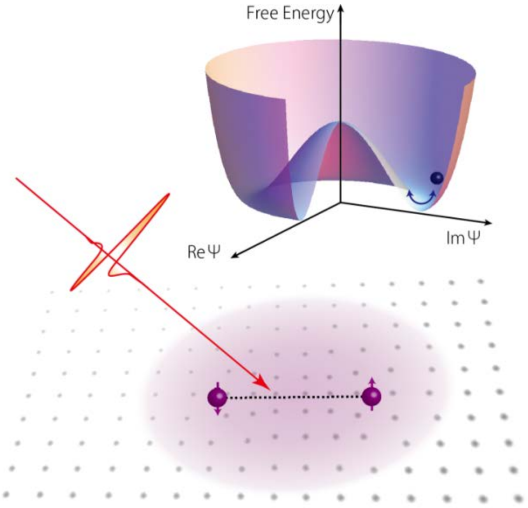
Observation of Higgs mode in s-wave superconductors

Ryo Shimano

Cryogenic Research Center,
Department of Physics,
The University of Tokyo



東京大学
THE UNIVERSITY OF TOKYO

Outline

(1) Higgs mode in superconductors: previous works

(2) Observation of Higgs mode in NbN
by THz pump and THz probe experiments:
non-adiabatic excitation with monocycle THz pulse

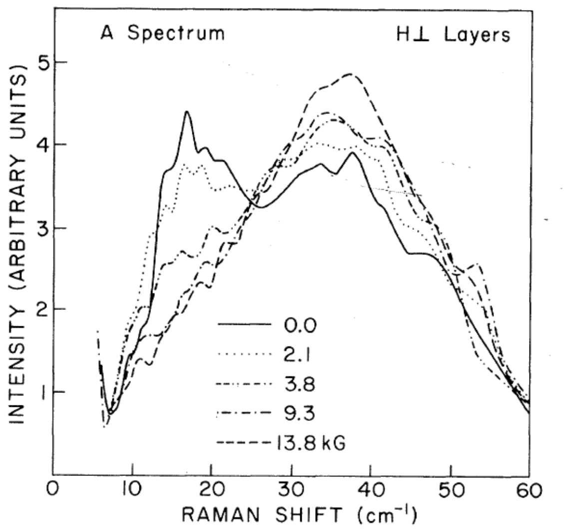
(3) Coherent order parameter oscillation in the presence of
multi-cycle THz pulse tuned below the superconducting gap:
coupling of Higgs mode with two-photon process.

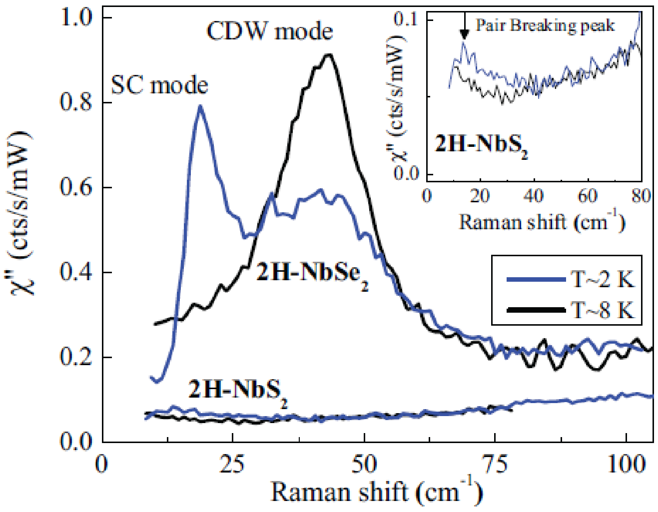
(4) Higher order harmonics generation from the collective
precession of Anderson's pseudospins

Higgs mode in superconductors : previous works

BCS-CDW coexistent compound NbSe$_2$

R. Sooryakumar and M. V. Klein, PRL **45**, 660 (1980).
P.B. Littlewood and C. M. Varma, PRL **47**, 811 (1982).
C. M. Varma, J. Low Temp. Phys.**126**, 901 (2002).





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

Cf.) *p*-wave superfluid ^{3}He

For a review, e.g., Lee, J. Phys. Chem. Sol. **59**, 1682 (1998).
G. E. Volovik, and M. A. Zubkov, J. Low Temp. Phys.**175**, 486 (2014)

Theoretical investigations: quench problem

Quenching the interaction U(t) much faster than

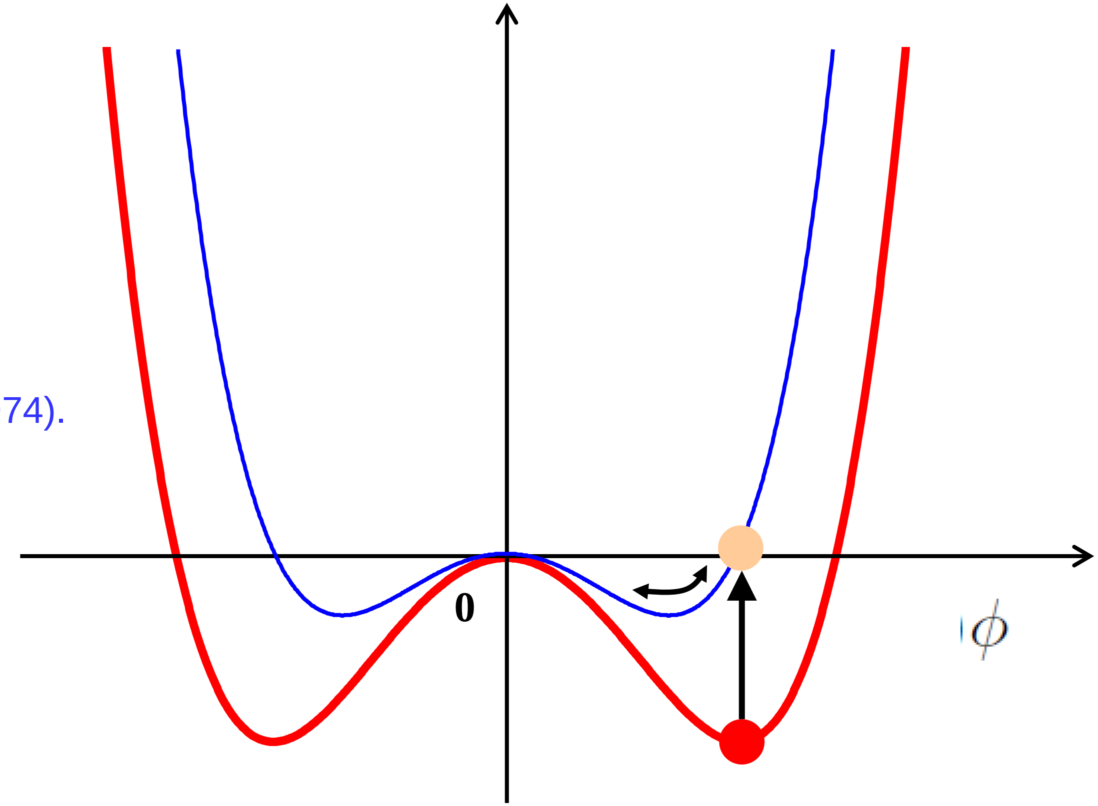
$\tau_{\Delta} \sim \hbar/\Delta$ （Δ:order parameter）

⇨ Emergence of order parameter oscillation (Higgs mode)

Theoretical studies for

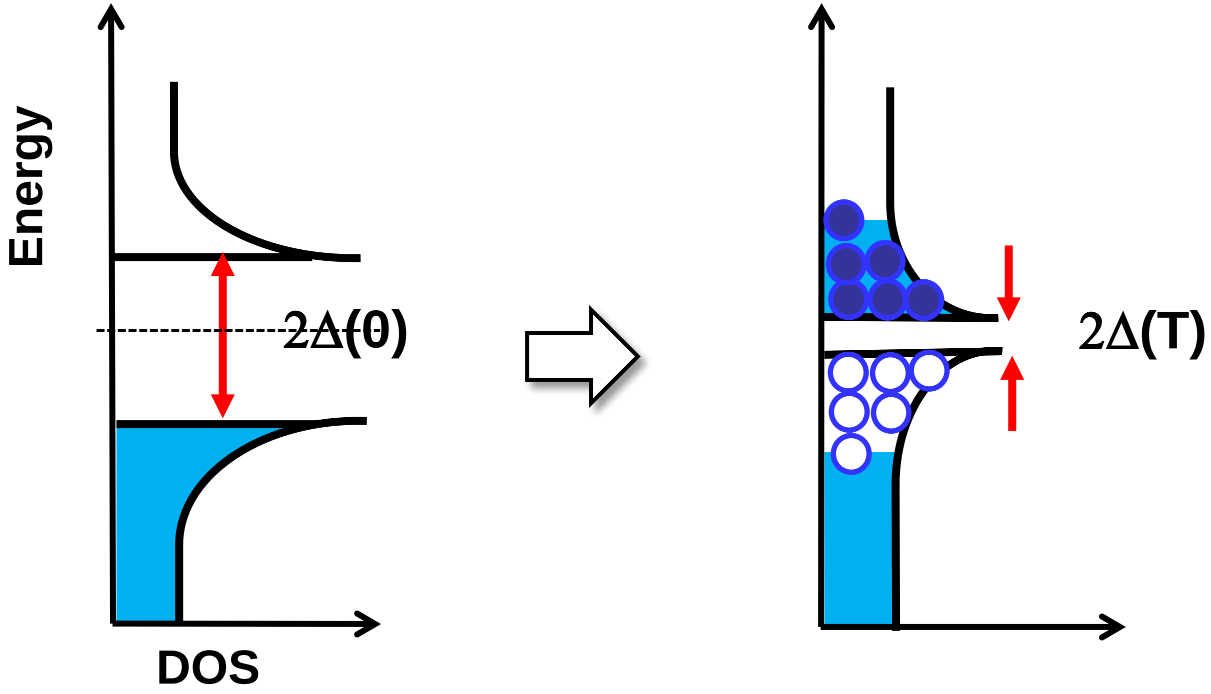
dynamics of nonequilibrium BCS state after *nonadiabatic* excitation

- Volkov *et al*., Sov. Phys. JETP **38**, 1018 (1974).
- Barankov *et al*., PRL **94**, 160401 (2004).
- Barankov *et al*., PRL **96**, 230403 (2006).
- Yuzbashyan *et al*., PRL **96**, 230404 (2006).
- Gurarie *et al*., PRL **103**, 075301 (2009).
- Tsuji *et al*., PRL **110**, 136404 (2013).

