Angle-Resolved Two-Photon Photoemission of Mott Insulator

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H. Onodera *et al.*, cond-mat/0503267

OUTLINE

Introduction

Mott insulator: Copper oxides

Why is the upper Hubbard band important?

Two-photon photoemission (2PPES)

The "ω" and "2ω" processes

The "simultaneous" and "cascade" processes

Angle-resolved 2PPES for insulating cuprates in two dimensions

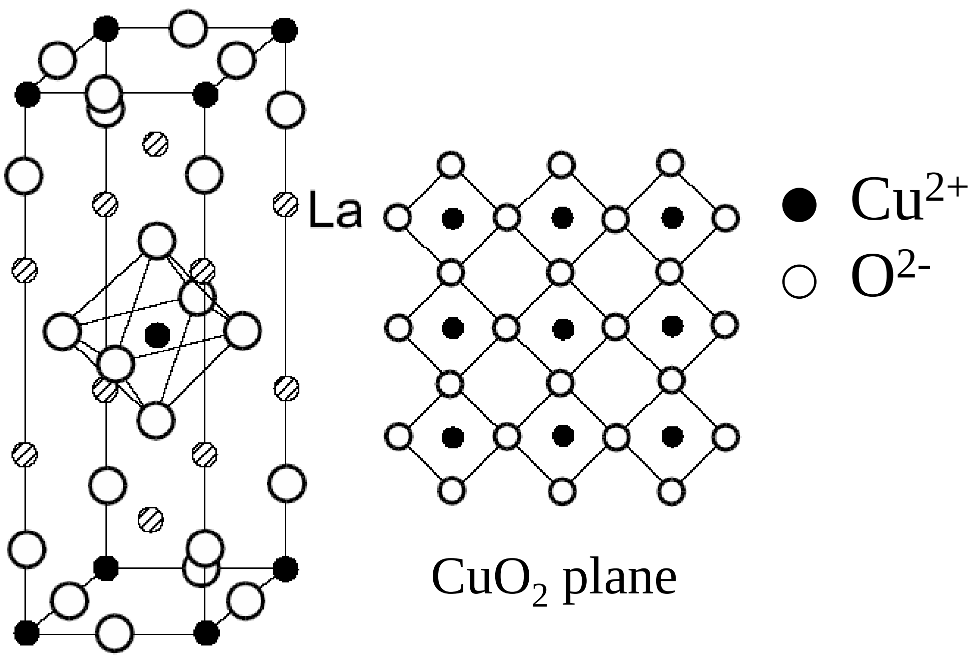
Pump photon

- Zhang-Rice singlet band
- Non-bonding oxygen band

Predictions for future experiments

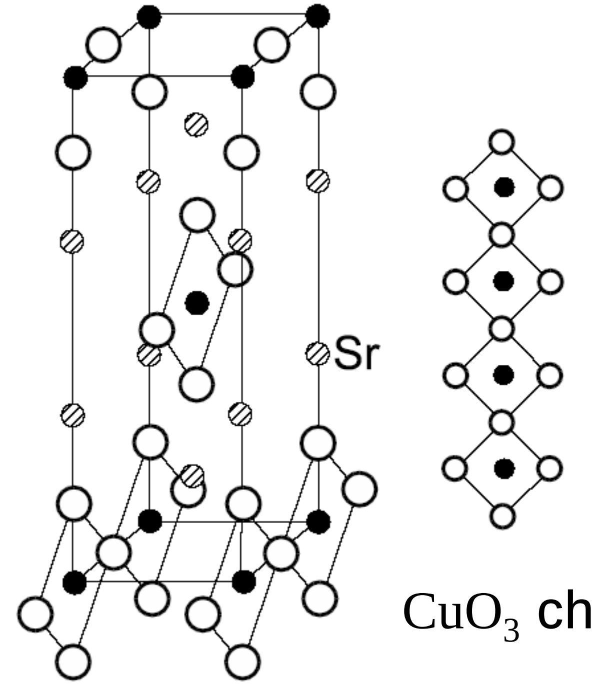
Crystal structures of insulating cuprates

La_2CuO_4 (two dimension)



CuO_2 plane

Sr_2CuO_3 (one dimension)



