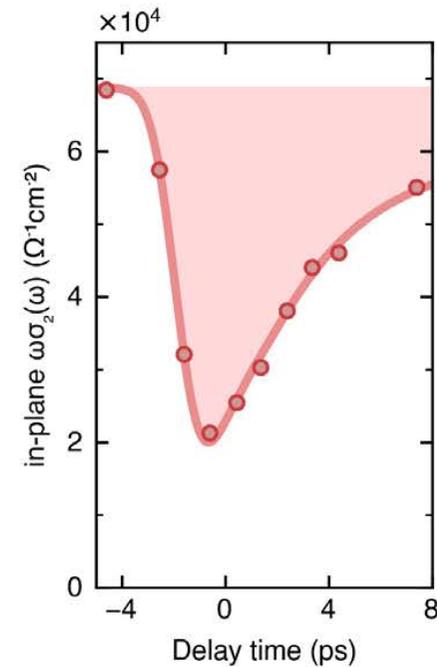
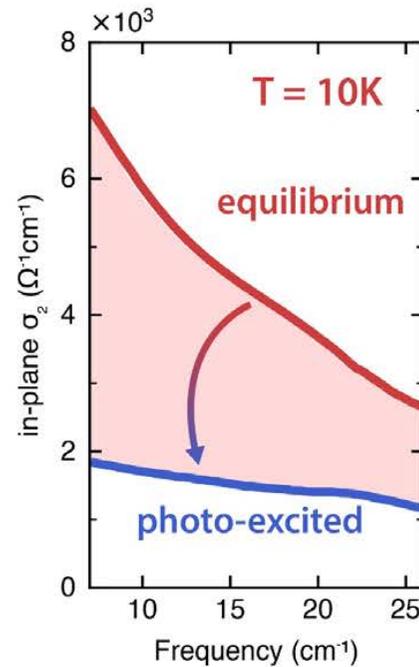
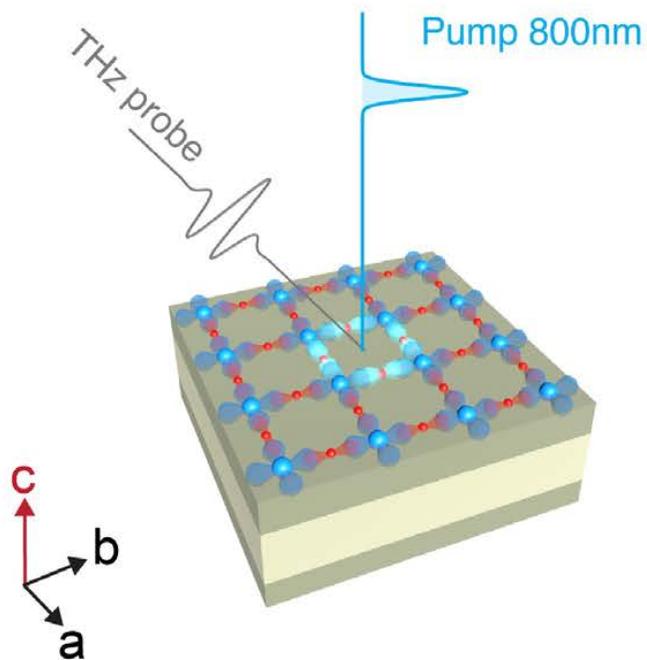


# Calibration 1: Static B-Field Expulsion



# Calibration 2: Disruption

## Disruption ( $T < T_C$ )

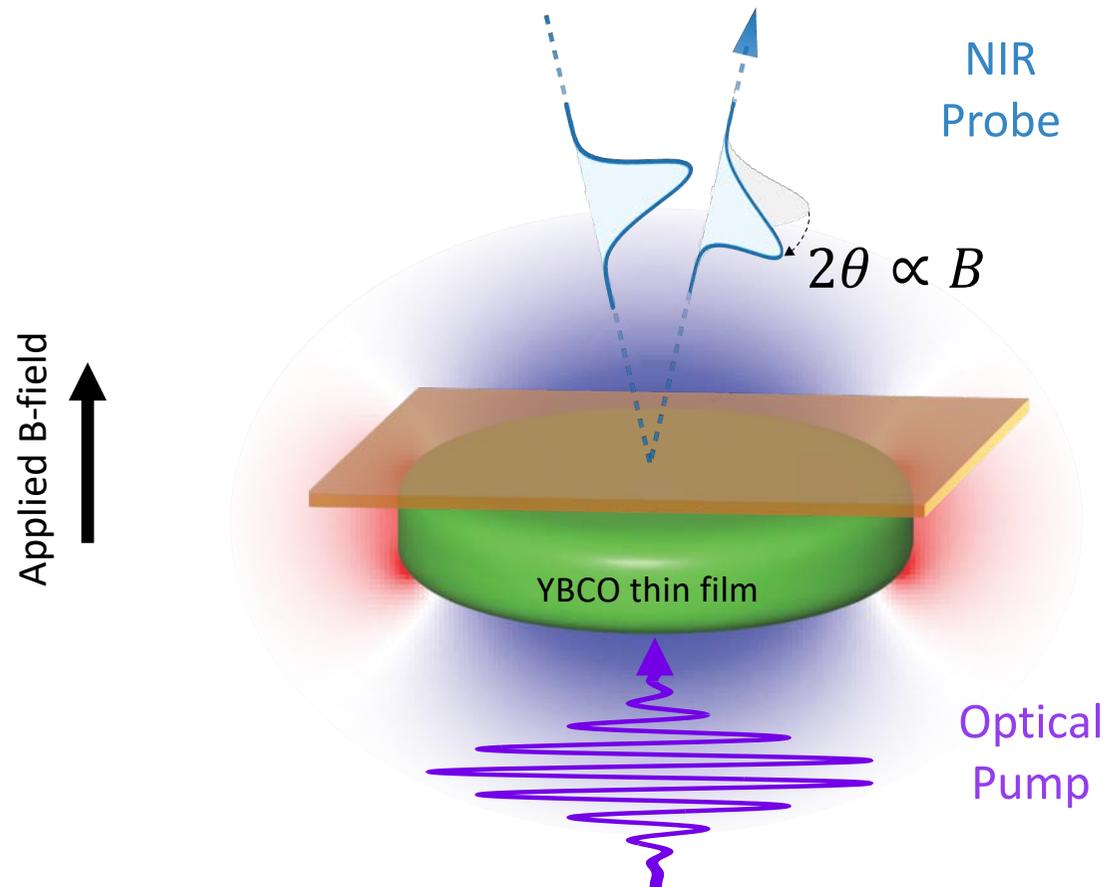


Averitt et al. PRB 2001



# Calibration 2: disruption of Superconductivity

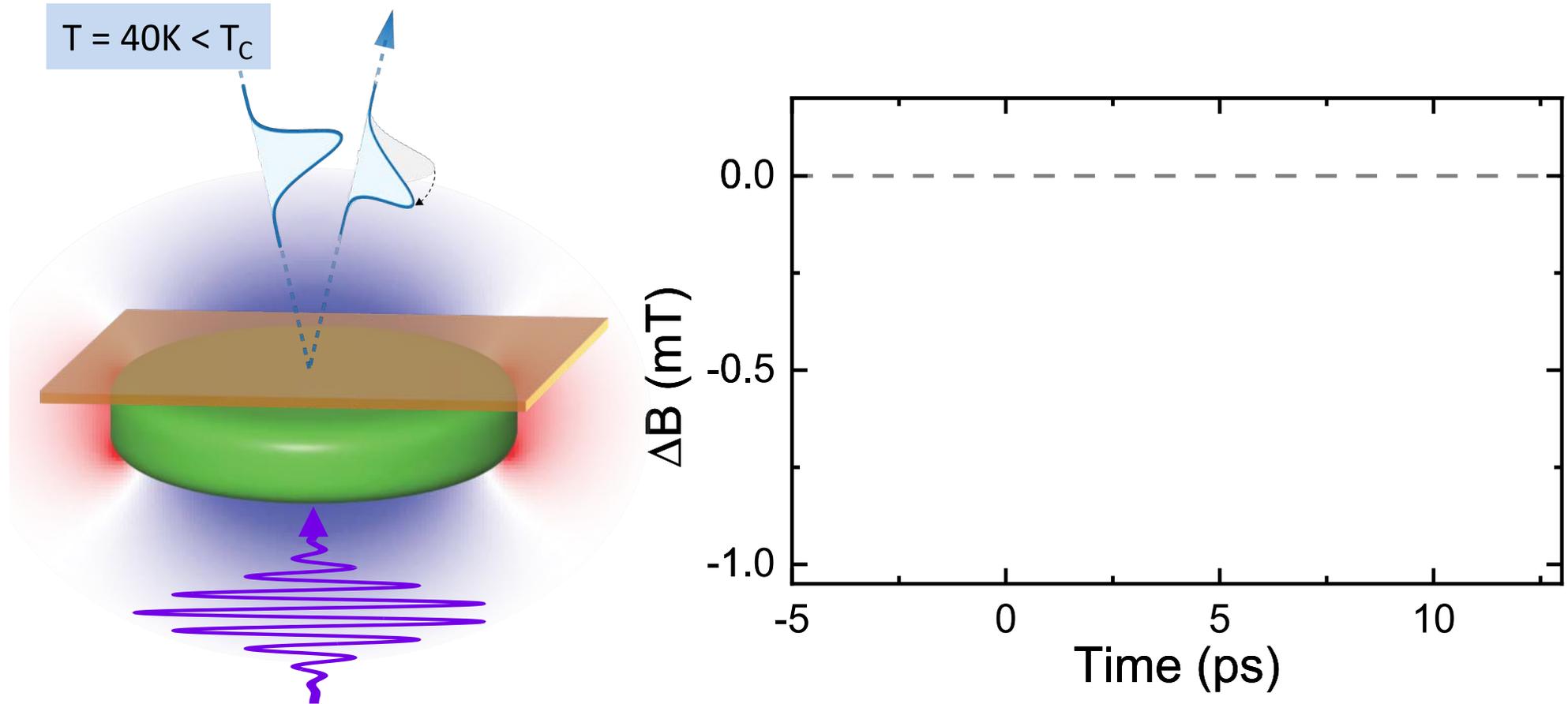
$$T = 40\text{K} < T_C$$



Giovanni De Vecchi

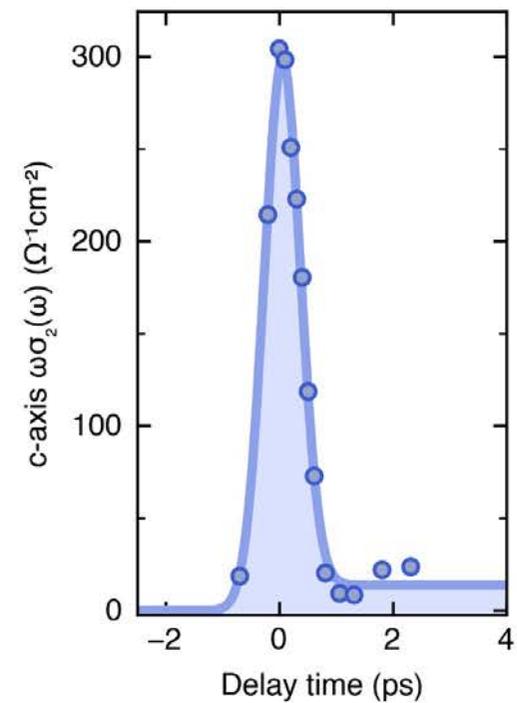
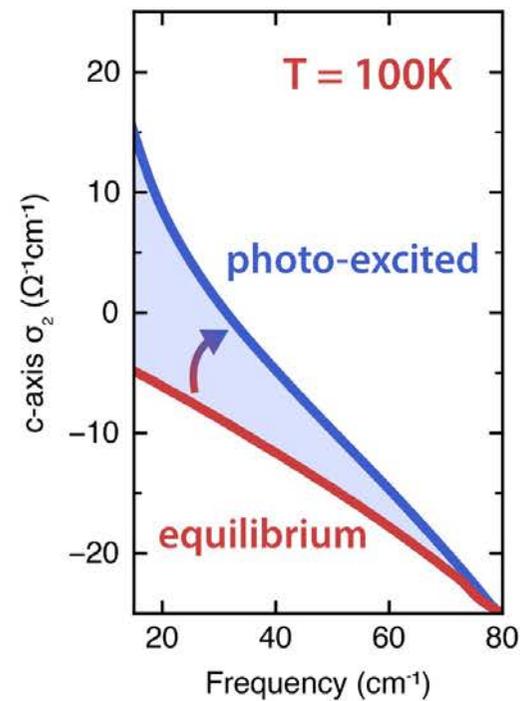
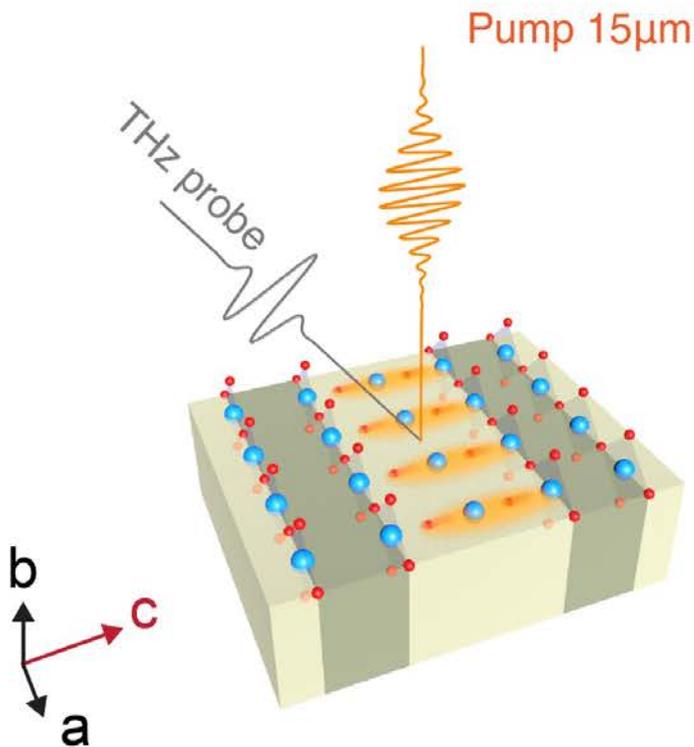


