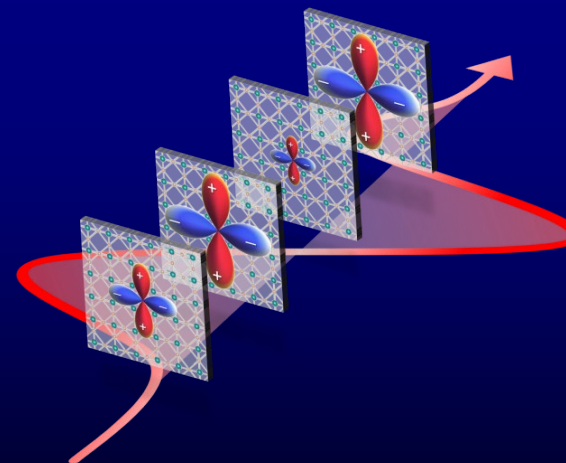
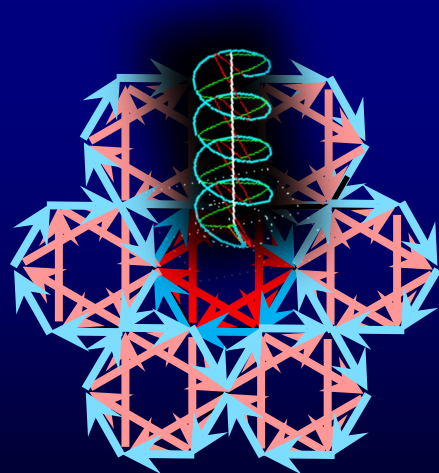


Floquet engineering and Higgs dynamics in superconductors

Hideo Aoki
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&
AIST, Tsukuba, Japan



FROM BEING TO BECOMING

TIME AND COMPLEXITY IN THE PHYSICAL SCIENCES

ILYA PRIGOGINE



1977

(Freeman, 1980)



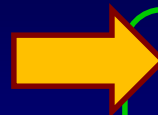
Preface: "I would like to have named it
Time, the Forgotten Dimension".

Today's talk

Many-body

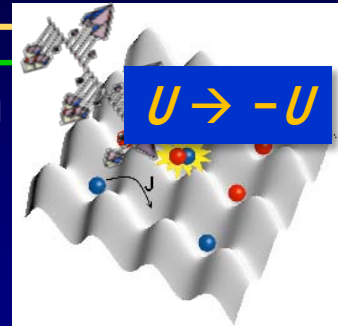
Floquet

Steady/transient

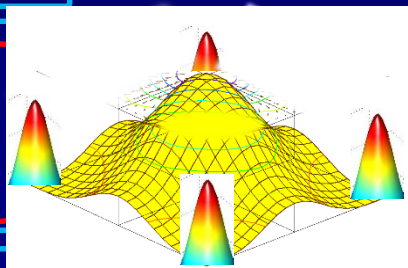


✓ Attraction-repulsion conversion

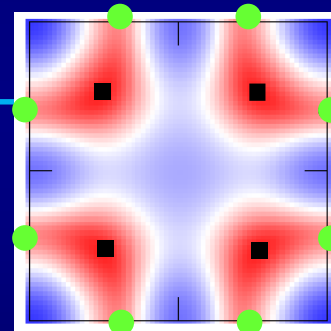
(Tsuji; Tsuji)



✓ η -pairing
(Kitamura)

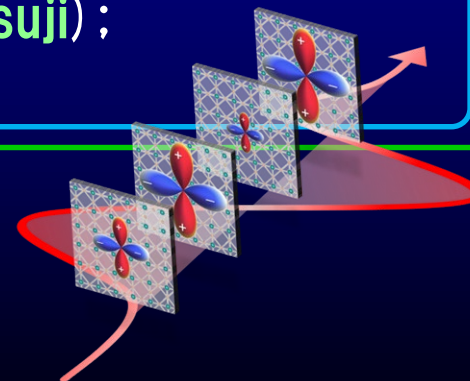


✓ k -dep relaxation
(Tsuji; Sayyad)



✓ Higgs dynamics:

- * phonon mechanism SC (Tsuji);
- * unconventional SC (Tsuji)



**Floquet engineering in
many-body (Hubbard) systems**

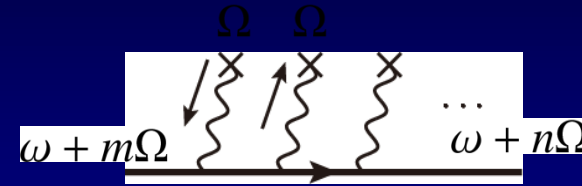
Floquet + DMFT

(Tsuji, Oka & Aoki, PRB 2008; PRL 2009)

- Ingredient 1: Floquet-Green function**

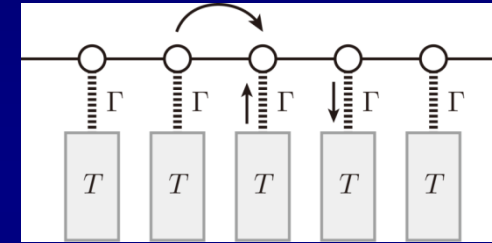
$$G(t + \mathcal{T}, t' + \mathcal{T}) = G(t, t') \quad (\text{Faisal, 1989})$$

$$\Rightarrow G(t, t') = \int_{-\Omega/2}^{\Omega/2} \frac{d\omega}{2\pi} \sum_{mn} e^{-i(\omega+m\Omega)t+i(\omega+n\Omega)t'} G_{mn}(\omega)$$

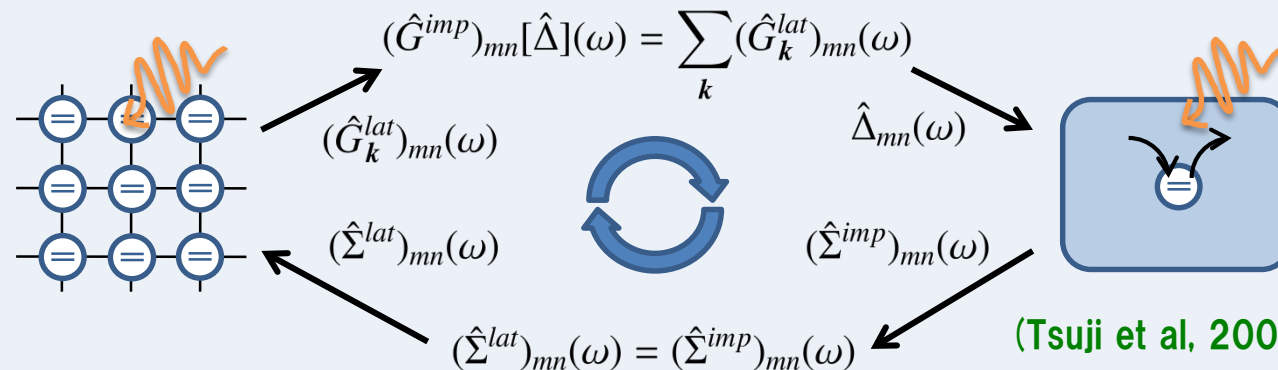


- Ingredient 2: Büttiker's heat-bath**

(Büttiker, 1985; 1986)



- Ingredient 3: Nonequilibrium DMFT formulated in a Floquet matrix form**



(Tsuji et al, 2008; 2009)

REVIEWS OF MODERN PHYSICS

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Nonequilibrium dynamical mean-field theory and its applications

Hideo Aoki, Naoto Tsuji, Martin Eckstein, Marcus Kollar, Takashi Oka, and Philipp Werner
Rev. Mod. Phys. 86, 779 (2014) – Published 24 June 2014

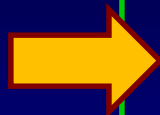
Advances in preparing and probing nonequilibrium systems, as exemplified by ultrafast spectroscopy and cold atoms in optical lattices, have enabled the systematic study of nonequilibrium phenomena in correlated lattice systems. This review provides a theoretical framework for describing these effects, with a focus on generalizations of dynamical mean-field methods to nonequilibrium situations and their applications.

Today's talk

Many-body

Floquet

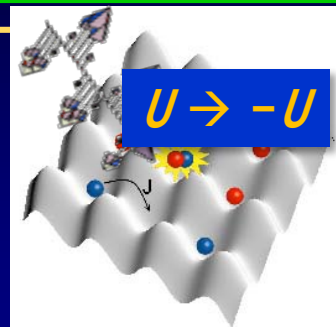
Steady/transient



Attraction-repulsion conversion

(Tsuji;

Tsuji)

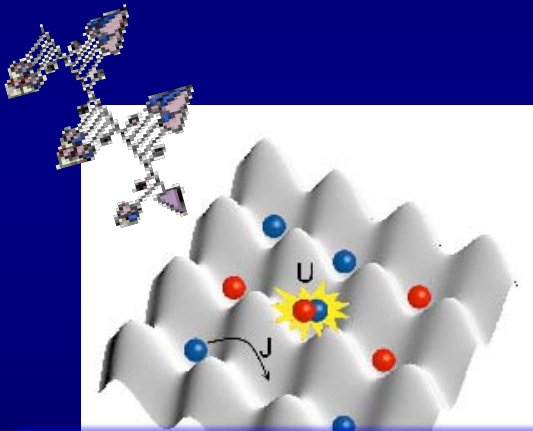


Repulsion–attraction conversion



(Tsuji, Oka, Werner & Aoki, PRL 2011)

Continuous laser:
 $U \rightarrow -U$



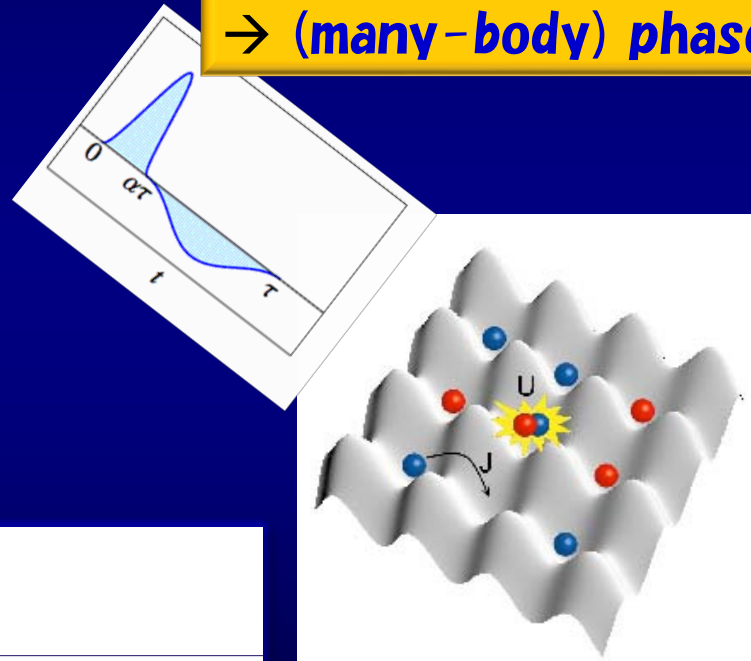
Physics

Viewpoint James K. Freericks

Changing repulsion into attraction with
the quantum Hippy Hippy Shake

(Tsuji, Oka, Aoki & Werner, PRB 2012)

Single monocycle pulse
 $\rightarrow$ (many-body) phase shift

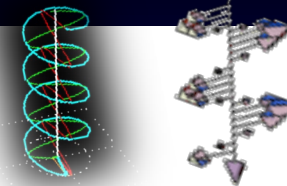


AC (t -periodic) modulation $\rightarrow$ Floquet theory

(for **arbitrarily strong AC**)

Gaston Floquet, 1883

– Temporal analog of Bloch theorem (**1928**) for spatial periodicity



$$i \frac{d}{dt} \Psi(t) = H(t) \Psi(t) \quad H(t + \tau) = H(t)$$

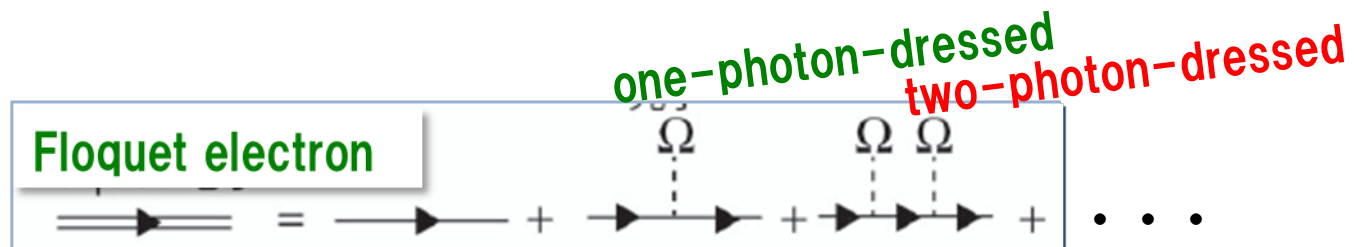
t -dep Schrodinger eqn



$$\begin{bmatrix} \ddots & \ddots & \vdots & \vdots & \ddots \\ \ddots & H_0 - \omega & H_{-1} & H_{-2} & \cdots \\ \cdots & H_1 & H_0 & H_{-1} & \cdots \\ \cdots & H_2 & H_1 & H_0 + \omega & \ddots \\ \ddots & \vdots & \vdots & \ddots & \ddots \end{bmatrix} \mathbf{u} = \varepsilon \mathbf{u}$$

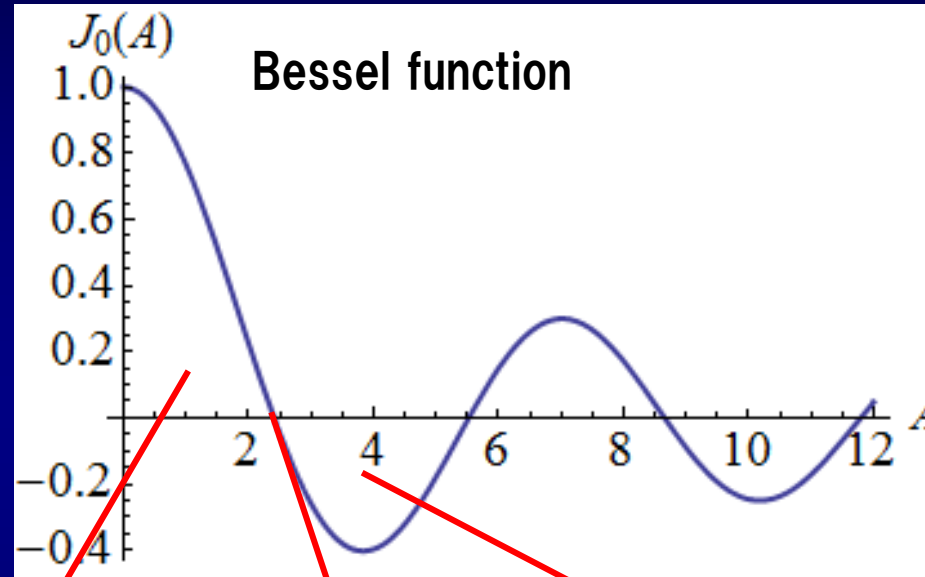
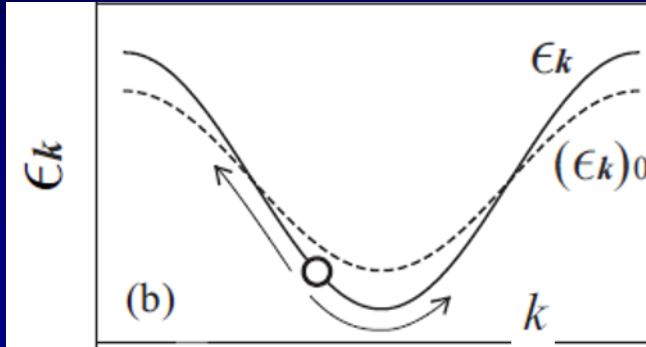
matrix eqn

Floquet electron

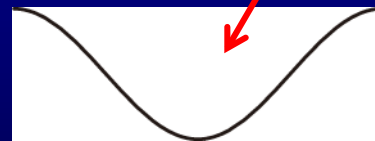


Floquet formalism for Bloch particles in ac fields

[Shirley, PR 1965; Sambe, PRA 1973;
analytically shown for noninteracting case
(Tsuji, Oka & Aoki, PRB, 2008)]



ude / Ω



band narrowing



dynamical localization

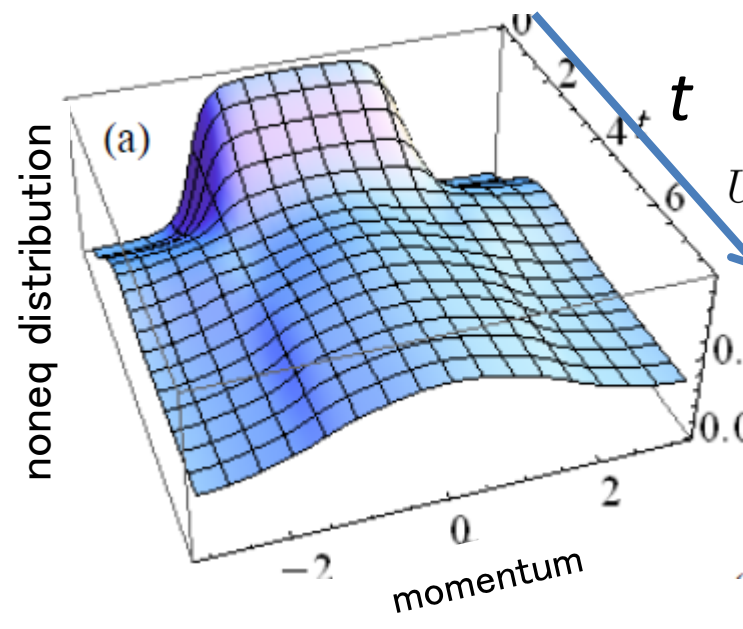
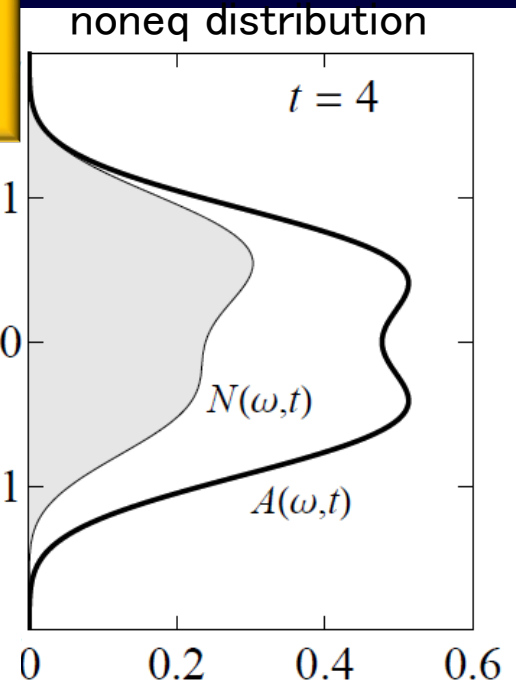
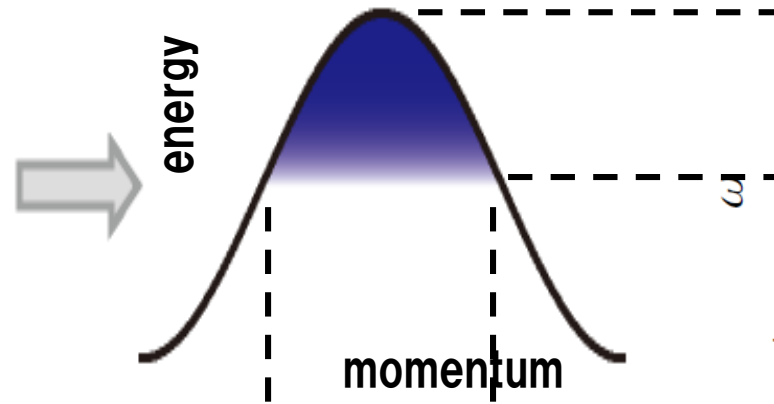
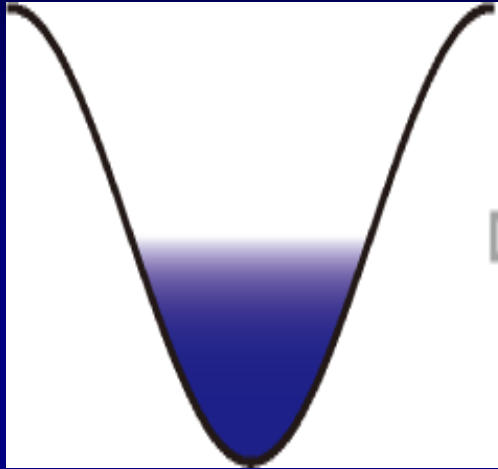


band flipping

Population inversion

(Tsuji, Oka, Werner & Aoki, PRL 2011)

Negative T , Positive U
 $\leftrightarrow$ Positive T , Negative U

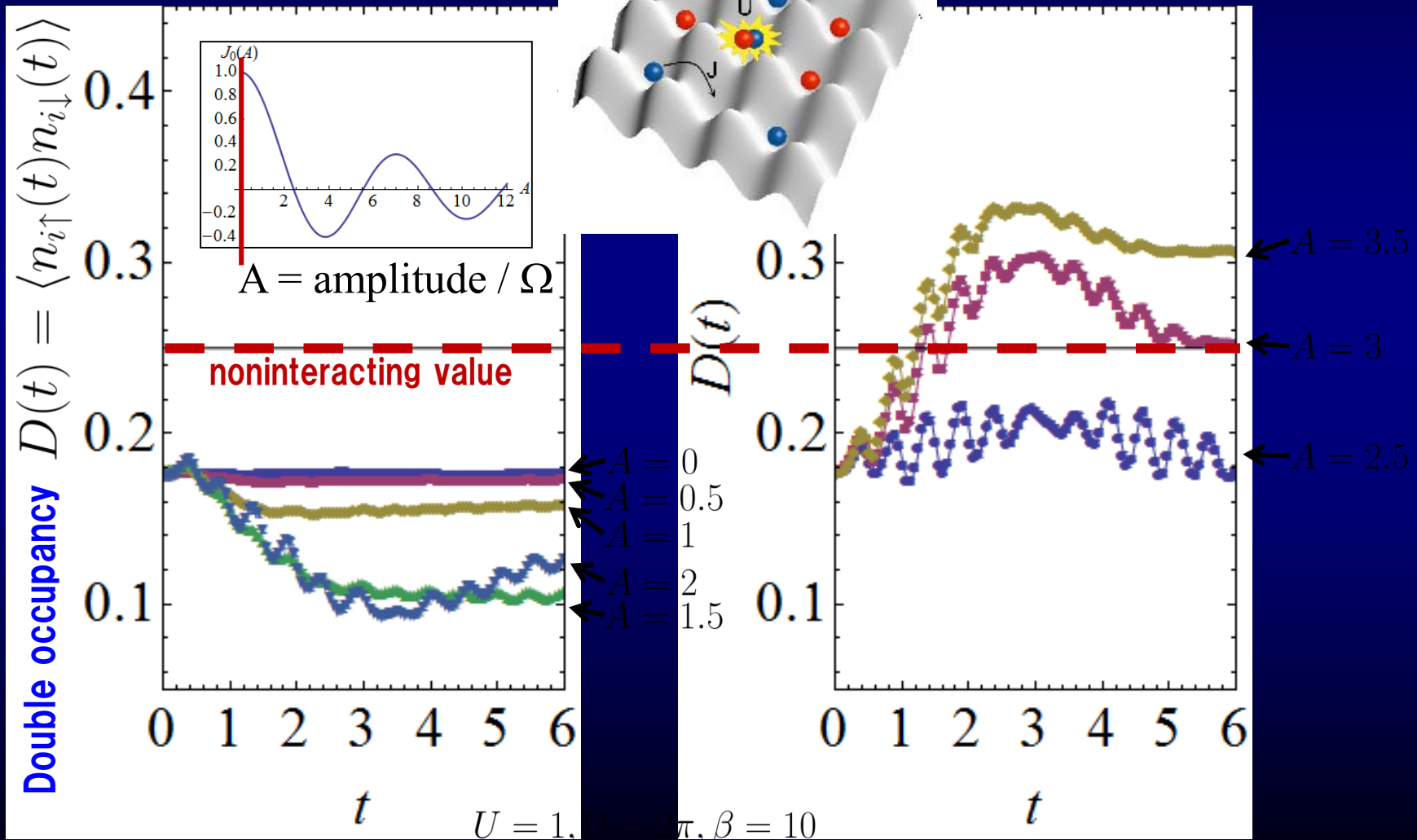
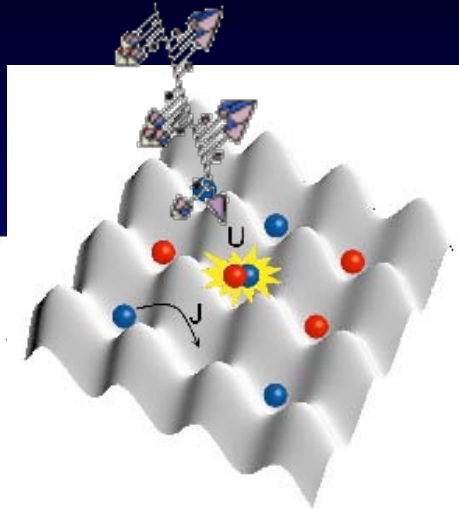


$$U = 1, \Omega = 2\pi, A = 4, \beta = 10$$

Noneq-induced repulsion-attraction conversion

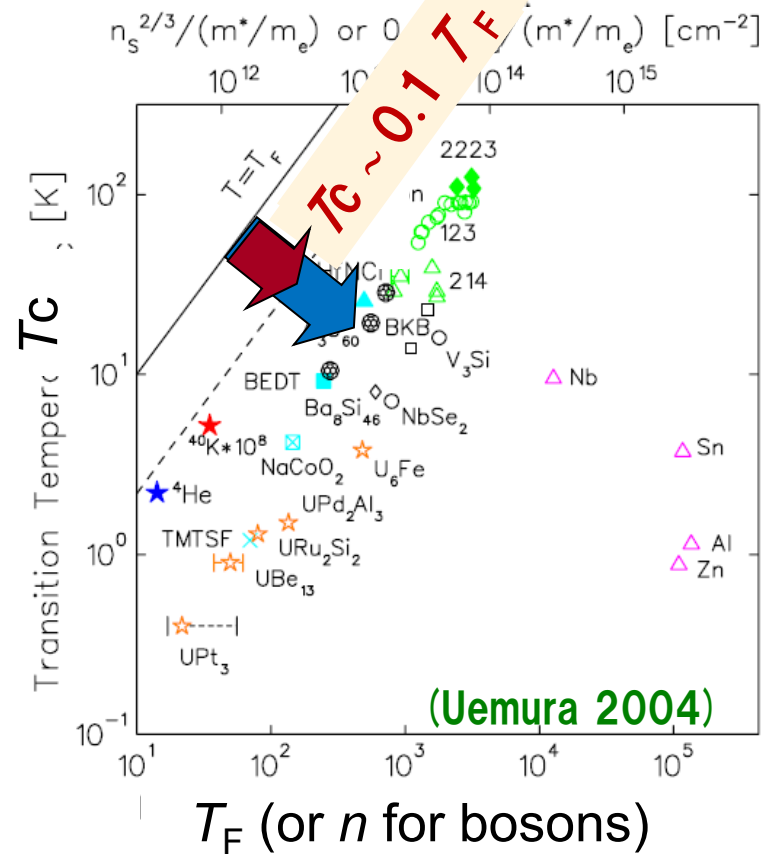
t -dep DMFT for
Hubbard model

(Tsuji et al, PRL 2011)

