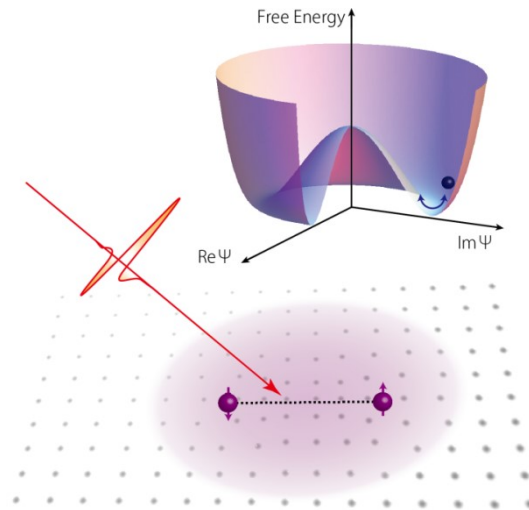


Nonequilibrium dynamics of superconductors

Ryo Shimano

Cryogenic Research Center and Department of Physics

University of Tokyo

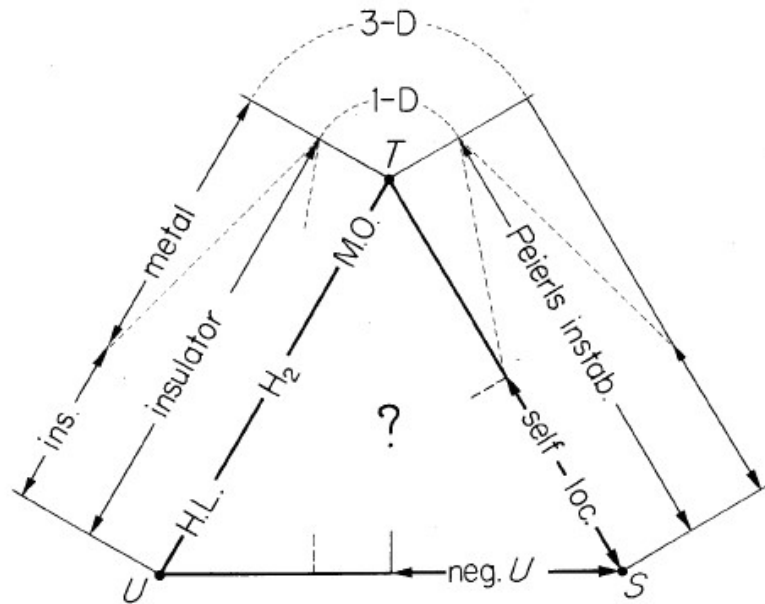


東京大学
THE UNIVERSITY OF TOKYO

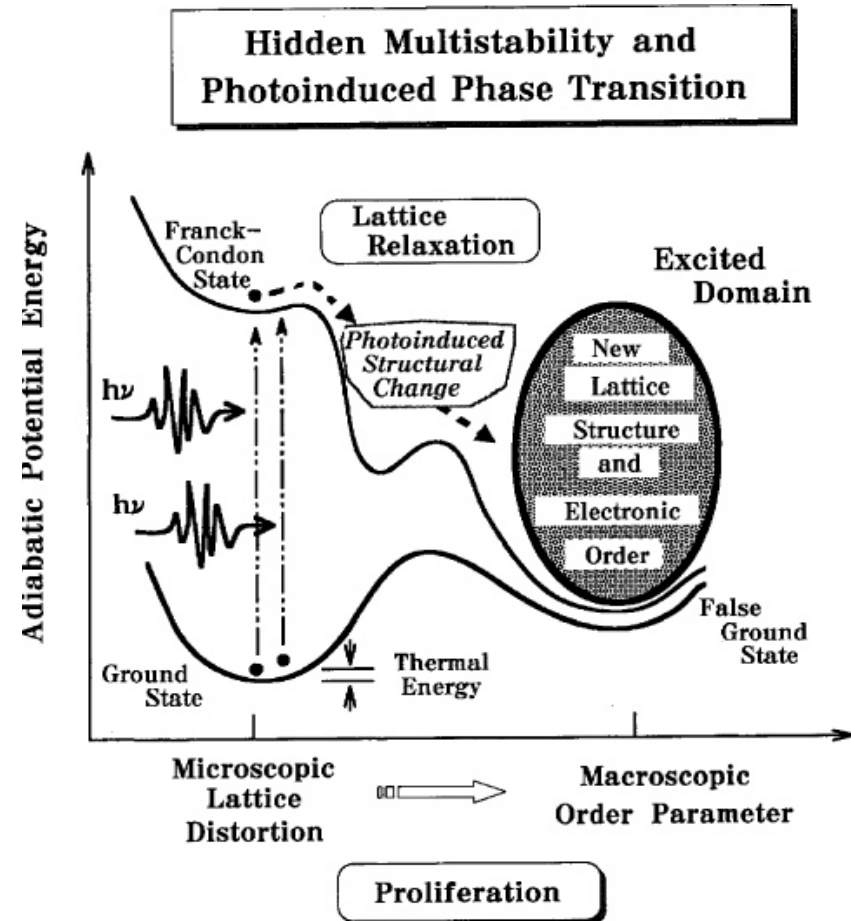
Outline

- (1) Introduction
- (2) Photoexcitation in s-wave superconductor
- (3) Higgs mode in a s-wave superconductor NbN
- (4) Higgs mode in d-wave cuprate superconductors
- (5) Photoinduced metastable phase

Concept of Photoinduced Phase Transition



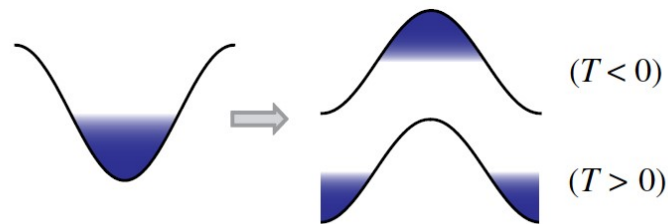
Yutaka Toyozawa,
 J. Phys. Soc. Jpn. **50**, 1861(1981)



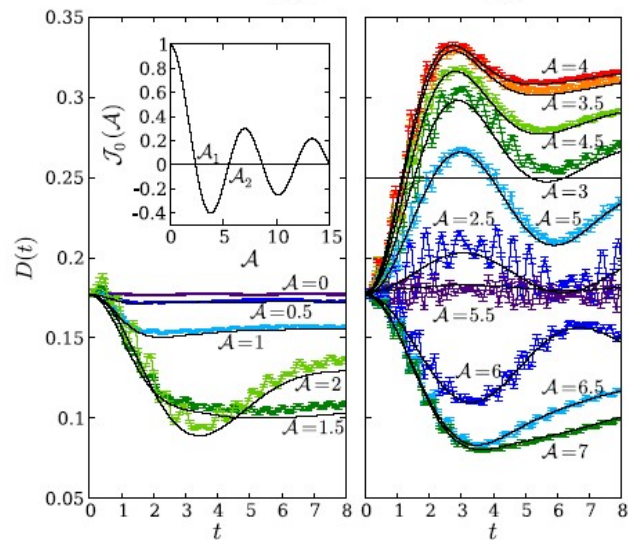
Keiichiro Nasu,
 Rep. Prog. Phys. **67**, 1607(2004)

Dynamical localization

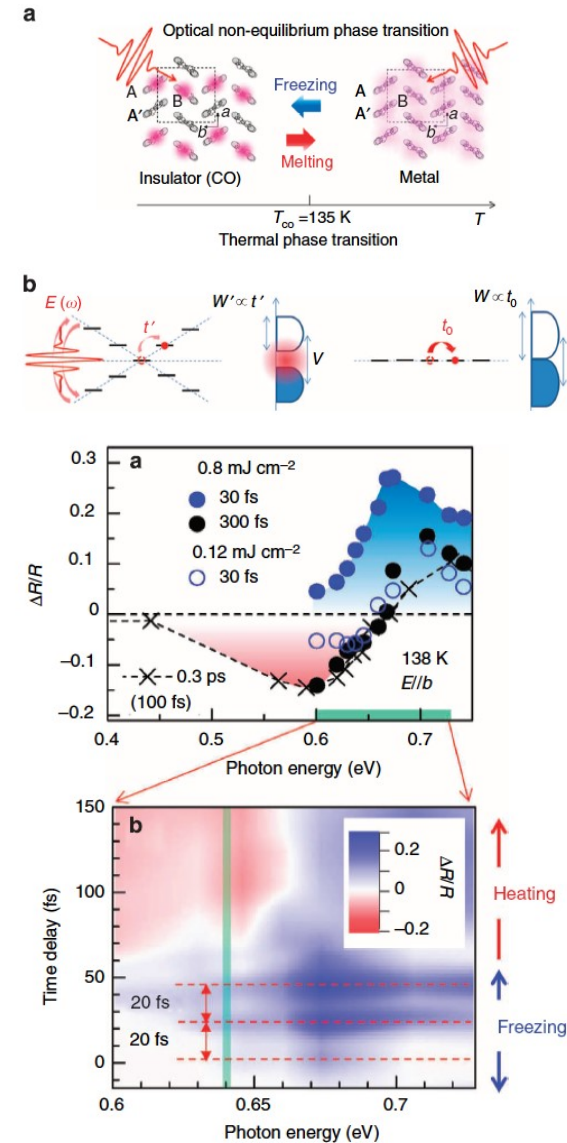
N. Tsuji, T. Oka, P. Werner, and H. Aoki
 Phys. Rev. Lett **106**, 236401(2011)



$$U \rightarrow U_{\text{eff}} = U/\mathcal{J}_0(\mathcal{A}).$$



Interaction Quench



T. Ishikawa et al., Nat. Commun. **5**, 5528(2014)

Photocreation of Berry phase

RAPID COMMUNICATIONS

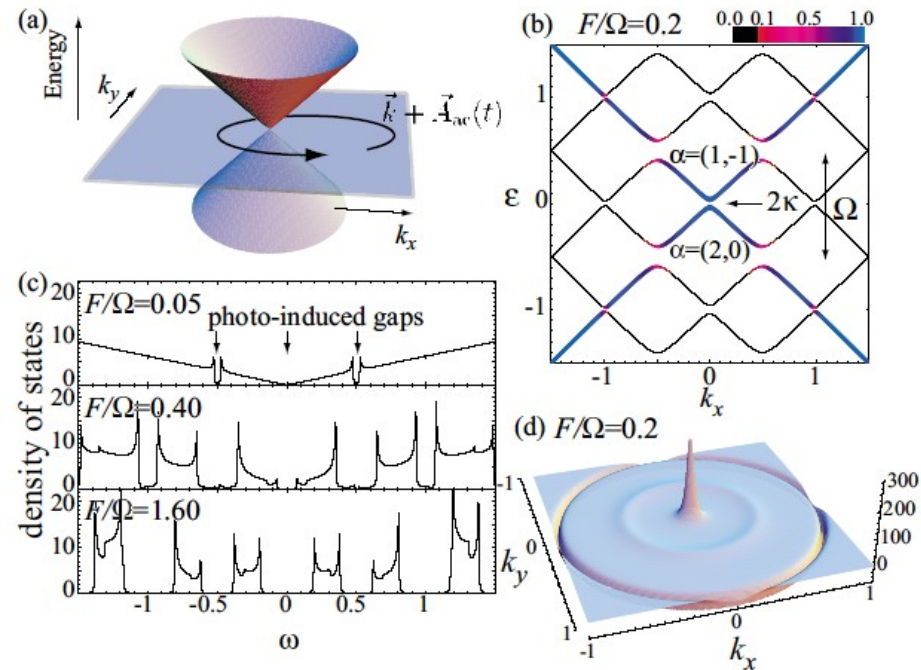
PHYSICAL REVIEW B 79, 081406(R) (2009)

Photovoltaic Hall effect in graphene

Takashi Oka and Hideo Aoki

Department of Physics, University of Tokyo, Hongo, Tokyo 113-0033, Japan

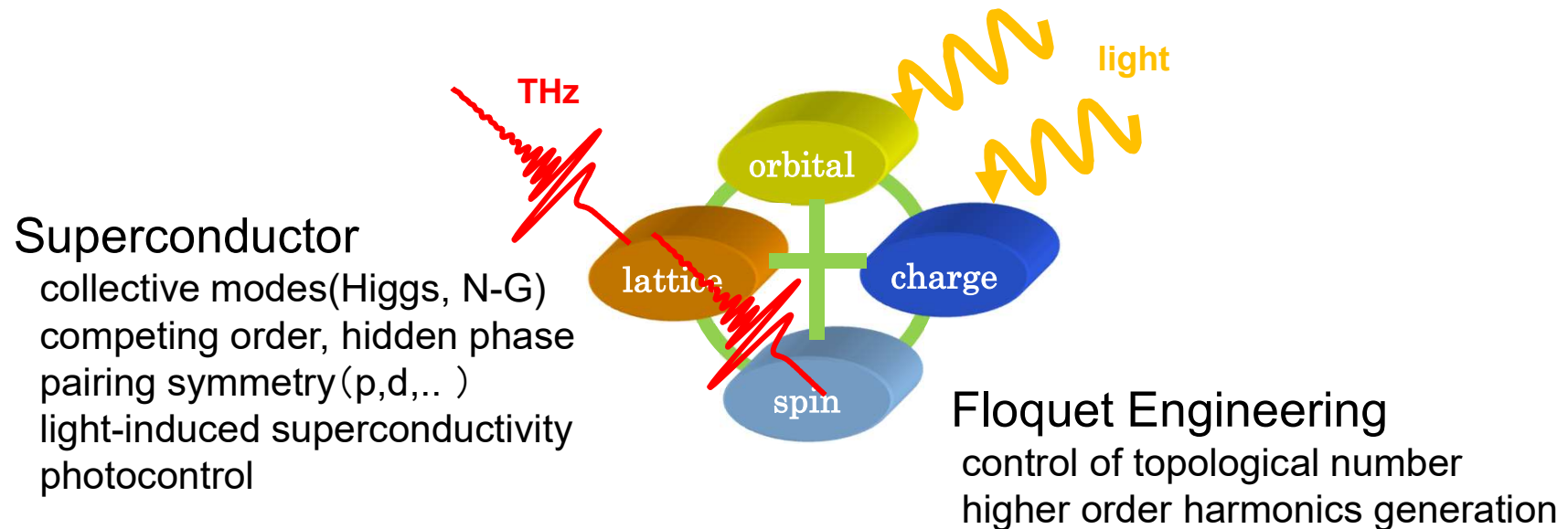
(Received 29 July 2008; revised manuscript received 15 January 2009; published 23 February 2009)



realized in cold atoms: “Experimental realization of the topological Haldane model with ultracold Fermions”, G. Jotzu et al., Nature **515**, 237 (2014)

Towards artificial light-control of quantum material

Nonequilibrium



Ultrafast control of multiferroics
electromagnon, skyrmion

Light-control of ferroelectricity

Light-control of magnetism

Advanced Light Source/Probe

Time resolved spectroscopy

ARPES

XRD

Electron Diffraction

Terahertz

+

Advanced light source

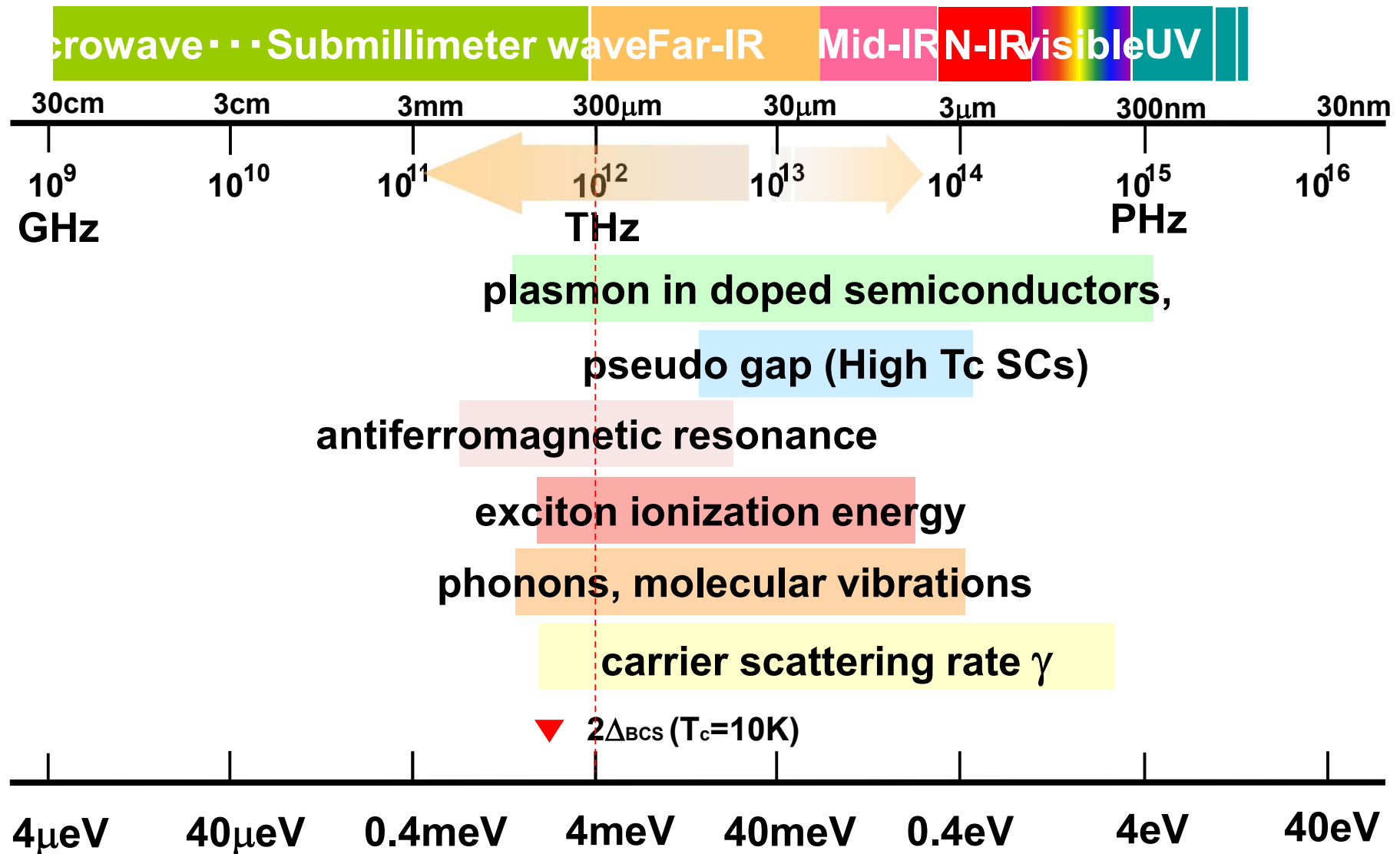
Intense THz pulse

mid-IR

fs~as optical pulse

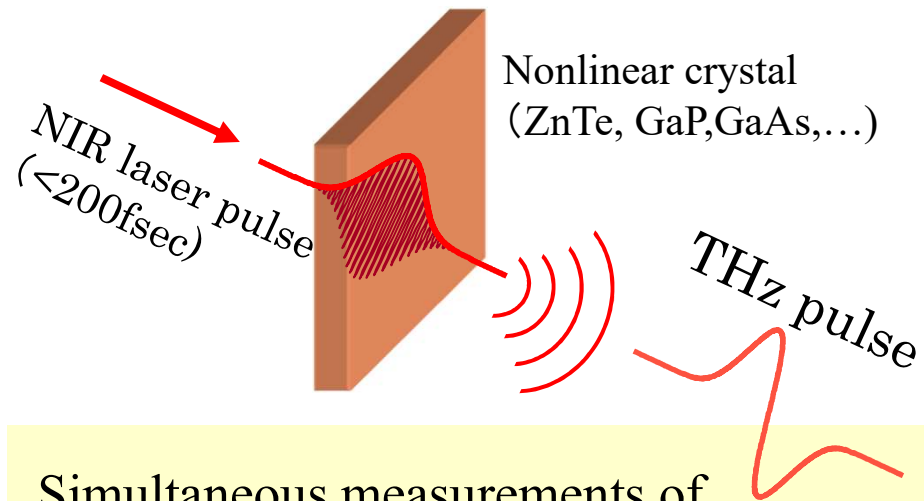
Elementary excitations in condensed matter systems

$$1\text{THz} = 4\text{meV} = 300\mu\text{m} = 33\text{cm}^{-1} \sim 50\text{K}$$



THz time-domain spectroscopy

THz generation by femtosecond laser pulse



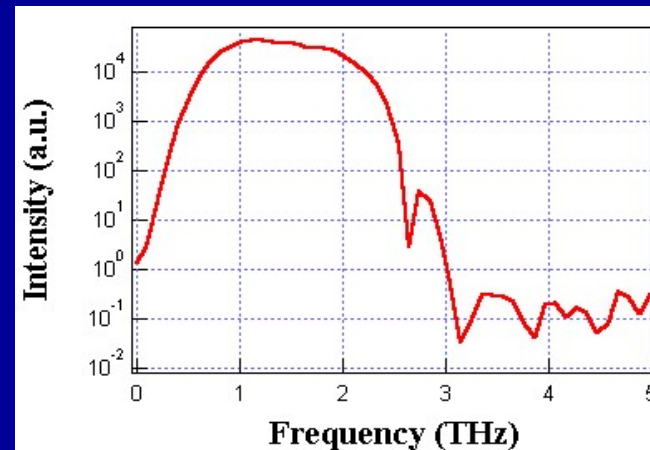
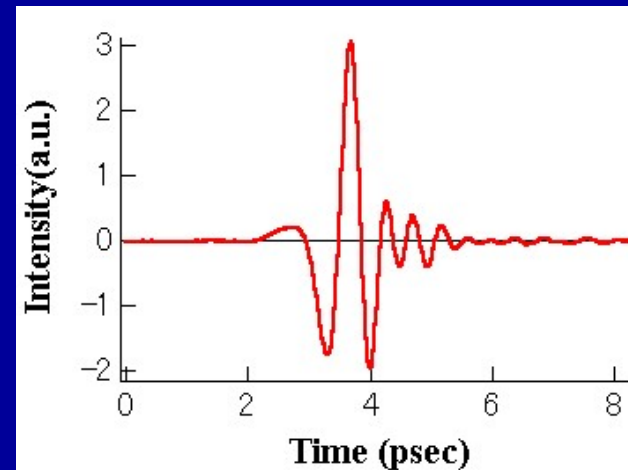
Simultaneous measurements of amplitude and phase of E-field

Determination of complex refractive Index without using Kramers-Kronig relation

Imaging applications

Time-resolved probe for ultrafast transient phenomena

Waveform and power spectrum



THz time-domain spectroscopy

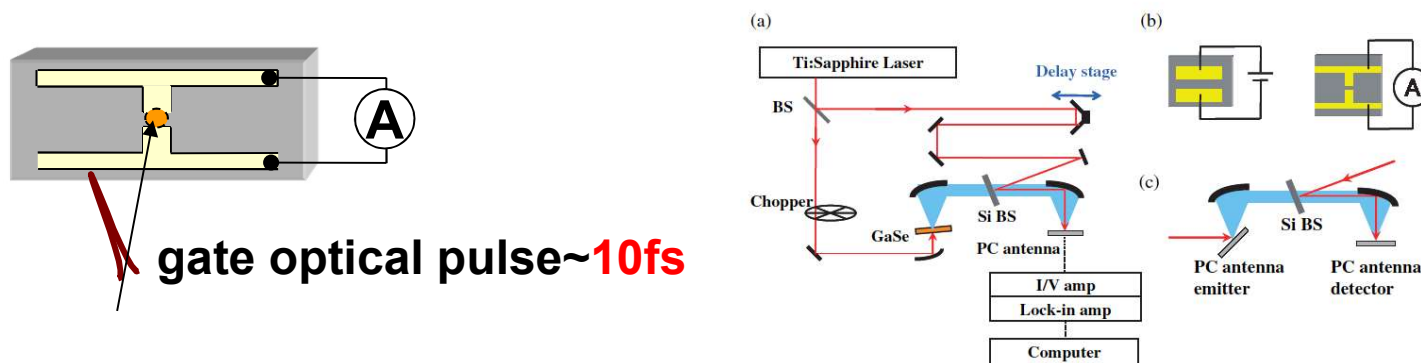


Fig. 1. Experimental setup for the first experiment: generation with GaSe crystal and detection with PC antenna. (b) Schematic geometry of PC antennas. Left: PC antenna with a large aperture used for generation. Right: Dipole-type PC antenna used for detection. (c) Modified part of the experimental arrangement for the second experiment: generation and detection using a PC antenna.

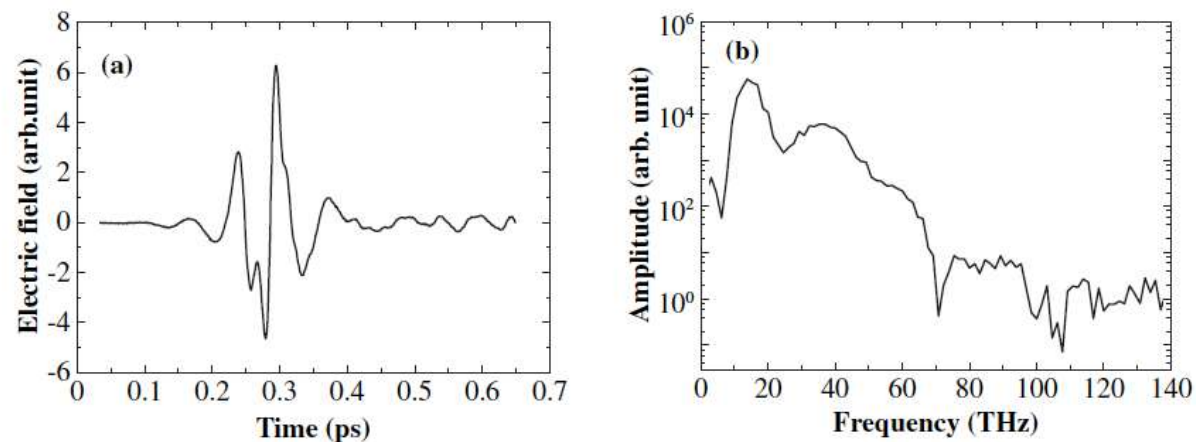


Fig. 2. (a) Temporal waveform of the THz electric field generated with the GaSe crystal and detected with the PC antenna. (b) Fourier-transformed amplitude spectrum of the waveform (a).

Intense THz pulse generation from LiNbO₃

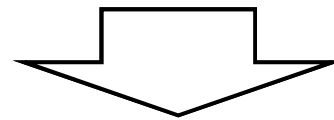
THz generation from LiNbO₃ : *tilted pulse-front method*

J. Hebling *et al.*, Opt. Express **10**, 1161 (2002).

J. Hebling *et al.*, J. Opt. Soc. Am. B **25**, B6 (2008).

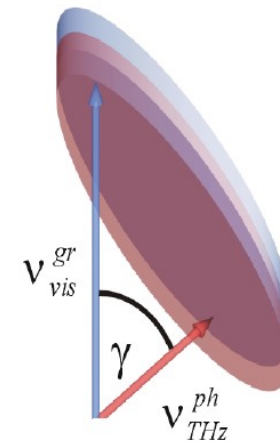
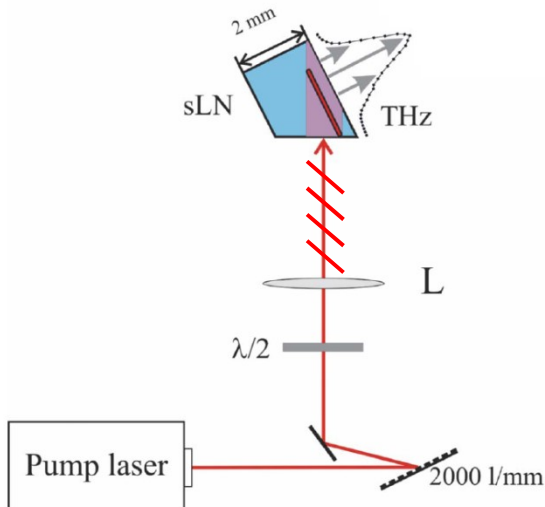
Nonlinear crystal	$\chi^{(2)}$ (pm/V)	$n_{800\text{nm}}^{\text{gr}}$	$n_{\text{THz}}^{\text{ph}}$
GaP	25	3.67	3.34
ZnTe	69	3.13	3.27
LiNbO ₃	168	2.25	4.96

Large $\chi^{(2)}$, but large phase mismatch



tilted pulse-front method

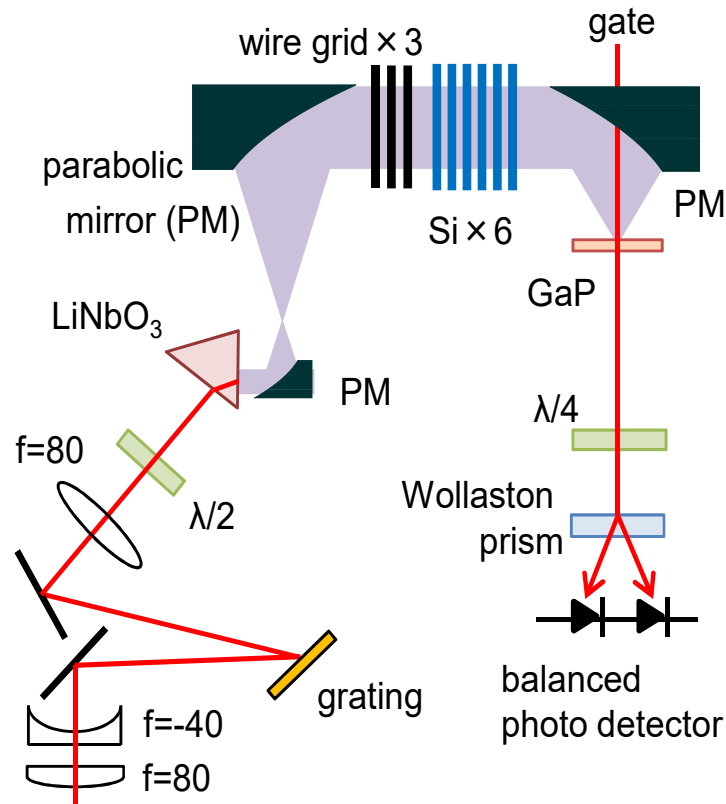
$$v_{\text{vis}}^{\text{gr}} \cdot \cos \gamma = v_{\text{THz}}^{\text{ph}}$$



Intense THz pulse generation

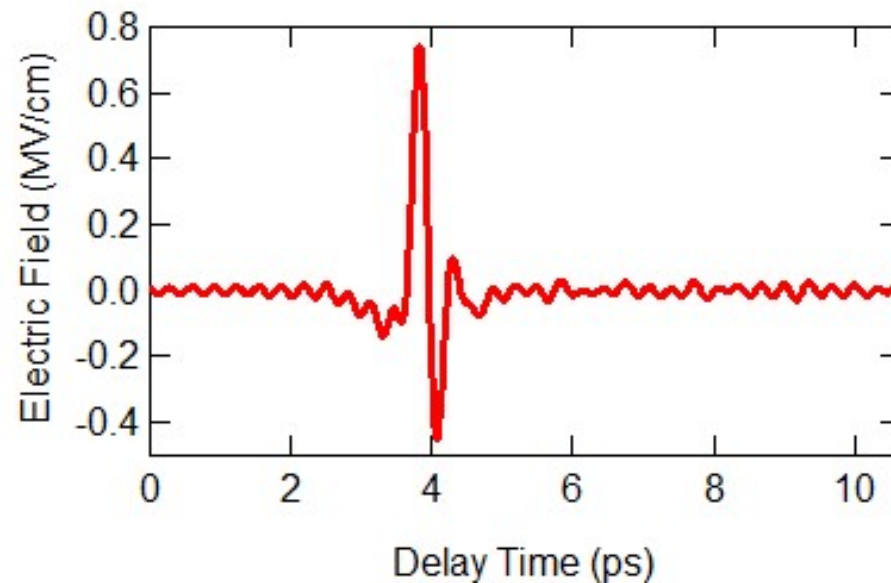
THz generation from LiNbO_3 : *tilted pulse-front method*

J. Hebling *et al.*,
Opt. Express **10**, 1161 (2002).

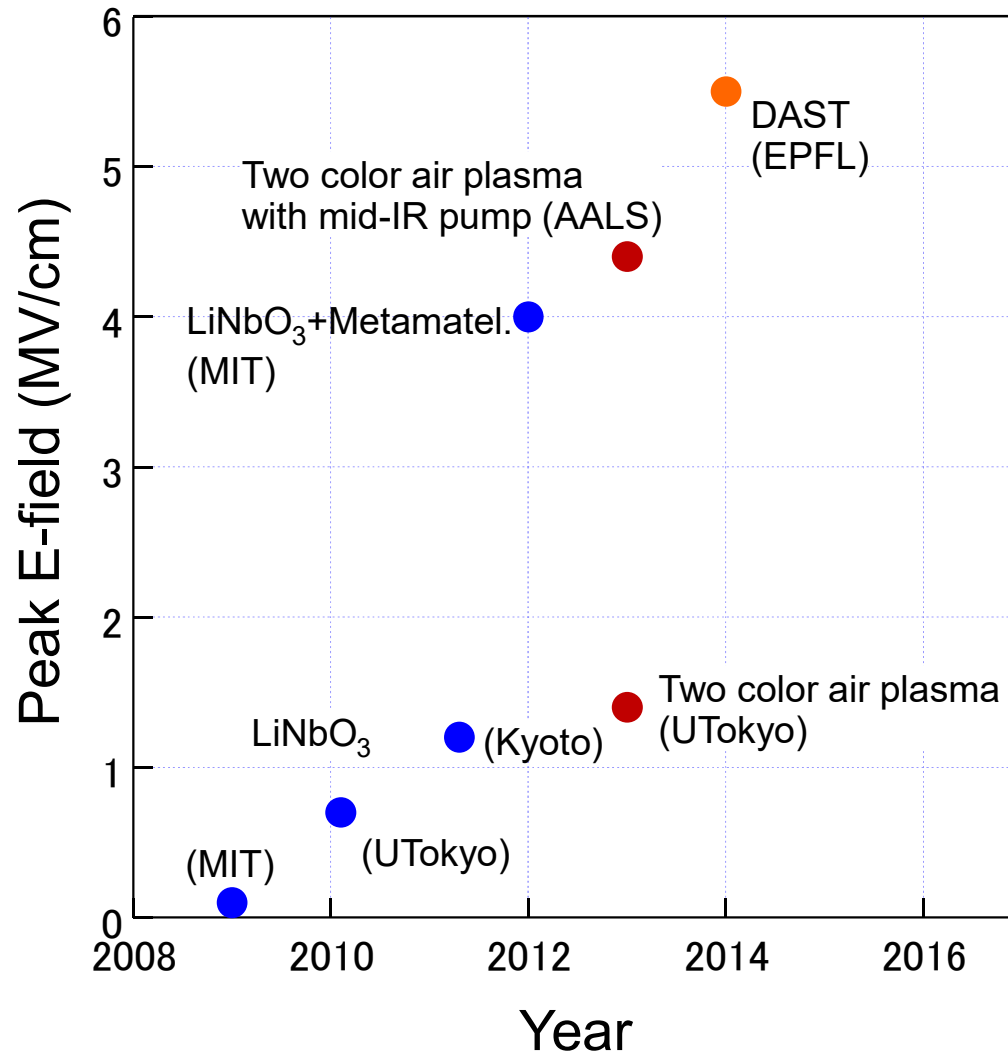


S. Watanabe, N. Minami, and R. Shimano,
Opt. Express **19**, 1528 (2011).

Tight focusing with small PM
100kV/cm \rightarrow 700kV/cm



Development laser-based table top THz pulse generation



$$E=6\text{MV/cm} \rightarrow B=2\text{T}$$

Outline

(1) Introduction

(2) Photoexcitation in s-wave superconductor

(3) Higgs mode in a s-wave superconductor NbN

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(5) Photocontrol of superconductors

Super-to-normal transition by quasiparticle injection

Two constraints:

Total electron density $\sum_{k,s} \langle c_{ks}^\dagger c_{ks} \rangle = N$

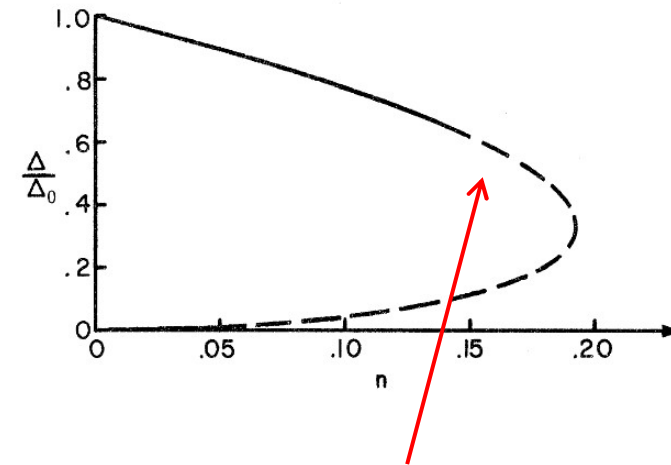
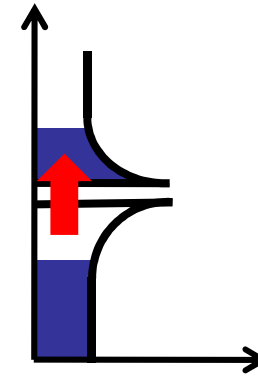
Total QP density $\sum_{k,s} f_{ks} = N_q \ll N$



μ^* model $f_{ks} = [1 + \exp \beta(E_k - \mu^*)]^{-1}$

Gap eq. $\frac{1}{N(0)V} = \int_{-\omega_c}^{\omega_c} \frac{d\varepsilon_k}{2E_k} \tanh \frac{1}{2} \beta(E_k - \mu^*)$

$$\left(\frac{\Delta}{\Delta_0}\right)^3 = \left\{ \sqrt{\left(\frac{\Delta}{\Delta_0}\right)^2 + n^2} - n \right\}$$



First order like transition

C. S. Owen *et al.*, Phys. Rev. Lett. 28, 1559 (1972).

Experiments in 1970's

PHYSICAL REVIEW B

VOLUME 4, NUMBER 7

1 OCTOBER 1971

Destruction of Superconductivity by Laser Light

L. R. Testardi

Bell Telephone Laboratories, Murray Hill, New Jersey 07974

(Received 27 January 1971)

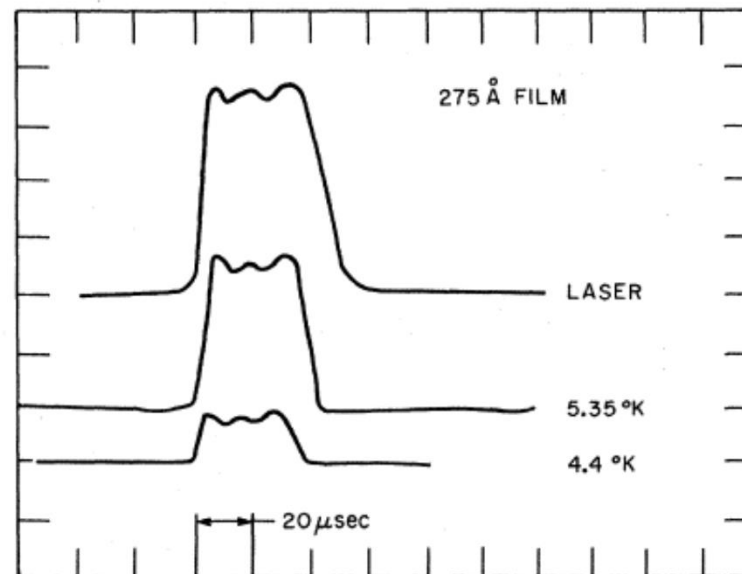


FIG. 3. 275-Å-thick sample resistance (lower curve) and laser output (upper curve) vs time. Horizontal scale 20 μsec/div. For $T=4.4$ °K the sample resistance during laser pulse is $0.06R_N$. For $T=5.35$ °K the sample resistance during laser pulse is $0.15R_N$.