# Dynamics: Superconductor to Metal

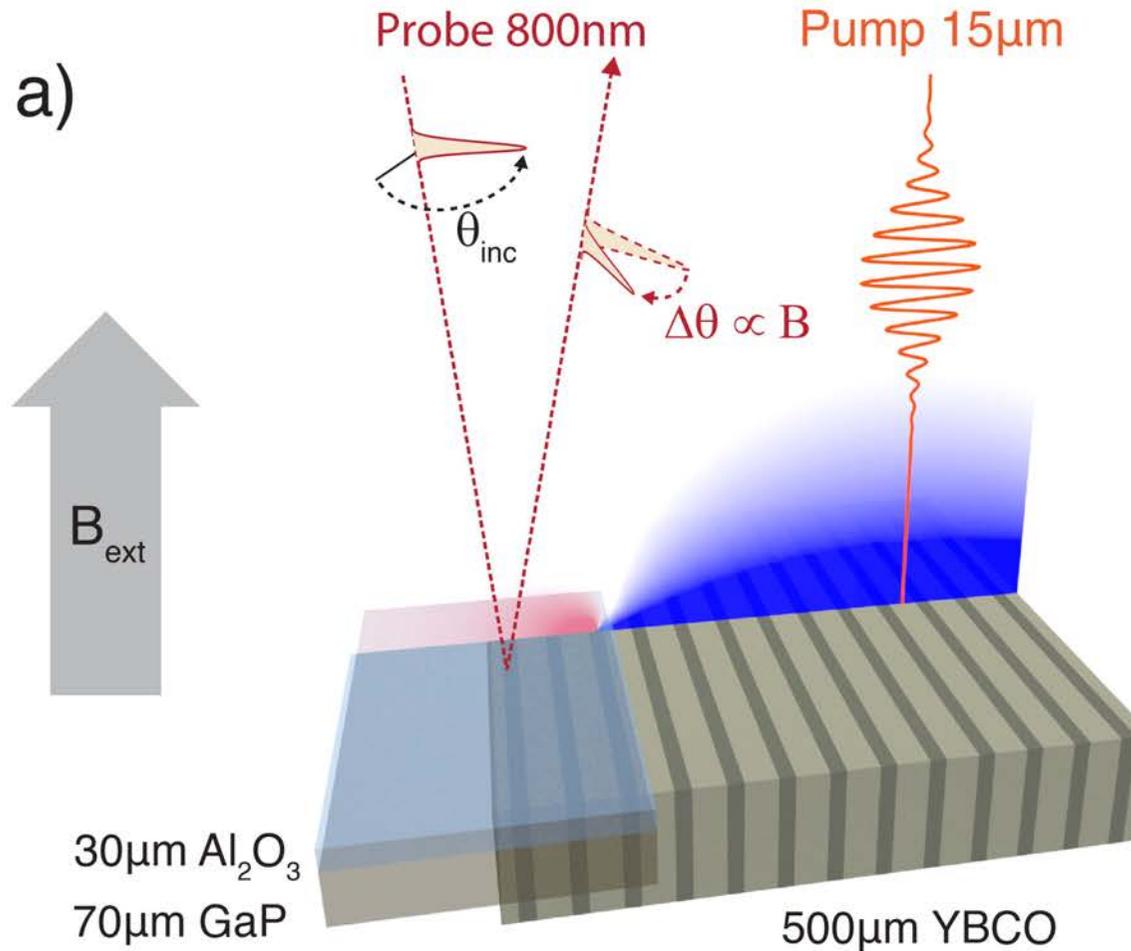


# Enhancement of Superconductivity YBCO<sub>6.48</sub>

## Enhancement ( $T > T_C$ )

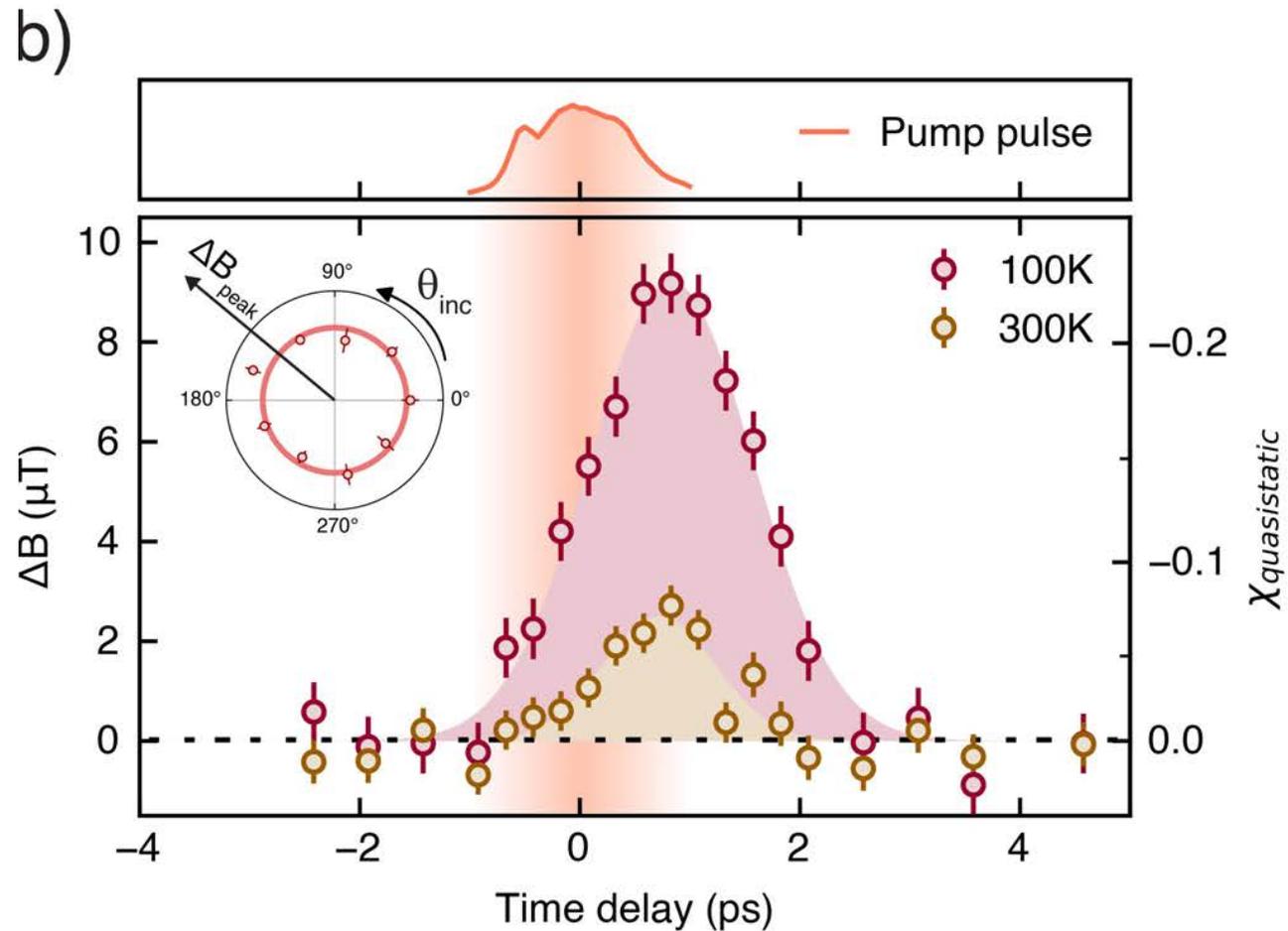


# Ultrafast Meissner Effect



Sebastian Fava

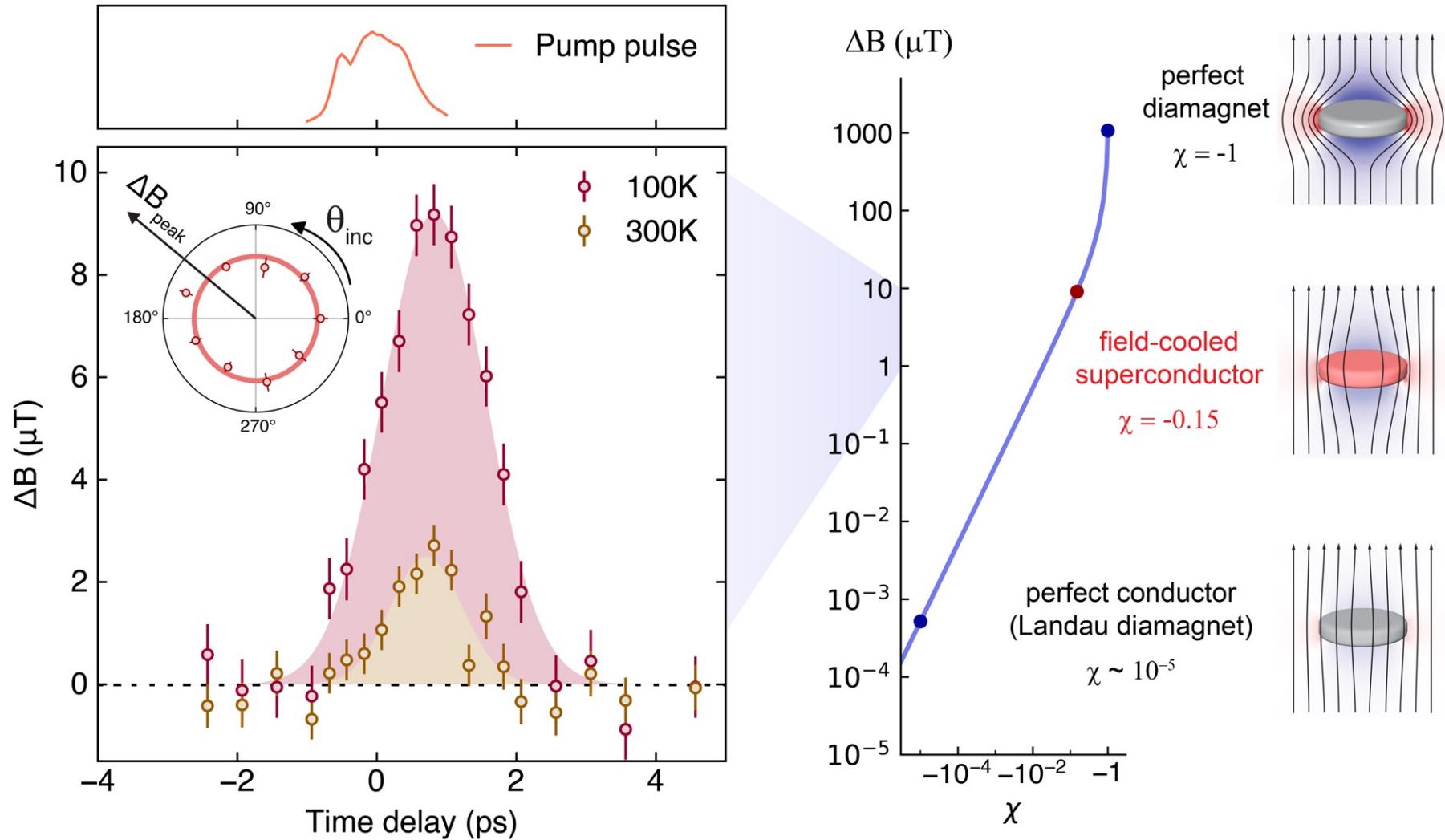
# Ultrafast Meissner Effect



S. Fava, G. DeVecchi, G. Jotzu, M. Buzzi et al. forthcoming



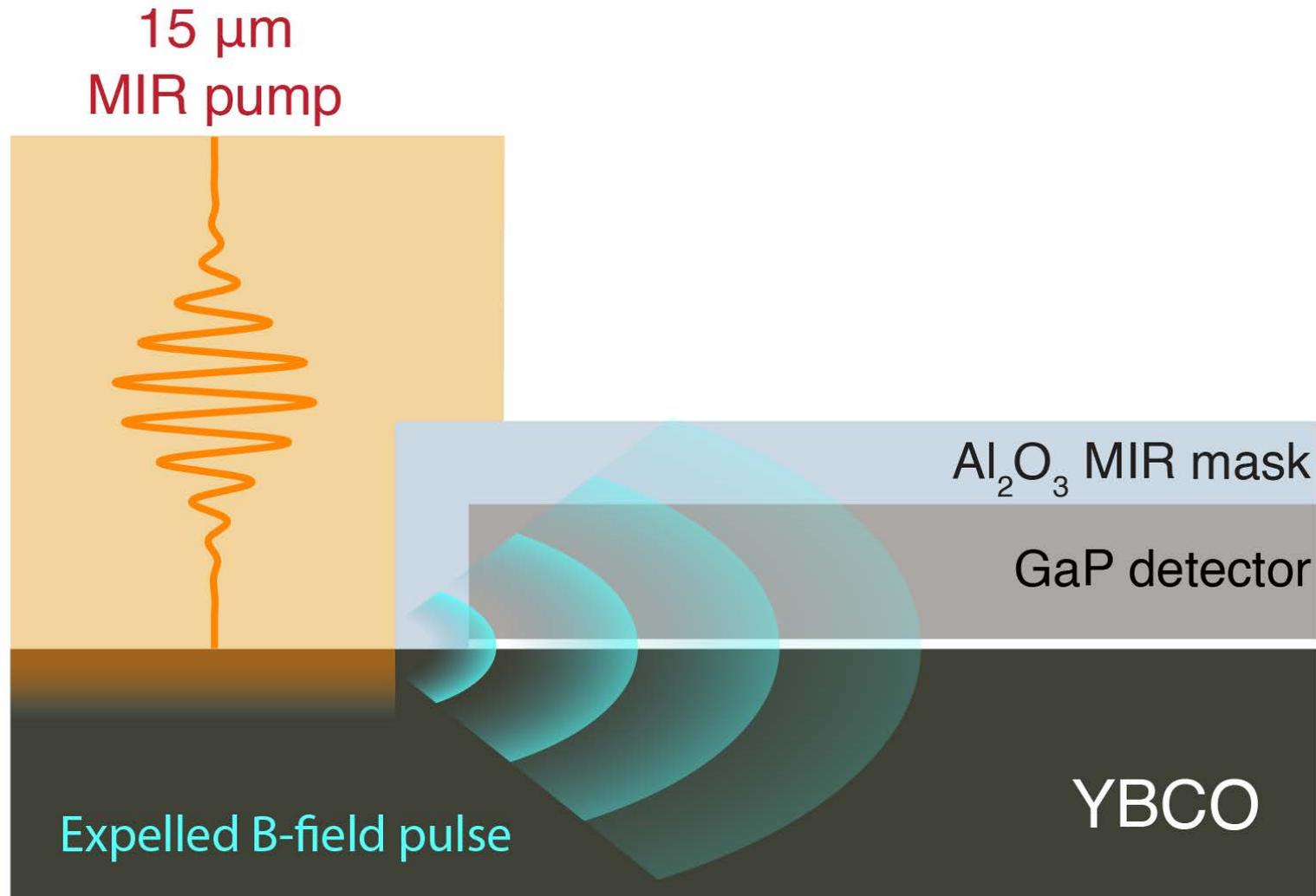
# A colossal diamagnetic response



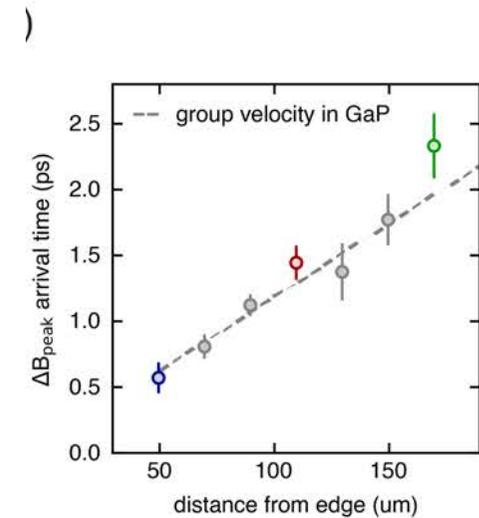
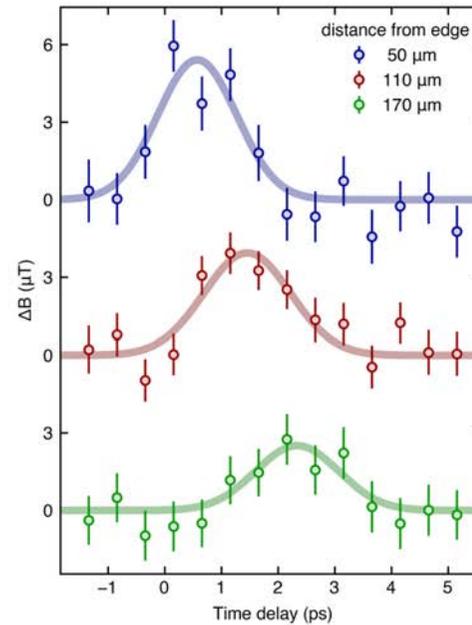
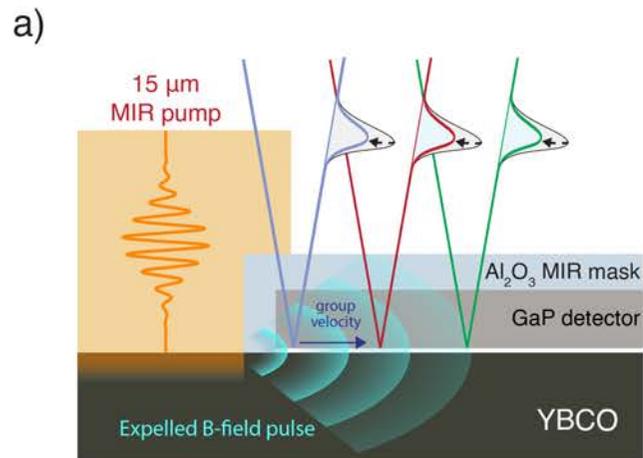
S. Fava, G. DeVecchi, G. Jotzu, M. Buzzi et al. forthcoming

# The Ultrafast Meissner effect: electrodynamics

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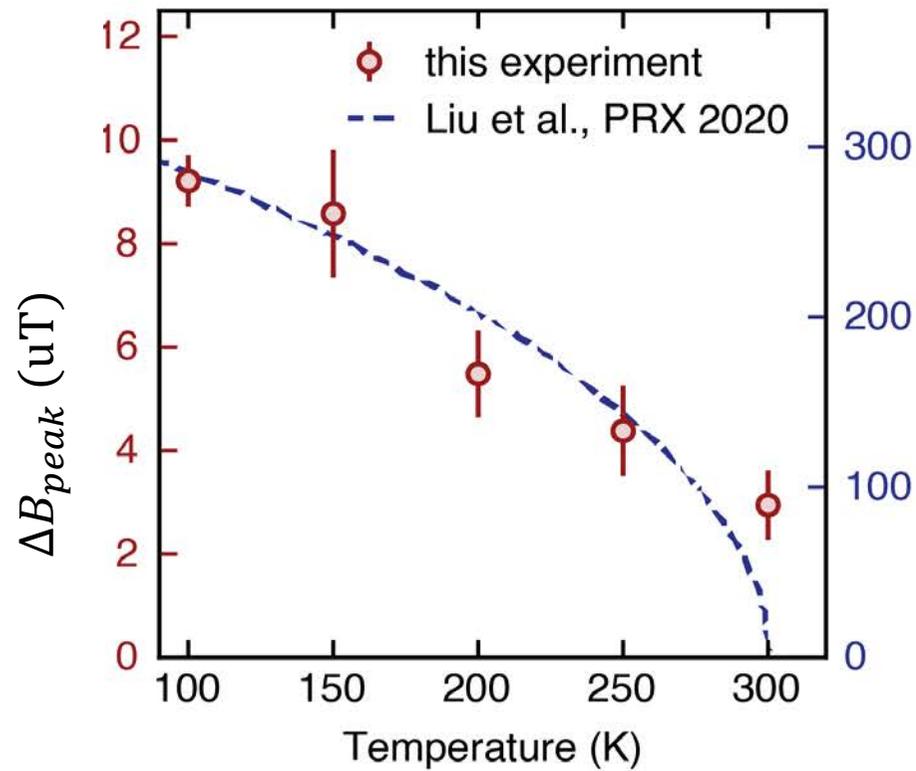