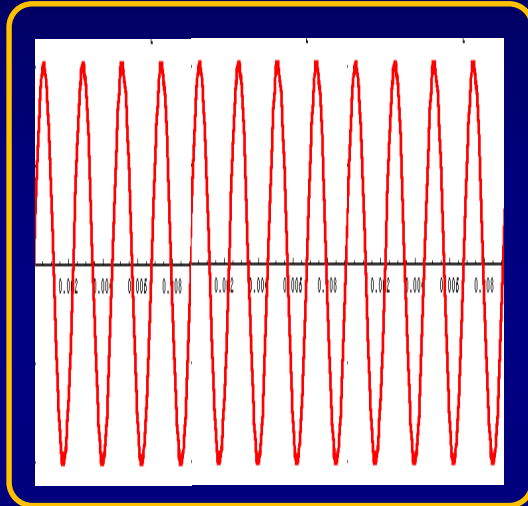


- Repulsive: $T_c \sim 0.01 t$

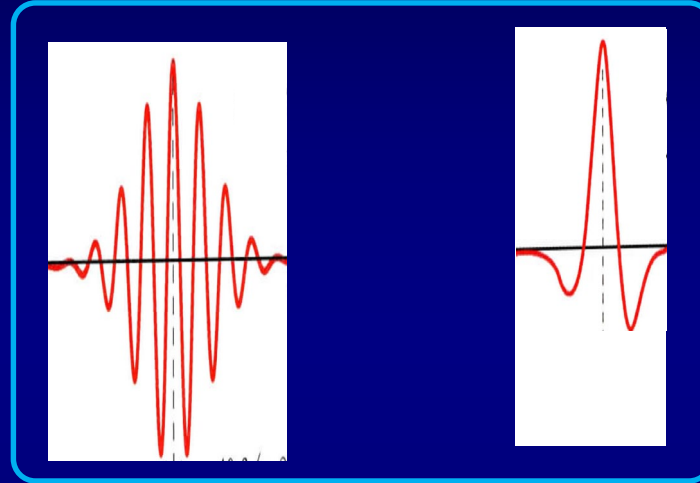
Attractive: $T_c \sim 0.1 t$



Steady/transient



continuous



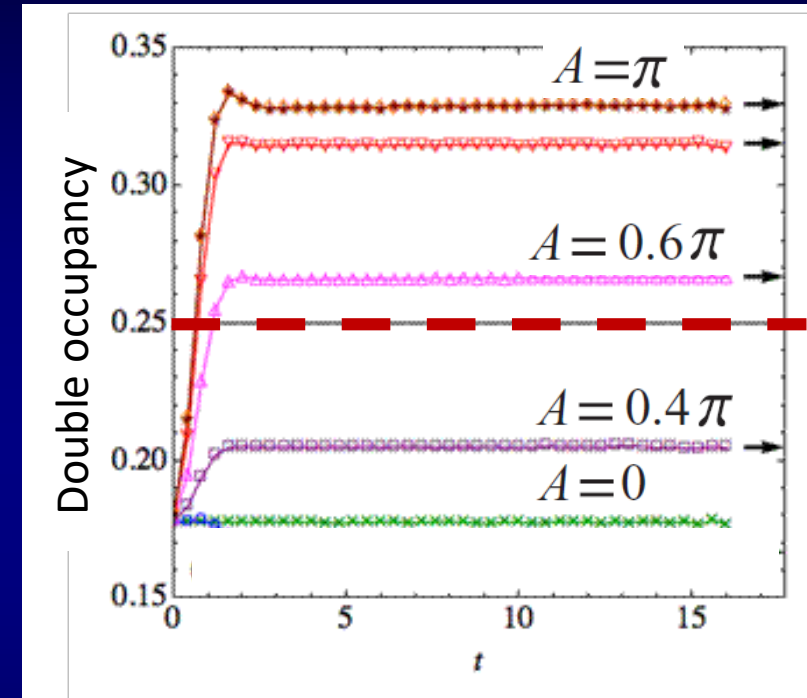
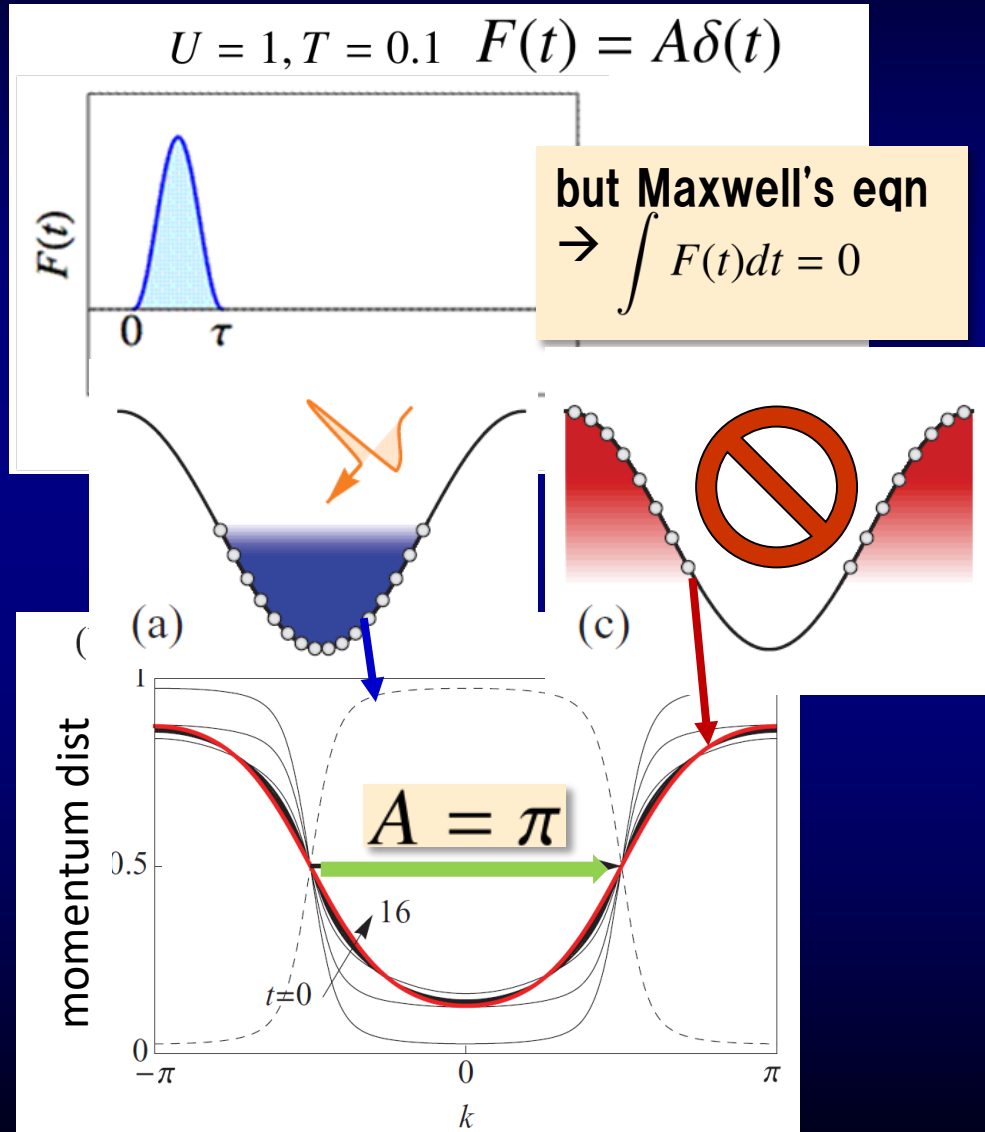
pulse

mono- or sub-cycle

A half-cycle pulse suffices

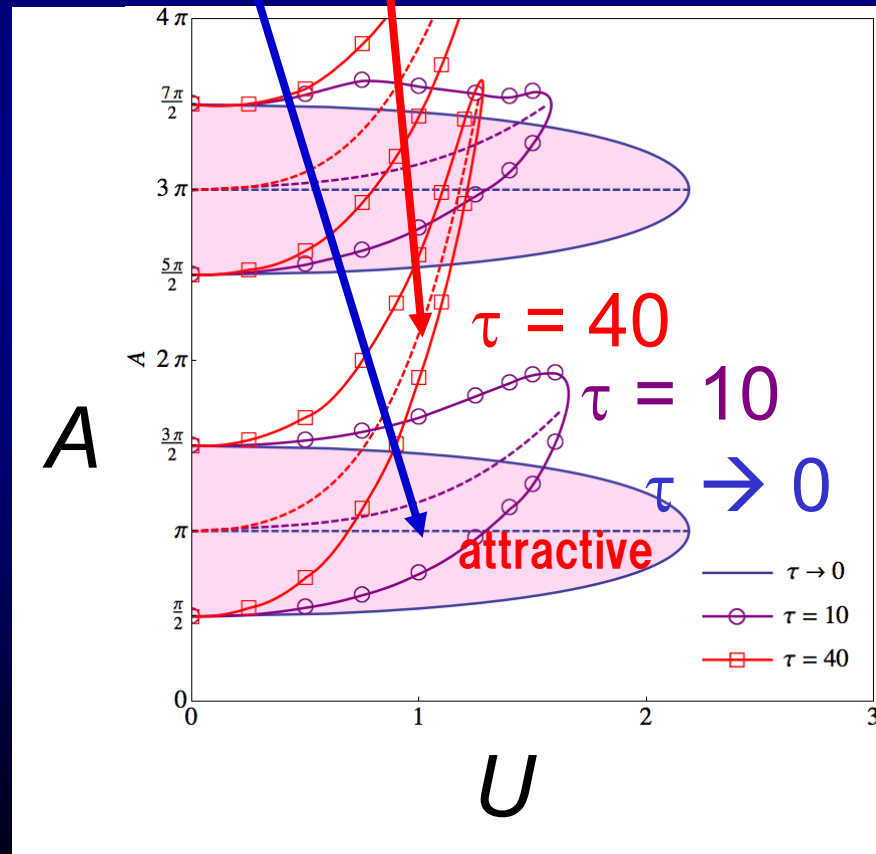
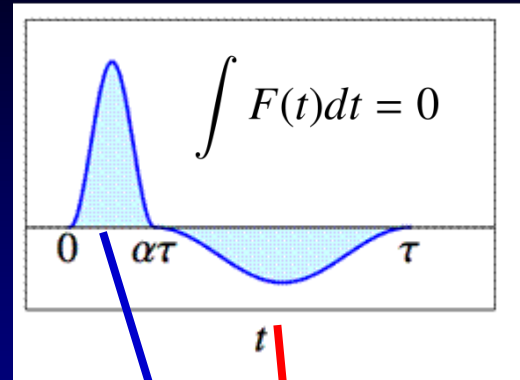
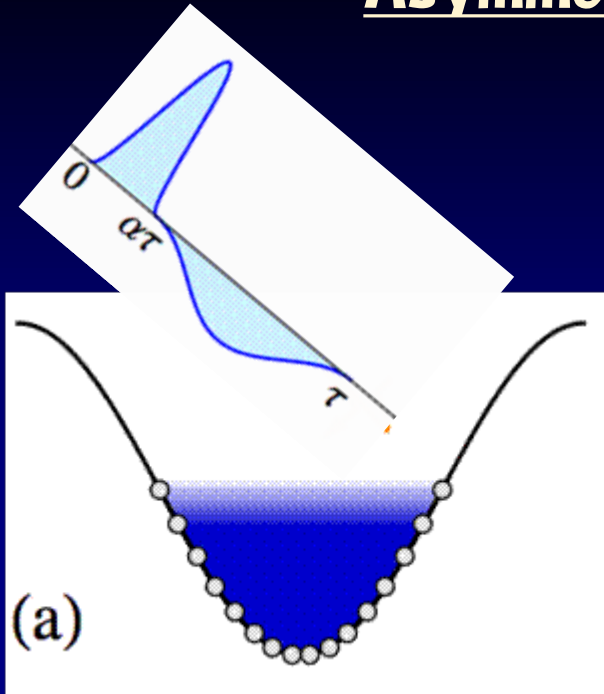
(Hubbard model, t -dep DMFT)

(Tsuji et al, PRB 2012)



Asymmetric monopulse realizes $\tau < 0$

(Tsuji et al, PRB 2012)



Experimental feasibility

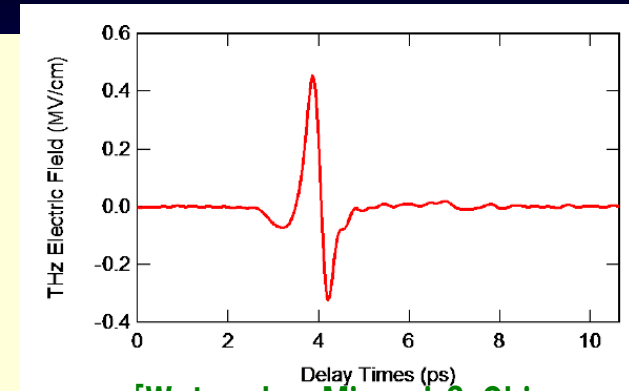
* Required strength of the laser field:

$$E \sim 1 \text{ MV/cm}$$

$$\tau \sim 1 \text{ ps}$$

$$\text{lattice const } a = 1 \text{ \AA}$$

$$\rightarrow \int F(t)dt \sim eE\tau a/\hbar \sim 10$$



[Watanabe, Minami & Shimano,
Optics Express (2011)]

Floquet + Hubbard $\rightarrow$ various energy scales

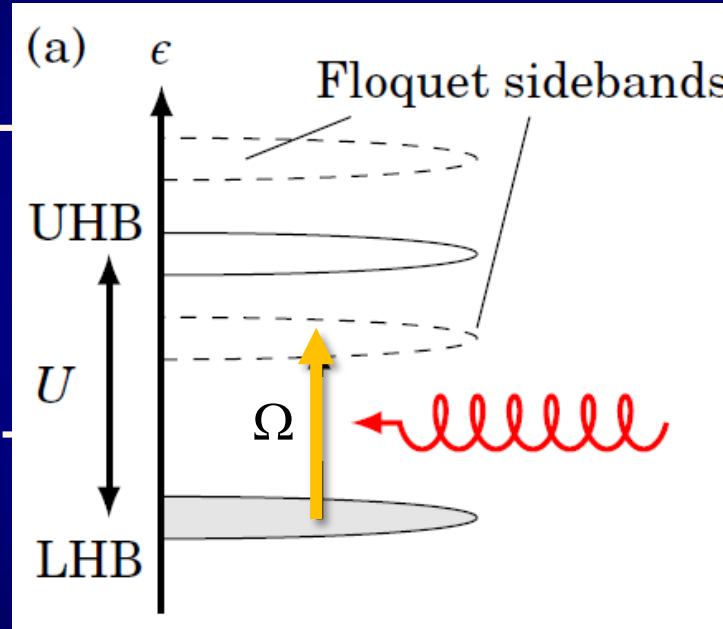
$$\frac{\Omega}{t} \sim \frac{U}{t}$$

Ω on-resonant with Hubbard

(Kitamura, Oka & Aoki, PRB 2017)

$$\frac{\Omega}{t} \ll \frac{U}{t}$$

$$\frac{\Omega}{t} \gg \frac{U}{t}$$

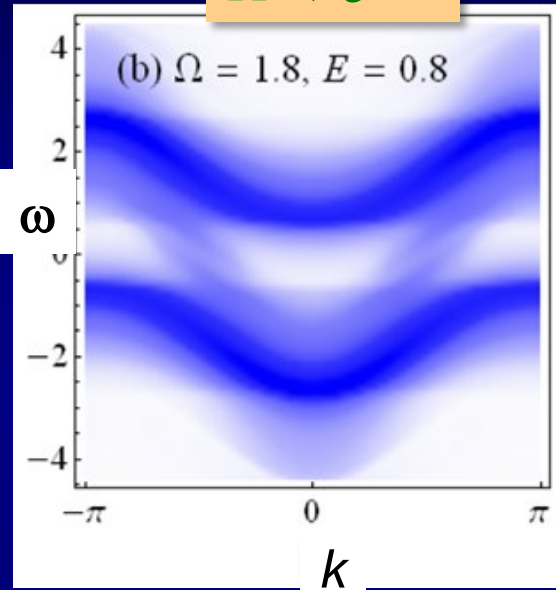


Floquet sidebands for $\Omega \sim U$

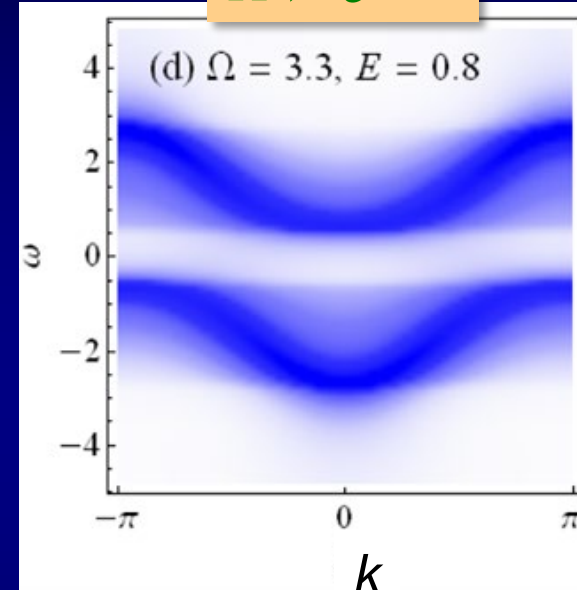
Falicov–Kimball model with Floquet–DMFT (Tsuji *et al*, PRB 2008)

- Momentum-resolved spectral function $\tilde{A}_k(\omega)$

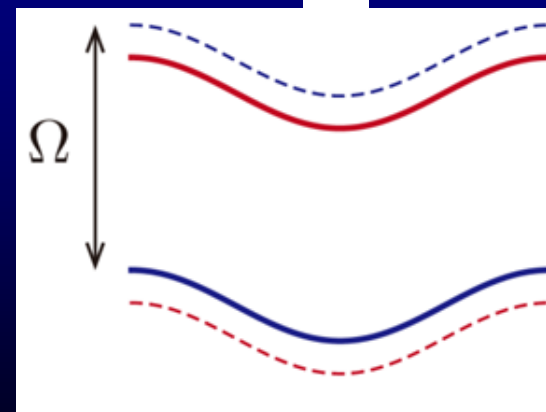
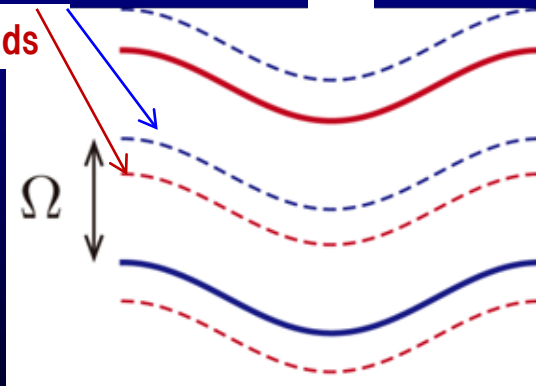
$\Omega < U$



$\Omega > U$



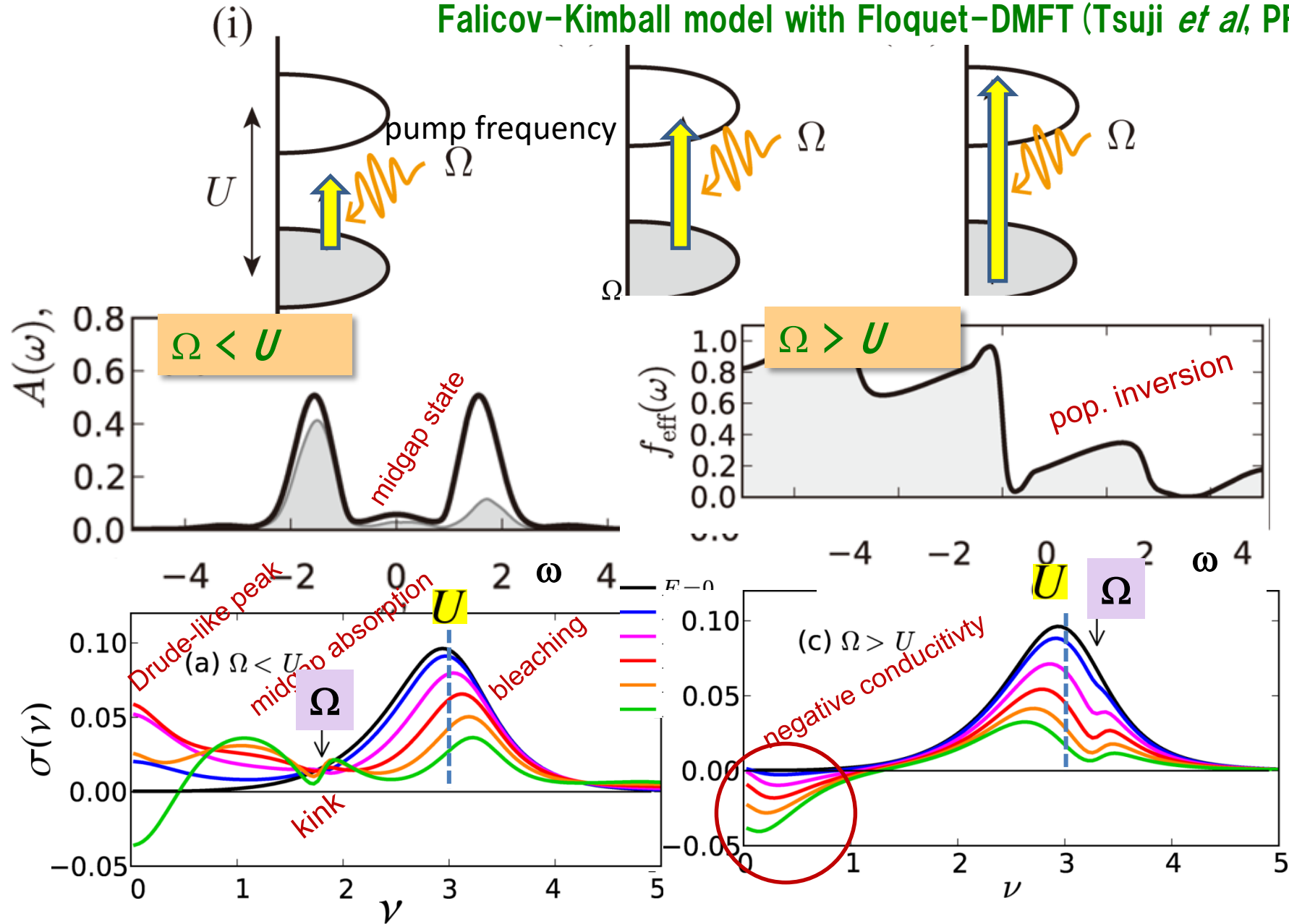
Floquet sidebands



$$U = 3, \Gamma = 0.05, T = 0.05$$

A remark: Floquet optical conductivity

Falicov-Kimball model with Floquet-DMFT (Tsuji *et al*, PRL 2009)



Today's talk

Many-body

Floquet

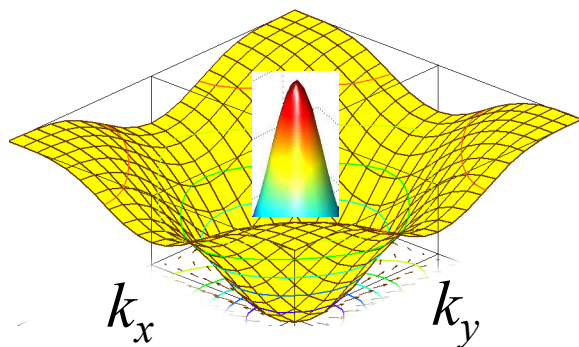
Steady/transient

✓ Attraction–repulsion conversion
(Tsuji; Tsuji)

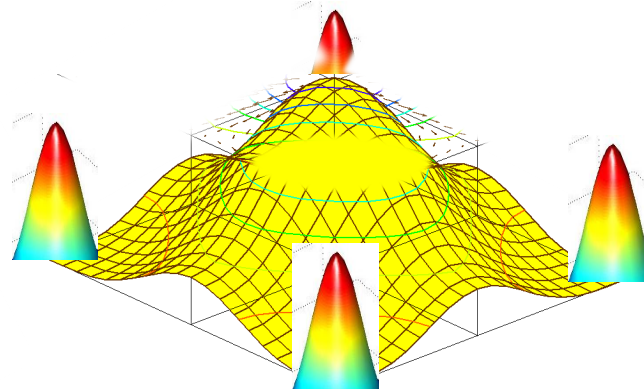
✓ η -pairing
(Kitamura)

Nonequilibrium $\rightarrow$ long sought-after η -pairing (C.N. Yang 1989)

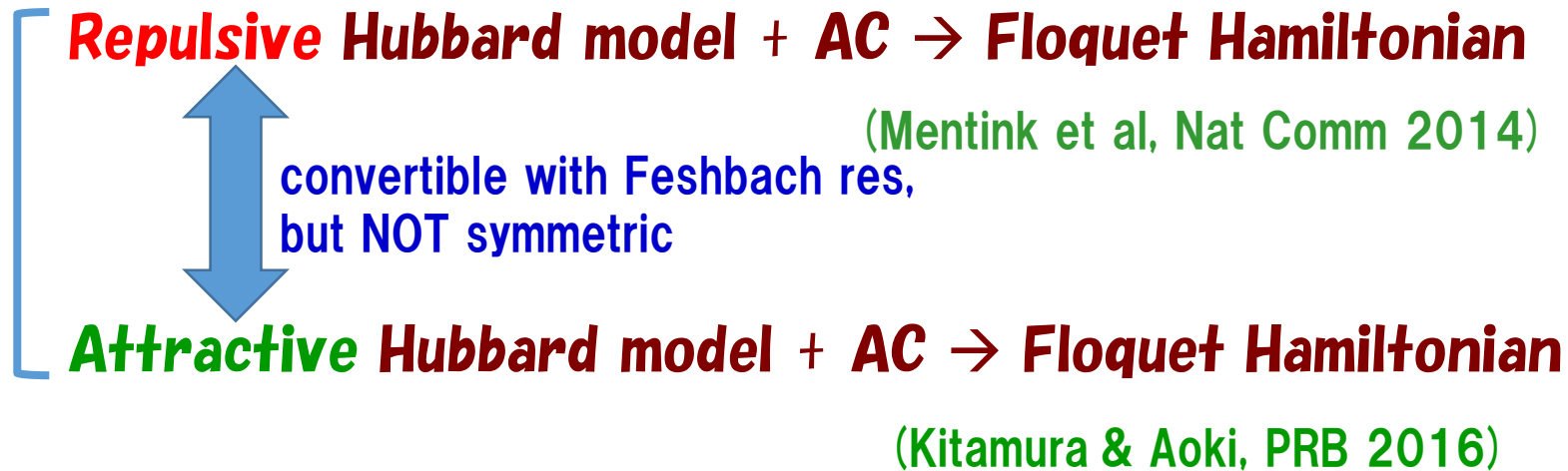
Pair hopping > 0



Pair hopping < 0



Strong-coupling



In equil: Repulsion-attraction

" η SU(2)" trsfm

Nonequil ?

