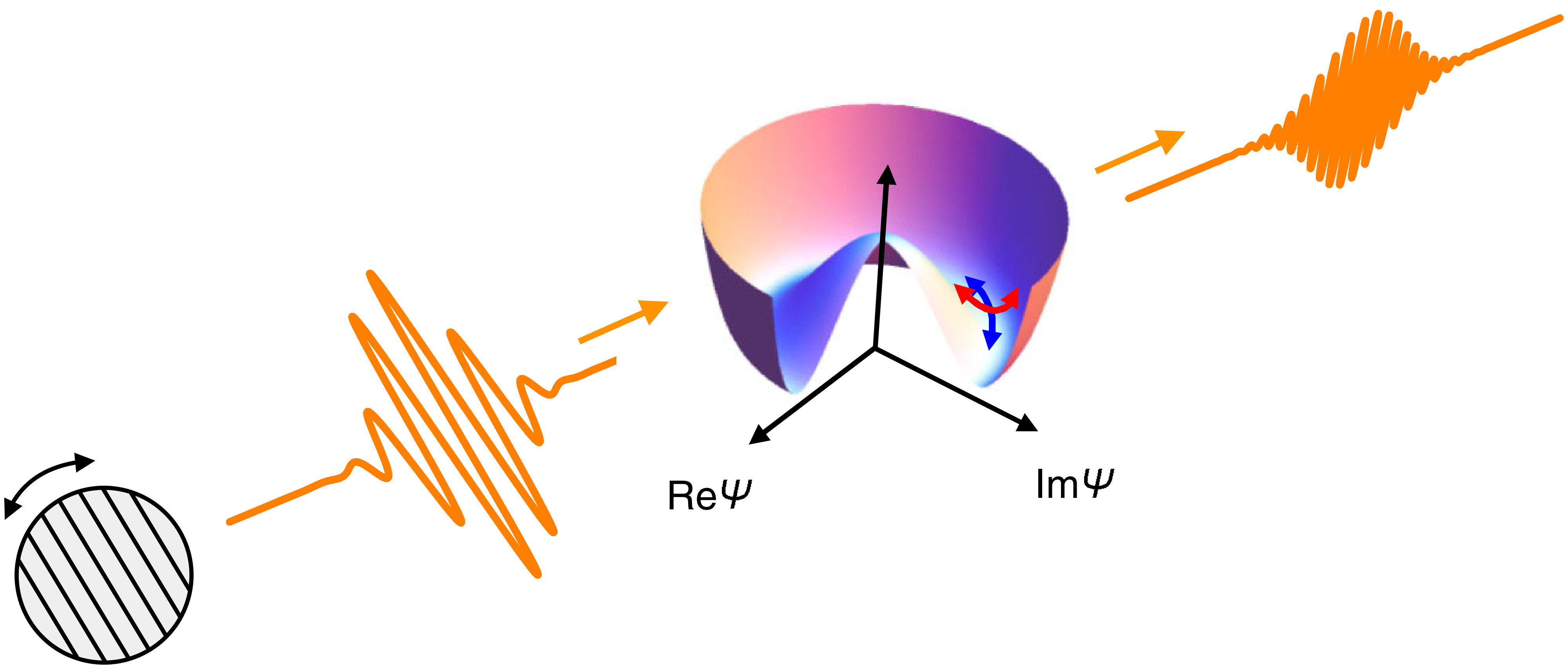
Higgs amplitude mode and charge density fluctuations in the nonlinear optical response of superconductors



Naoto Tsuji (RIKEN CEMS)

Collaborators

[Theory] Y. Murakami (Fribourg Univ.)
H. Aoki (Univ. Tokyo, AIST)

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R. Matsunaga (Univ. Tokyo)
K. Makise (NICT)
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[1] Tsuji, Murakami, Aoki, Phys. Rev. B 94, 224519 (2016).
[2] Matsunaga, Tsuji, Makise, Terai, Aoki, Shimano, arXiv:1703.02815.
(To be published in Phys. Rev. B, Rapid Communications.)

Outline of the talk

- Introduction: Higgs amplitude mode in superconductors

- Higgs mode vs charge density fluctuation (CDF)

- Experimental discrimination by polarization-resolved THG

- A mechanism of enhancing the light-Higgs coupling
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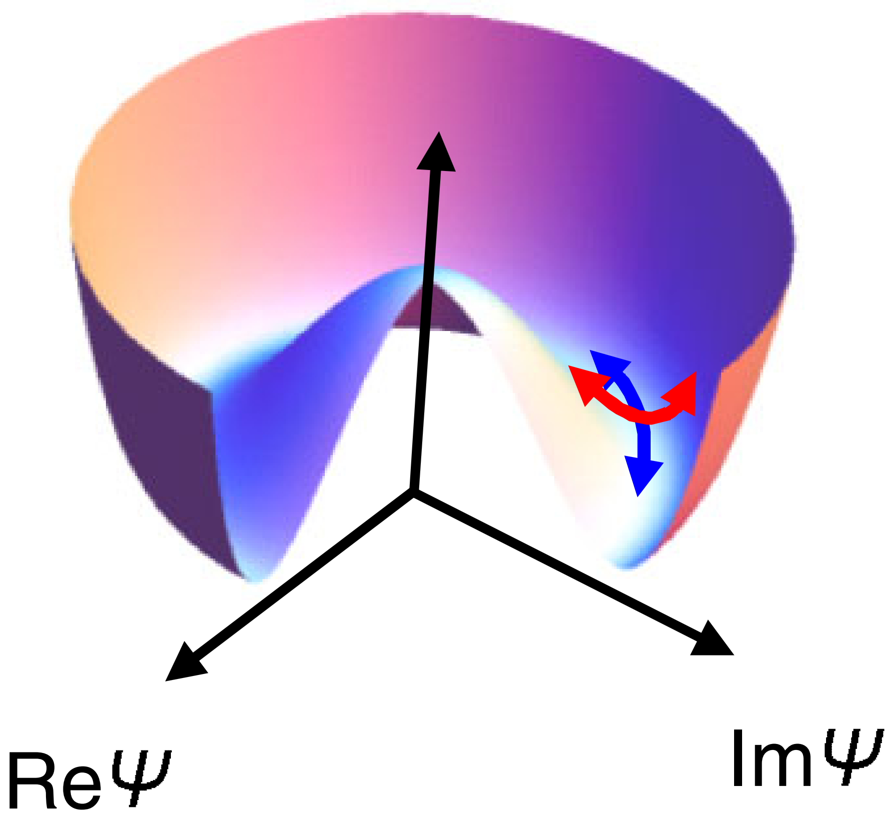
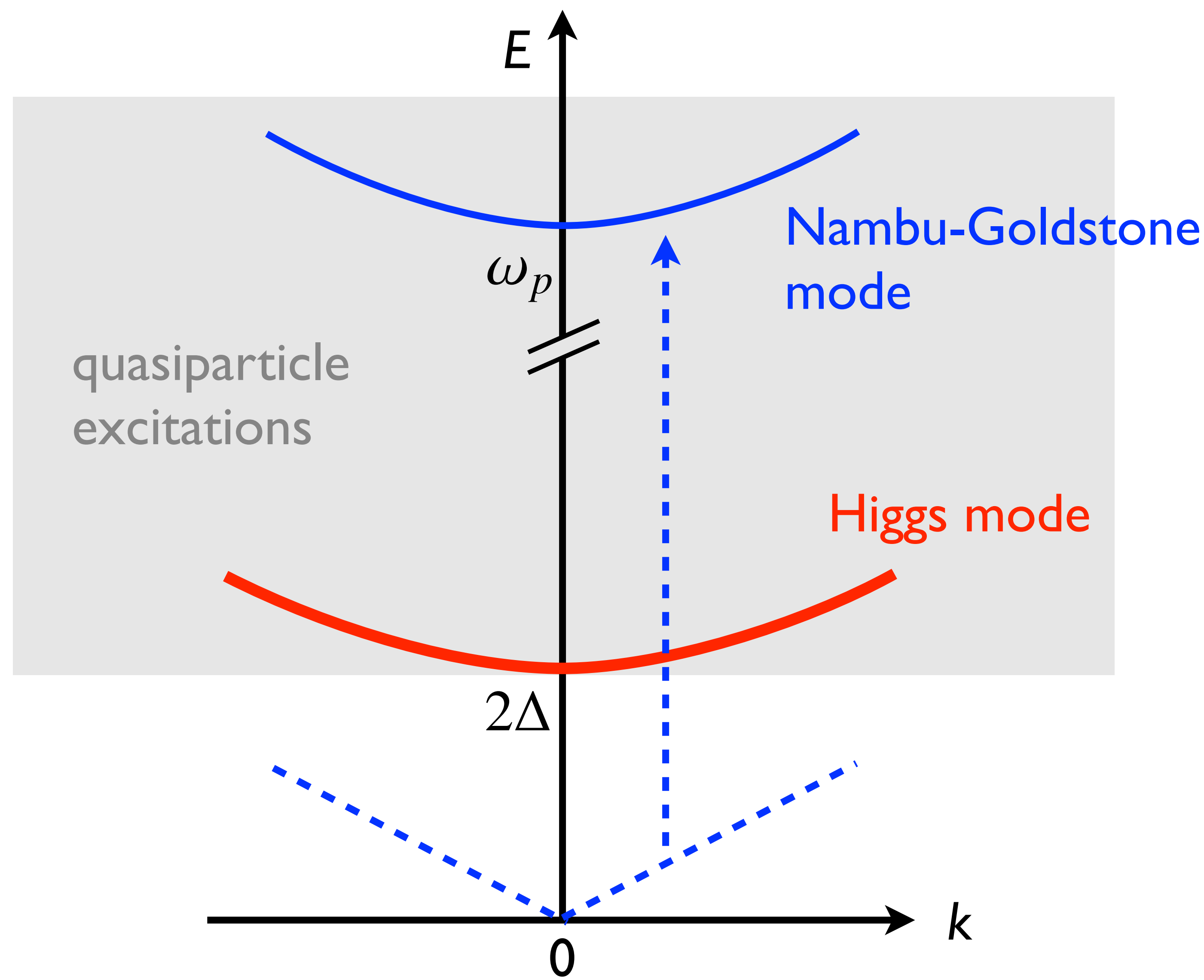
Collective modes in superconductors

Anderson (1958), Volkov, Kogan (1973), Littlewood, Varma (1981, 1982), …



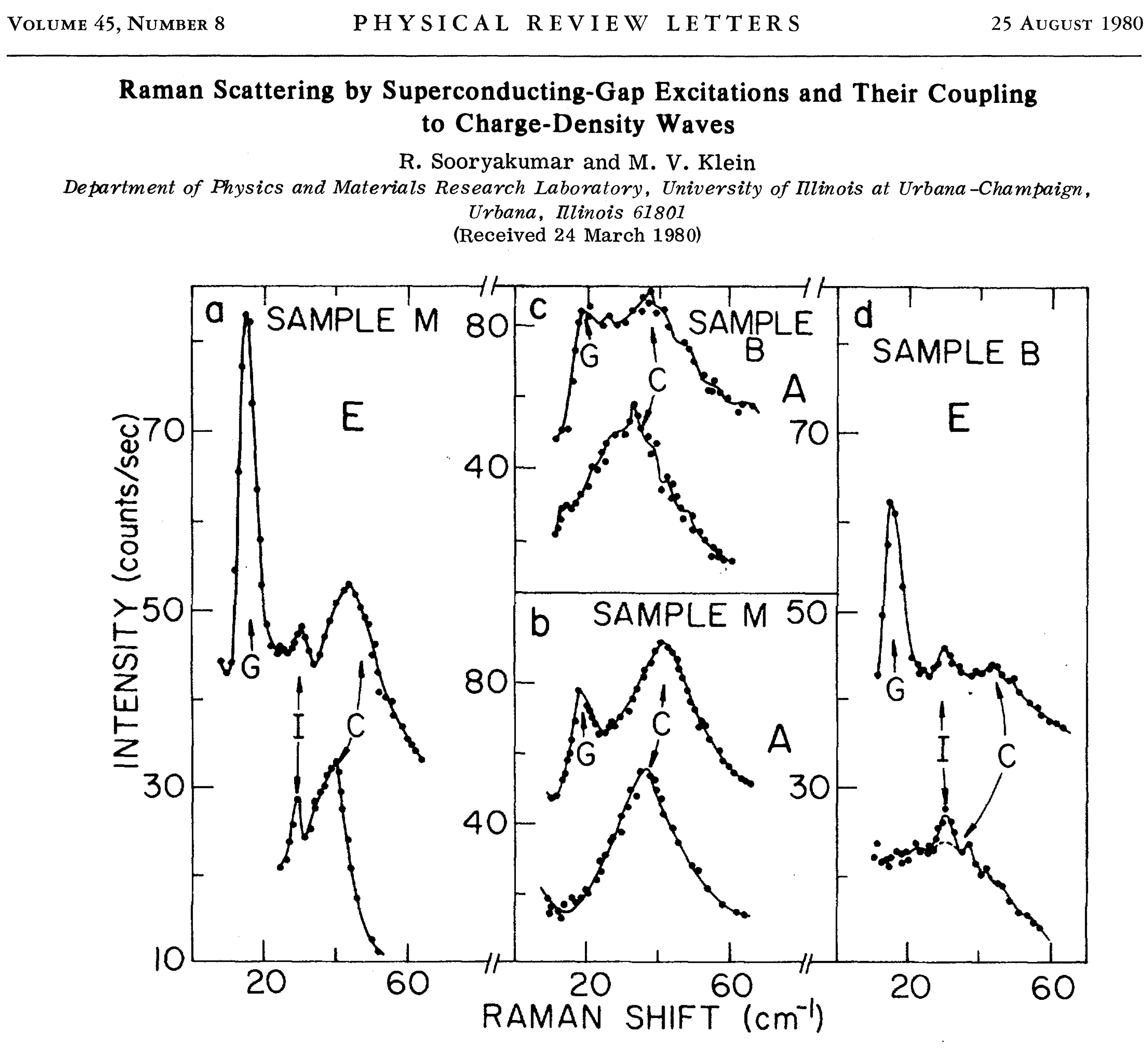
$$\Psi(\boldsymbol{r}) = \sqrt{n_s(\boldsymbol{r})}e^{i\theta(\boldsymbol{r})}$$