Experiments in 1990's: THz spectroscopy

PHYSICAL REVIEW B

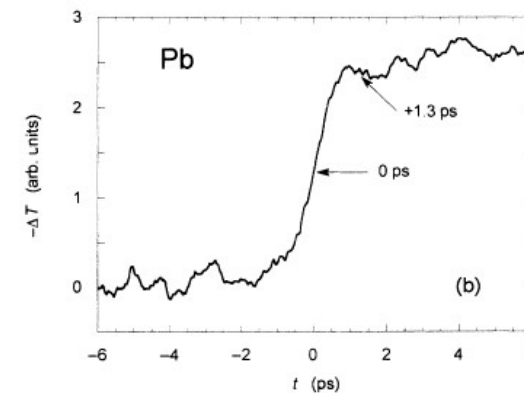
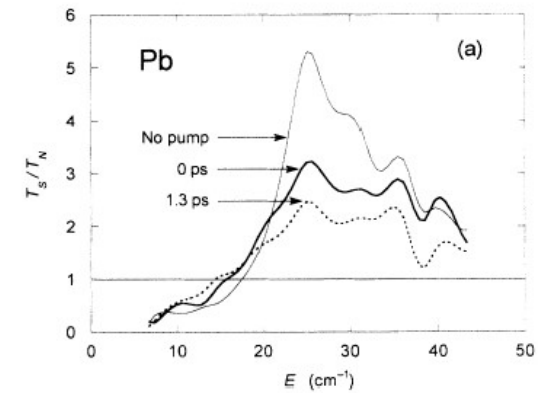
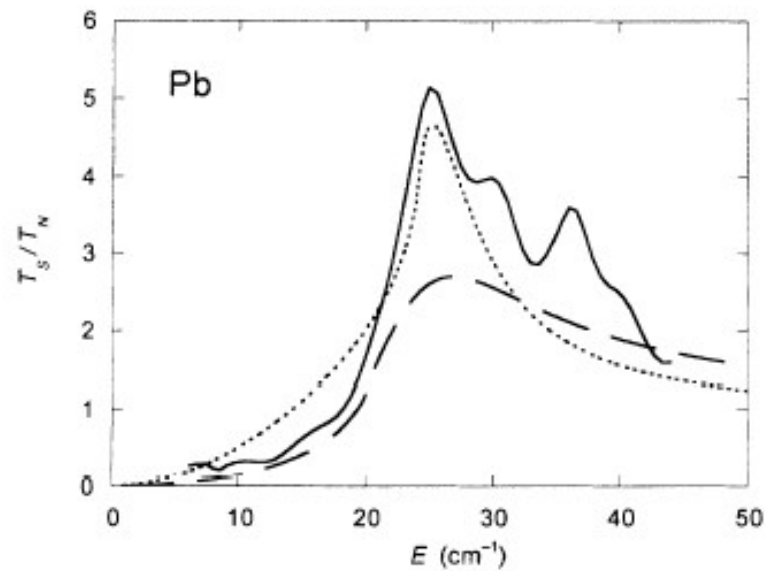
VOLUME 46, NUMBER 17

1 NOVEMBER 1992-I

Direct picosecond measurement of photoinduced Cooper-pair breaking in lead

J. F. Federici, B. I. Greene, P. N. Saeta, and D. R. Dykaar
AT&T Bell Laboratories, 600 Mountain Avenue, Murray Hill, New Jersey 07974

F. Sharifi and R. C. Dynes
Department of Physics, University of California at San Diego, San Diego, California 92093
(Received 4 May 1)



SDW gap dynamics in quasi-1D organic conductor

RAPID COMMUNICATIONS

PHYSICAL REVIEW B **80**, 220408(R) (2009)



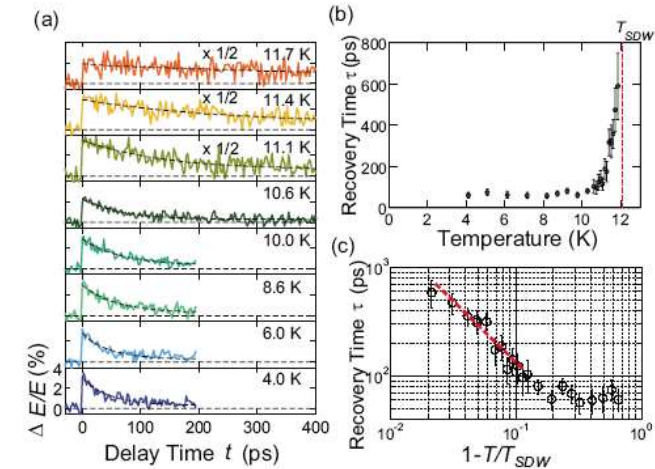
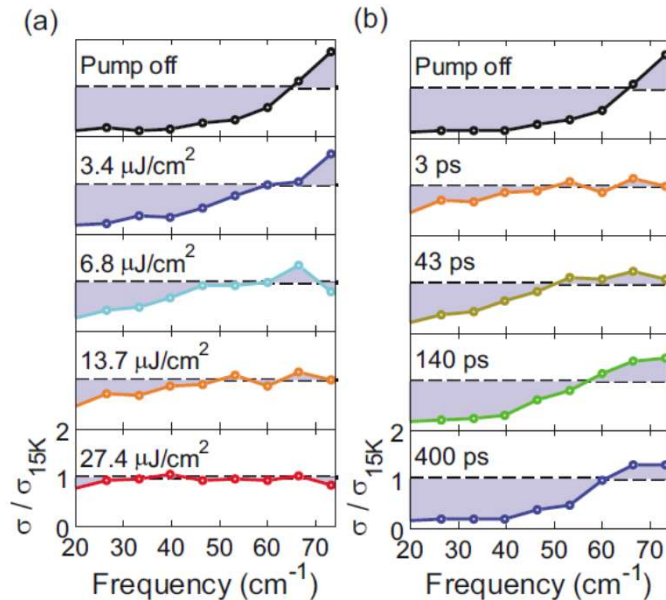
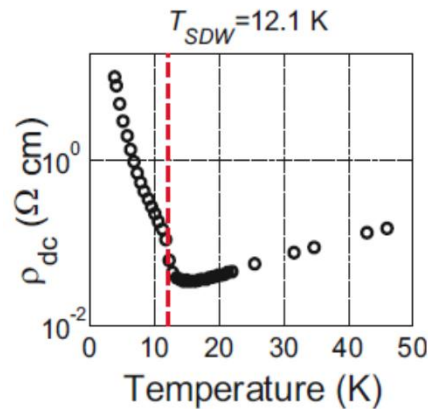
Observation of ultrafast photoinduced closing and recovery of the spin-density-wave gap in $(\text{TMTSF})_2\text{PF}_6$

Shinichi Watanabe,¹ Ryusuke Kondo,² Seiichi Kagoshima,² and Ryo Shimano^{1,*}

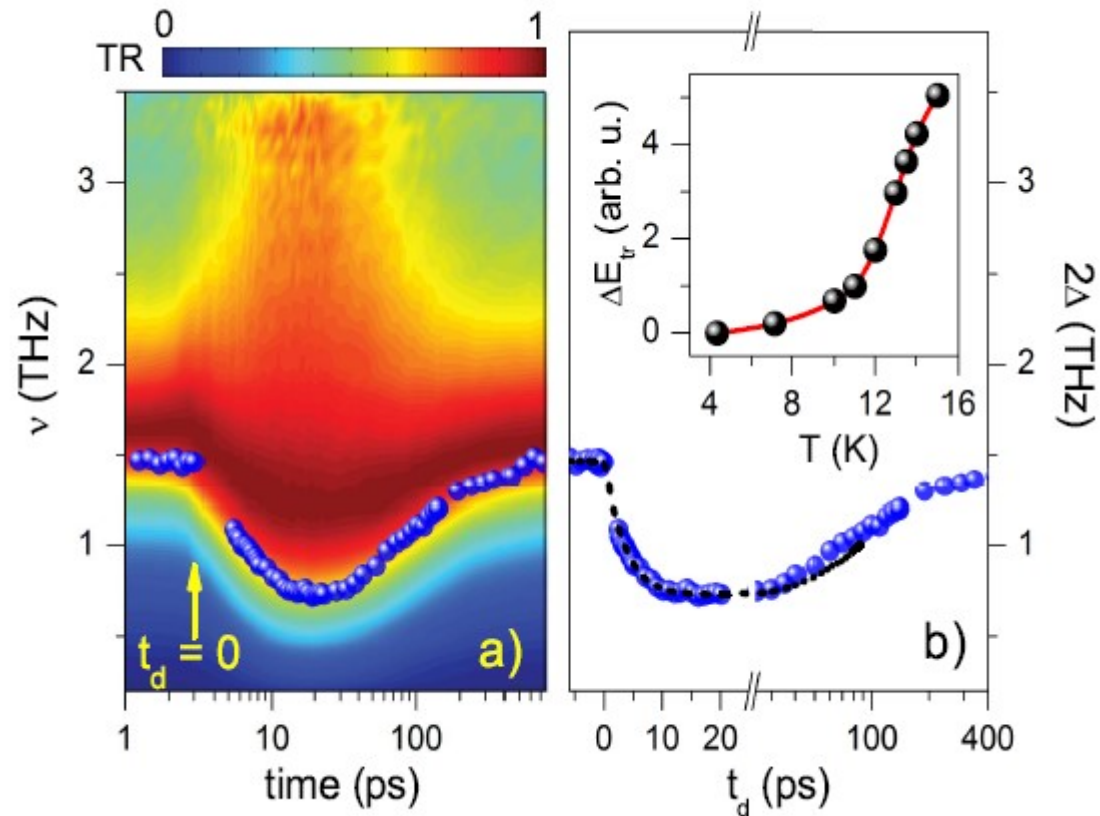
¹Department of Physics, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-0033, Japan

²Department of Basic Science, The University of Tokyo, 3-8-1 Komaba, Meguro-ku, Tokyo 153-8902, Japan

(Received 13 October 2009; published 17 December 2009)



Optical pump and THz probe experiment in a s-wave superconductor NbN

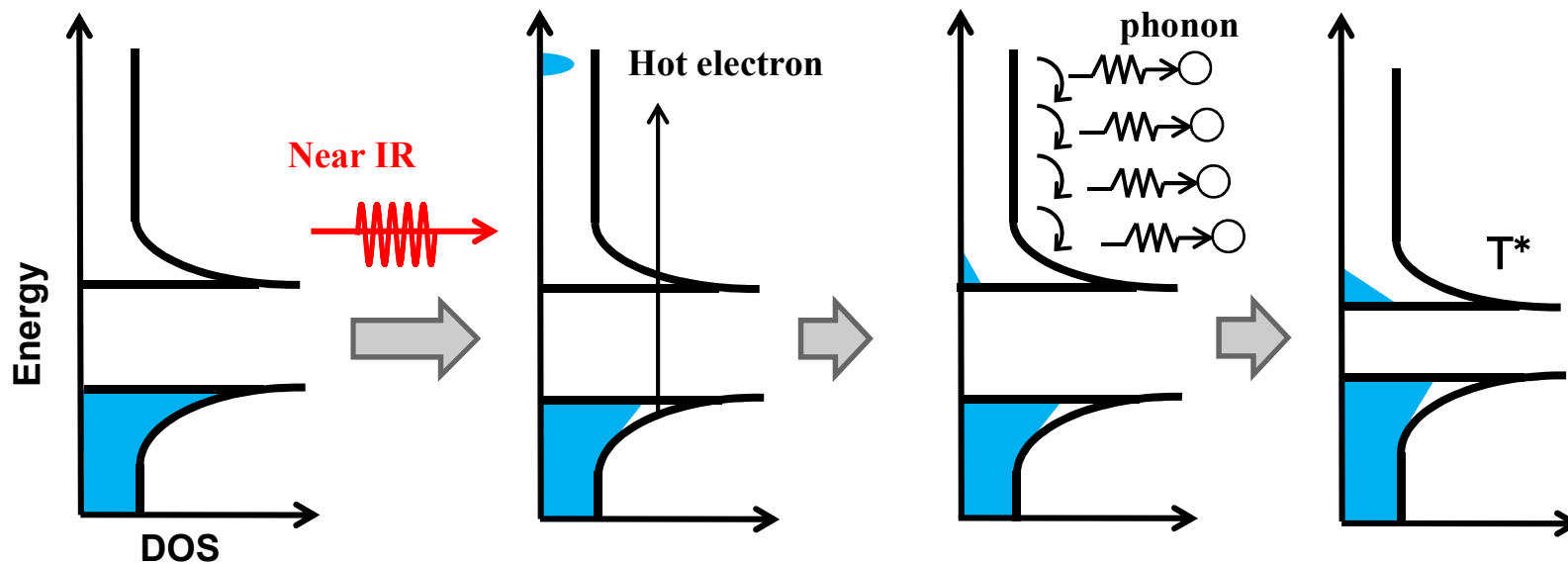


M. Beck *et al.*, Phys. Rev. Lett. **107**, 177007 (2011).

Near infrared excitation

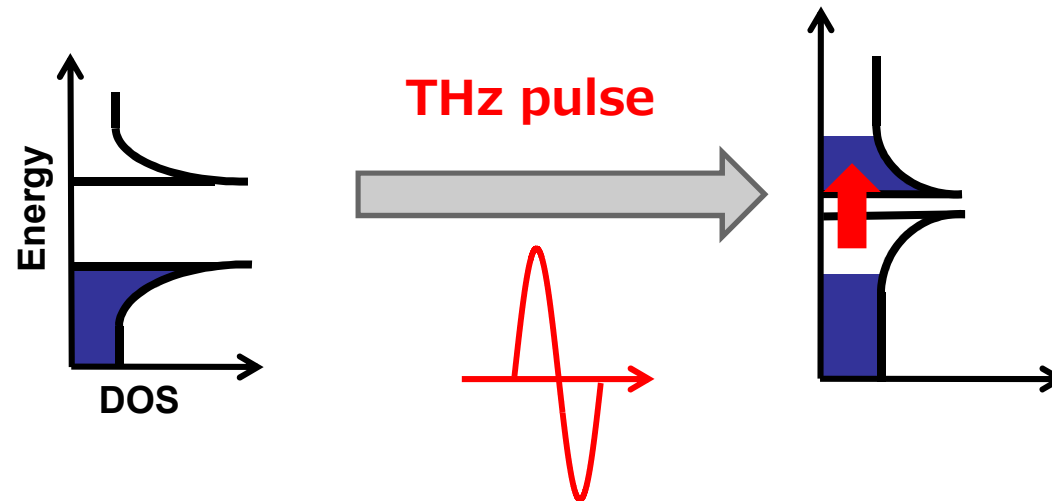
$$\Delta = V \int_{\Delta}^{\hbar\omega_D} d\varepsilon \frac{\Delta}{\sqrt{\varepsilon^2 - \Delta^2}} [1 - 2f(\varepsilon)]$$

- ① hot electron excitation by near infrared light
- ② relaxation of hot electrons through high energy emission
- ③ Cooper pair breaking by phonons
- ④ gradual suppression of superconductivity



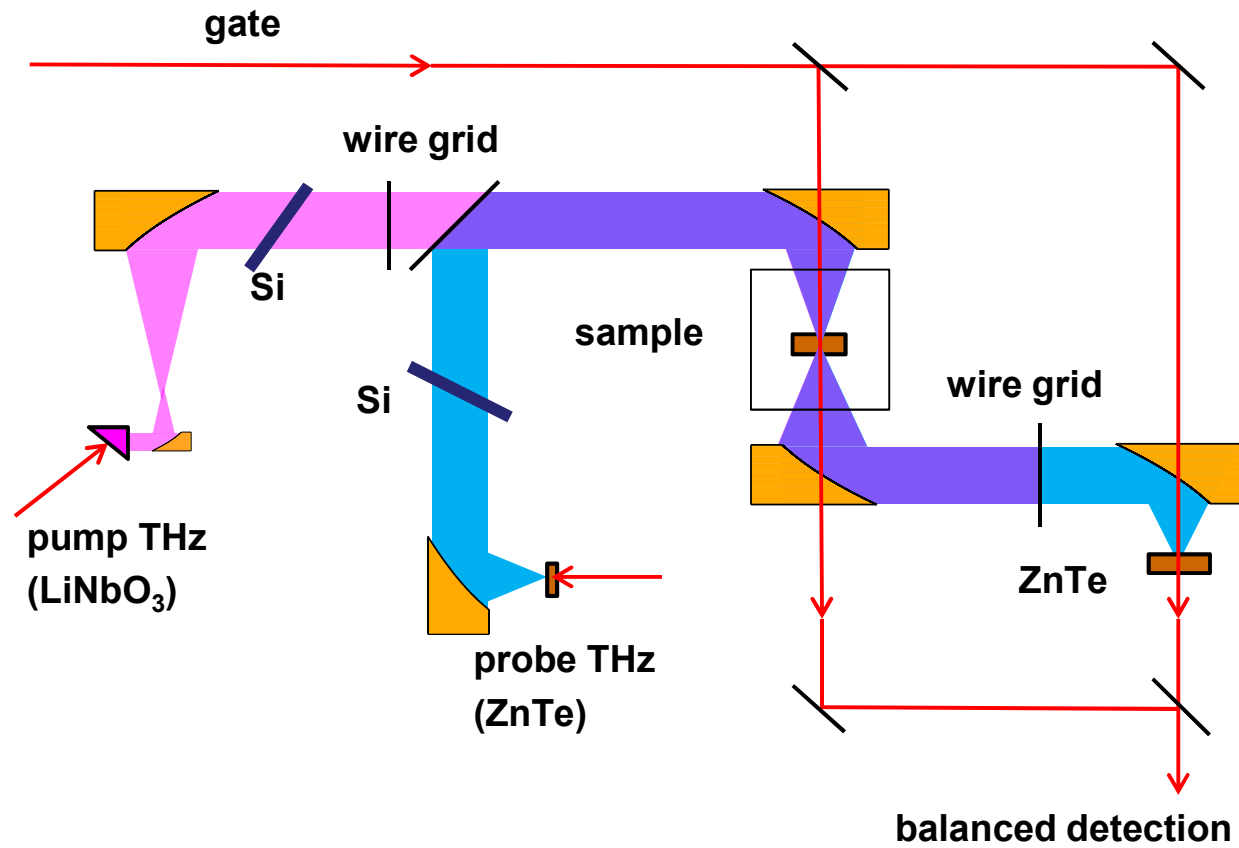
THz pumping: high density QP injection at the gap edge

$$\Delta = V \int_{\Delta}^{\hbar\omega_D} d\varepsilon \frac{\Delta}{\sqrt{\varepsilon^2 - \Delta^2}} [1 - 2f(\varepsilon)]$$



- direct injection of QPs at the gap edge
- nonequilibrium SC state dynamics

THz pump THz probe experiment



THz pump and THz probe in NbN

PRL 109, 187002 (2012)

PHYSICAL REVIEW LETTERS

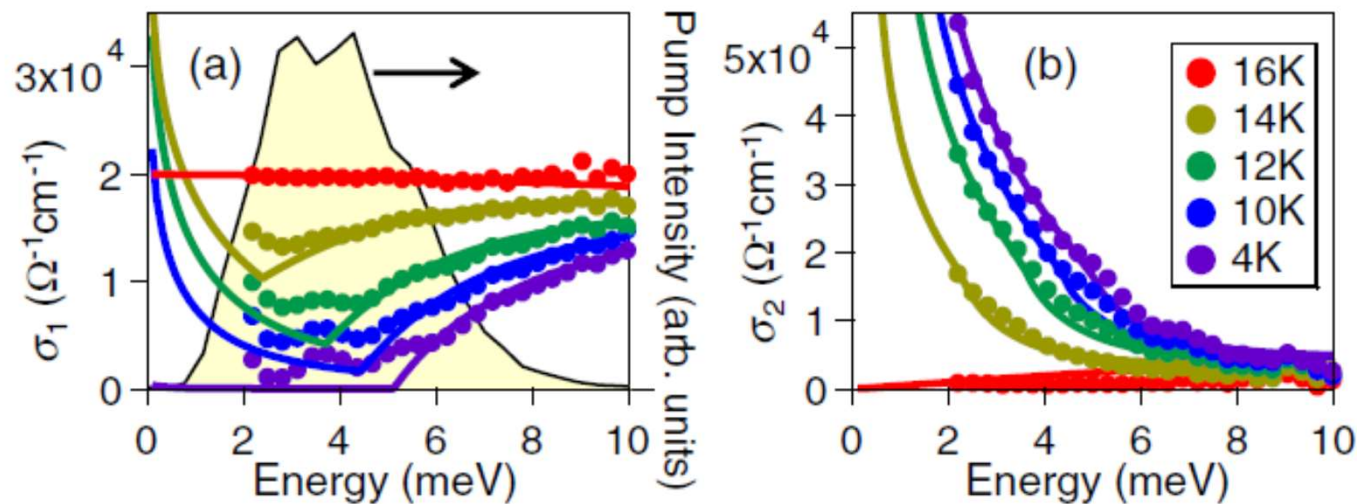
week ending
2 NOVEMBER 2012

Nonequilibrium BCS State Dynamics Induced by Intense Terahertz Pulses in a Superconducting NbN Film

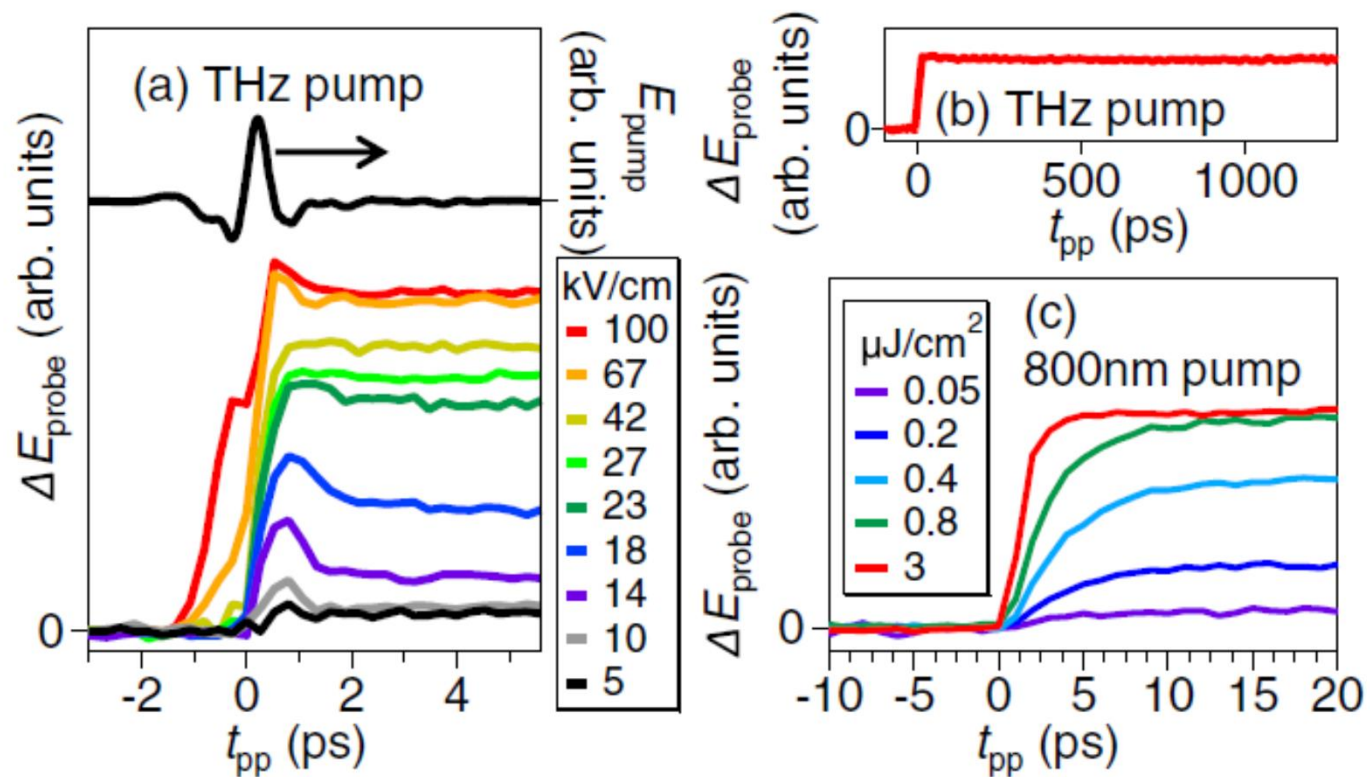
Ryusuke Matsunaga and Ryo Shimano

Department of Physics, The University of Tokyo, Tokyo, 113-0033, Japan

(Received 10 July 2012; published 31 October 2012)



THz pump and THz probe dynamics



Order parameter dynamics in the BCS approximation

Quench Problem:

rapid switching of the orientation of $\mathbf{b}_k^{\text{eff}}$ faster than the response time of the pseudospin

$$\frac{d}{dt} \boldsymbol{\sigma}_k = 2\mathbf{b}_k^{\text{eff}} \times \boldsymbol{\sigma}_k$$

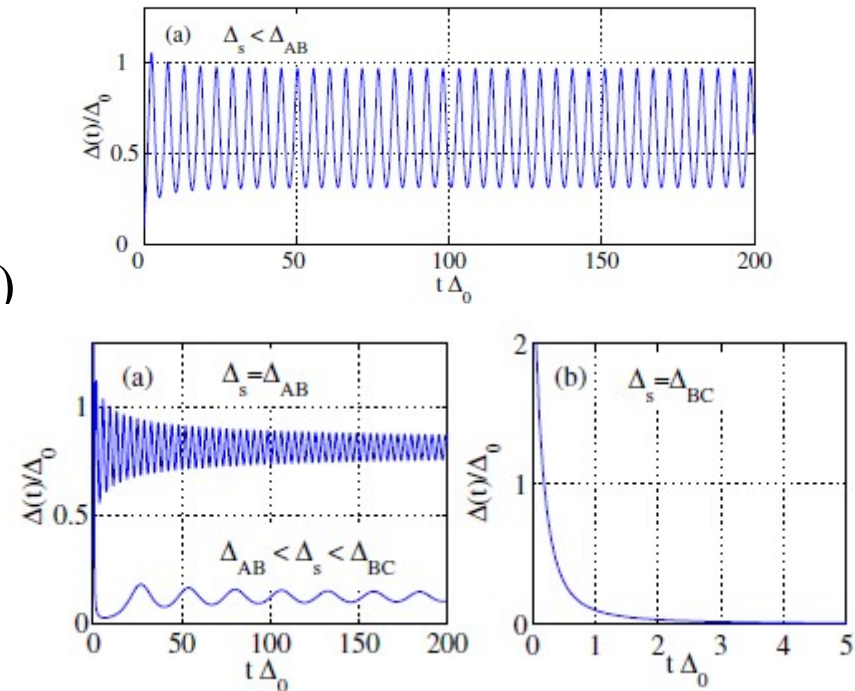
$$\Delta'(t) + i\Delta''(t) = -V \sum_{\mathbf{k}} (\sigma_k^x(t) + i\sigma_k^y(t))$$

$$\mathbf{b}_k^{\text{eff}} = (-\Delta'(t), -\Delta''(t), \varepsilon_k)$$

Order parameter change induced by external perturbation

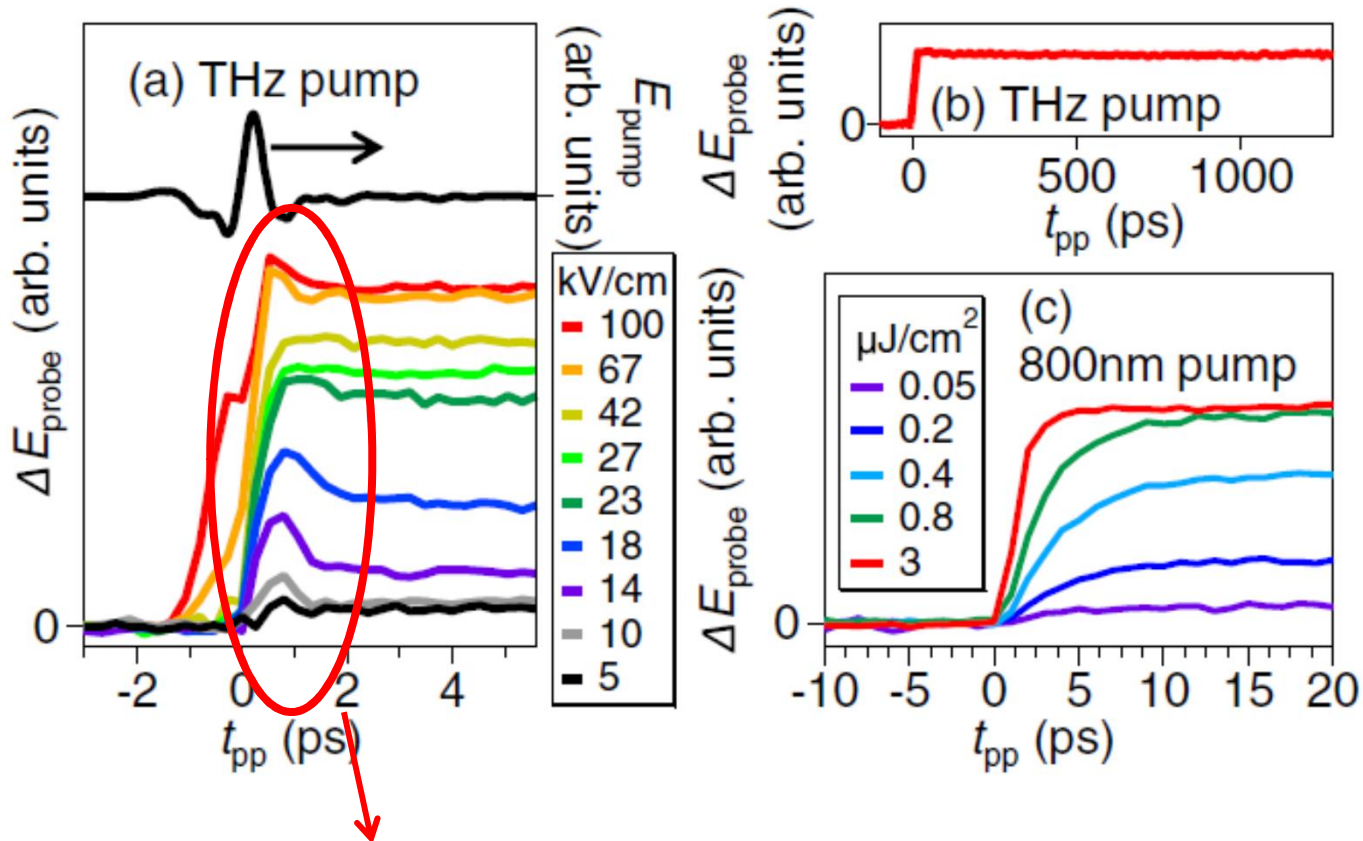
= change in the orientation of $\mathbf{b}_k^{\text{eff}}$

⇒ Collective precession of the pseudospin
= order parameter oscillation (**Higgs mode**)



Barankov and Levitov,
PRL **96**, 230403 (2006)

THz pump and THz probe dynamics



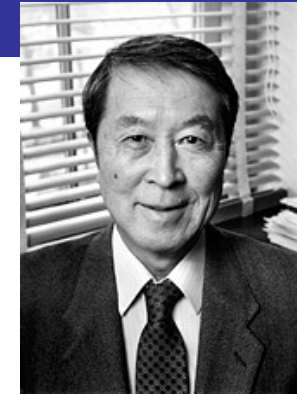
- What is this overshooting signal? Higgs?