$$\hat{c}_j^\dagger \rightarrow (-1)^j \hat{c}_{j\uparrow}$$

$$\hat{S}_j \rightarrow \hat{\eta}_j$$

$$\text{AF}(\vec{Z}) \rightarrow \text{CDW}$$

$$\text{AF}(\text{XY}) \rightarrow \text{BCS}$$

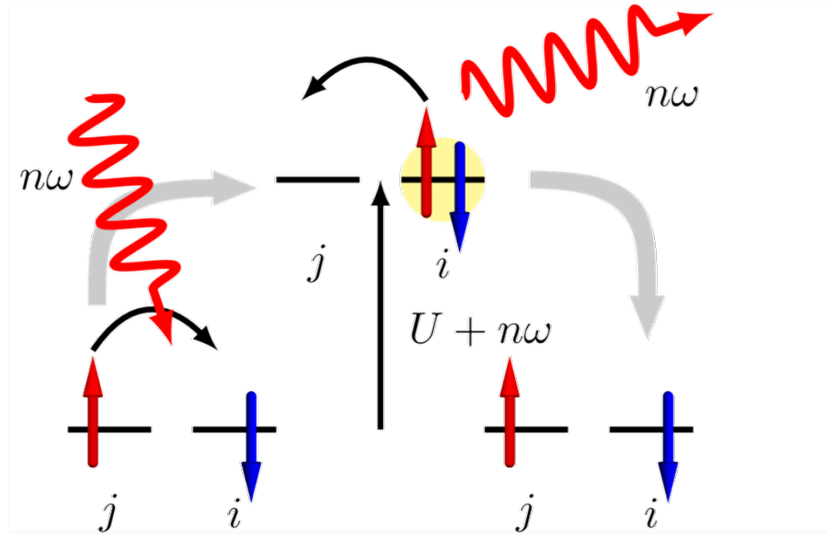
$$\text{F}(\text{XY}) \rightarrow \eta\text{-pairing}$$

$$\hat{\eta}_j^x + i\hat{\eta}_j^y = (-1)^j \hat{c}_{j\uparrow} \hat{c}_{j\downarrow}, \quad \hat{\eta}_j^z = \frac{1}{2}(1 - \hat{n}_{j\uparrow} - \hat{n}_{j\downarrow})$$

Strong-coupling

Repulsive Hubbard + AC → Floquet Hamiltonian

(Mentink et al, Nat Comm 2014)



$$H_{\text{eff}} = -J_{\text{eff}} \sum_{ij}^{\text{n.n.}} \mathbf{S}_i \cdot \mathbf{S}_j$$

$$J_{\text{eff}} = 2 \sum_{n=-\infty}^{\infty} \frac{\mathcal{J}_n(A)^2}{n\omega - U}$$

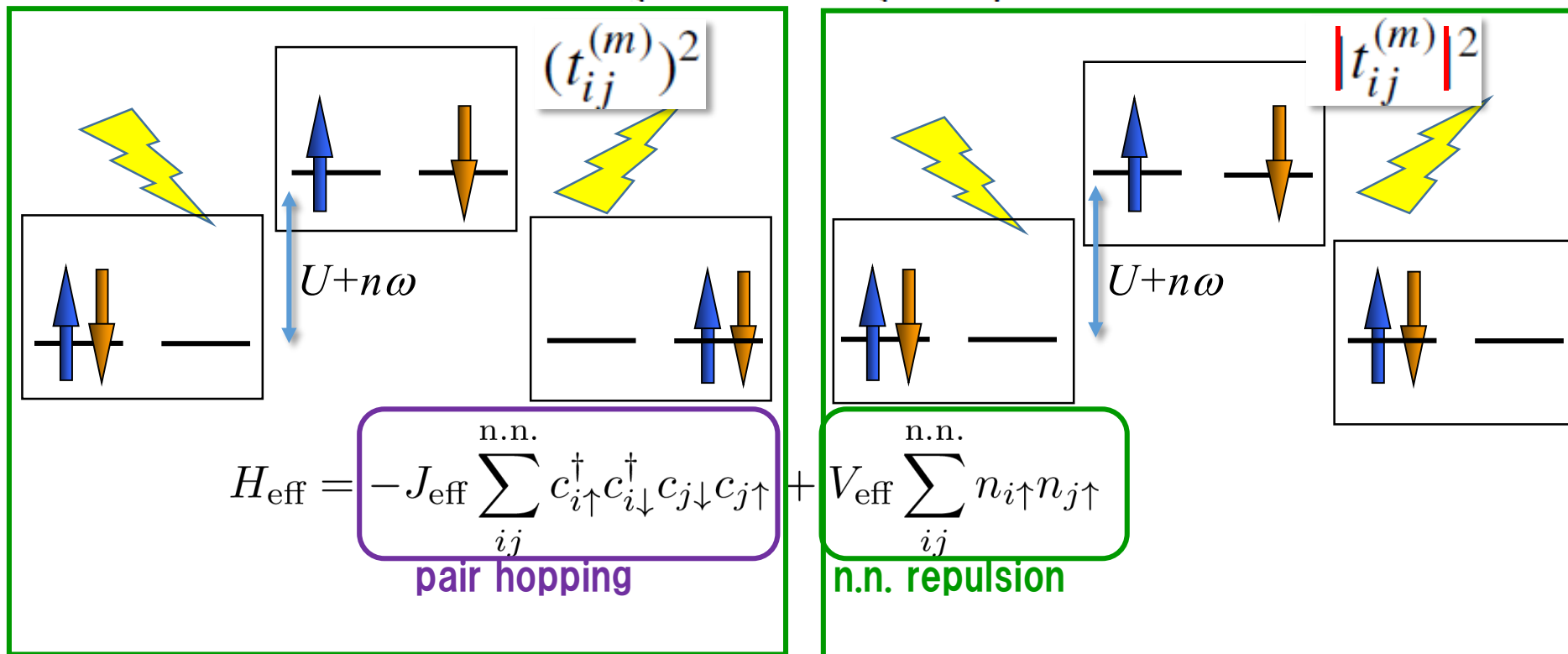
Strong-coupling

Attractive Hubbard + AC → Floquet Hamiltonian

— η SU(2) (repulsion–attraction symm) destroyed

(Kitamura & Aoki, PRB 2016)

$$t_{ij}^{(m)} = i^m \mathcal{J}_m(A \cdot R_{ji})$$



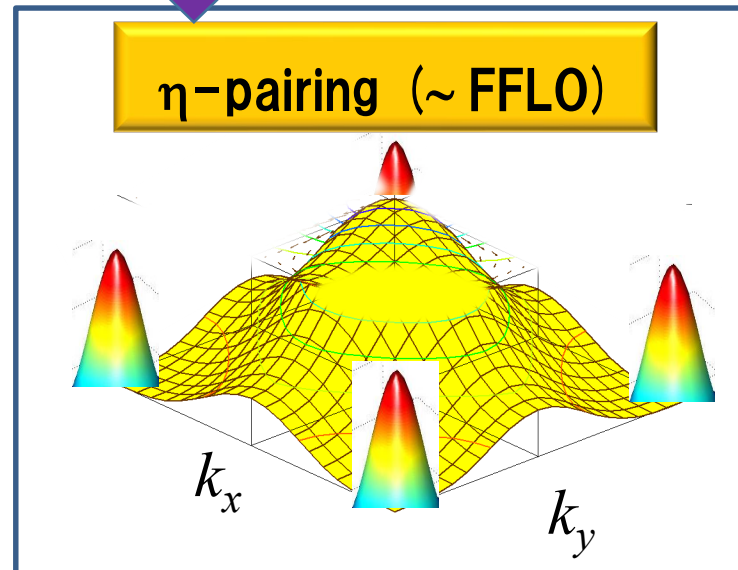
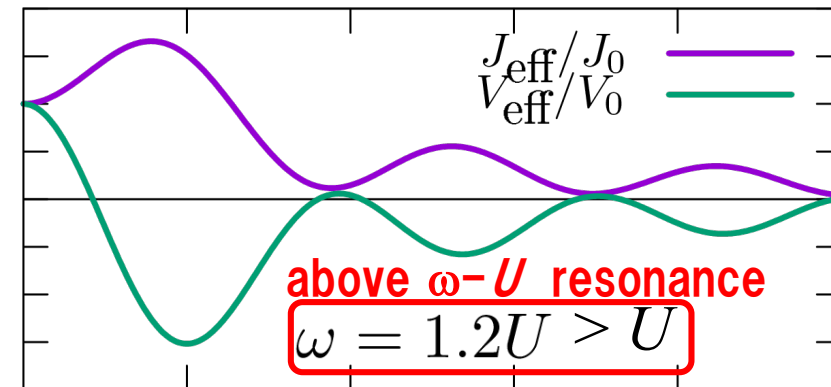
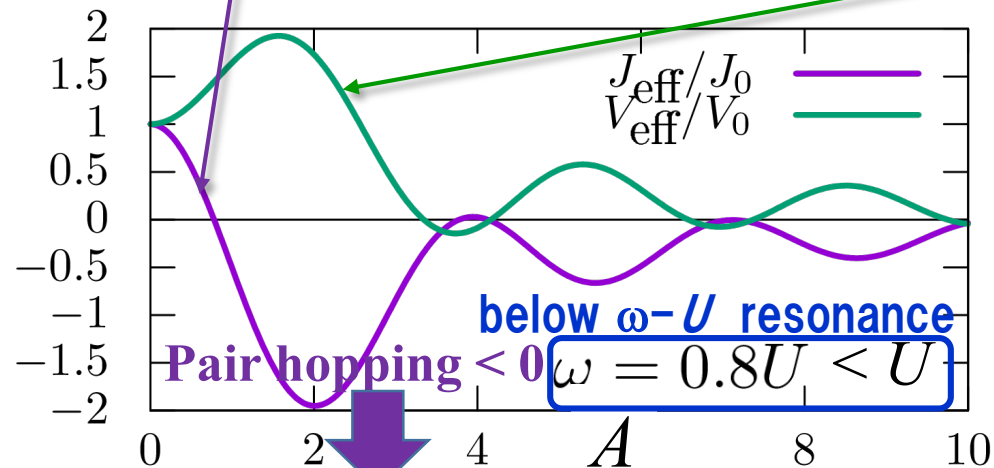
Strong-coupling **Attractive** Hubbard model + AC

pair hopping

$$J_{\text{eff}} = -2 \sum_{n=-\infty}^{\infty} \frac{(-)^n \mathcal{J}_n(A)^2}{n\omega - U}$$

n.n. repulsion

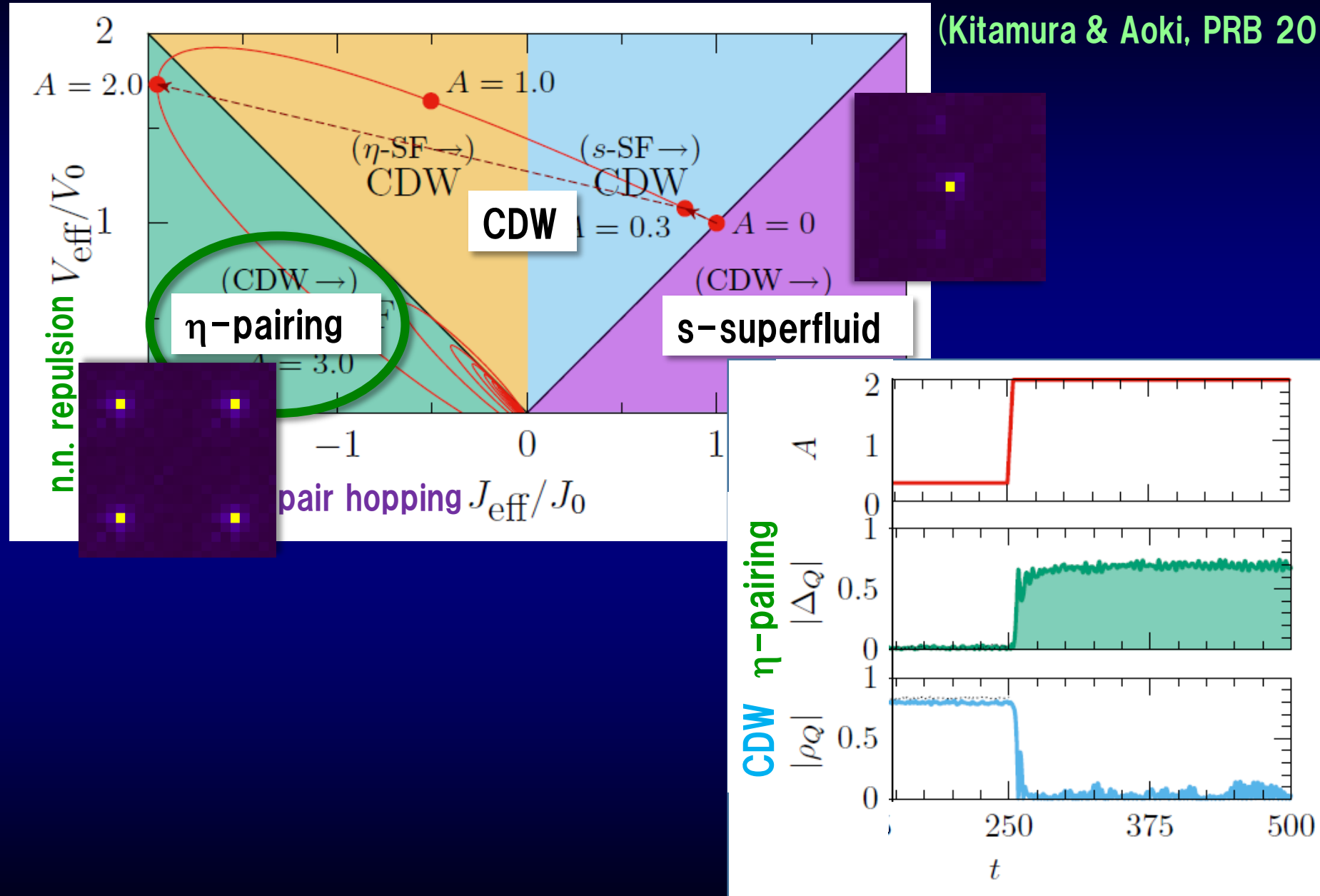
$$V_{\text{eff}} = -2 \sum_{n=-\infty}^{\infty} \frac{\mathcal{J}_n(A)^2}{n\omega - U}$$



(Kitamura & Aoki, PRB 2016)

Floquet "phase diagram"

(Kitamura & Aoki, PRB 2016)

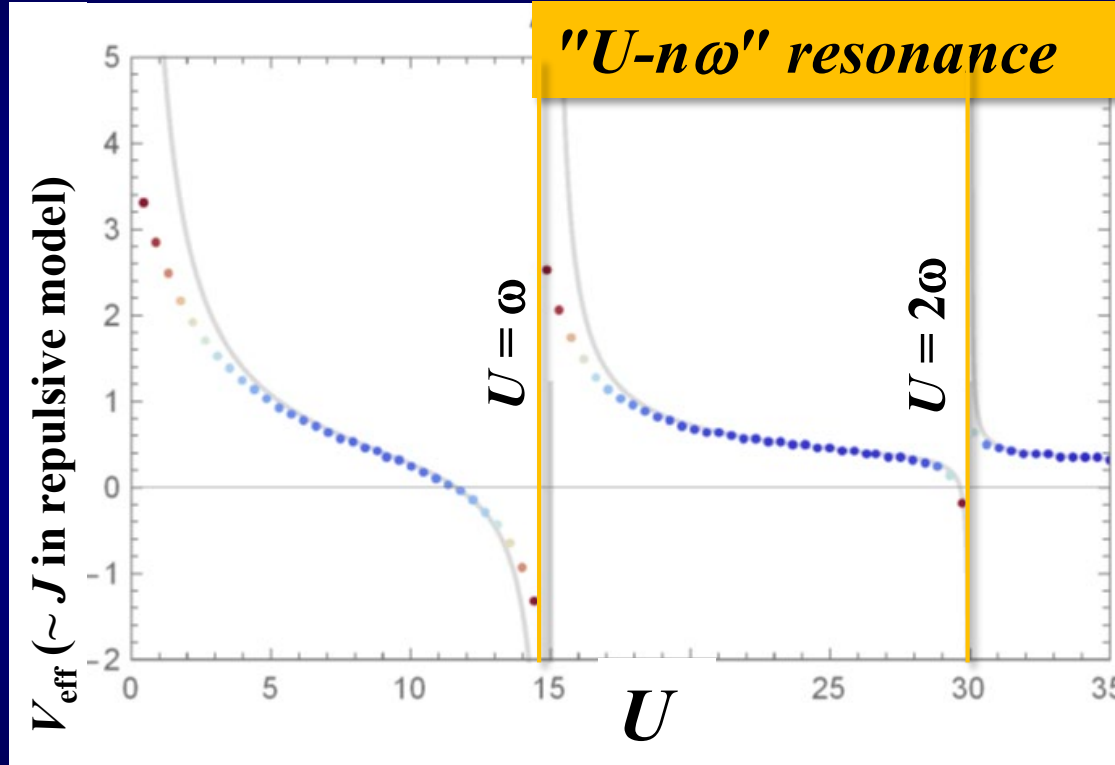


Floquet states on-resonant with Hubbard U

(Kitamura, Oka & Aoki, PRB 2016)

$$V_{\text{eff}} = -2 \sum_{n=-\infty}^{\infty} \frac{\mathcal{J}_n(A)^2}{n\omega - U}$$

(See also: cold-atom experiments,
Desbuquois et al, PRA 2017;
Görg et al, Nature 2018;
Theory: Herrmann et al,
Europhys Lett 2018)



Today's talk: Superconductors in nonequil

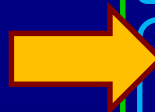
Many-body

Steady/transient

✓ Attraction–repulsion conv

(Tsuji; Tsuji)

✓ η -pairing
(Kitamura)

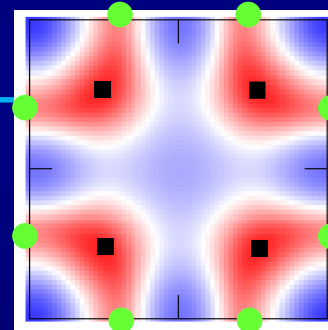


✓ k -dep relaxation
(Tsuji; Sayyad)

✓ Higgs dynamics:

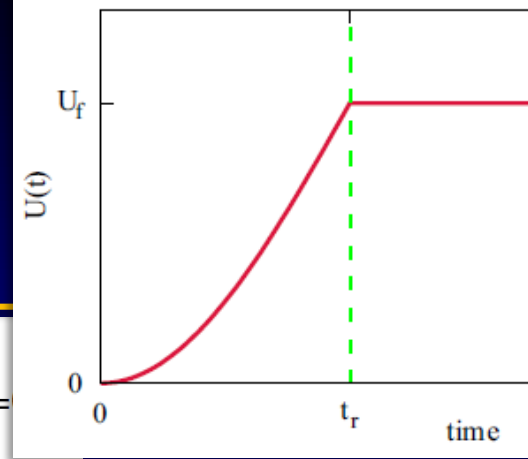
* phonon mechanism SC (Tsuji);

* unconventional SC (Tsuji)

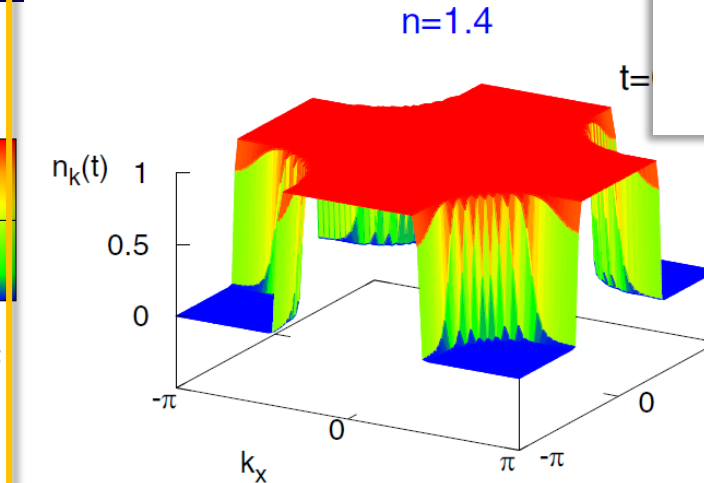
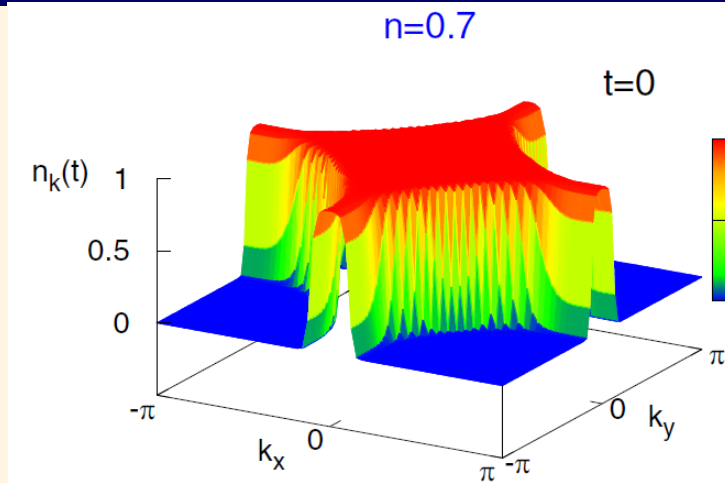


Momentum-dependent noneq states $\leftarrow$ Noneq DMFT+FLEX
 (Sayyad, Tsuji, Vaezi, Capone, Eckstein & Aoki, PRB 2019)

Repulsive Hubbard model on square lattice ($U = 0 \rightarrow 3$)
 hole-doped el-doped

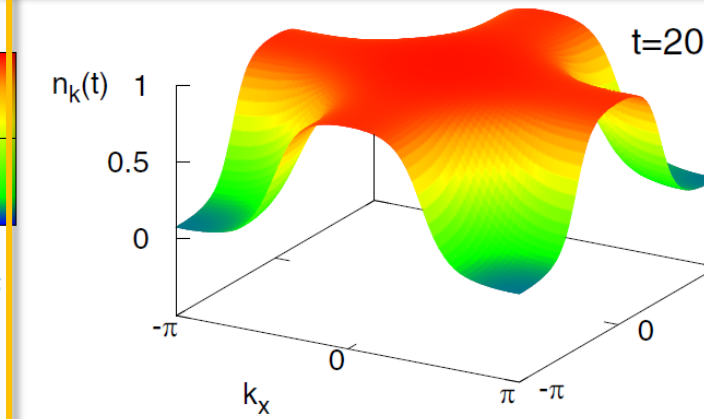
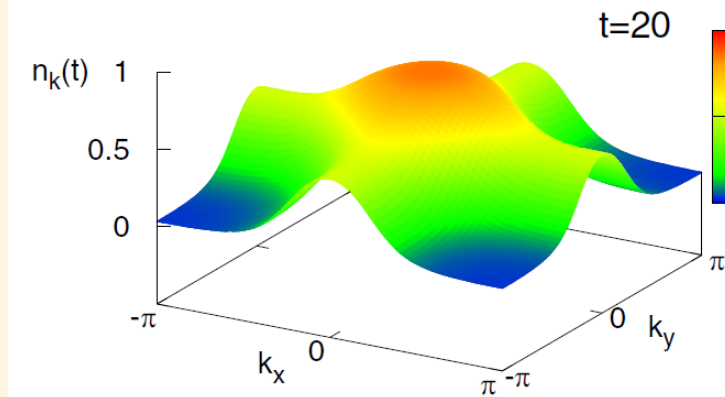


before
quench



$t' = -0.2t$, $t'' = 0.16t$
 (cuprate)

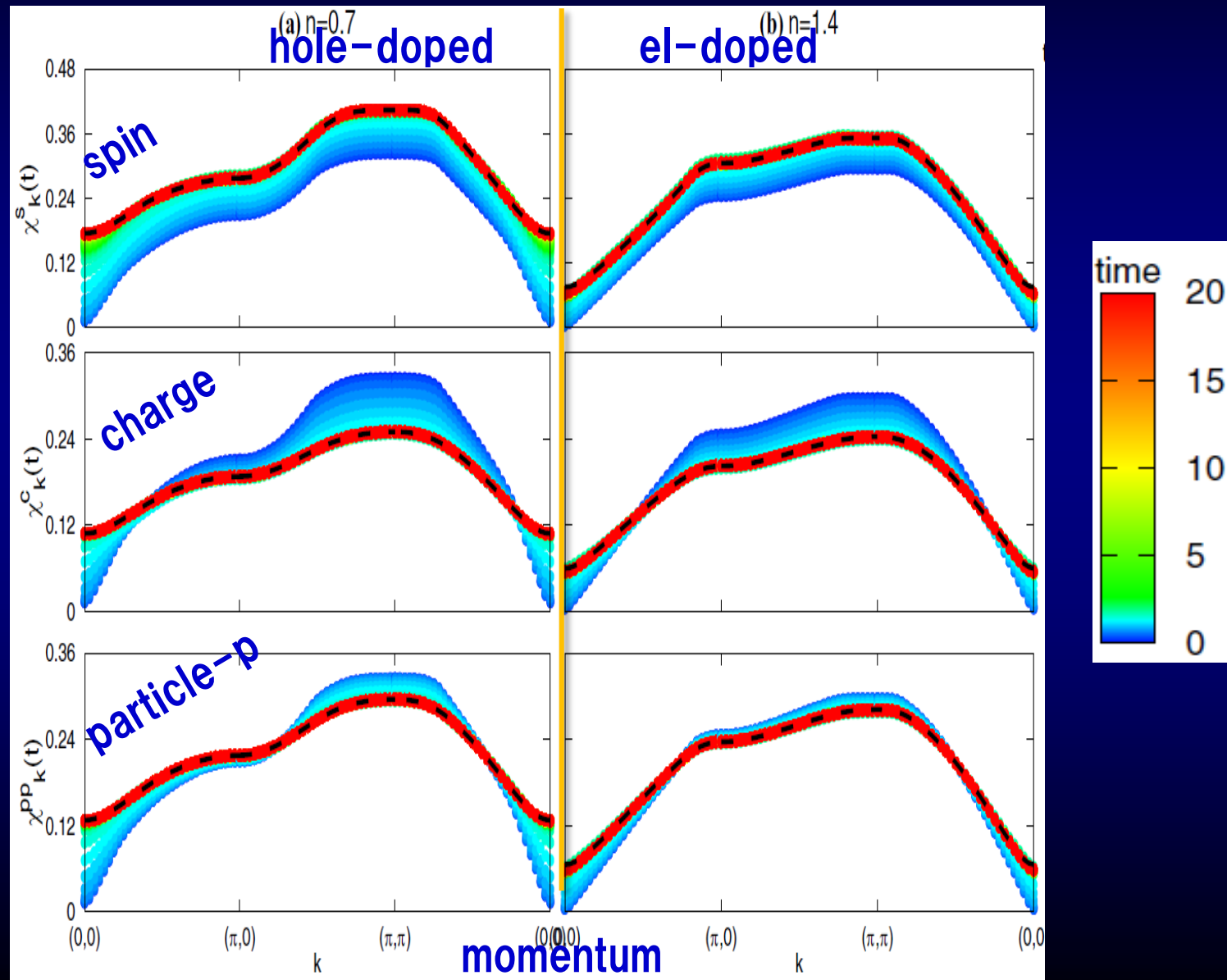
after
quench



$\beta = 30$

Momentum-dep nonequilibrium states

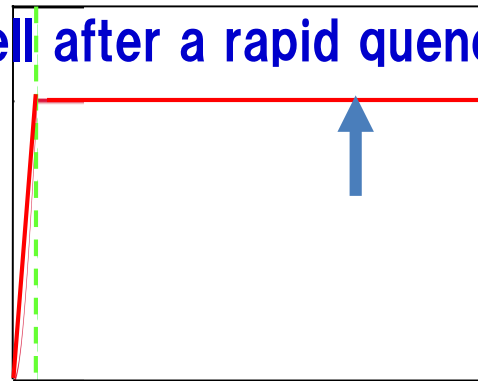
Noneq DMFT+FLEX (Sayyad et al, PRB 2019)



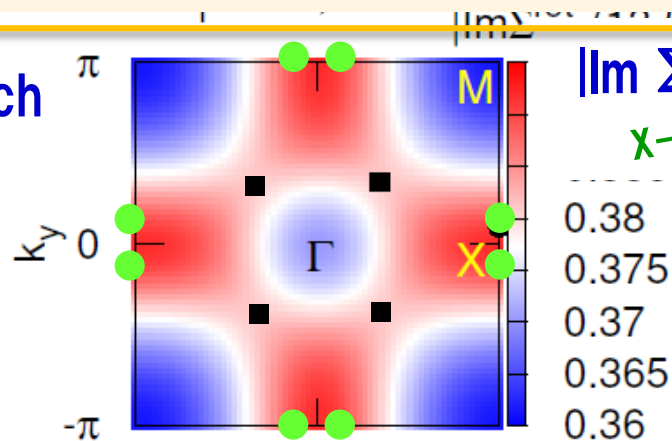
Momentum-dependent noneq states $\leftarrow \text{Im } \Sigma \sim 1/\tau$