# The Ultrafast Meissner effect: electrodynamics

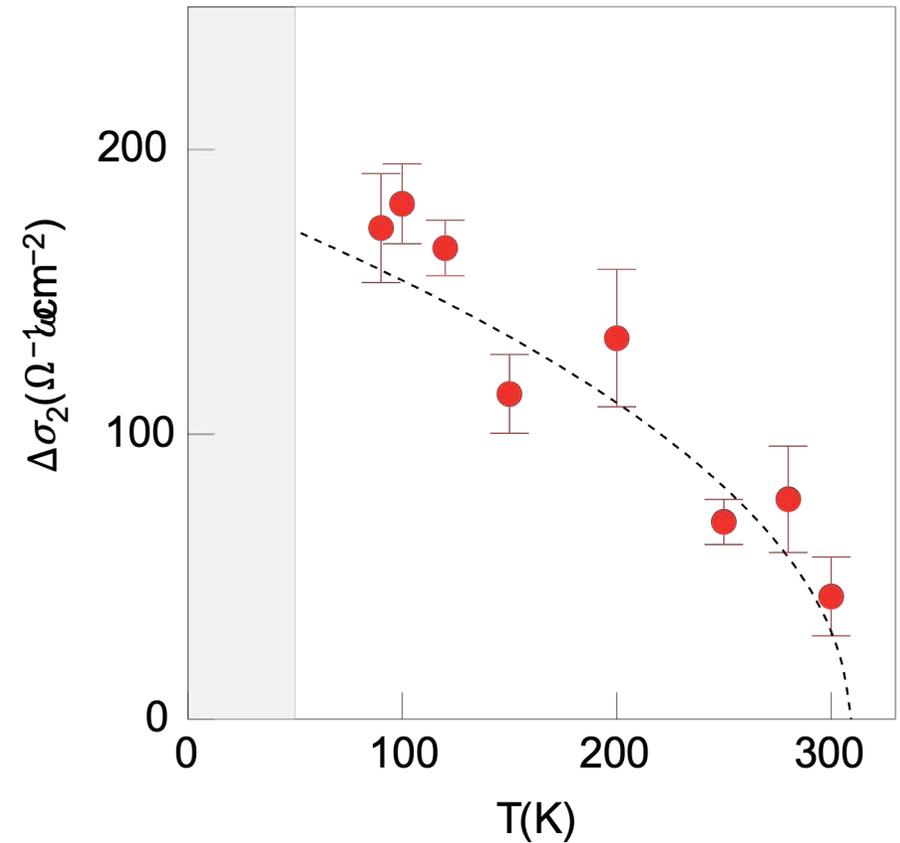


# Temperature Dependence

## Magnetic



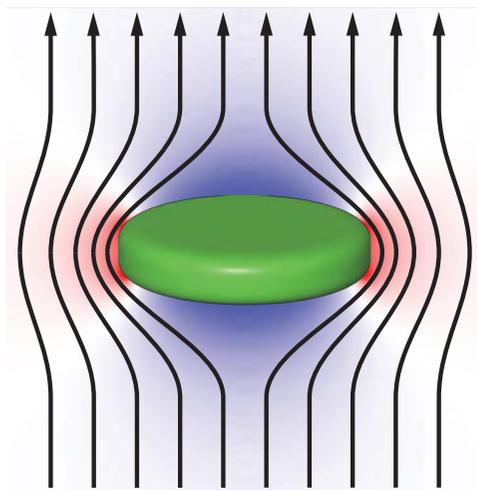
## Optical



S. Fava, G. DeVecchi, G. Jotzu, M. Buzzi et al. forthcoming



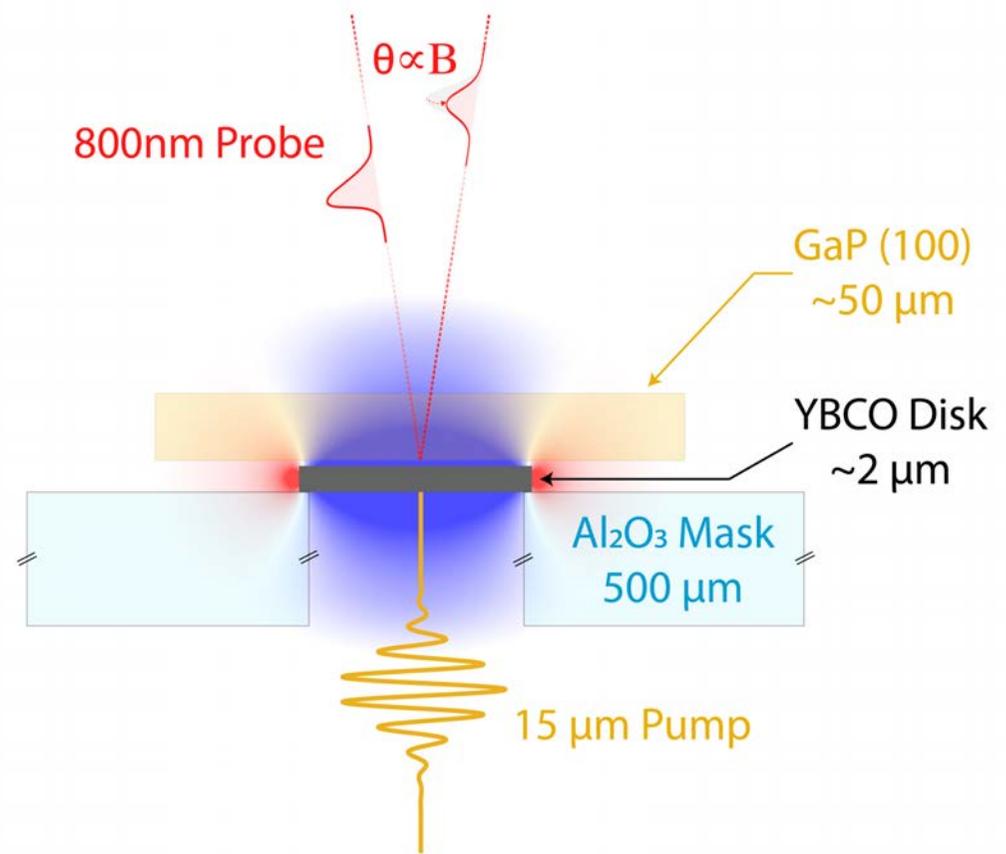
# Outlook: $\text{YBa}_2\text{Cu}_3\text{O}_x$ disk-shaped lamellas



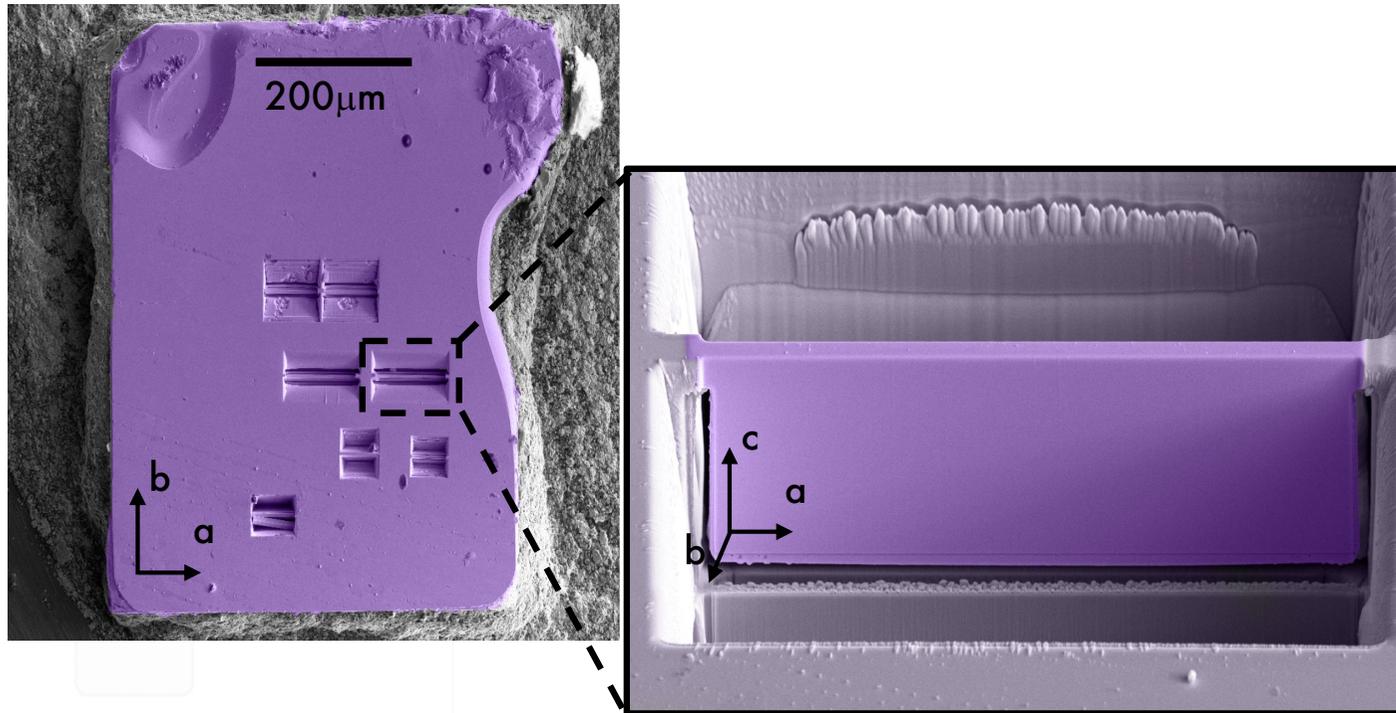
150  $\mu\text{m}$  diameter  
2  $\mu\text{m}$  thick YBCO disk

$B_{\text{app}}$

A vertical arrow pointing upwards, labeled  $B_{\text{app}}$ , indicating the direction of the applied magnetic field.



# Lamellas through microstructuring



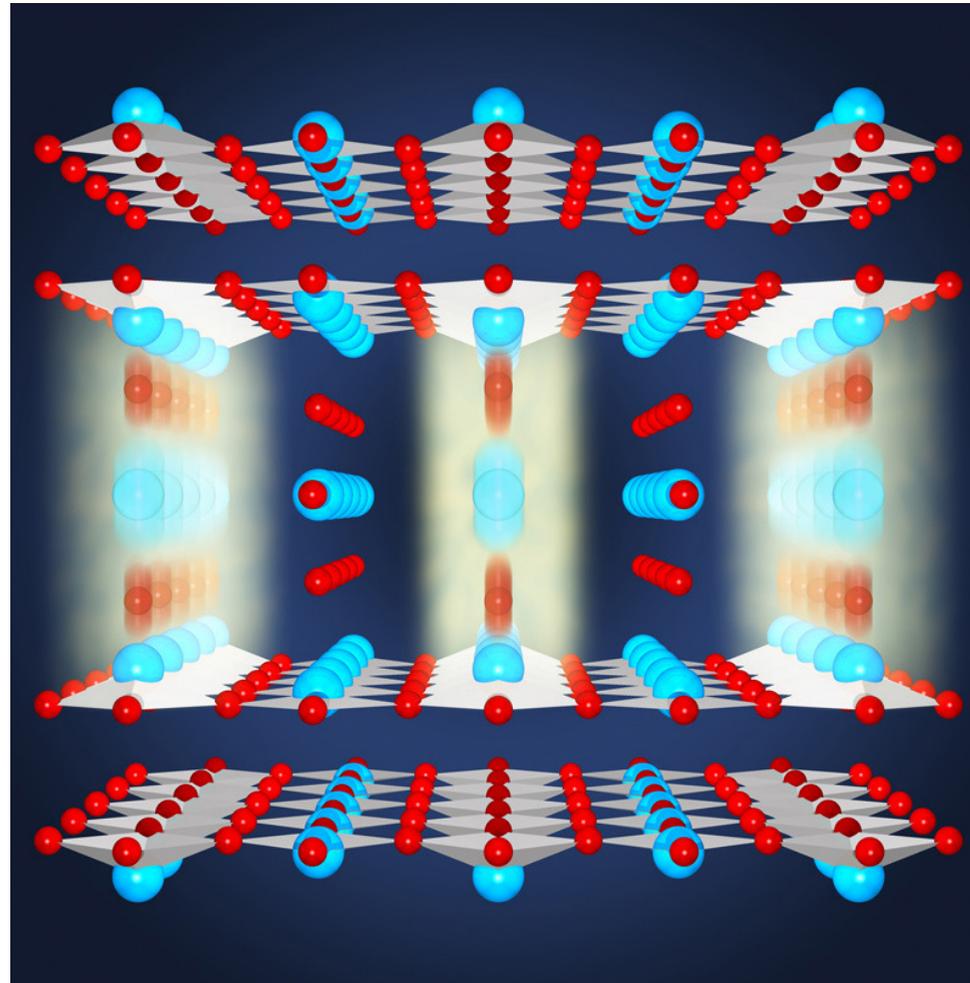
Phillip Moll



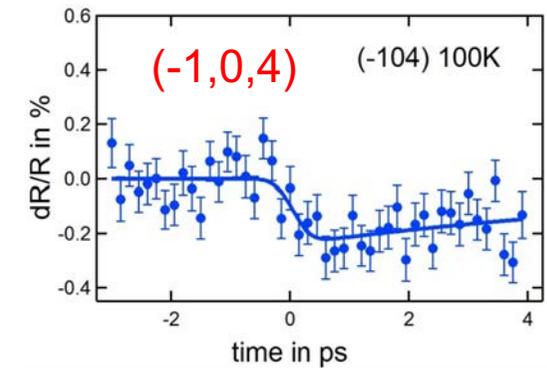
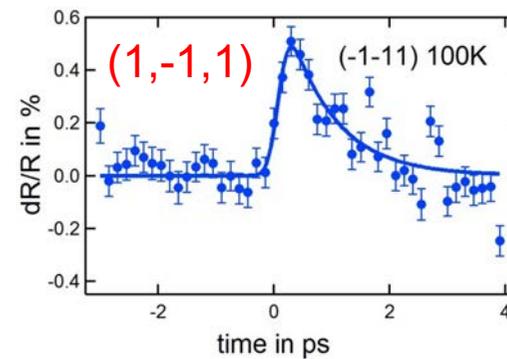
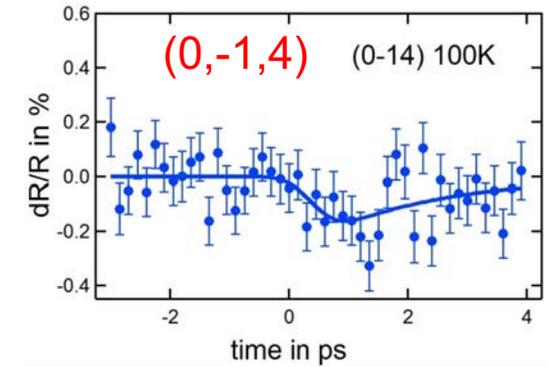
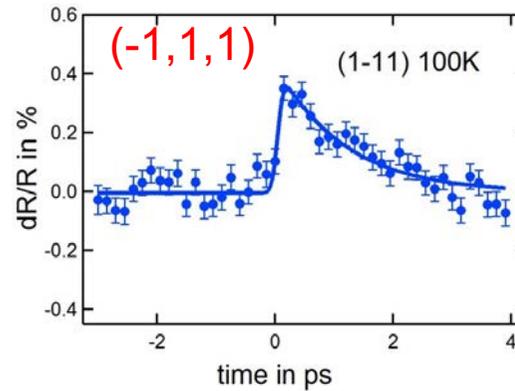
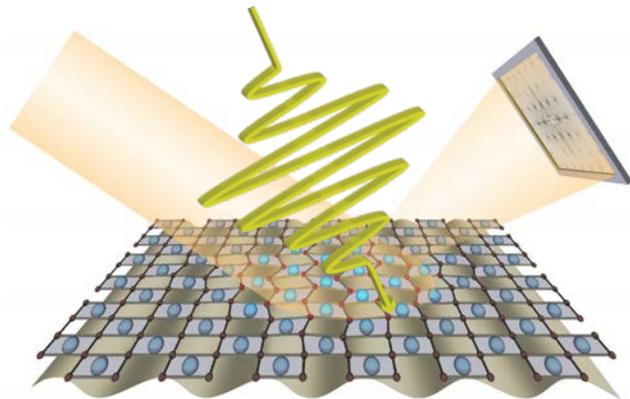
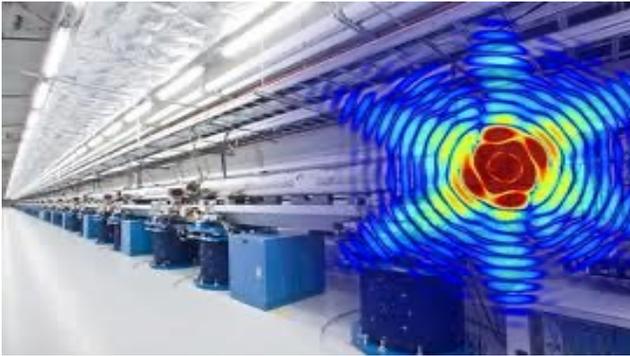
Carsten Putzke