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- (1) Introduction
- (2) Photoexcitation in s-wave superconductor
- (3) Higgs mode in a s-wave superconductor NbN
- (4) Higgs mode in d-wave cuprate superconductors
- (5) Photocontrol of superconductors

History

- 1957 BCS theory of superconductor (Bardeen, Cooper & Schrieffer)
- 1958 Prediction of amplitude mode in superconductors (Anderson)
- 1960 Theory of spontaneous symmetry breaking (Nambu)
- 1960-61 Nambu-Goldstone theorem
- 1963-66 Anderson-Higgs mechanism (Anderson, Higgs)



<http://www.nobelprize.org/>

BCS theory:

the nonzero order parameter

$$\Delta(\mathbf{k}) = - \sum_{\mathbf{k}'} V(\mathbf{k}, \mathbf{k}') \langle c_{\mathbf{k}'\uparrow} c_{-\mathbf{k}'\downarrow} \rangle \neq 0$$

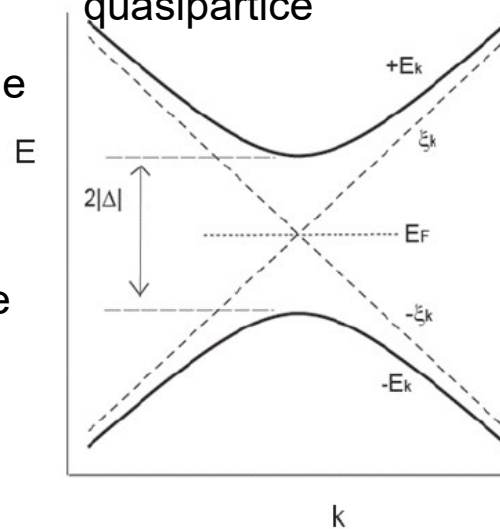
breaks the invariance of the gauge transformation

$$c \rightarrow c e^{i\theta}, c^\dagger \rightarrow c^\dagger e^{-i\theta}$$

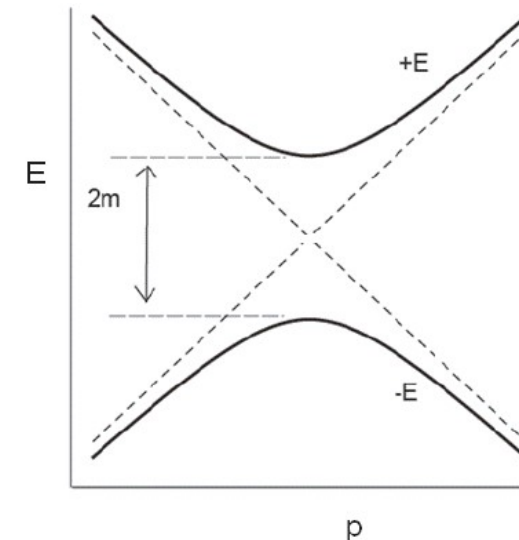
The dispersion of the quasiparticle

$$E(\mathbf{k}) = \sqrt{\xi(\mathbf{k})^2 + |\Delta(\mathbf{k})|^2}$$

Energy dispersion of BCS Bogoliubov quasiparticle



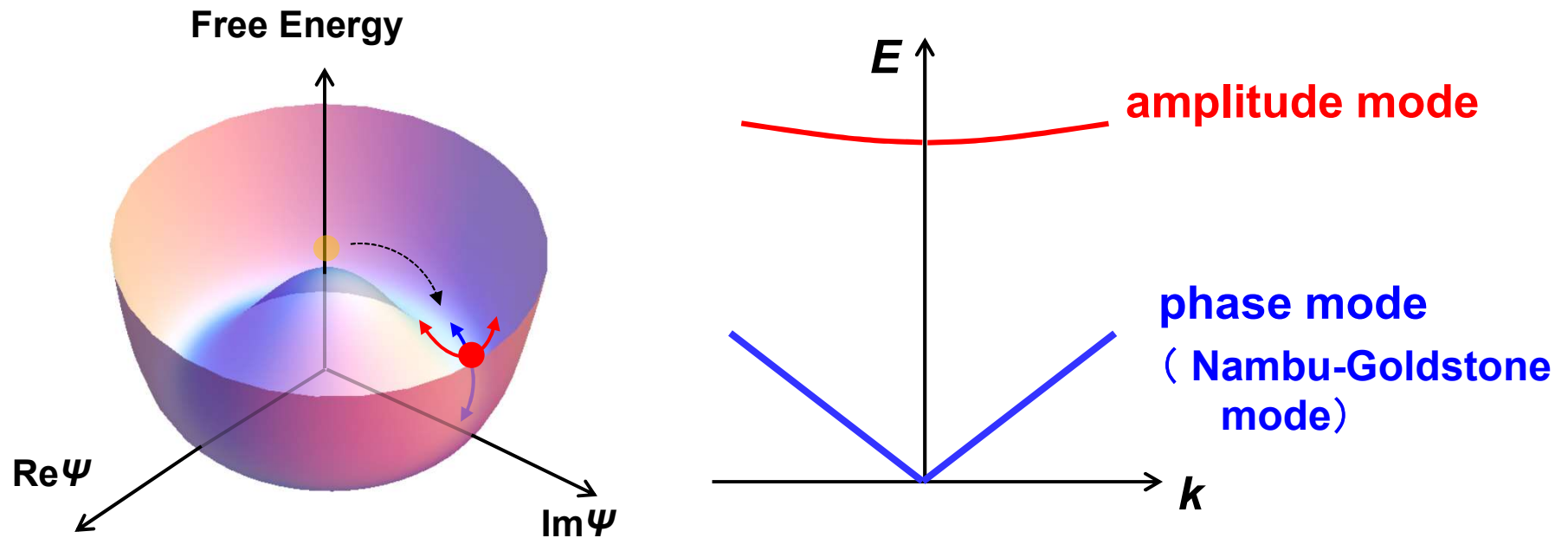
Energy dispersion of particle and antiparticle



$$E(\mathbf{k}) = \pm \sqrt{k^2 + m^2}$$

Goldstone Theorem

When spontaneous symmetry breaking occurs, massless collective mode with respect to the order parameter appears

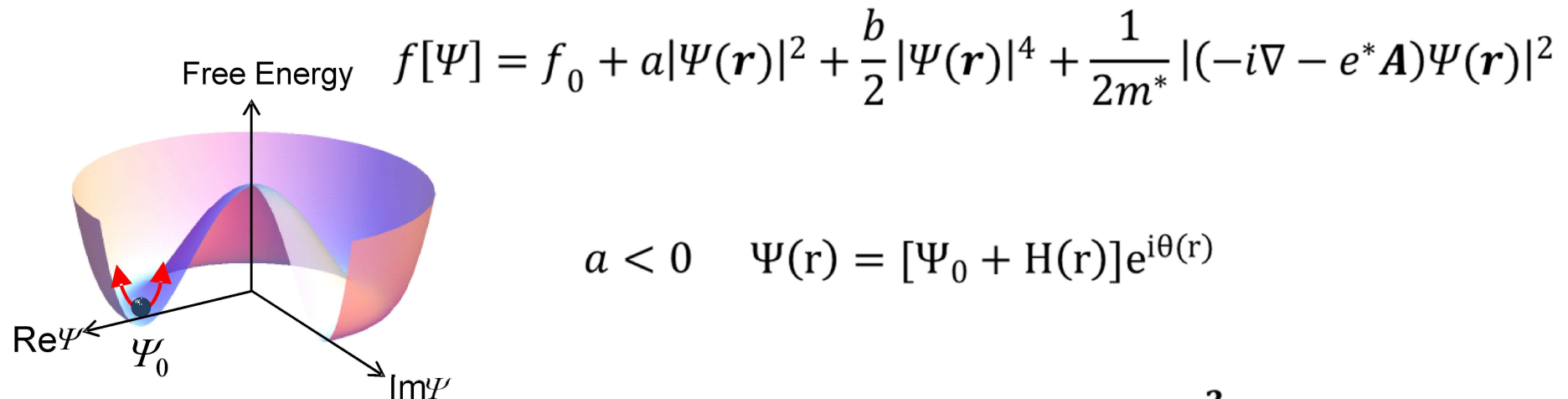


In particle physics: such a massless Nambu-Goldstone boson has never been observed. Instead, massive gauge bosons (W, Z) were found.



Is N-G theorem wrong?

Anderson-Higgs mechanism



$$f[\Psi] = f_0 + a|\Psi(\mathbf{r})|^2 + \frac{b}{2}|\Psi(\mathbf{r})|^4 + \frac{1}{2m^*}|(-i\nabla - e^*\mathbf{A})\Psi(\mathbf{r})|^2$$

$$a < 0 \quad \Psi(\mathbf{r}) = [\Psi_0 + H(\mathbf{r})]e^{i\theta(\mathbf{r})}$$

$$f = -2aH^2 + \frac{1}{2m^*}(\nabla H)^2 + \frac{e^{*2}}{2m^*}\left(\mathbf{A} - \frac{1}{e^*}\nabla\theta\right)^2(\Psi_0 + H)^2 + \dots$$

Local gauge transformation $A' = A - \nabla\theta/e^*$ $A' \rightarrow A$

$$f = -2aH^2 + \frac{1}{2m^*}(\nabla H)^2 + \frac{e^{*2}\Psi_0^2}{2m^*}A^2 + \frac{e^{*2}\Psi_0}{m^*}A^2H + \dots$$

massive amplitude mode

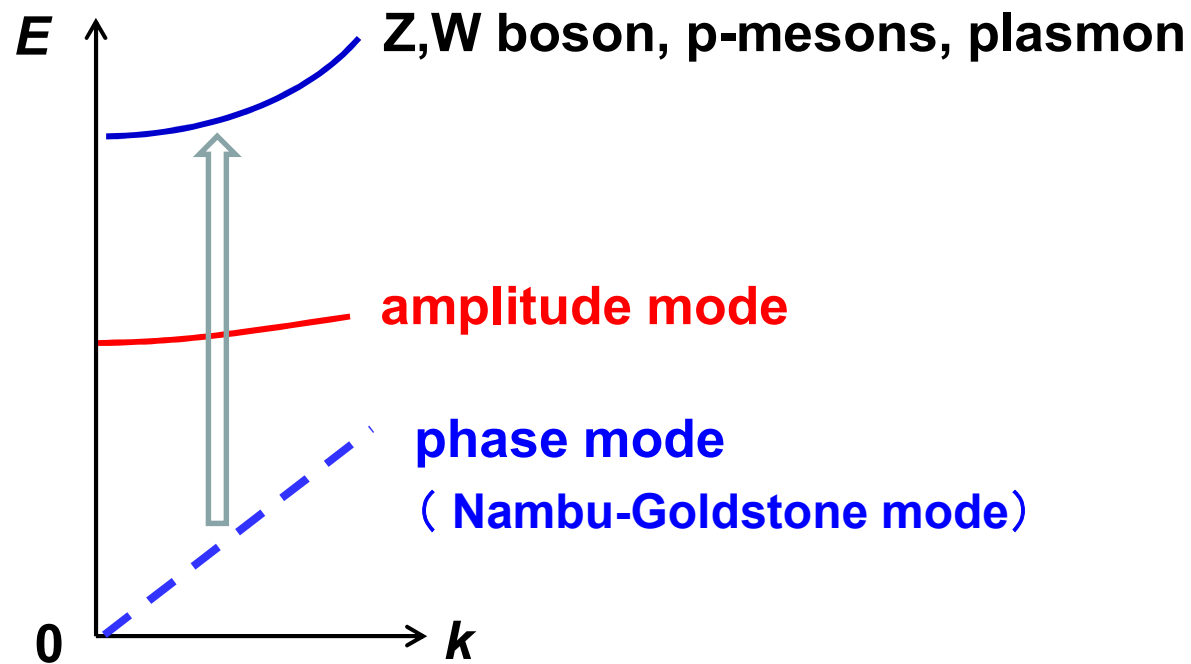
massive gauge boson

Anderson-Higgs mechanism

“Anderson-Higgs mechanism” or “Brout-Englert-Higgs mechanism”

“ABEGHHK'tH mechanism”

[for Anderson, Brout, Englert, Guralnik, Hagen, Higgs, Kibble and 't Hooft]

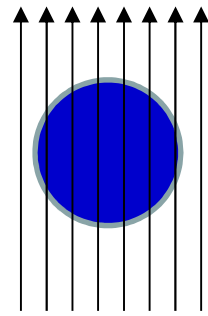


Massive gauge boson(photon) eating N-G mode in superconductors

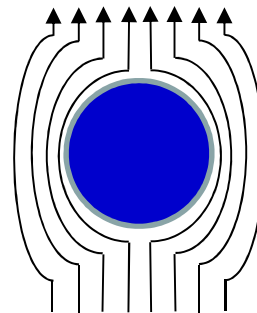
Meissner-Ochsenfeld effect 1933



Meissner



$T > T_c$



$T < T_c$

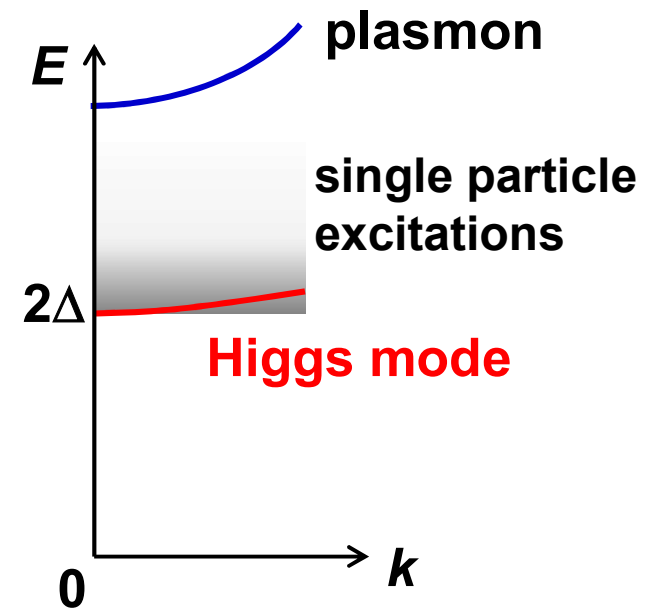
Mass of transverse component of photon

$$\nabla^2 B = \frac{B}{\lambda^2}$$

“Plasmons, Gauge Invariance, and Mass”
 Phys. Rev. 130, 439 (1963)



Anderson



Random-Phase Approximation in the Theory of Superconductivity*

P. W. ANDERSON

Bell Telephone Laboratories, Murray Hill, New Jersey

(Received July 28, 1958)

A generalization of the random-phase approximation of the theory of Coulomb correlation energy is applied to the theory of superconductivity. With no further approximations it is shown that most of the elementary excitations have the Bardeen-Cooper-Schrieffer energy gap spectrum, but that there are collective excitations also. The most important of these are the longitudinal waves which have a velocity $v_F \{ \frac{1}{3} [1 - 4N(0)|V|] \}^{\frac{1}{2}}$ in the neutral Fermi gas, and are essentially unperturbed plasma oscillations in the charged case. Other collective excitations resembling higher bound pair states may or may not exist but do not seriously affect the energy gap. The theory obeys the sum rules and is gauge invariant to an adequate degree throughout.

PRB 1958

Theoretical investigations: quantum quench problem

Quenching the interaction $U(t)$ much faster than

$$\tau_{\Delta} \sim \hbar/\Delta \quad (\Delta: \text{order parameter})$$

⇒ Emergence of order parameter oscillation (Higgs mode)

Theoretical studies for
dynamics of nonequilibrium BCS
state after *nonadiabatic*
excitation

Volkov *et al.*, Sov. Phys. JETP **38**, 1018 (1974).

Barankov *et al.*, PRL **94**, 160401 (2004).

Yuzbashyan *et al.*, PRL **96**, 230404 (2006).

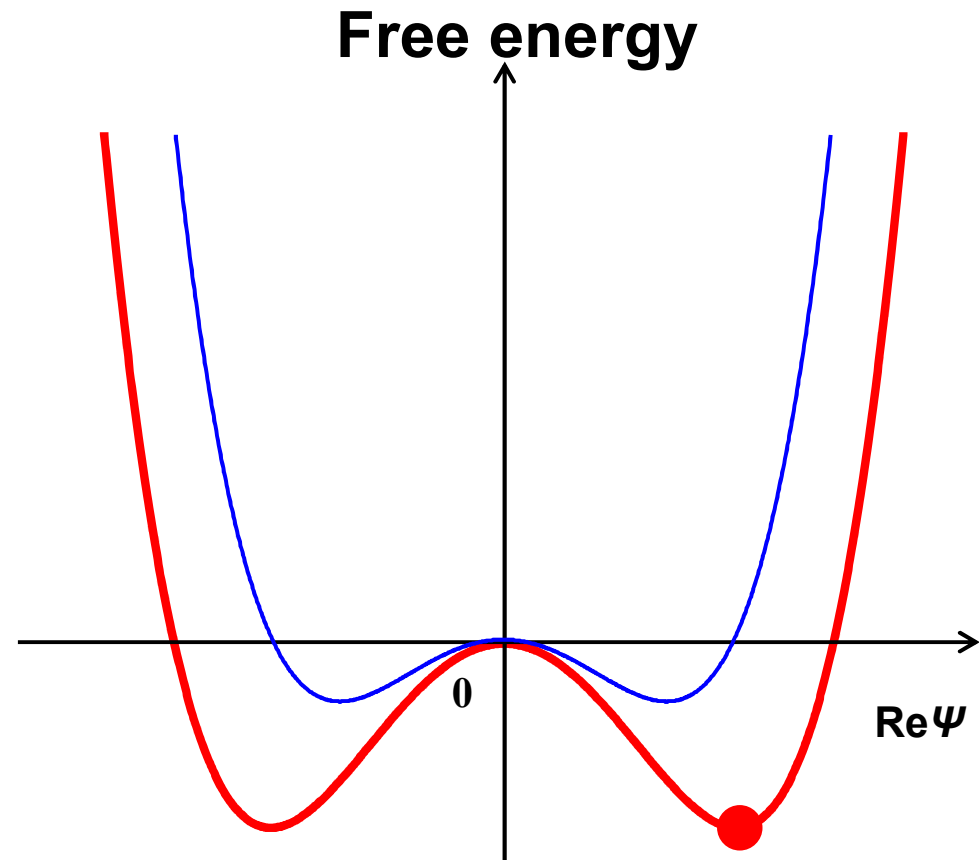
Gurarie *et al.*, PRL **103**, 075301 (2009).

Podolsky, PRB **84**, 174522 (2011).

A. P. Schnyder *et al.*, PRB **84**, 214513 (2011)

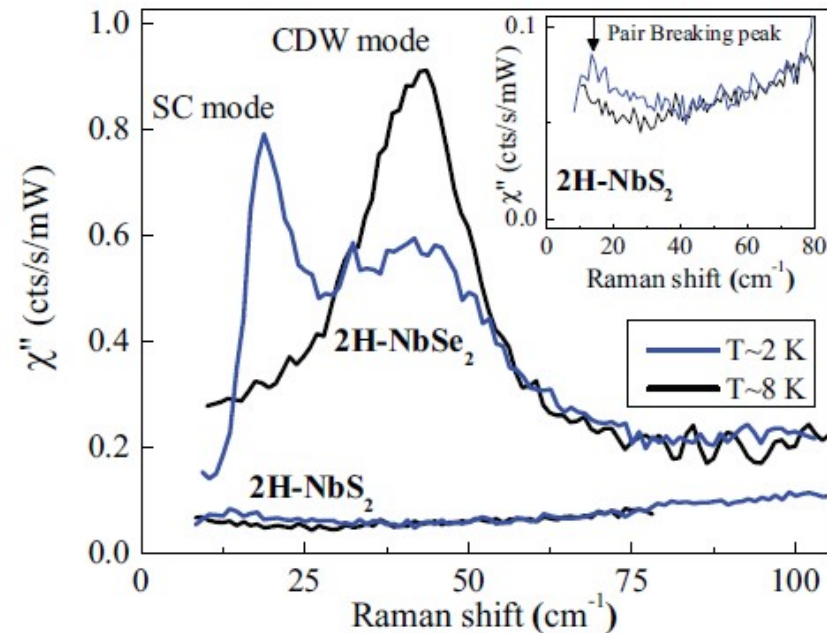
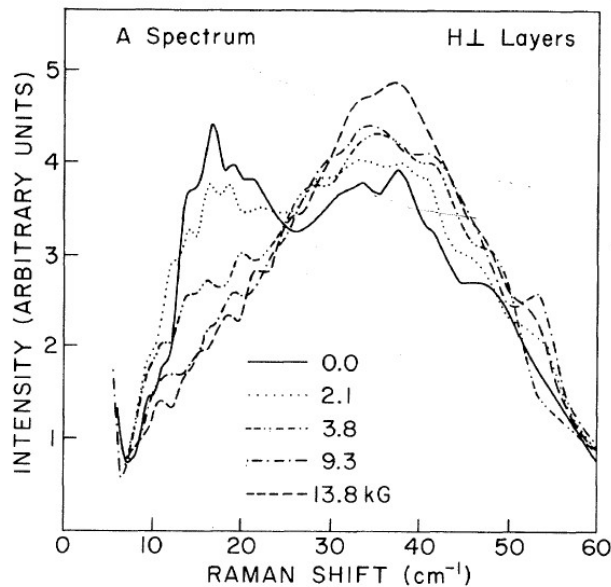
N. Tsuji *et al.*, PRB **88**, 165115 (2013).

N. Tsuji *et al.*, PRL **110**, 136404 (2013).



Higgs mode in superconductors: NbSe₂

BCS-CDW coexistent compound



R. Sooryakumar and M. V. Klein, PRL **45**, 660 (1980).
P.B. Littlewood and C. M. Varma, PRL **47**, 811 (1982).

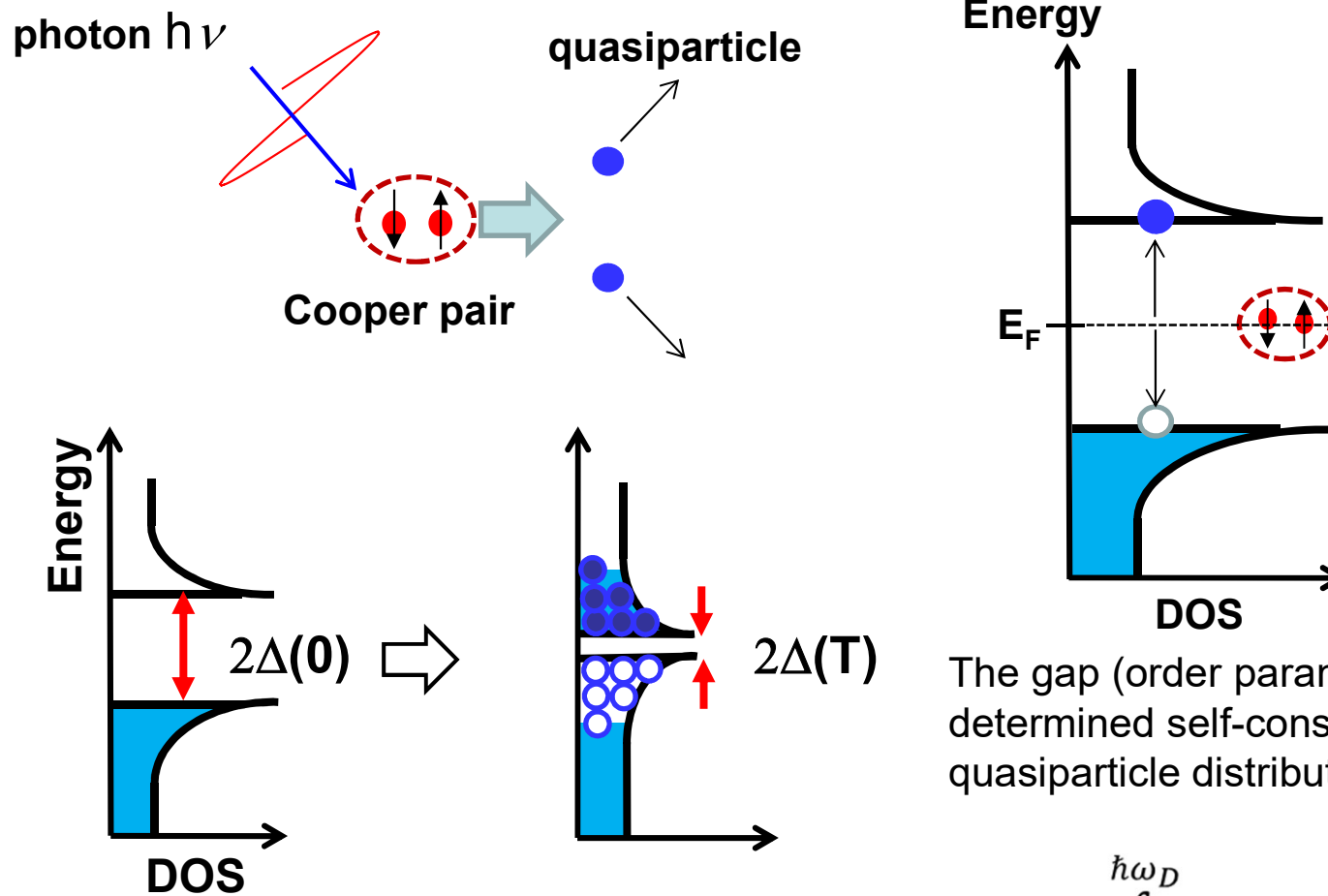
M.-A. Measson, et al.,
PRB **89**, 060503 (2014).

For a recent review:

D. Pekker and C. M. Varma, Annual Review of Condensed Matter Physics **6**, 269 (2015)

Instead of quenching the interaction,...

Quasiparticle injection by ultrafast optical pulse

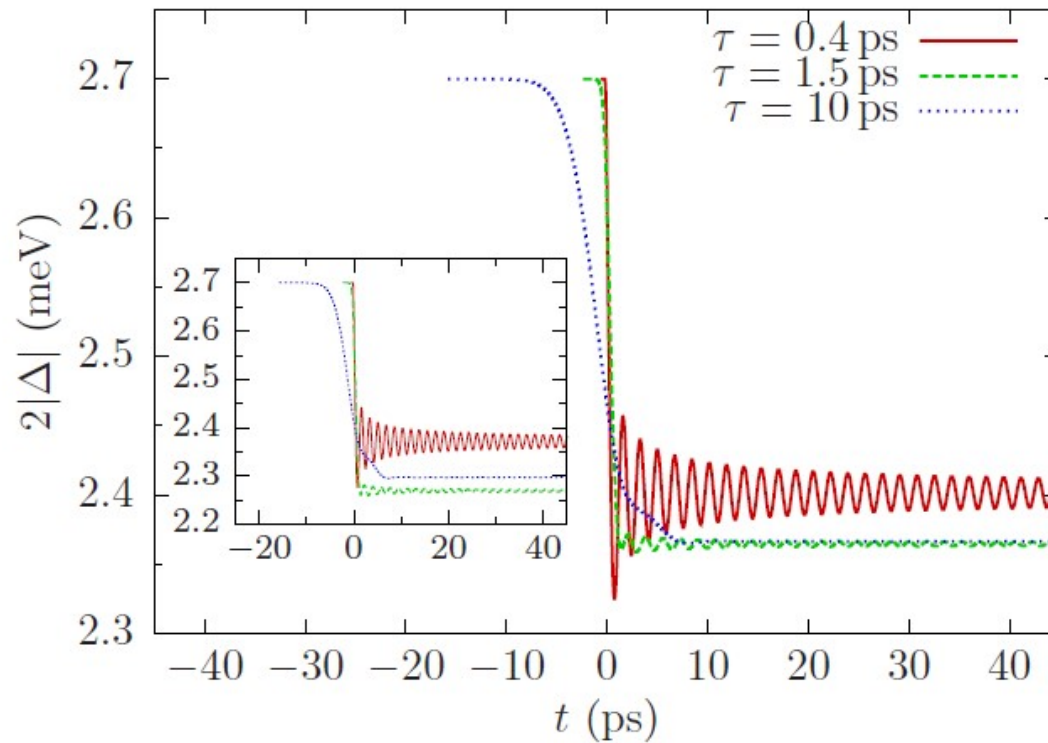


The gap (order parameter) is determined self-consistently with the quasiparticle distribution $f(\varepsilon)$

$$\Delta = V \int_{\Delta}^{\hbar\omega_D} d\varepsilon \frac{\Delta}{\sqrt{\varepsilon^2 - \Delta^2}} [1 - 2f(\varepsilon)]$$

Order parameter oscillation after instantaneous excitation near the gap edge

$$\tau_{\text{pump}} < \Delta^{-1}$$



T. Papenkort, V. M. Axt, and T. Kuhn,
Phys. Rev. B **76**, 224522 (2007).

THz pump and THz probe experiment in NbN

Sample



Nb_{0.8}Ti_{0.2}N film (12nm)/Quartz

$T_C = 8.5$ K,

$2\Delta(T=4$ K) = 3.0 meV = 0.72 THz

response time : $\tau_\Delta = \Delta^{-1} \sim 2.8$ ps

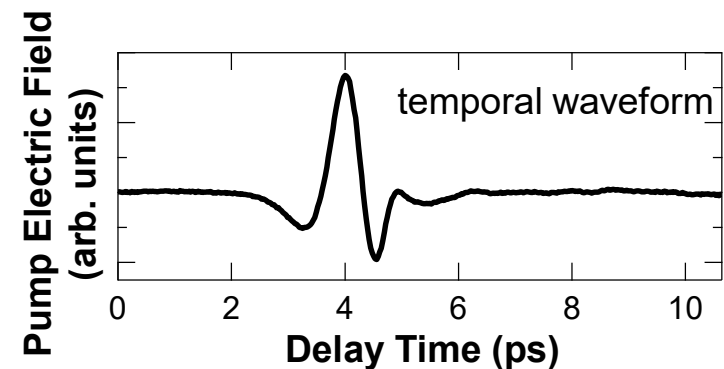
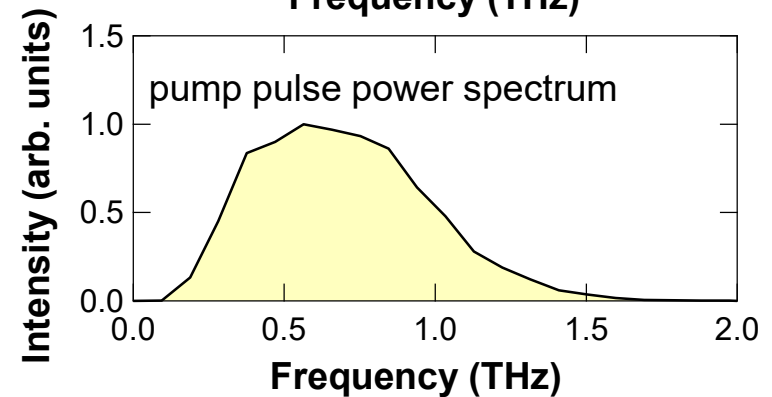
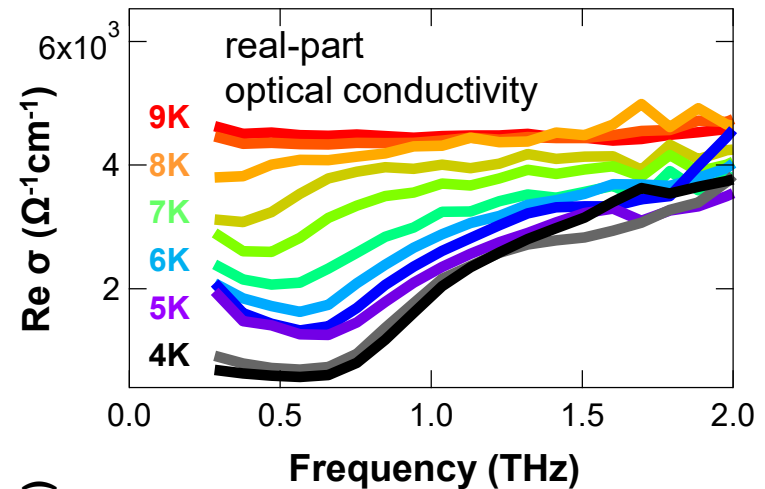
THz pump pulse