[Sayyad et al, PRB 2019 $\leftarrow$ consistent with noneq DCA (Tsuji et al, PRB 2014)]

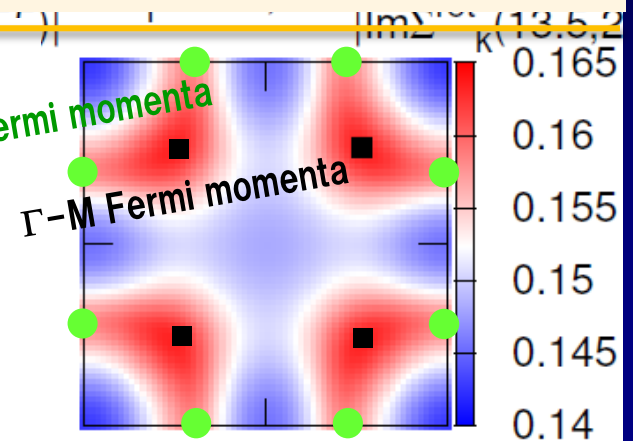
well after a rapid quench



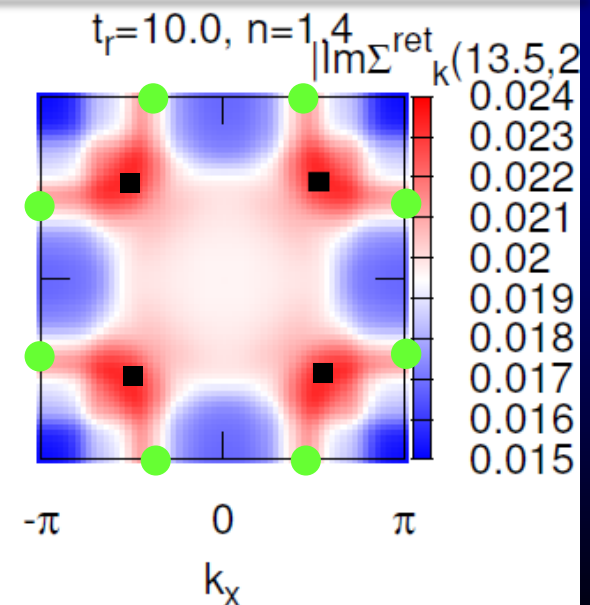
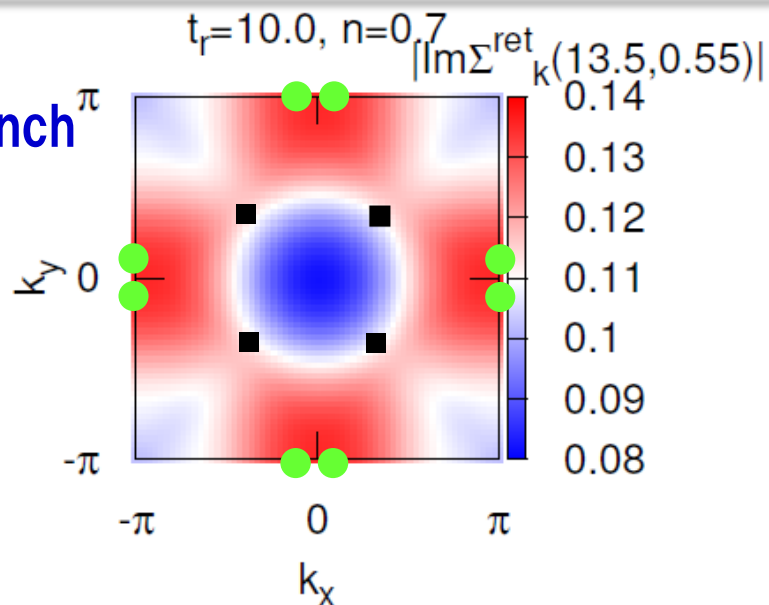
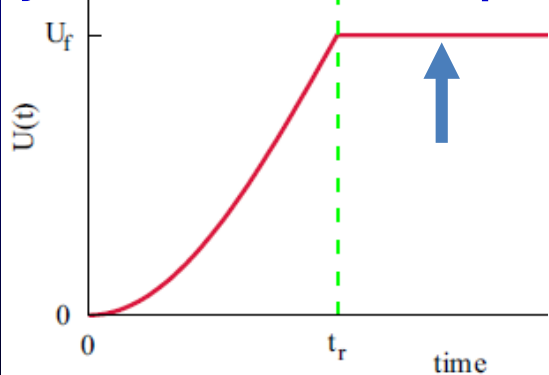
hole-doped



el-doped



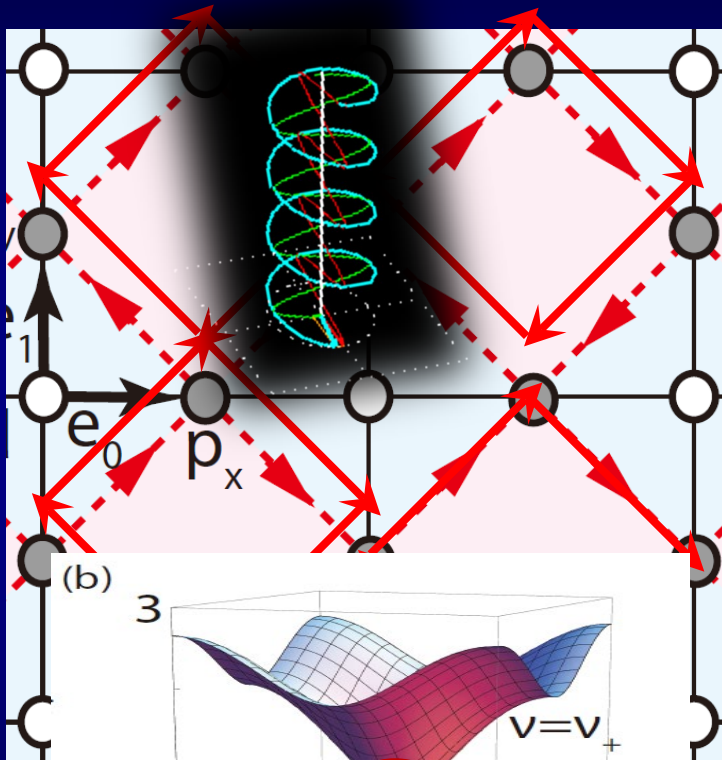
just after a slower quench



Mom-dep manipulation in Floquet

Lieb lattice

(Mikami, ..., Aoki, PRB 2016)

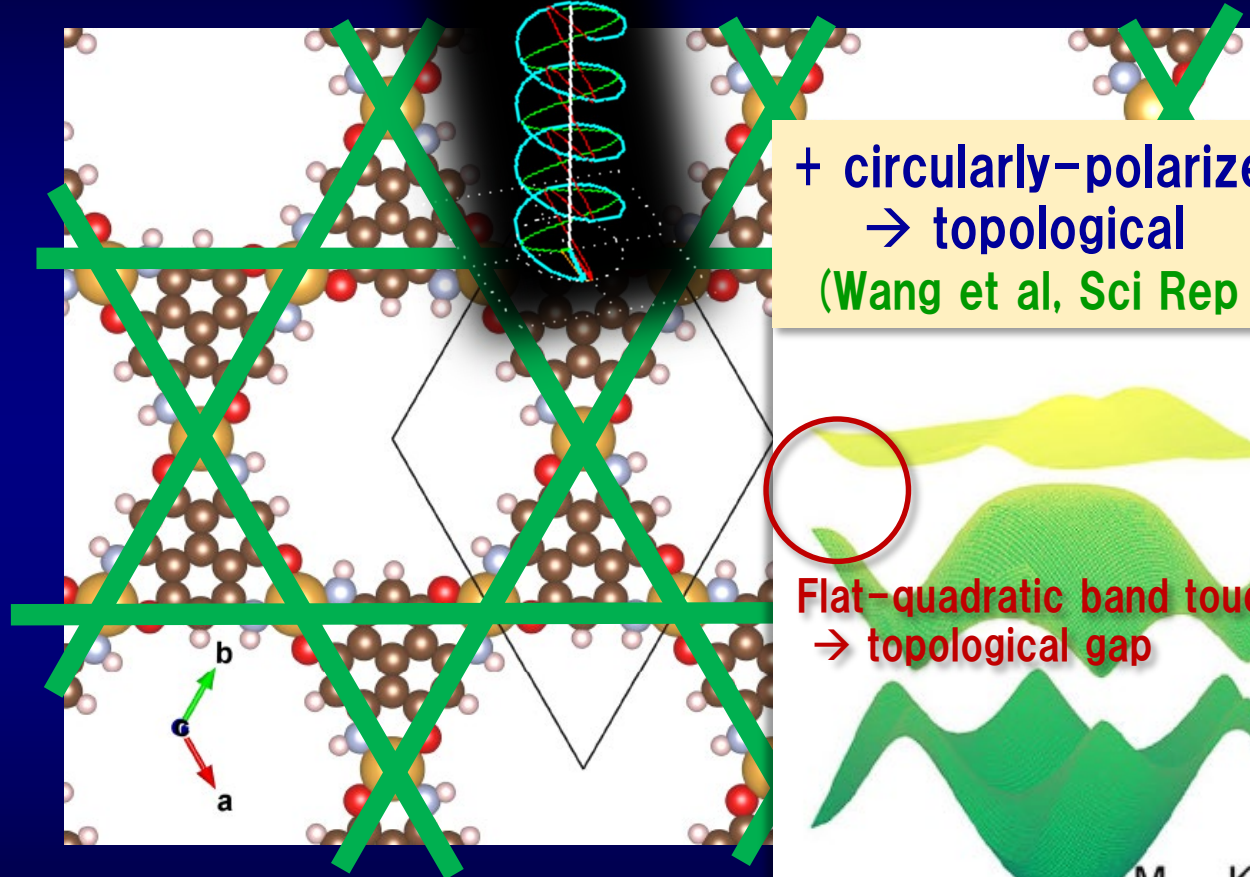


Flat band + Dirac cone
→ topological gap

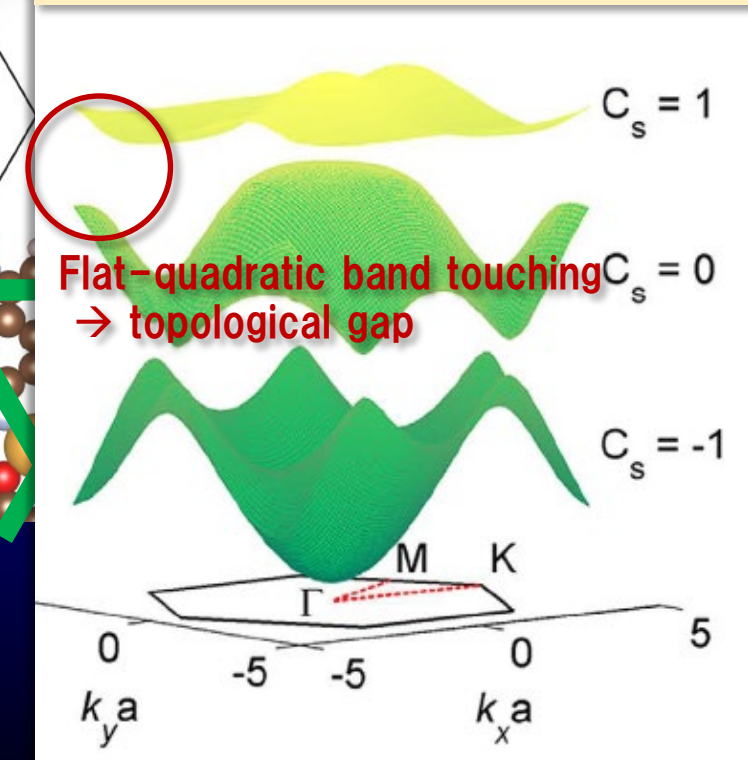
→ large superfluid weight
(Julku, ..., Torma, PRL 2016)

Kagome flat band in a metal-organic framework

(Yamada et al, PRB (R) 2016)

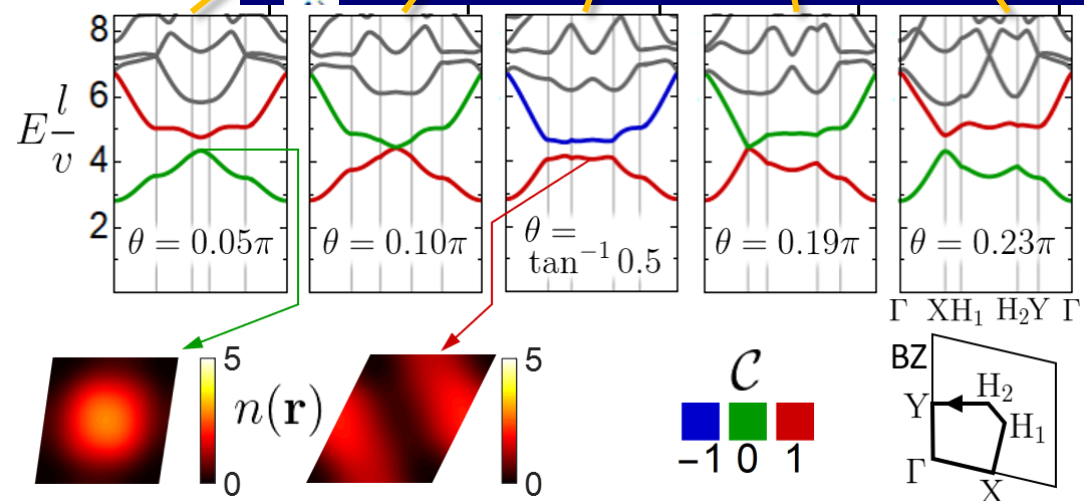
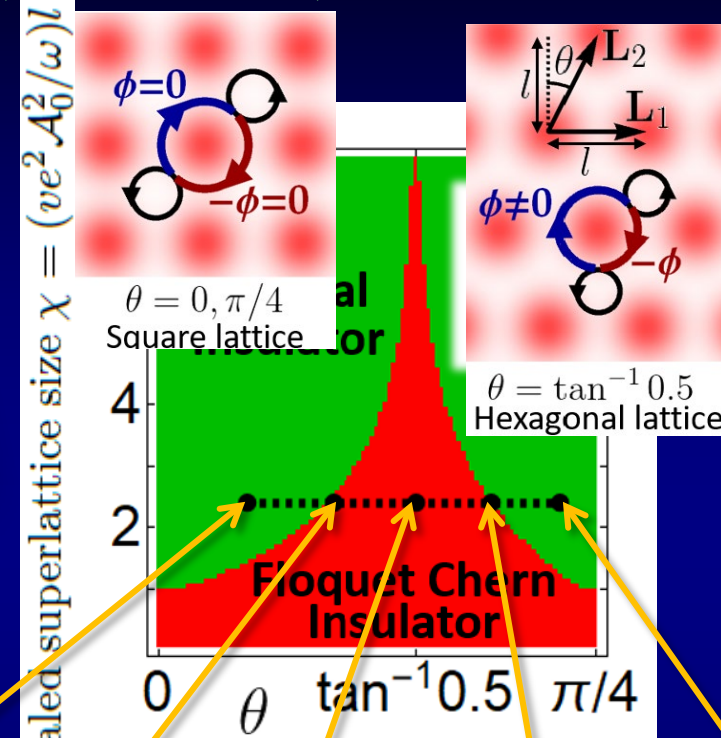
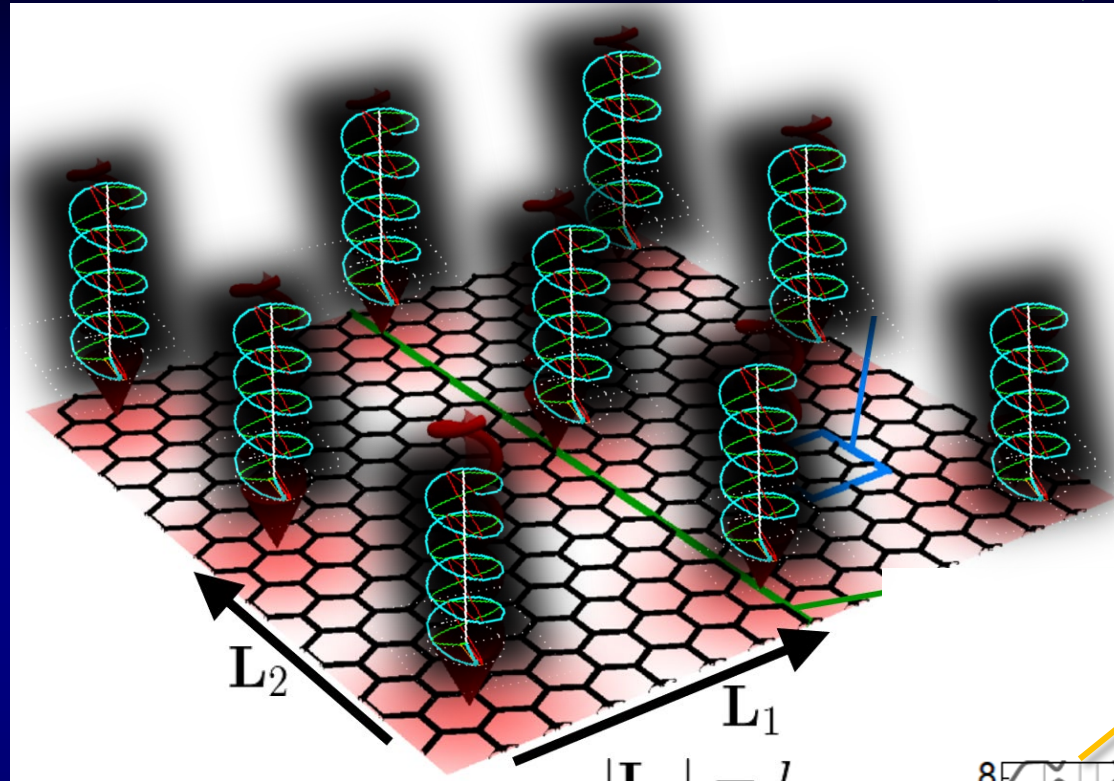


+ circularly-polarized light
→ topological
(Wang et al, Sci Rep 2017)



Optical imprinting of superlattices in 2D materials

(Kim, ..., Aoki & Hafezi, arXiv:1912.13059)



Today's talk

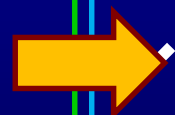
Many-body

Steady/transient

✓ Attraction–repulsion conversion
(Tsuji; Tsuji)

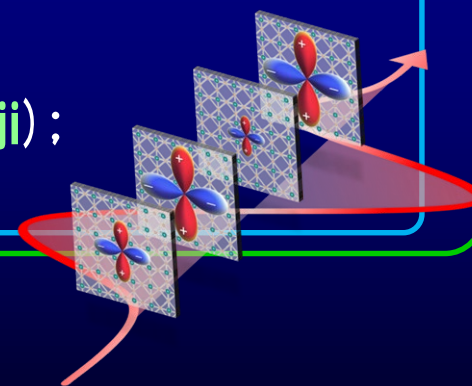
✓ η -pairing
(Kitamura)

✓ k -dep relaxation
(Tsuji; Sayyad)



♦ Higgs dynamics:

- * phonon mechanism SC (Tsuji);
- * unconventional SC (Tsuji)





(© Nobel F)

(© H. Aoki)

Collective modes in various classes of SSBs

	phase modes	amplitude modes	SSB
Crystals	acoustic phonon	optical phonon	translation
Magnets	magnon	amplitude mode	spin rotation
CDW	phason	amplitudon	discrete tr
BEC	Bogoliubov mode	amplitude mode	phase rotation
SF ^3He	Bogoliubov mode	amplitude mode	$\mathcal{S}, \mathcal{L}, \text{U}(1)$ phase
SC	–	Higgs mode	$\text{U}(1)$ phase
atomic nuclei	pion	σ particle	chiral symm

Anderson–Higgs mechanism

"Higgs" defined here as an amplitude mode in a gauge–coupled system

✓ Attraction–repulsion conversion

(Tsuji; Tsuji)

✓ η -pairing
(Kitamura)

✓ k -dep relaxation
(Tsuji; Sayyad)

✓ Higgs dynamics:

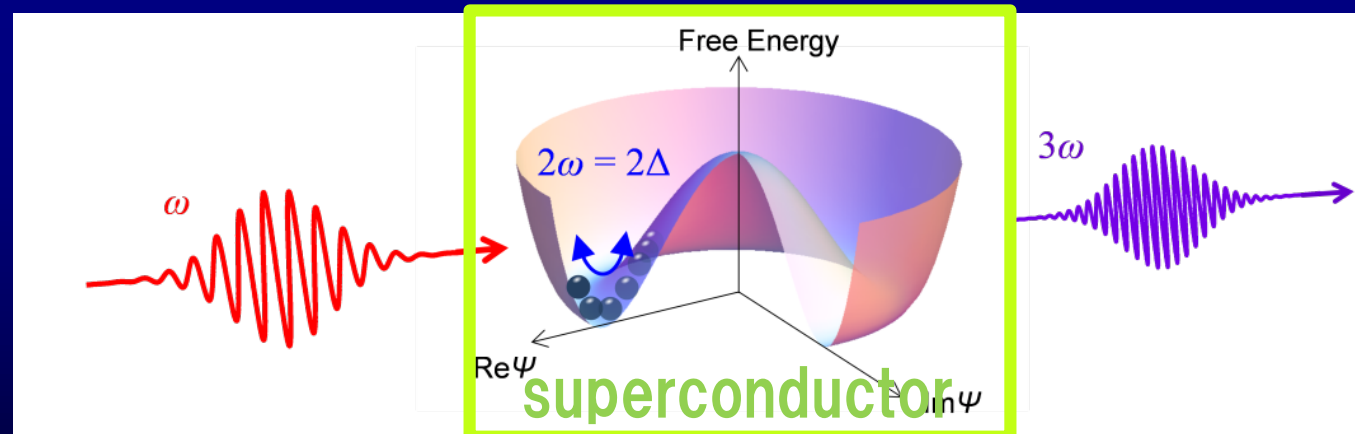
- 
- * phonon mechanism SC (Tsuji);
 - * unconventional SC (Tsuji)

SUPERCONDUCTIVITY

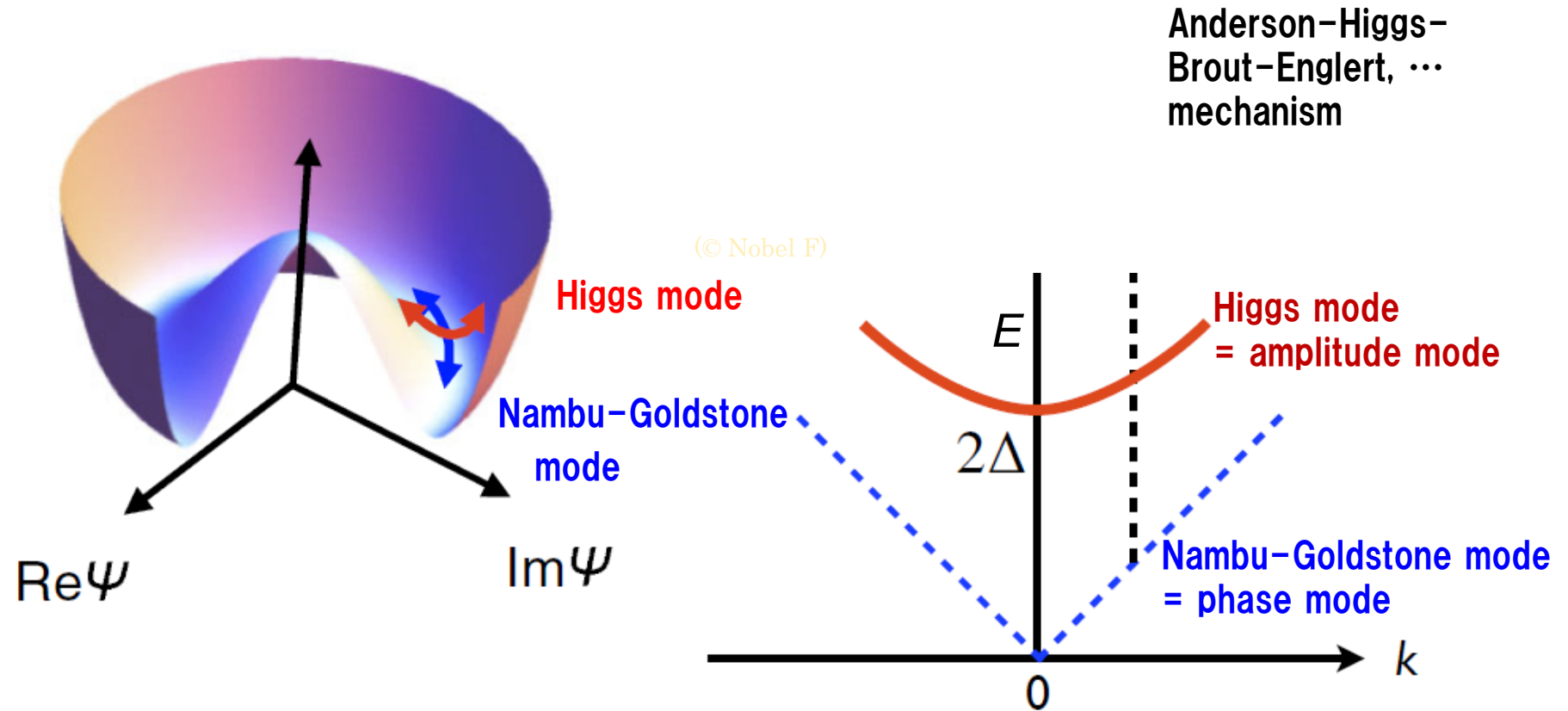
✓ s-wave conventional SC

Light-induced collective pseudospin precession resonating with Higgs mode in a superconductor

Ryusuke Matsunaga,^{1*} Naoto Tsuji,¹ Hiroyuki Fujita,¹ Arata Sugioka,¹ Kazumasa Makise,² Yoshinori Uzawa,^{3,†} Hirotaka Terai,² Zhen Wang,^{2,‡} Hideo Aoki,^{1,4} Ryo Shimano^{1,5*}