- Higgs mode = collective amplitude oscillation of the superfluid density.
- One of the lowest energy excitation modes.
- Universal phenomenon in spontaneous symmetry broken phases

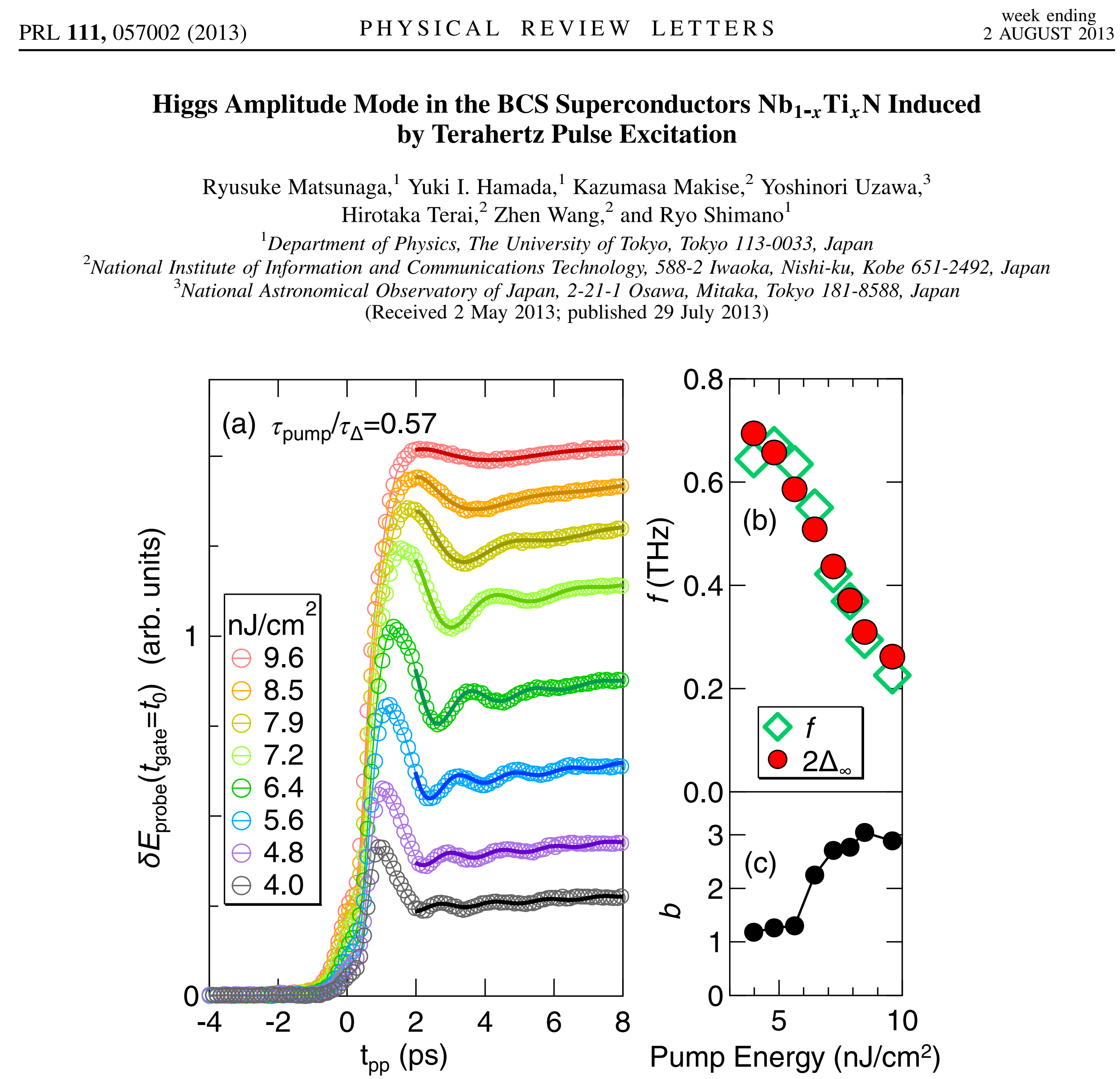
Observation of Higgs mode

- 2H-$NbSe_2$ (SC+CDW), Raman
- $Nb_{1-x}Ti_xN$ (SC only), THz pump-probe


VOLUME 45, NUMBER 8 PHYSICAL REVIEW LETTERS 25 AUGUST 1980

Raman Scattering by Superconducting-Gap Excitations and Their Coupling to Charge-Density Waves

R. Sooryakumar and M. V. Klein
Department of Physics and Materials Research Laboratory, University of Illinois at Urbana-Champaign, Urbana, Illinois 61801
(Received 24 March 1980)


PRL 111, 057002 (2013) PHYSICAL REVIEW LETTERS week ending 2 AUGUST 2013

Higgs Amplitude Mode in the BCS Superconductors $Nb_{1-x}Ti_xN$ Induced by Terahertz Pulse Excitation

Ryusuke Matsunaga,[1] Yuki I. Hamada,[1] Kazumasa Makise,[2] Yoshinori Uzawa,[3] Hirotaka Terai,[2] Zhen Wang,[2] and Ryo Shimano[1]
[1]*Department of Physics, The University of Tokyo, Tokyo 113-0033, Japan*
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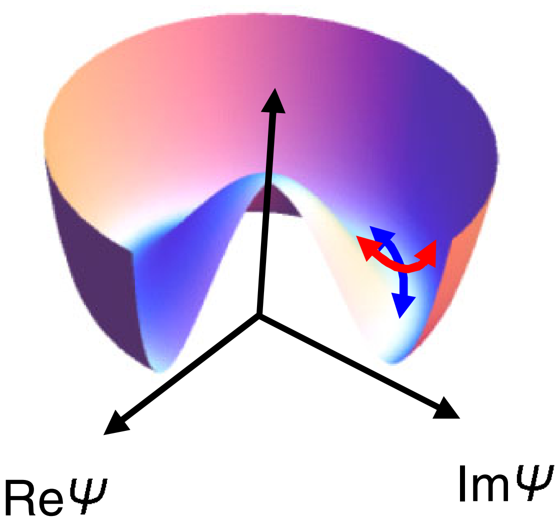
(Received 2 May 2013; published 29 July 2013)

See also Measson et al., PRB 89, 060503 (2014).

Light-Higgs coupling

- Ginzburg-Landau theory

$$F = a|\Psi|^2 + \frac{b}{2}|\Psi|^4 + \frac{1}{2m^*}|(-i\nabla - e^* \boldsymbol{A})\Psi|^2$$



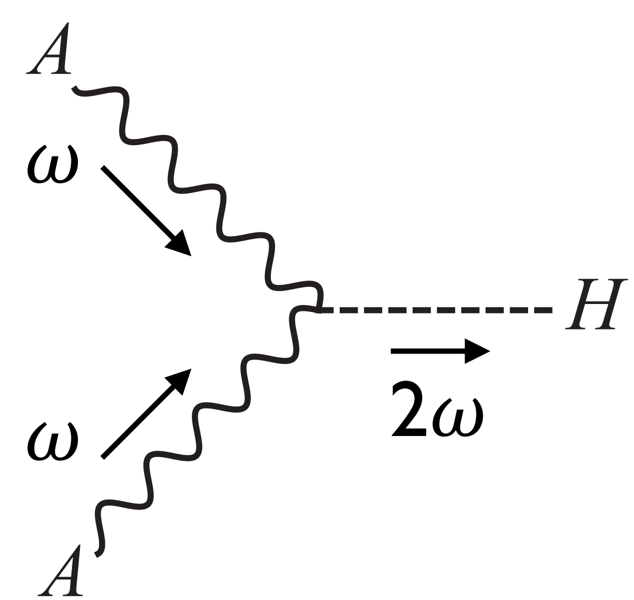
- Amplitude oscillation

$$\Psi(\boldsymbol{r}, t) = \Psi_0 + H(\boldsymbol{r}, t)$$

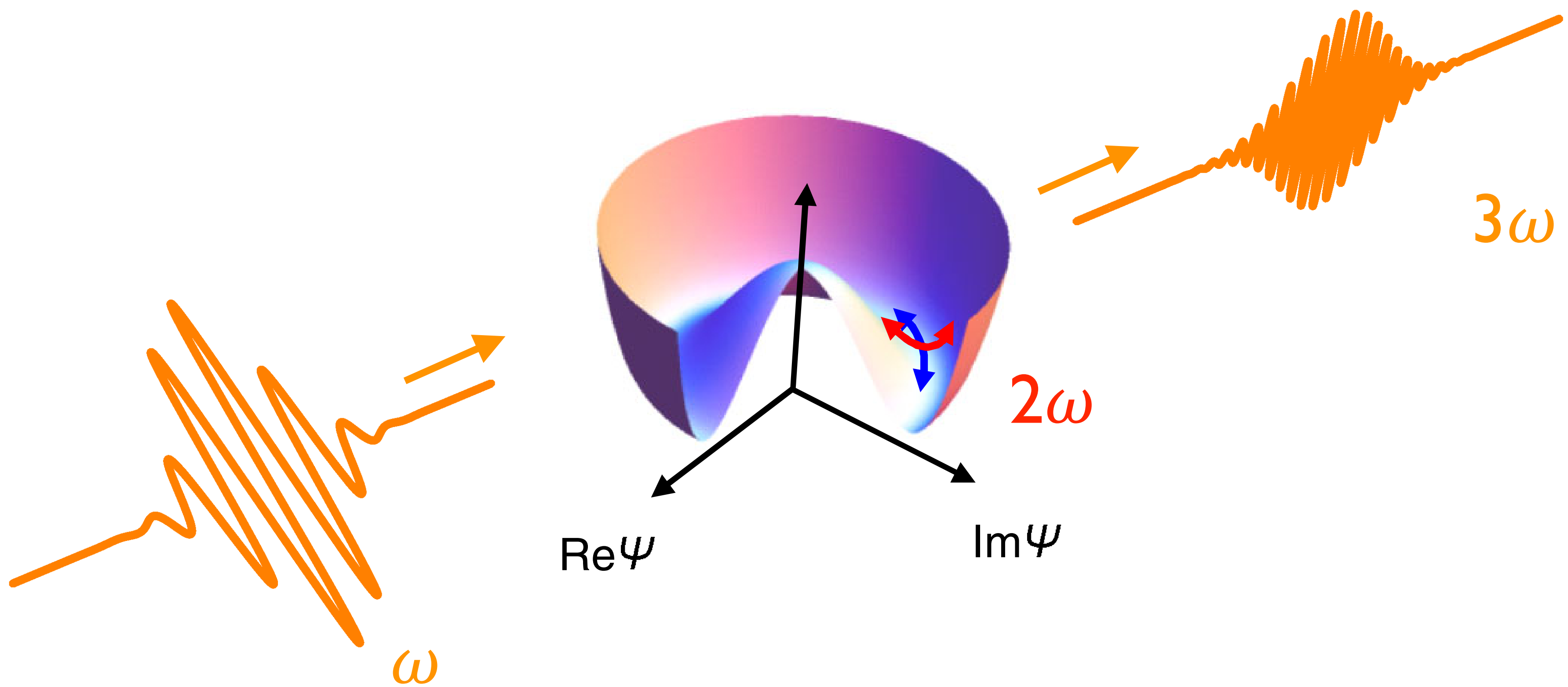
$$F = -2aH^2 + \frac{1}{2m^*}(\nabla H)^2 - \underbrace{\frac{e^{*2}\Psi_0^2}{2m^*}\boldsymbol{A}^2}_{\text{mass of EM field (Meissner effect)}} + \underbrace{\frac{e^{*2}\Psi_0}{m^*}\boldsymbol{A}^2 H}_{\text{Coupling between Higgs and EM fields}} + \cdots$$

mass of EM field
(Meissner effect)

Coupling between Higgs and EM fields



Third-harmonic generation from Higgs



- Current: $$j(t) = -\frac{e^{*2} n_s(t)}{m^*} A(t)$$ (London equation)