# What is the physics of nonlinear phonons ?

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# Femtosecond X-ray Scattering: New Crystal Structure

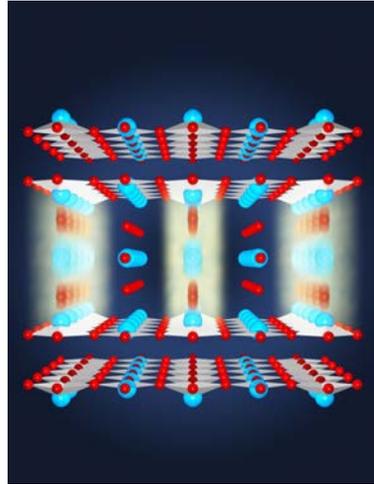


with A. Subedi, A. Georges

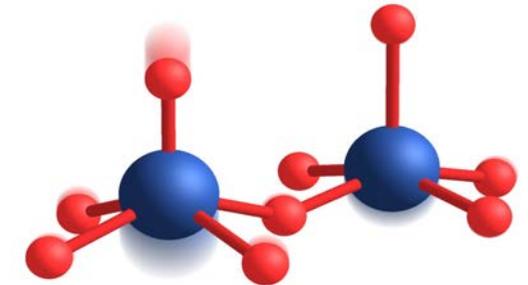
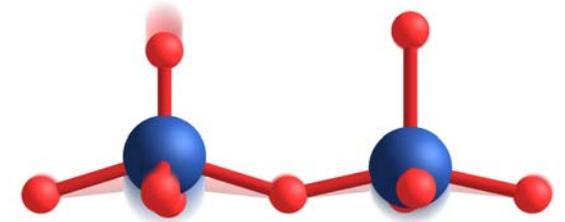
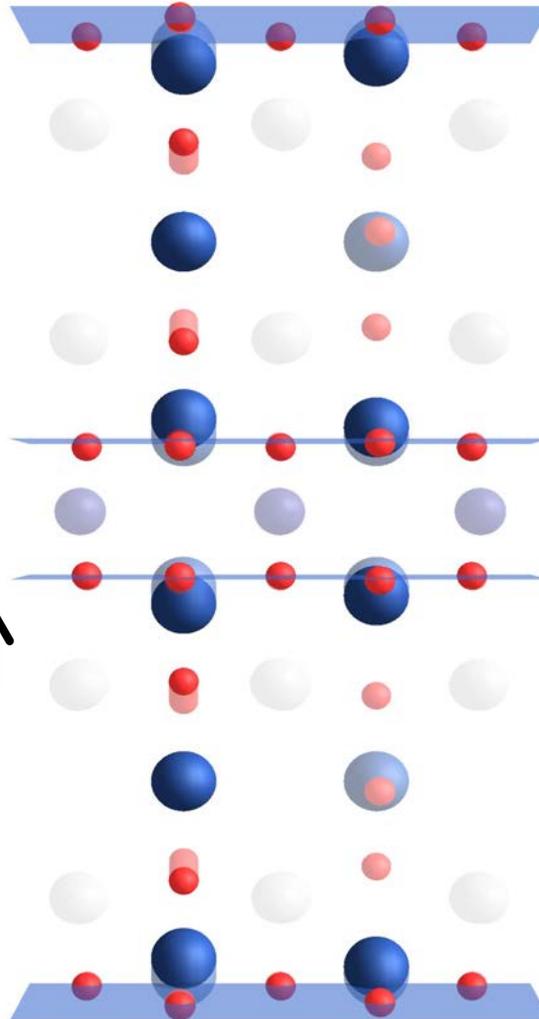
R. Mankowsky et al. *Nature* 516,71 (2014)



# New crystal structure in $\text{YB}_2\text{Cu}_3\text{O}_{6+x}$



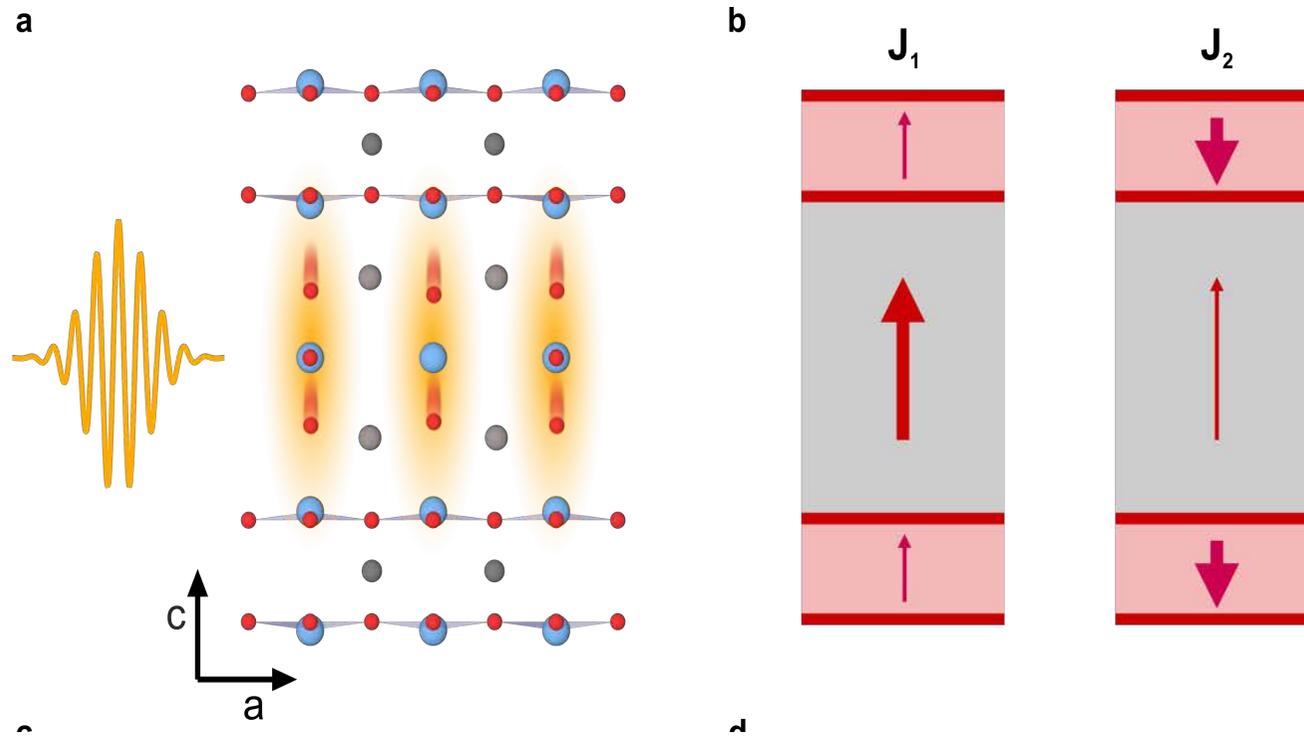
$d \sim 3\%$



R. Mankowsky et al. *Nature* 516, 71 (2014)

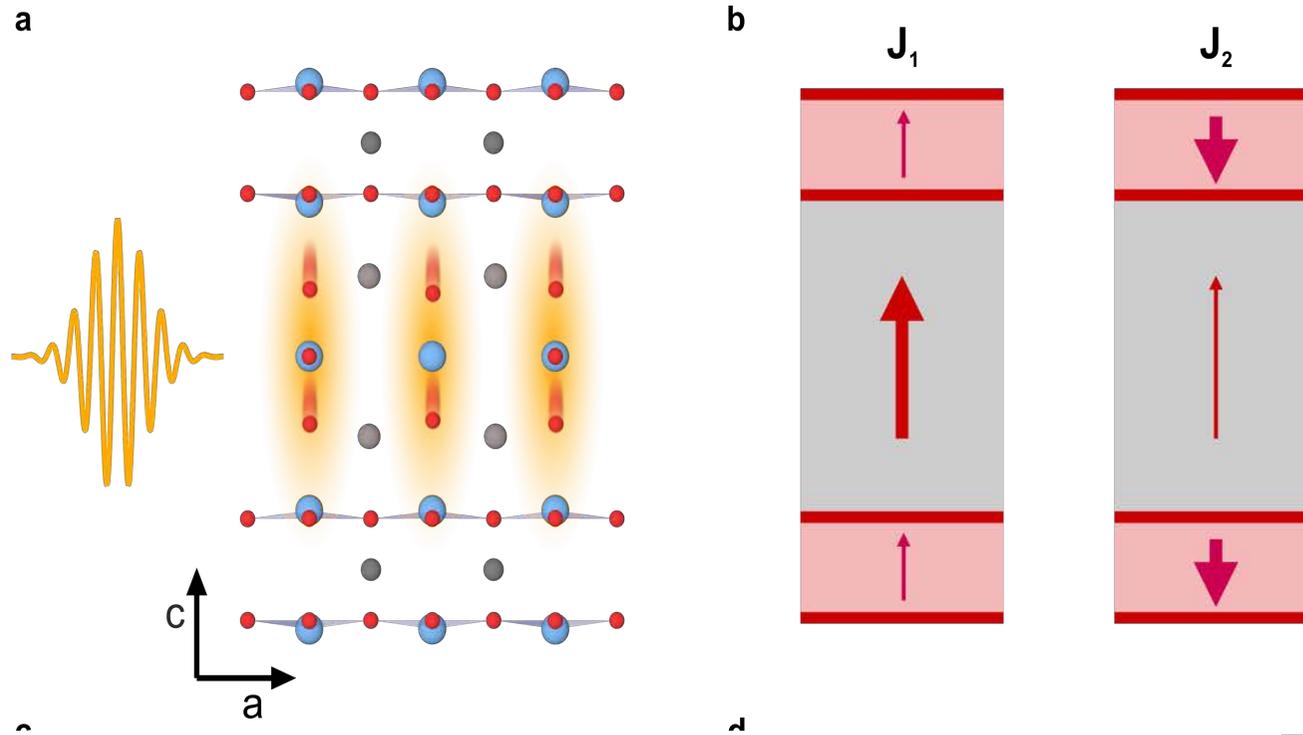


# How does the driven mode couple to interlayer tunneling



# Tri-linear coupling: one phonon and two plasmons

$$U_{non-linear} = \frac{1}{2} \omega_{IR}^2 Q_{IR}^2 + \frac{1}{2} \omega_{J_1}^2(q) J_1^2 + \frac{1}{2} \omega_{J_2}^2(q) J_2^2 + \mathbf{A} q^2 Q_{IR} J_1 J_2$$



M. Michael *et al.*, Phys. Rev B 102, 174505 (2020)

M. Michael *et al.*, Phys. Rev B 105, 17301 (2022)



with Marios Michael, Eugene Demler



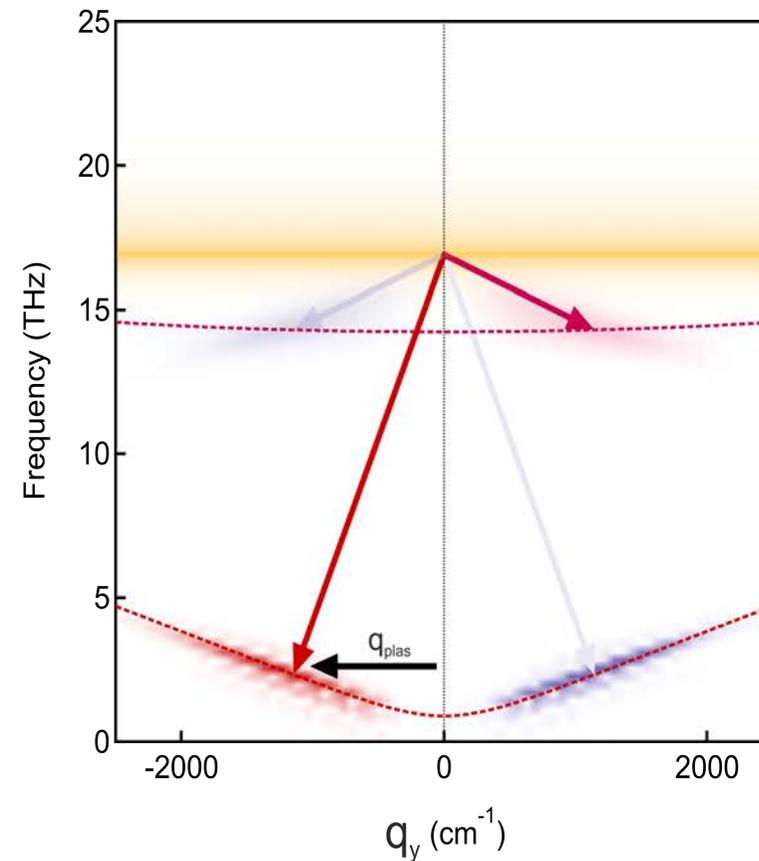
# Three mode mixing – one phonon and two plasmons

$$\ddot{Q}_{IR} + 2\gamma_{IR}\dot{Q}_{IR} + \omega_{IR}^2 Q_{IR} = Z^* E(t)$$

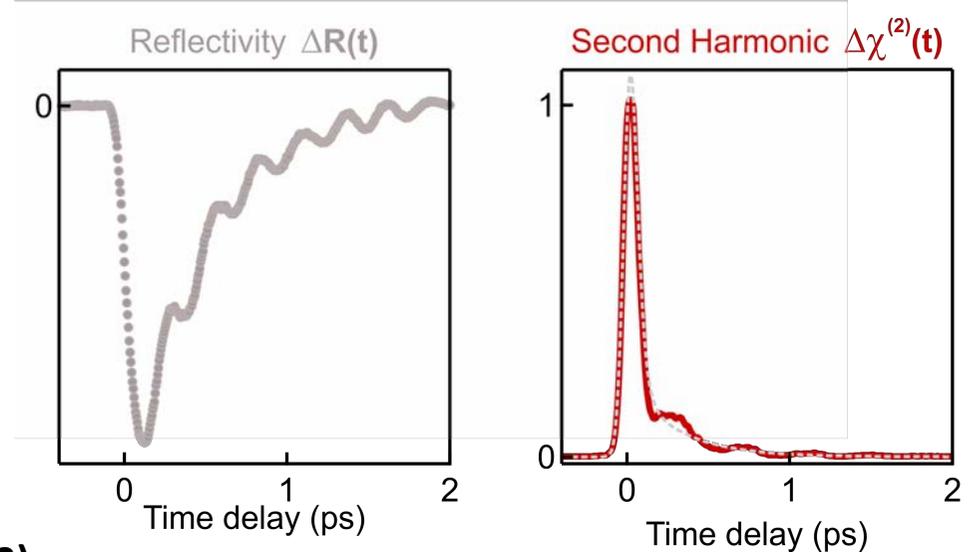
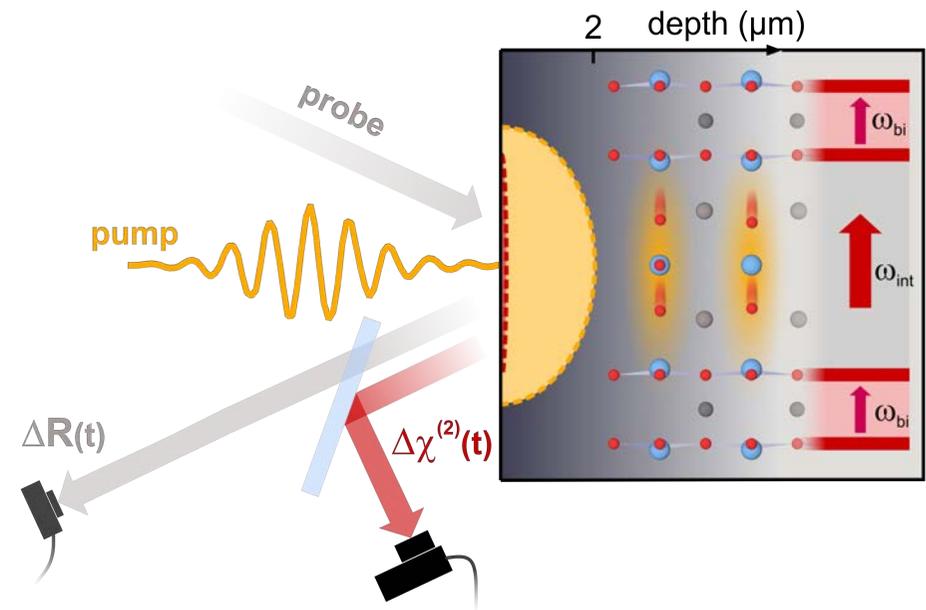
$$\ddot{J}_1 + 2\gamma_{J_1}\dot{J}_1 + \omega_{J_1}^2(q)J_1 = -aq^2 Q_{IR}J_2$$