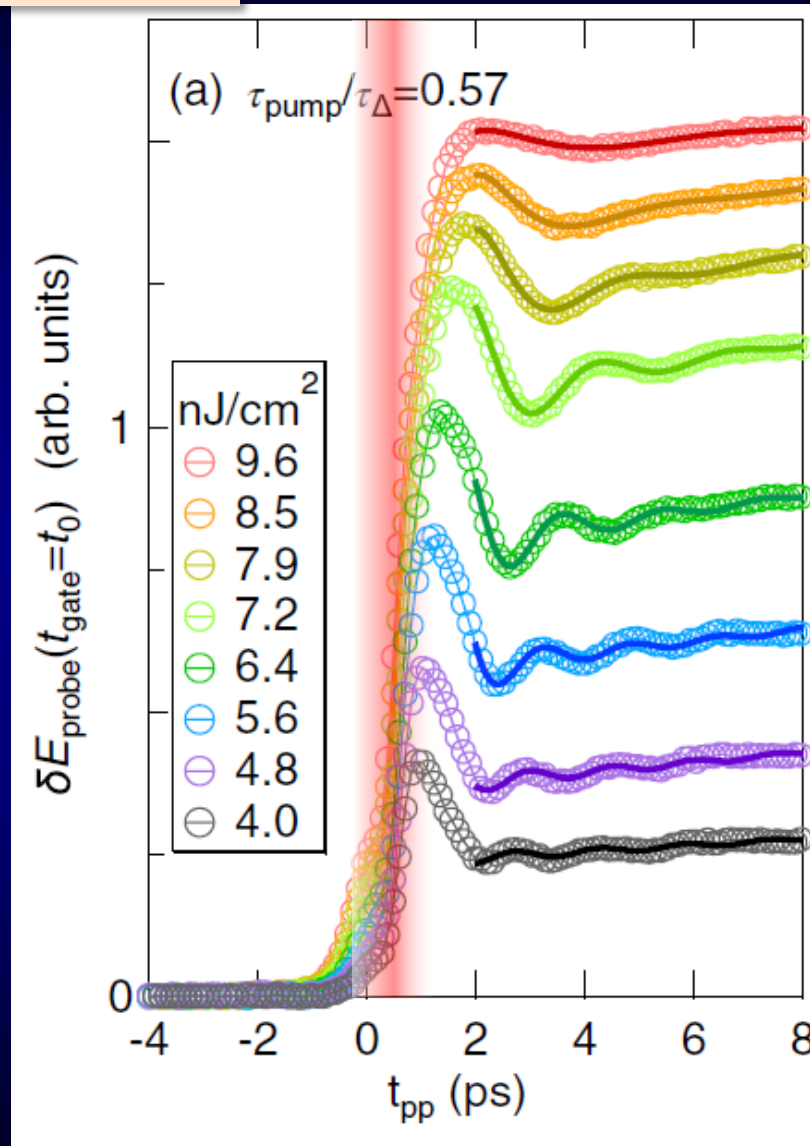
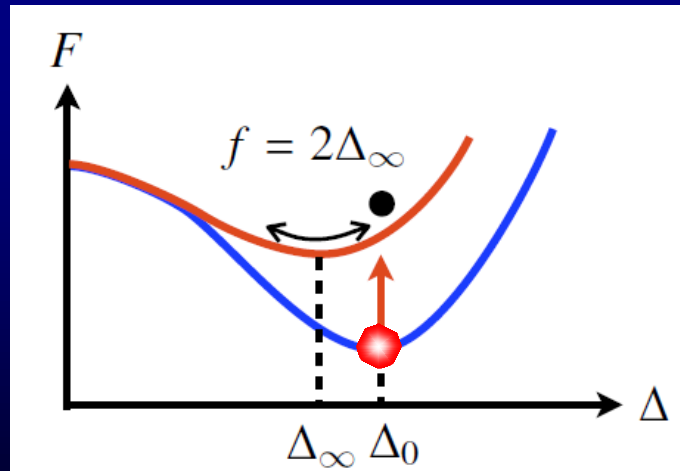
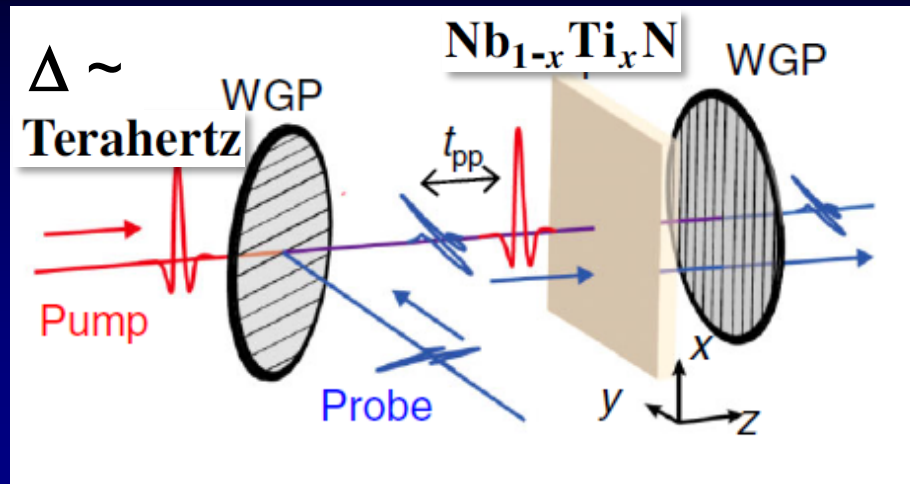


Collective excitations in superconductors

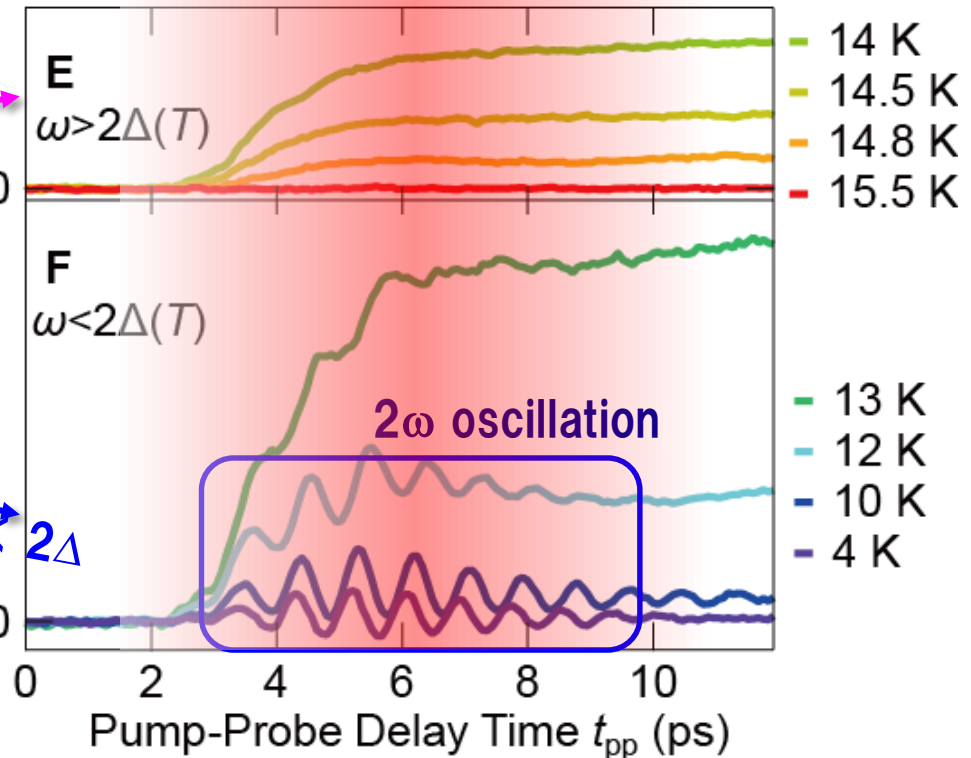
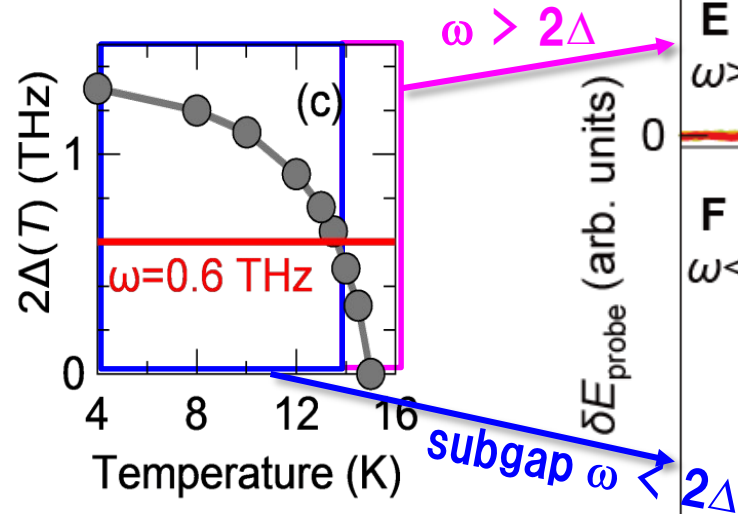
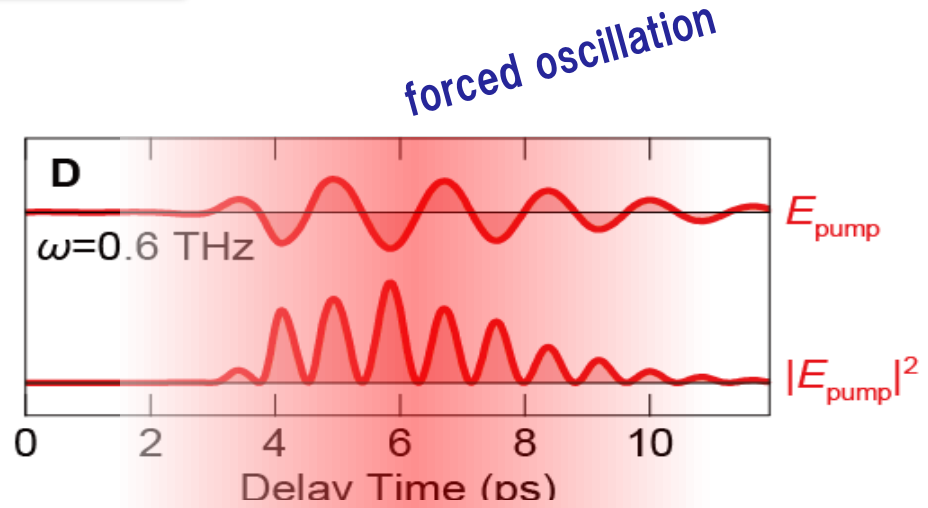
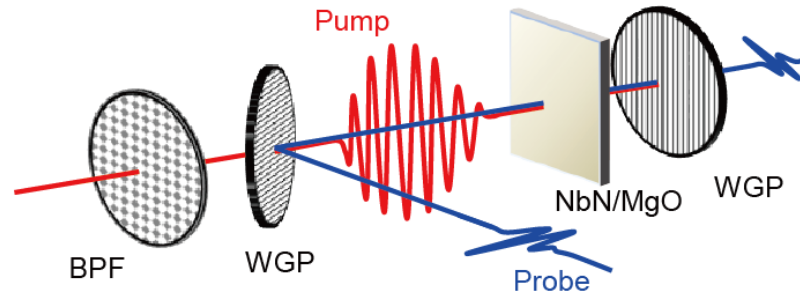


Experiment with THz laser: pump-probe

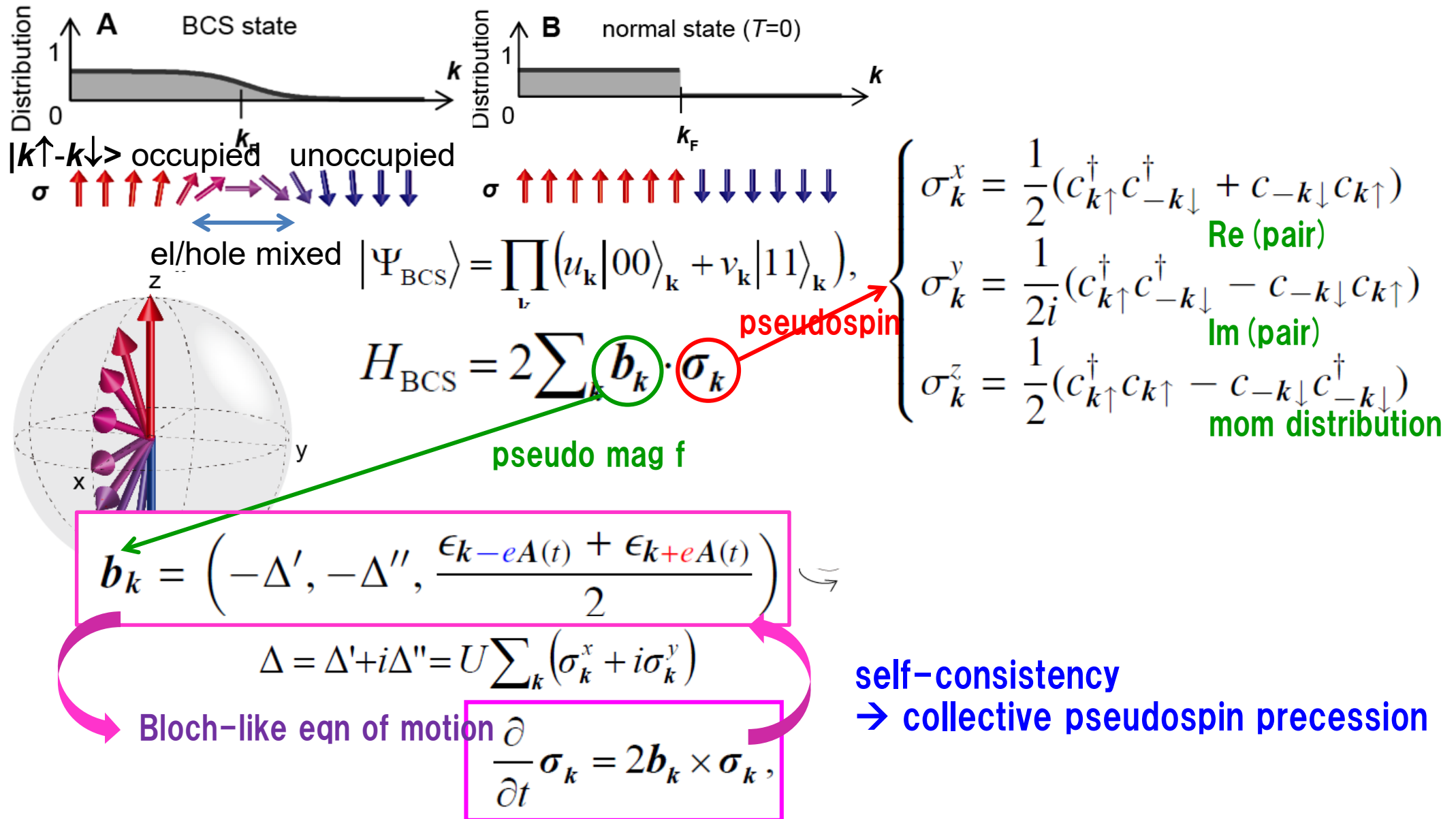
(Matsunaga et al, PRL 2013)



Experiment with THz laser: **during pump** (Matsunaga et al, Science 2014)



Anderson (1958) pseudospin description



Bogoliubov-de Gennes eqn

$$i\partial_t \Psi_k = \begin{pmatrix} \epsilon_{k-eA(t)} & -\Delta^* \\ -\Delta & -\epsilon_{k+eA(t)} \end{pmatrix} \Psi_k$$

↑
electron

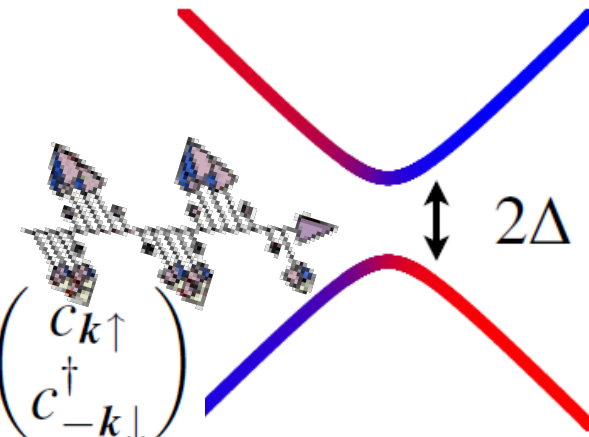
↑
hole

$$b_k = \left(-\Delta', -\Delta'', \frac{\epsilon_{k-eA(t)} + \epsilon_{k+eA(t)}}{2} \right)$$

↑
electron

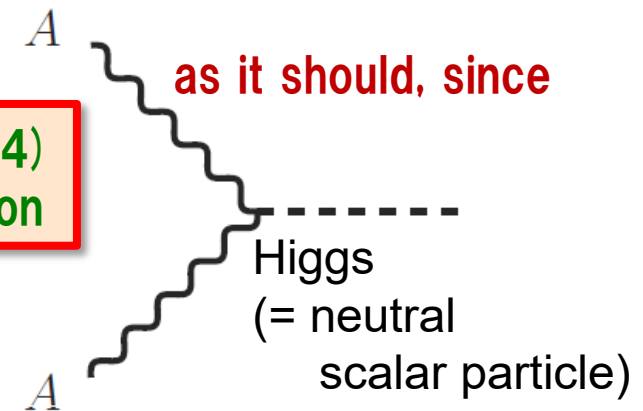
↑
hole

$$\Psi_k = \begin{pmatrix} c_{k\uparrow} \\ c_{-k\downarrow}^\dagger \end{pmatrix}$$



$$\frac{\epsilon_{k-eA(t)} + \epsilon_{k+eA(t)}}{2} = \epsilon_k + \frac{e^2}{2} \sum_{i,j} \frac{\partial^2 \epsilon_k}{\partial k_i \partial k_j} \overbrace{A_i(t) A_j(t)}^{\exp(i2\omega t)} + O(A^4)$$

Laser-SC coupling $\sim A^2$ (Matsunaga et al, Science 2014)
 ← one reason why it took half a century for the observation



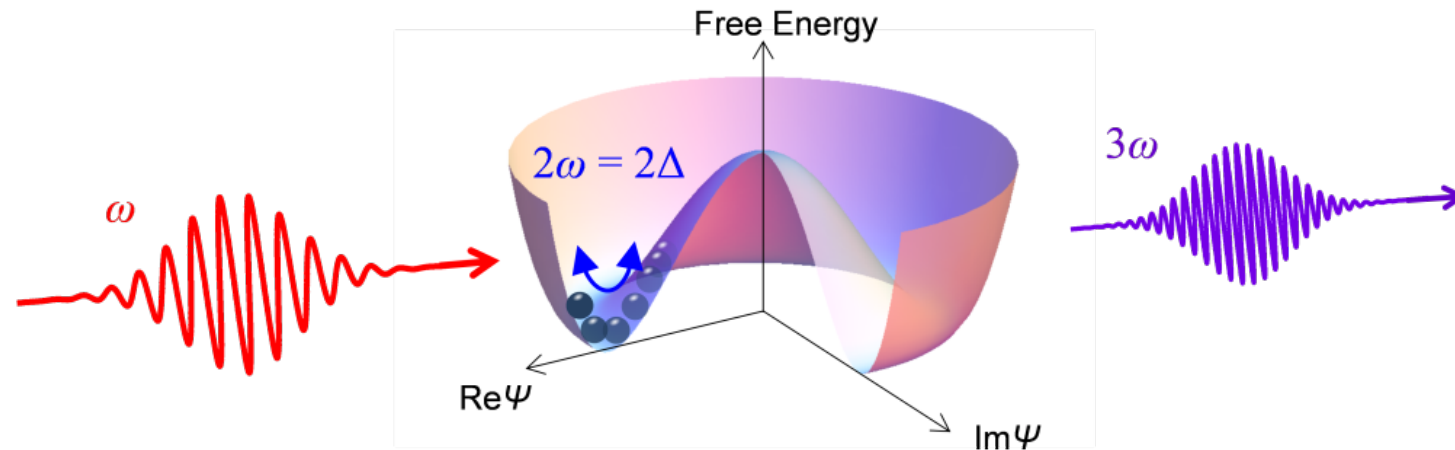
Nonlinear “London eqn”

(Matsunaga et al, Science 2014)

$$j_{\text{NL}}^{(3)}(t) \propto \frac{e^2 \Delta}{U} \underline{A(t)} \underline{\delta\Delta(t)}$$

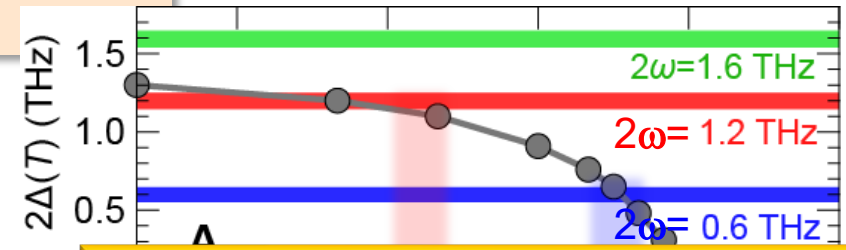
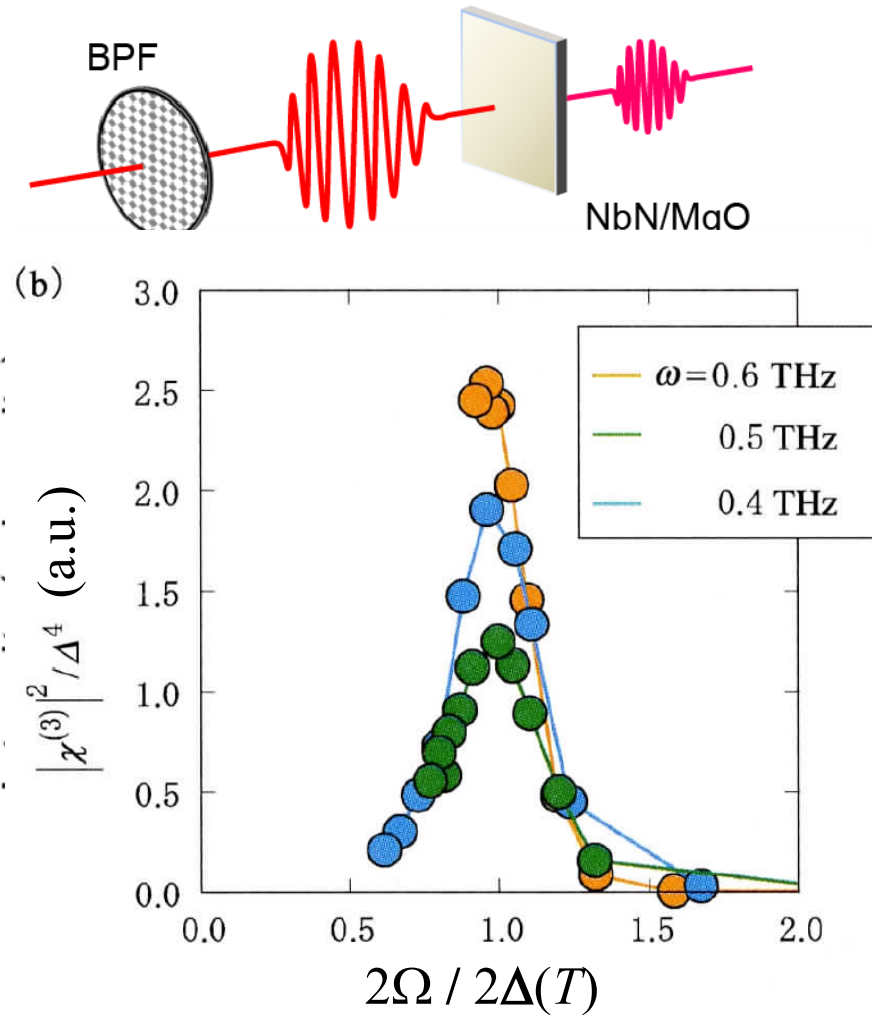
$\omega \otimes 2\omega = 2\Delta$ resonance $\rightarrow 3\omega$

($\rightarrow$ 3rd-harmonic generation)

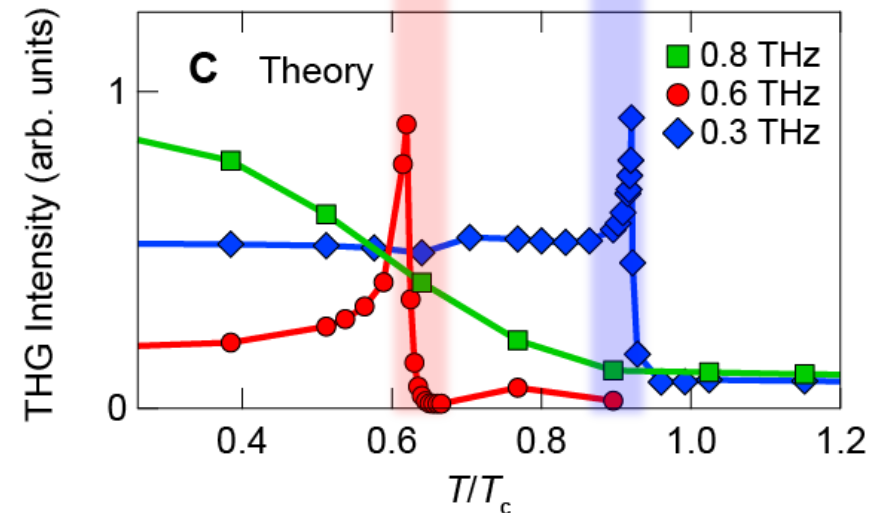
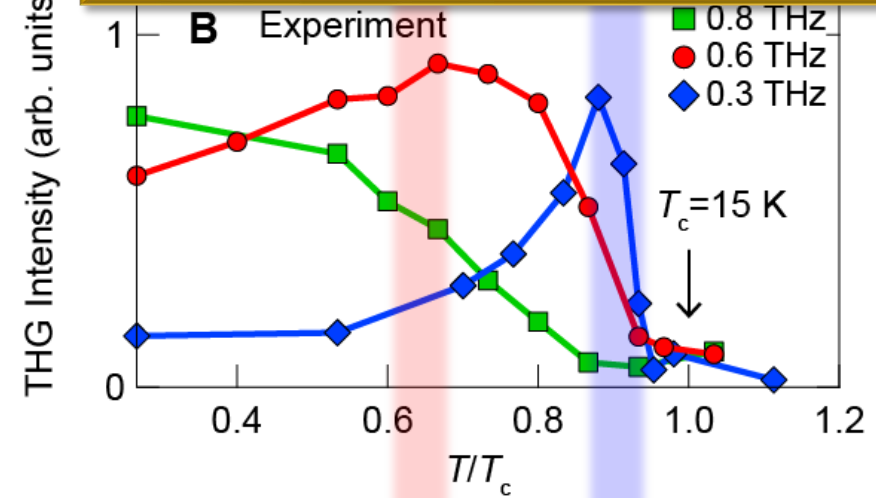


Third-harmonics generation: Exp & theory

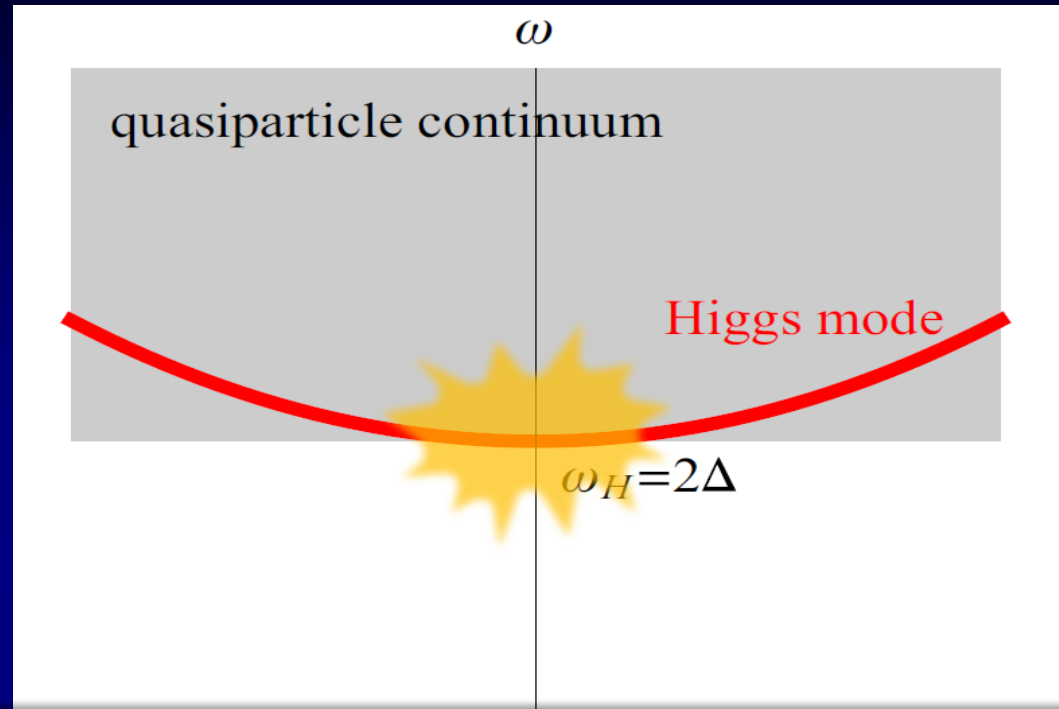
(Matsunaga et al, Science 2014)



resonant with Higgs ($2\Omega = 2\Delta$)