Center frequency 0.7THz $\sim 2\Delta$

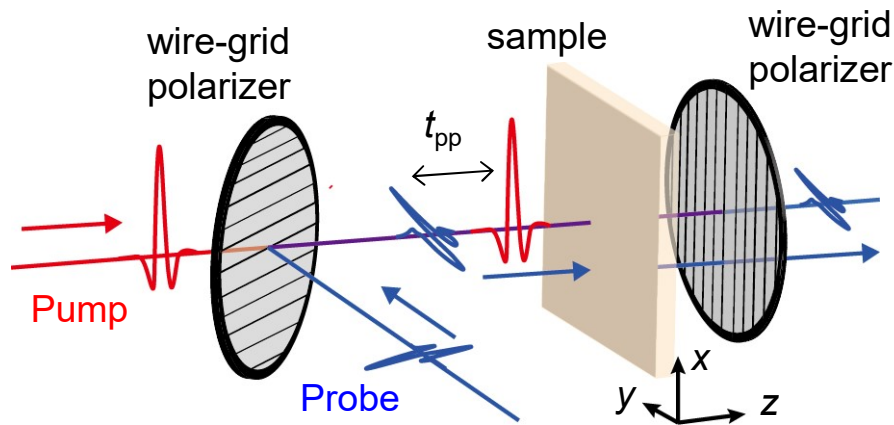
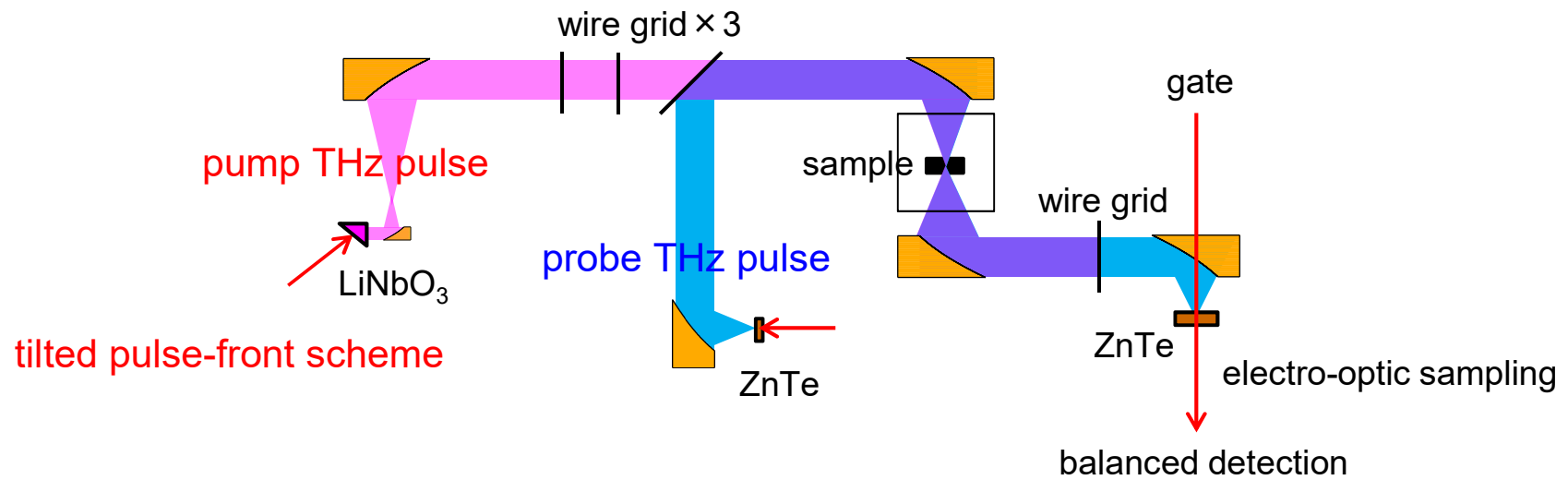
pulse width: $\tau_{\text{pump}} \sim 1.5$ ps

$\tau_{\text{pump}}/\tau_\Delta \sim 0.57 < 1$

 **nonadiabatic excitation condition**



THz pump and THz probe experiment in NbN



Pump : $E_{\text{pump}} // x$

Probe: $E_{\text{probe}} // y$

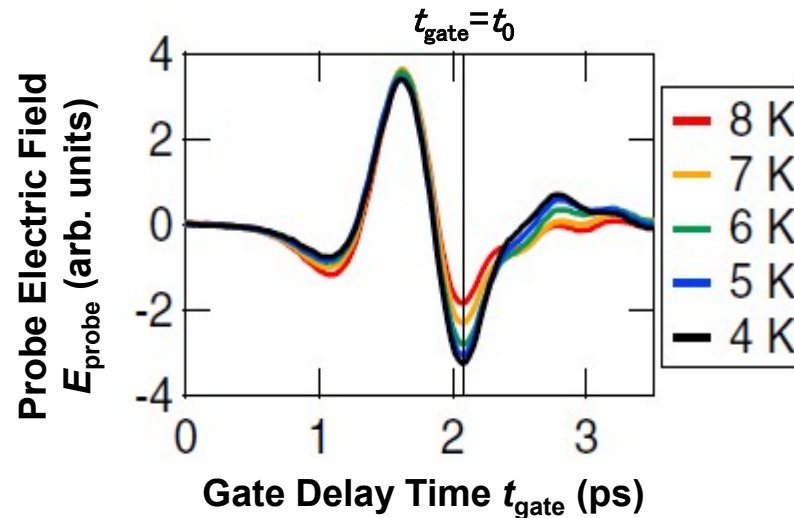
t_{pp} : pump-probe delay

Transmitted probe THz electric field:

Free space EO sampling

t_{gate} : gate pulse delay

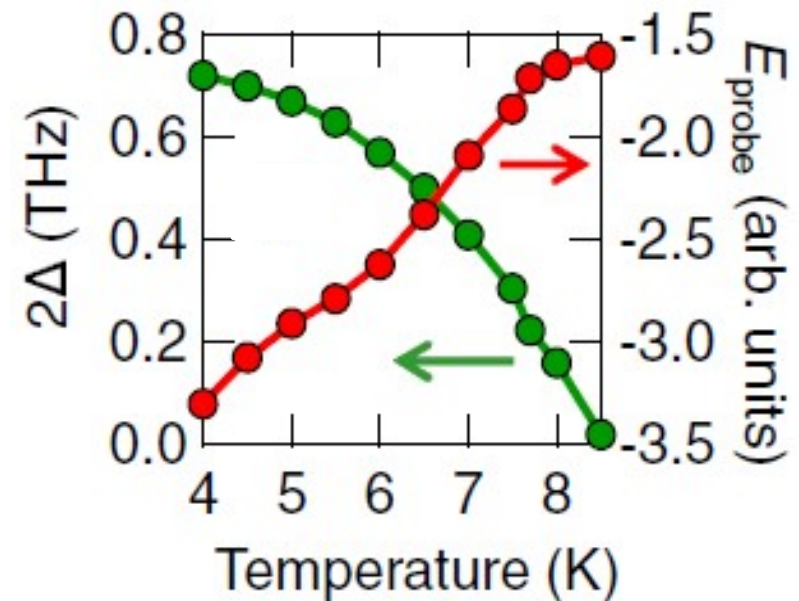
Detection of order parameter dynamics



Temperature dependence of the probe
E-field without pump $E_{\text{probe}}(t_{\text{gate}})$

At $t_{\text{gate}} = t_0$, the change in E_{probe} is proportional to the change in the order parameter Δ .

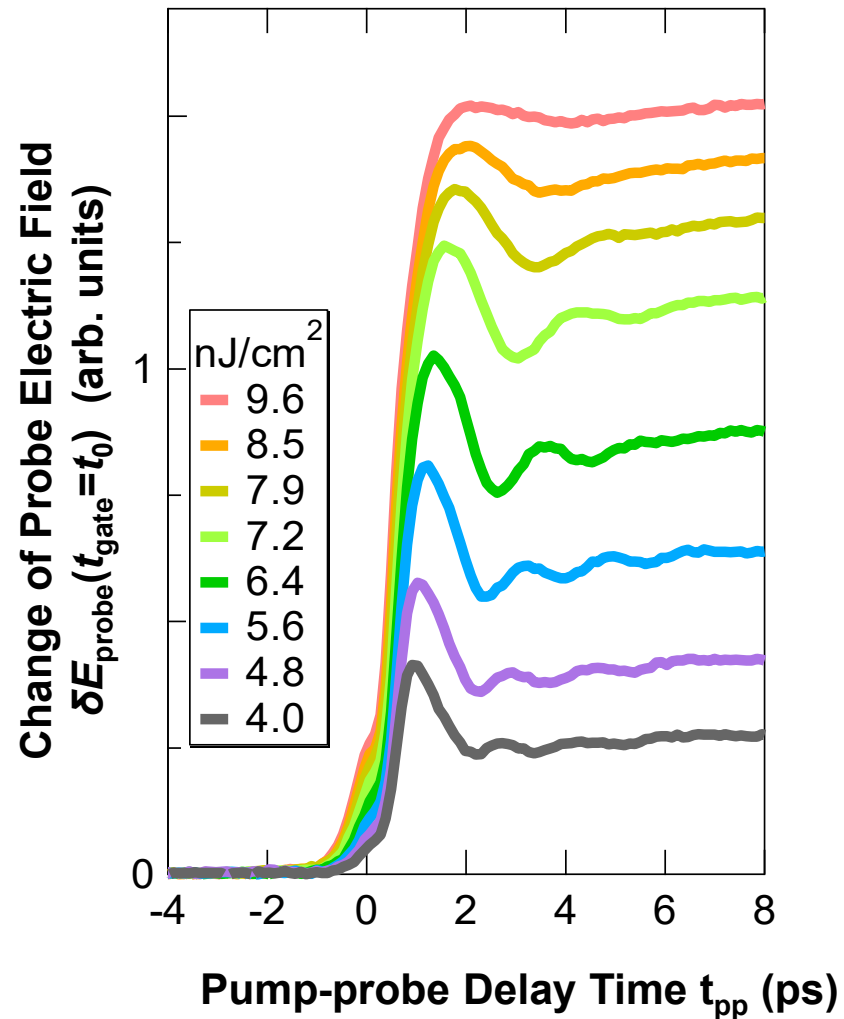
We fixed the gate delay at $t_{\text{gate}} = t_0$ and measure the pump-probe delay dependence



Dynamics after the THz pump pulse

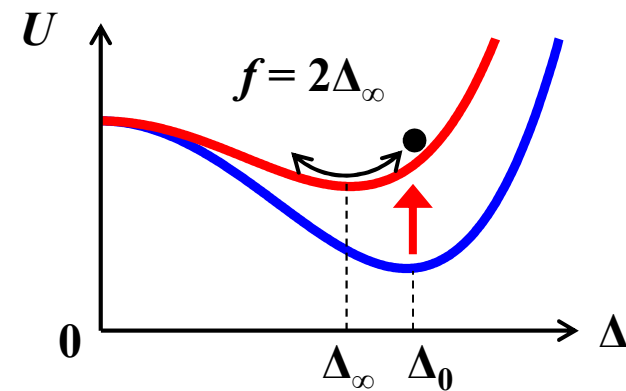
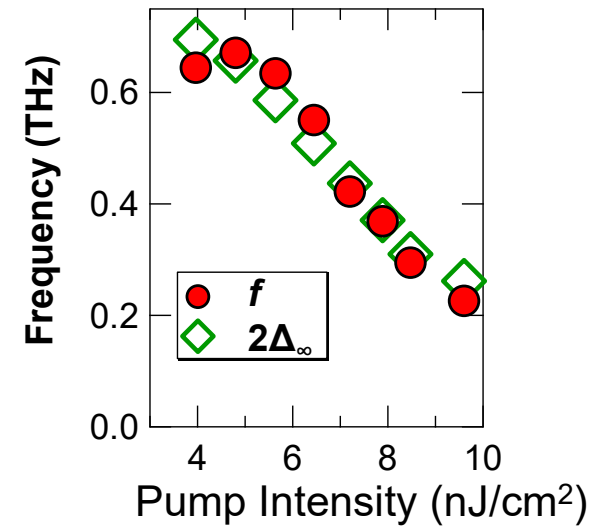
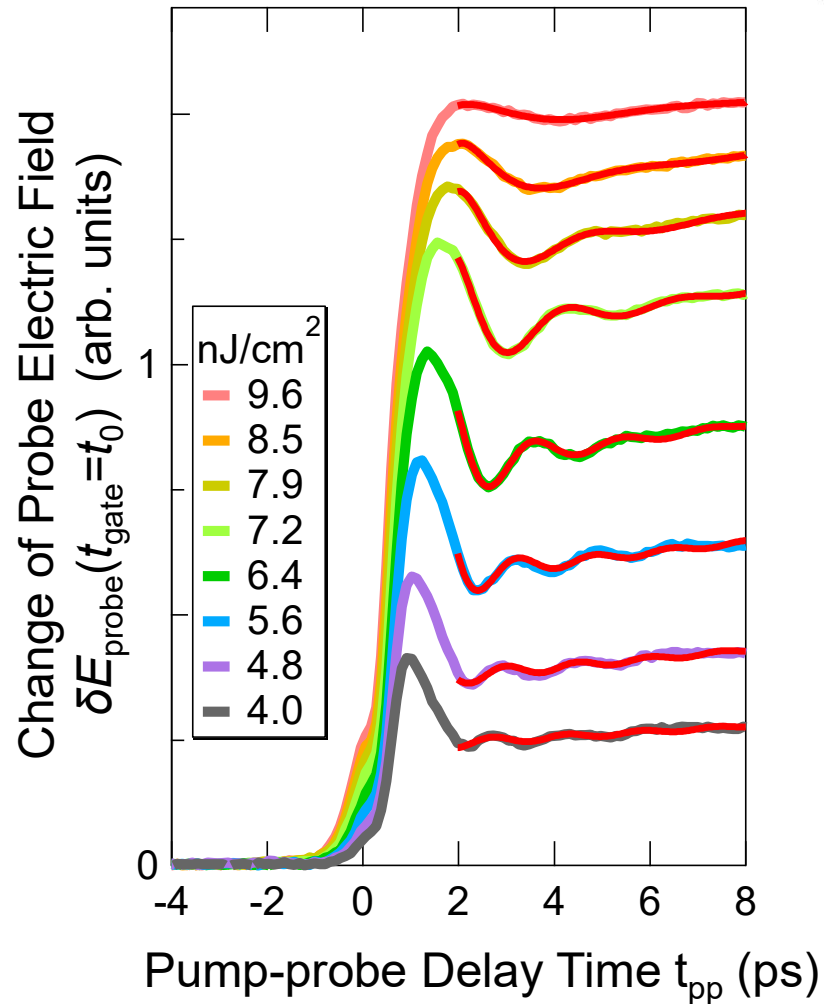
THz pump-induced change in the probe E-field $\delta E_{\text{probe}}(t_{\text{gate}}=t_0)$

$$\tau_{\text{pump}}/\tau_{\Delta}=0.57$$

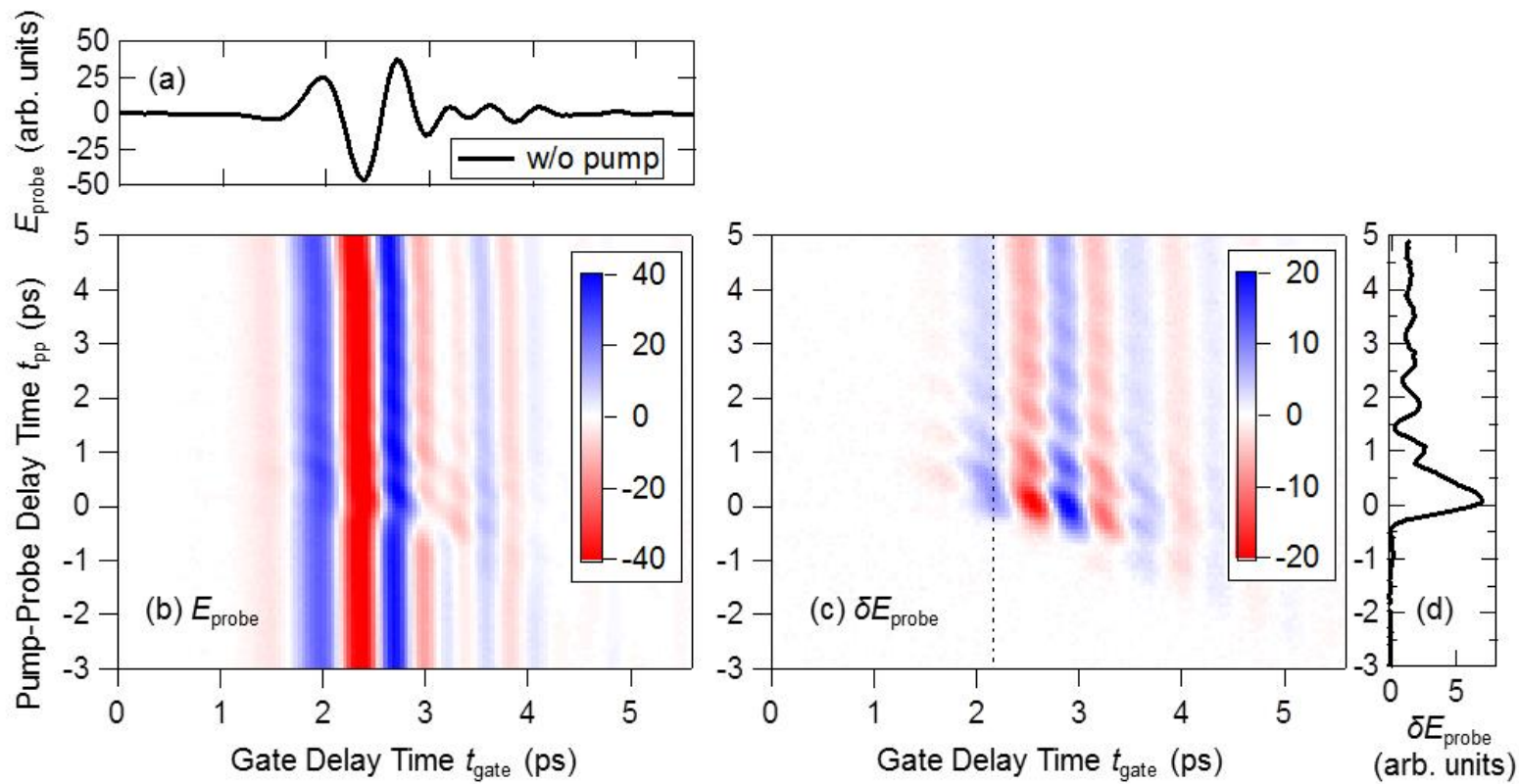


Order parameter dynamics

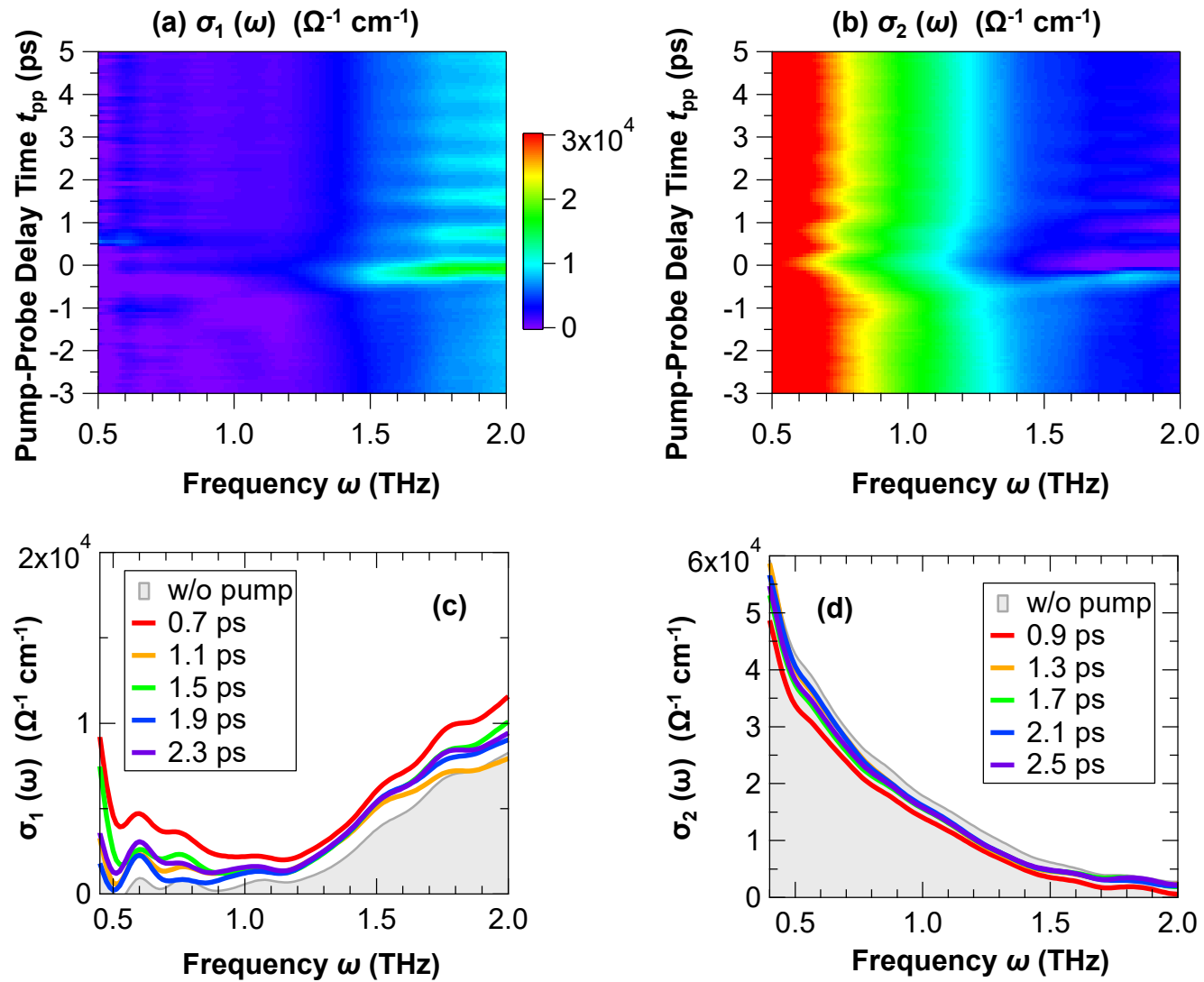
$$\delta\Delta(t_{pp}) = C_1 + C_2 t_{pp} + \frac{a}{(t_{pp})^b} \cos(2\pi f t_{pp} + \phi)$$



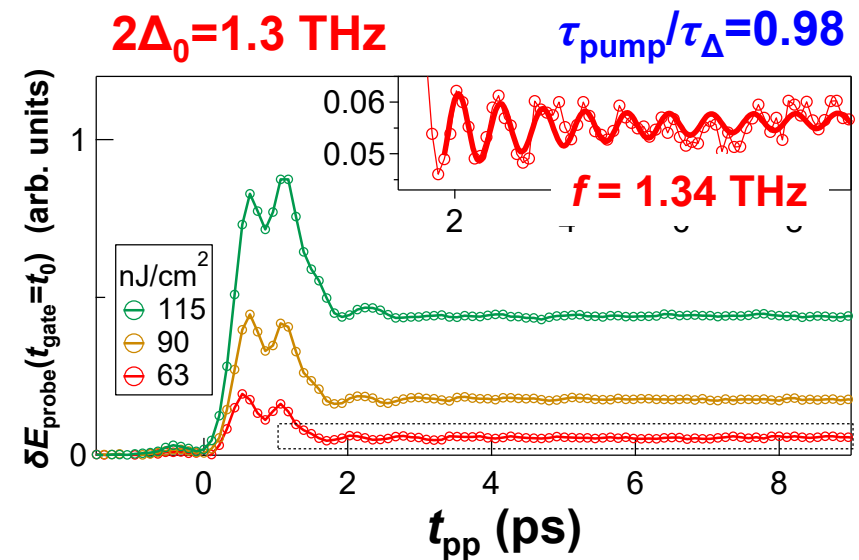
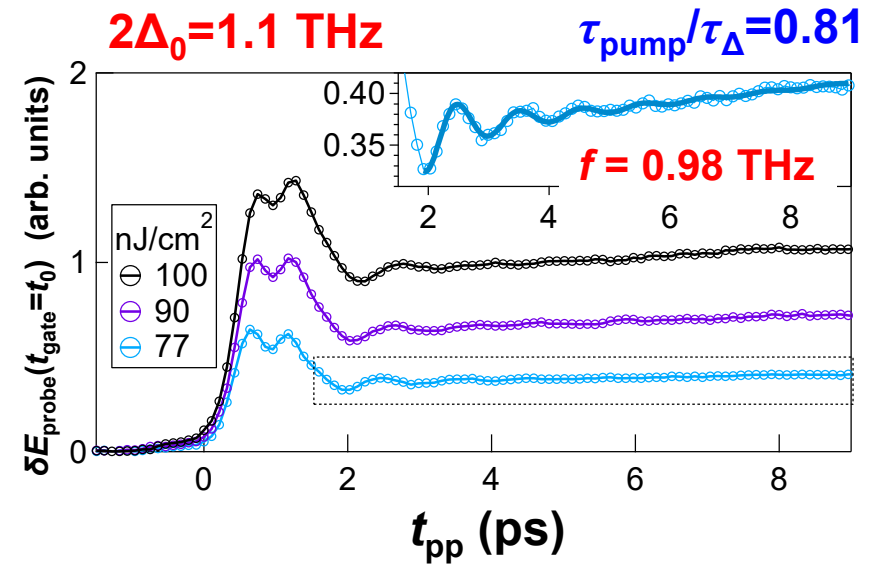
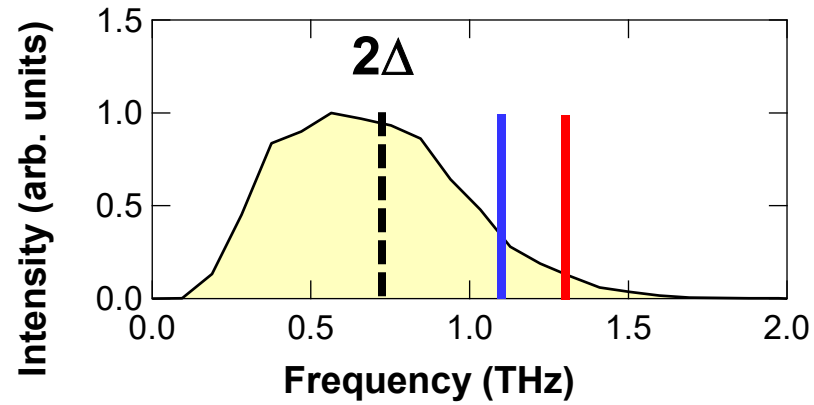
2D-scan of THz pump and THz probe measurement



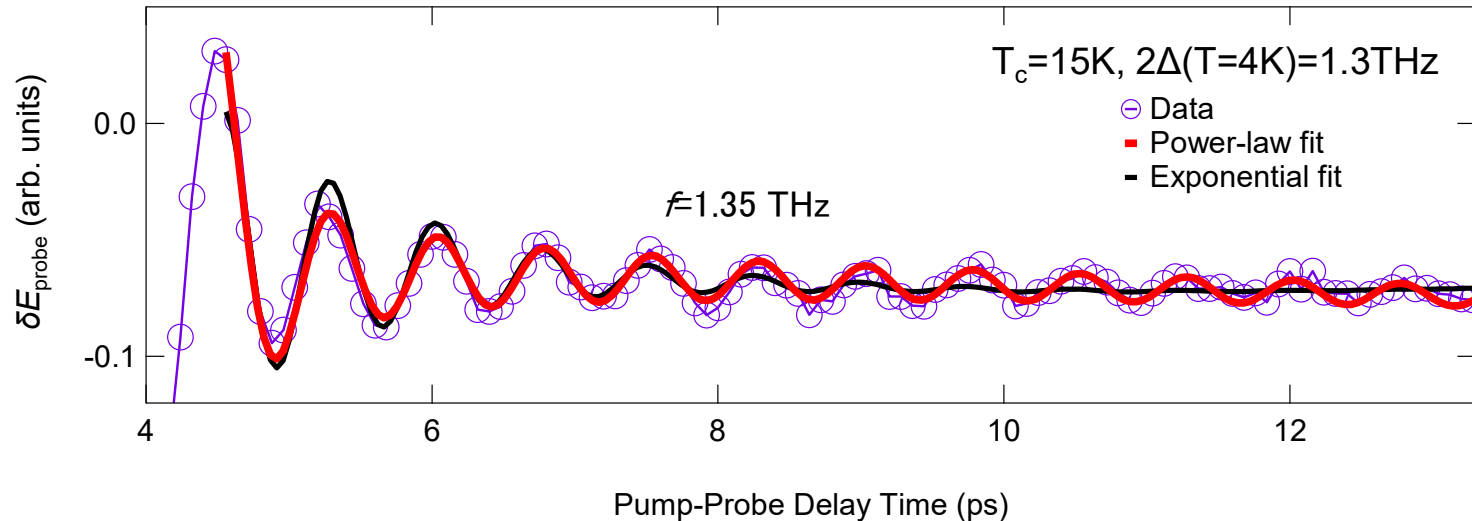
Time evolution of conductivity spectrum $\sigma_1(\omega; t_{pp})$



Higgs mode in larger gap samples $\tau_{\text{pump}}/\tau_{\Delta} \lesssim 1$



Power law decay



Weak coupling case (BCS)

$$\frac{\Delta(t)}{\Delta_\infty} = 1 + a \frac{\cos(2\Delta_\infty t + \pi/4)}{\sqrt{\Delta_\infty t}}$$

Volkov *et al.*, Sov. Phys. JETP 38, 1018 (1974).
 Yuzbashyan *et al.*, PRL 96, 097005 (2006).

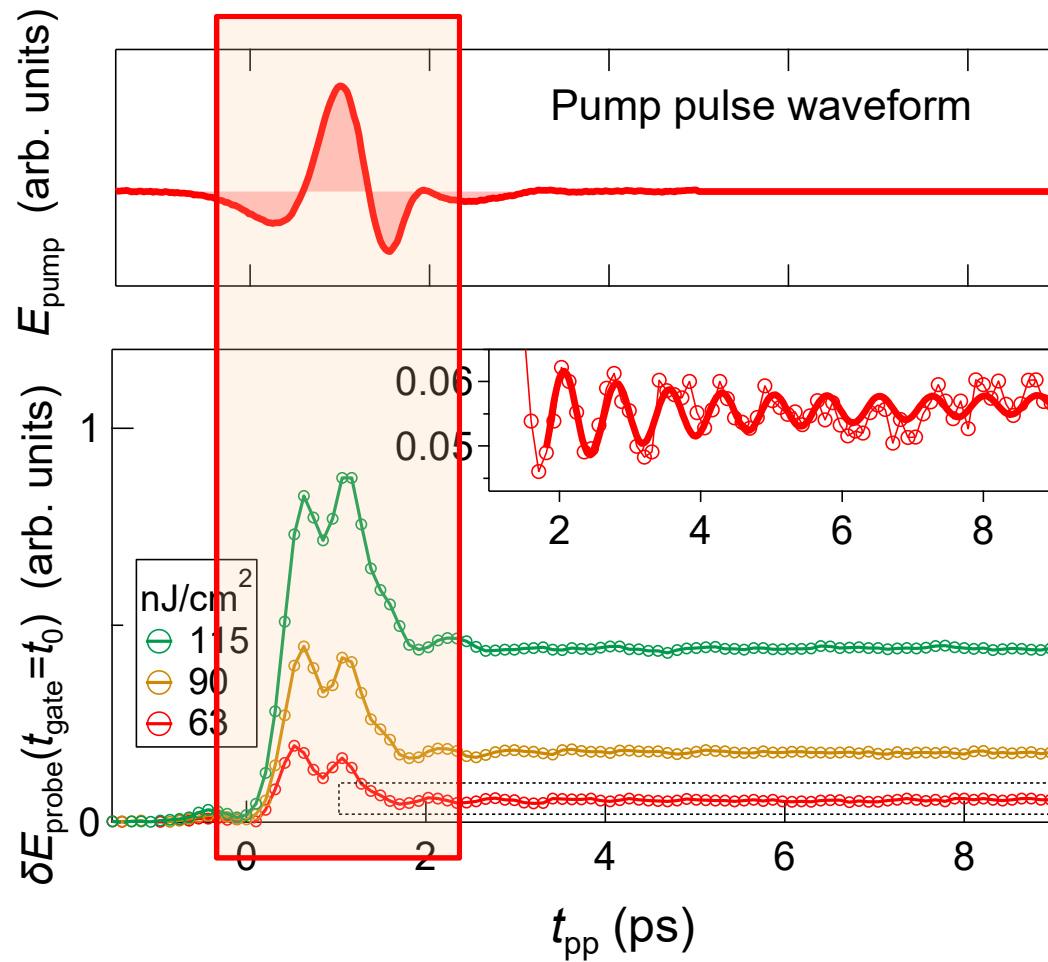
exponential decay $\delta E_{\text{probe}}(t_{\text{pp}}) = C + A \exp\left(-\frac{t}{\tau}\right) \cos(2\pi f t_{\text{pp}} + \phi)$

$\tau = 1.3\text{ ps}$ $\chi^2 = 3.6 \times 10^{-4}$

power-law decay $\delta E_{\text{probe}}(t_{\text{pp}}) = C + \frac{A}{(t_{\text{pp}} - t_0)^b} \cos(2\pi f t_{\text{pp}} + \phi)$

$b = 0.71$ $\chi^2 = 2.8 \times 10^{-4}$

Dynamics in the coherent excitation regime

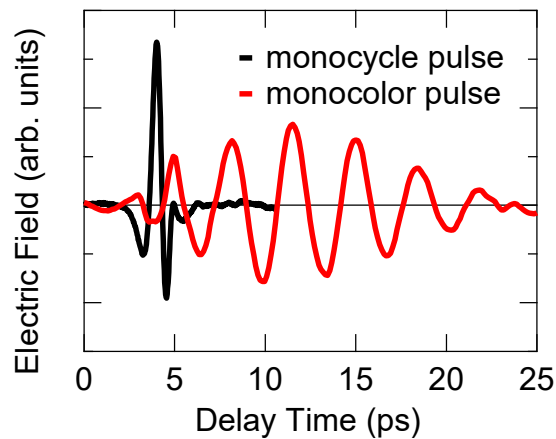


What is happening during the irradiation of AC driving field?