$$\ddot{J}_2 + 2\gamma_{J_2}\dot{J}_2 + \omega_{J_2}^2(q)J_2 = -aq^2 Q_{IR}J_1$$

**Resonant if  $\omega_{IR} = \omega_{IP1} + \omega_{IP2}$**



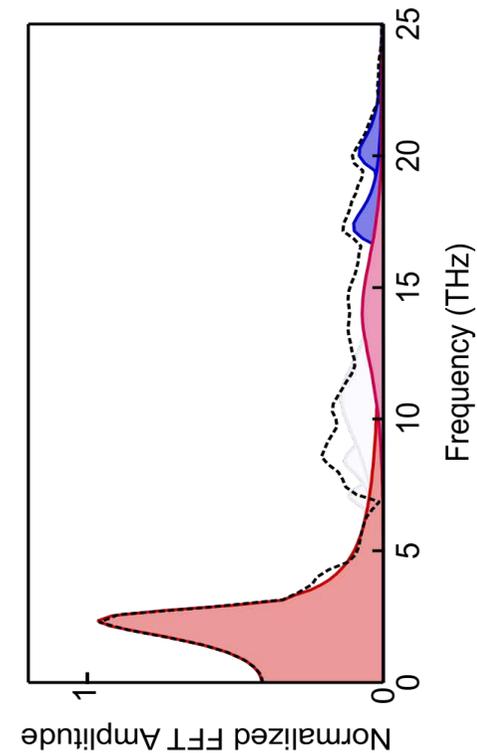
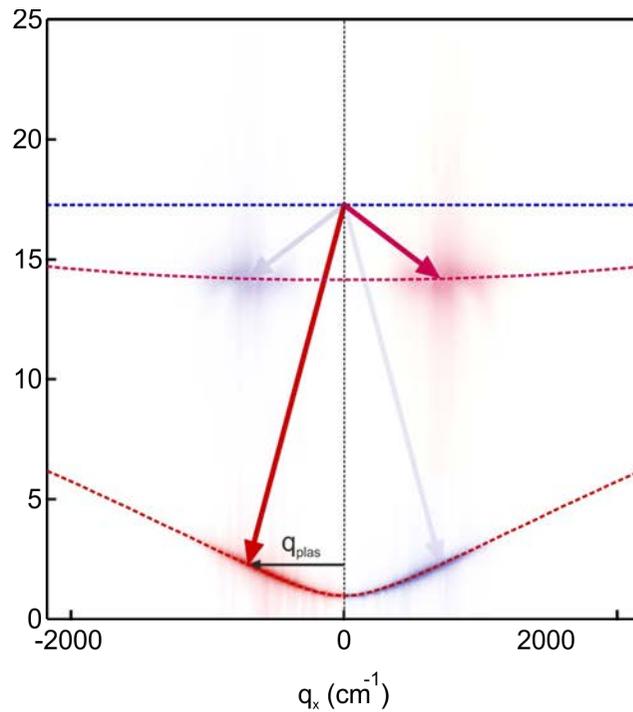
# Measuring coherent dynamics: time resolved SHG



A. Von Hoegen et al. Phys. Rev. X 12, 031008 (2022)



# 1) Frequency resonant three mode mixing

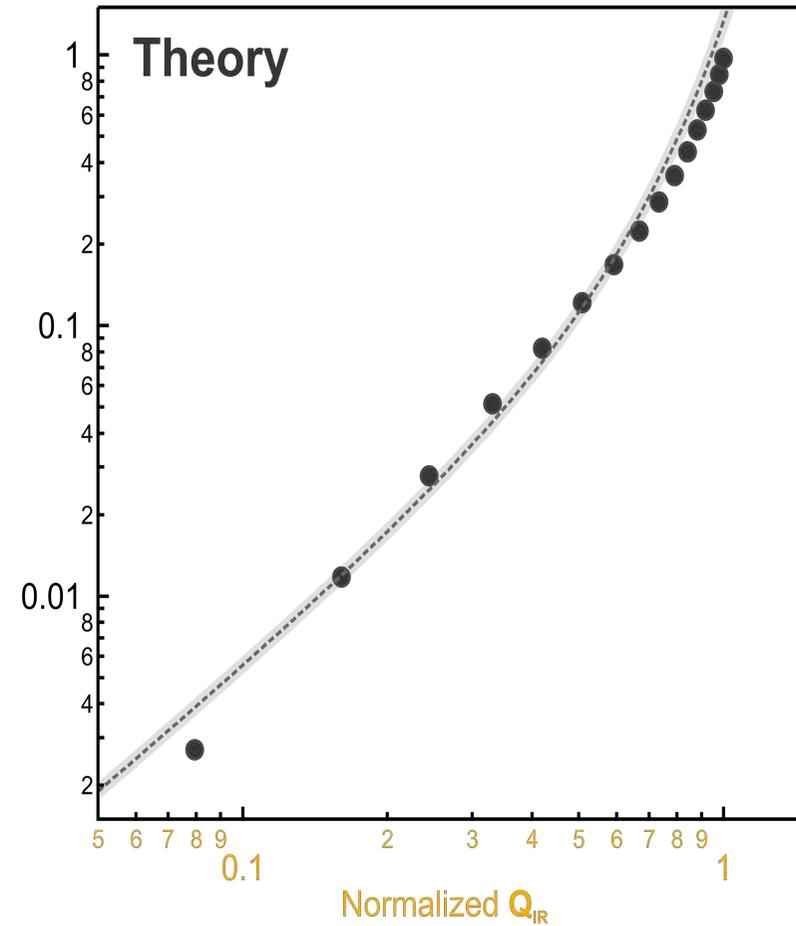
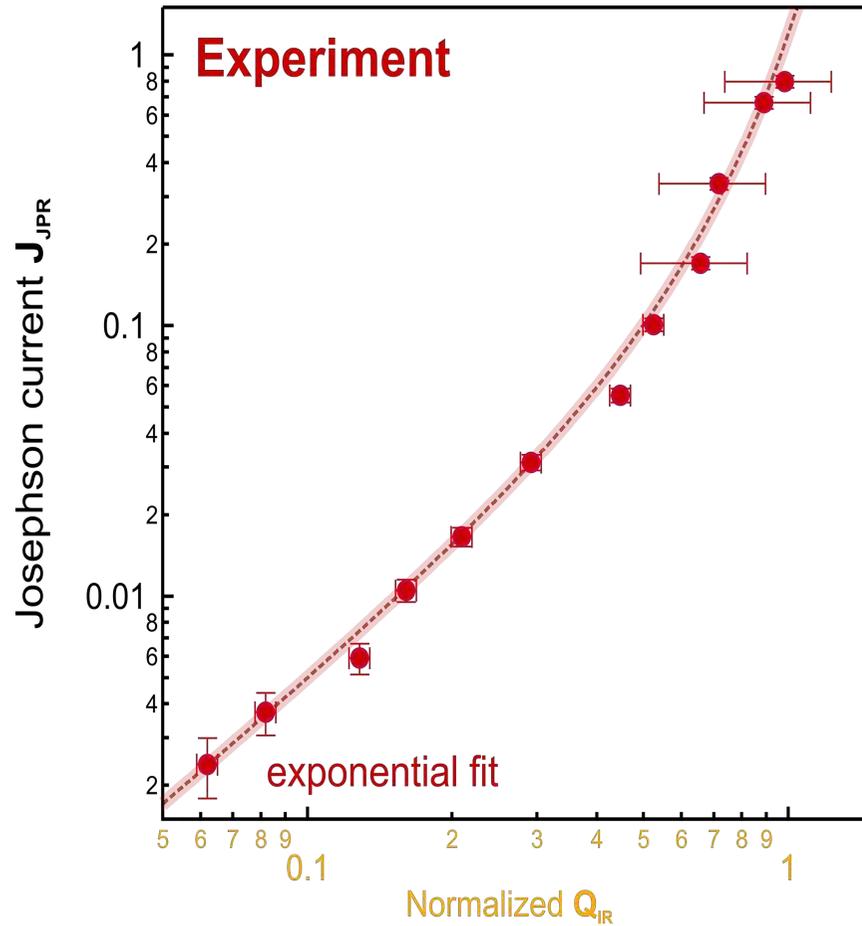


M. Michael *et al.*, Phys. Rev B 102, 174505 (2020)

A. Von Högen *et al.* Phys Rev X 12, 031008 (2022)



## 2) Exponential amplification of the plasma mode



A. Von Högen et al. Phys Rev X 12, 031008 (2022)

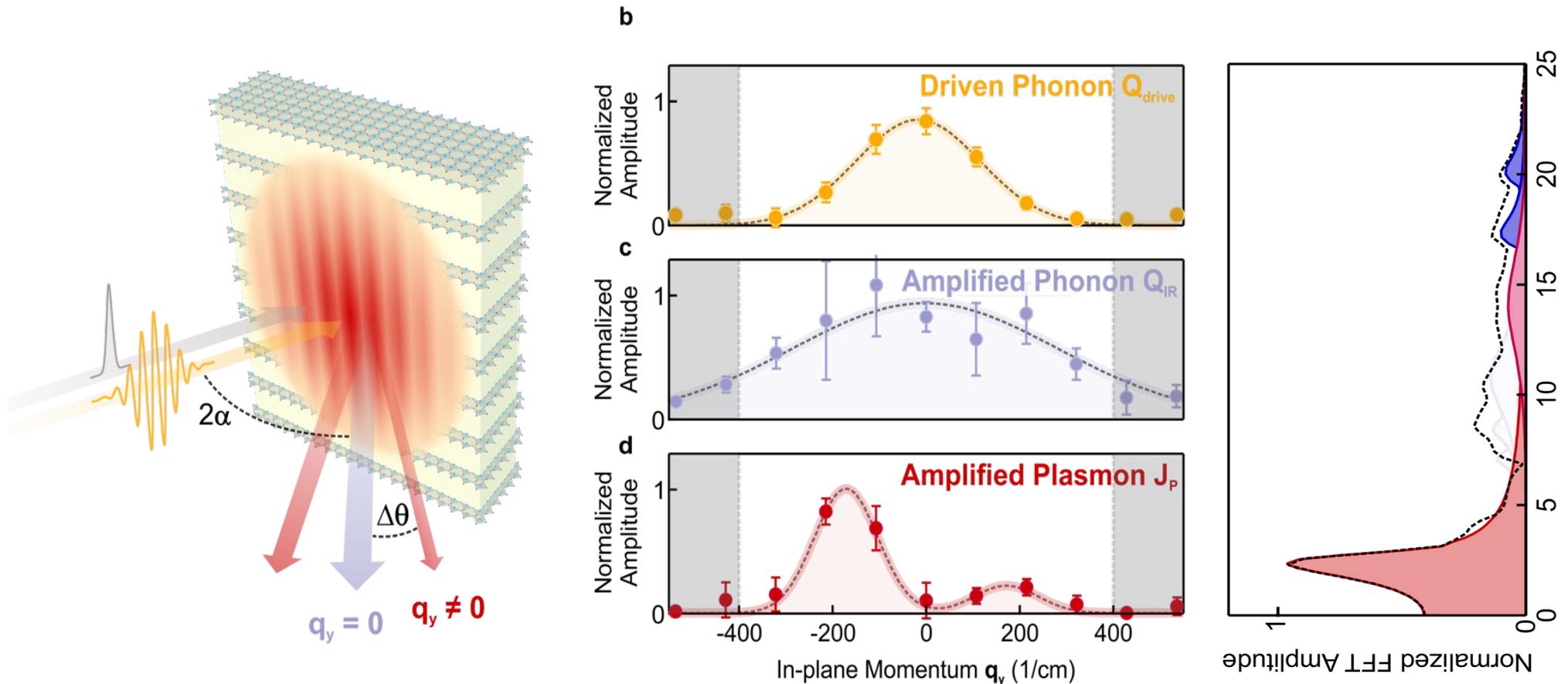
M. Michael et al., Phys. Rev B 102, 174505 (2020)