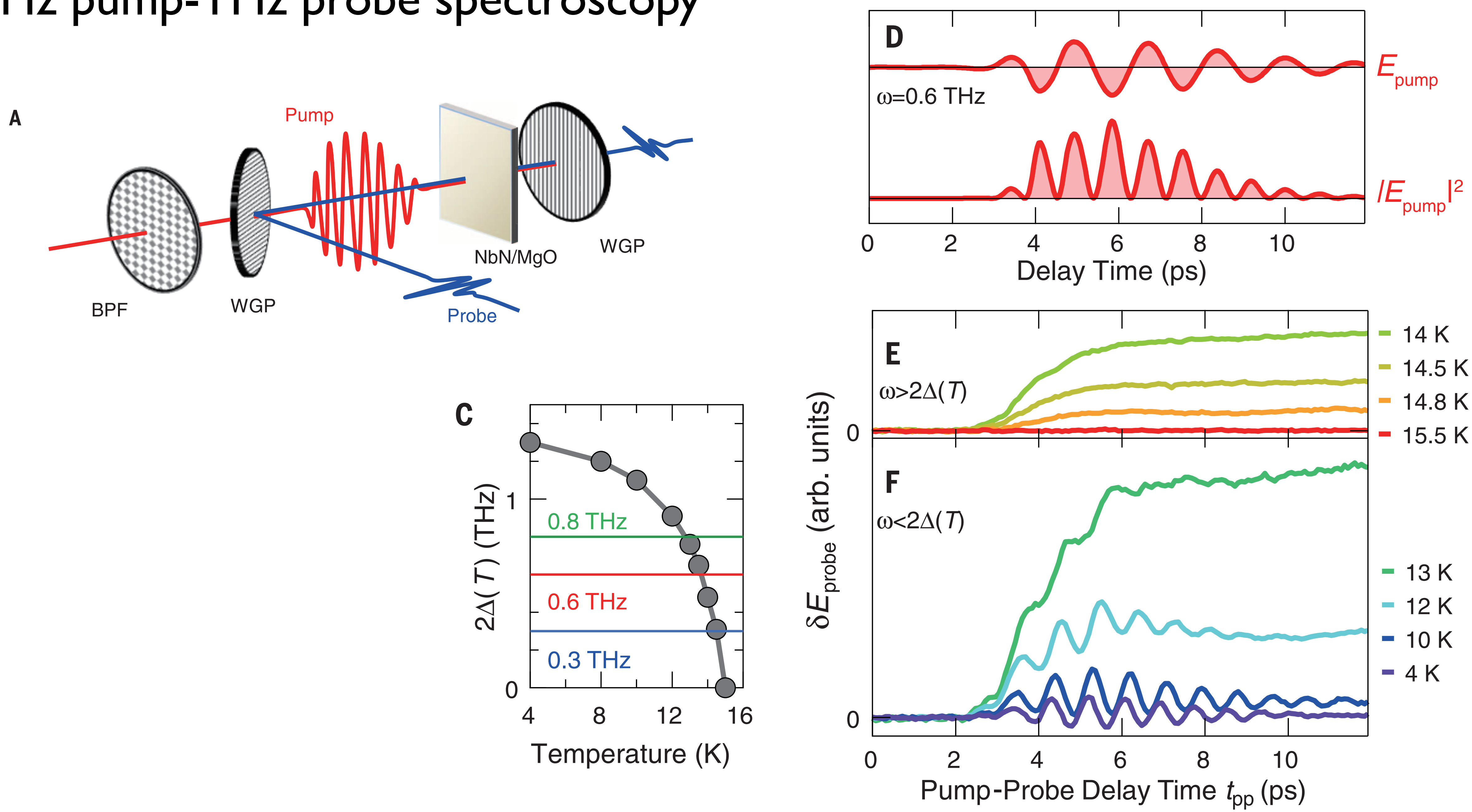
$$j_{\mathrm{NL}}(t) = -\frac{2e^{*2}\Psi_0}{m^*} \underline{H(t)}\,\underline{A(t)}$$

2ω + ω = 3ω oscillation

THz laser experiment

Matsunaga, Tsuji, Aoki, Shimano, et al., Science 345, 1145 (2014)

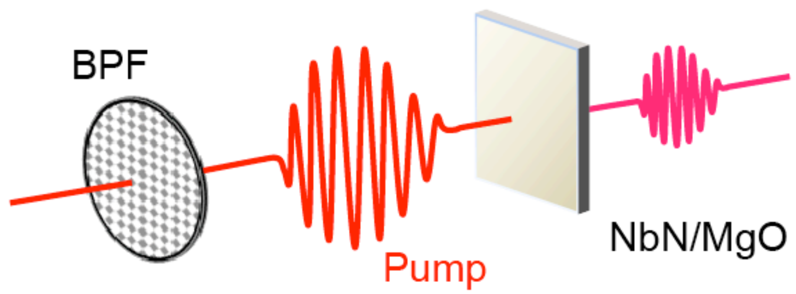
- THz pump-THz probe spectroscopy

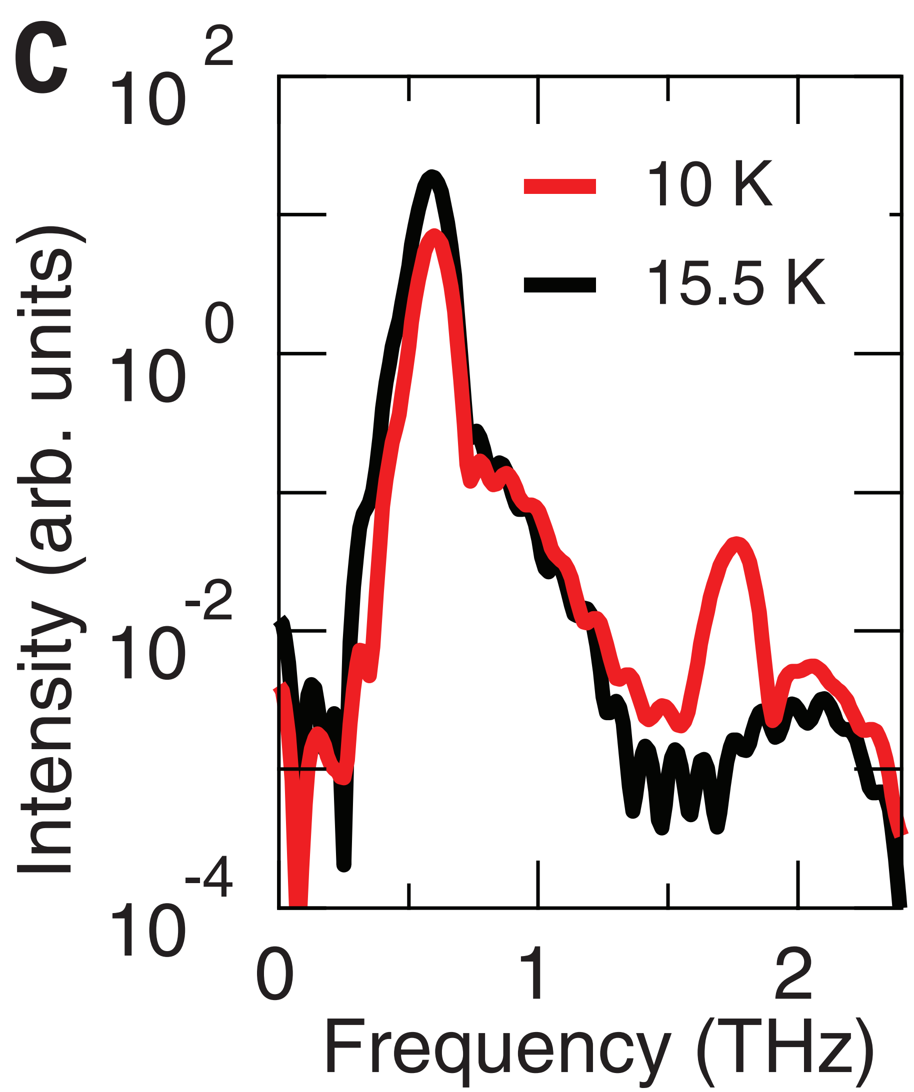


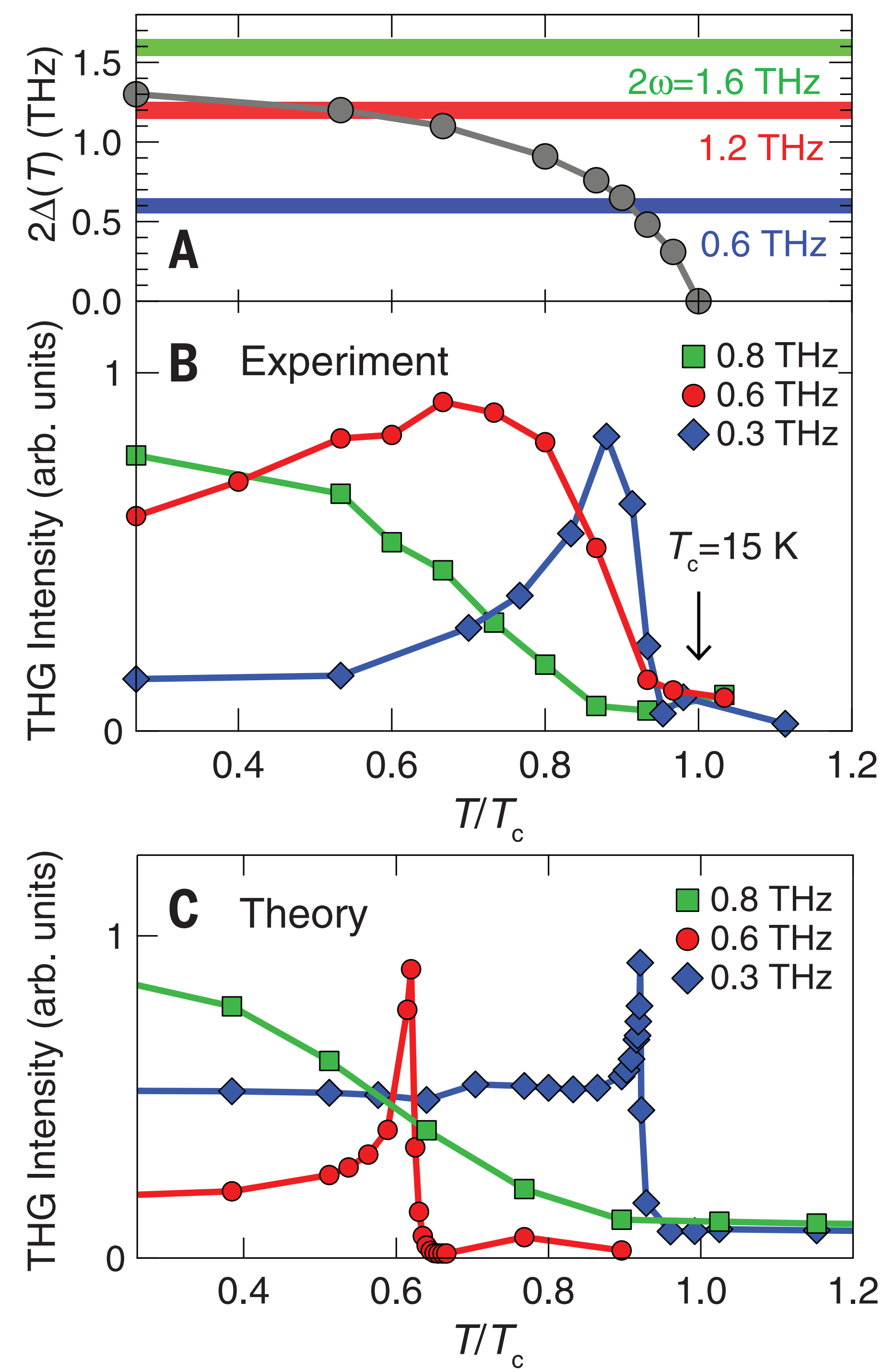
2ω oscillation of the superfluid density

THG resonance experiment

Matsunaga, Tsuji, Aoki, Shimano, et al., Science 345, 1145 (2014)







Resonance at $2\omega=2\Delta$

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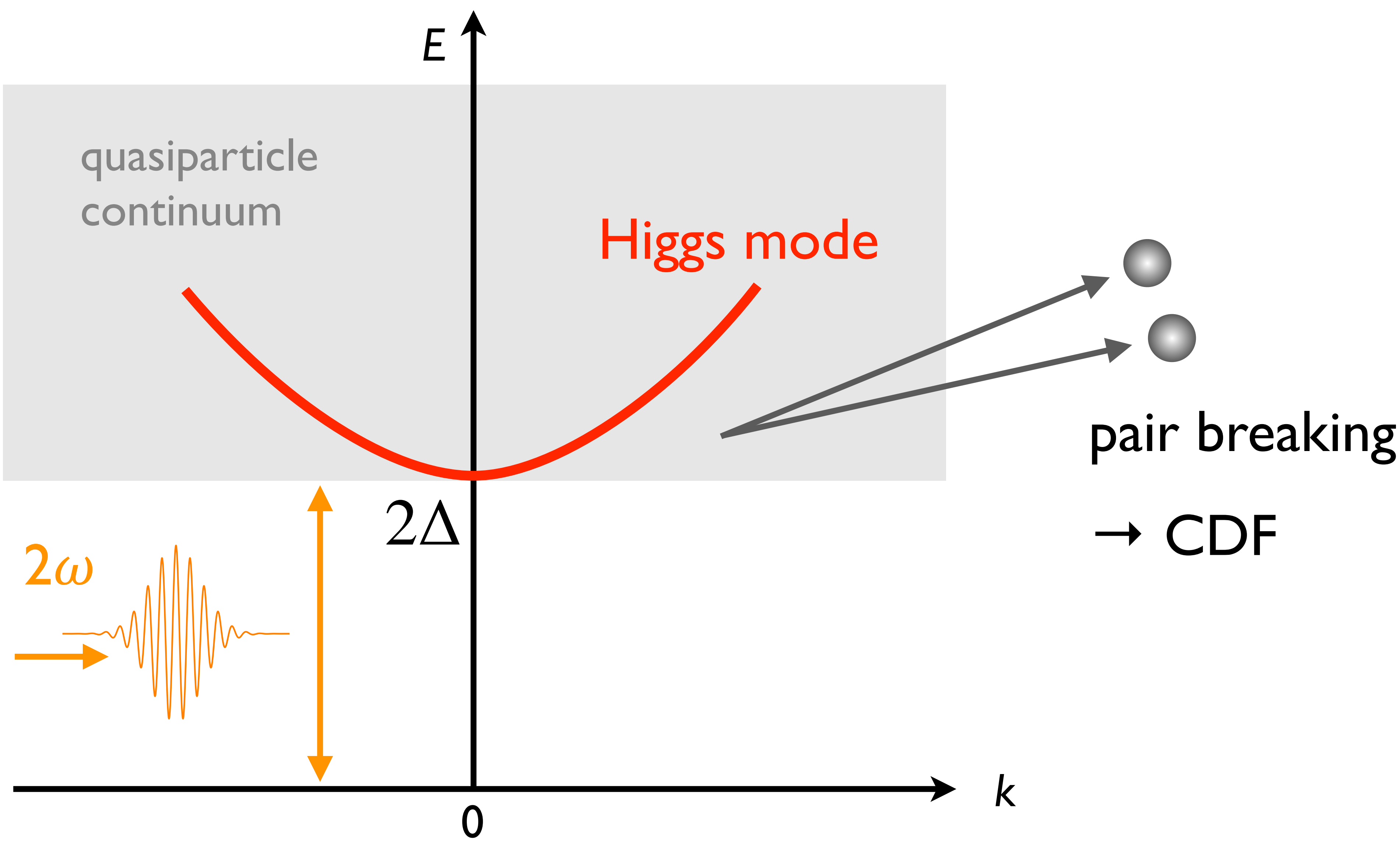
Higgs mode vs CDF