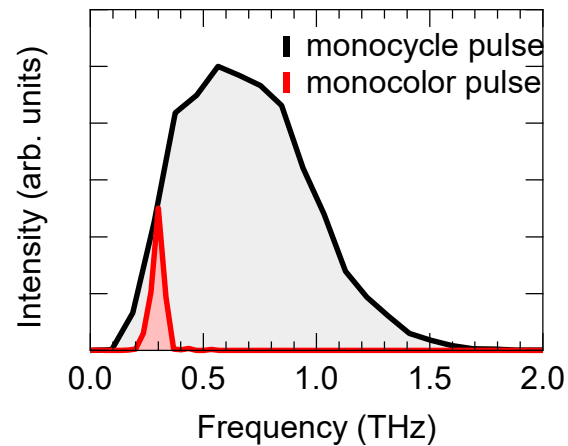
Coherent excitation regime with multicycle THz pulse

Quasi-monochromatic THz pulse (0.3THz, pulsewidth ~ 13 ps)

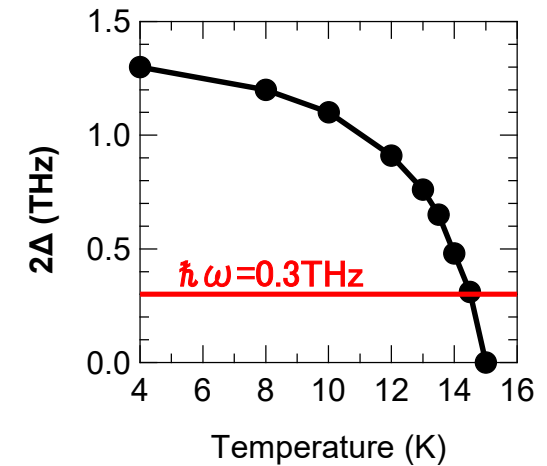
E-field waveform



Power Spectrum



Photon energy vs BCS gap



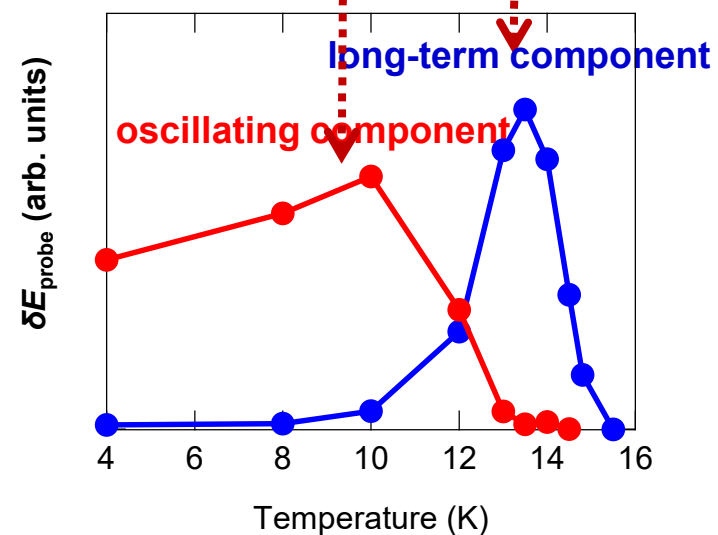
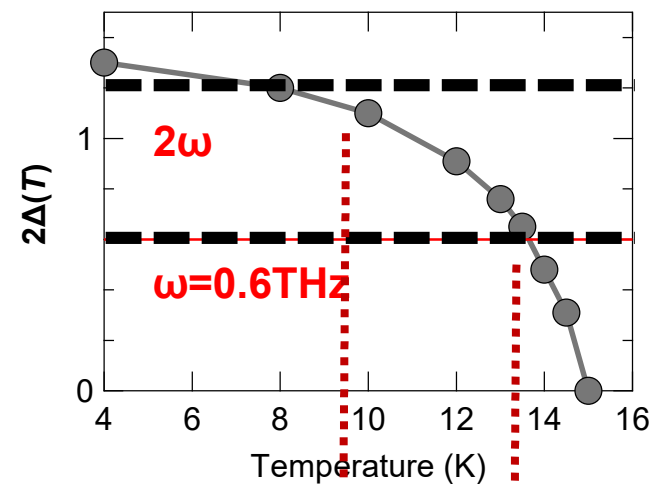
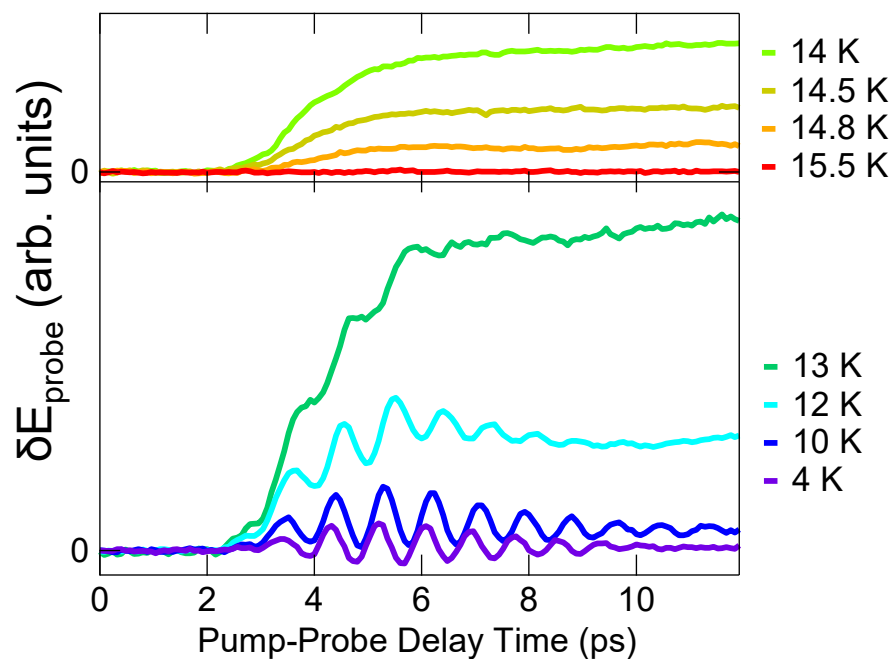
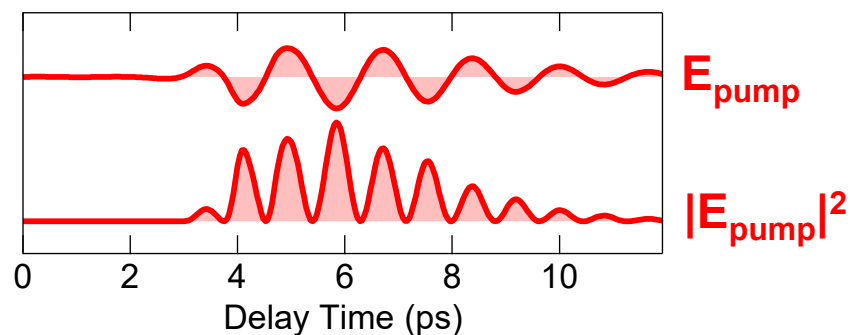
How does the BCS ground state respond to the strong electromagnetic field with $\hbar\omega < 2\Delta$?

Coherent Excitation Regime Experiments

R. Matsunaga et al., Science **345**, 1145 (2014)

$\omega=0.6\text{THz}$

$E=3.5\text{ kV/cm @ peak}$



Anderson's pseudospin (σ_k) representation

Anderson, Phys. Rev. **112**, 1900 (1958)

$$|\Psi_{\text{BCS}}\rangle = \prod_{\mathbf{k}} (u_{\mathbf{k}} + v_{\mathbf{k}} c_{\mathbf{k}\uparrow}^+ c_{-\mathbf{k}\downarrow}^+) |0\rangle$$

Pseudospin up : ($k, -k$) both empty

Pseudospin down: ($k, -k$) both occupied

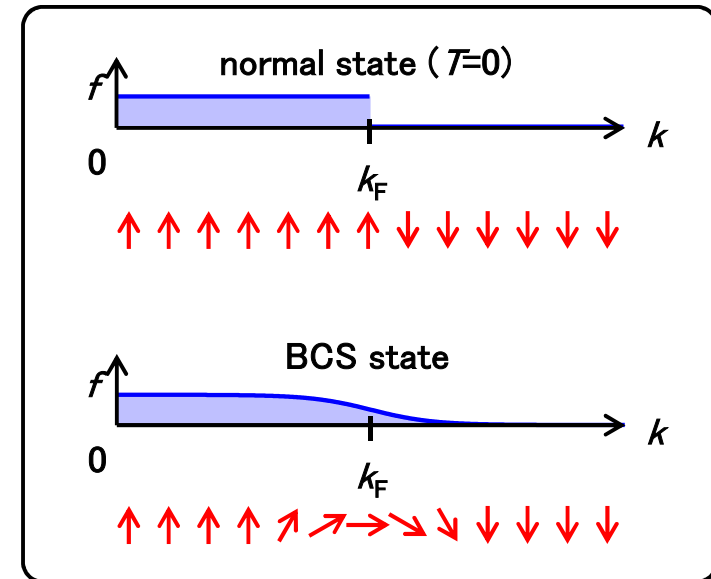
$$\mathcal{H}^{\text{BCS}} = \sum_{\mathbf{k}} \mathbf{b}_{\mathbf{k}}^{\text{eff}} \cdot \boldsymbol{\sigma}_{\mathbf{k}}$$

$$\mathbf{b}_{\mathbf{k}}^{\text{eff}} = (-\Delta', -\Delta'', \varepsilon_{\mathbf{k}})$$

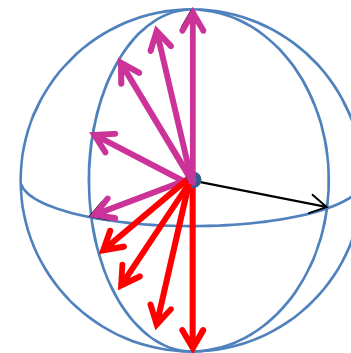
: effective magnetic field for k

$$\Delta = \Delta' + i\Delta'' = U \sum_{\mathbf{k}} (\sigma_{\mathbf{k}}^x + i\sigma_{\mathbf{k}}^y)$$

$$\frac{d}{dt} \boldsymbol{\sigma}_{\mathbf{k}} = i[\mathcal{H}^{\text{BCS}}, \boldsymbol{\sigma}_{\mathbf{k}}] = 2\mathbf{b}_{\mathbf{k}}^{\text{eff}} \times \boldsymbol{\sigma}_{\mathbf{k}}$$



$k, -k$ empty



$k, -k$ occupied

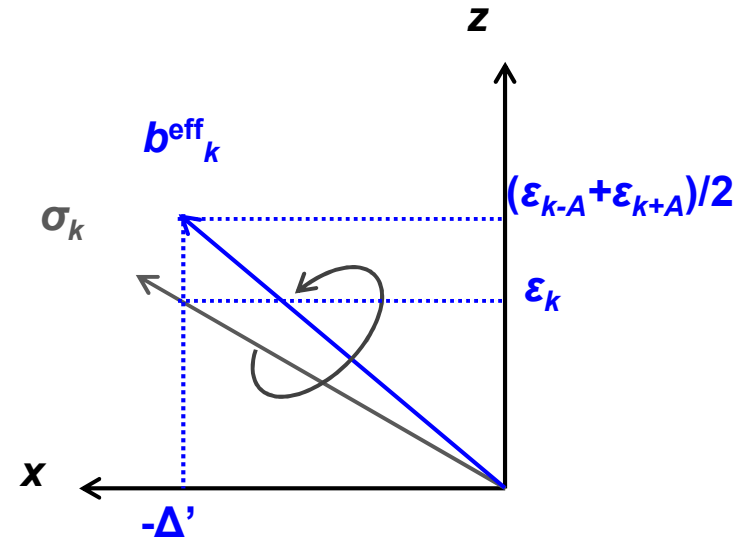
Time evolution of BCS state = motion of pseudospins under effective magnetic field

Pseudospin dynamics under the presence of vector potential $A(t)$

$$\frac{d}{dt} \boldsymbol{\sigma}_k = i [\mathcal{H}^{\text{BCS}}, \boldsymbol{\sigma}_k] = 2\mathbf{b}_k^{\text{eff}} \times \boldsymbol{\sigma}_k$$

$$\Delta = \Delta' + i\Delta'' = U \sum_k (\sigma_k^x + i\sigma_k^y)$$

$$\mathbf{b}_k^{\text{eff}} = (-\Delta', -\Delta'', \boxed{\varepsilon_k})$$



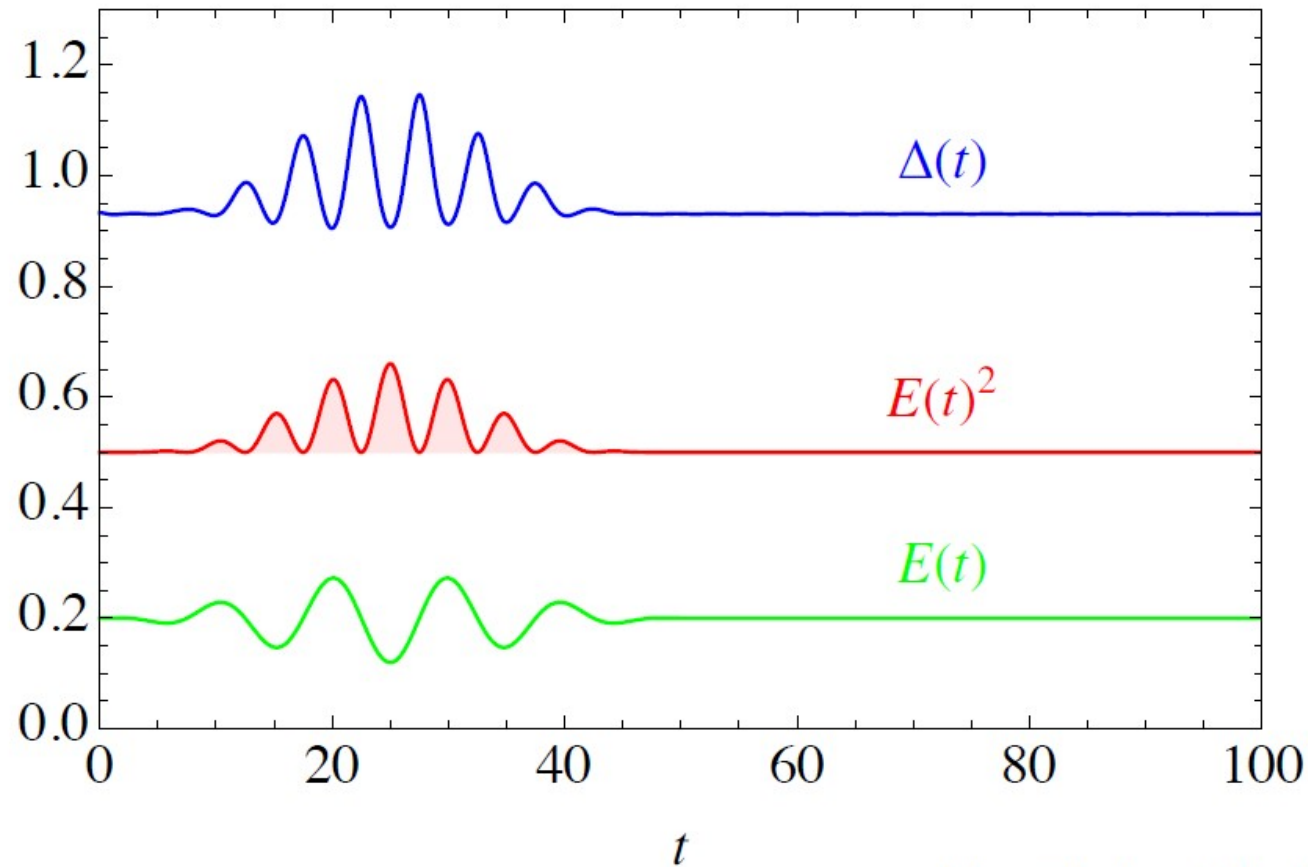
In the presence of EM field (vector potential)

$$\begin{aligned} \frac{1}{2} (\varepsilon_{\mathbf{k}-e\mathbf{A}(t)} + \varepsilon_{-\mathbf{k}-e\mathbf{A}(t)}) &= \varepsilon_{\mathbf{k}} + \frac{e^2}{2} \sum_{i,j} \frac{\partial^2 \varepsilon_{\mathbf{k}}}{\partial k_i \partial k_j} A_i(t) A_j(t) + O(A^4) \\ &= \varepsilon_{\mathbf{k}} - \frac{e^2}{2} \sum_{i,j} \frac{\partial^2 \varepsilon_{\mathbf{k}}}{\partial k_i \partial k_j} \frac{E_i E_j}{\omega^2} e^{i2\omega t} + O(A^4). \end{aligned}$$

z-component of effective magnetic field oscillates at 2ω

\Rightarrow precession of Anderson's pseudospins

Pseudospin dynamics : simulation with BdG equation



$$U = 3, \beta = 12, A = 0.2,$$
$$\Omega = 0.628, 2\Delta(0) = 1.87$$

THz THG by Higgs mode

Current density

$$\mathbf{j}(t) = e \sum_{\mathbf{k}} \mathbf{v}_{\mathbf{k}-A} n_{\mathbf{k}} = e \sum_{\mathbf{k}} \frac{\partial \varepsilon_{\mathbf{k}-eA(t)}}{\partial \mathbf{k}} \left(\sigma_{\mathbf{k}}^z(t) + \frac{1}{2} \right)$$
$$\sim \mathbf{j}_{\text{linear}}(t) - \frac{e^2 \Delta}{U} A(t) \delta \Delta(t)$$

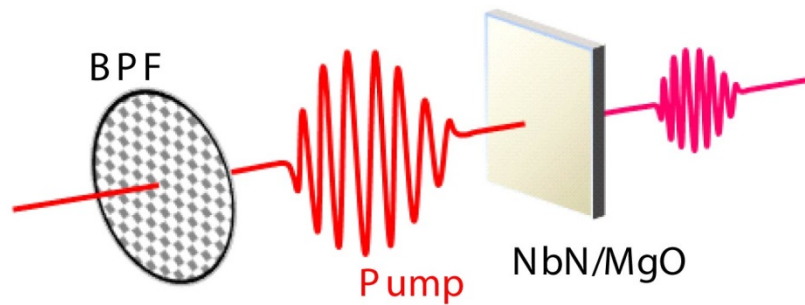
London equation for nonlinear current \mathbf{j}_{nl}

$$\begin{array}{l} \delta \Delta(t) \sim e^{i2\omega t}, \\ A(t) \sim e^{i\omega t} \end{array} \quad \Rightarrow \quad j(t) \sim e^{i3\omega t}$$

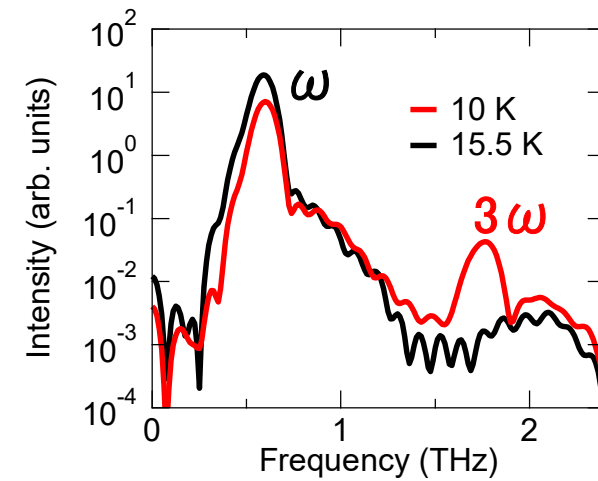
Does superconductor emit THz third harmonics?

Efficient THG from superconductor

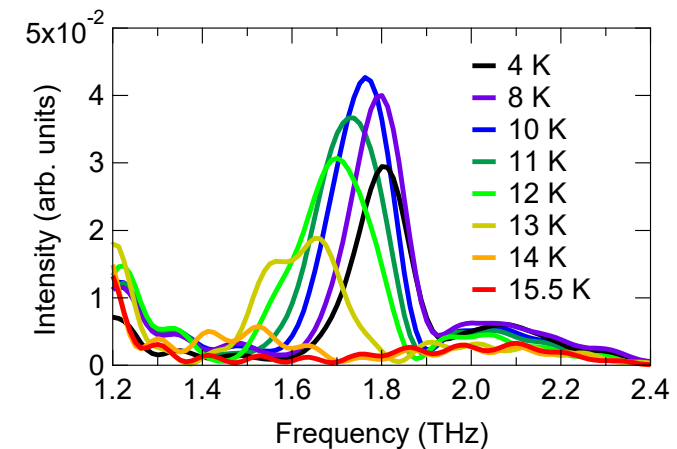
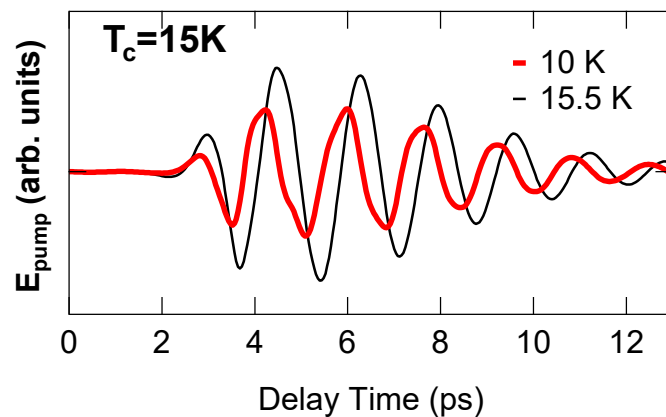
Nonlinear transmission experiment



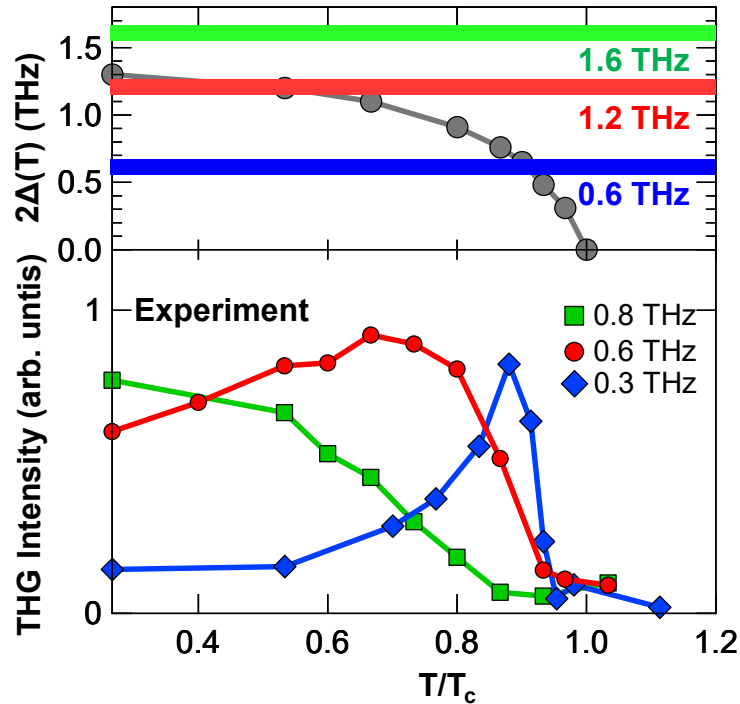
Power spectrum of the transmitted pulse



Waveform of the transmitted pulse



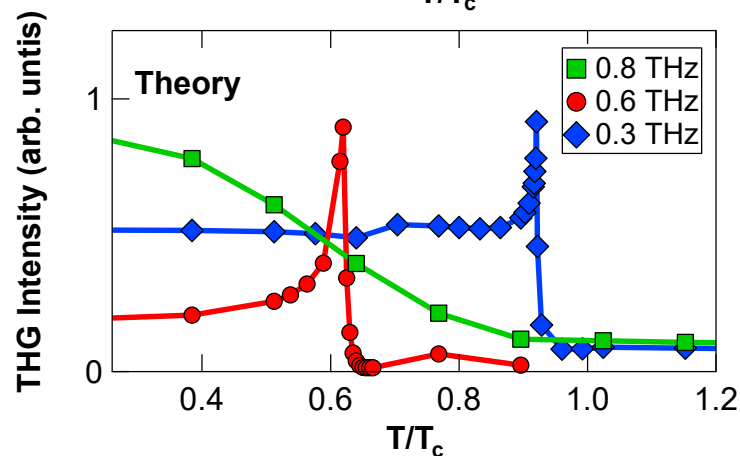
Temperature dependence of THG



Experiments with different frequencies
 $\omega=0.3, 0.6, 0.8$ THz

THG shows a peak at $2\omega=2\Delta(T)$,
but not at $\omega=2\Delta(T)$!

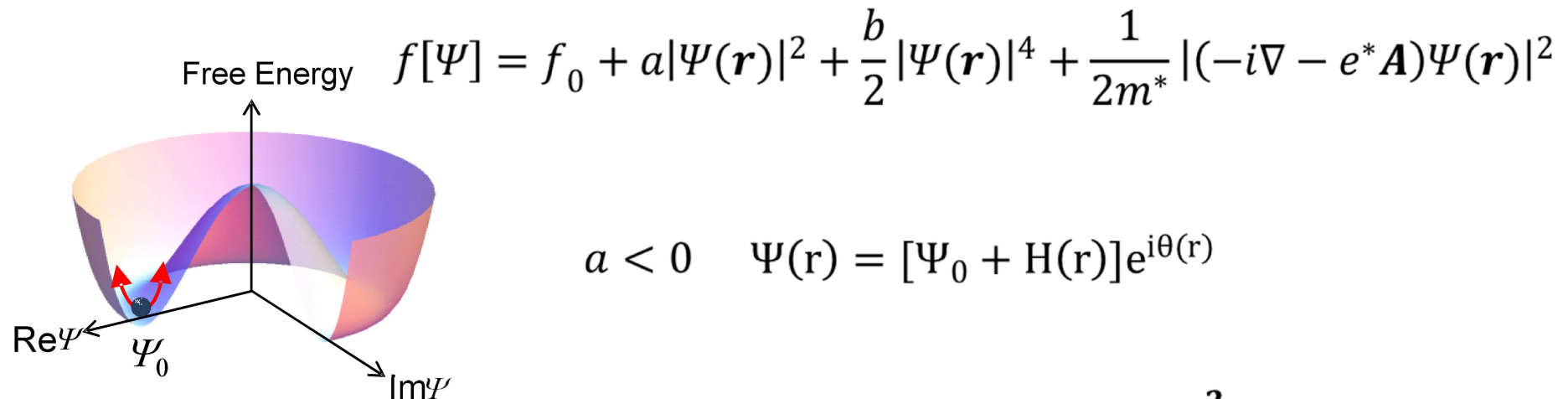
Collective precession of Anderson's
pseudospin resonating with the Higgs
mode



R. Matsunaga et al.,
Science **345**, 1145 (2014)

Theory: N. Tsuji and H. Aoki,
Phys. Rev. B **92**, 064508(2015)

Ginzburg-Landau picture



$$a < 0 \quad \Psi(\mathbf{r}) = [\Psi_0 + H(\mathbf{r})]e^{i\theta(\mathbf{r})}$$

$$f = -2aH^2 + \frac{1}{2m^*}(\nabla H)^2 + \frac{e^{*2}}{2m^*}\left(\mathbf{A} - \frac{1}{e^*}\nabla\theta\right)^2 (\Psi_0 + H)^2 + \dots$$

Local gauge transformation $A' = A - \nabla\theta/e^*$ $A' \rightarrow A$

$$f = -2aH^2 + \frac{1}{2m^*}(\nabla H)^2 + \frac{e^{*2}\Psi_0^2}{2m^*}A^2 + \frac{e^{*2}\Psi_0}{m^*}A^2H + \dots$$

PHYSICS

Particle physics in a superconductor

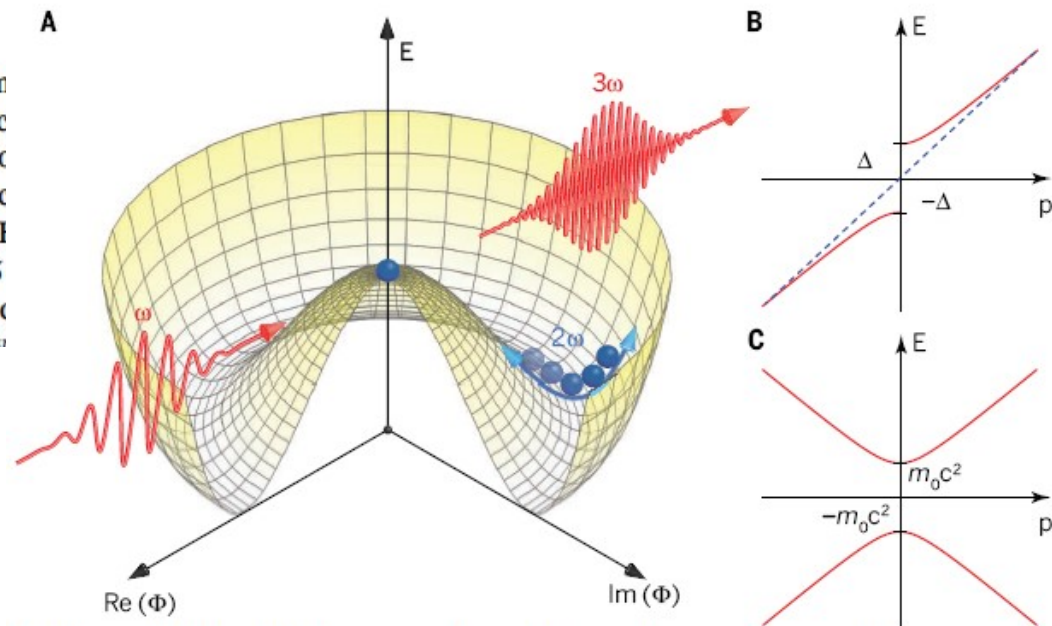
Science 345, 1121 (2014)

A superconducting condensate can display analogous behavior to the Higgs field

By Alexej Pashkin and Alfred Leitenstorfer

The recent discovery of the Higgs boson has created a lot of excitement among scientists. Celebrated as one of the most fundamental results in experimental physics (1), the observation of this particle confirms the existence of

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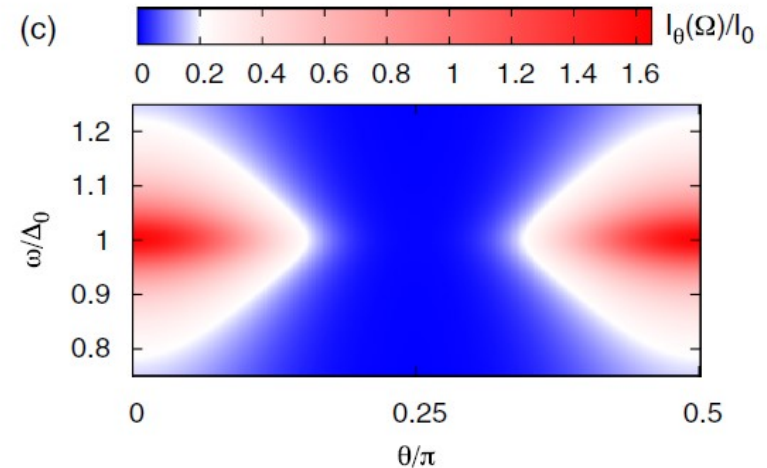
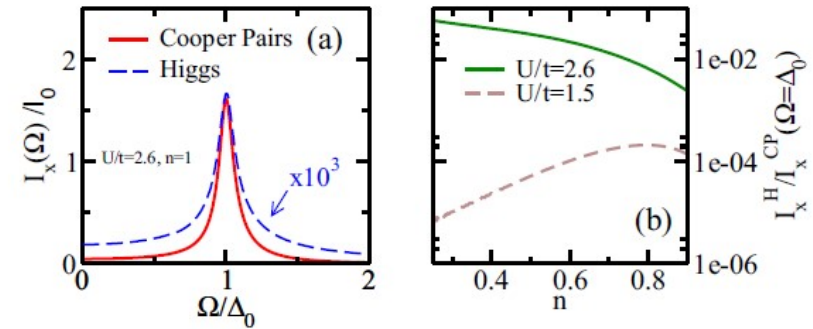
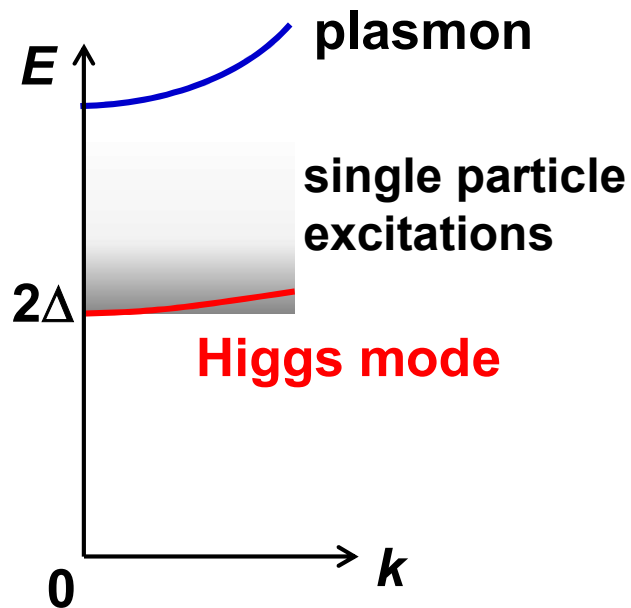
The Higgs amplitude mode. (A) Energy of a system as a function of the complex order parameter Φ in a state with spontaneously broken symmetry. The Higgs mode corresponds to the amplitude oscillations of Φ shown by the blue arrow. The excitation by a light pulse at half the resonance frequency starts a coherent oscillation of the order parameter. The induced superconducting current is nonlinear and leads to emission of the third harmonic of the excitation wave. (B) Energy of quasi-particles as a function of their momentum near the Fermi energy of a normal metal (dashed blue line) and a superconductor with energy gap 2Δ (solid red line). (C) Energy of a relativistic particle-antiparticle system with rest mass m_0 as a function of its momentum.

Higgs vs Charge Density Fluctuation

T. Cea, C. Castellani, and L. Benfatto,
 Phys. Rev. B **93**, 180507 (2016)

BCS with 2D square lattice model

BCS mean field:
 Higgs \ll Charge density fluctuation



Pump polarization dependence

Beyond BCS with retardation

N. Tsuji, Y. Murakami, and H. Aoki, Phys. Rev. B **94**, 224519 (2016)

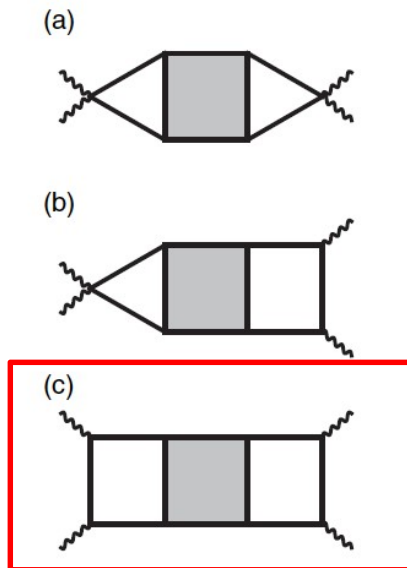
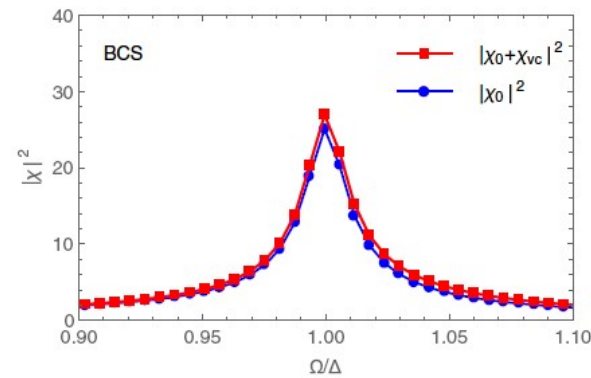
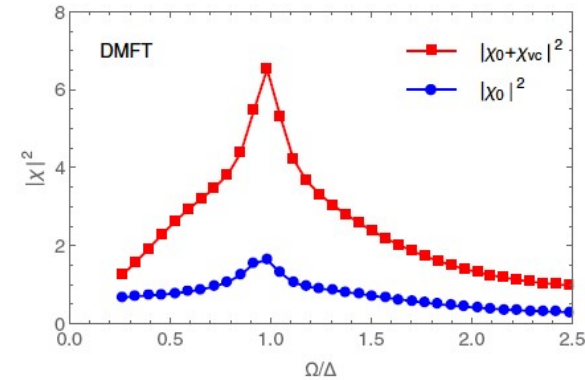


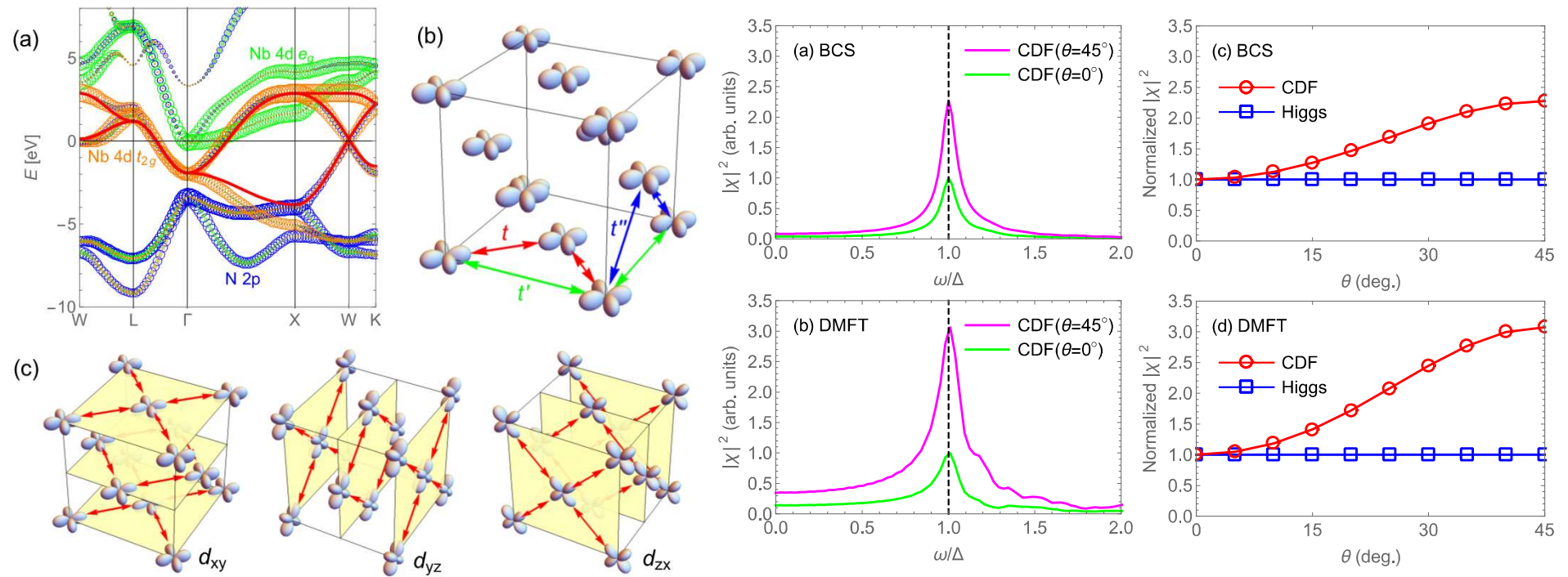
FIG. 1. Feynman diagrams for the nonresonant (a), mixed (b), and resonant (c) contributions to the THG susceptibility containing the effect of collective modes as vertex corrections. The solid (wavy) lines represent the electron (photon) propagators, while the shaded boxes represent the reducible four-point vertex function. Among the four photon lines, one is outgoing with an energy 3Ω , and the other three are incoming with an energy Ω .