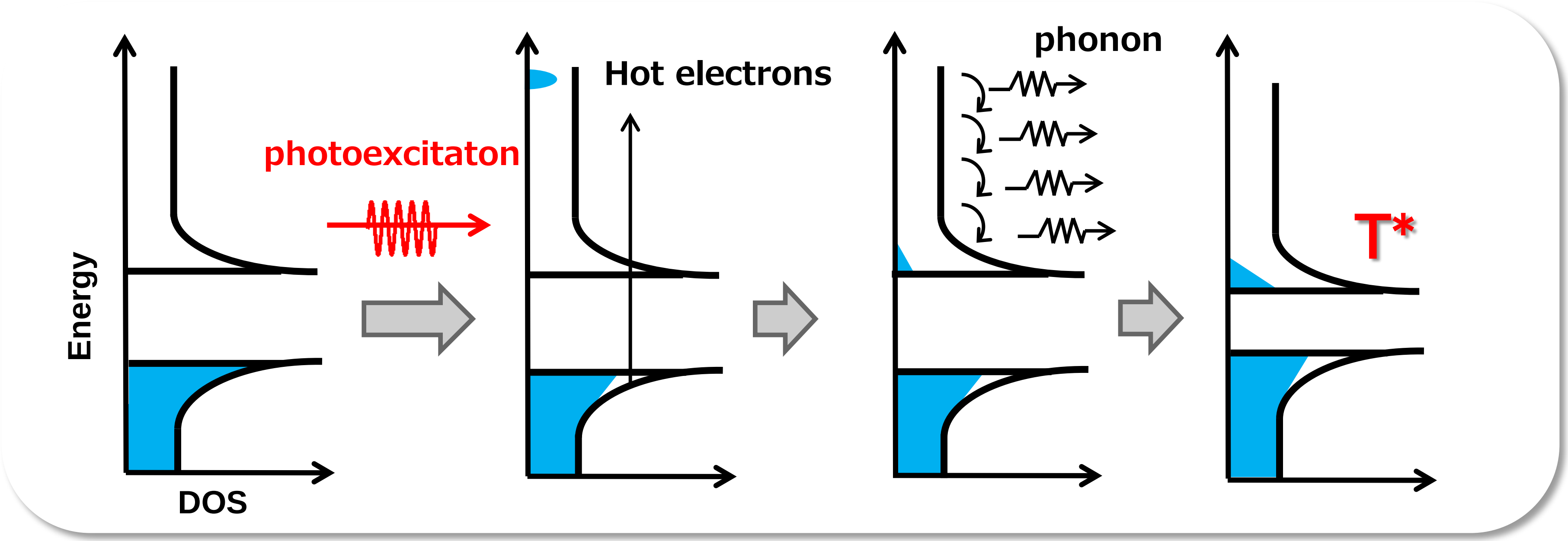


Quench by Quasiparticle injection

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



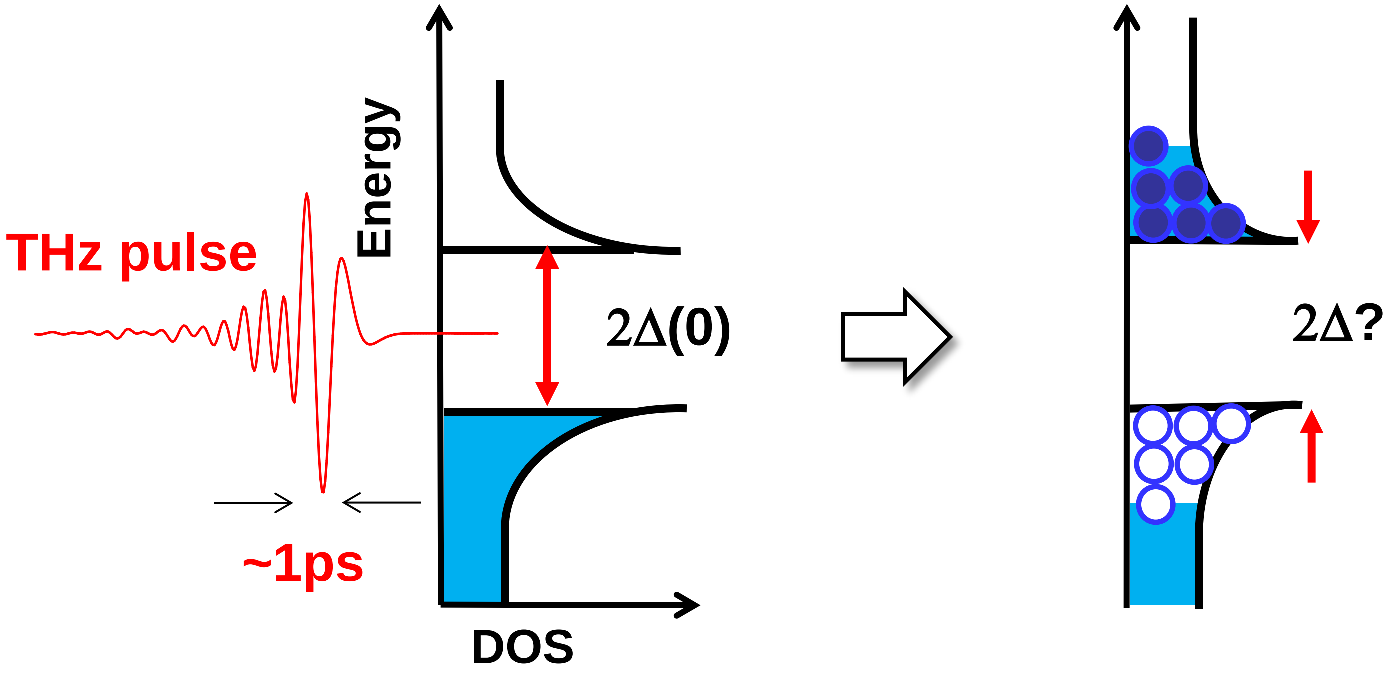
Gap quenching dynamics after photoexcitation



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

①photoexcitation of hot electrons far above the SC gap
②phonon emission
③pair breaking by emitted phonons
④gradual reduction of the SC gap