# 3) Amplification at finite momentum

## Momentum Resolved Nonlinear Scattering

$$H_I \sim A q^2 Q_{IR} J_1 J_2$$

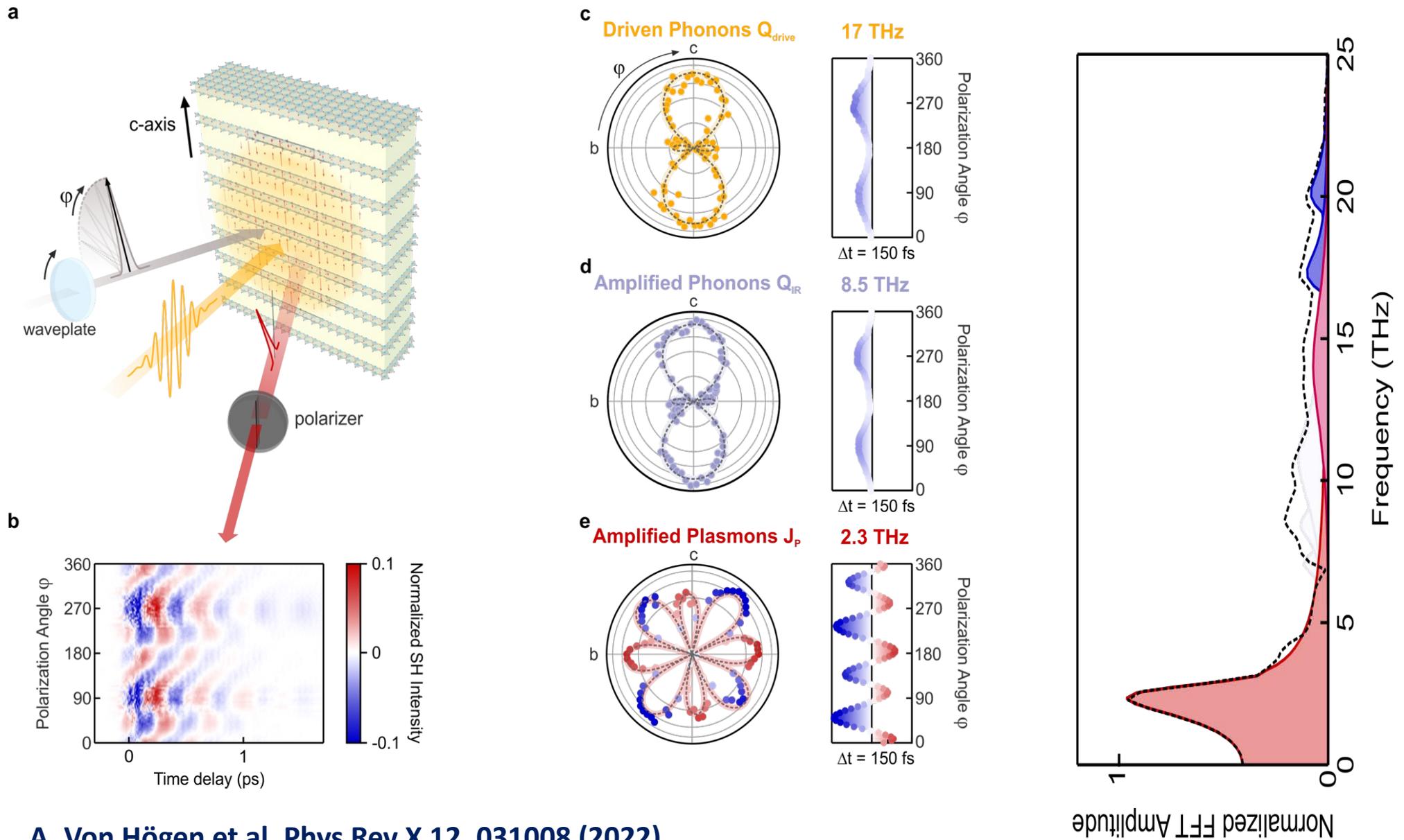


A. Von Högen et al. Phys Rev X 12, 031008 (2022)

M. Michael et al., Phys. Rev B 102, 174505 (2020)



# 4) Complex Mode symmetry – not a phonon

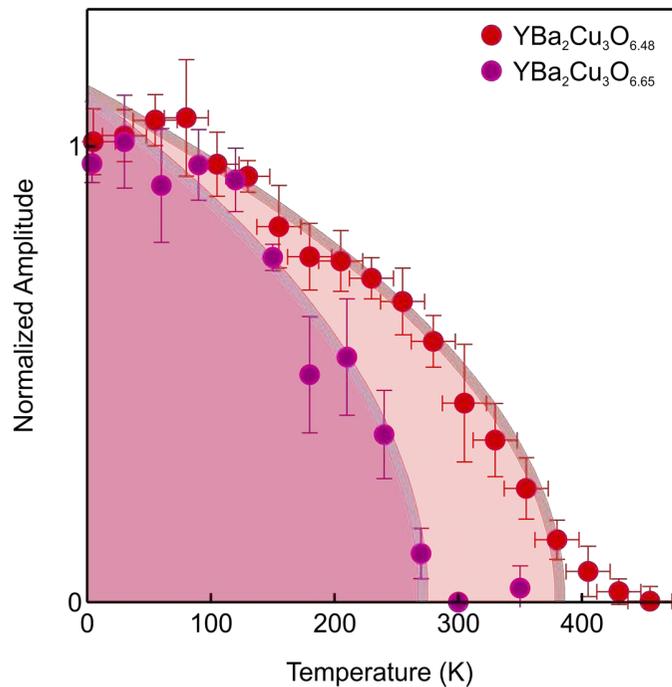


A. Von Högen et al. Phys Rev X 12, 031008 (2022)



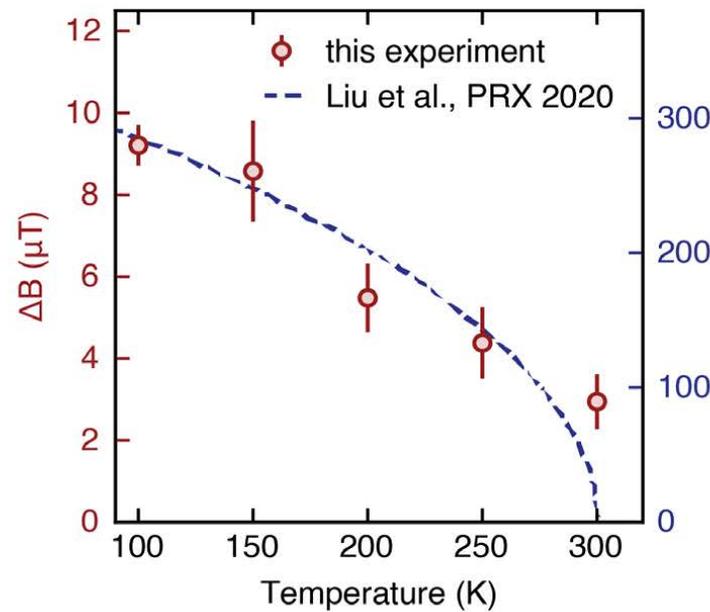
# 5) Anomalous temperature dependence (up to $T^*$ )

## Coherent Plasmons

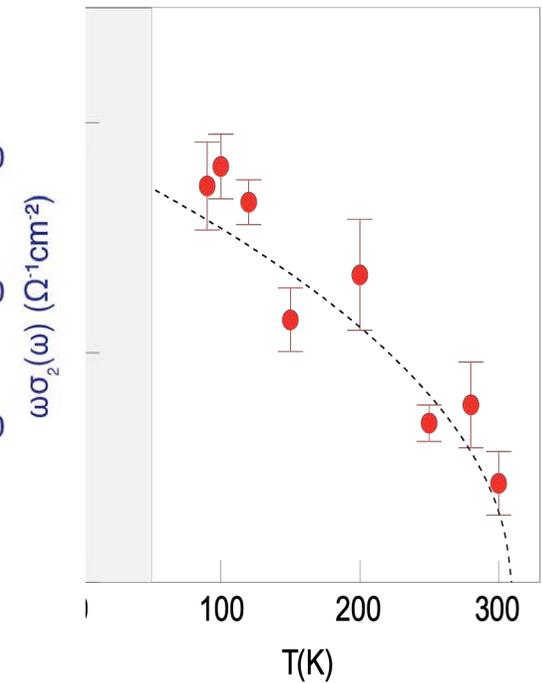


## Meissner

a)



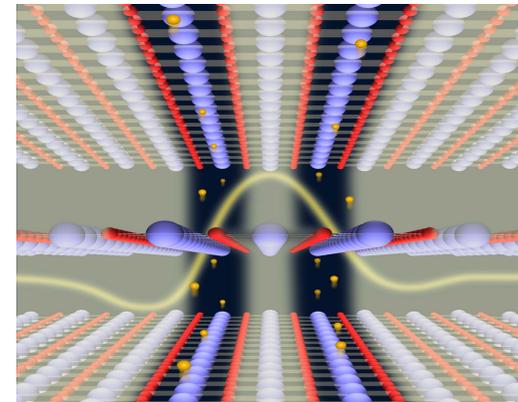
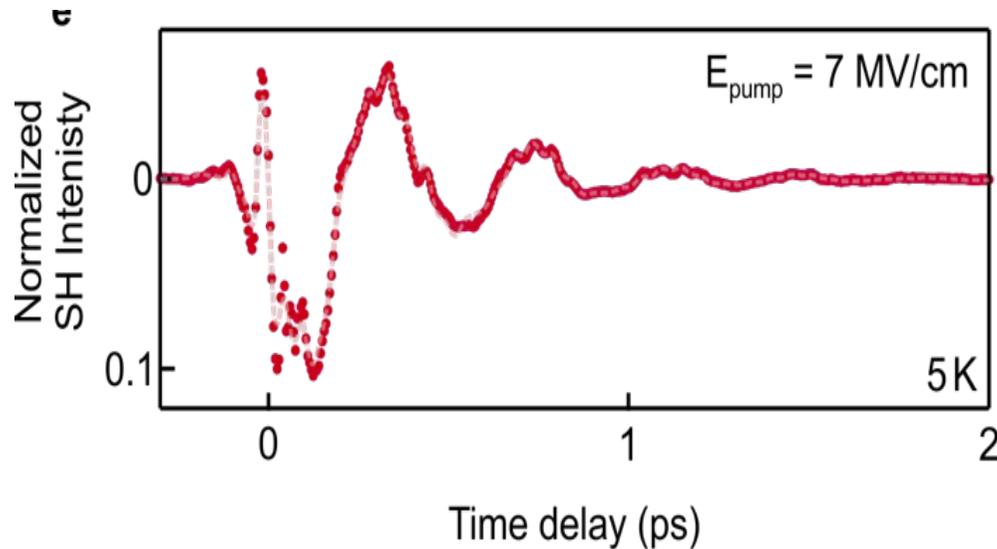
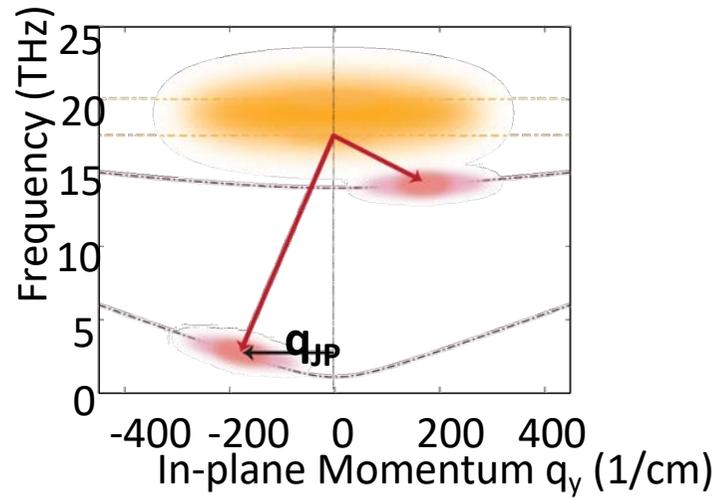
## Optical



A. Von Högen et al. Phys Rev X 12, 031008 (2022)



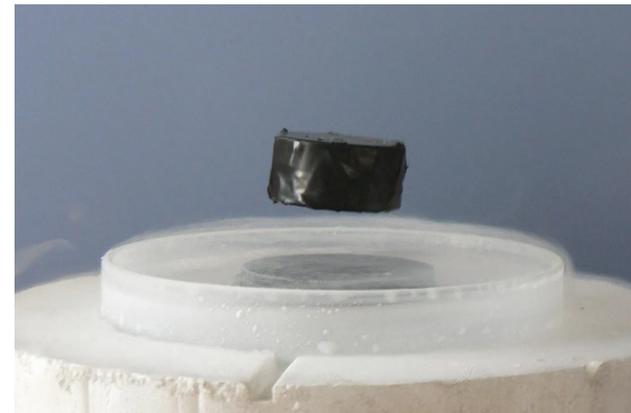
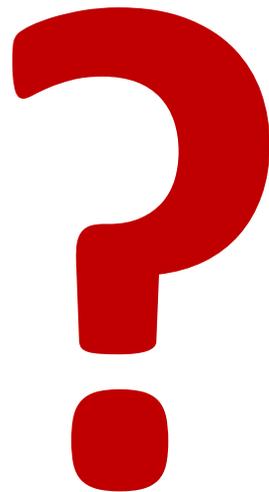
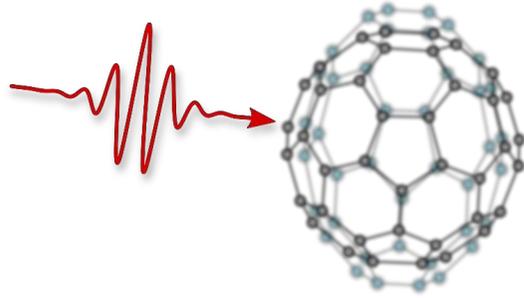
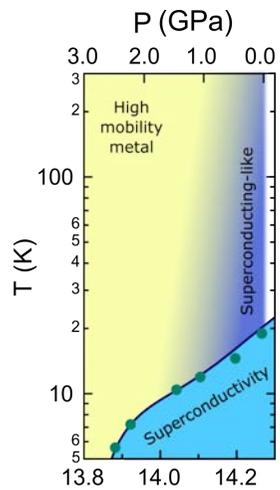
# Similarities with polariton condensates, time crystals.....



A. Von Högen et al. Phys Rev X 12, 031008 (2022)



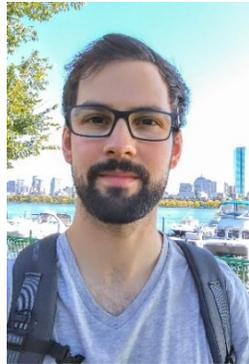
# Do these explain the optical and magnetic properties?



# Acknowledgements



N. Taherian



Alexander von Hoegen



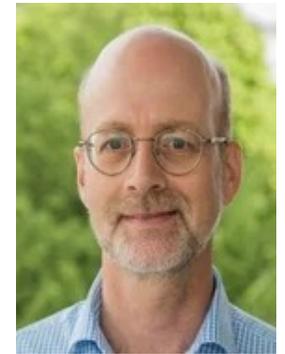
Michael Först



Albert Liu



Michael Fechner



Bernhard Keimer



Sebastian Fava



Giovanni De Vecchi



Michele Buzzi



Gregor Jotzu

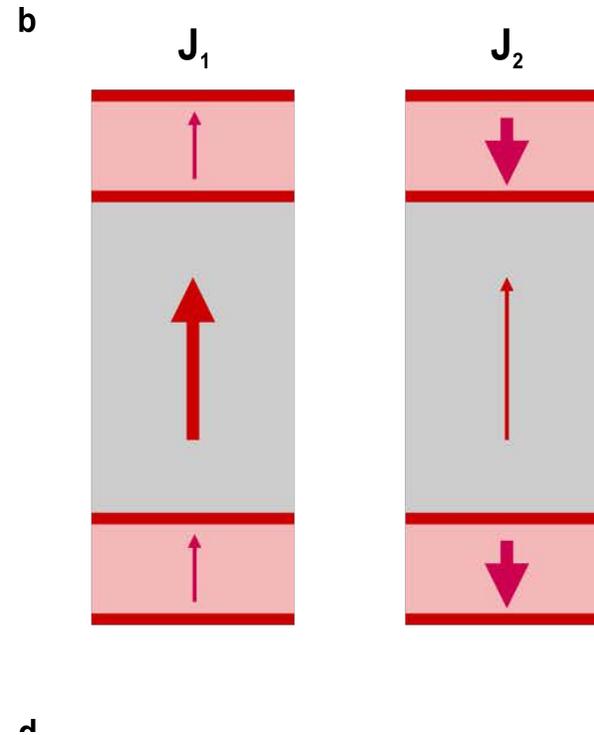
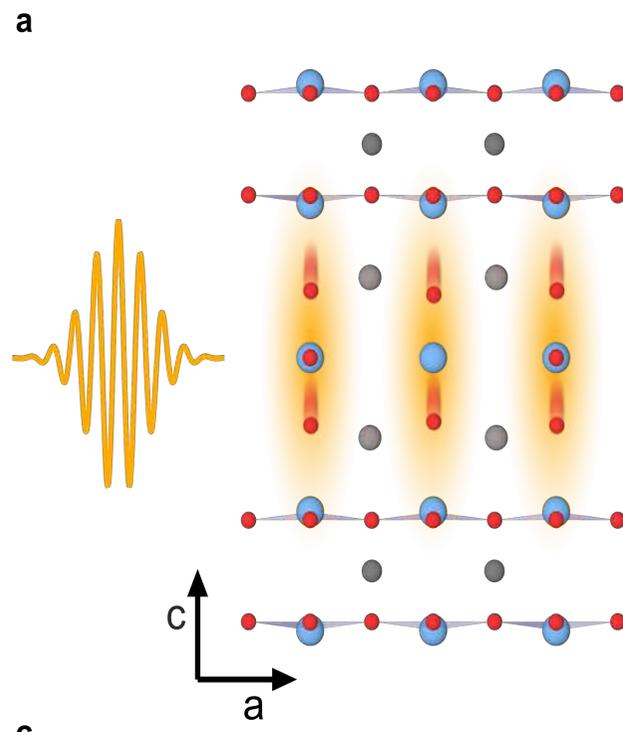


Marios H. Michael



Eugene Demler

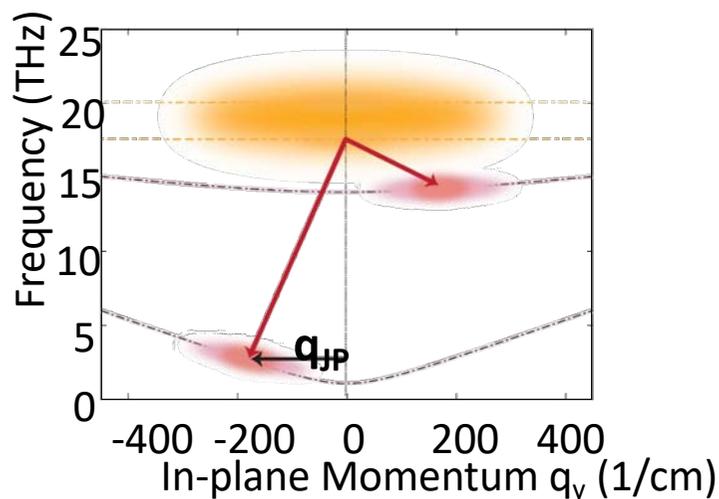
# Is this model unique?