Beyond BCS:

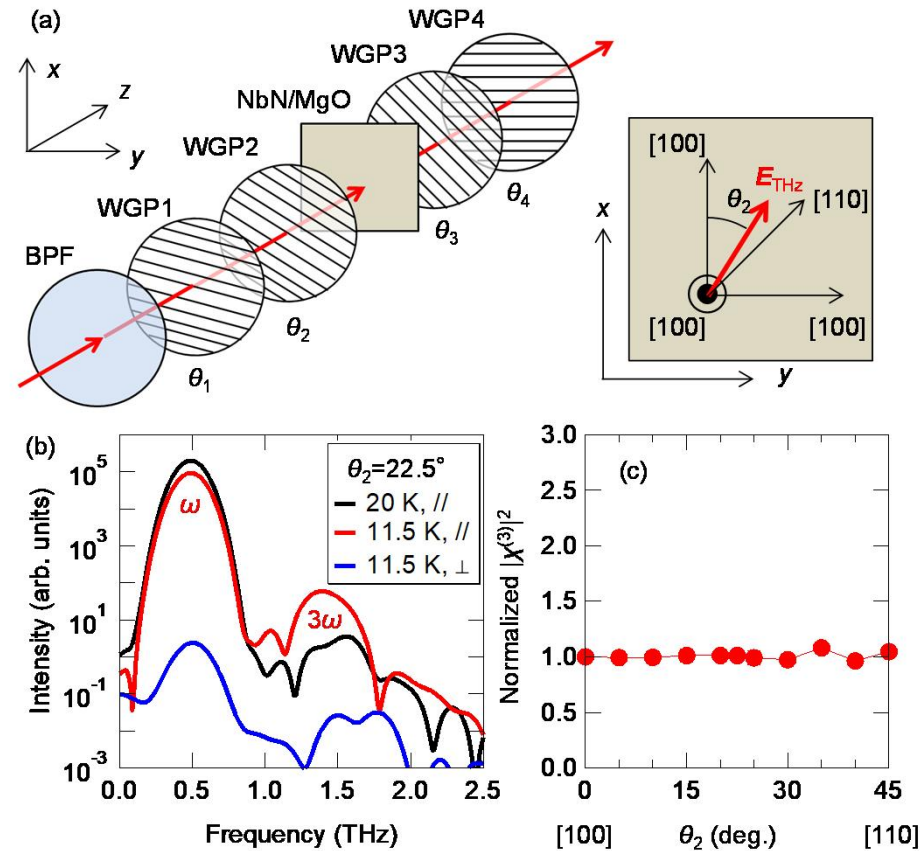
When retarded interaction is taken into account, Higgs term can become larger than the charge density fluctuation.

Polarization dependence of THG



Polarization dependence of THG

R. Matsunaga, et al. Phys. Rev. B 96, 020505(R) (2017).



Polarization of THG is always in parallel with the incident light polarization and its intensity is irrespective to the crystal axis.

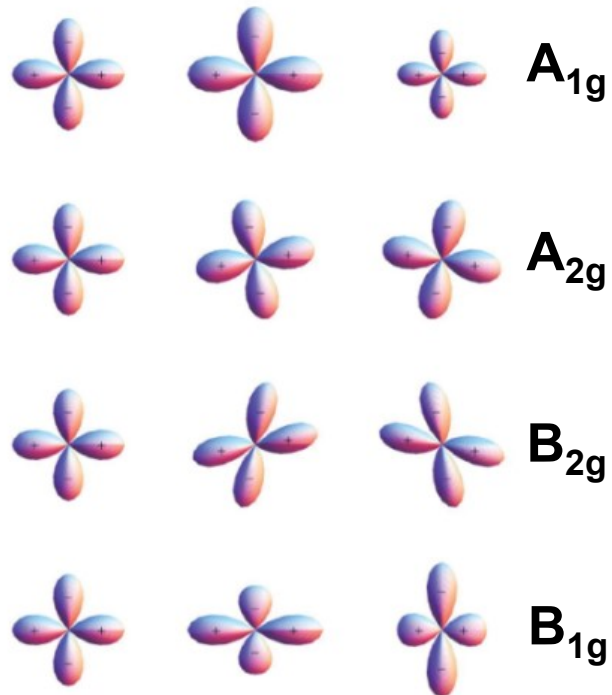
Higgs mode is the dominant origin of THG
→ Retardation effect beyond BCS

Outline

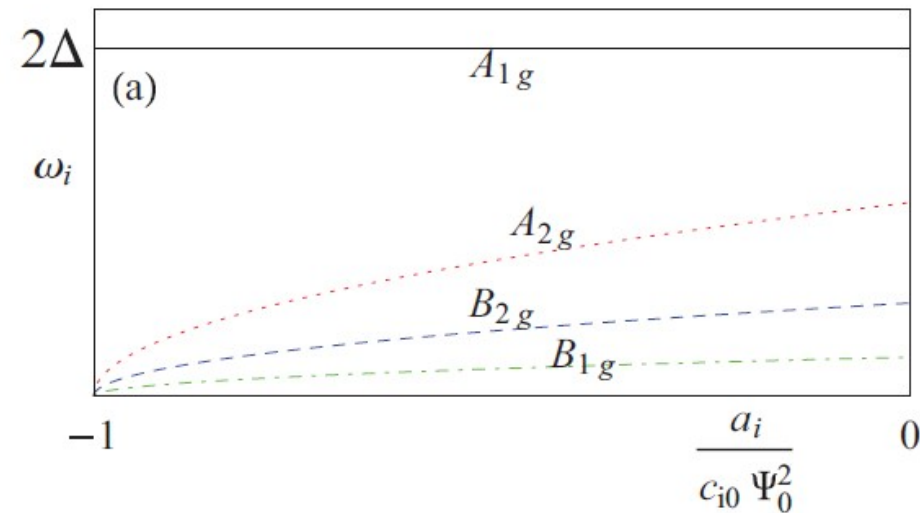
- (1) Introduction
- (2) Photoexcitation in s-wave superconductor
- (3) Higgs mode in a s-wave superconductor NbN
- (4) Higgs mode in d-wave cuprate superconductors
- (5) Photocontrol of superconductors

Higgs modes in d-wave SC













Barlas and Varma,
PRB **87**, 054503 (2013)



$$\mathcal{L} = \sum_{i=0}^3 |\partial_t \phi_i|^2 + a_i |\phi_i|^2 - b_i |\phi_i|^4 - \sum_{i < j} \left(c_{ij} |\phi_i|^2 |\phi_j|^2 + \frac{d_{ij}}{2} (\phi_i^* \phi_j - \phi_j^* \phi_i)^2 \right)$$



Higgs modes in d-wave SC: nonequilibrium

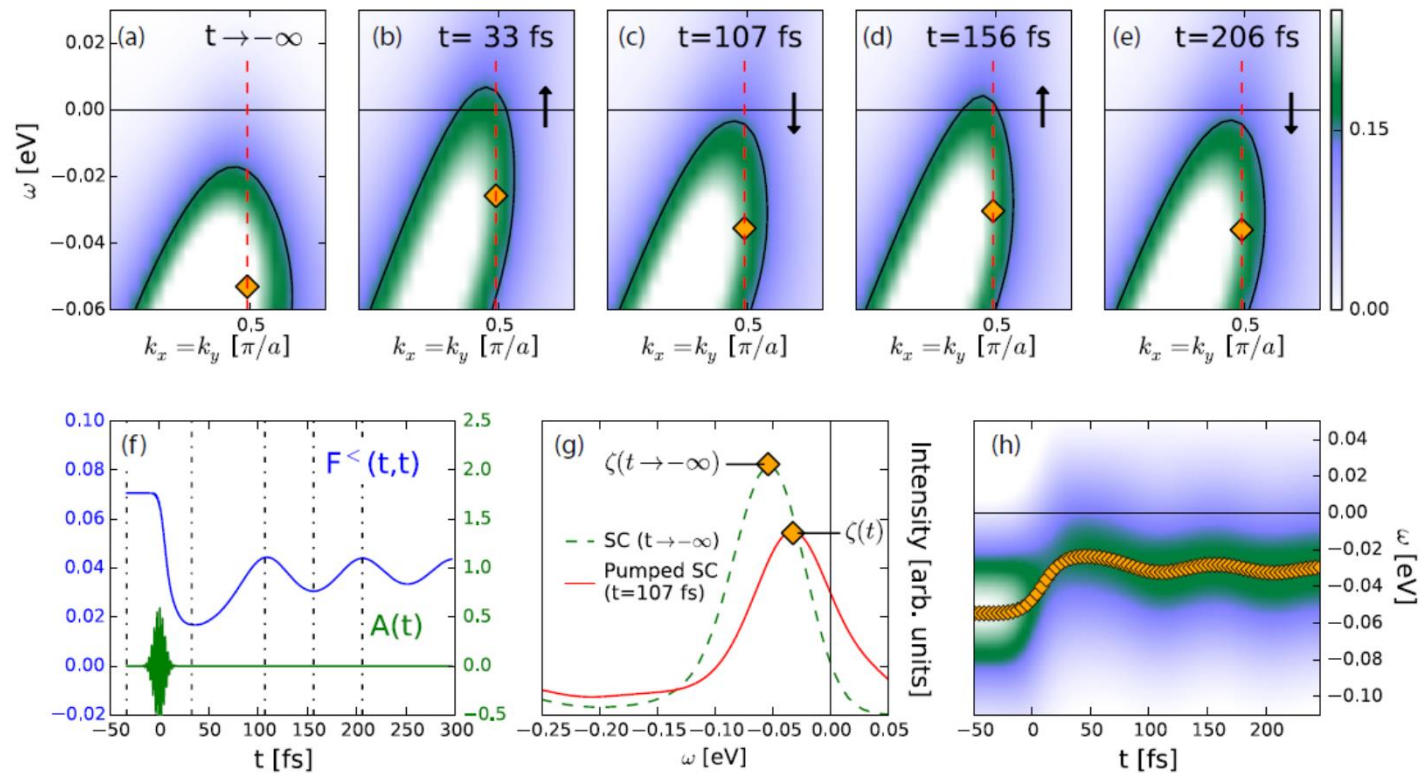
Gap Function	Symmetry		Modes	Excitation geometry
	Oscillation	Quench		
s	A_s^{1g}	$[2(x^2 - y^2)^2 - 1]$		$\phi = \text{any}$
	A_s^{2g}	$[xy(x^2 - y^2)]$		$\phi = \text{any}$
	B_s^{1g}	$[x^2 - y^2]$		$\phi = \text{any}$
	B_s^{2g}	$[xy]$		$\phi = \text{any}$
$d_{x^2-y^2}$	$A_{x^2-y^2}^{1g}$	$[x^2 - y^2]$		$\phi = \pi/4$
	$A_{x^2-y^2}^{2g}$	$[xy]$		$\phi = 0$
	$B_{x^2-y^2}^{1g}$	$[2(x^2 - y^2)^2 - 1]$		$\phi = 0$
	$B_{x^2-y^2}^{2g}$	$[xy(x^2 - y^2)]$		$\phi = \pi/4$
d_{xy}	A_{xy}^{1g}	$[xy]$		$\phi = 0$
	A_{xy}^{2g}	$[x^2 - y^2]$		$\phi = \pi/4$
	B_{xy}^{1g}	$[2(x^2 - y^2)^2 - 1]$		$\phi = \pi/4$
	B_{xy}^{2g}	$[xy(x^2 - y^2)]$		$\phi = 0$

B. Fauseweh, et al., arXiv:1712.0798

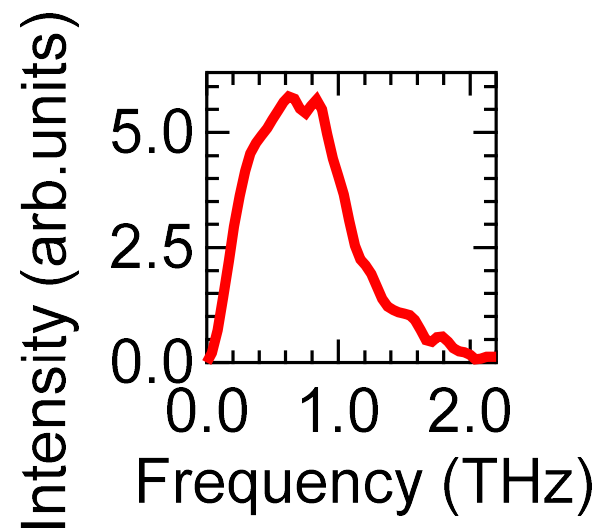
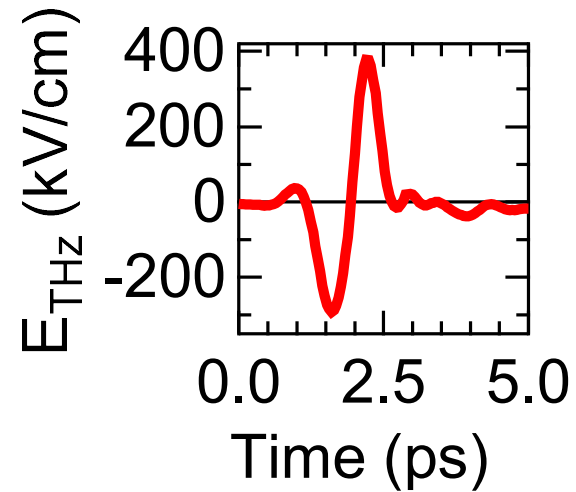
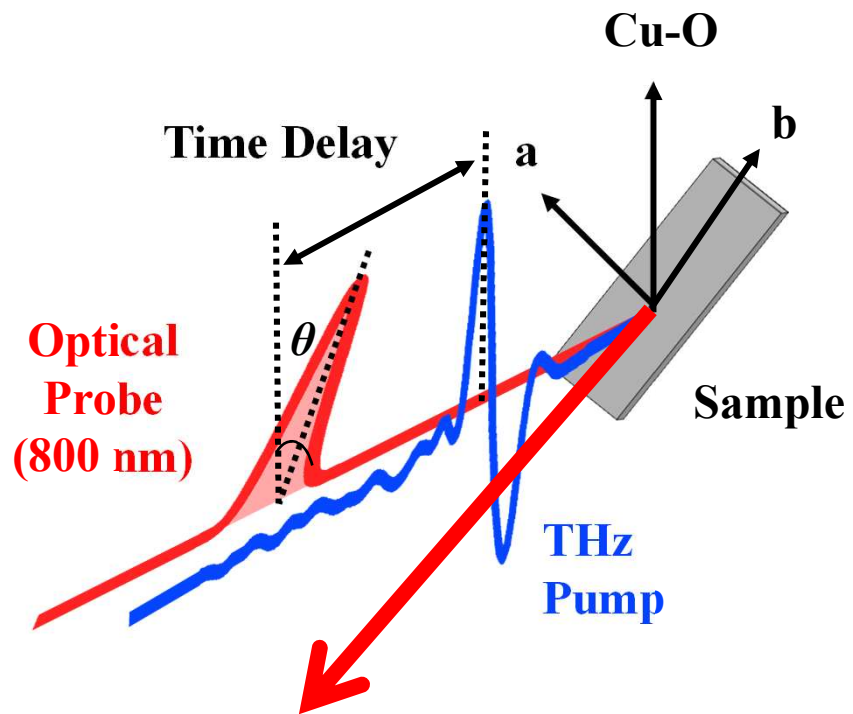
Time-resolved ARPES (Theory)

A. F. Kemper, M. A. Sentef, B. Moritz, J. K. Freericks, and T. P. Deveraux,
Phys. Rev. B 92, 224517 (2015)

B. Nosarzewski, B. Moritz, J. K. Freericks, A. F. Kemper,
and T. P. Deveraux *et al.*, arXiv 1609.04080(2016)



THz pump and optical probe experiments in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$

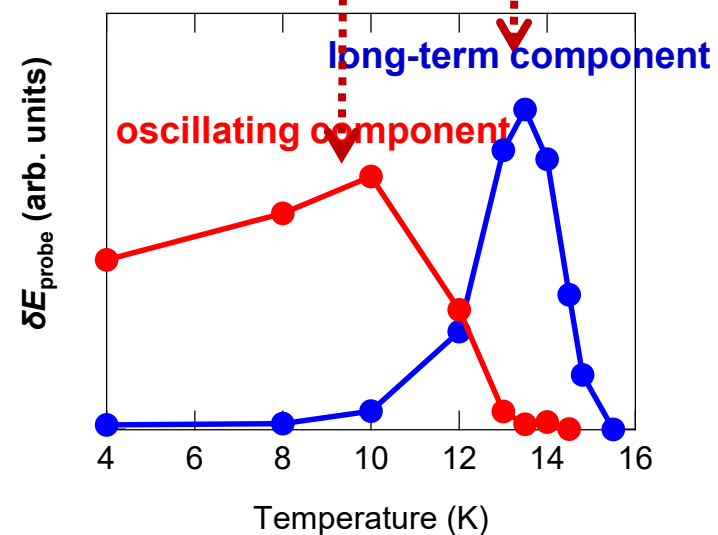
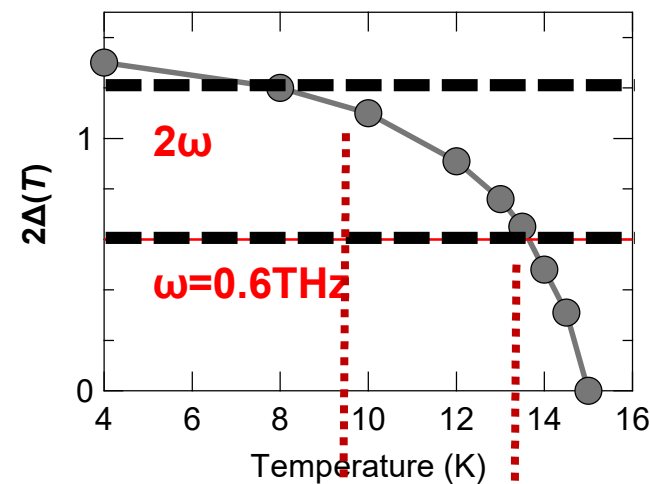
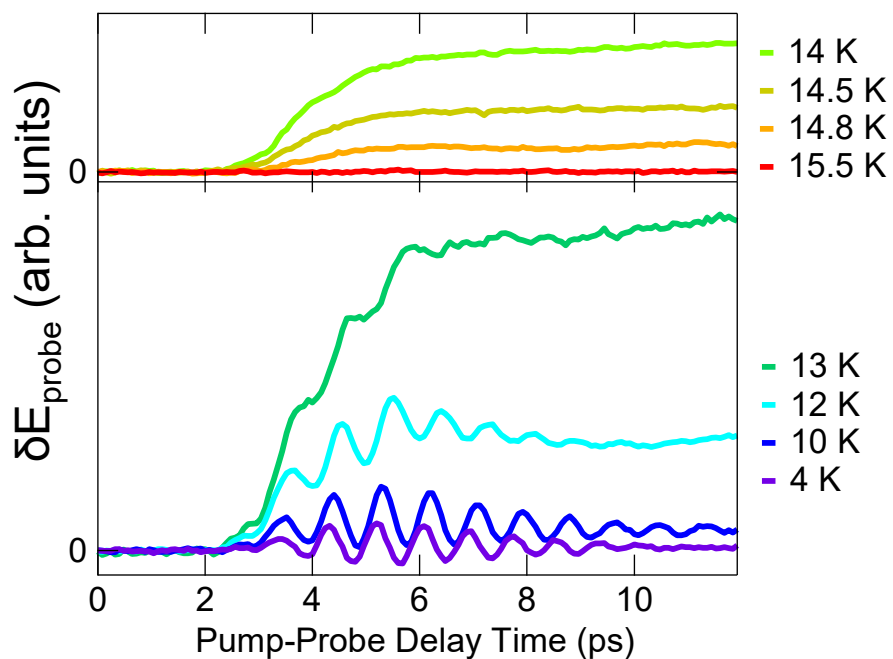
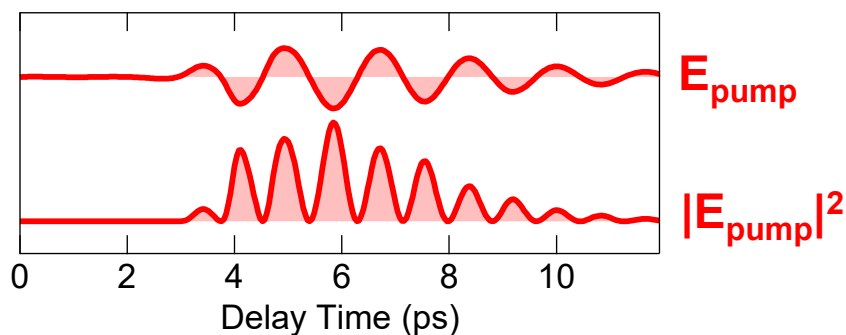


Coherent Excitation Regime Experiments

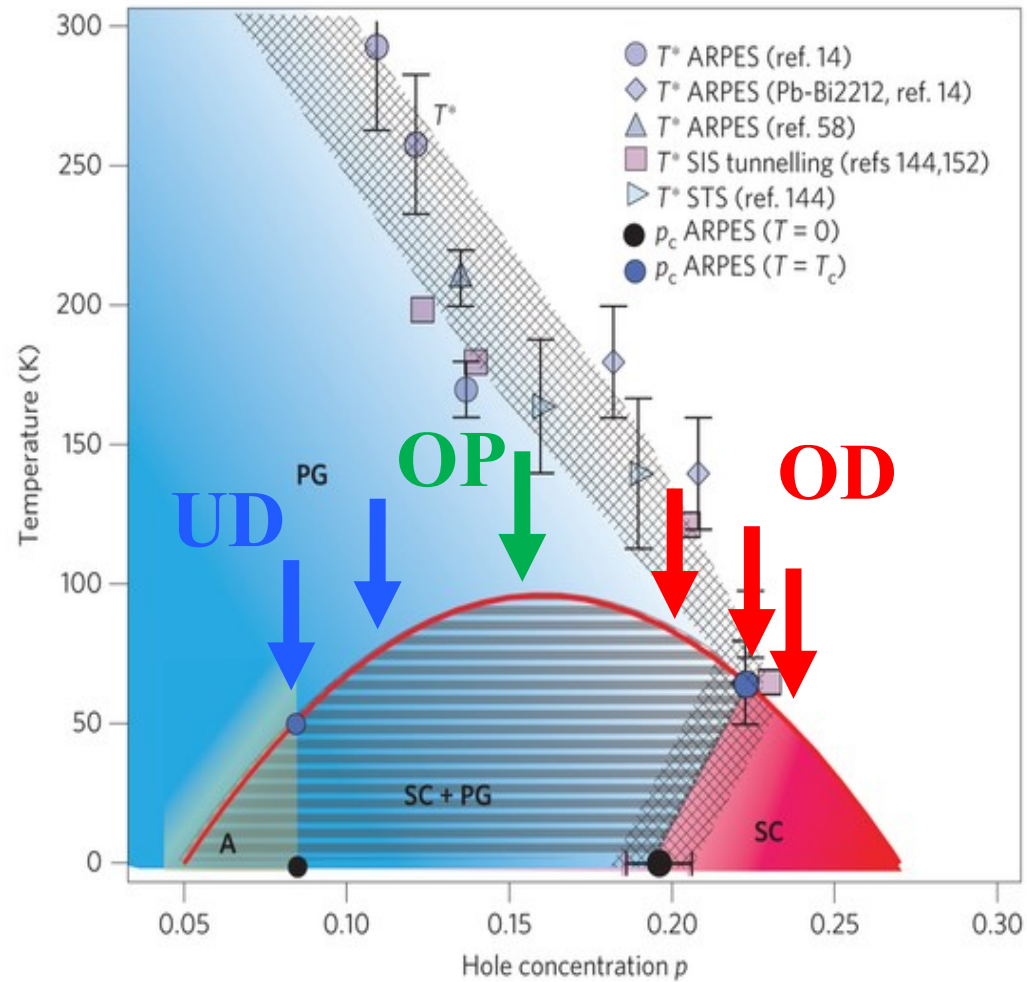
R. Matsunaga et al., Science **345**, 1145 (2014)

$\omega=0.6\text{THz}$

$E=3.5\text{ kV/cm @ peak}$

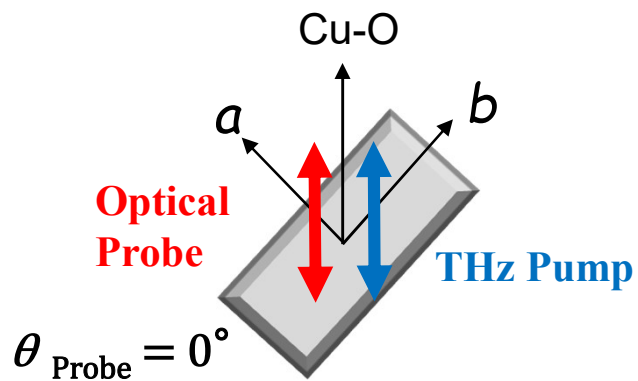
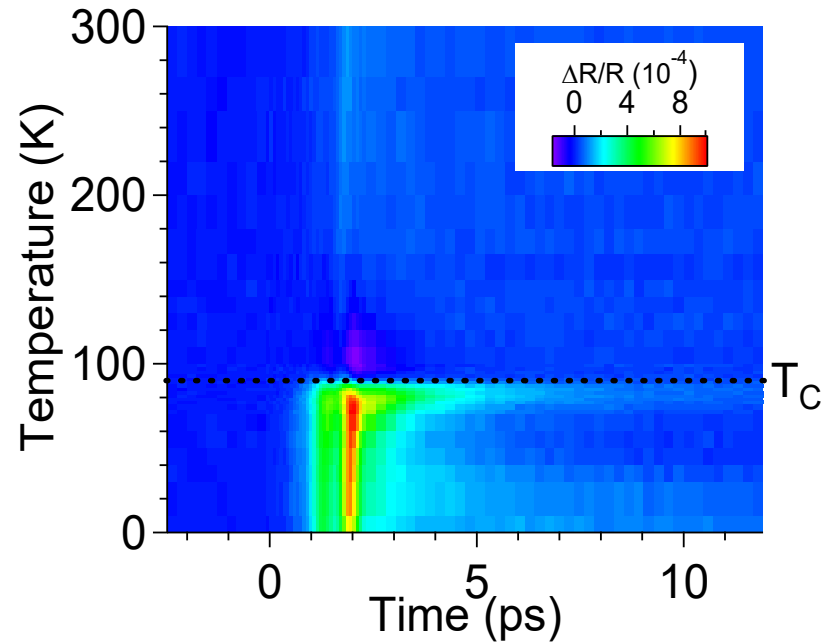
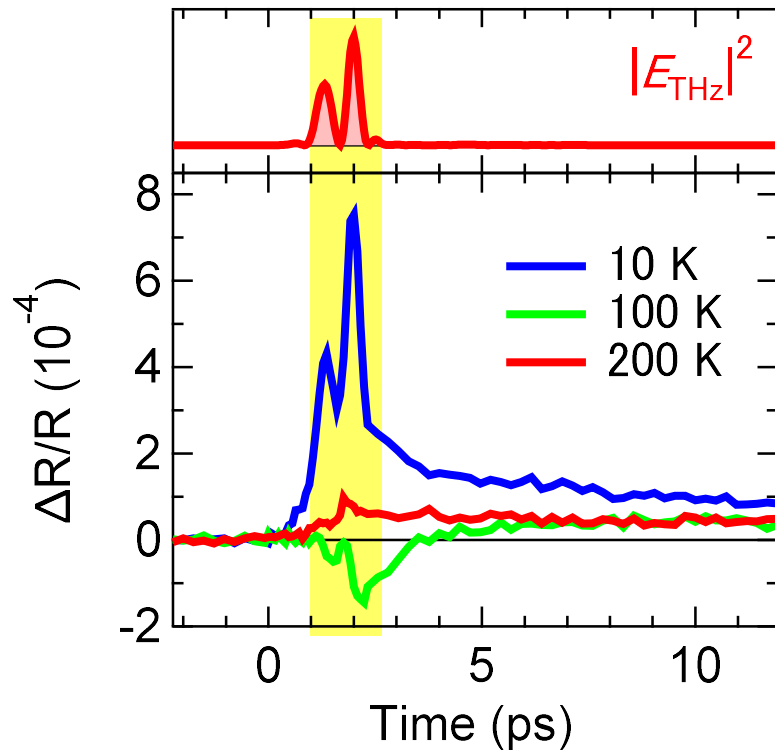


Phase diagram of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$

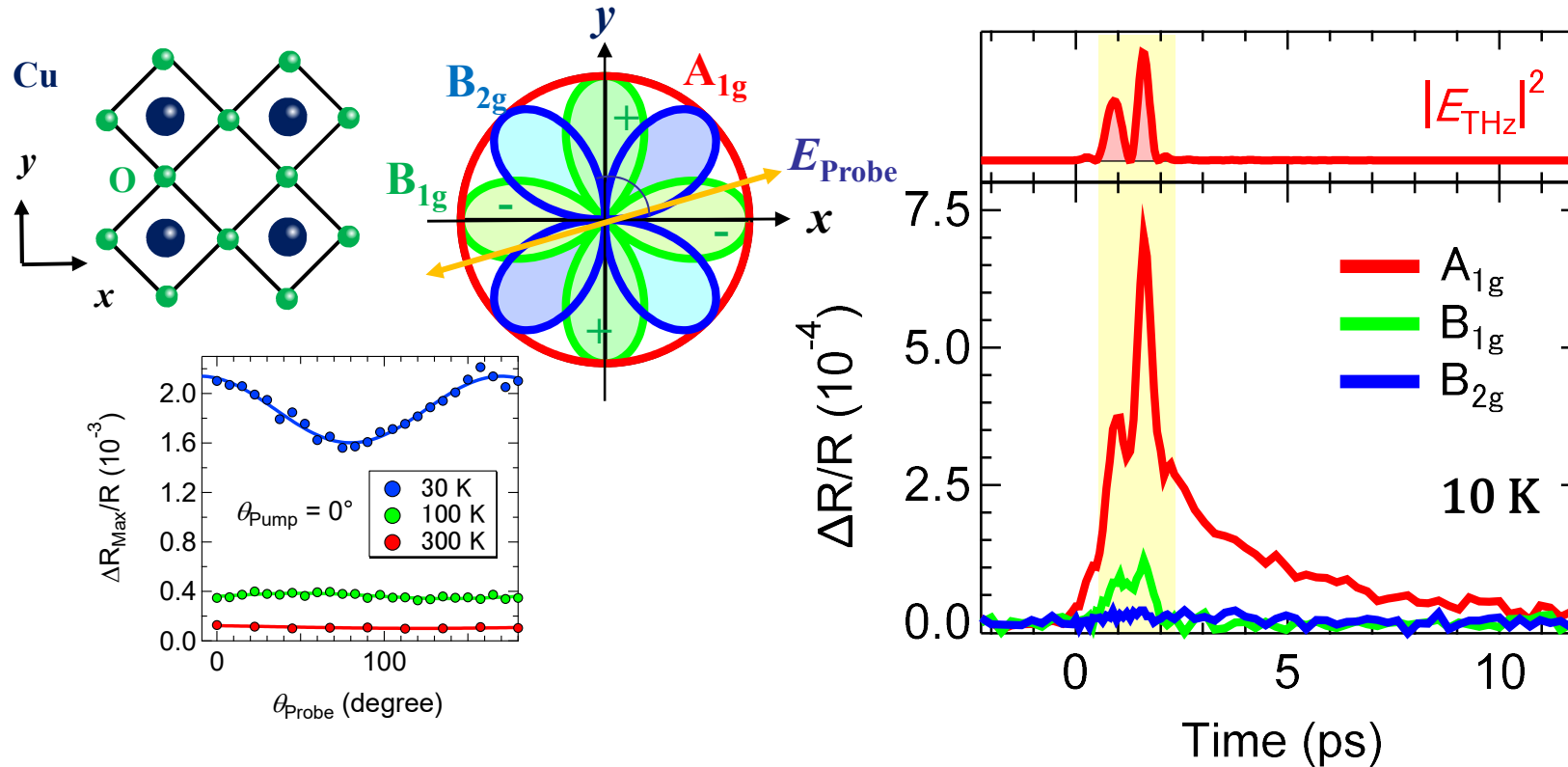


M. Hashimoto et al., Nat. Phys. 10, 483 (2014)

Transient reflectivity change



Symmetry of the signal

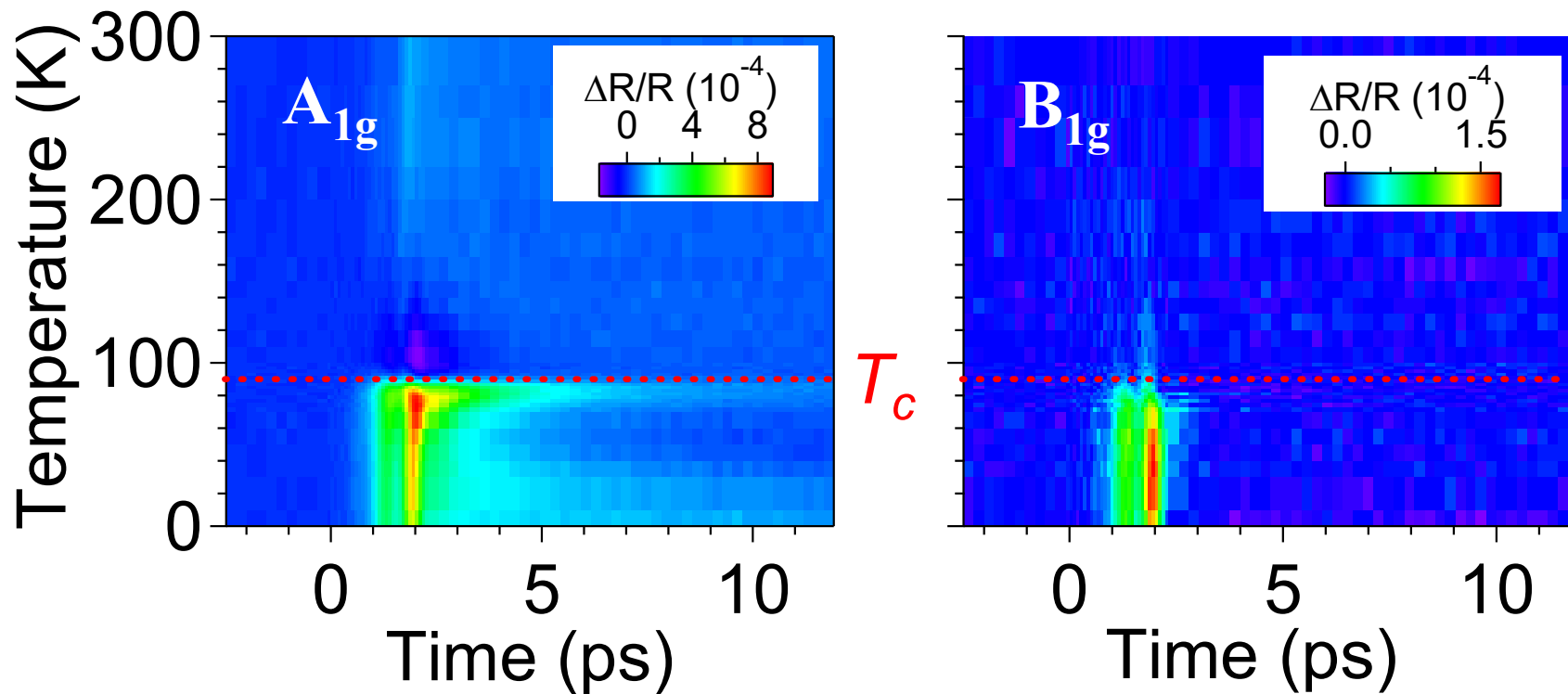


$$\frac{\Delta R}{R}(E_i^{probe}, E_j^{probe}) \sim \frac{1}{R} \frac{\partial R}{\partial \epsilon_1} \epsilon_0 \text{Re} \chi_{ijkl}^{(3)} E_k^{pump} E_l^{pump} \quad \text{THz-induced Kerr effect}$$