What happens if one create quasiparticle instantaneously, $\tau < \Delta^{-1}$

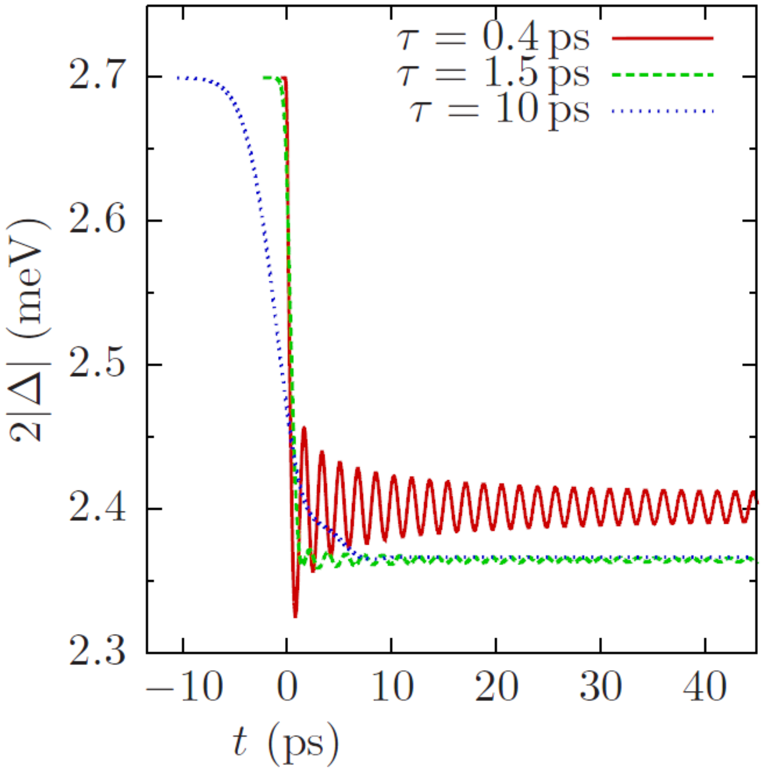


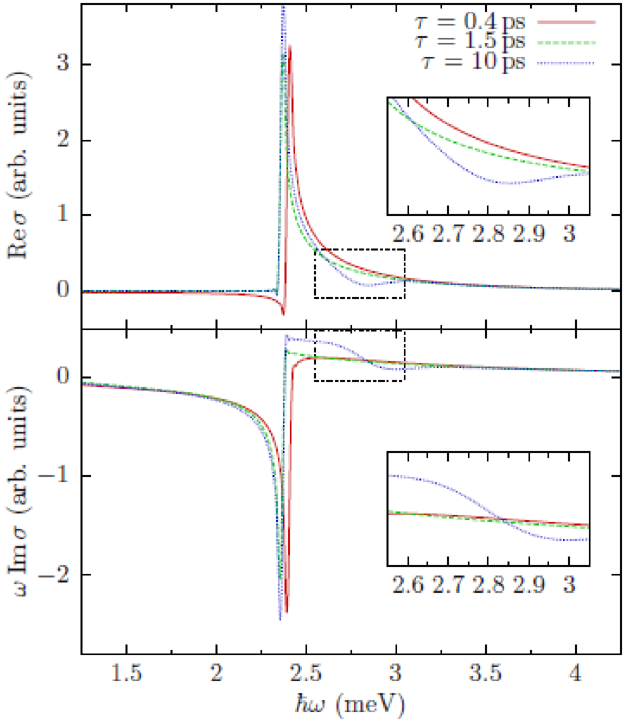
$2\Delta(0)$ in typical superconductors: ~1THz

We need an intense monocycle-like THz pulse, avoiding the injection of excess energy to the system that cause the phonon-induced pair breaking.

Non-adiabatic excitation by light pulse

Papenkort *et al*., PRB 76, 224522 (2007).





Invisible in $\sigma(\omega)$ spectra?

See also, A. P. Schnyder, D. Manske, and A. Avella, Phys. Rev. B84, 214513 (2011)