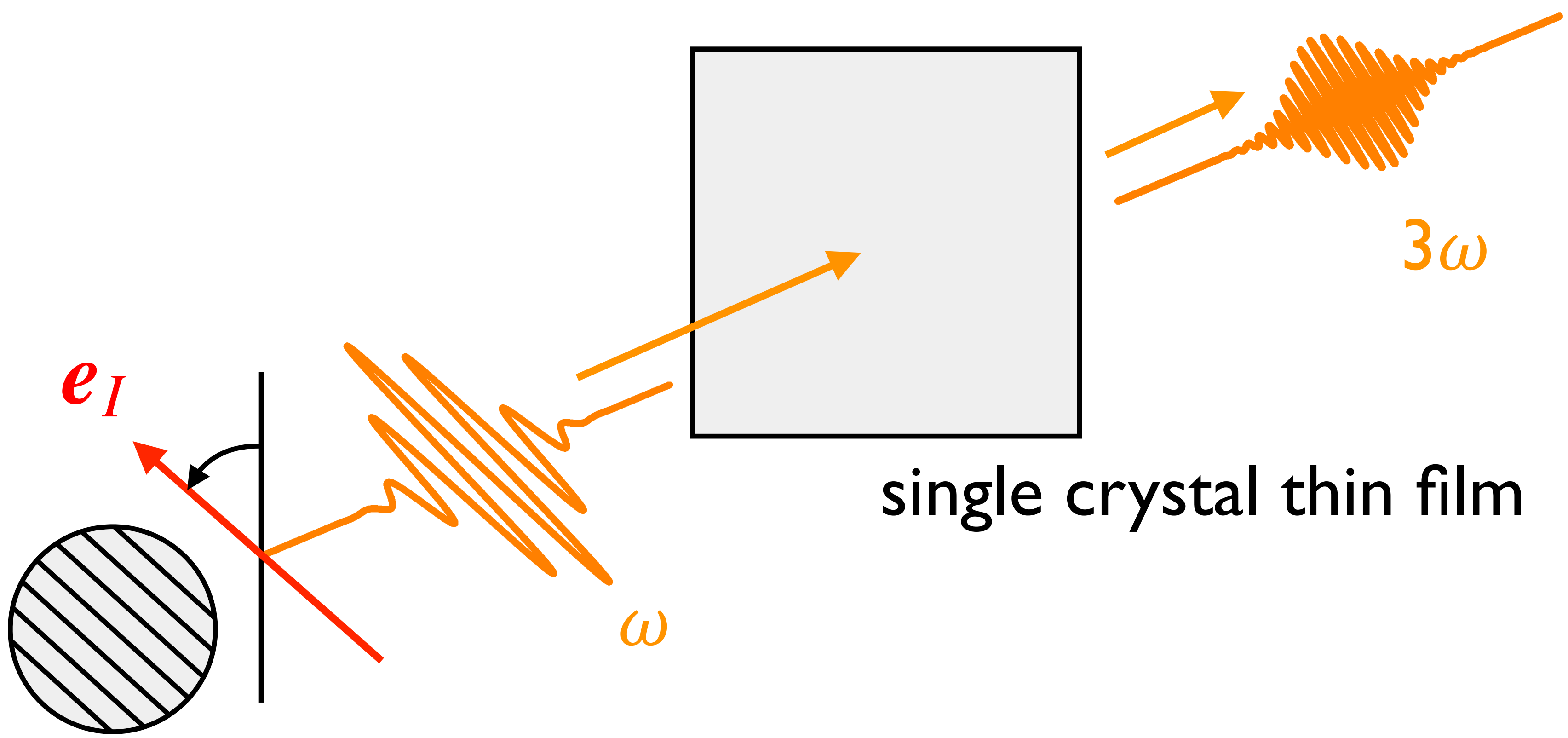
- According to BCS theory, Higgs-mode energy is degenerate with the lower bound of the quasiparticle continuum (2Δ).



- Laser may excite not only Higgs but also charge density fluctuation (CDF).

Polarization dependence of THG

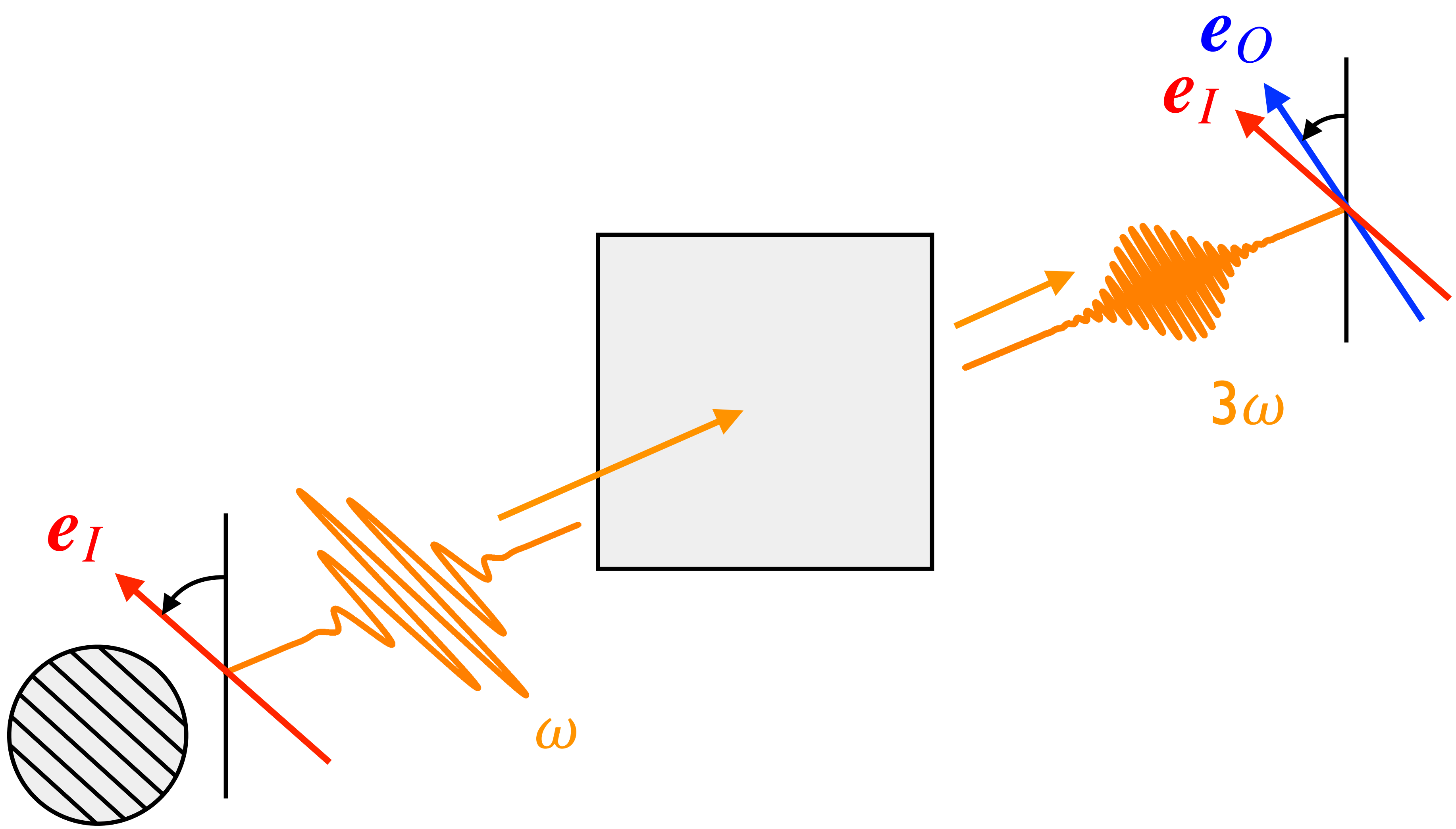
- CDF and Higgs have different polarization-angle dependence for THG resonance. Cea, Castellani, Benfatto, Phys. Rev. B 93, 180507 (2016)



Polarization dependence of CDF

- In general, CDF has polarization dependence relative to the crystal axis since it depends on the band structure of the material.

- CDF also has a THG component with polarization perpendicular to that of the incoming wave. Matsunaga, Tsuji et al., arXiv:1703.02815.



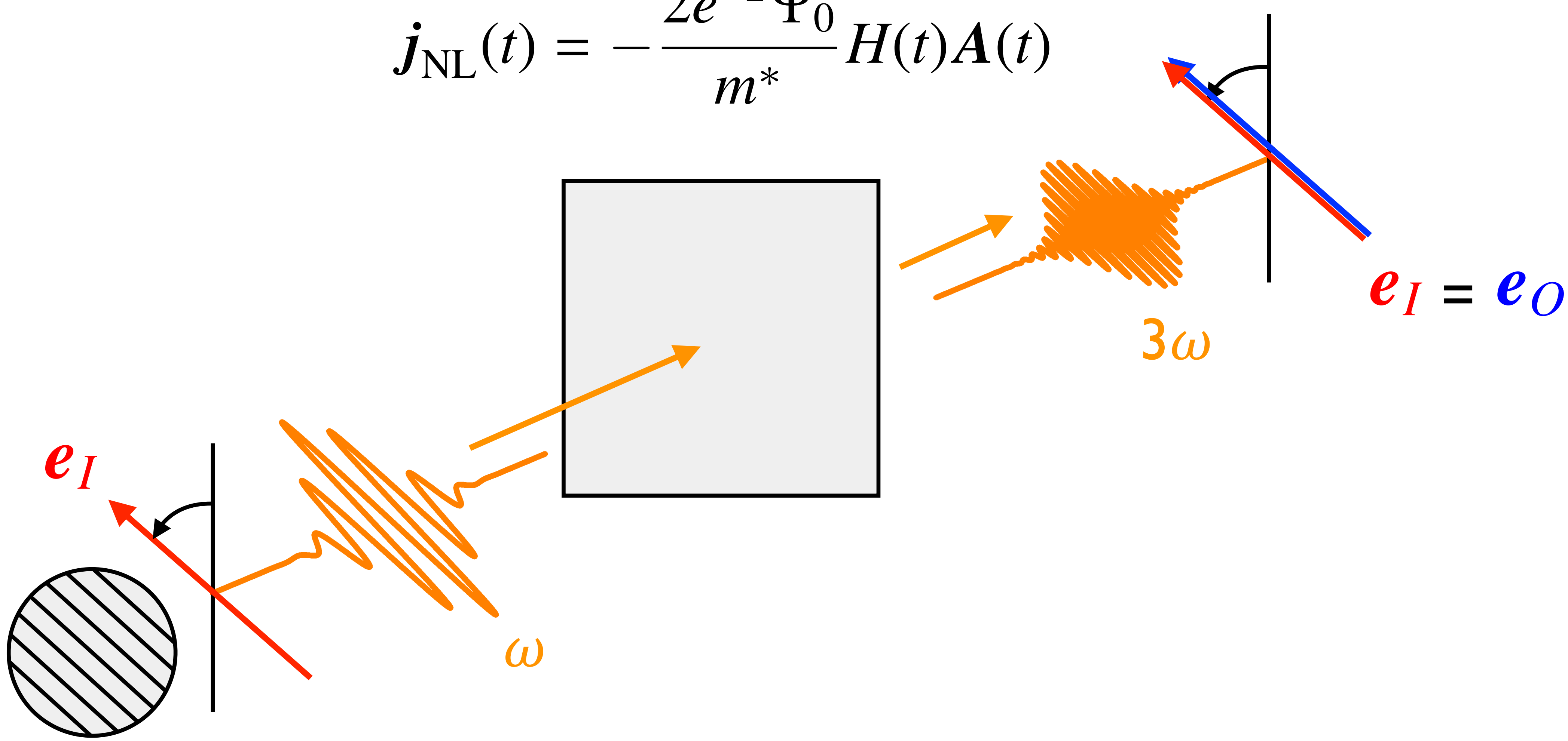
Polarization dependence of Higgs

- Higgs is a scalar boson, and does not have polarization dependence.

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

- Higgs always induces THG with polarization parallel to that of the incoming wave.