Bi2212: D_{4h} point group

$$\chi^{(3)} = \frac{1}{2} (\chi_{A_{1g}}^{(3)} + \chi_{B_{1g}}^{(3)} \cos 2\theta_{pump} \cos 2\theta_{probe} + \chi_{B_{2g}}^{(3)} \sin 2\theta_{pump} \sin 2\theta_{probe})$$

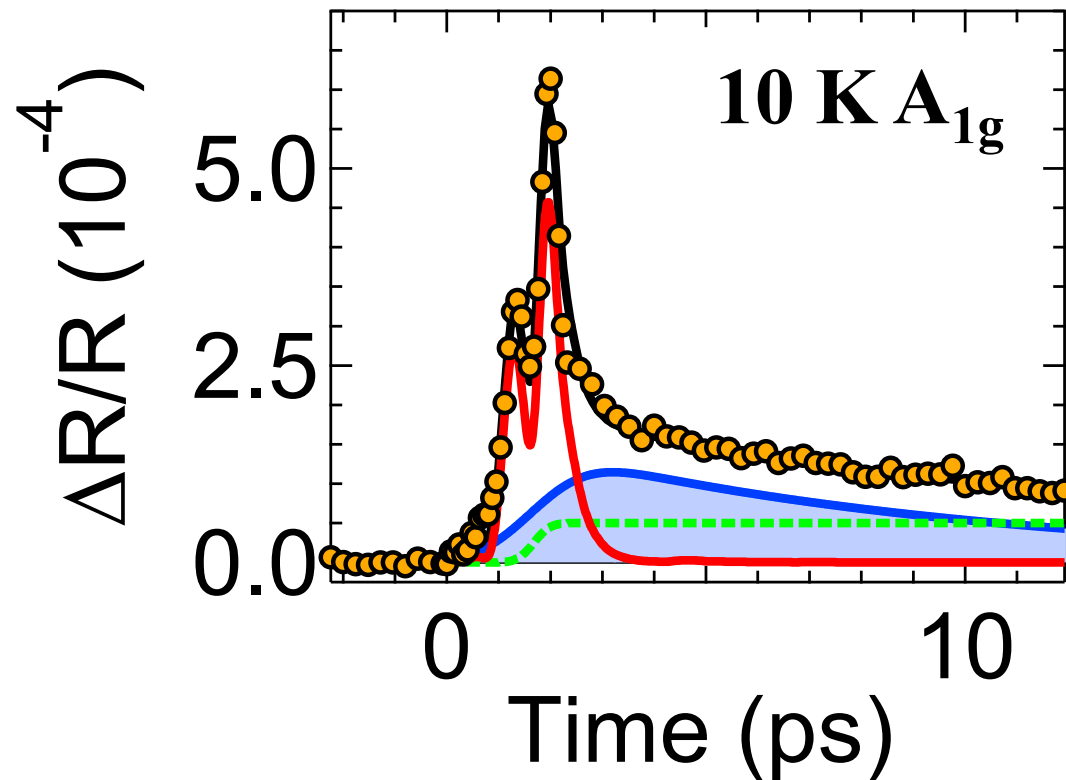
Temperature dependence of A_{1g} and B_{1g}



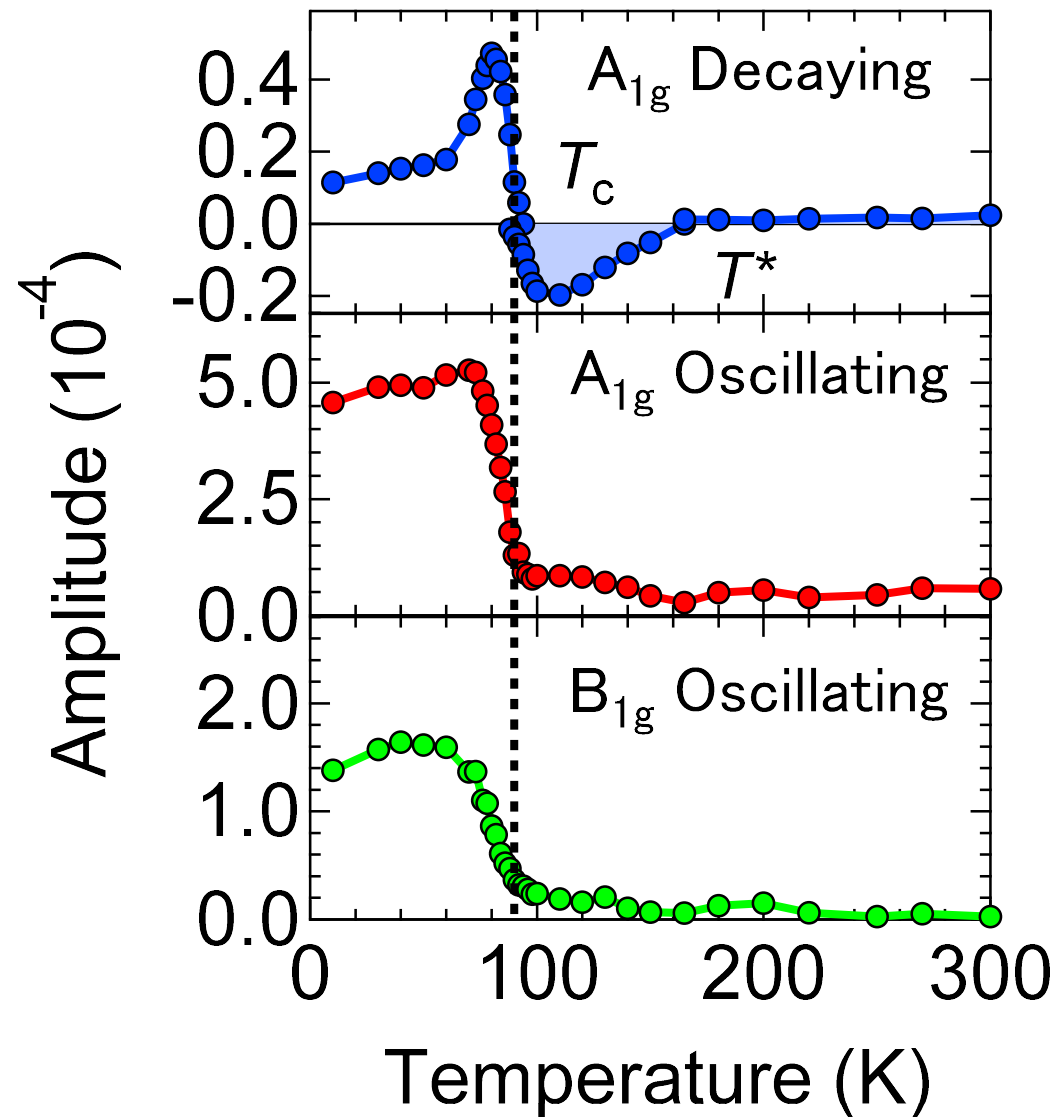
A_{1g} : oscillatory(coherent) component + decay(incoherent) component
 B_{1g} : only oscillatory(coherent) component

Decomposition into coherent and incoherent part

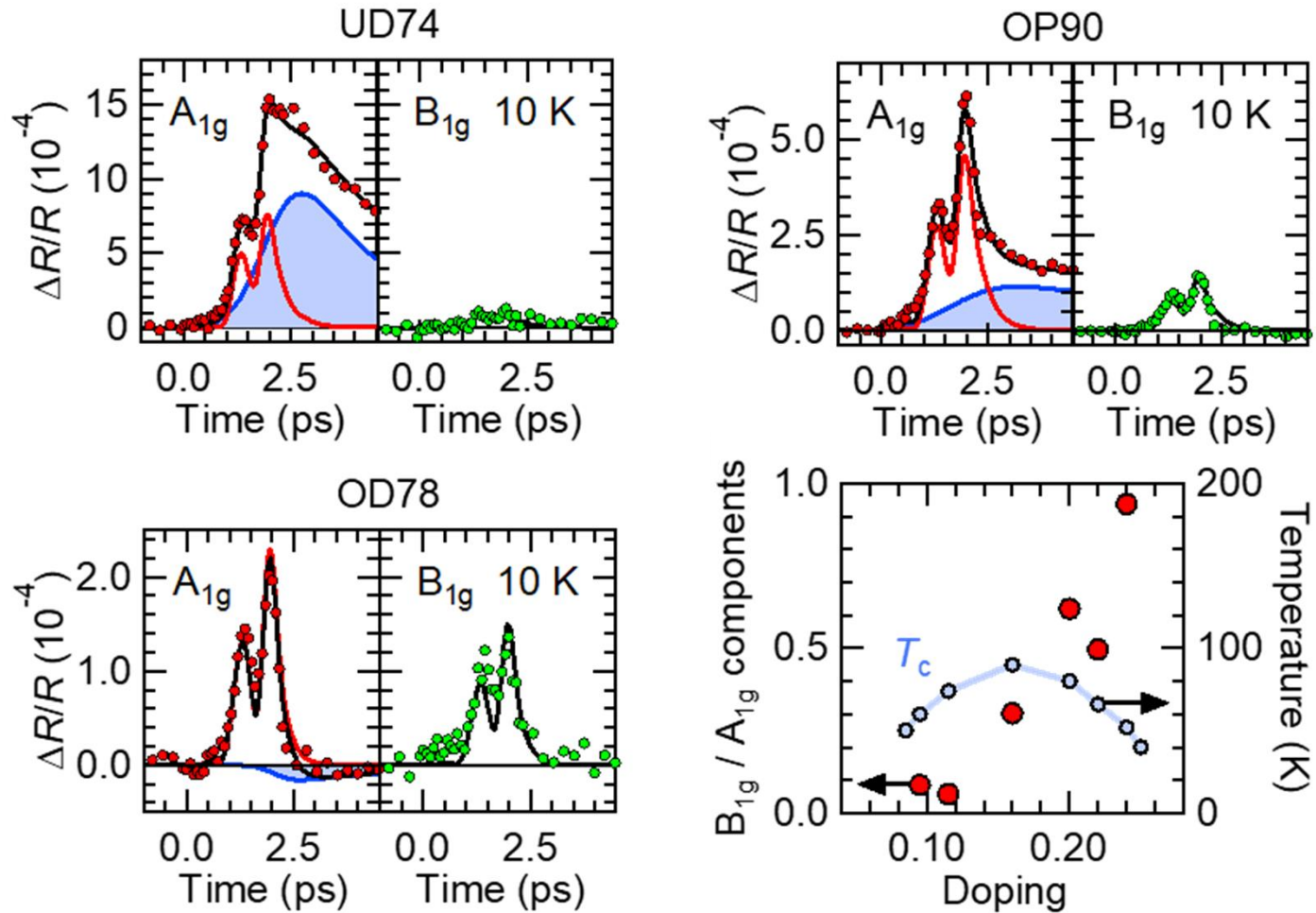
$$\frac{\Delta R}{R}(t) = A \int_{-\infty}^{\infty} |E_{\text{Pump}}(t - \tau)|^2 e^{-\frac{\tau}{\tau_0}} d\tau + B \int_{-\infty}^{\infty} e^{-\frac{\tau^2}{\tau_p^2}} e^{-\frac{t-\tau}{\tau_I}} d\tau + \text{Offset}$$



Temperature dependence of each component



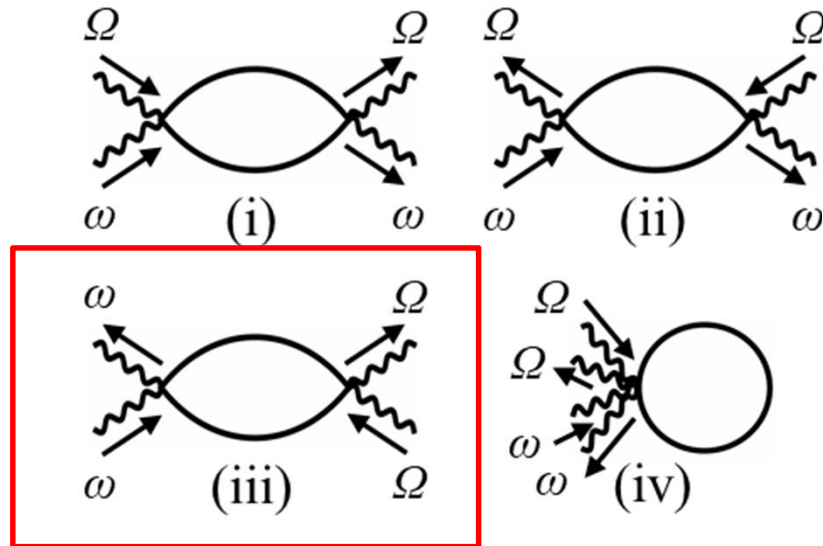
Doping dependence



A_{1g} signal is always dominant.

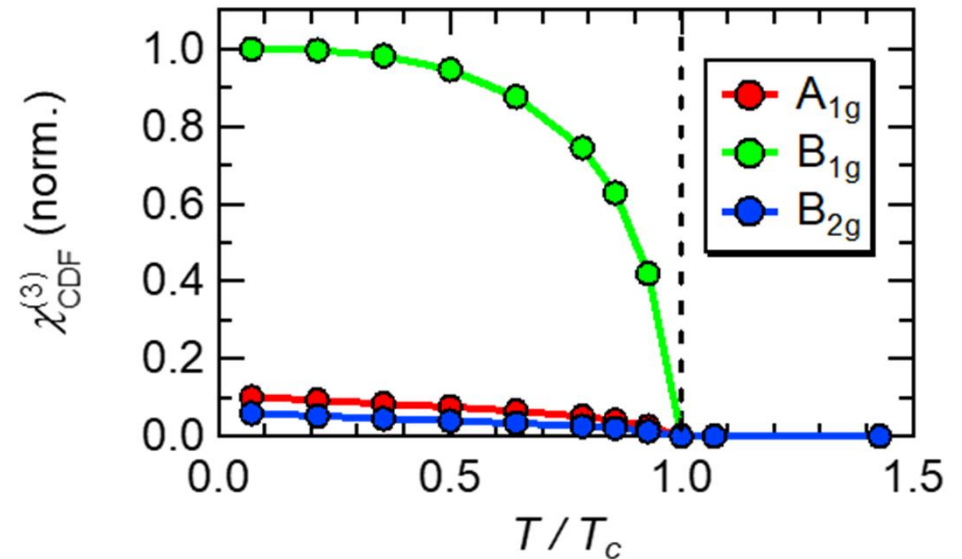
Polarization dependence of CDF

mean field(BCS) theory with d-wave symmetry



THz pump-optical probe

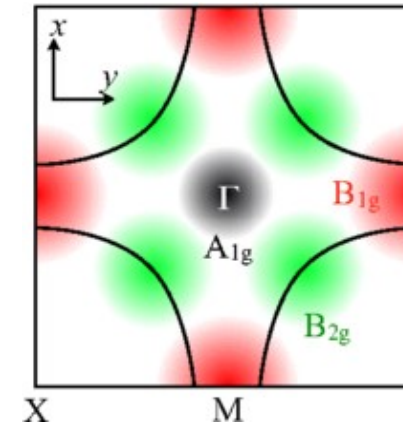
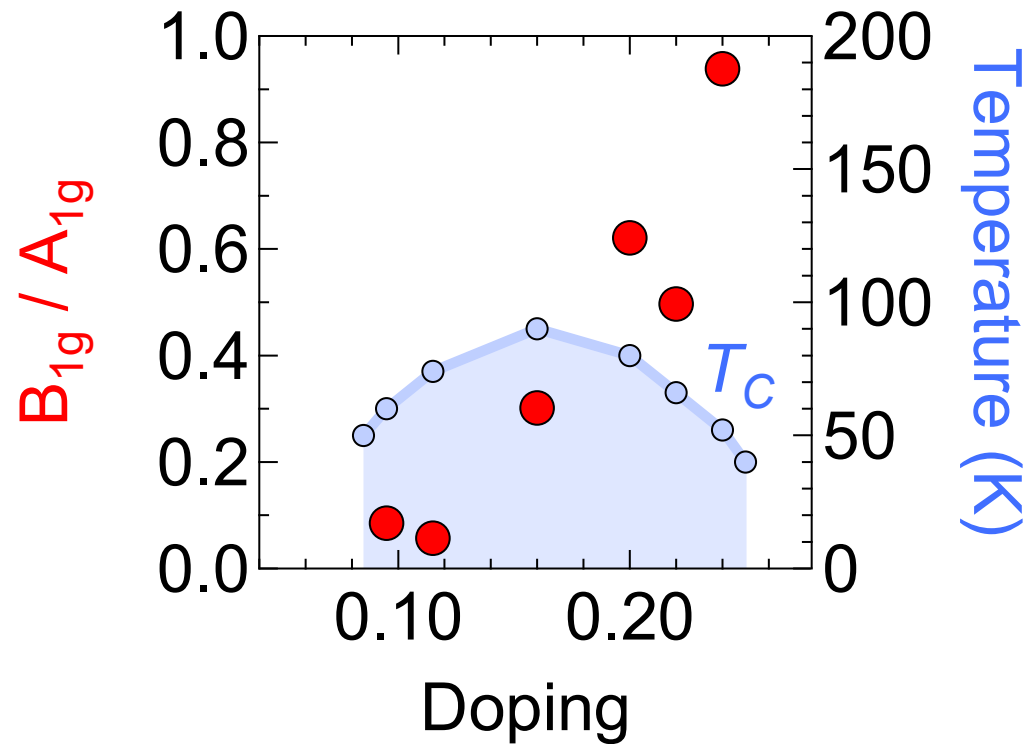
Ω : 4meV ω : 1.5 eV



CDF: B_{1g} is dominant

The dominance of A_{1g} signal cannot be explained by CDF.

Doping dependence of the oscillating component



A_{1g} signal is attributed to Higgs.

B_{1g} is most likely CDF.

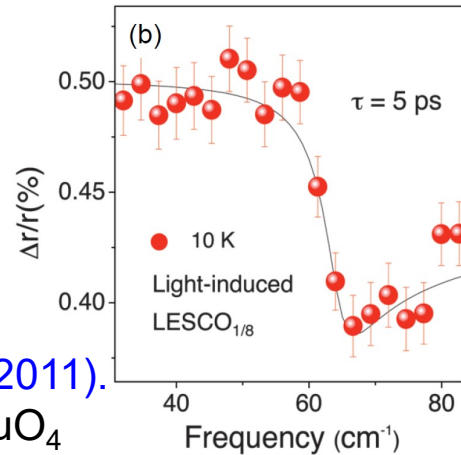
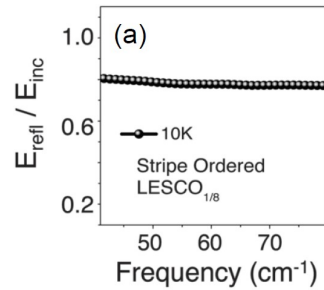
K. Katsumi et al., arXiv:1711.04923 (to be published in PRL)

poster presentation

Outline

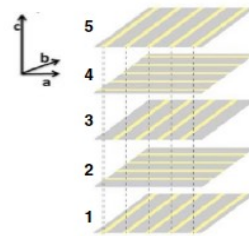
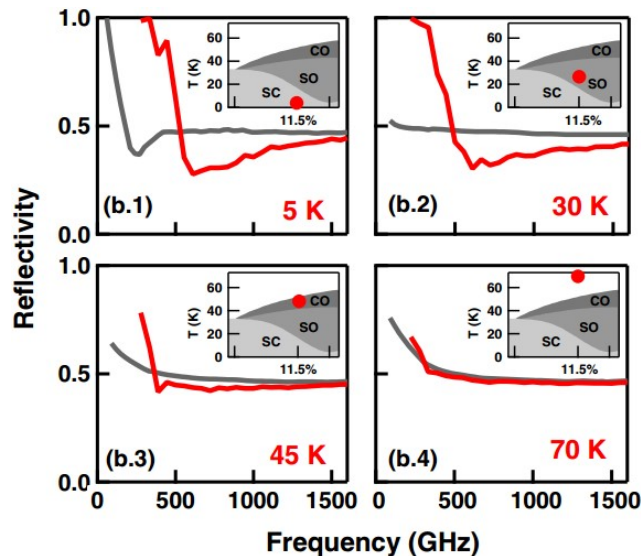
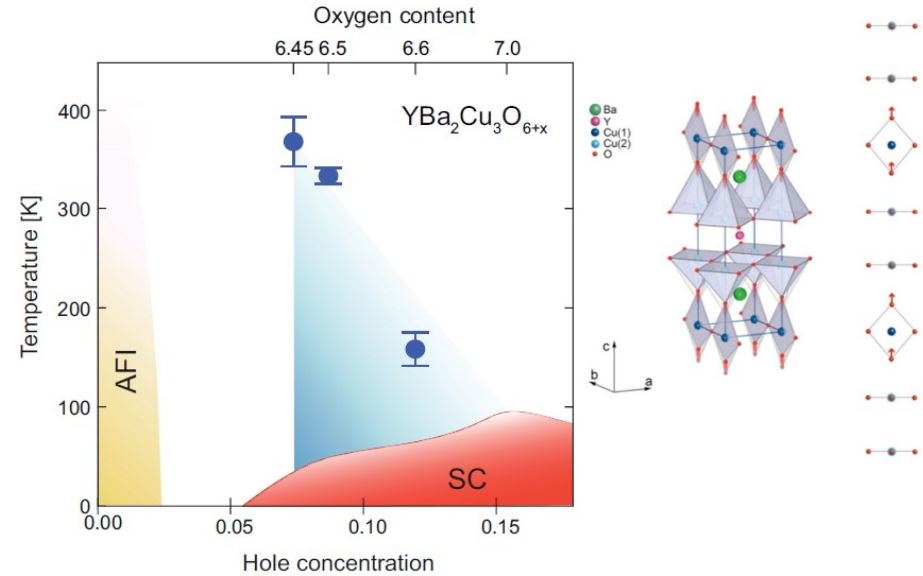
- (1) Introduction
- (2) Photoexcitation in s-wave superconductor
- (3) Higgs mode in a s-wave superconductor NbN
- (4) Higgs mode in d-wave cuprate superconductors
- (5) Photocontrol of superconductors

Photoinduced superconductivity



D. Fausti *et al.*,
Science 331, 189 (2011).

$\text{La}_{1.675}\text{Eu}_{0.2}\text{Sr}_{0.125}\text{CuO}_4$
Pump ($//ab$): $15 \mu\text{m}$



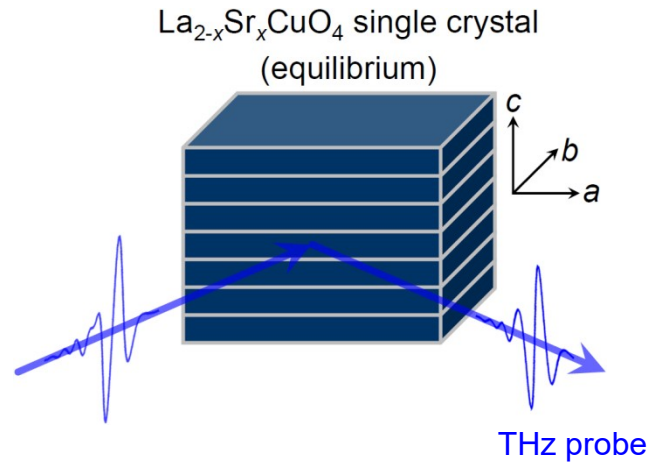
S. Kaiser *et al.*,
PRB 89, 184516 (2014).

$\text{YBa}_2\text{Cu}_3\text{O}_{6.45}$
Pump ($//c$): $15 \mu\text{m}$

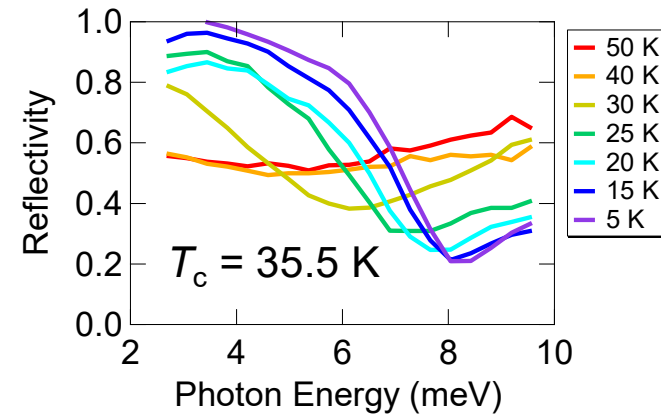
D. Nicoletti *et al.*,
PRB 90, 100503(R) (2014).

$\text{La}_{1.885}\text{Ba}_{0.115}\text{CuO}_4$
Pump ($//c$): 800 nm

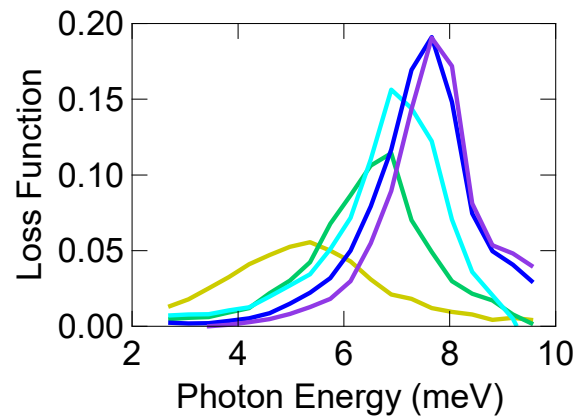
c-axis spectra of optimally doped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$



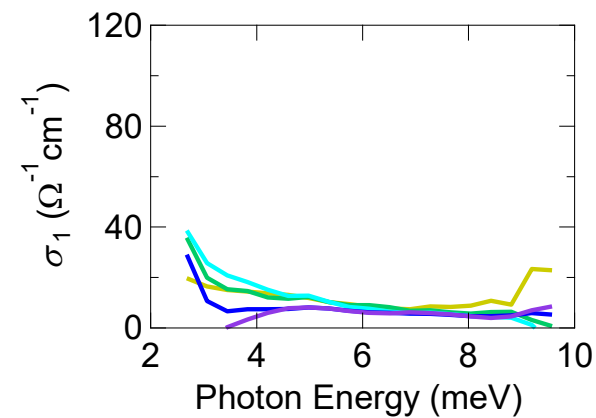
Reflectivity



Loss function : $\text{Im}(-1/\epsilon(\omega))$

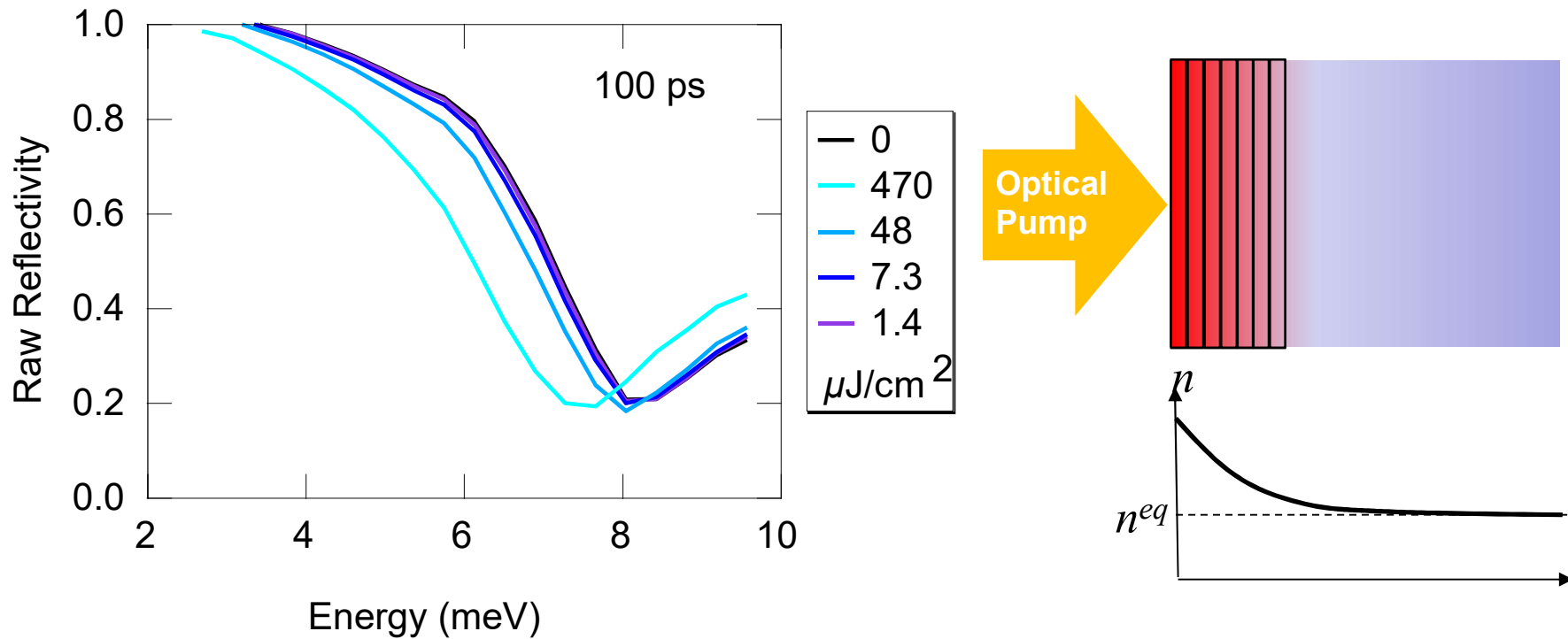


Optical conductivity $\sigma_1(\omega)$



In equilibrium: only one longitudinal mode

Reflectivity spectra under the 800-nm pump

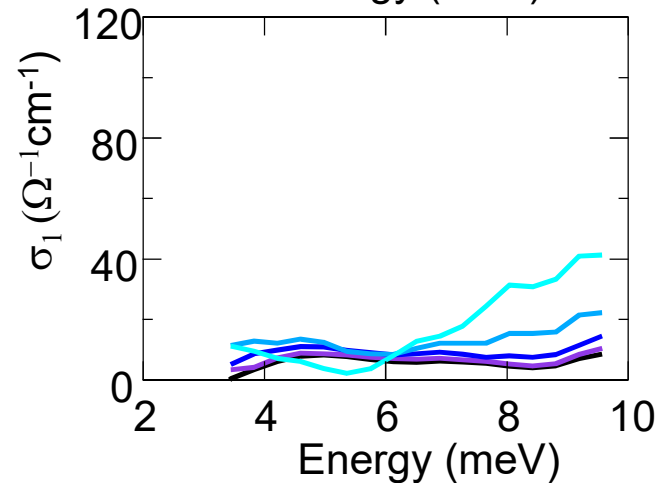
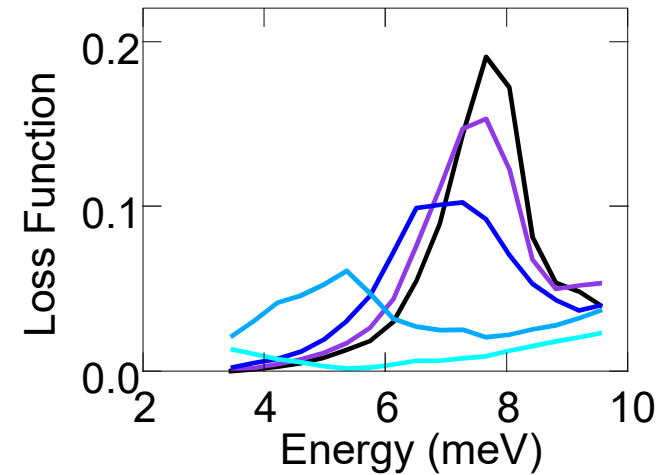
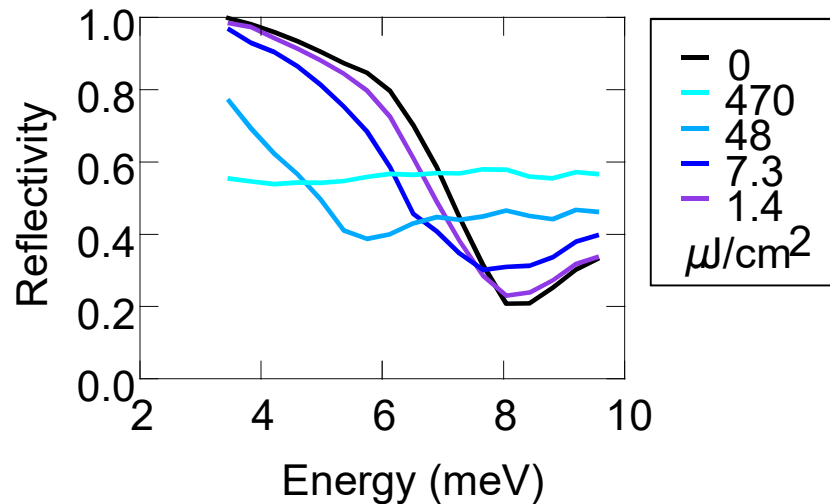
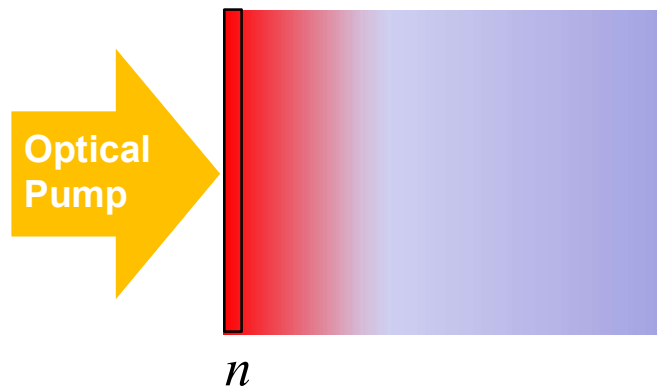


$$n(\nu, z) = n^{eq}(\nu) + [n^{ex}(\nu) - n^{eq}(\nu)] \exp(-z/d_{\text{pump}})$$