Sample and pump pulse

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

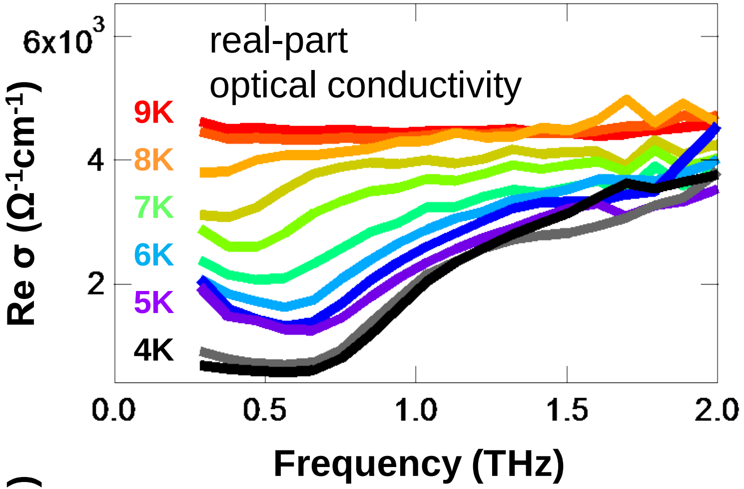
THz pump pulse

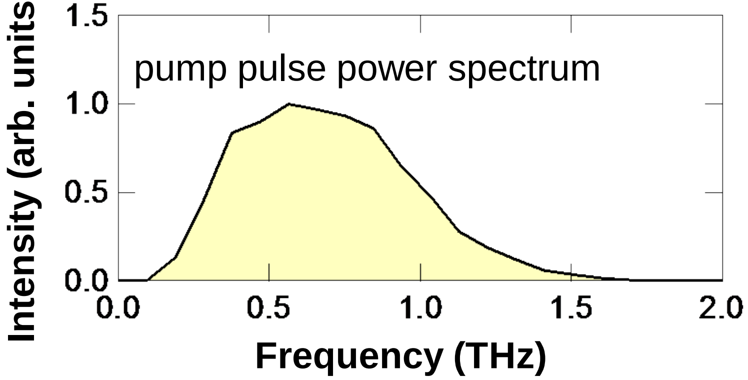
Center frequency 0.7THz～2Δ

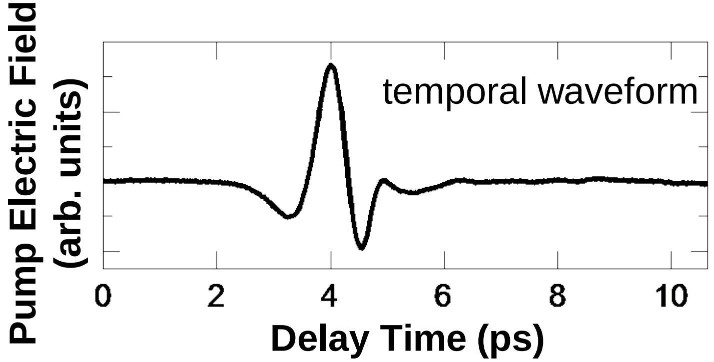
pulse width: $\tau_{pump} \sim 1.5$ ps

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

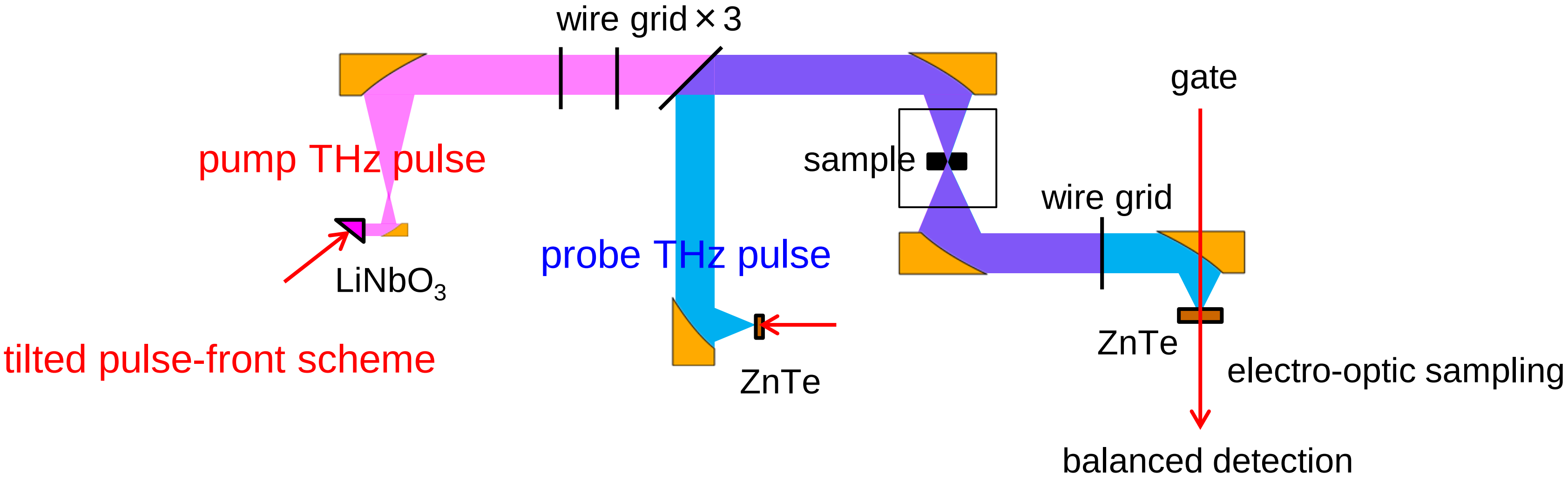
nonadiabatic excitation condition

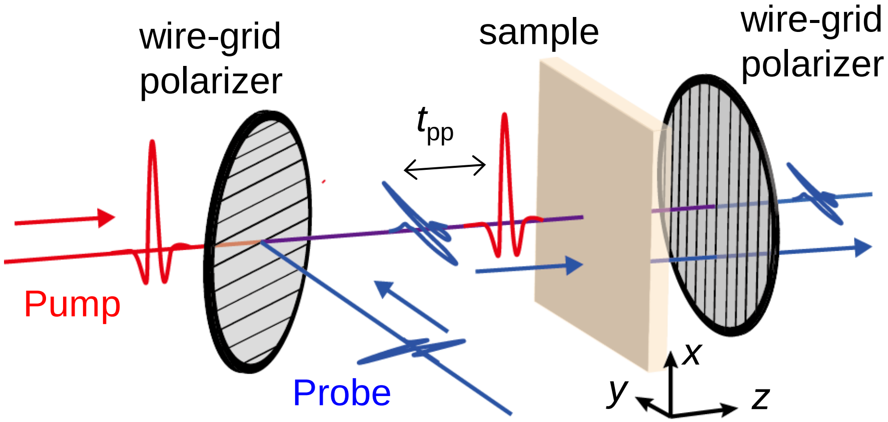






THz pump and THz probe spectroscopy





Pump : $E_{pump}//x$

Probe : $E_{probe}//y$

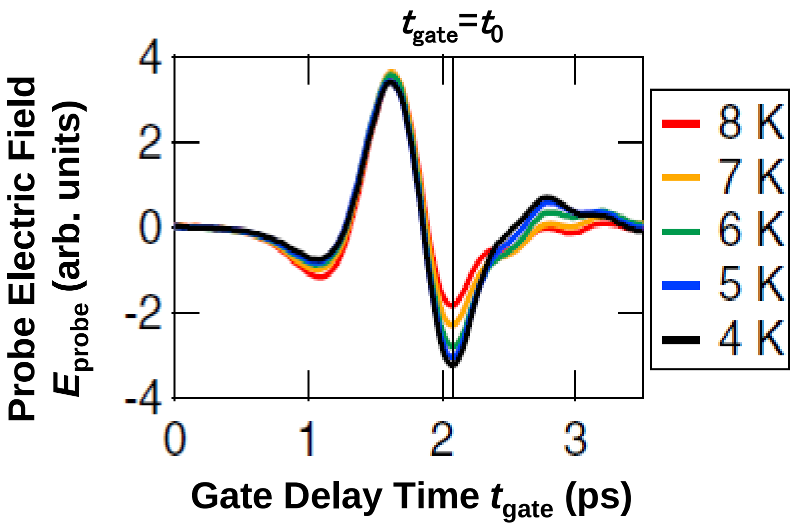
t_{pp}: pump-probe delay

Transmitted probe THz electric field:

Free space EO sampling

t_{gate}: gate pulse delay

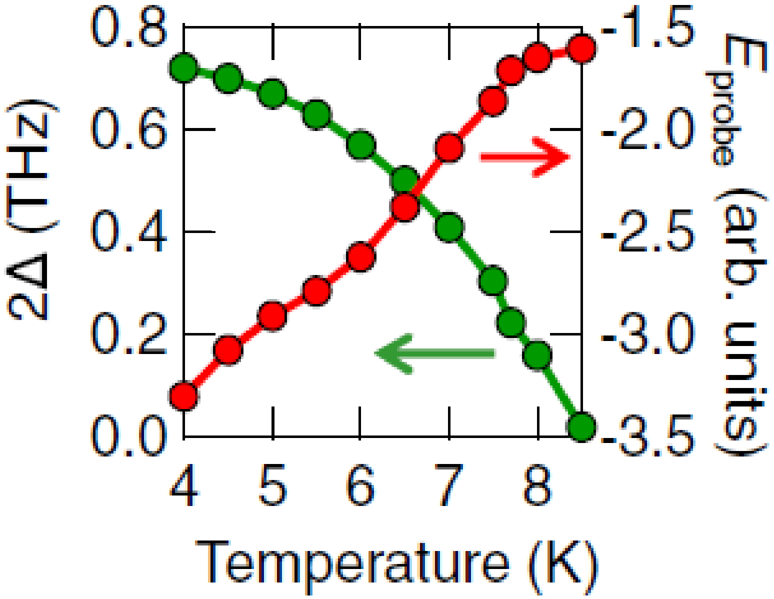
Detection scheme of the Higgs mode



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

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

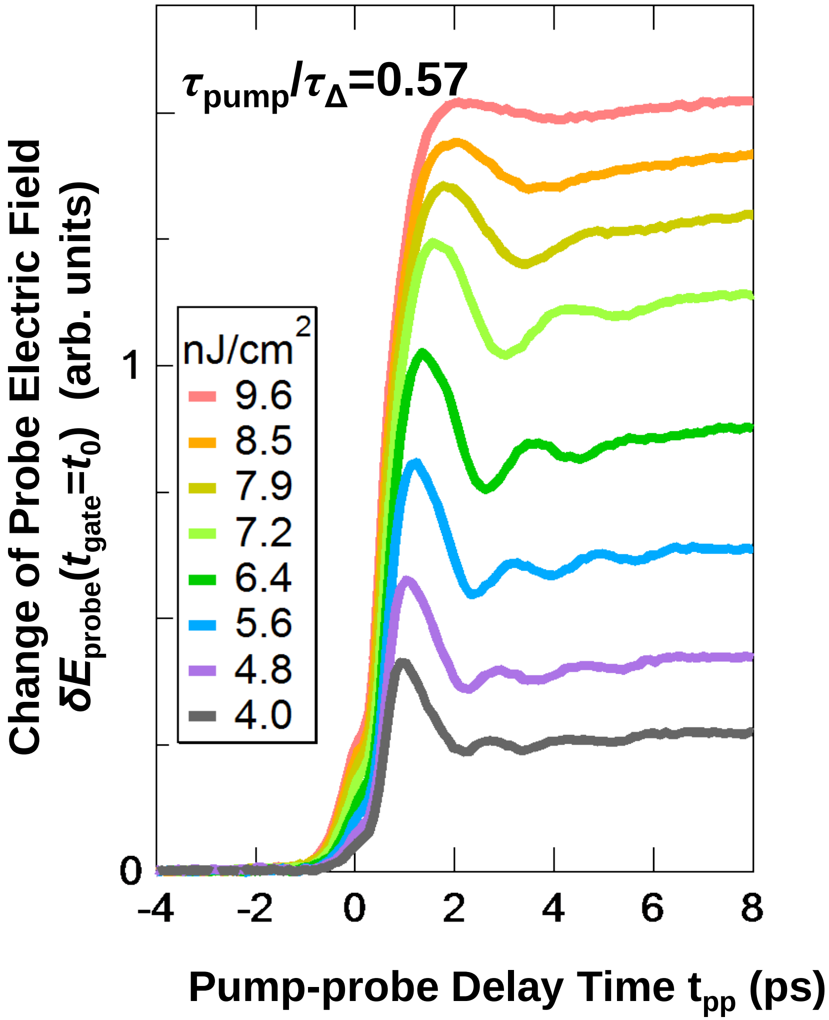
We fixed the gate delay at $t_{gate}=t_0$ and measure the pump-probe delay dependence



Dynamics after the THz pump pulse

Pump-induced change in the probe E-field

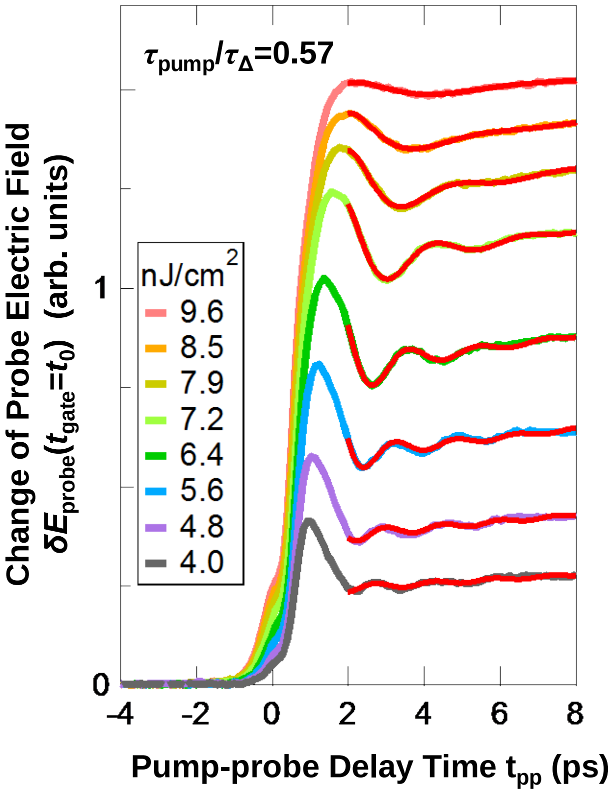
$\delta E_{\mathrm{probe}}(t_{\mathrm{gate}}=t_0)$



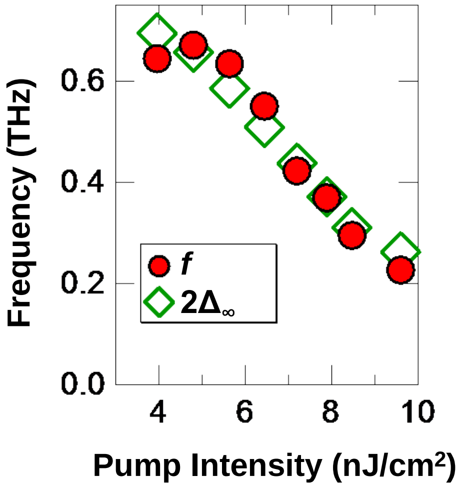
THz Pump irradiatin (t_{pp}>0)
⇒ rapid increase in E_{probe}
⇔ reduction of Δdue to the quasipaticle excitation

A clear damped oscillation is observed

Oscillation frequency

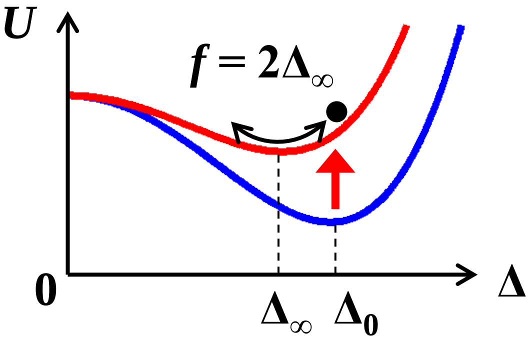


$$\delta E_{\mathrm{probe}}(t_{\mathrm{pp}}) = C_1 + C_2 t_{\mathrm{pp}} + \frac{a}{(t_{\mathrm{pp}})^b}\cos(2\pi f t_{\mathrm{pp}} + \phi)$$