Higgs vs quasiparticle (charge density fl) excitations



Collective Higgs mode and **quasiparticle excitations (CDF)**

-- both contribute to the 2Δ mode (Cea, ..., Benfatto, PRB2016)

→ go beyond BCS

(Tsuji, Murakami & Aoki, PRB 2016; Matsunaga et al, PRB (R) 2017)

THG susceptibility beyond BCS

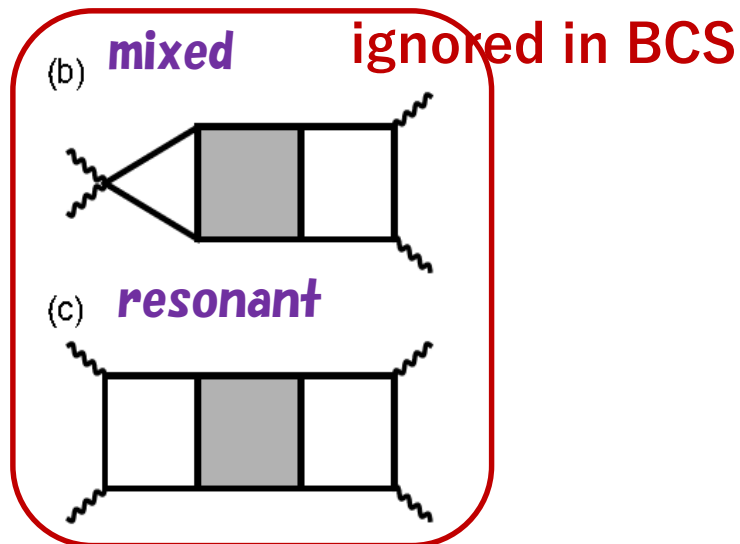
(Tsuji, Murakami & Aoki, PRB 2016)

$$j^{(3)} = \chi_{\text{tot}}^{(3)} A^3 = (\chi_0^{(3)} + \chi_{\text{vc}}^{(3)}) A^3$$

χ_{THG} diagrams with vertex corrections $\supset$ collective modes



beyond BCS



terminology borrowed from Raman scattering

DMFT extended for treating THG

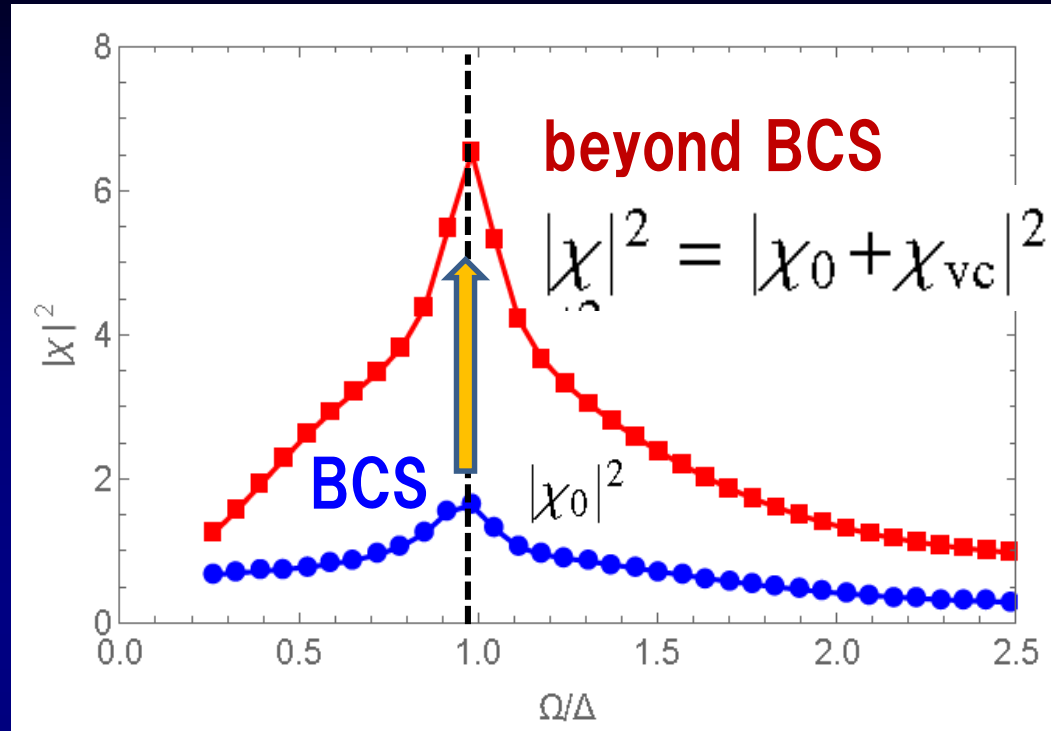
$$\chi_{\text{THG}}(\Omega) = \sum_{i=1}^5 \chi_0^{(i)}(\Omega) + \sum_{i=1}^2 \chi_{\text{vc}}^{(i)}(\Omega)$$

$$\chi_0^{(1)} = \text{circle}, \quad \chi_0^{(2)} = \text{bubble}, \quad \chi_0^{(3)} = \text{fish}$$

$$\chi_0^{(4)} = \text{triangle}, \quad \chi_0^{(5)} = \text{square}$$

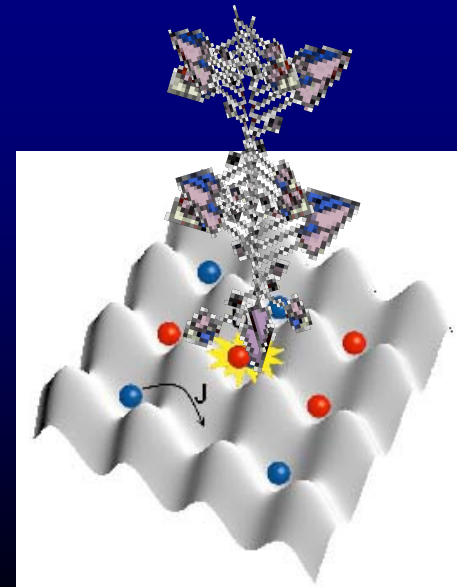
$$\chi_{\text{vc}}^{(1)} = \text{triangle with dot}, \quad \chi_{\text{vc}}^{(2)} = \text{square with dot}$$

THG intensity (Tsuji, Murakami & Aoki, PRB 2016)



Conclusion:

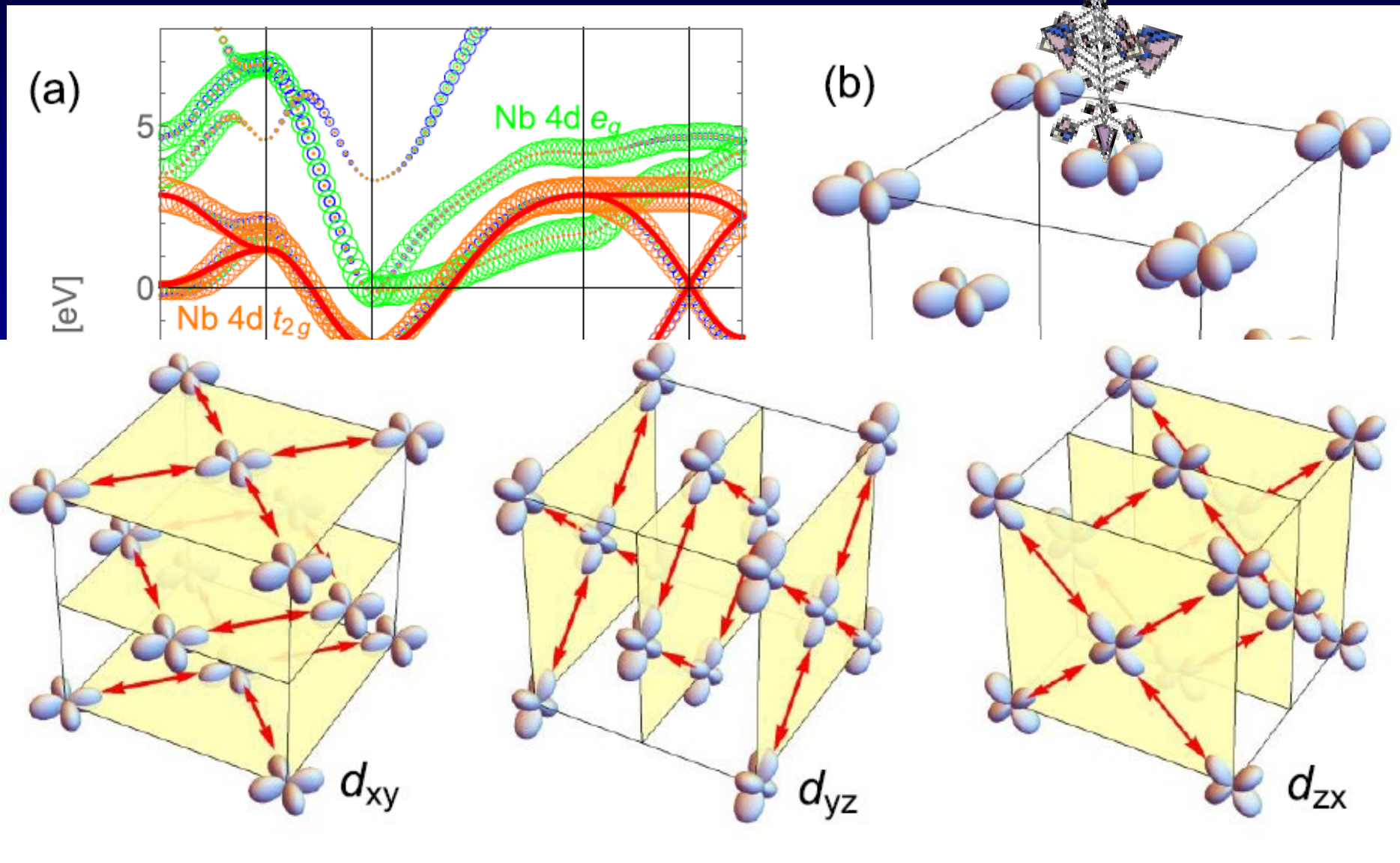
- (1) Higgs mode can significantly contribute if we go beyond BCS,
- (2) Strongly depends on angle between light and crystal



Higgs vs charge density fl

--- polarization-resolved THG in NbN

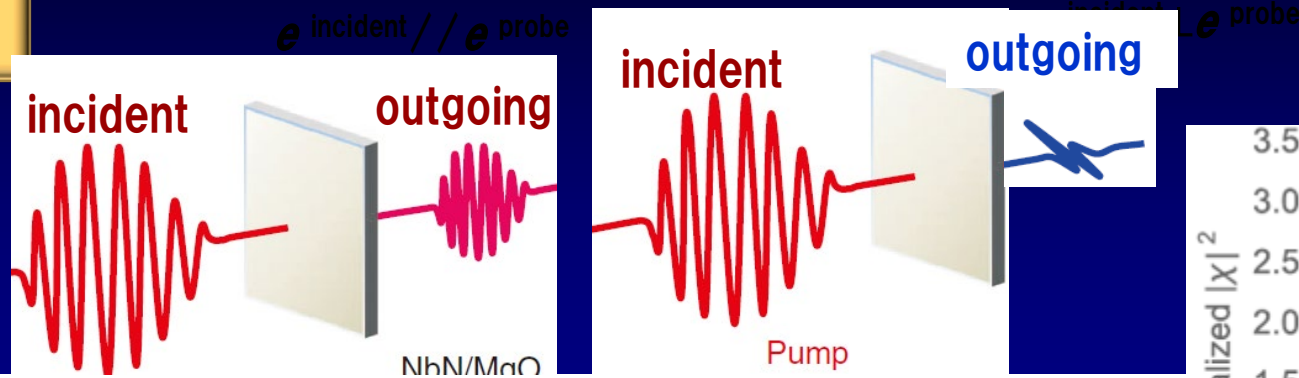
(Matsunaga et al, PRB 2017)



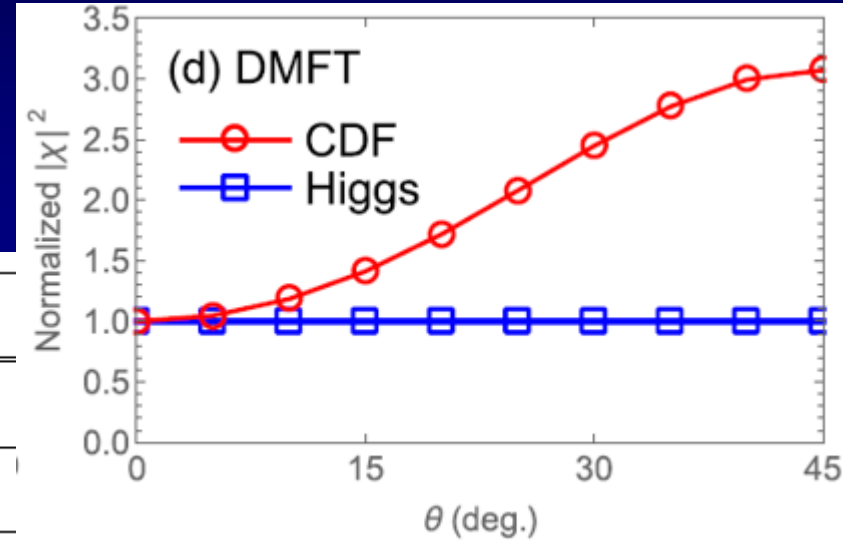
Polarization-resolved Higgs vs charge density fl contributions

(Matsunaga, Tsuji, ..., Aoki & Shimano, PRB (R) 2017)

Theory



	$e^I = e^O = e_\theta$	$e^I = e_\theta, e^O = e_{\theta+90^\circ}$
CDF	$A(\omega) + 2B(\omega)\sin^2 2\theta$	$B(\omega)\sin 4\theta$
Higgs	$C(\omega)$	0

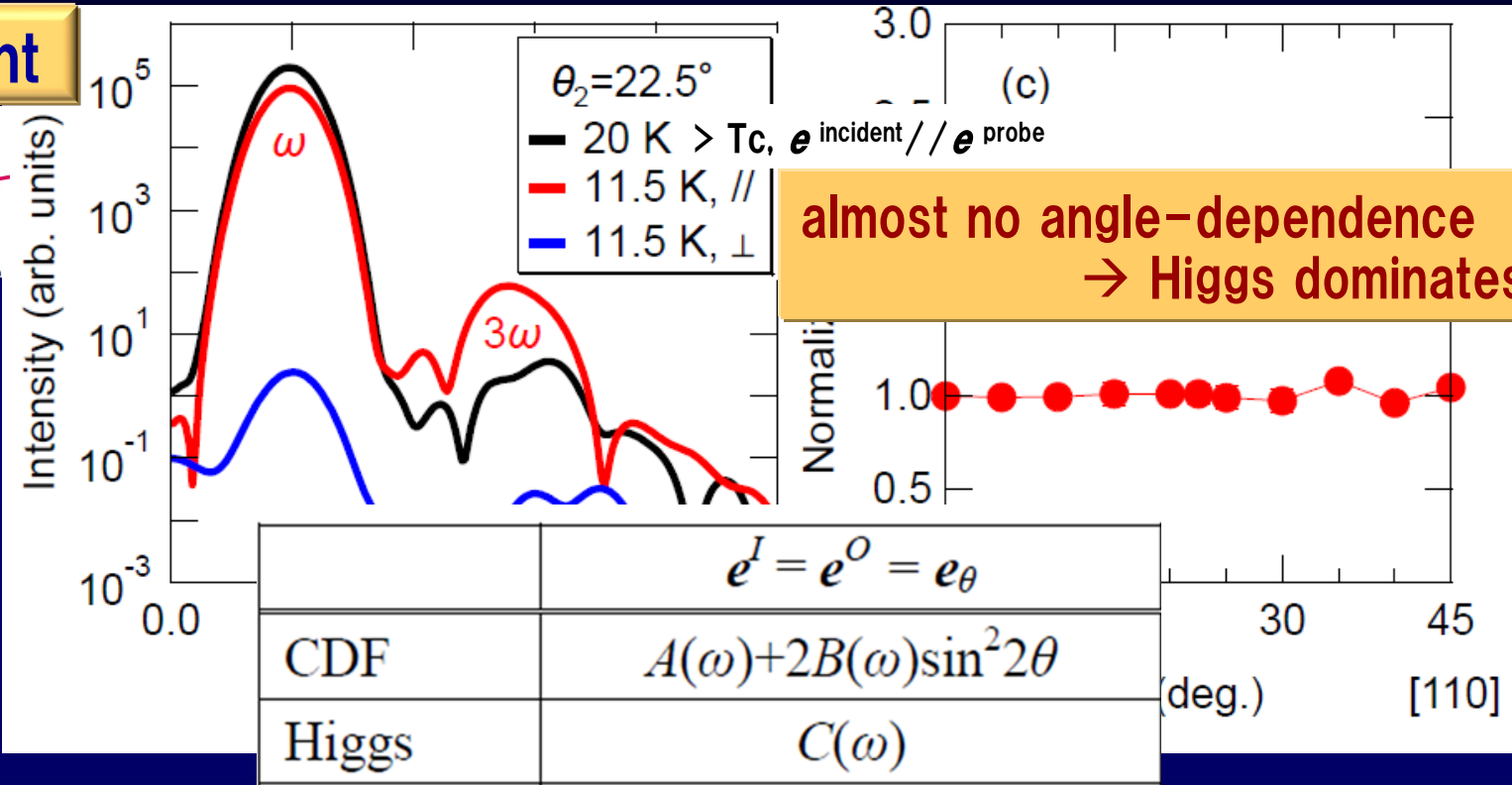
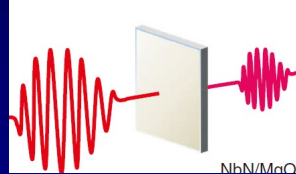


angle-dependent $\rightarrow$ CDF,
angle-independent $\rightarrow$ Higgs

Polarisation-resolved THz THG in NbN

(Matsunaga, Tsuji, ..., Aoki & Shimano, PRB (R) 2017)

Experiment



almost no angle-dependence
→ Higgs dominates

- Higgs $\gg$ CDF (opposite to BCS) $|C(\omega)| \gg |B(\omega)|$ ($\sim |A(\omega)|$)
 possible reason: NbN = strongly coupled el-ph
 → large retardation effects → BCS invalidated

✓ Attraction–repulsion conversion

(Tsuji; Tsuji)

✓ η -pairing
(Kitamura)

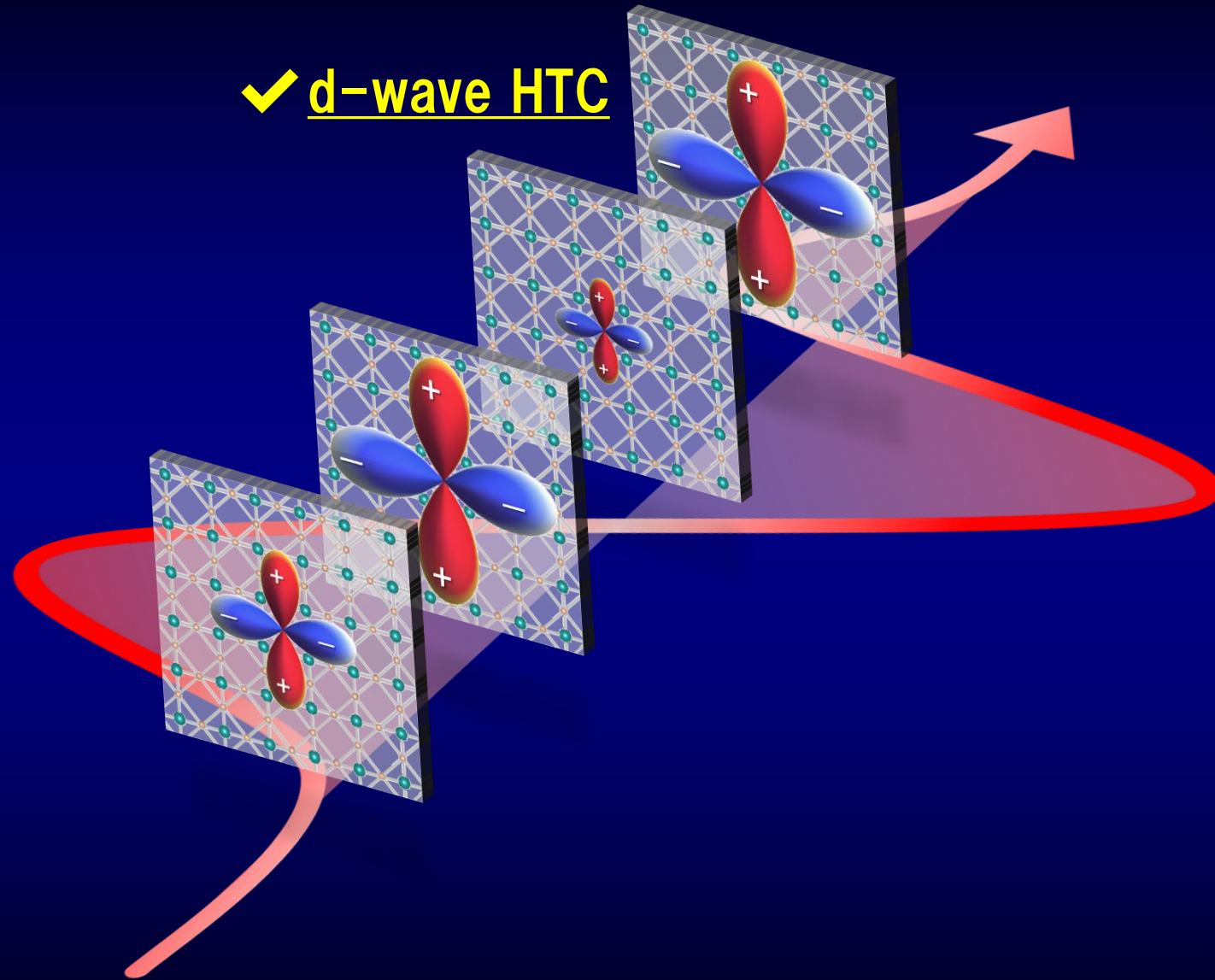
✓ k -dep relaxation
(Tsuji; Sayyad)

✓ Higgs dynamics:

* phonon mechanism SC (Tsuji);

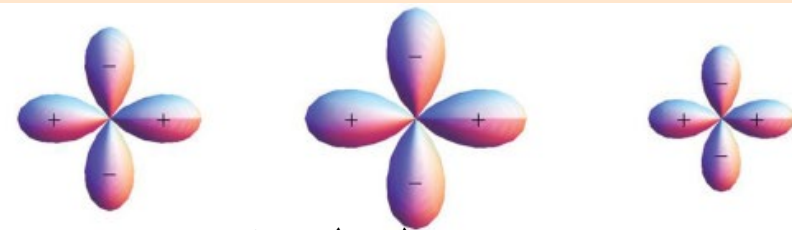
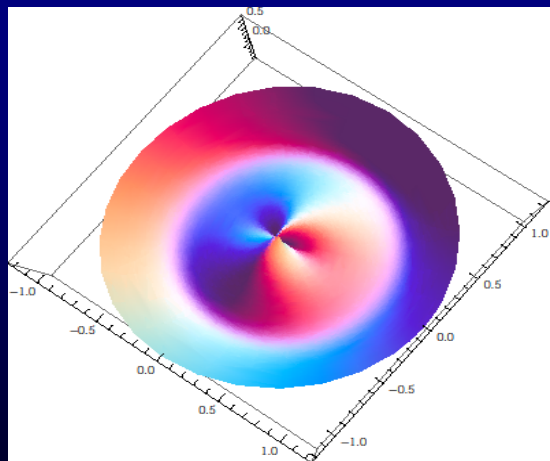
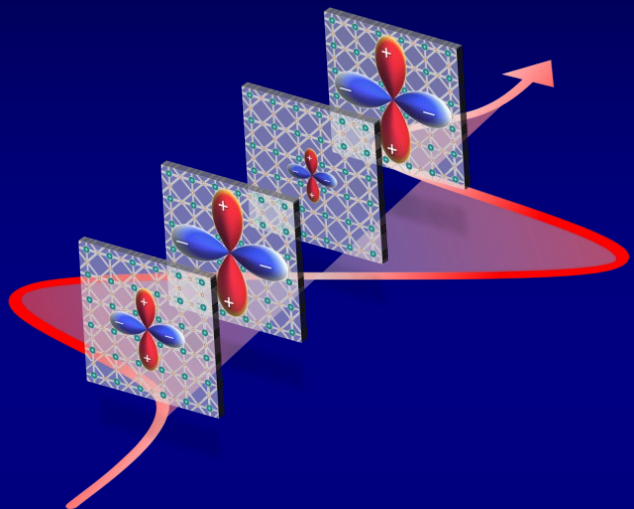
 * unconventional SC (Tsuji)

✓ d-wave HTC



Higgs modes in unconventional (*d*-wave) SC

Group-theoretical classification for amplitude oscillations for $d_{x^2-y^2}$ -wave
(Barlas & Varma, PRB 2013)



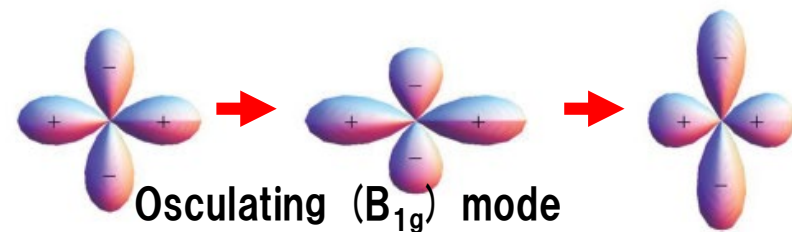
Breathing (A_{1g}) mode



Rotating (A_{2g}) mode



Clapping (B_{2g}) mode



Osculating (B_{1g}) mode

Actual detection of the Higgs modes in a HTC cuprate

Experiment + theory on $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$ (Bi2212)
(Katsumi, ..., Aoki & Shimano, PRL 2018)

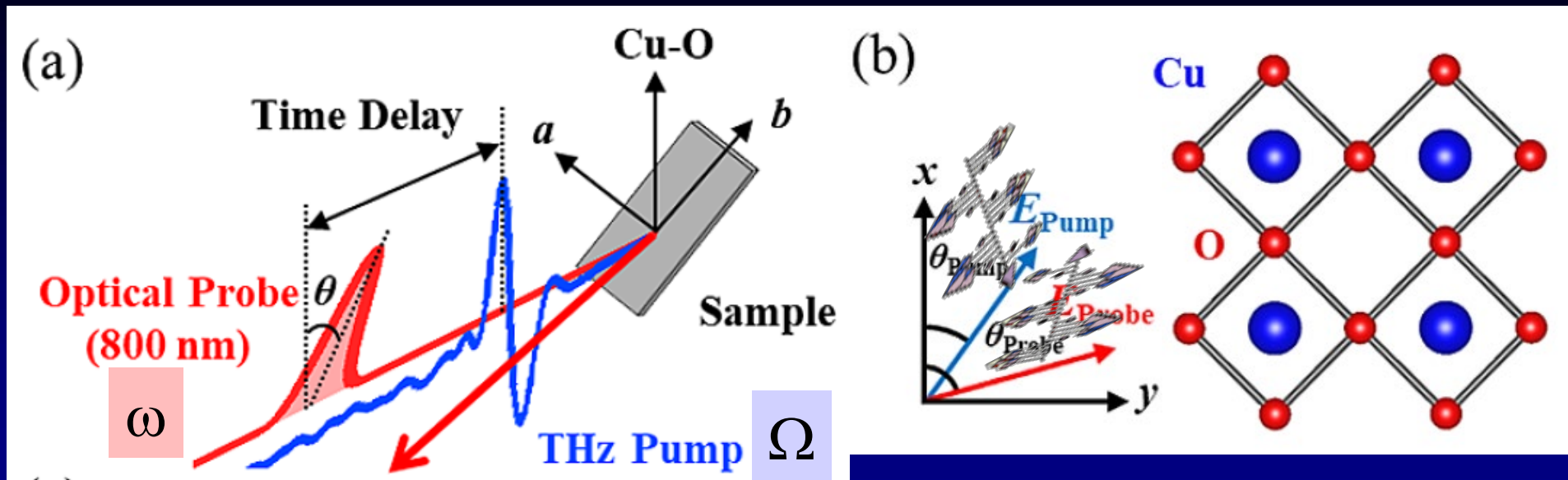
PHYSICAL REVIEW LETTERS 120, 117001 (2018)