# With two phonons – I have TWO possible resonances

## THREE MODE MIXING

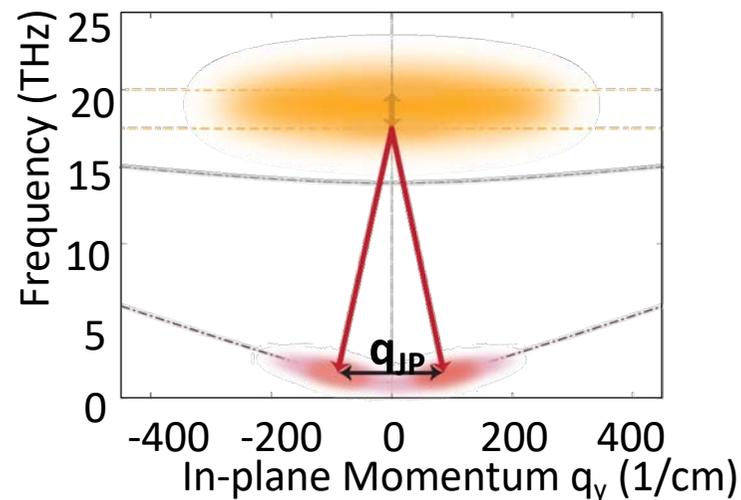
$$gq^2(Q_{1,IR} + Q_{2,IR})J_1 J_2$$



## FOUR MODE MIXING

$$g(Q_{1,IR} + Q_{2,IR})^2 J_1^2$$

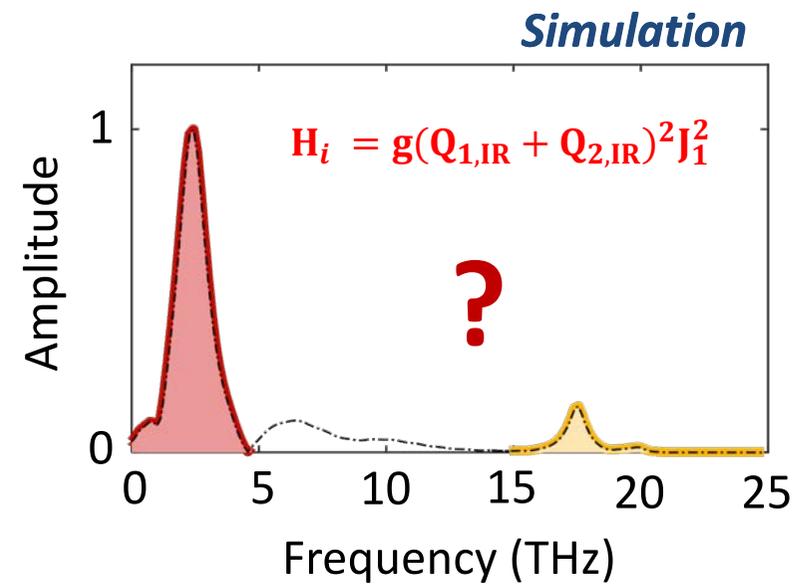
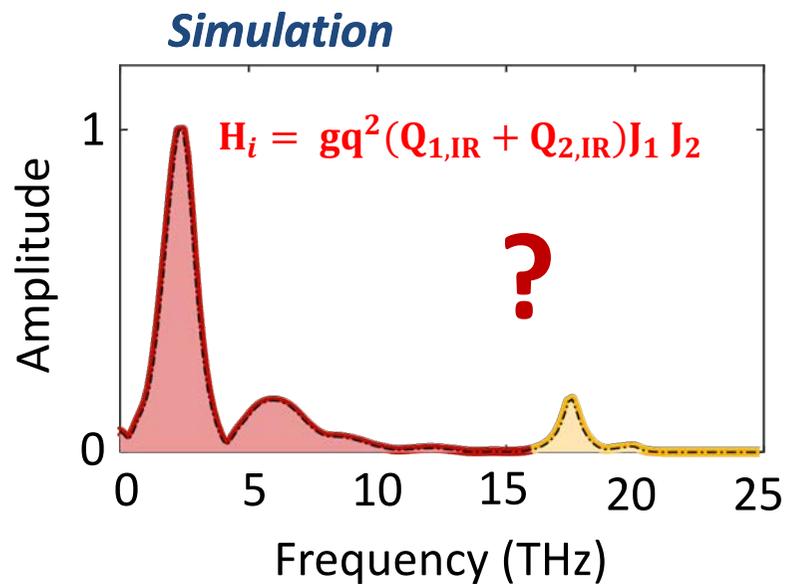
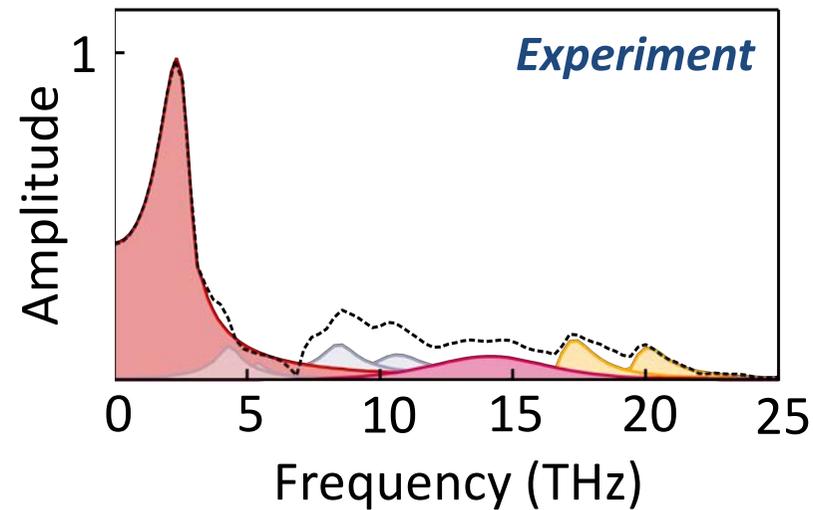
$$gQ_{1,IR}^2 J_1^2 + Q_{2,IR}^2 J_1^2 + Q_{1,IR} Q_{2,IR} J_1^2$$



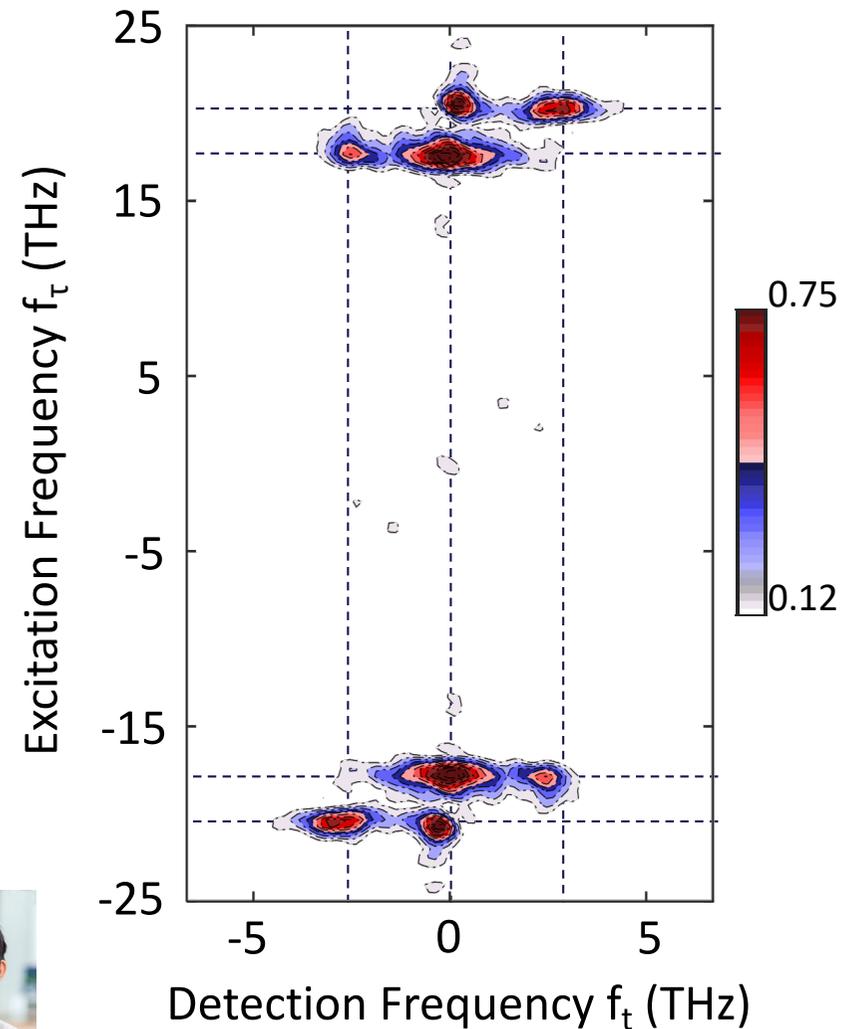
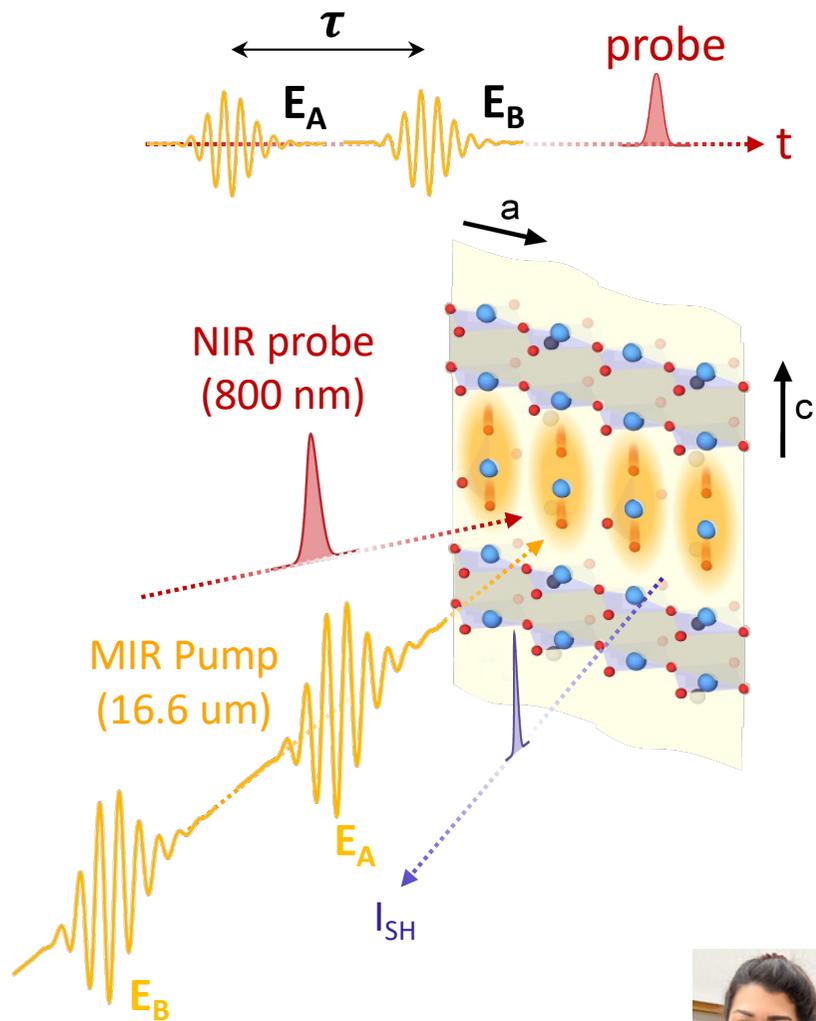
N. Taherian



# One-dimensional pump probe: ambiguous assignment



# Two dimensional spectroscopy to resolve ambiguity



N.Taherian, et al. (in preparation).



N. Taherian



Albert Liu

# Experiment vs theory: **four waves** and not three waves

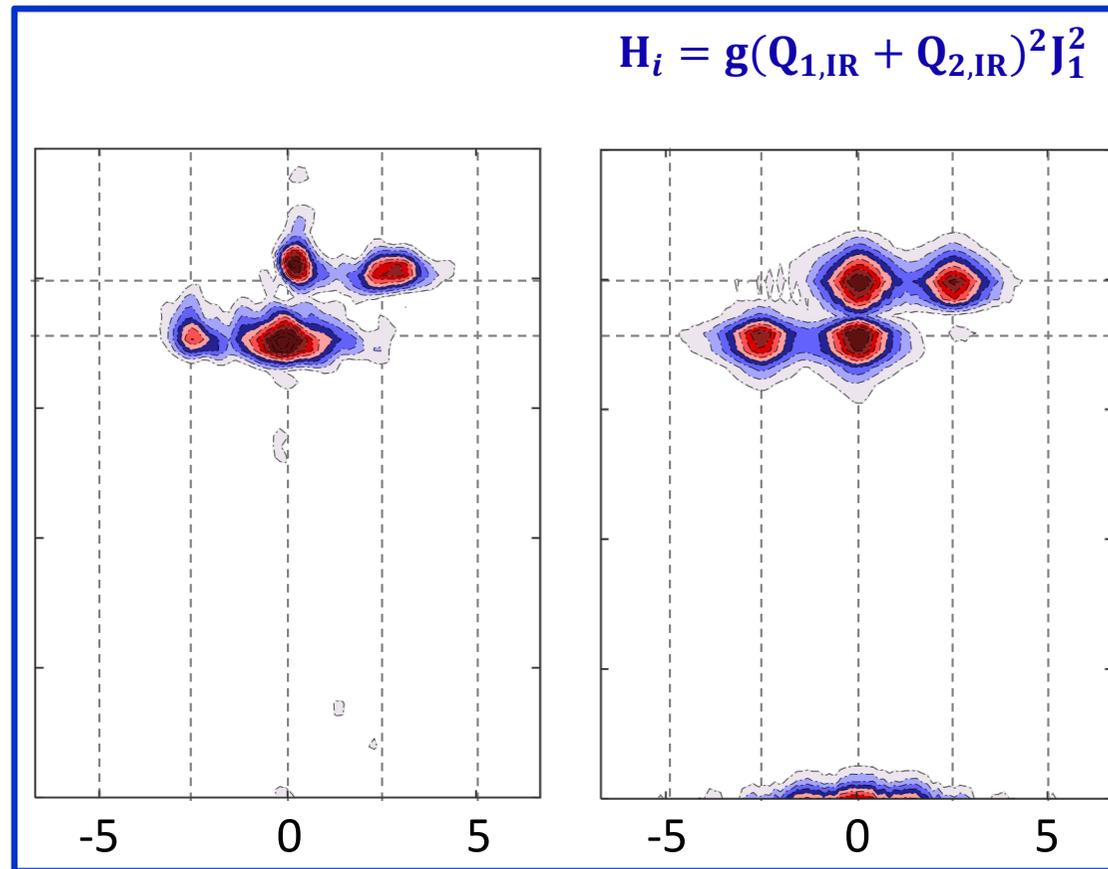
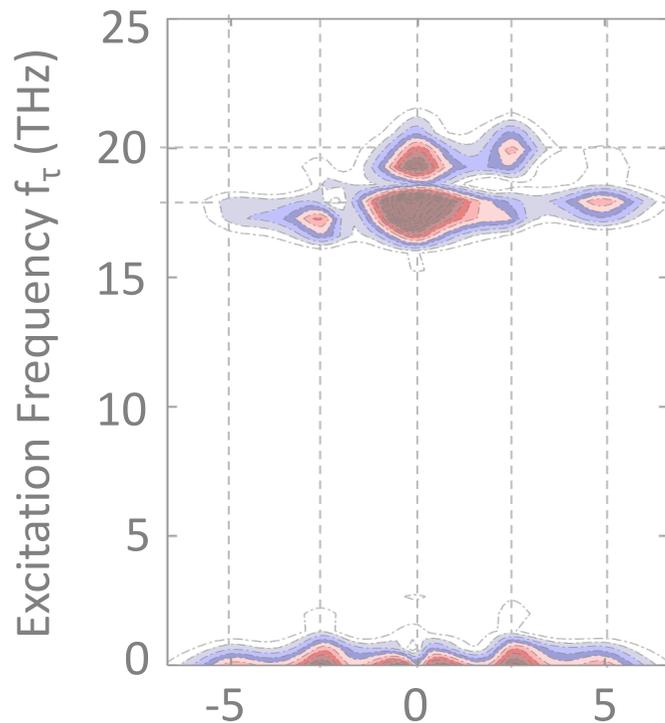
2D Simulation:  
*three waves*