$2\Delta_\infty$:
asymptotic value of
the order parameter
estimated at t_{pp}=8ps

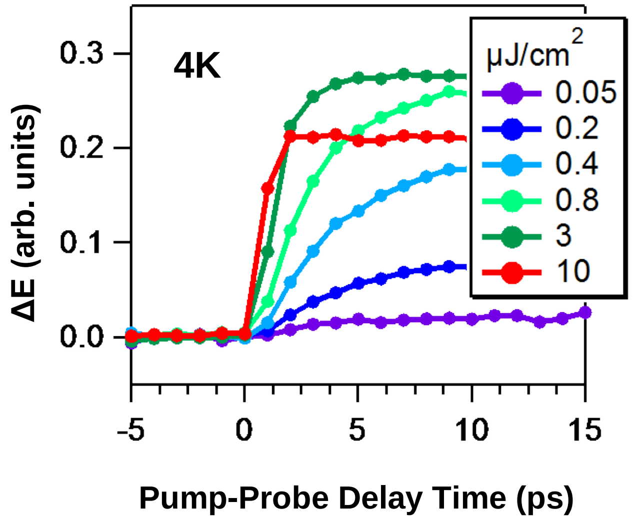
oscillation frequency $f = 2\Delta_\infty$



characteristic to the Higgs mode behavior

R. Matsunaga, Y. Hamada, R. Shimano *et al*.,
PRL **111**, 057002 (2013).

Optical pump vs THz pump



Optical Pump（800nm, 1.5eV 90fs）

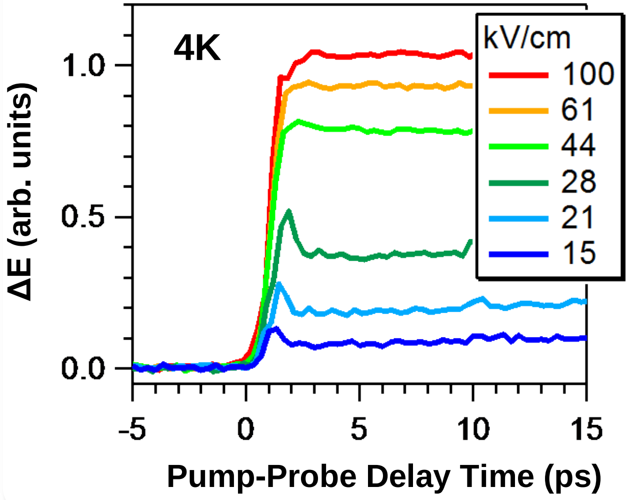
Slow rise , dependent of pump intensity

Photoexcitation of hot electron

⇒ phonon emission

⇒ pair breaking

⇒ reduction of order parameter



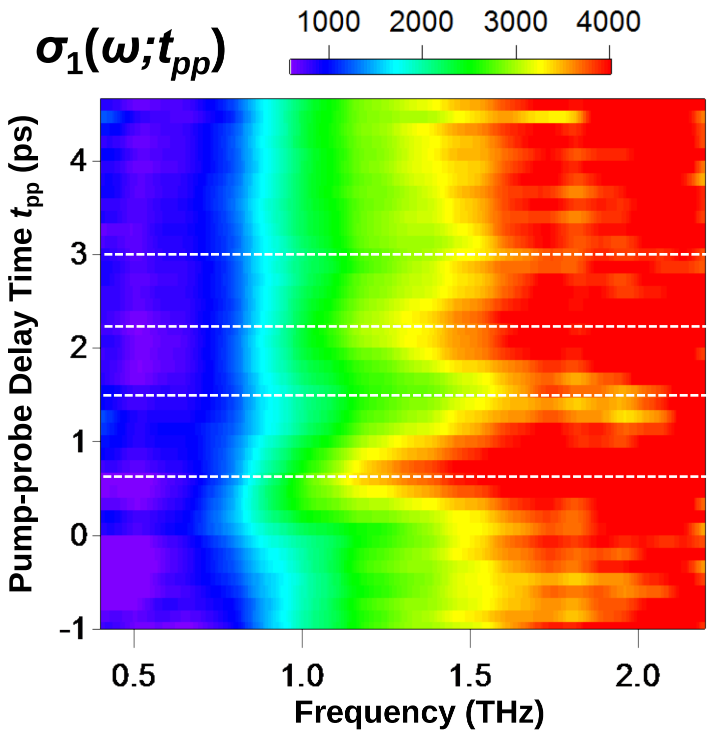
THz pump

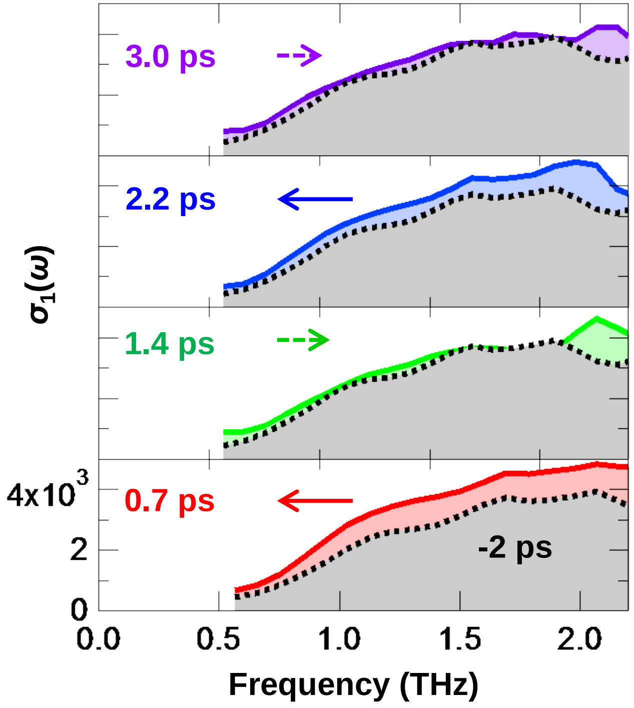
Fast rise, independent of pump intensity

Direct excitation of quasiparticle near the gap

R. Matsunaga and R. Shimano, PRL **109**, 187002 (2012).

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



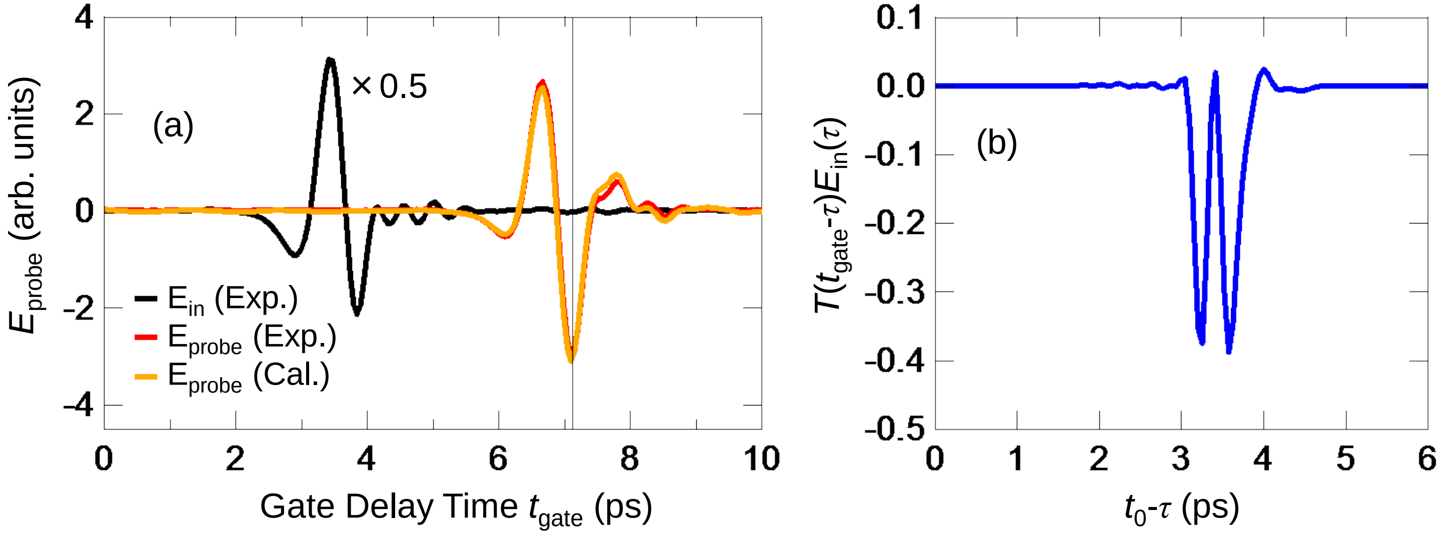


Temporal oscillation in the optical conductivity spectrum near the gap frequency

Why the Higgs mode oscillation $(2\Delta)^{-1}$ is not smeared out in the probe transmittance?