Editors' Suggestion

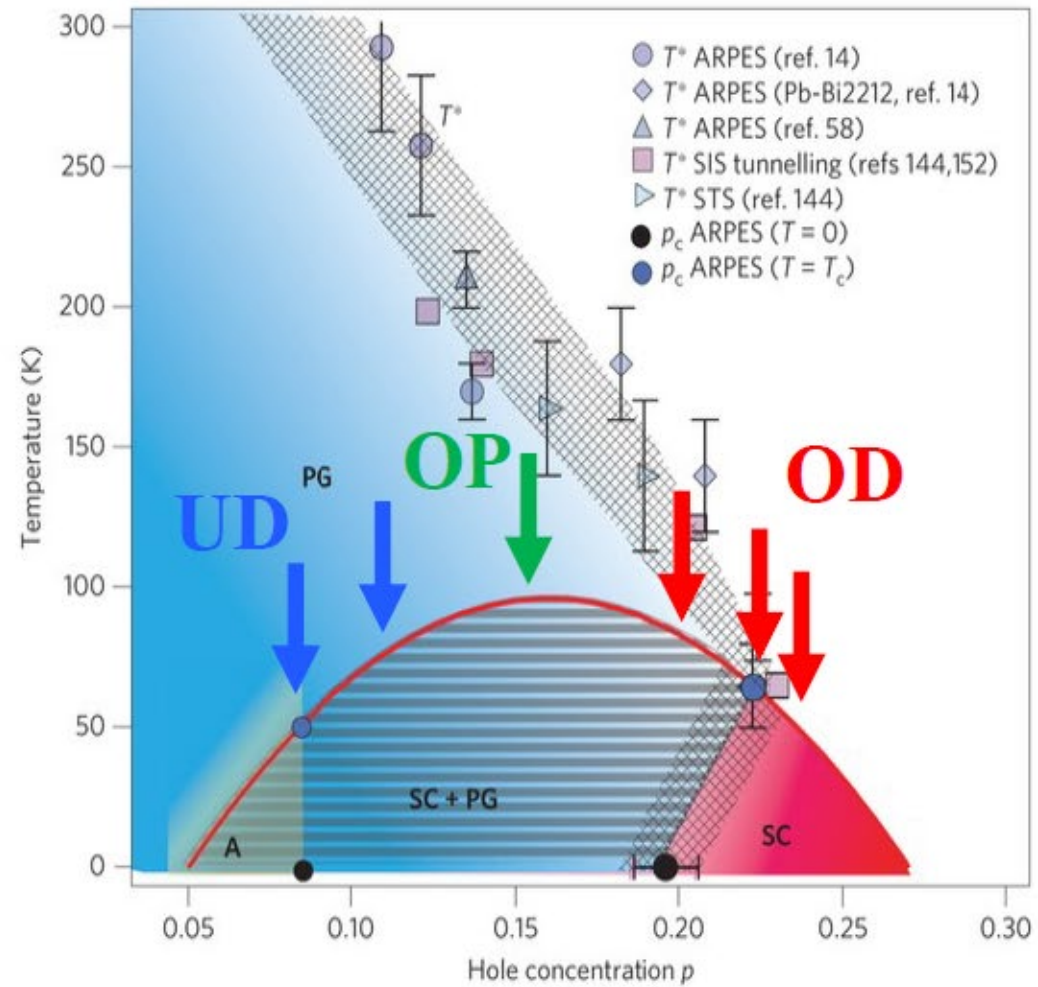
Higgs Mode in the d -Wave Superconductor $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$ Driven by an Intense Terahertz Pulse

Kota Katsumi,¹ Naoto Tsuji,² Yuki I. Hamada,¹ Ryusuke Matsunaga,^{1,3} John Schneeloch,⁴ Ruidan D. Zhong,
Genda D. Gu,⁴ Hideo Aoki,^{1,5,6} Yann Gallais,^{1,7,8} and Ryo Shimano^{1,8}
¹*Department of Physics, The University of Tokyo, Tokyo 113-0033, Japan*

THz-pump, optical (near IR) -probe



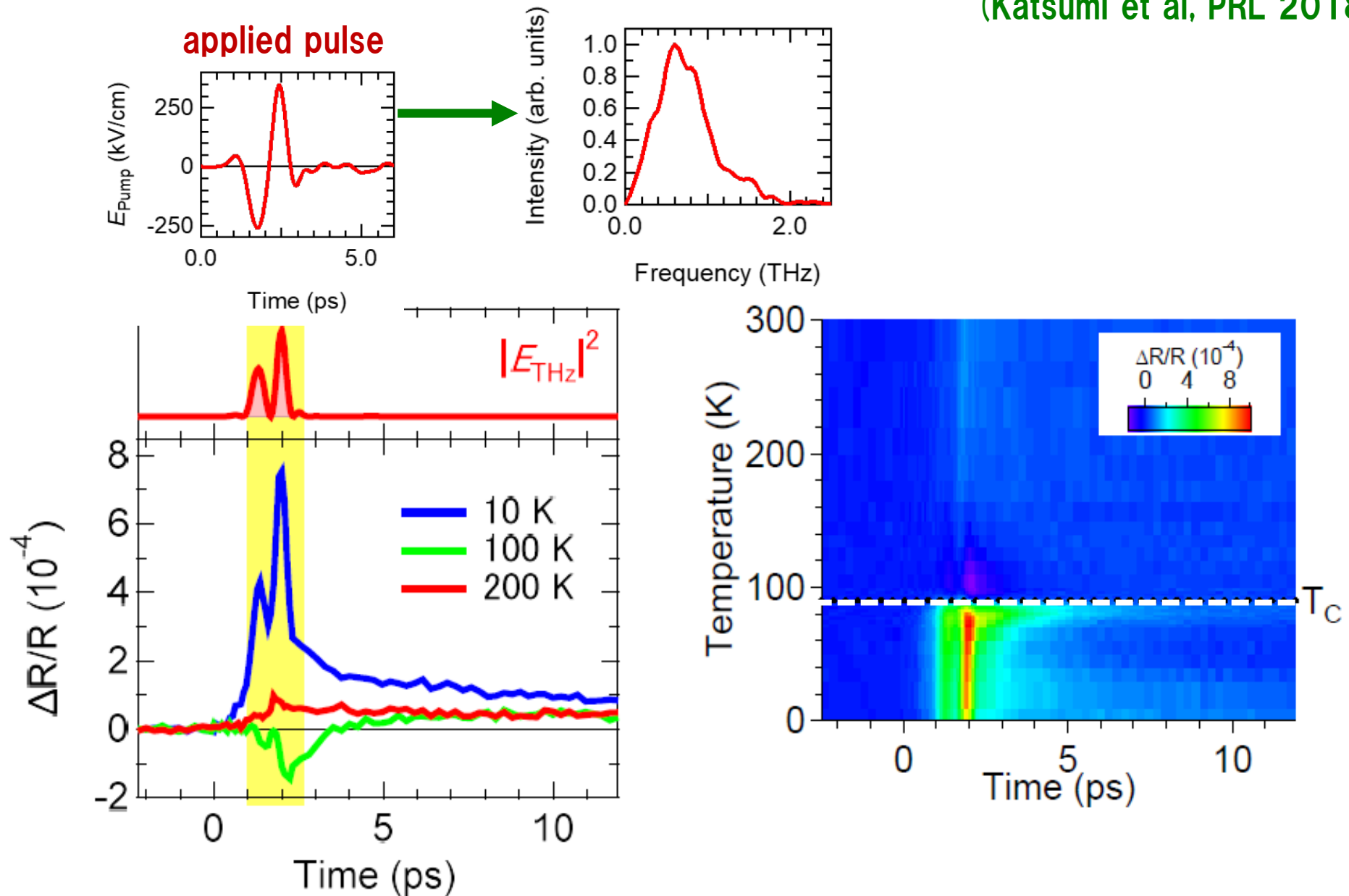
Employed samples of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$



(Hashimoto et al, Nat Phys 2014)

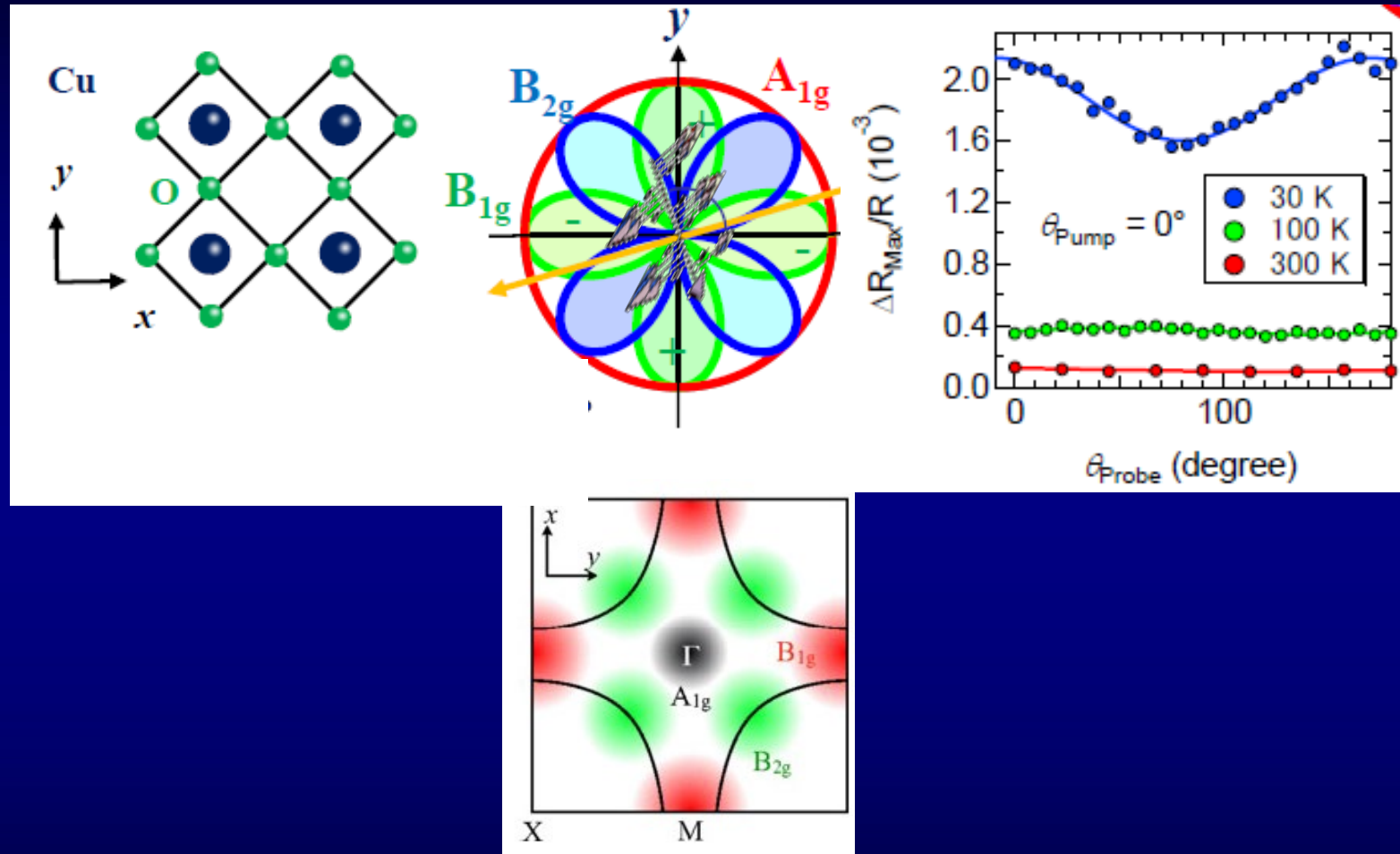
Transient reflectivity change

(Katsumi et al, PRL 2018)



Symmetry of the signal

(Katsumi et al, PRL 2018)



We can decompose into irred reps

(Katsumi et al, PRL 2018)

Ω

ω

THz-pump, optical-probe (reflectivity change) measures THz Kerr signal
= 3rd-order nonlinear susceptibility $\chi^{(3)}$

$$e_{\text{Probe}} j^{(3)}(t) = \chi^{(3)}(\omega; \omega, +\Omega, -\Omega) A_{\text{Pump}}^2 A_{\text{Probe}} e^{-i\omega t}.$$

$$\frac{\Delta R}{R}(E_i^{\text{probe}}, E_j^{\text{probe}}) \sim \frac{1}{R} \frac{\partial R}{\partial \epsilon_1} \epsilon_0 \text{Re} \chi_{ijkl}^{(3)} E_k^{\text{pump}} E_l^{\text{pump}} \quad (i, j, k, l = x, y)$$

3rd-order nonlinear susc for D_{4h} point group $\in$ Bi2212

$$\chi^{(3)}(\theta_{\text{Pump}}, \theta_{\text{Probe}}) = \frac{1}{2} (\overset{\text{A1g}}{\chi_{A_{1g}}^{(3)}} + \overset{\text{B1g}}{\chi_{B_{1g}}^{(3)}} \cos 2\theta_{\text{Pump}} \cos 2\theta_{\text{Probe}} + \overset{\text{B2g}}{\chi_{B_{2g}}^{(3)}} \sin 2\theta_{\text{Pump}} \sin 2\theta_{\text{Probe}})$$

$\chi_{A_{1g}}^{(3)} = \chi_{xxxx}^{(3)} + \chi_{xyyy}^{(3)}, \chi_{B_{1g}}^{(3)} = \chi_{xxxx}^{(3)} - \chi_{xyyy}^{(3)} \text{ and } \chi_{B_{2g}}^{(3)} = \chi_{xyxy}^{(3)} + \chi_{yyxx}^{(3)}$



A1g: indep of θ_{Pump}

B1g $\sim \cos(2\theta_{\text{Pump}})$

$$\frac{\Delta R}{R}(\theta_{\text{Pump}}, \theta_{\text{Probe}}) = \frac{\Delta R_{A_{1g}}}{R} + \frac{\Delta R_{B_{1g}}}{R} \cos 2\theta_{\text{Pump}} \cos 2\theta_{\text{Probe}} + \frac{\Delta R_{B_{2g}}}{R} \sin 2\theta_{\text{Pump}} \sin 2\theta_{\text{Probe}}$$

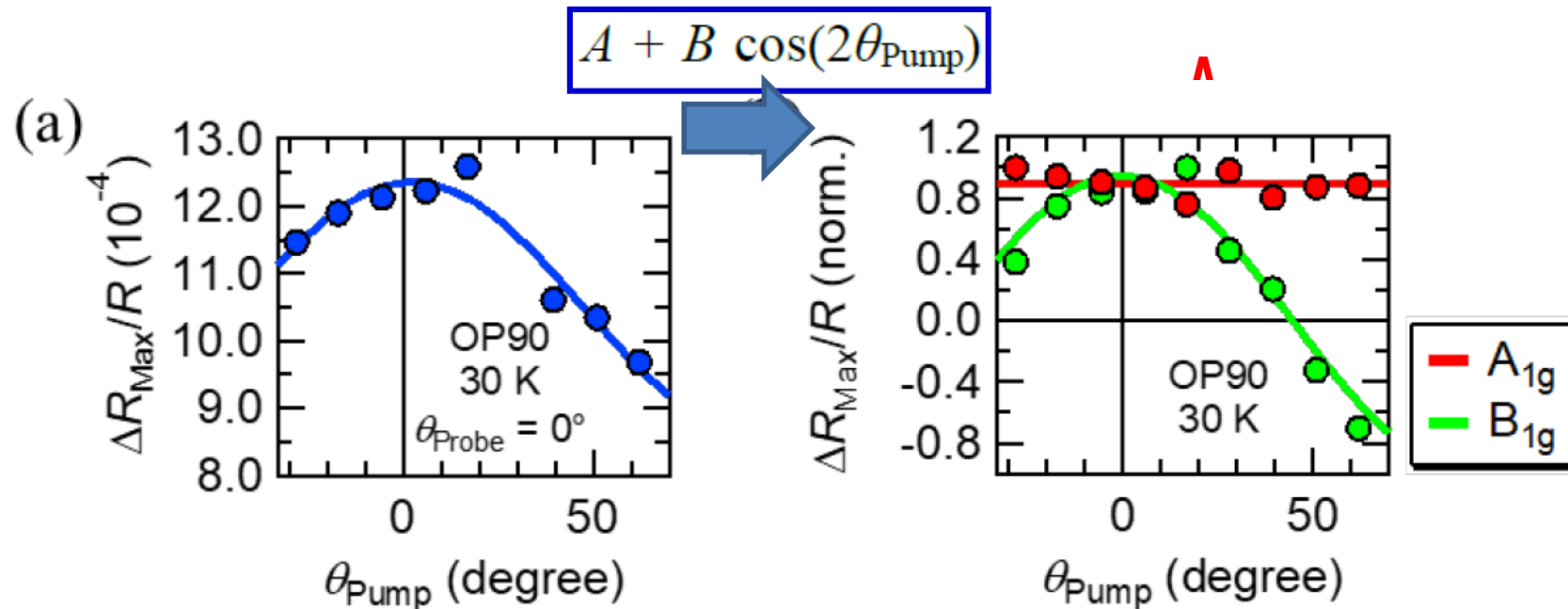
Pump-polarization dependence: experiment

(Katsumi et al, PRL 2018)

$$\frac{\Delta R}{R}(\theta_{\text{Pump}}, \theta_{\text{Probe}}) = \frac{\Delta R_{A_{1g}}}{R} + \frac{\Delta R_{B_{1g}}}{R} \cos 2\theta_{\text{Pump}} \cos 2\theta_{\text{Probe}} + \frac{\Delta R_{B_{2g}}}{R} \sin 2\theta_{\text{Pump}} \sin 2\theta_{\text{Probe}}$$

A_{1g} : indep of θ_{Pump}

$B_{1g} \sim \cos(2\theta_{\text{Pump}})$



Theoretical nonlinear susceptibility

A general property

(Katsumi et al, PRL 2018)

THz-pump optical-probe Kerr susceptibility ($\chi^{(3)}$)
(not to be confused with amplitude osc modes)

	A_{1g}	B_{1g}	B_{2g}
CDF	✓	✓	✓
Higgs	✓	0	0

Pump-polarization dependence: theory

--- everything now dep on k

(Katsumi et al, PRL 2018)

$$H = \sum_{k\sigma} \xi_k c_{k\sigma}^\dagger c_{k\sigma} - \frac{1}{N} \sum_{kk'} V(k, k') c_{k\uparrow}^\dagger c_{-k\downarrow}^\dagger c_{-k'\downarrow} c_{k'\uparrow}$$

$\propto (\cos k_x - \cos k_y)(\cos k'_x - \cos k'_y)$ for d-wave

$$\Delta_k = \frac{1}{N} \sum_{k'} V(k, k') \langle c_{k'\uparrow}^\dagger c_{-k'\downarrow}^\dagger \rangle = \frac{1}{N} \sum_{k'} V(k, k') \frac{\Delta_{k'}}{2E_{k'}} \tanh\left(\frac{E_{k'}}{2T}\right) \propto (\cos k_x - \cos k_y)$$

k-dep Pseudospin $\sigma_k = \frac{1}{2} \langle \Psi_k^\dagger \boldsymbol{\tau} \Psi_k \rangle$ $\Psi_k^\dagger = (c_{k\uparrow}^\dagger, c_{-k\downarrow})$

$\frac{\partial}{\partial t} \sigma_k(t) = 2\mathbf{b}_k(t) \times \sigma_k(t)$ **Bloch-like eqn motion**

$\mathbf{b}_k(t) = (-\Delta'_k(t), -\Delta''_k(t), \frac{\xi_{k-A(t)} + \xi_{k+A(t)}}{2})$

$\sigma_k(t) = \sigma_{k, \text{eq}} + \delta\sigma_k(t)$

Laser-induced current

$$\mathbf{j}^{(3)}(t) = -\frac{2}{N} \sum_{k,i} \frac{\partial \mathbf{v}_k}{\partial k_i} A_i(t) \delta\sigma_k^z(t) - \frac{1}{3N} \sum_{k,ijk} \frac{\partial^3 \mathbf{v}_k}{\partial k_i \partial k_j \partial k_k} A_i(t) A_j(t) A_k(t) \left(\sigma_{k, \text{eq}}^z + \frac{1}{2} \right)$$

Theoretical THG susceptibility in HTC cuprate

(Katsumi et al, PRL 2018)

$$e_{\text{Probe}} j^{(3)}(t) = \chi^{(3)}(\omega; \omega, +\Omega, -\Omega) A_{\text{Pump}}^2 A_{\text{Probe}} e^{-i\omega t}.$$

Higgs

$$j^{(3)}(t) = -\frac{2}{N} \sum_{k,i} \frac{\partial v_k}{\partial k_i} A_i(t) \delta \sigma_k^z(t) - \frac{1}{3N} \sum_{k,ijk} \frac{\partial^3 v_k}{\partial k_i \partial k_j \partial k_k} A_i(t) A_j(t) A_k(t) \left(\sigma_{k,\text{eq}}^z + \frac{1}{2} \right)$$

- (i) $A_i(t)$ is $A_{\text{Pump}} e^{i\Omega t}$ with $\delta \sigma_k^z(t)$ containing $A_{\text{Pump}} e^{-i\Omega t}$ and $A_{\text{Probe}} e^{-i\omega t}$.
- (ii) $A_i(t)$ is $A_{\text{Pump}} e^{-i\Omega t}$ with $\delta \sigma_k^z(t)$ containing $A_{\text{Pump}} e^{i\Omega t}$ and $A_{\text{Probe}} e^{-i\omega t}$.
- (iii) $A_i(t)$ is $A_{\text{Probe}} e^{-i\omega t}$ with $\delta \sigma_k^z(t)$ containing $A_{\text{Pump}} e^{i\Omega t}$ and $A_{\text{Pump}} e^{-i\Omega t}$.

(iv)

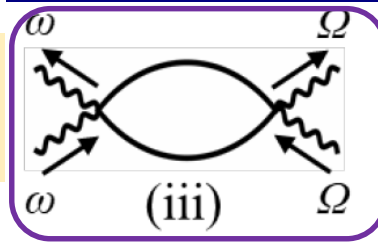
THz pump Ω -optical probe ω :
 $\omega \gg \Omega, 2\Delta, T_c \rightarrow$ (i)(ii) $\sim 1/\omega^2$

(i)

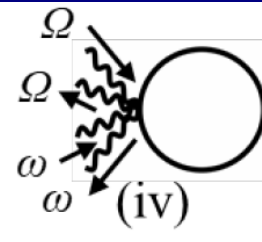
ω ω

(ii)

ω



(iii)



(iv)

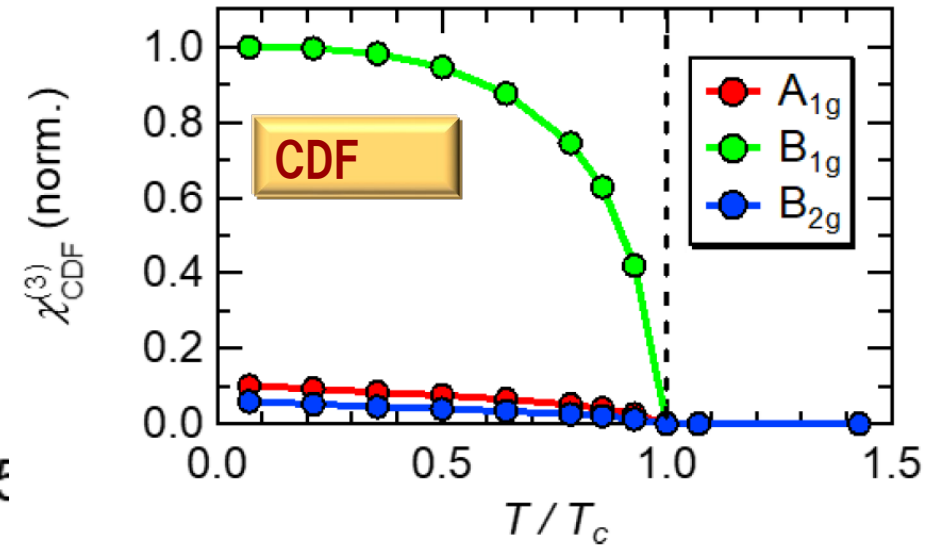
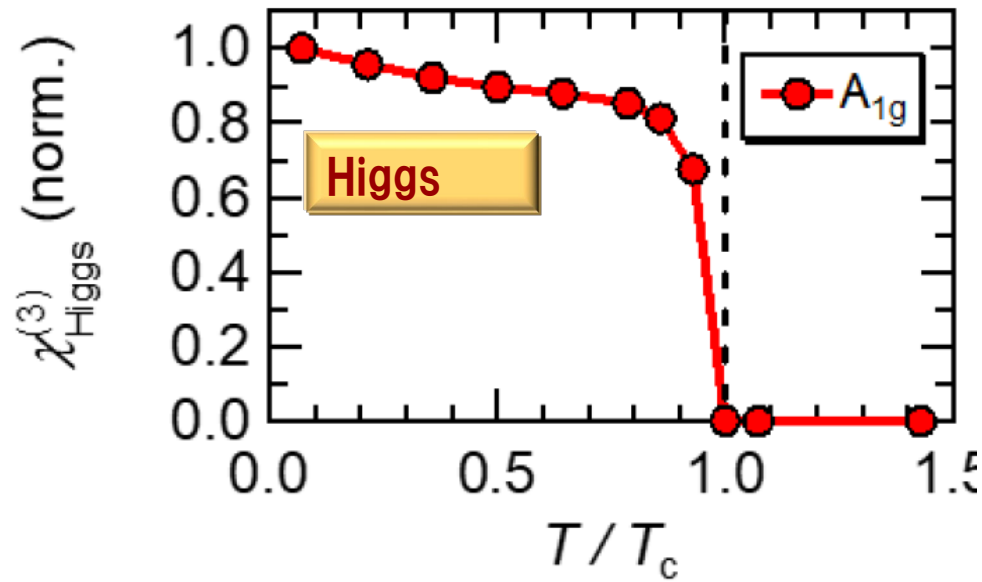
CDF

$$\chi_{\text{CDF}}^{(3)} = \frac{1}{N} \sum_{k,ijkl} \frac{\partial^2 \xi_k}{\partial k_i \partial k_j} e_i^{\text{Pump}} e_j^{\text{Pump}} \frac{\partial^2 \xi_k}{\partial k_k \partial k_l} e_k^{\text{Probe}} e_l^{\text{Probe}} \chi_{33}(k, 0)$$

$$- \frac{\left[\frac{1}{N} \sum_{k,ij} \frac{\partial^2 \xi_k}{\partial k_i \partial k_j} e_i^{\text{Pump}} e_j^{\text{Pump}} \chi_{33}(k, 0) \right] \left[\frac{1}{N} \sum_{k,kl} \frac{\partial^2 \xi_k}{\partial k_k \partial k_l} e_k^{\text{Probe}} e_l^{\text{Probe}} \chi_{33}(k, 0) \right]}{\frac{1}{N} \sum_k \chi_{33}(k, 0)}$$