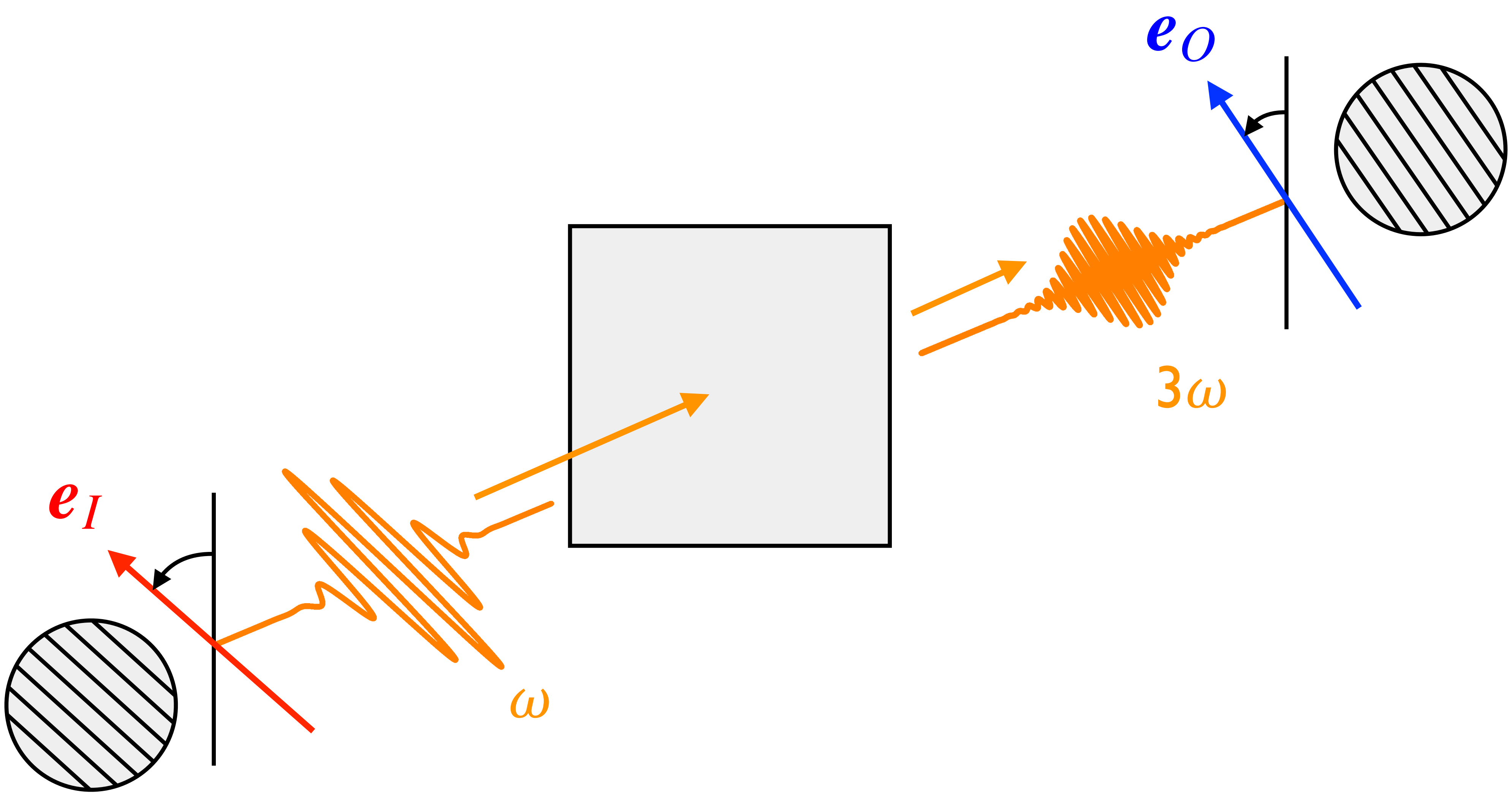
$$\boldsymbol{j}_{\mathrm{NL}}(t) = -\frac{2e^{*2}\Psi_0}{m^*}H(t)\boldsymbol{A}(t)$$



General Polarization dependence

Matsunaga, Tsuji et al., arXiv:1703.02815.

	$\boldsymbol{e}_I \parallel \boldsymbol{e}_O$	$\boldsymbol{e}_I \perp \boldsymbol{e}_O$
CDF	$A(\omega)+2B(\omega)\sin^2 2\theta$	$B(\omega)\sin 4\theta$
Higgs	$C(\omega)$	0



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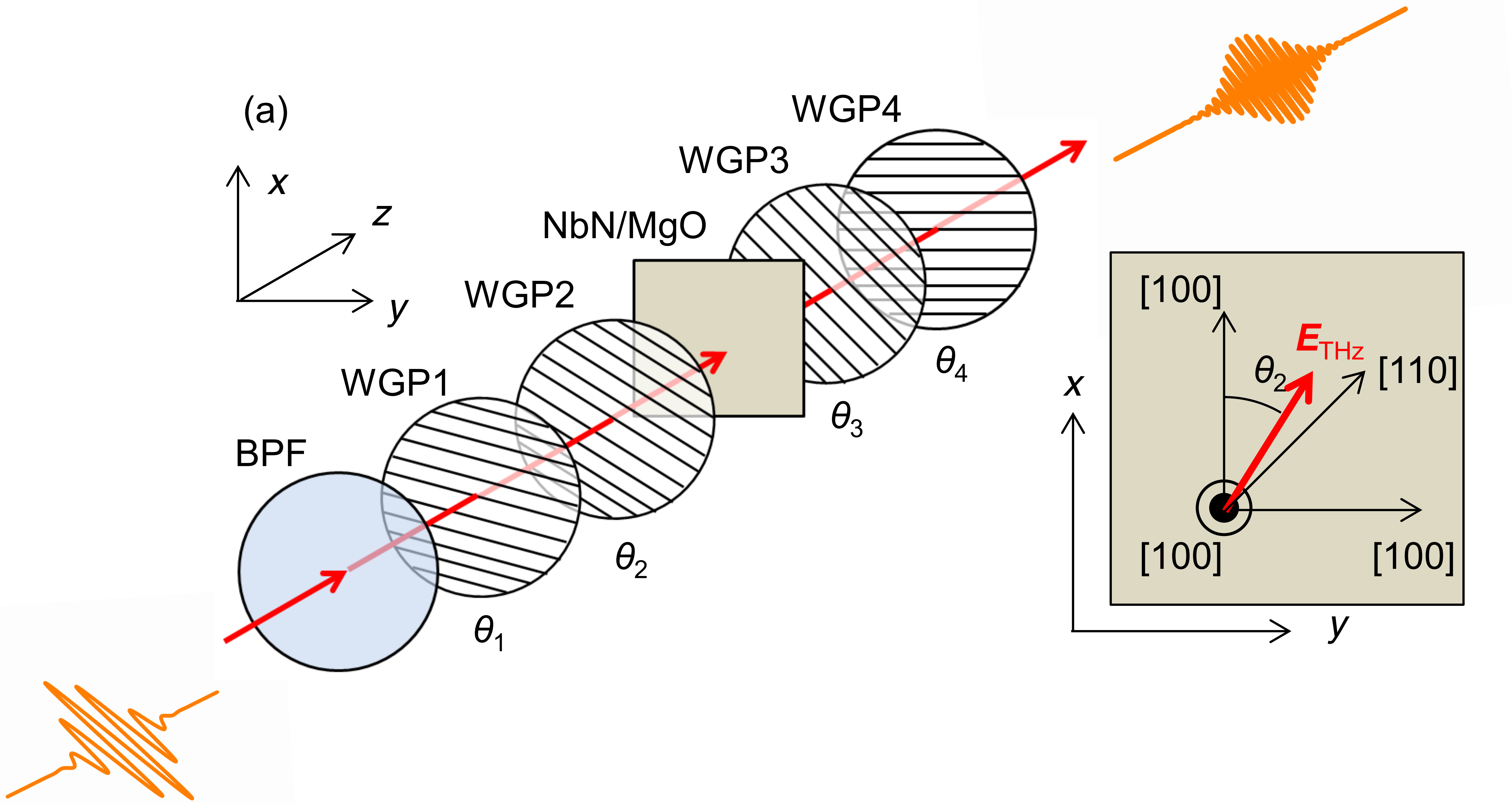
Polarization-resolved spectroscopy

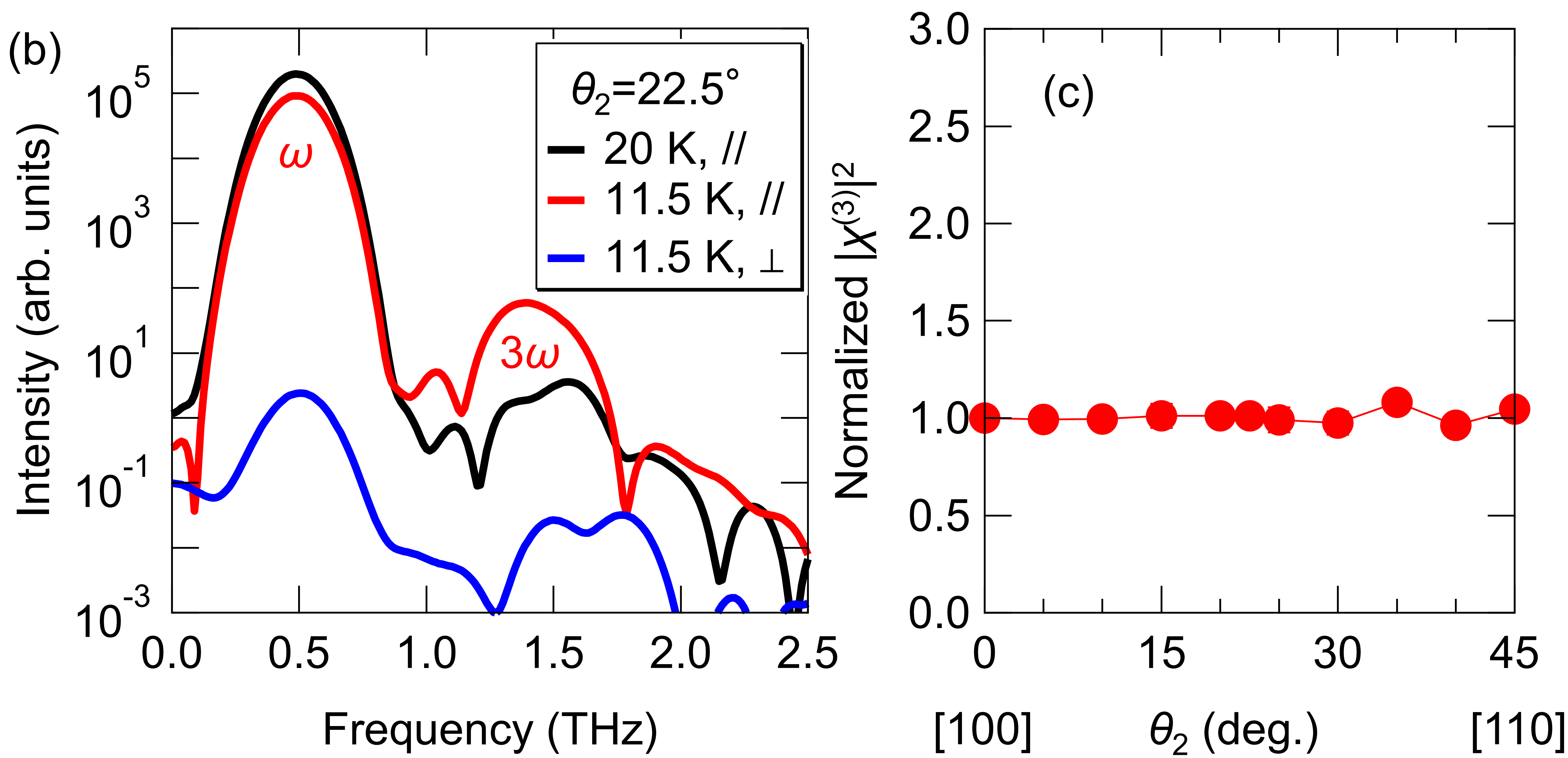
Matsunaga, Tsuji et al., arXiv:1703.02815.



Polarization dependence of THG

Matsunaga, Tsuji et al., arXiv:1703.02815.

- Experiment:

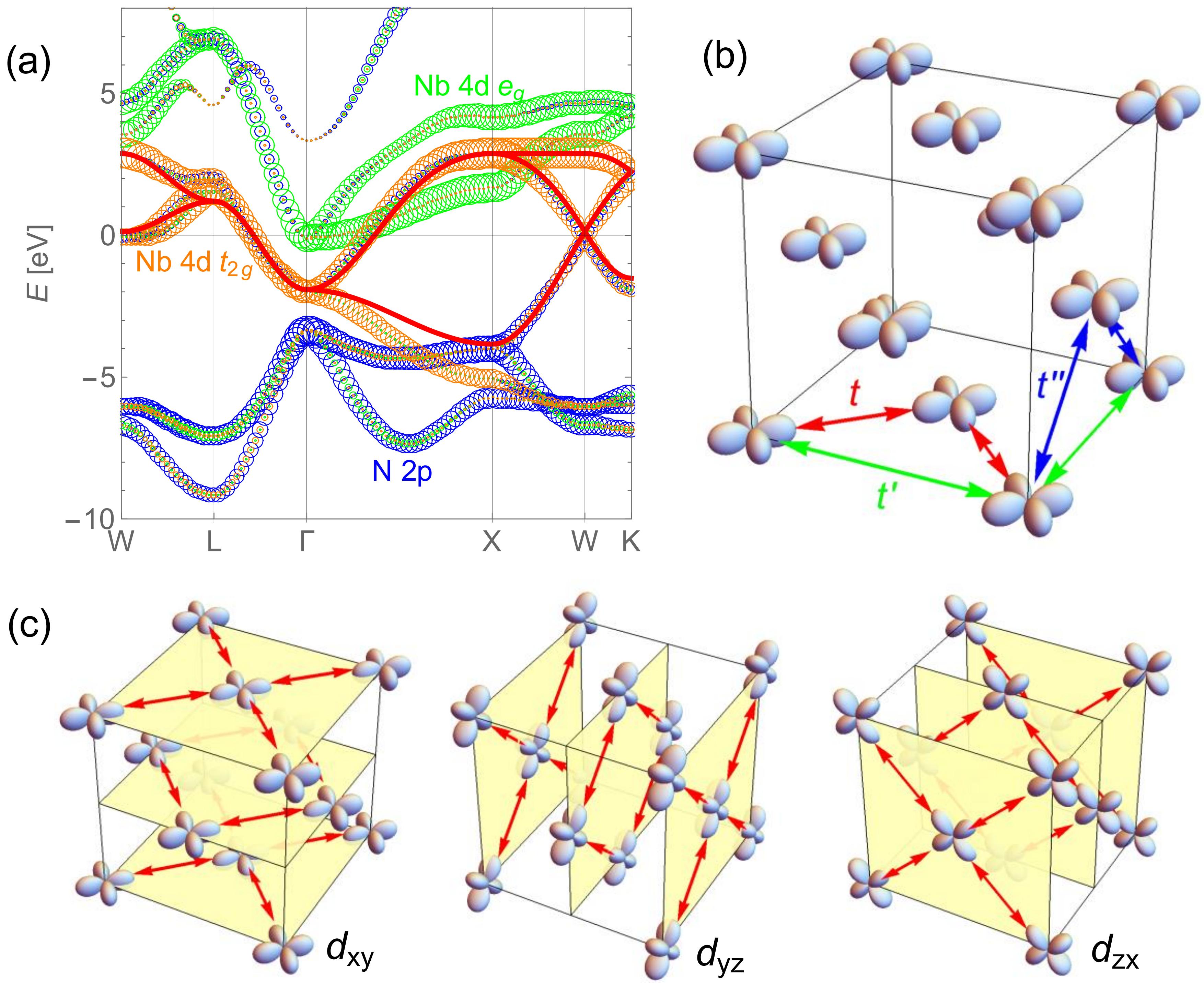


- THG intensity is quite isotropic, and does not depend on the polarization.