$$T(\omega;\Delta)=\frac{1}{1+n_{\mathrm{sub}}+Z_0\,d\,\sigma(\omega;\Delta)}\frac{4n_{\mathrm{sub}}e^{i\Phi(\omega)}}{1+n_{\mathrm{sub}}}$$

$$E_{\mathrm{probe}}\left(t_{\mathrm{gate}}\right)=\int_{-\infty}^{t_{\mathrm{gate}}}T\left(t_{\mathrm{gate}}-\tau;\Delta\right)E_{\mathrm{in}}\left(\tau\right)d\tau$$



Samples with larger gaps

Nb$_{0.8}$Ti$_{0.2}$N
12nm / Quartz

$2\Delta_0$=0.72 THz

τ_{pump}/τ_Δ=0.57

Nb$_{0.8}$Ti$_{0.2}$N
30nm / Quartz

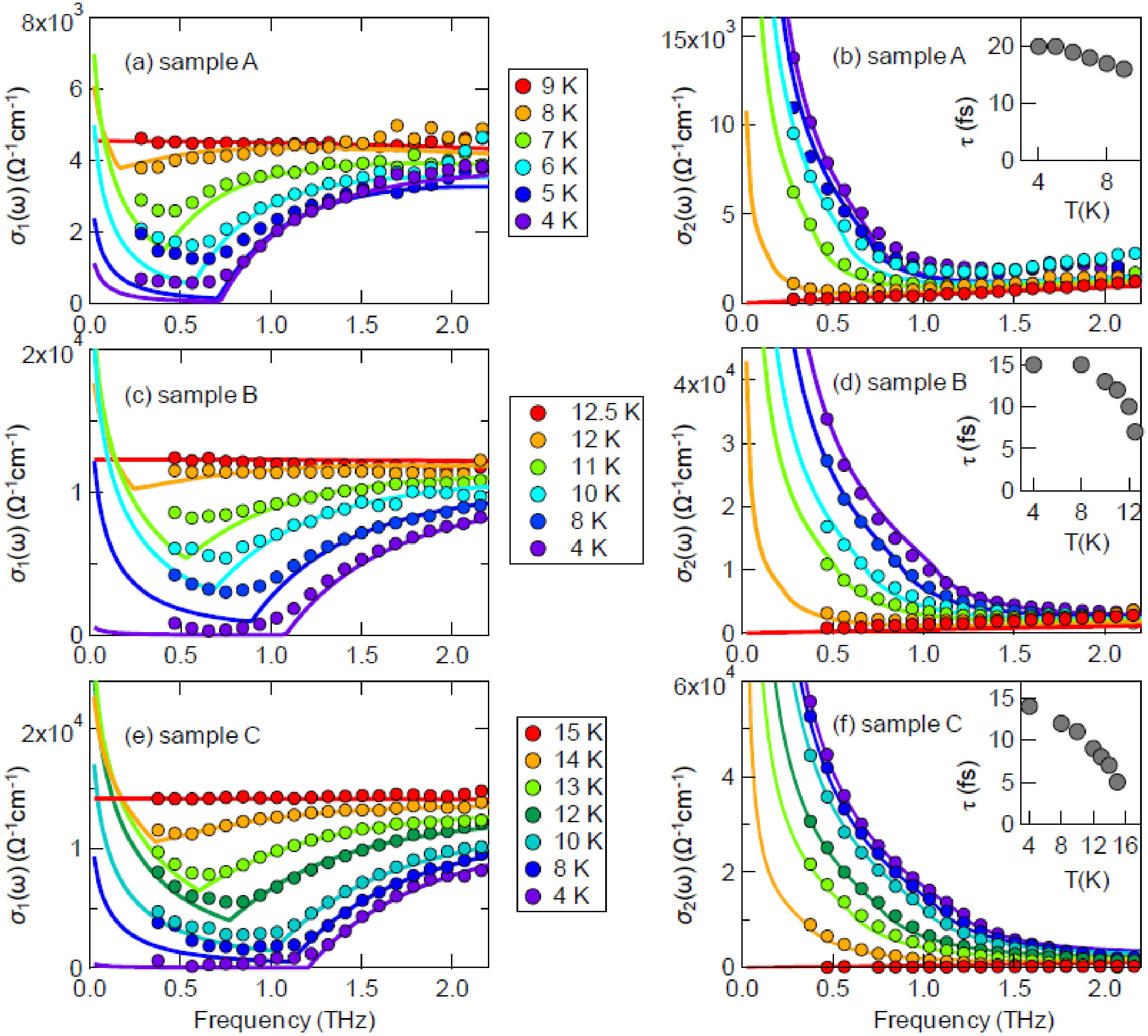
$2\Delta_0$=1.1 THz

τ_{pump}/τ_Δ=0.81

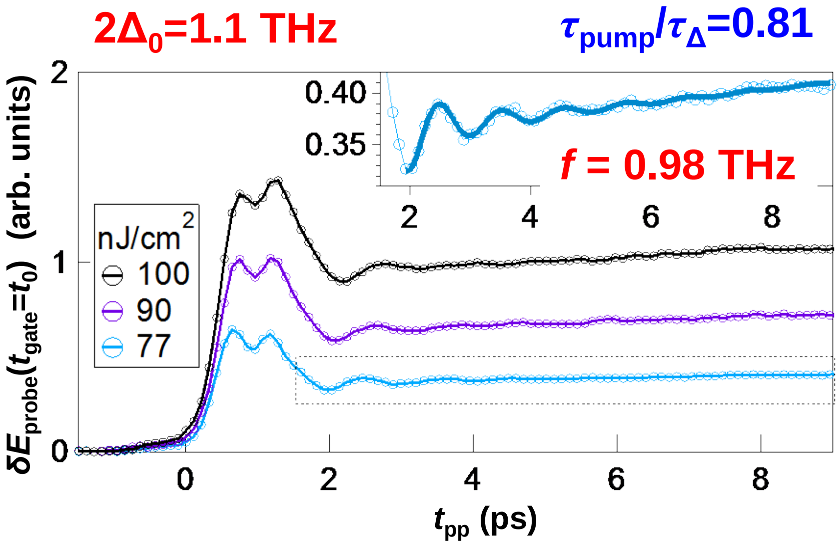
NbN
24nm / MgO

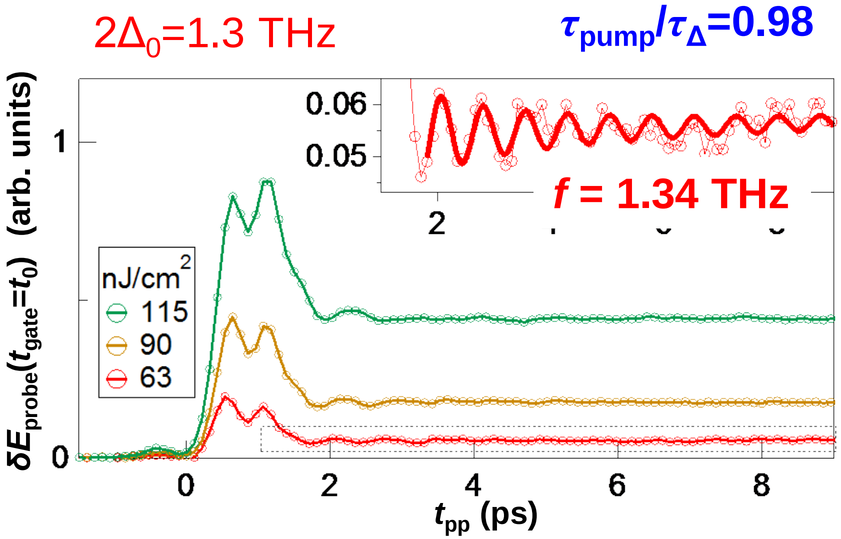
$2\Delta_0$=1.3 THz

τ_{pump}/τ_Δ=0.98



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



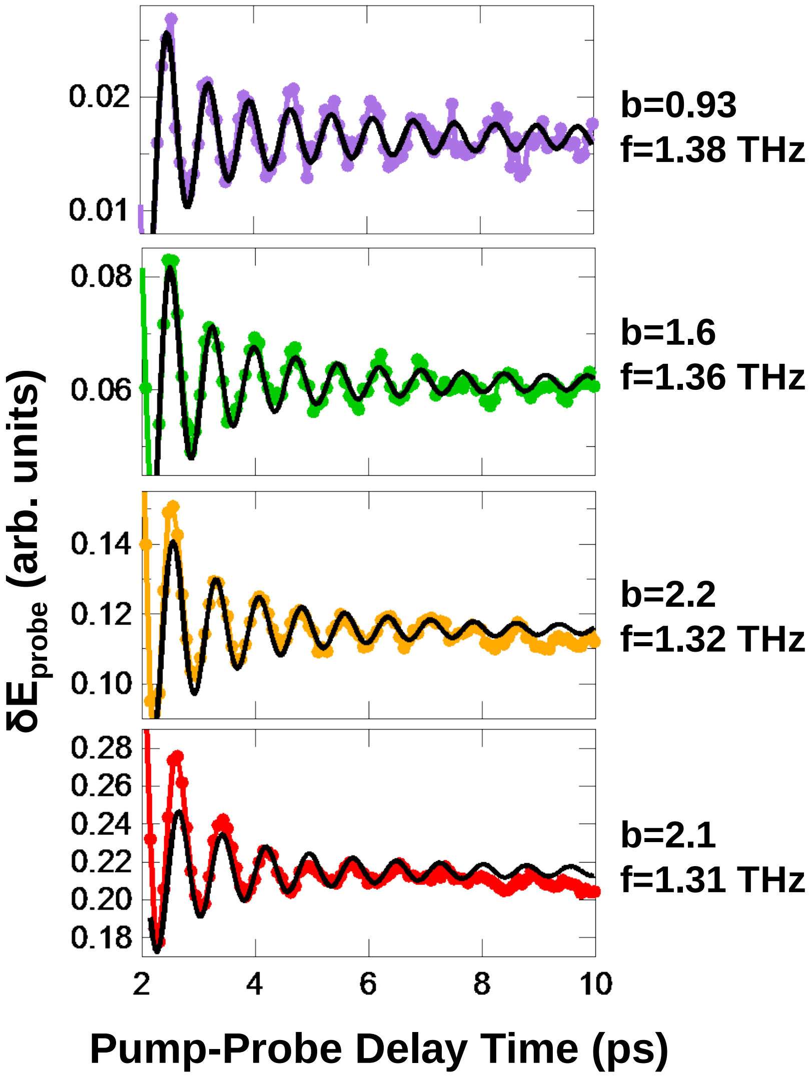


The Higgs mode is identified, although it is less prominent than the previous sample.