800-nm pump: $d_{\text{pump}} = 600 \text{ nm}$

THz: $d_{\text{THz}} \sim 20 \mu\text{m}$

Optical spectra at the surface region under weak excitation

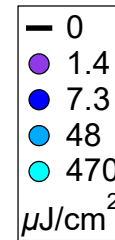
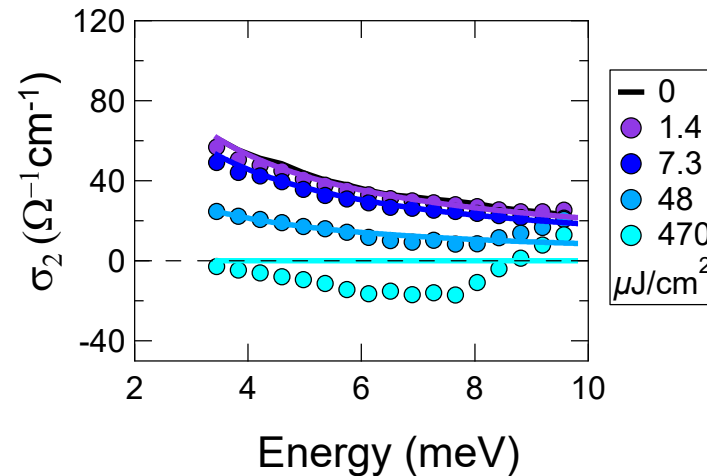
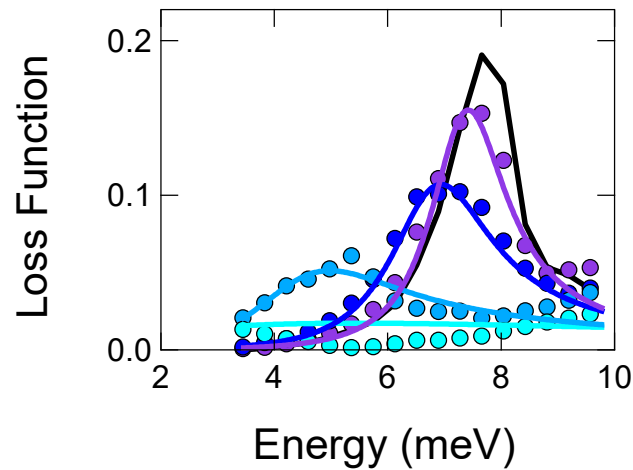
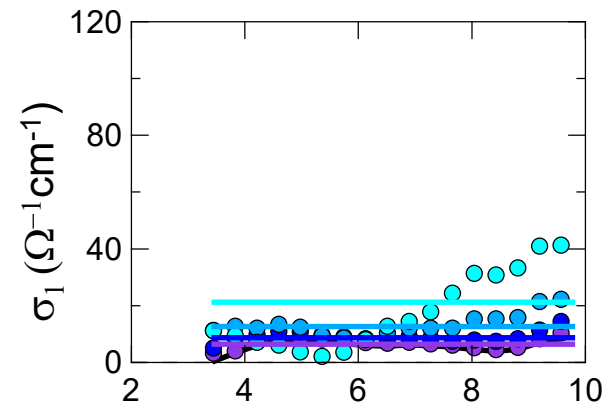
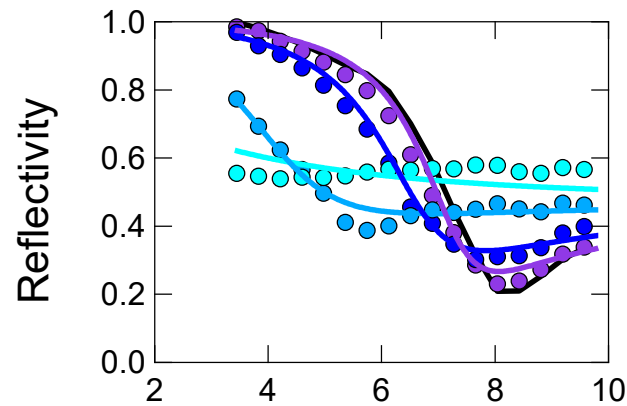


Consistent with optical pump optical probe at $t=100\text{ps}$ [M. Beyer, et al., Phys. Rev. B 83, 214515 (2011).]. Energy required to destruct SC~14 K/Cu~150 $\mu\text{J}/\text{cm}^2$.

Fitting by two fluid model

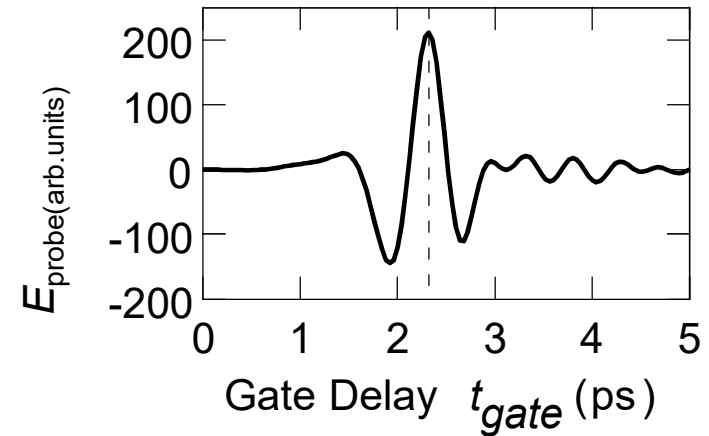
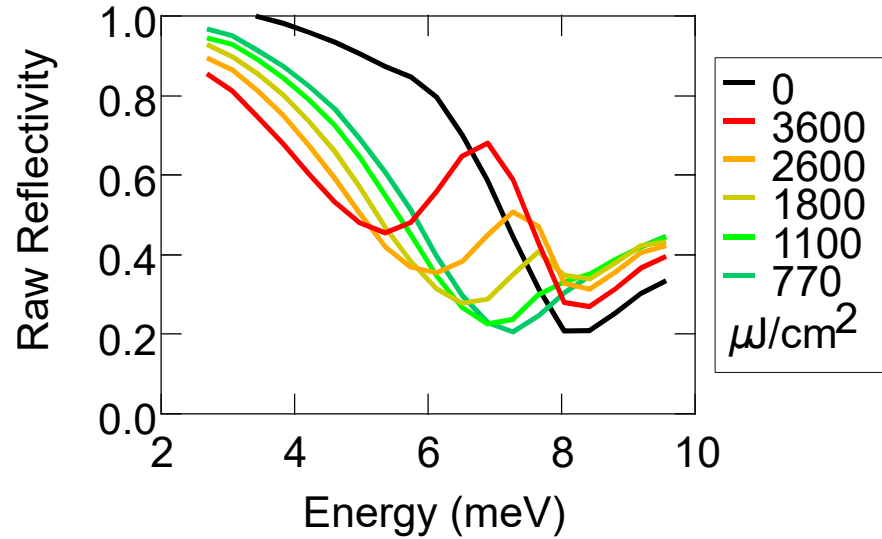
$$\varepsilon(\omega) = \varepsilon_\infty \left[1 - \frac{\omega_0^2}{\omega^2} + i \frac{\sigma_n}{\varepsilon_0 \omega} \right]$$

Continuous suppression of the Josephson plasma resonance

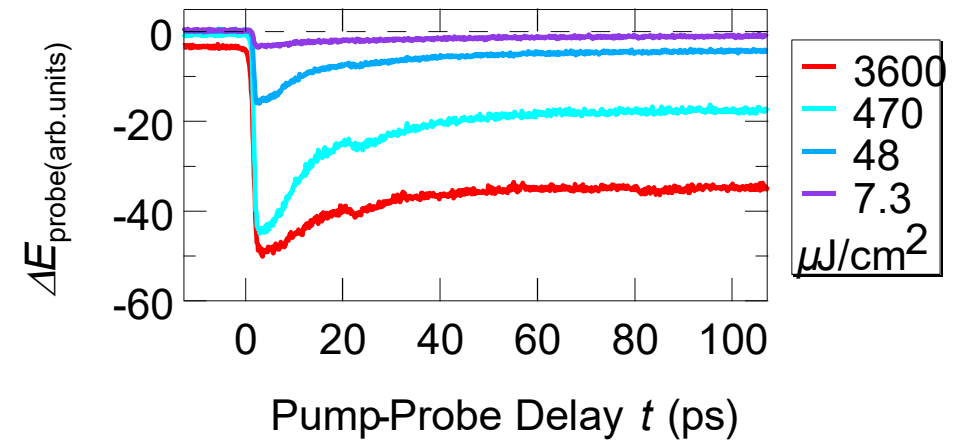
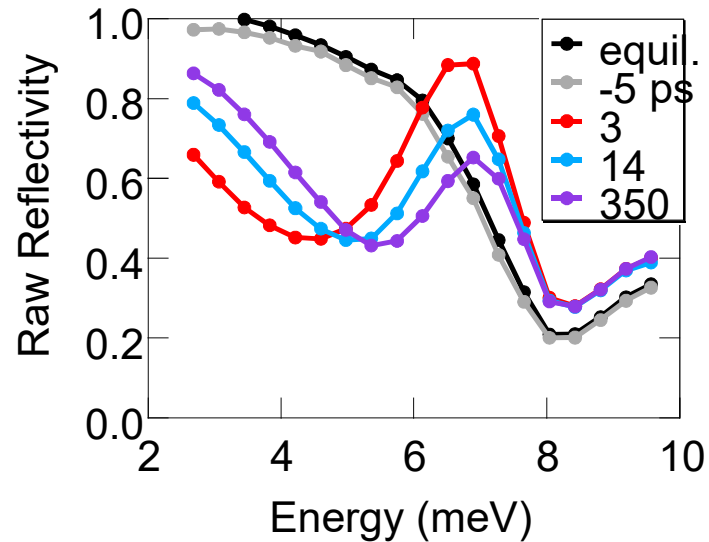


Reflectivity spectra under strong excitation

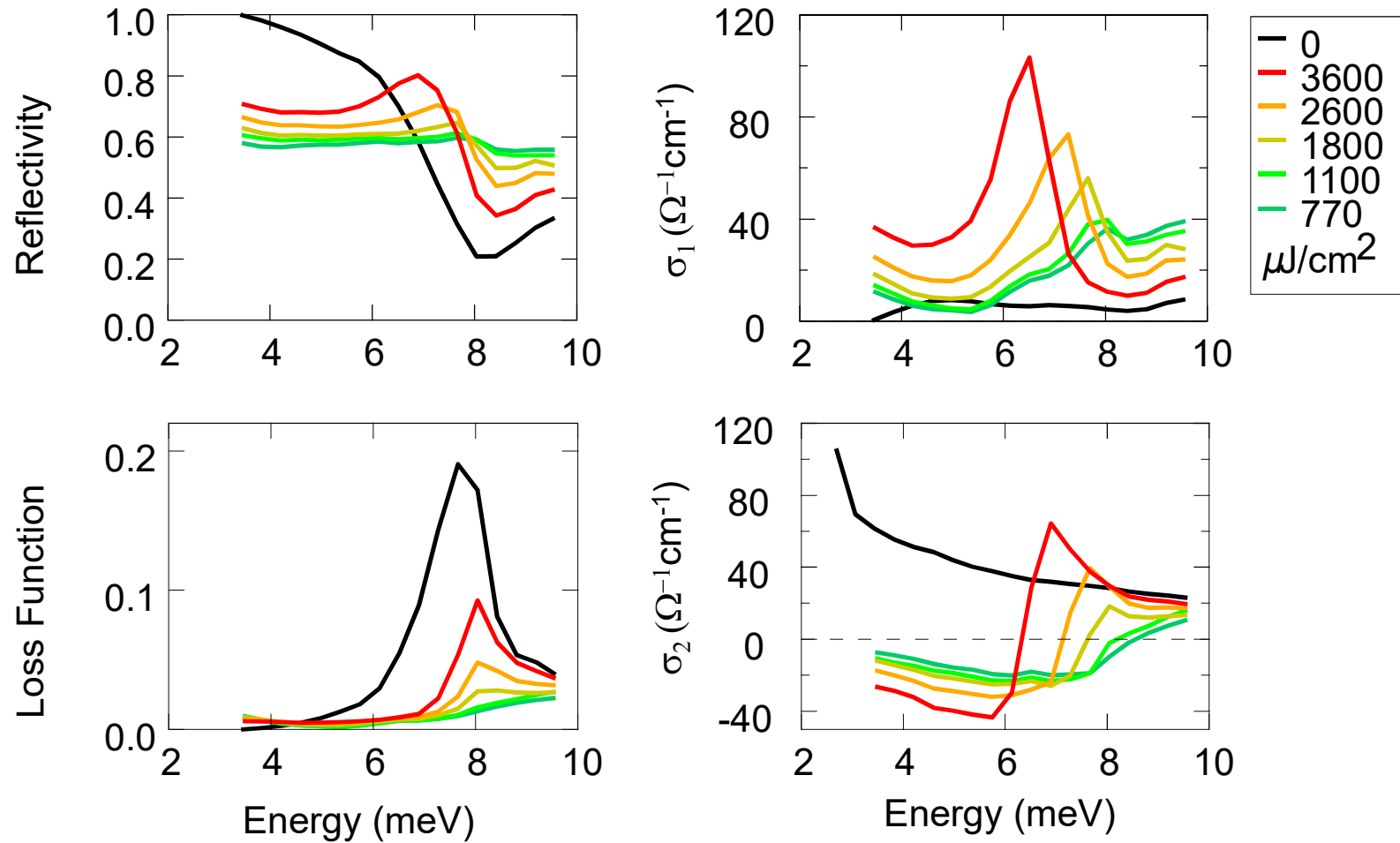
Excitation fluence dependence at 100 ps



Dynamics



Optical spectra at the surface region under strong excitation



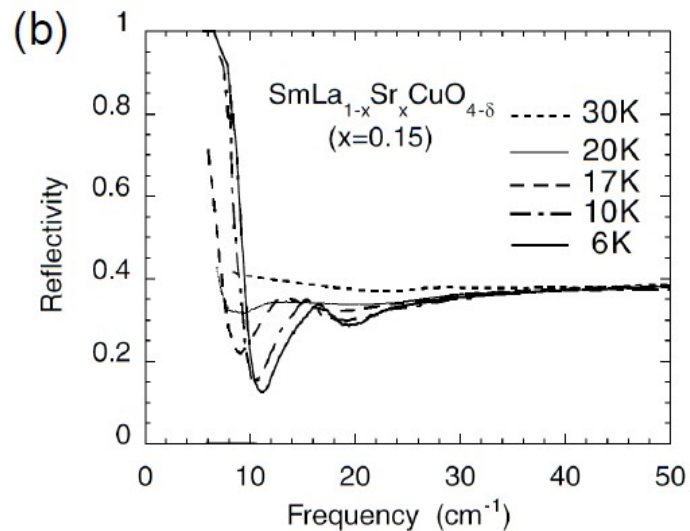
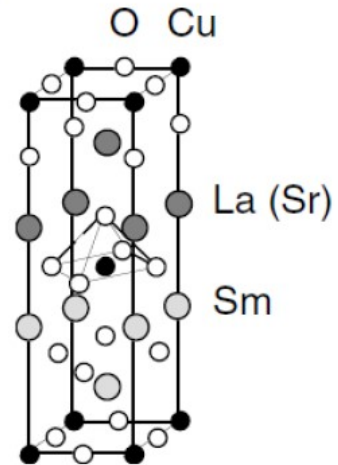
A new longitudinal mode

A new transverse mode

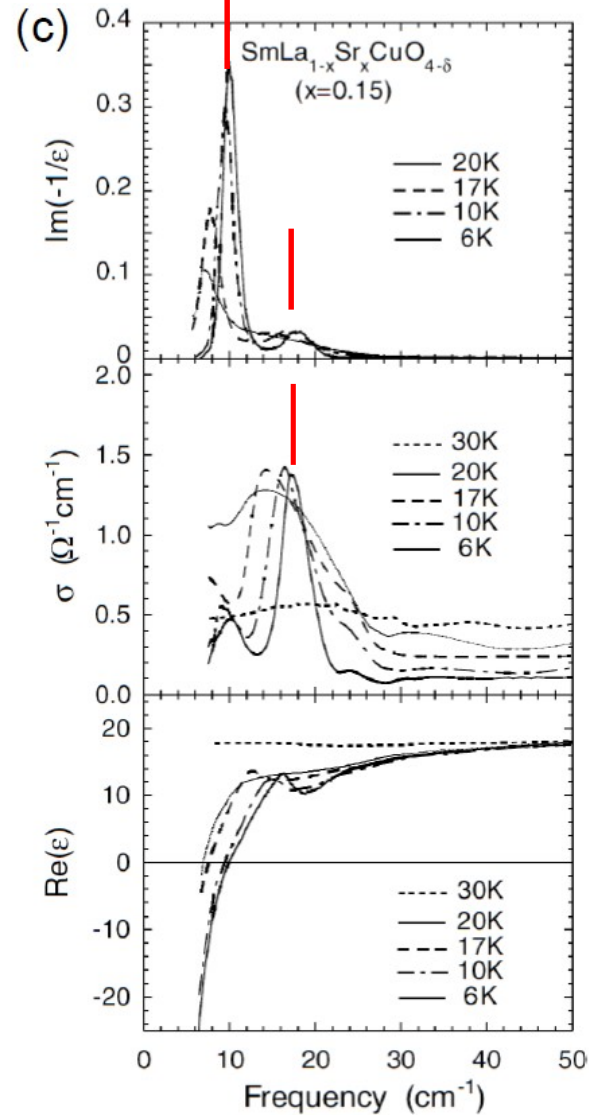
Double JPR in T*214



(a)

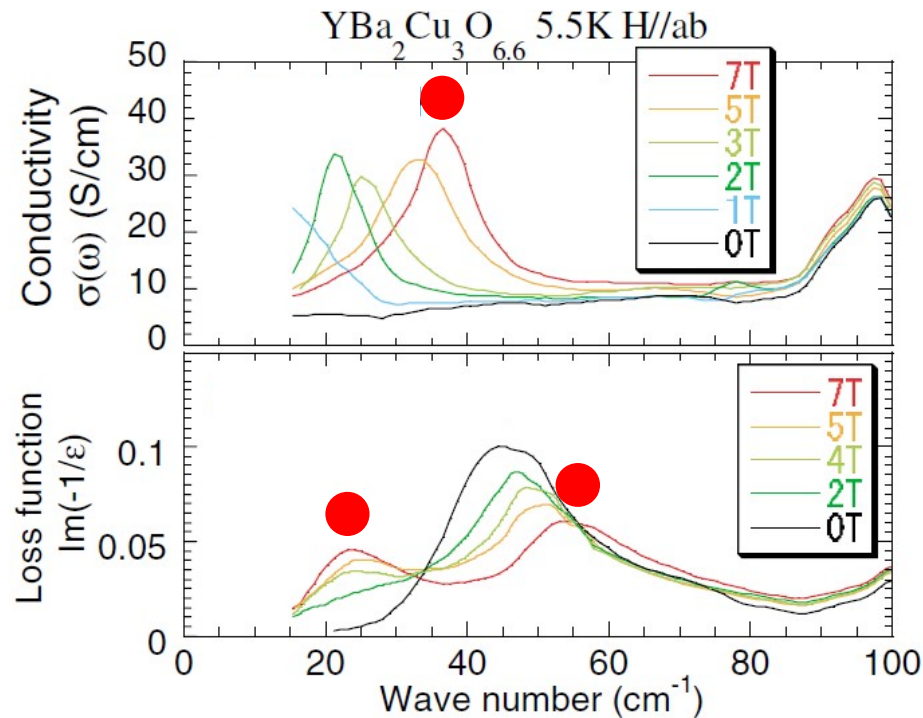


T. Kakeshita et al., Phys. Rev. Lett. 86, 2122 (2001).



Two longitudinal
and one Transverse
JPR modes

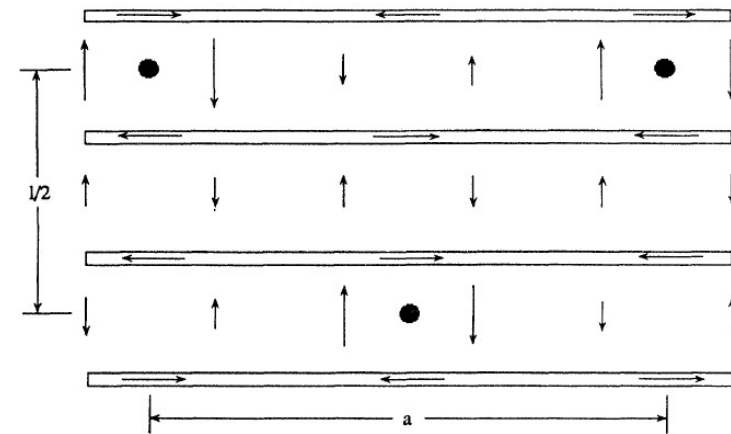
Double JPR of $\text{YBa}_2\text{Cu}_3\text{O}_{6.6}$ under B-field



K. M. Kojima *et al.*, PRL **89**, 247001 (2002).

- Splitting of longitudinal JPR
- Emergence of transverse JPR

Josephson vortex



Bulaevskii and Clem, PRB **44**, 10234 (1991).

Two kinds of Josephson coupling



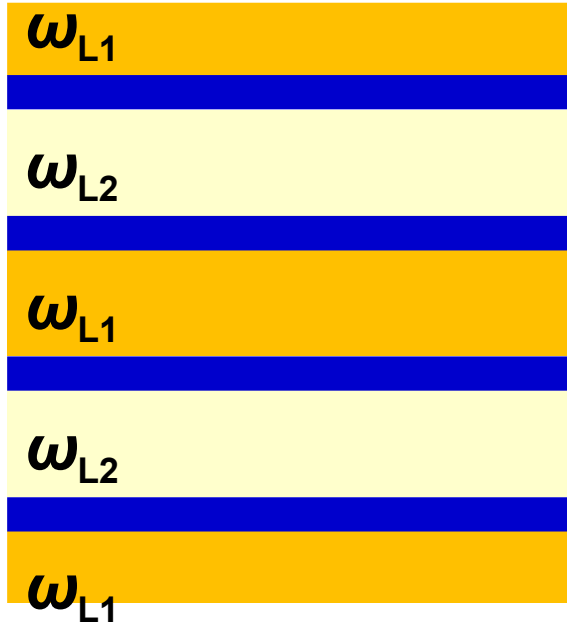
“multilayer model”

Multilayer model

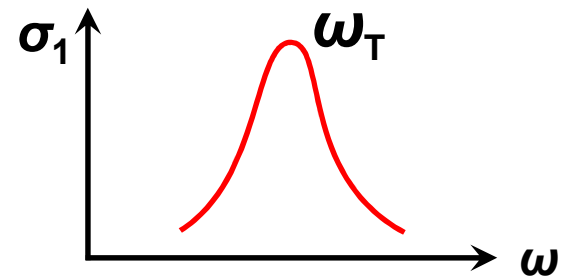
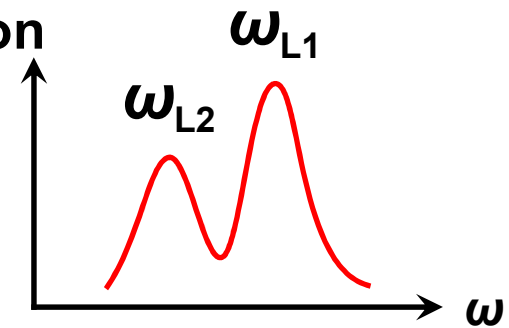
D. van der Marel and A. Tsvetkov,
Czech. J. Phys. 46, 3165 (1996);
PRB 64, 024530 (2001).

$$\varepsilon_j(\omega) = \varepsilon_\infty \left[1 - \frac{\omega_j^2}{\omega^2} + i \frac{\sigma_{n,j}}{\varepsilon_0 \omega} \right]$$

$$\frac{1}{\varepsilon_{\text{MLM}}(\omega)} = \sum_j \frac{z_j}{\varepsilon_j(\omega)}$$

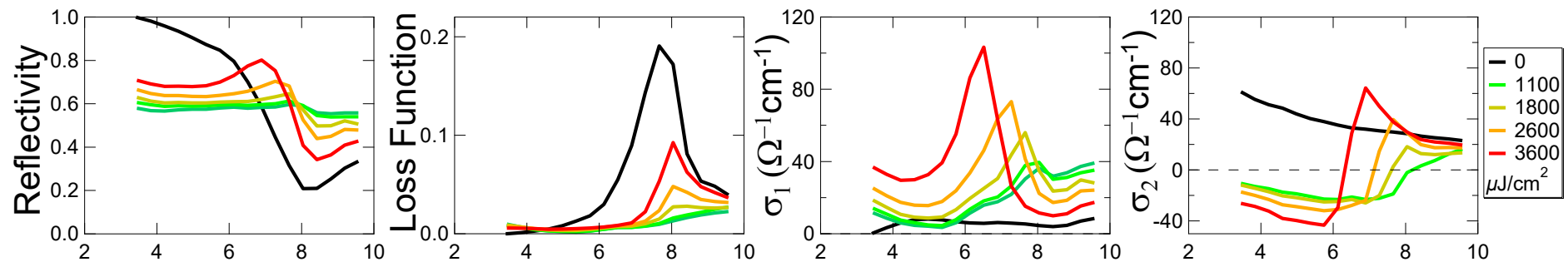


Loss Function

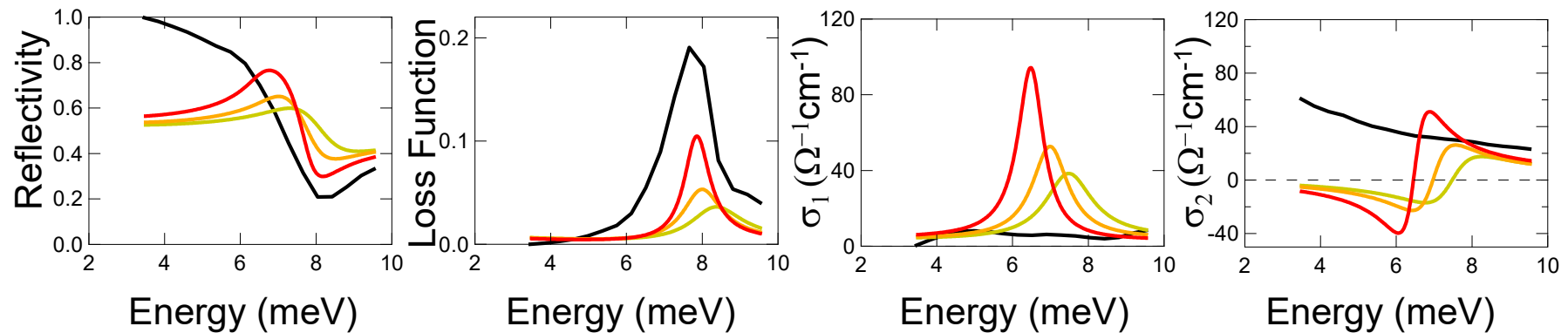


Fitting by the extended multilayer model in the strong photoexcitation regime

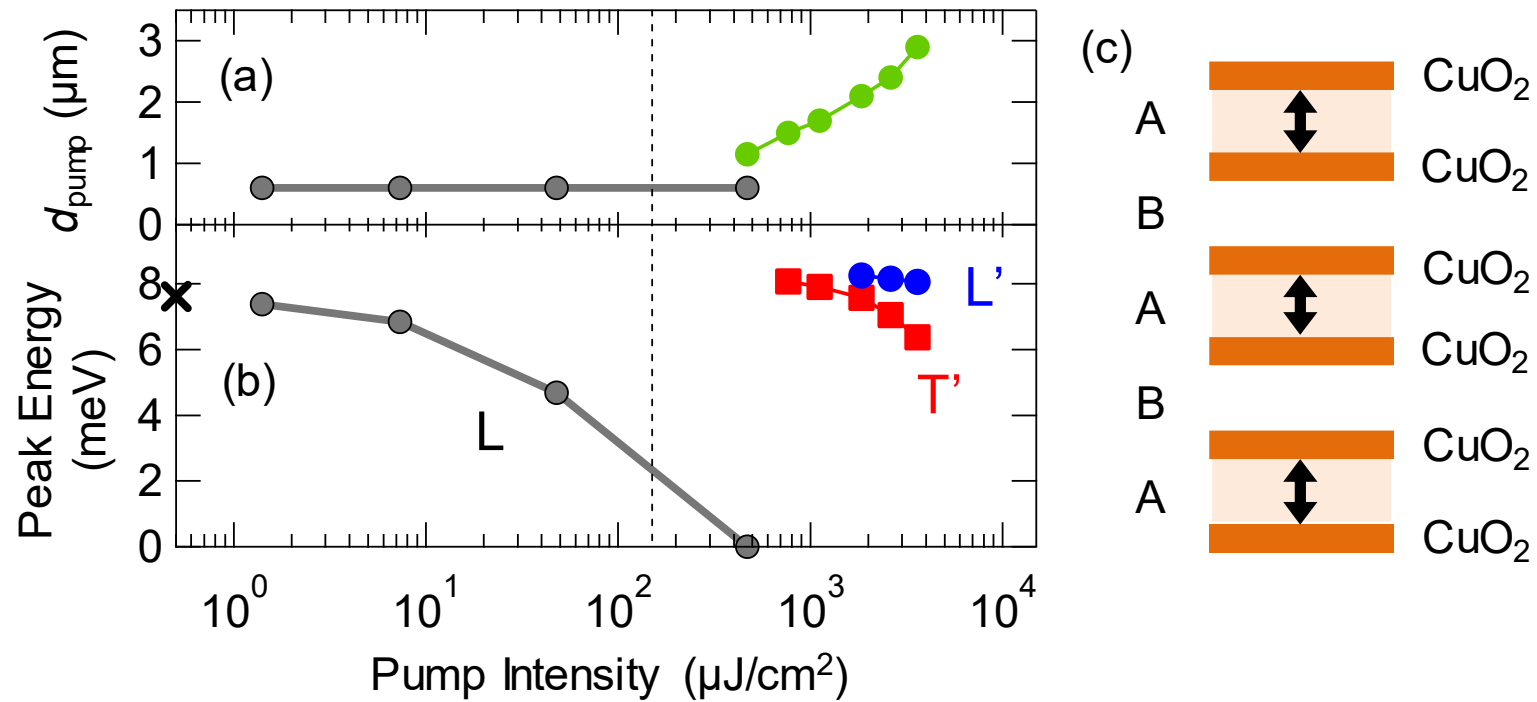
Experiments



Calculation



Pump fluence dependence of each JPR modes

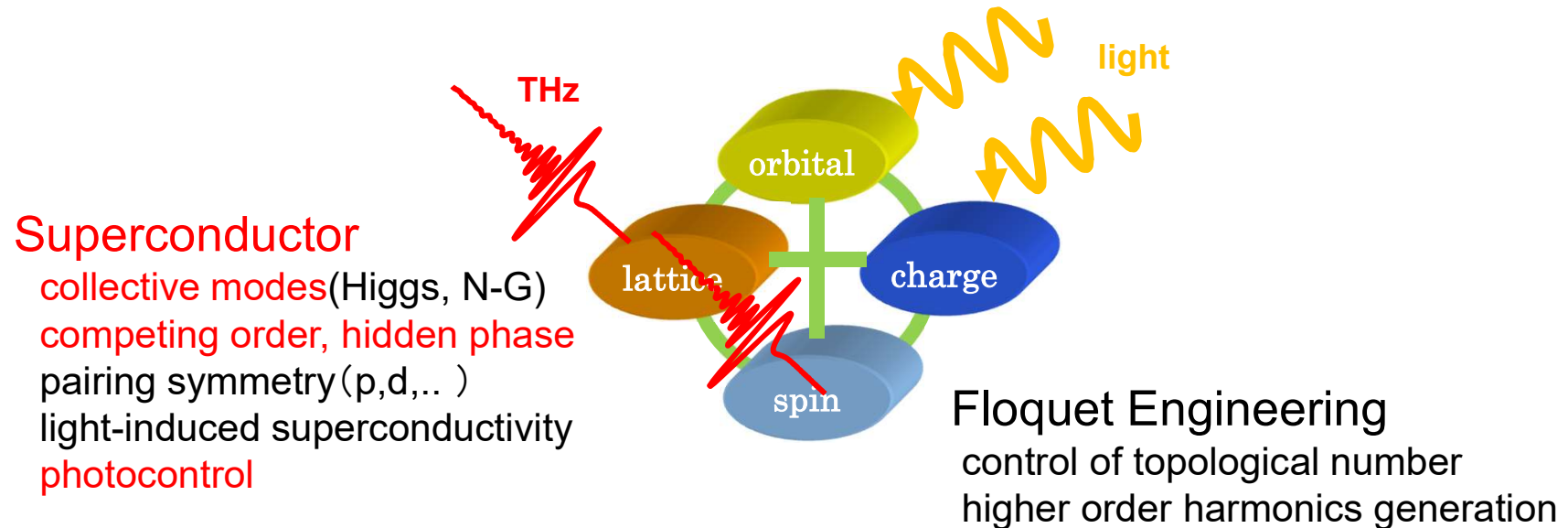


K. Tomari et al., arXiv:1712.05086

H. Niwa, poster presentation

Towards light-control of quantum material

Nonequilibrium



Ultrafast control of multiferroics
electromagnon, skyrmion

Light-control of ferroelectricity

Light-control of magnetism

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AIST

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H. Eisaki