← It implies that the Higgs is a dominant contribution to THG.

Band structure of NbN

Matsunaga, Tsuji et al., arXiv:1703.02815.



t = -0.72 eV

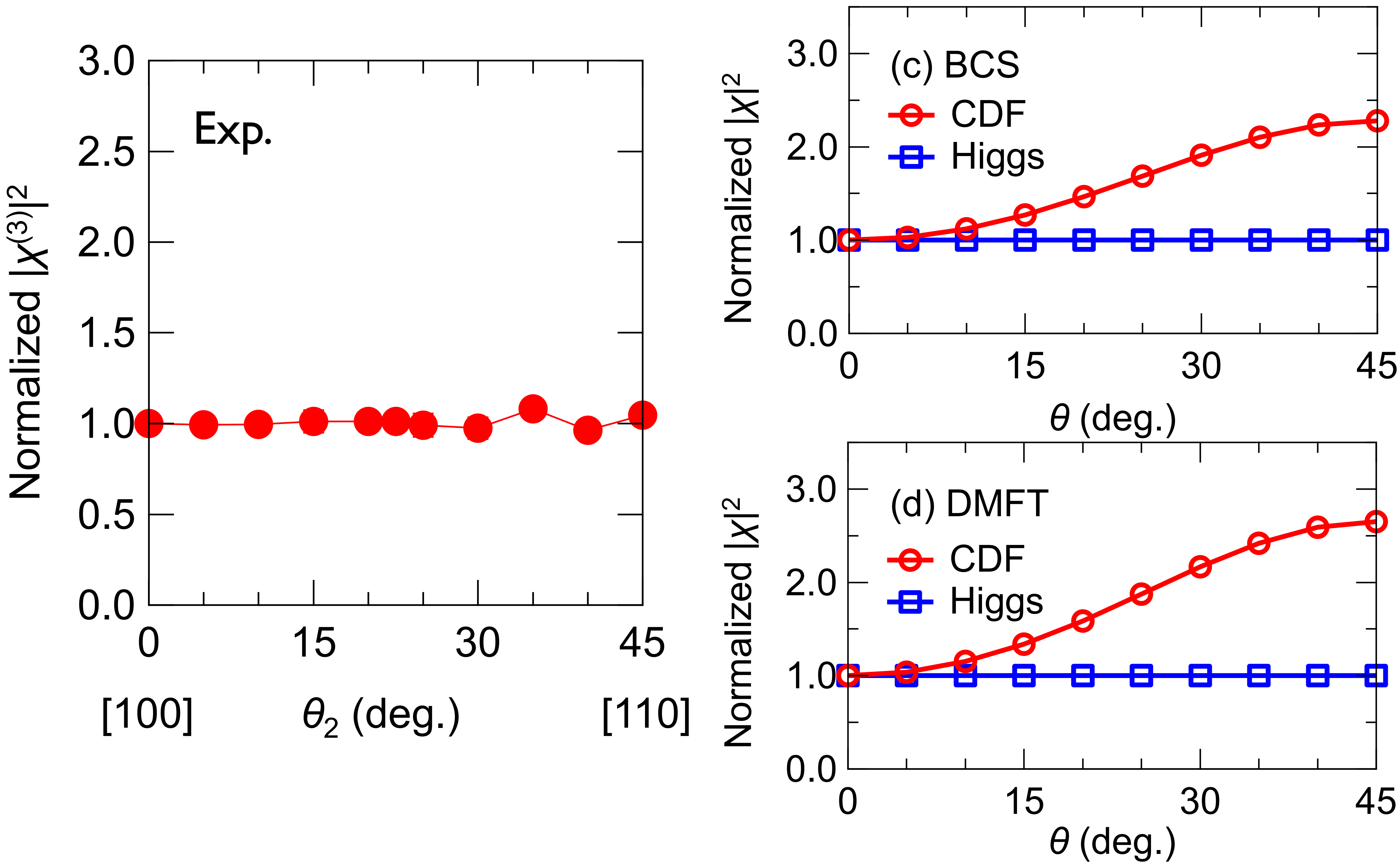
t' = -0.15 eV

t'' = 0.12 eV

μ = -0.6 eV

Polarization dependence of THG

Matsunaga, Tsuji et al., arXiv:1703.02815.



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Higgs mode vs CDF

- In BCS theory, for a general lattice with a general laser polarization CDF has much stronger coupling to light than Higgs.

Cea, Castellani, Benfatto, Phys. Rev. B 93, 180507 (2016)

- Why is the Higgs mode contribution to THG so dominant for NbN?

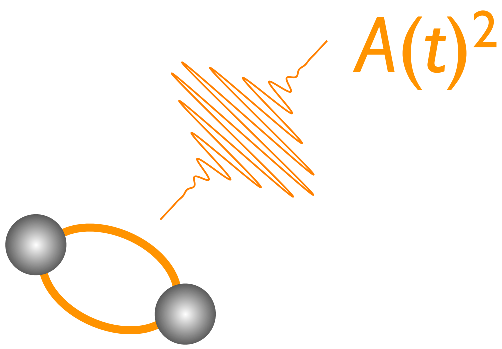
- NbN is known to have a strong electron-phonon coupling ($\lambda \sim 1$).

Kihlstrom, Simon, Wolf, PRB 32, 1843 (1985); Bronson et al., PRL 64, 2172 (1990); Chockalingam et al., PRB 77, 214503 (2008).

- Is there any mechanism to enhance the light-Higgs coupling?

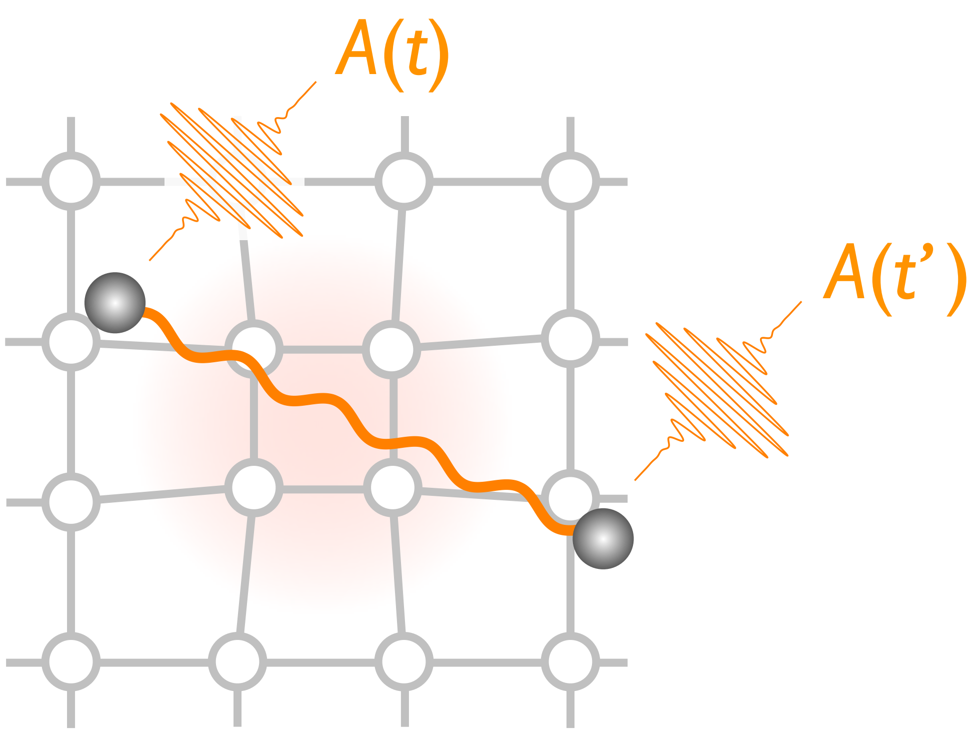
Retardation effect

- Un-retarded pairing

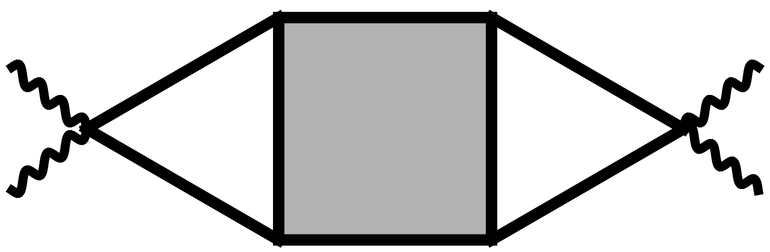
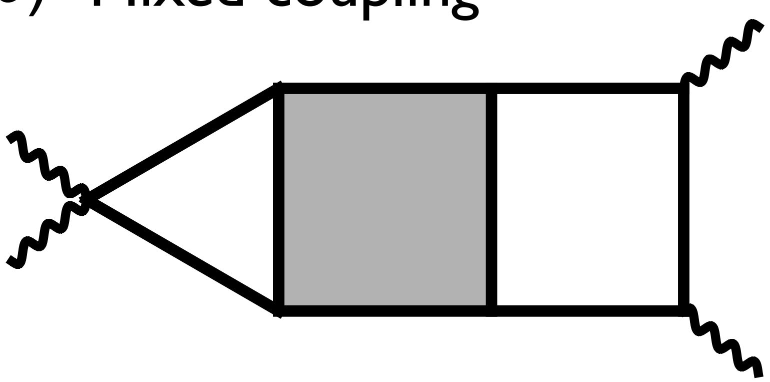
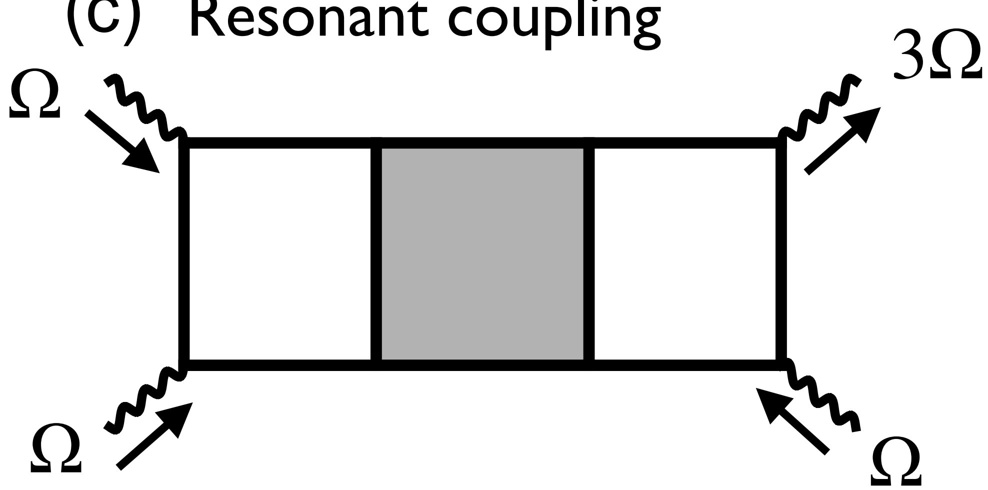


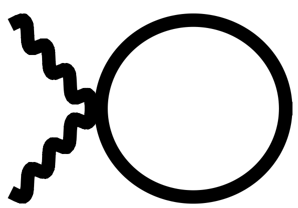
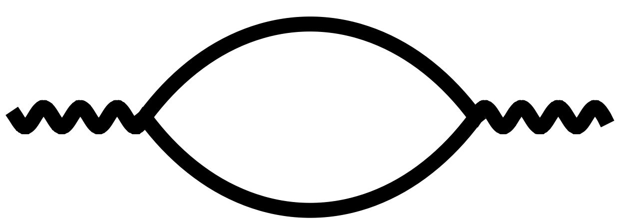
non-resonant
light-matter coupling