The oscillation frequency perfectly matches with the gap energy.

Decay dynamics of Higgs mode

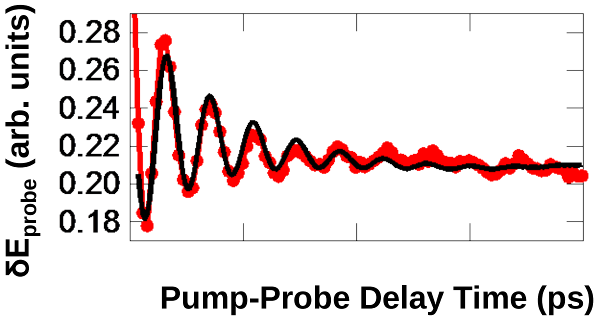
Weak
excitation



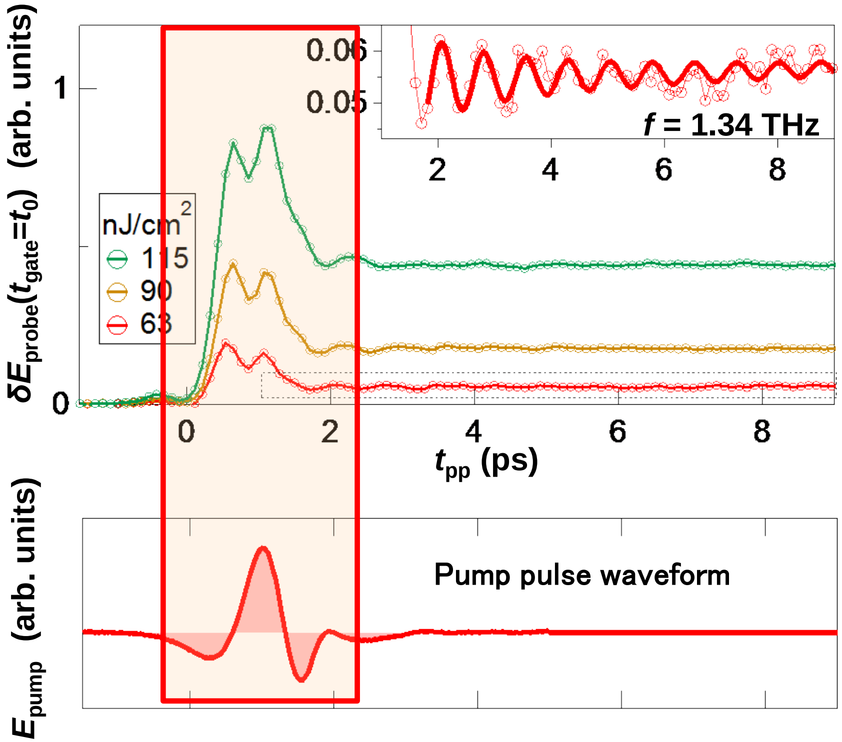
Strong
excitation

Power law decay

Exponential decay



Dynamics in the coherent excitation regime

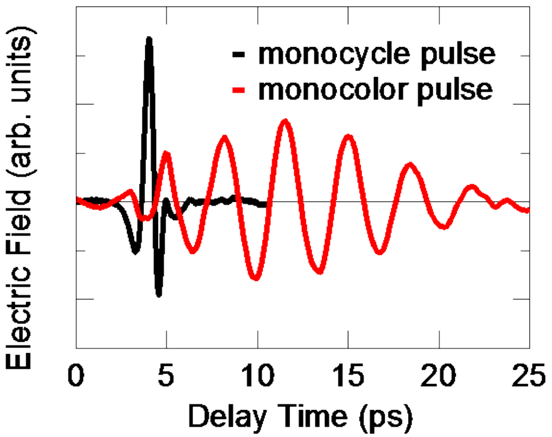


What is happening during the irradiation of coherent pump field?

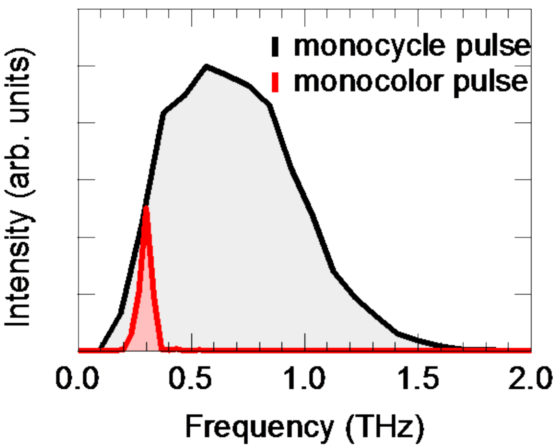
Quasi-Monochromatic Excitation Experiments (Coherent Excitation Regime)

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

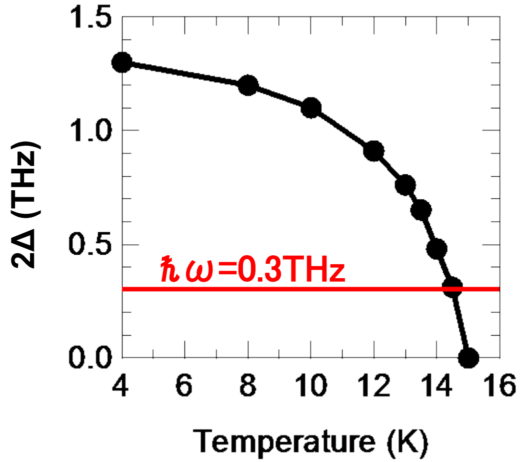
E-field waveform



Power Spectrum



Photon energy vs BCS gap

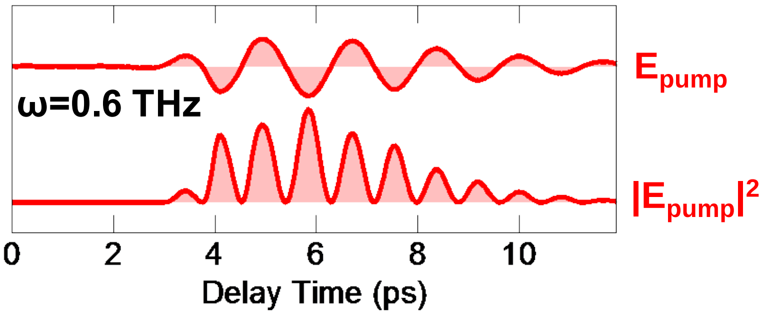


How does the BCS ground state respond to the strong electromagnetic field with $\hbar\omega < 2\Delta$?