THG susceptibility in HTC cuprate: theory

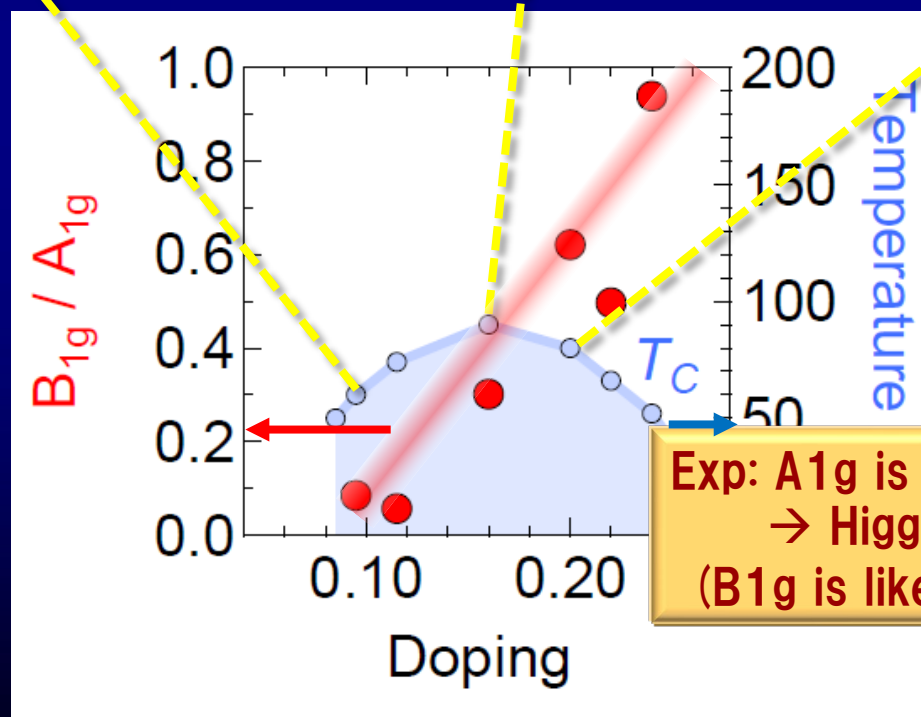
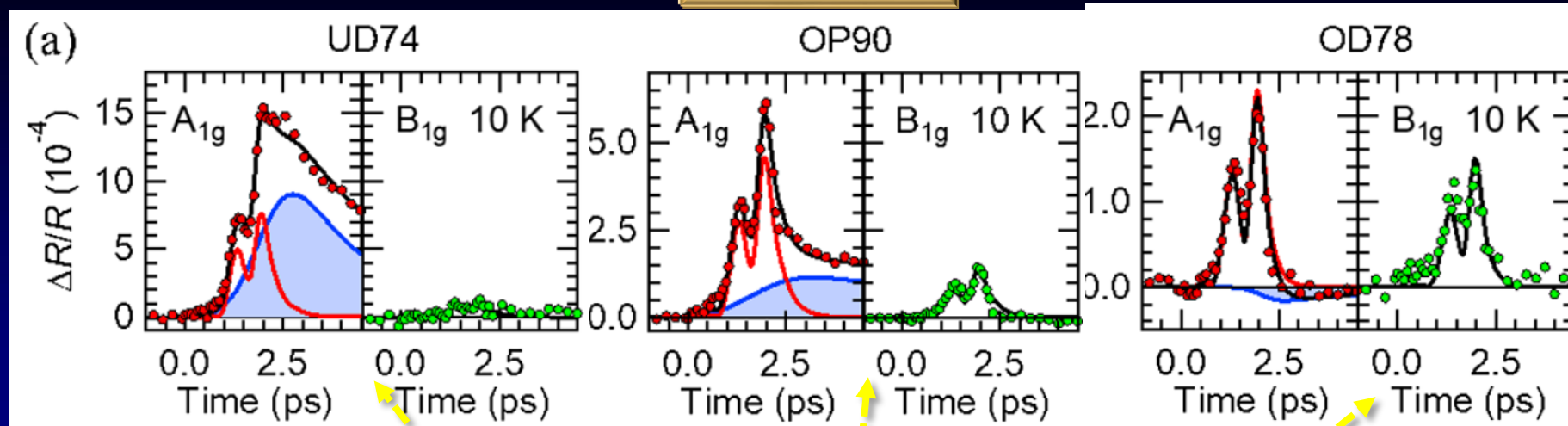
(Katsumi et al, PRL 2018)



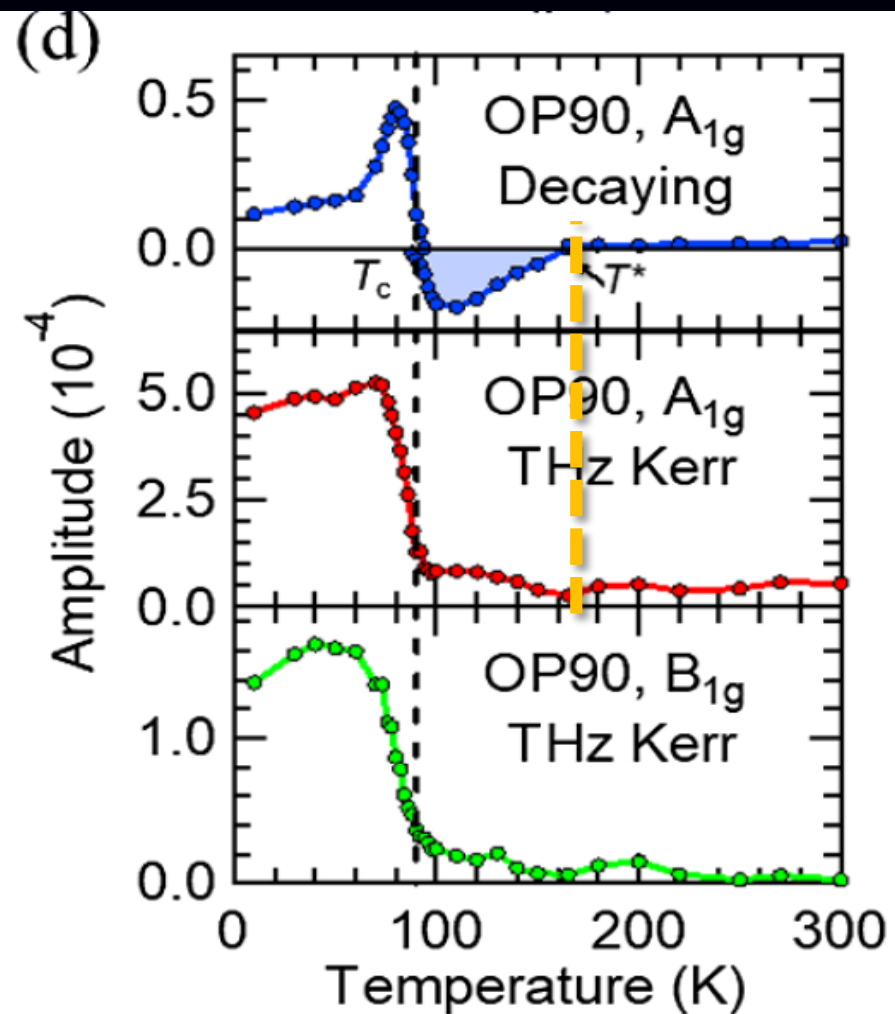
Theory: if A_{1g} dominates $\rightarrow$ Higgs dominates

Experiment

(Katsumi et al, PRL 2018)

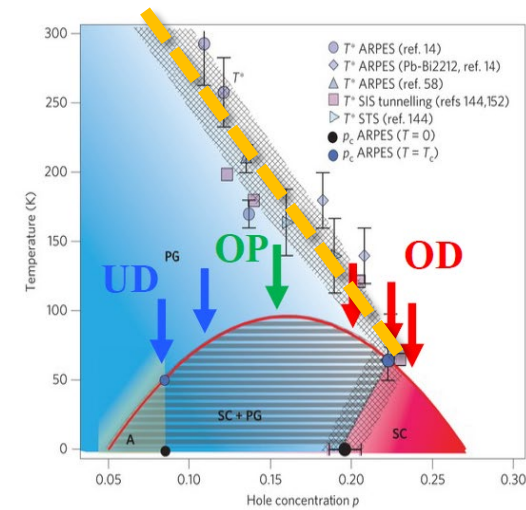


Exp: A_{1g} is always dominant
→ Higgs dominates,
(B_{1g} is likely due to CDF)



(Katsumi et al, PRL 2018)
Science 2011

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



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

SC current $\rightarrow$ linear coupling between Higgs & light

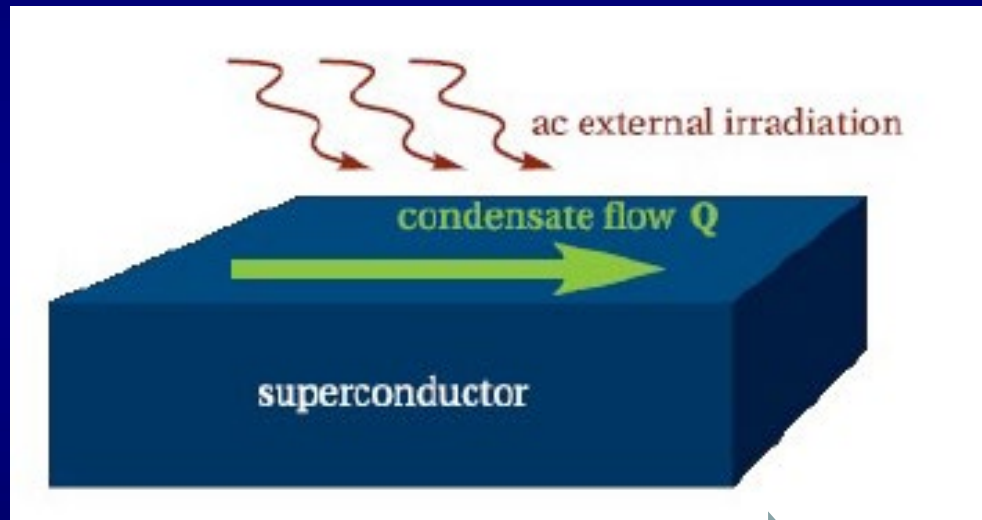
PRL 118, 047001 (2017)

PHYSICAL REVIEW LETTERS

week ending
27 JANUARY 2017

Amplitude Higgs Mode and Admittance in Superconductors with a Moving Condensate

Andreas Moor, Anatoly F. Volkov, and Konstantin B. Efetov



Condensate momentum



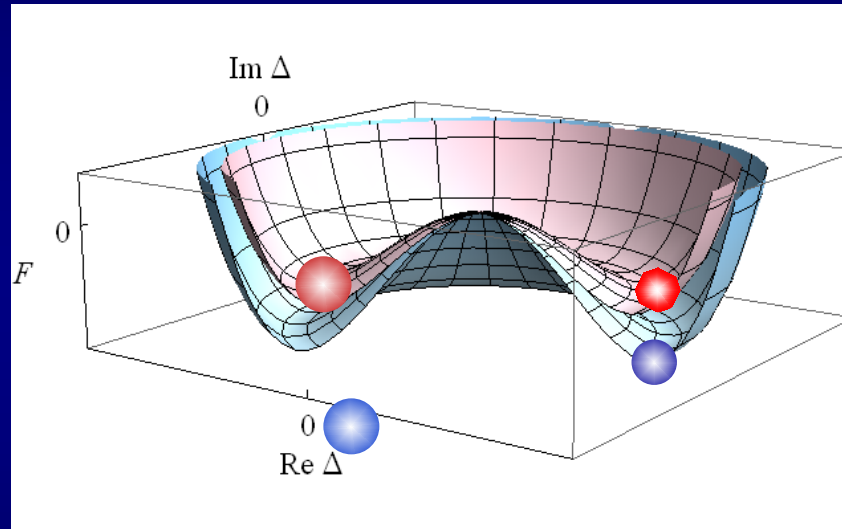
Higgs couples linearly with E_Ω

$$S_{\text{int}} = S_0 + 4C\text{Re} \int \delta|\Delta|_\Omega |\overline{\Delta}| \mathbf{Q}_0 \mathbf{Q}_{-\Omega} d\mathbf{r} \\ + C\text{Re} \int [2\delta|\Delta|_{2\Omega} |\overline{\Delta}| + (\delta|\Delta|_\Omega)^2] \mathbf{Q}_{-\Omega}^2 d\mathbf{r},$$

Collective modes in two-band superconductors (as in MgB_2)

→ Leggett mode & Higgs modes

(Krull, ..., Manske, Nature commun 2016;
Murotani, Tsuji & Aoki, PRB 2017)



Two-band formulation

(Murotani et al, PRB 2017)

Two-band Nambu spinors

$$\Psi_{\alpha k} \equiv \langle \alpha_{k\uparrow}^\dagger \alpha_{-k\downarrow}^\dagger \rangle, \quad \Psi_{\beta k} \equiv \langle \beta_{k\uparrow}^\dagger \beta_{-k\downarrow}^\dagger \rangle,$$

$$\Delta_\alpha = -V_{\alpha\alpha} \sum_k \Psi_{\alpha k} - V_{\alpha\beta} \sum_k \Psi_{\beta k},$$

$$\Delta_\beta = -V_{\beta\alpha} \sum_k \Psi_{\alpha k} - V_{\beta\beta} \sum_k \Psi_{\beta k},$$

inter-band

Two-band pseudospin rep

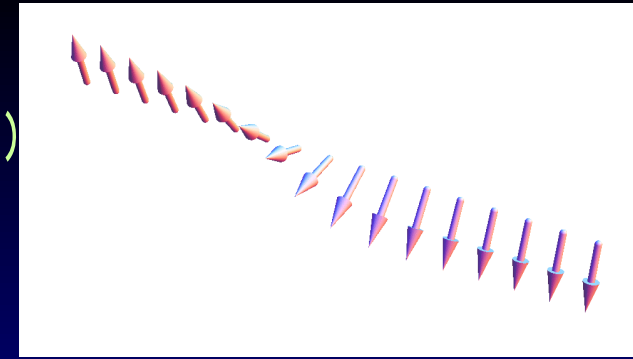
$$\mathcal{H}_{\text{BCS}} = \sum_{\gamma=\alpha,\beta} \sum_k 2\mathbf{b}_{\gamma k} \cdot \boldsymbol{\sigma}_{\gamma k},$$

$$\mathbf{b}_{\gamma k} = \left(-\Delta'_\gamma, -\Delta''_\gamma, \frac{\epsilon_{\gamma(k-eA(t))} + \epsilon_{\gamma(k+eA(t))}}{2} \right)$$

$$\frac{\partial \langle \boldsymbol{\sigma}_{\gamma k} \rangle}{\partial t} = 2\mathbf{b}_{\gamma k} \times \langle \boldsymbol{\sigma}_{\gamma k} \rangle.$$

$$\Delta'_\gamma = -V_{\gamma\alpha} \sum_k \langle \sigma_{\alpha k}^x \rangle - V_{\gamma\beta} \sum_k \langle \sigma_{\beta k}^x \rangle,$$

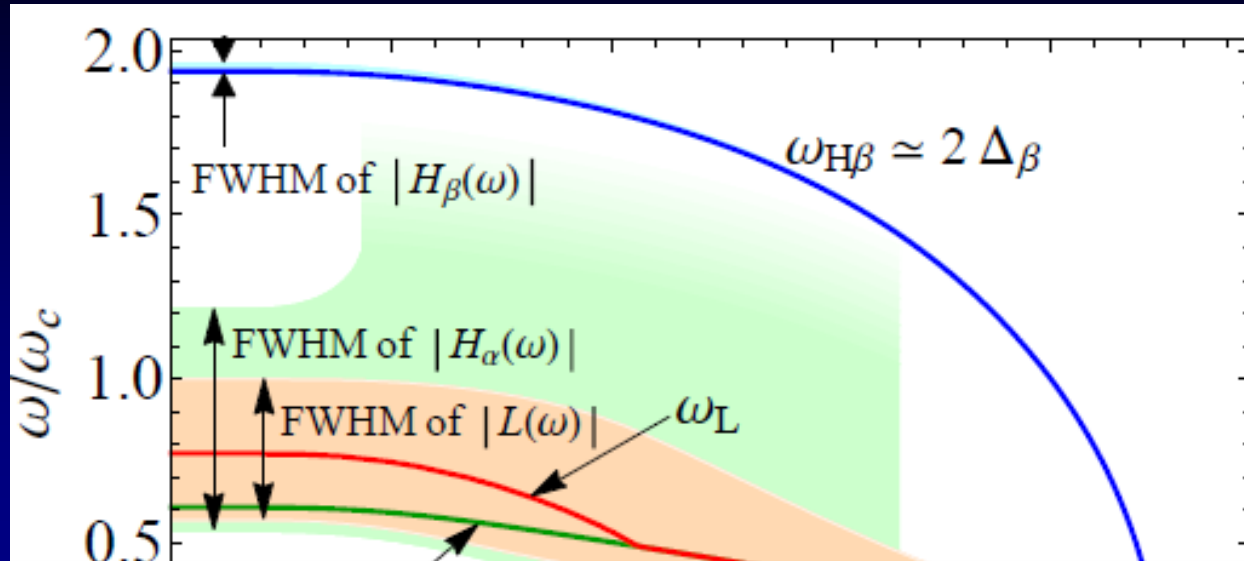
$$\Delta''_\gamma = -V_{\gamma\alpha} \sum_k \langle \sigma_{\alpha k}^y \rangle - V_{\gamma\beta} \sum_k \langle \sigma_{\beta k}^y \rangle,$$



Eqn of motion for the phase difference

$$\delta[\theta_\alpha(\omega) - \theta_\beta(\omega)] = -e^2 A^2(\omega) i \omega L(\omega),$$

Leggett & Higgs modes vs T (Murotani et al, PRB 2017)



Higgs and Leggett modes

→ *very good probe for superconductors*

(with even Leggett mode optically-excitabile)

Higgs/Leggett nonlinear spectroscopy

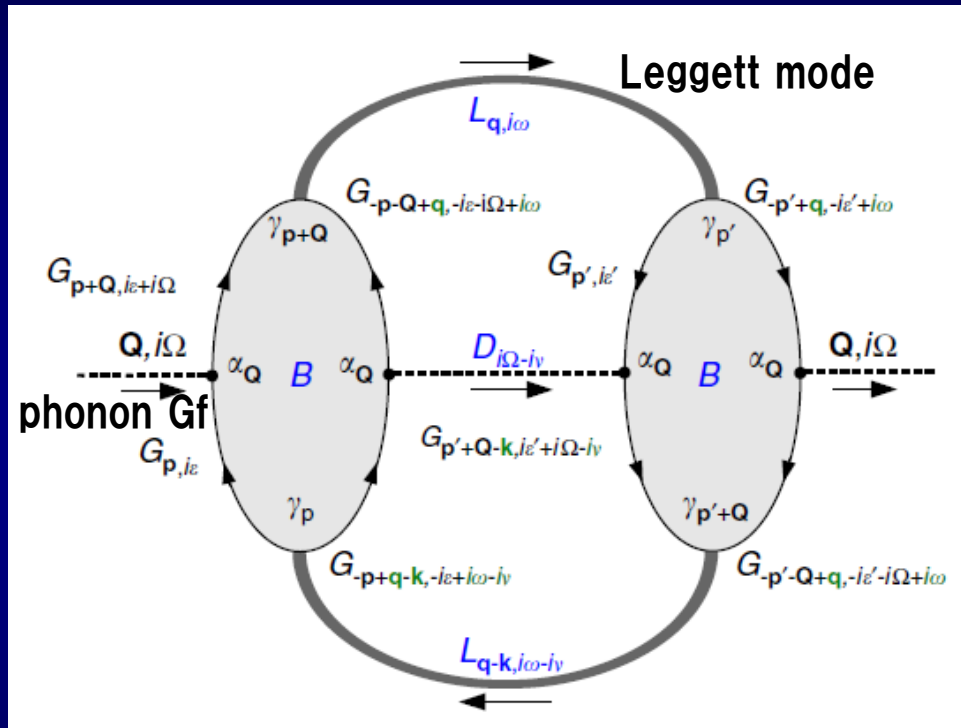
Cf. Jujo, JPSJ 2017; Murotani & Shimano, PRB 2019

Higgs-light nonlinear coupling ← enhanced by nonmag impurities

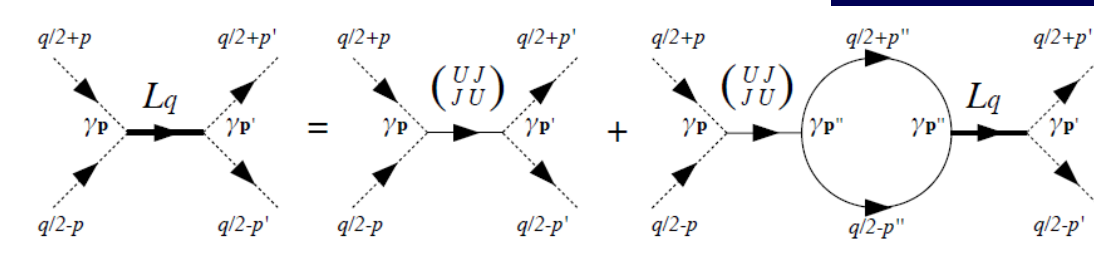
Leggett-light nonlinear coupling ← unaffected by nonmag impurities

Can collective modes actually affect SC itself ?

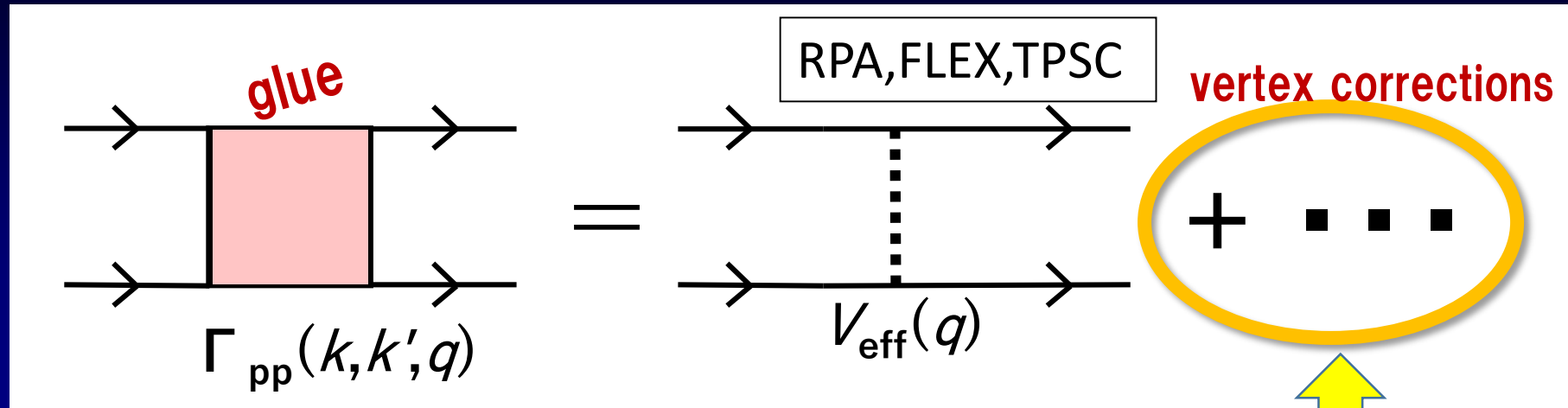
Σ^{phonon} in el-ph system (Liu et al, Nat Commun 2016)



Bethe-Salpeter eqn for the Leggett mode



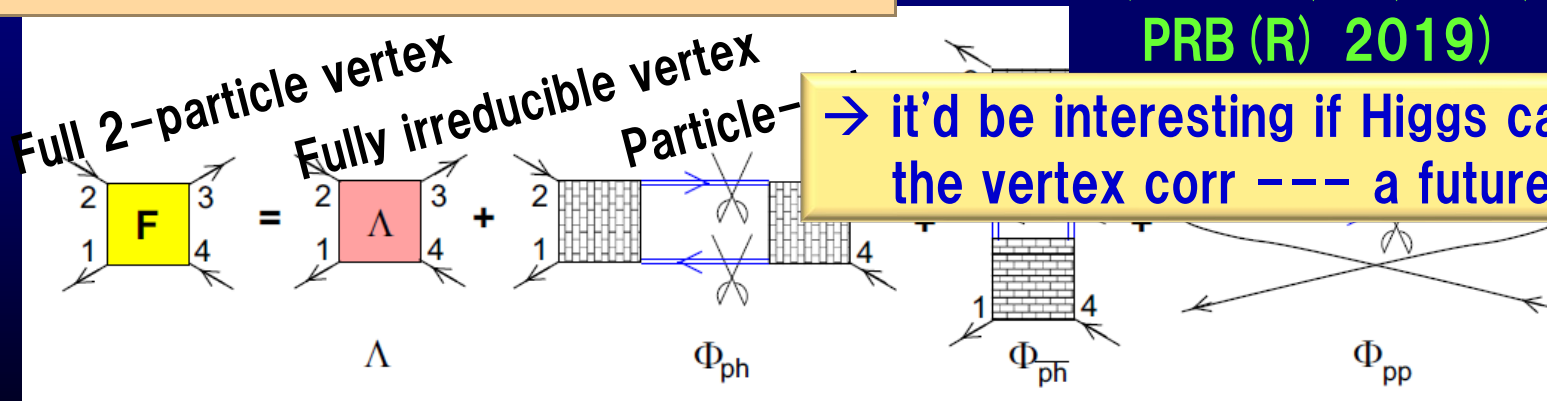
Vertex corrections usually neglected



- If vertex corr at work for SC $\rightarrow$ non-bosonic exchange

main reason why T_c is so low in electron mechanisms SC (Kitatani, ..., Aoki, Held, PRB (R) 2019)

DGA (Rohringer, ..., Held, RMP 2018)



$\rightarrow$ it'd be interesting if Higgs can affect the vertex corr --- a future problem

Summary: Superconductors in nonequilibrium

Many-body

Floquet

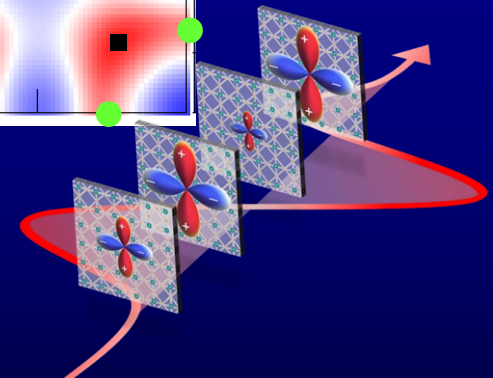
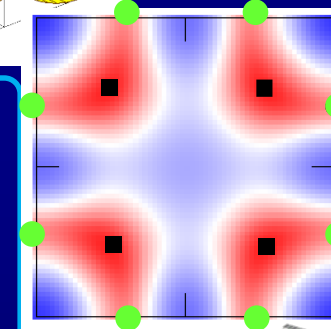
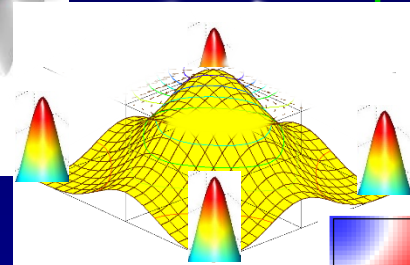
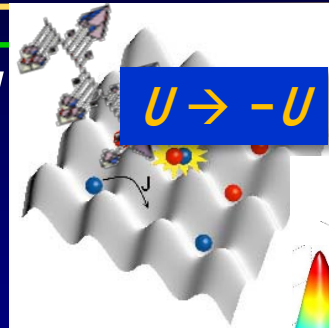
Steady/transient

✓ Attraction-repulsion conversion
(Tsuji; Tsuji)

✓ η -pairing
(Kitamura)

✓ k -dep relaxation
(Tsuji; Sayyad)

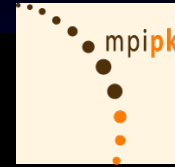
✓ Higgs dynamics: phonon mechanism SC (Tsuji)
unconventional SC (Tsuji)



→ A lot of future directions along
Many-body, Floquet, steady/transient

fiat lux

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