- Retarded pairing



resonant
light-matter coupling

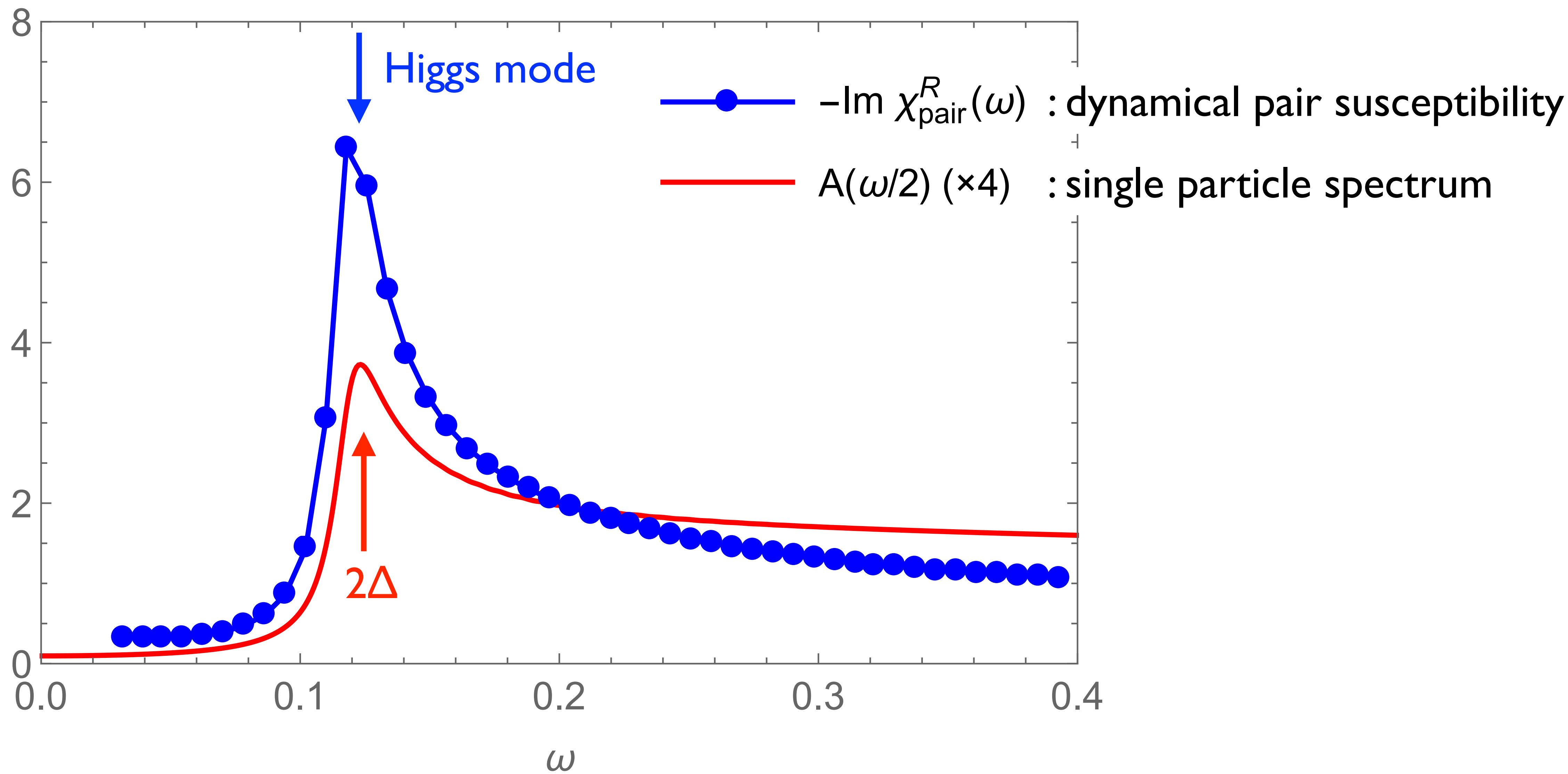
Higgs diagrams	BCS (clean limit)	Beyond BCS
(a) Non-resonant coupling 	✔	✔
(b) Mixed coupling 	🚫	✔
(c) Resonant coupling 	🚫	✔

Optical conductivity	BCS (clean limit)	Beyond BCS
(a) Diamagnetic term		
	✔	✔
(b) Paramagnetic term		
	🚫	✔
$\mathrm{Re}\,\sigma(\omega \neq 0)$	$= 0$	$\neq 0$

Model of el-ph system

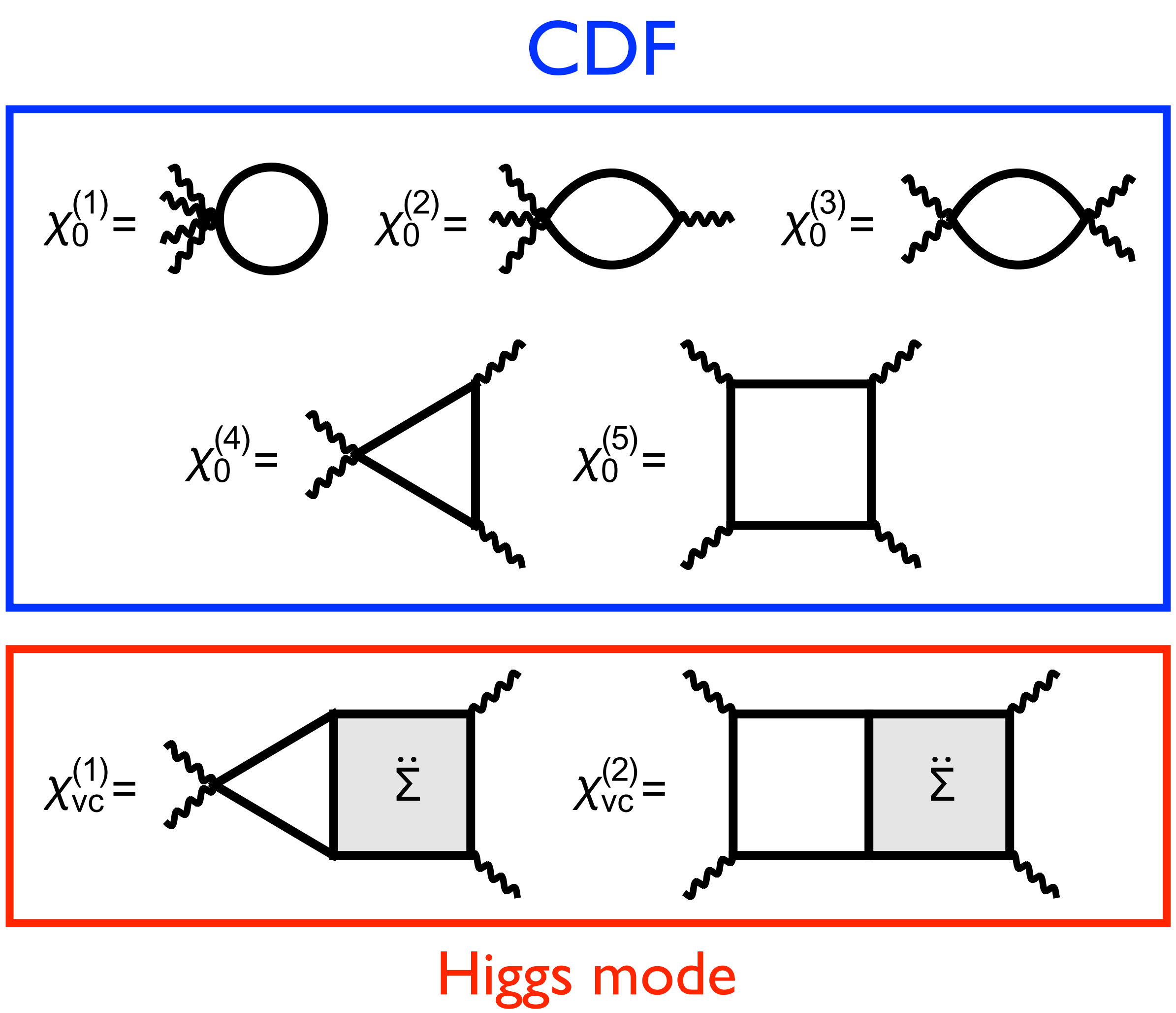
- Holstein model

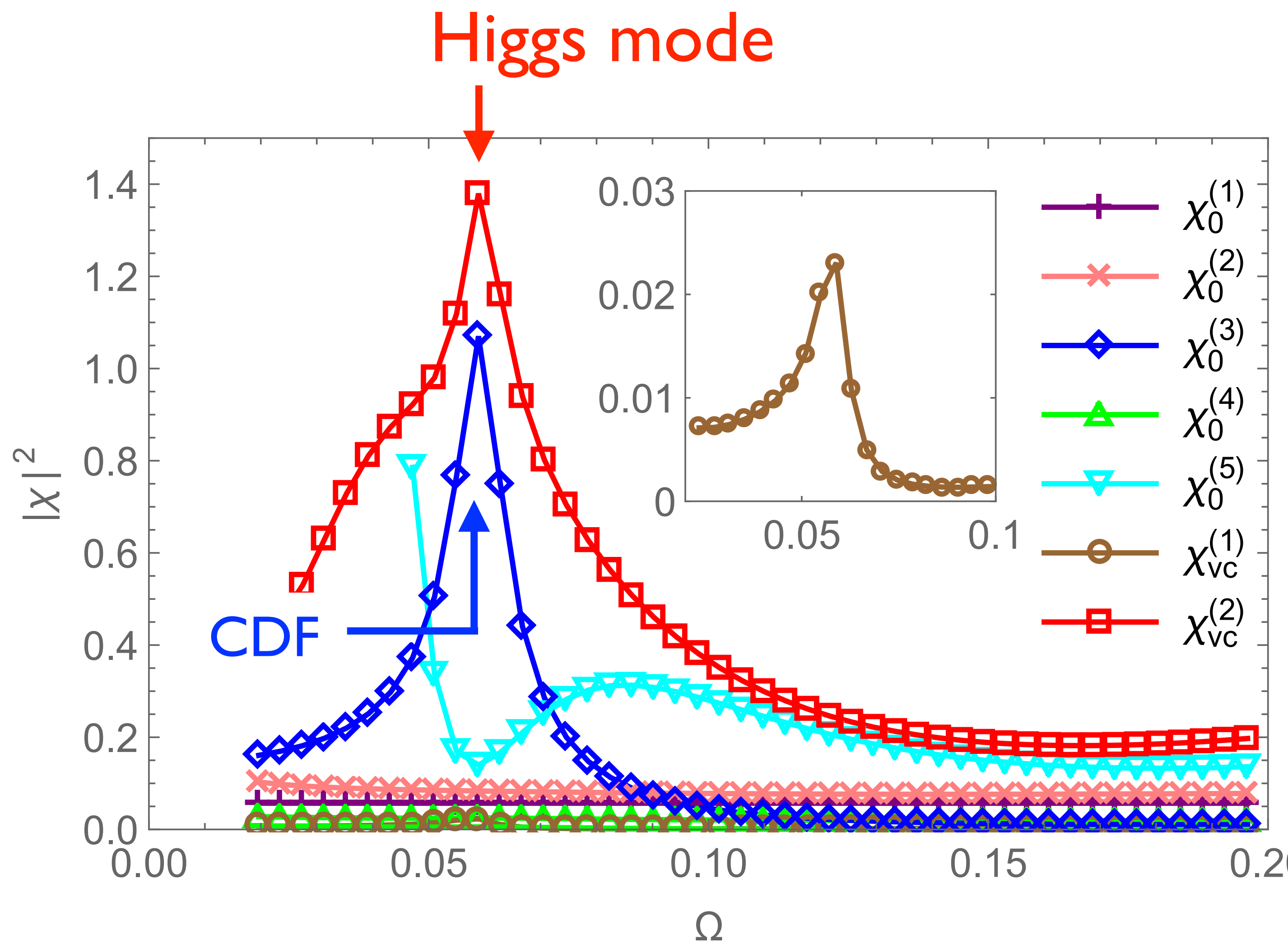
$$H = \sum_{ij,\sigma} t_{ij}(c^\dagger_{i\sigma}c_{j\sigma} + \mathrm{h.c.}) - \mu\sum_i n_i + \omega_0 \sum_i b^\dagger_i b_i + g\sum_i (b_i + b^\dagger_i)(n_i - 1)$$



$g = 0.8, \omega_0 = 0.6, T = 0.02, \gamma = 0.2, \delta = 0.005 \quad (\lambda = 0.77)$ Tsuji, Murakami, Aoki, Phys. Rev. B 94, 224519 (2016)

THG susceptibility (DMFT calc.)





$g = 0.8, \omega_0 = 0.6, T = 0.02, \gamma = 0.2, \delta = 0.005$

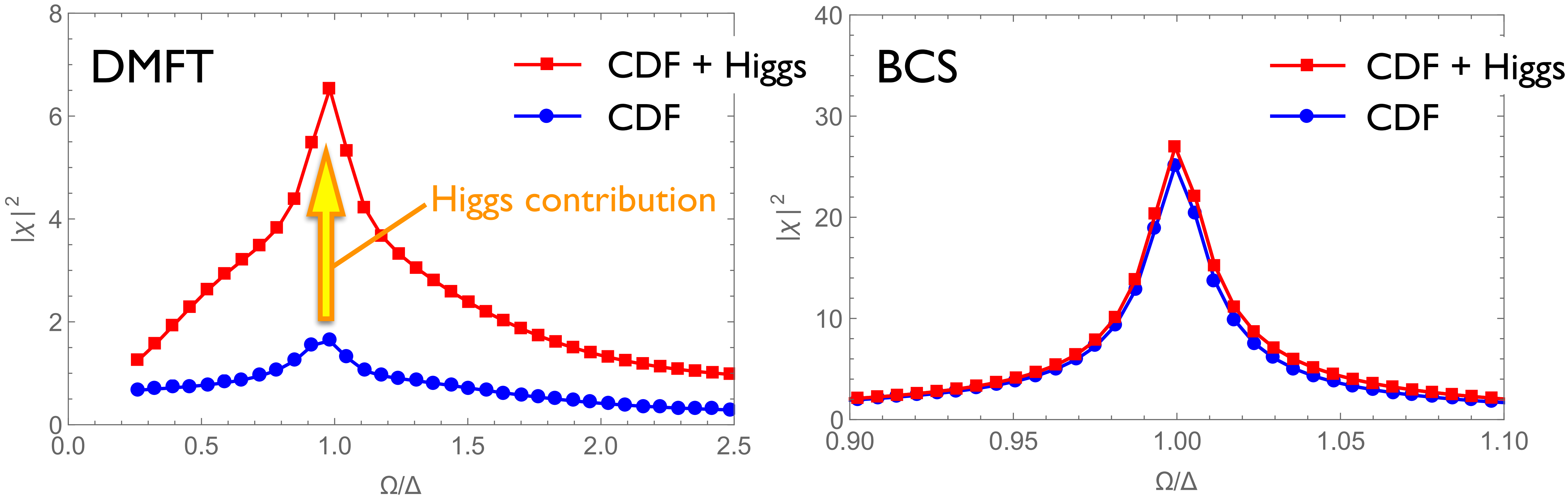
Polarization: $\alpha = 0.5$

Tsuji, Murakami, Aoki, Phys. Rev. B 94, 224519 (2016)

Higgs mode can contribute to THG resonance with an order of magnitude comparable or even larger than the contribution of CDF.