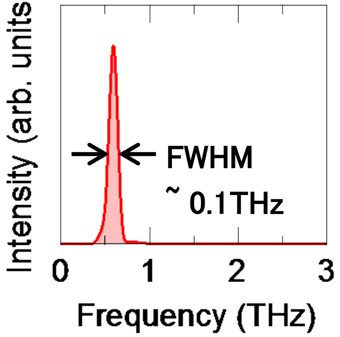
Coherent Excitation Regime Experiments

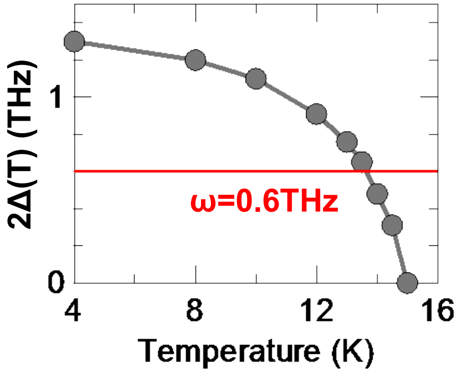
ω=0.6THz

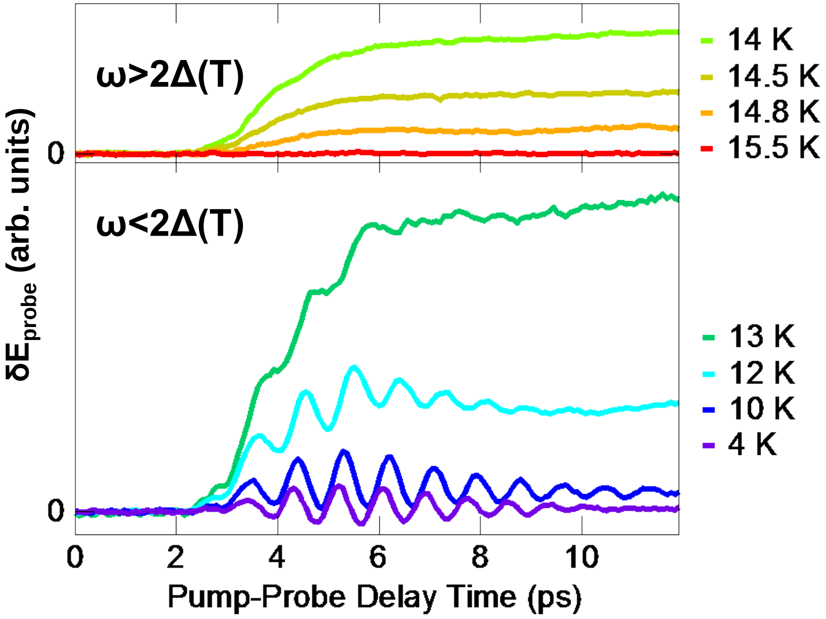
3.5 kV/cm@peak



Pump power spectru,







In case of $\hbar\omega > 2\Delta(T)$

Reduction of order parameter due to QPs excitation

In case of $\hbar\omega < 2\Delta(T)$

2ω coherent oscillation of order parameter driven by the AC field of ω?

Anderson’s pseudospin ($\boldsymbol{\sigma}_{\mathbf{k}}$) representation

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

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

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

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

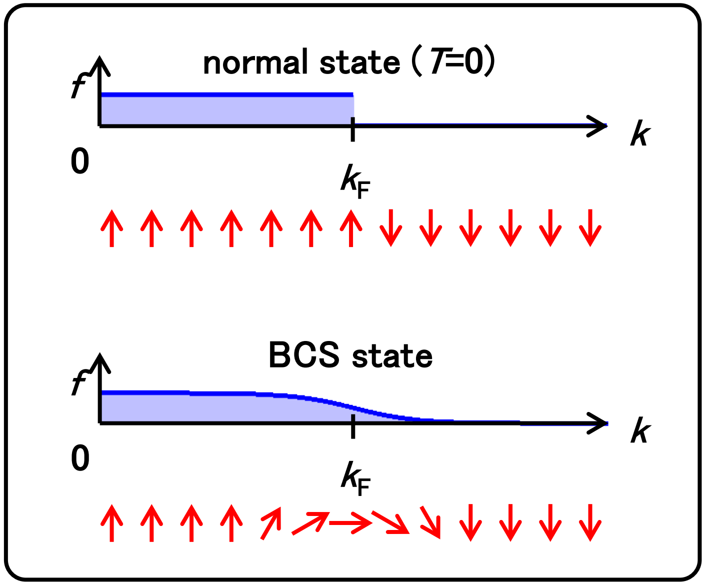
$$\mathcal{H}^{\mathrm{BCS}} = \sum_{k} \boldsymbol{b}_{k}^{\mathrm{eff}} \cdot \boldsymbol{\sigma}_{k}$$

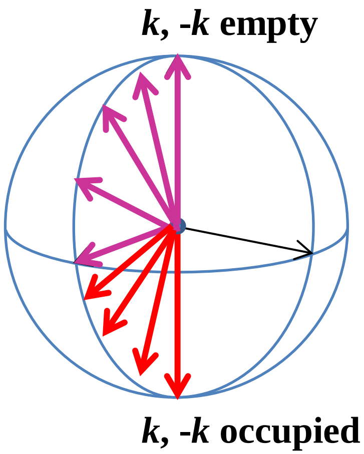
$$\boldsymbol{b}_{k}^{\mathrm{eff}} = (-\Delta', \ -\Delta'', \ \varepsilon_{k})$$

: effective magnetic field for *k*

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

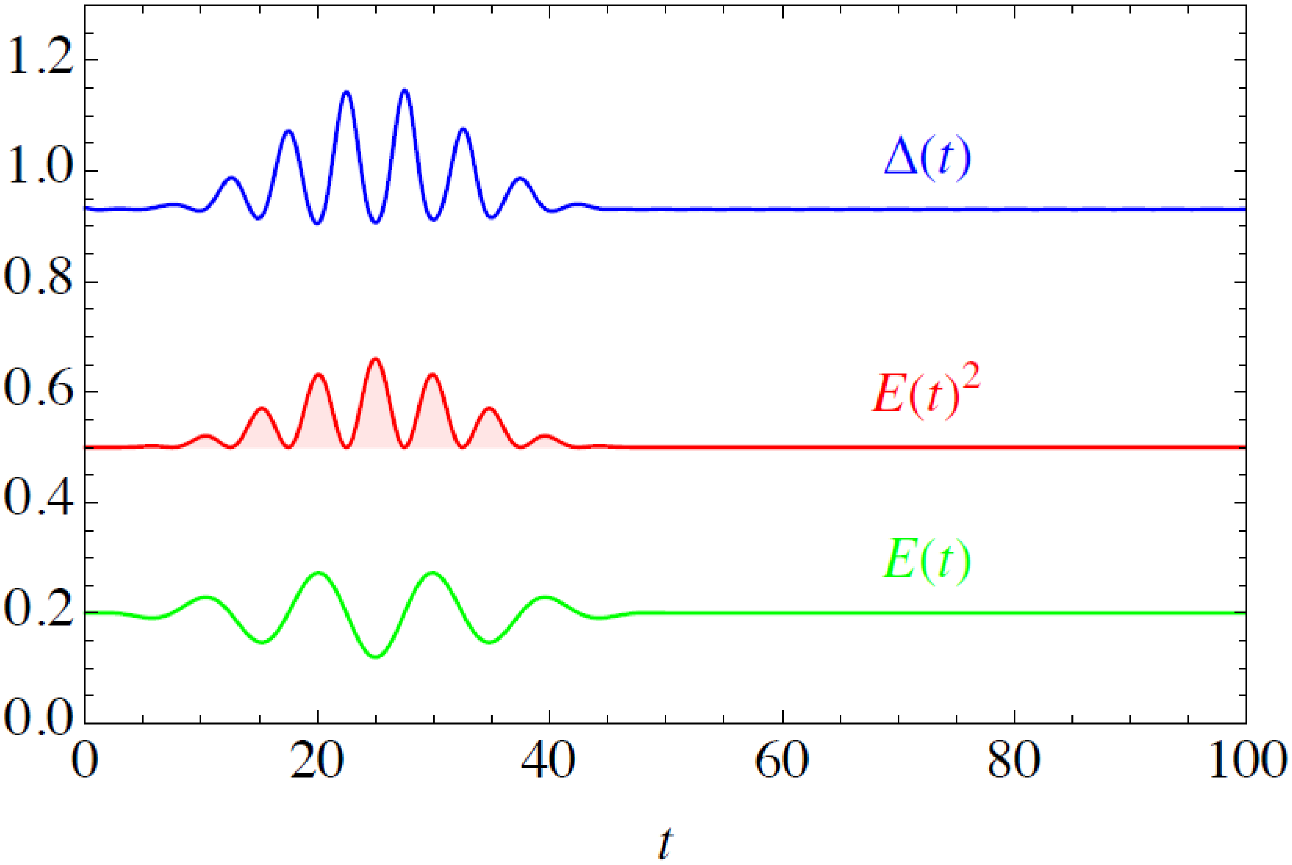
$$\boxed{\frac{d}{dt}\boldsymbol{\sigma}_{k} = i[\mathcal{H}^{\mathrm{BCS}}, \boldsymbol{\sigma}_{k}] = 2\boldsymbol{b}_{k}^{\mathrm{eff}} \times \boldsymbol{\sigma}_{k}}$$





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

Pseudospin dynamics : simulation with BdG equation



$U = 3,\ \beta = 12,\ A = 0.2,$

$\Omega = 0.628,\ 2\Delta(0) = 1.87$

Summary

(1) We have observed the Higgs amplitude mode in s-wave superconductors NbTiN in a non-adiabatic excitation regime with a monocycle THz pulse.

R. Matsunaga, Y. Hamada, R. Shimano *et al*., PRL **111**, 057002 (2013).

R. Matsunaga and R. Shimano, PRL **109**, 187002 (2012).

(2) Coherent 2ω oscillation of the order parameter was observed during the below-gap irradiation of multicycle THz pulse, which is well described by the precession of Anderson pseudospin.

(3) Strong THG is observed at $2\omega=2\Delta$
(two-photon resonance of the Higgs mode).

R. Matsunaga et al., to be published in Science

Collaborators

Experiment, Analysis & Simulation

- R. Matsunaga
- Y. I. Hamada
- A. Sugioka
- H. Fujita

Theory

- N. Tsuji
- H. Aoki

Sample Fabrication

- K. Makise
- Y. Uzawa
- H. Terai
- Z. Wang

東京大学
THE UNIVERSITY OF TOKYO

NICT
独立行政法人
情報通信研究機構