2D Experiment

2D Simulation  
*four waves*

$$H_i = gq^2(Q_{1,IR} + Q_{2,IR})J_1 J_2$$

$$H_i = g(Q_{1,IR} + Q_{2,IR})^2 J_1^2$$

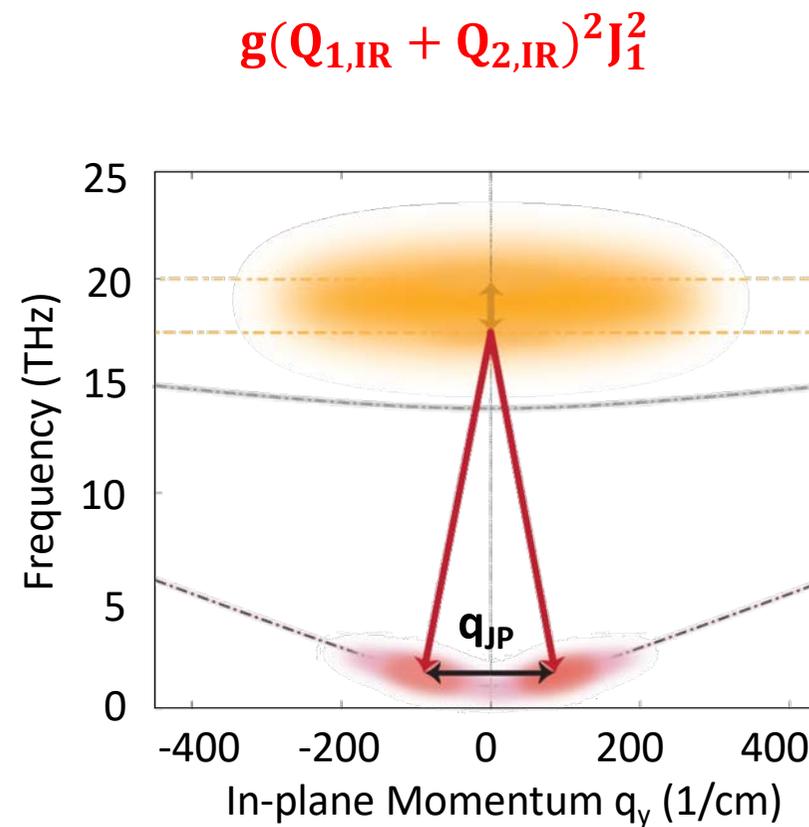
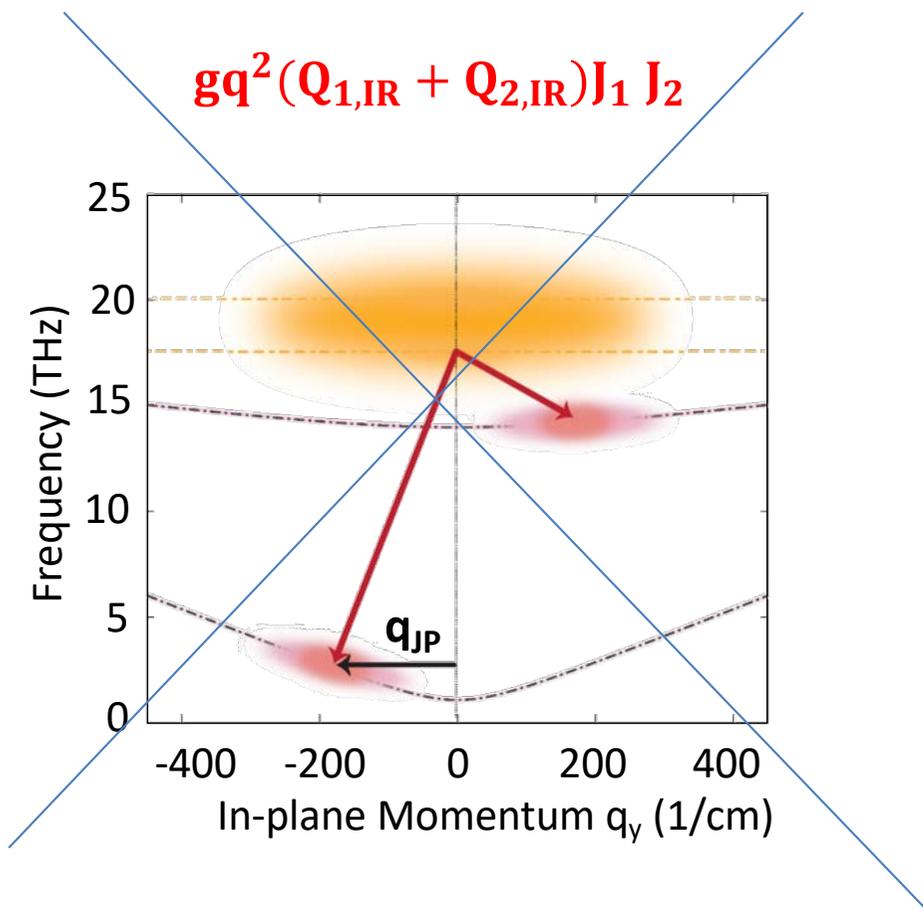


Detection Frequency  $f_t$  (THz)

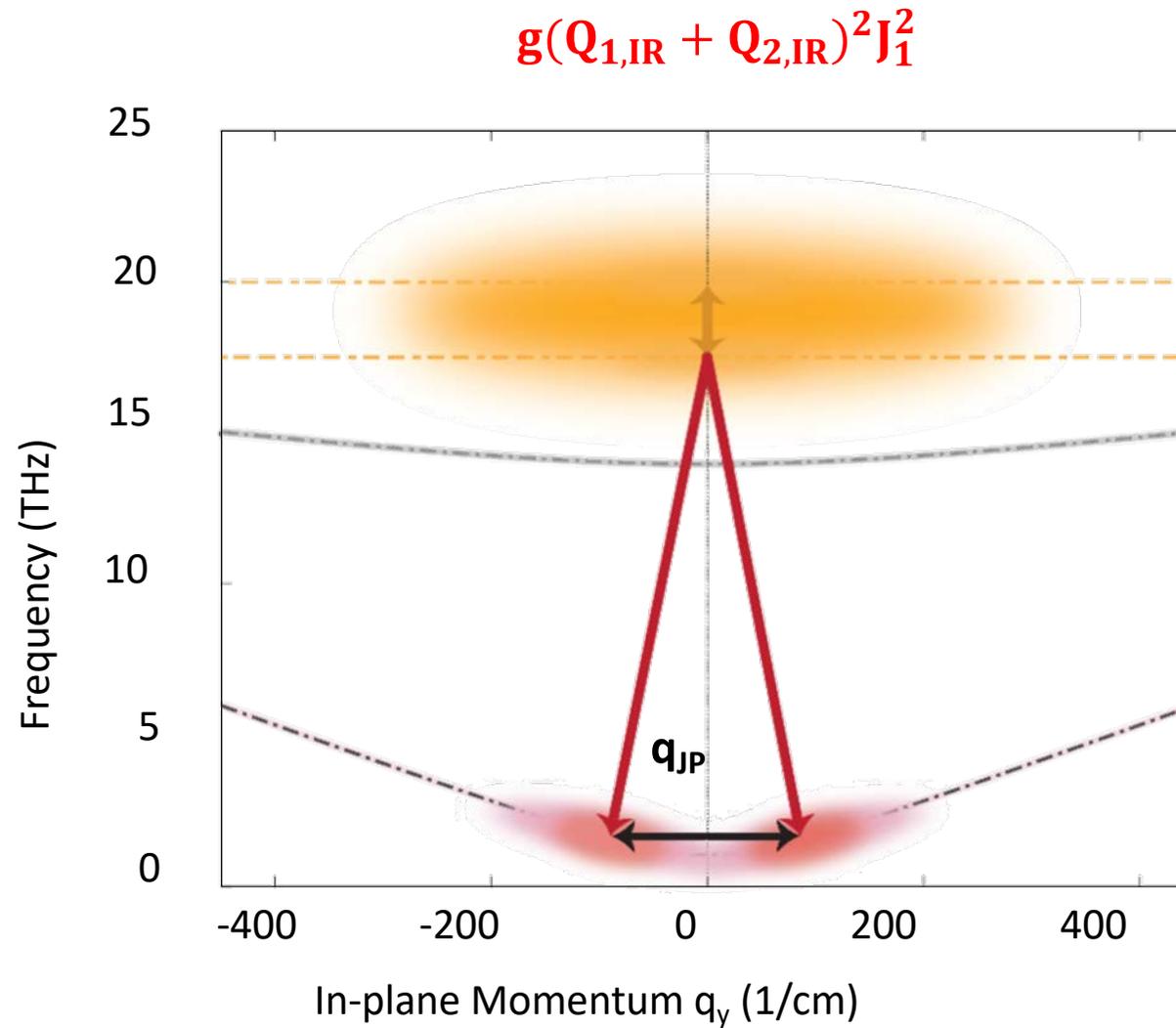
N.Taherian, et al. (in preparation).



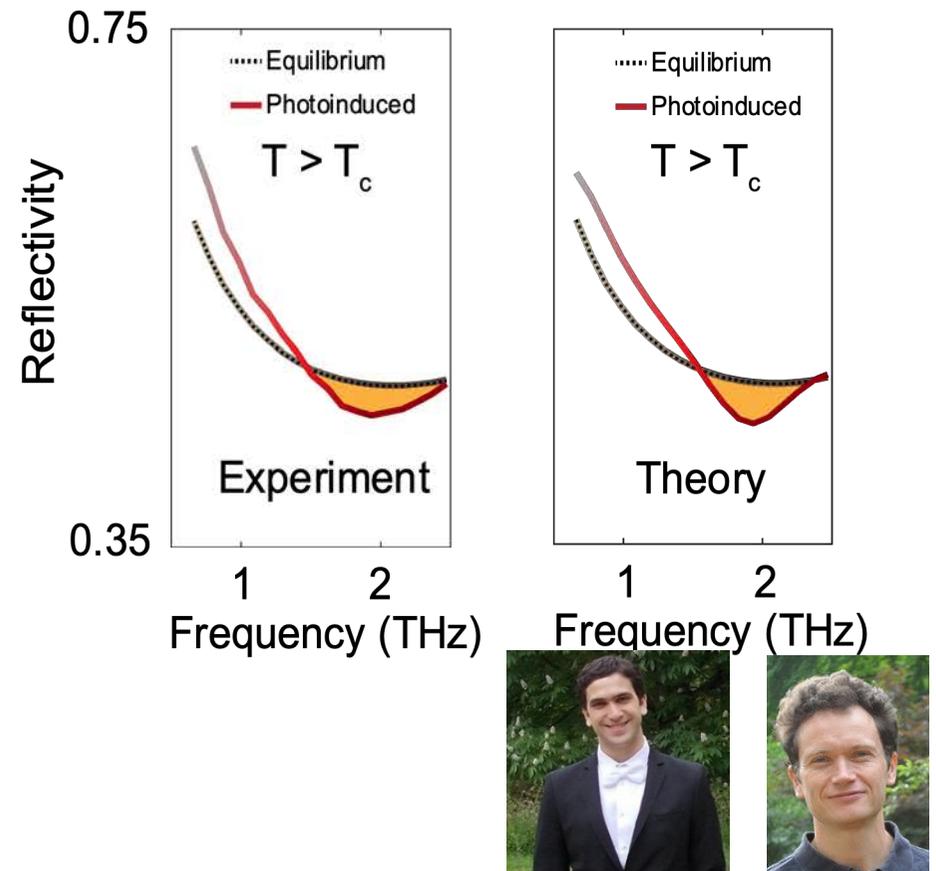
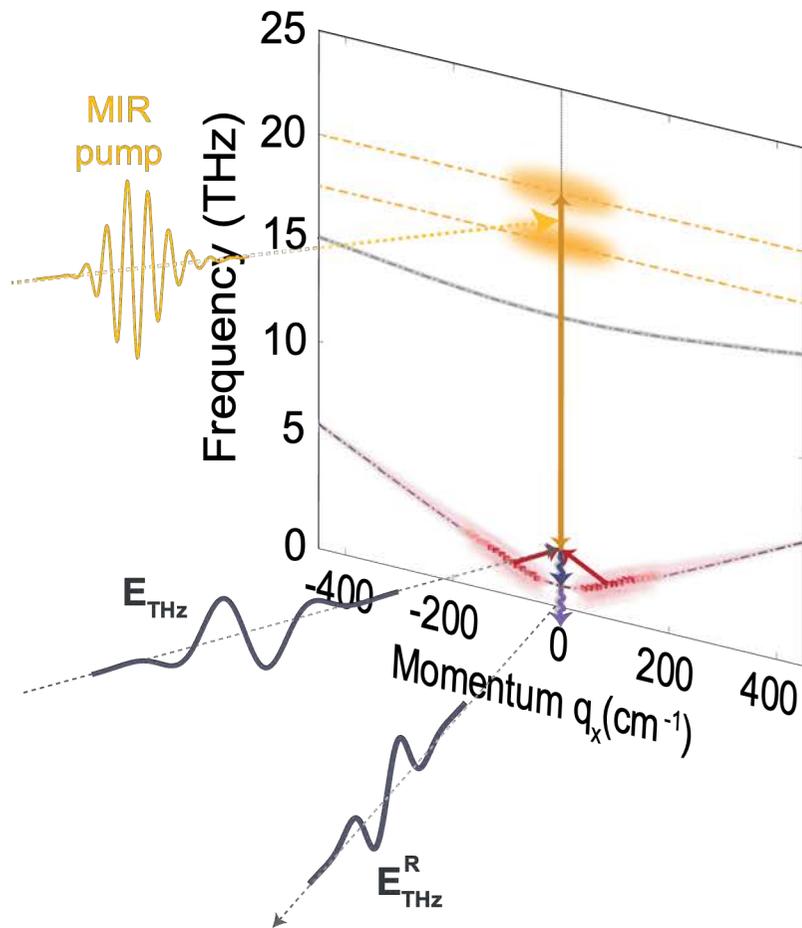
# Four mode parametric coupling



# Squeezed Josephson Plasmons



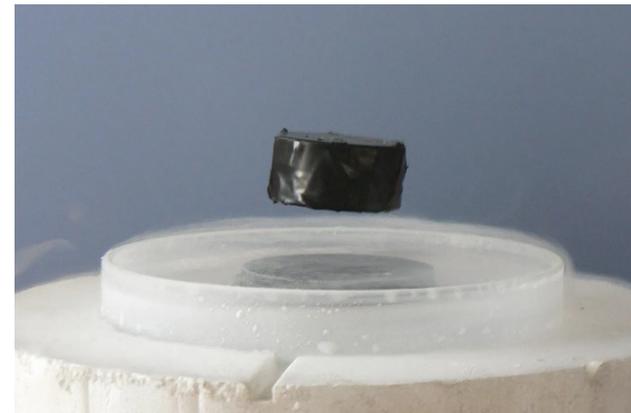
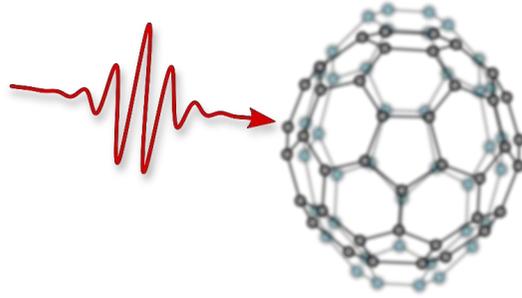
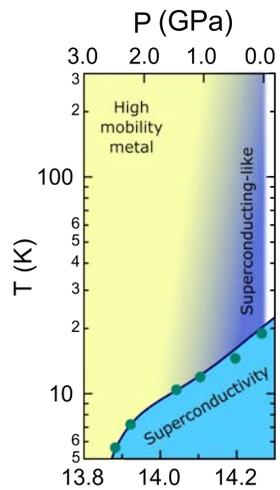
# Coherent squeezed mode explains optical properties



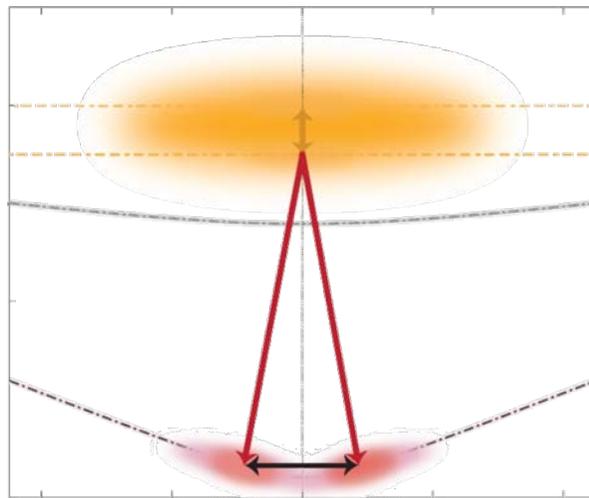
with Marios Michael, Eugene Demler



# Do these explain the optical and magnetic properties?

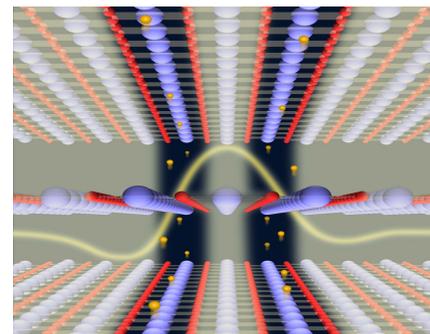
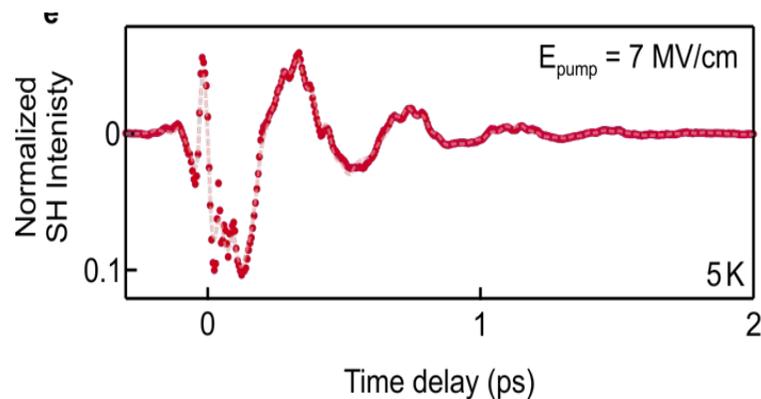


# Squeezed current/phase oscillations



Rather than amplification of the superconducting currents  $J_{q_x}$

The underlying physics may be connected to oscillations in the „noise“ of the current  $\langle J_{q_x} J_{-q_x} \rangle$



# Acknowledgements



N. Taherian



Alexander von Hoegen



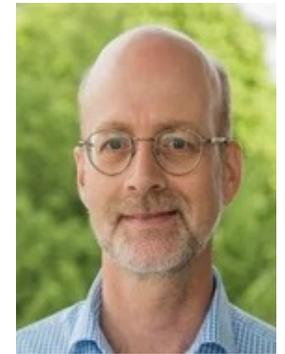
Michael Först



Albert Liu



Michael Fechner



Bernhard Keimer



Sebastian Fava



Giovanni De Vecchi



Michele Buzzi



Gregor Jotzu



Marios H. Michael



Eugene Demler