Comparison to BCS theory

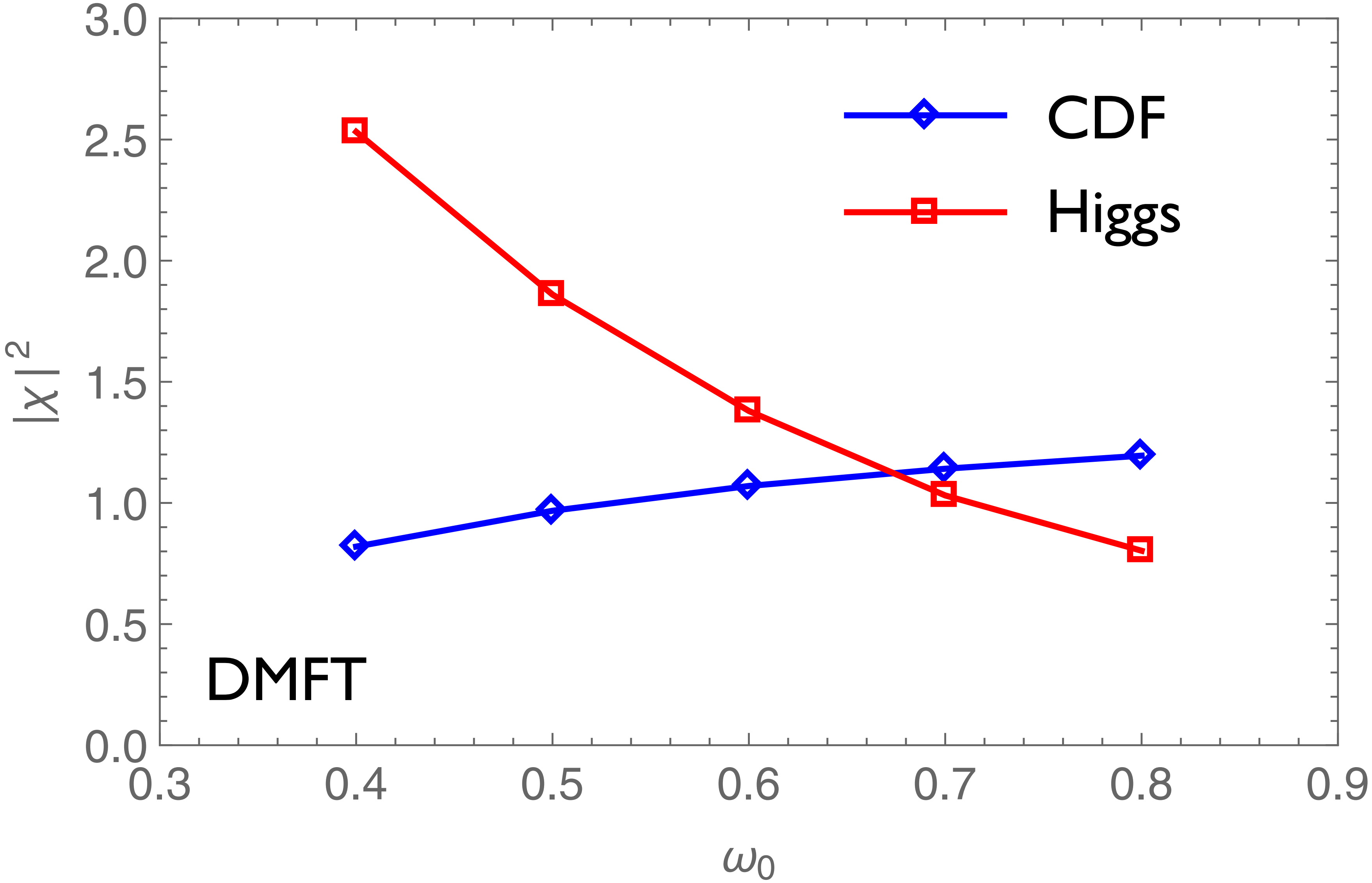
Tsuji, Murakami, Aoki, Phys. Rev. B 94, 224519 (2016)



$g = 0.8, \omega_0 = 0.6, T = 0.02, \gamma = 0.2, \delta = 0.005$ Polarization: $\alpha = 0.5$

In DMFT the Higgs-mode contribution is enhanced due to the retardation effect mediated by phonons, which is in sharp contrast to BCS.

Phonon frequency dependence



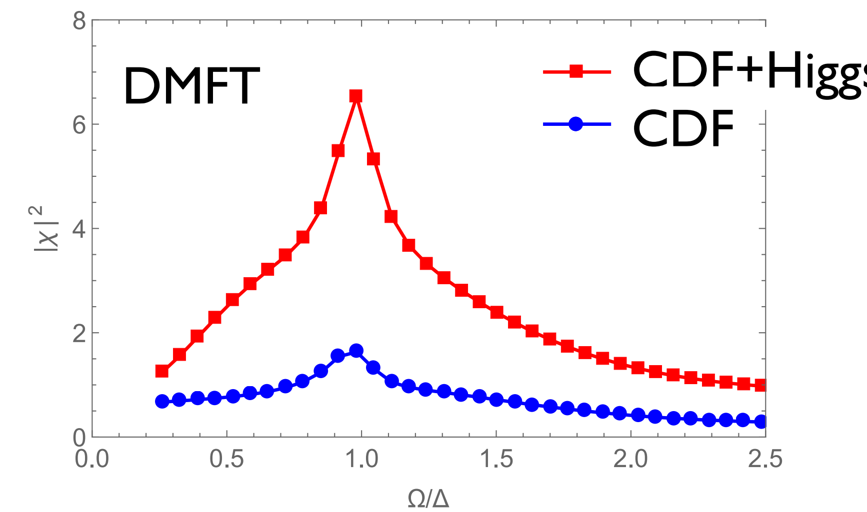
$T = 0.02, \gamma = 0.2, \delta = 0.005, \alpha = 0.5$

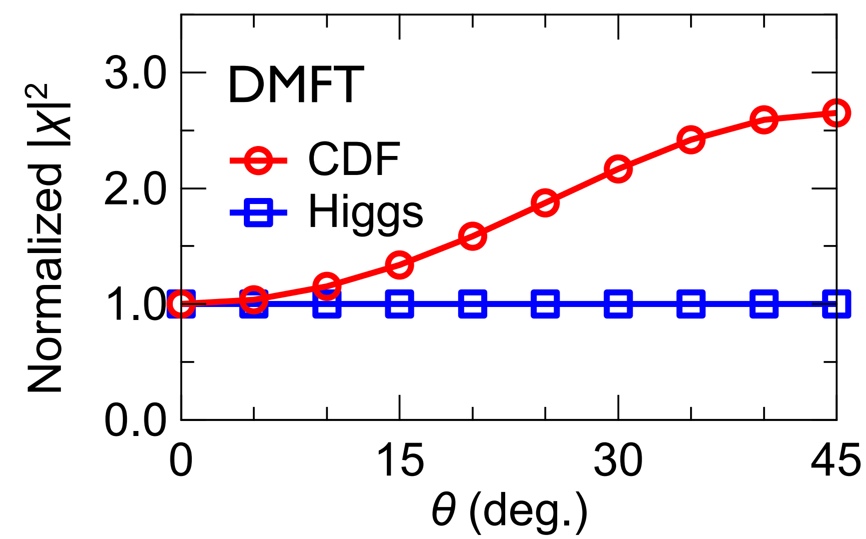
Tsuji, Murakami, Aoki, Phys. Rev. B 94, 224519 (2016)

We tune g for each ω_0 such that the superconducting gap is fixed to be $2\Delta \approx 0.12$.

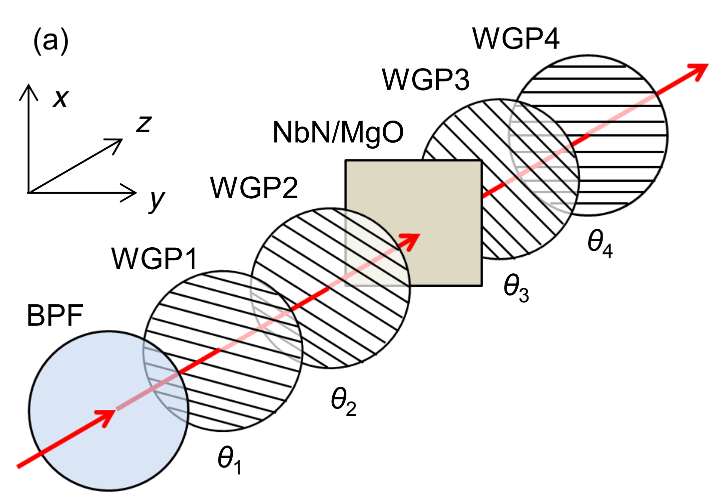
Summary

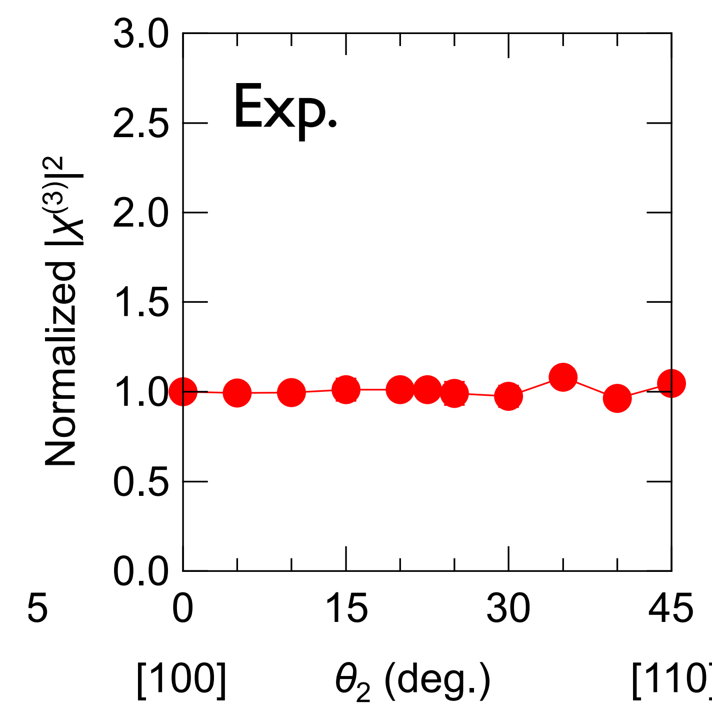
- Enhancement of Higgs mode due to the phonon retardation effect.
- Strong light-Higgs interaction through the resonant coupling.
- Polarization-angle independence of THG resonance in NbN superconductor.
- Dominant contribution to THG from Higgs mode rather than from CDF.





[1] Tsuji, Murakami, Aoki, Phys. Rev. B 94, 224519 (2016).
[2] Matsunaga, Tsuji et al., arXiv: 1703.02815. (To be published in Phys. Rev. B, Rapid Comm.)

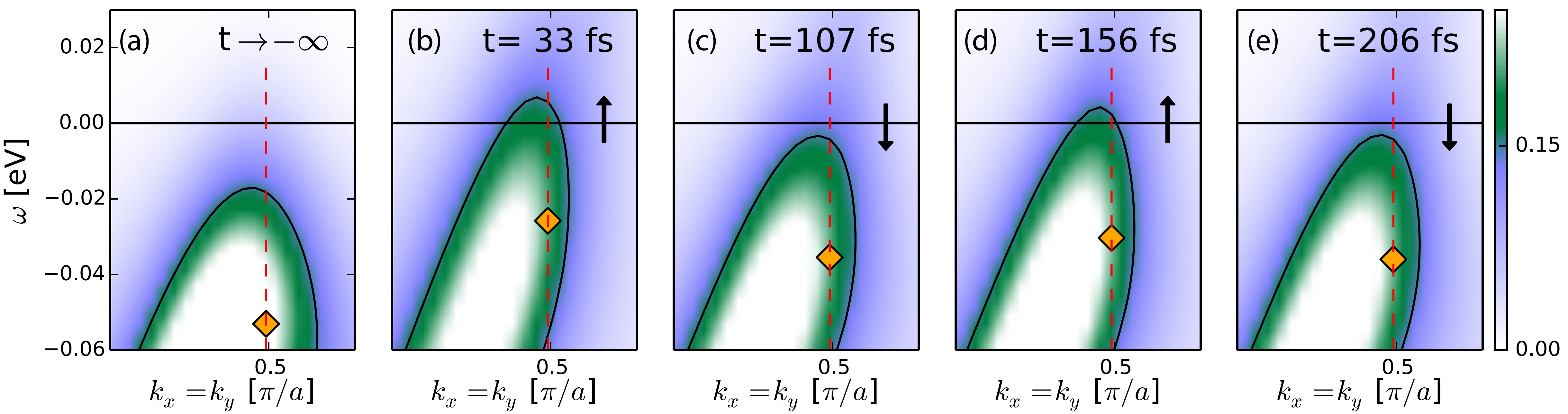




Big challenge

- Can we directly observe the Higgs mode by tr-ARPES?

Noneq. DMFT calculation: Kemper et al., Phys. Rev. B 92, 224517 (2015).



- THz pump(?). Energy resolution ~ 1meV.
- We are waiting for the news!