CuO_3 chain

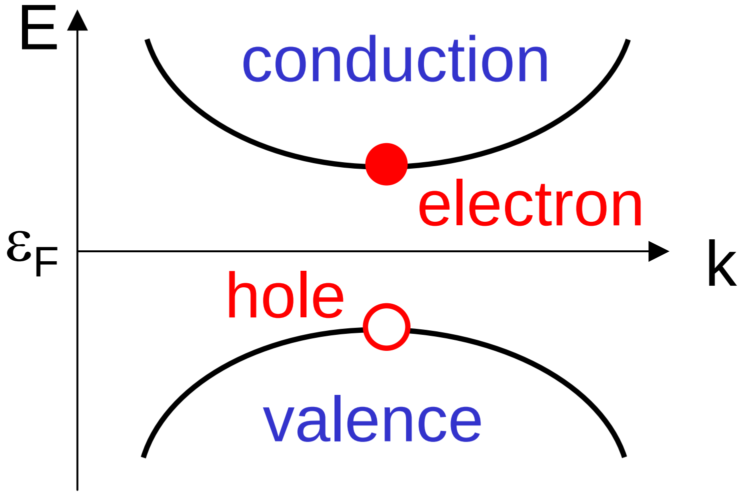
Cu^{2+} $3d^9$ 1 hole on each x^2-y^2 orbital

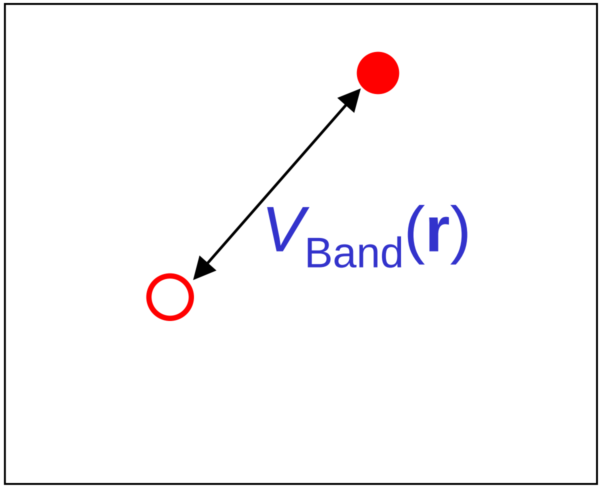
localized spin → antiferromagnetic exchange interaction

J~1000K-2000K

Photo excitation: Band insulator vs. Mott insulator

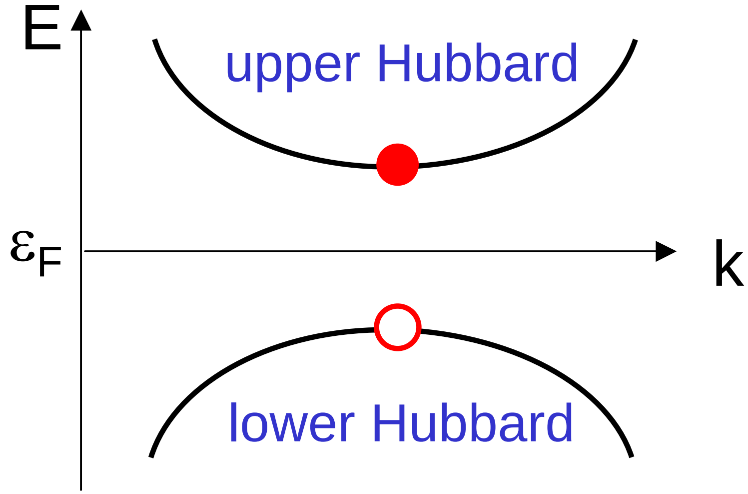
Band

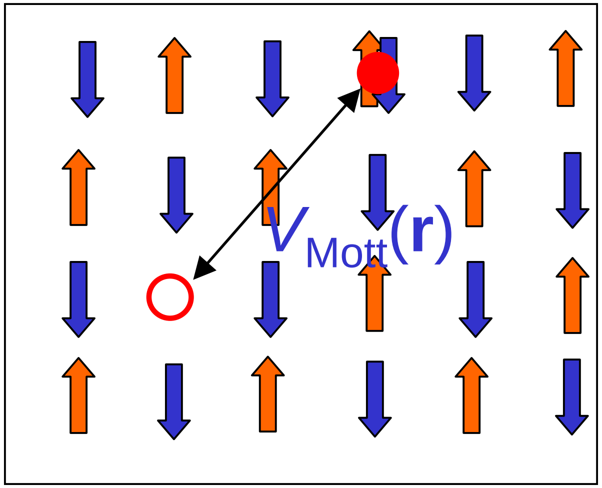




exciton picture

Mott

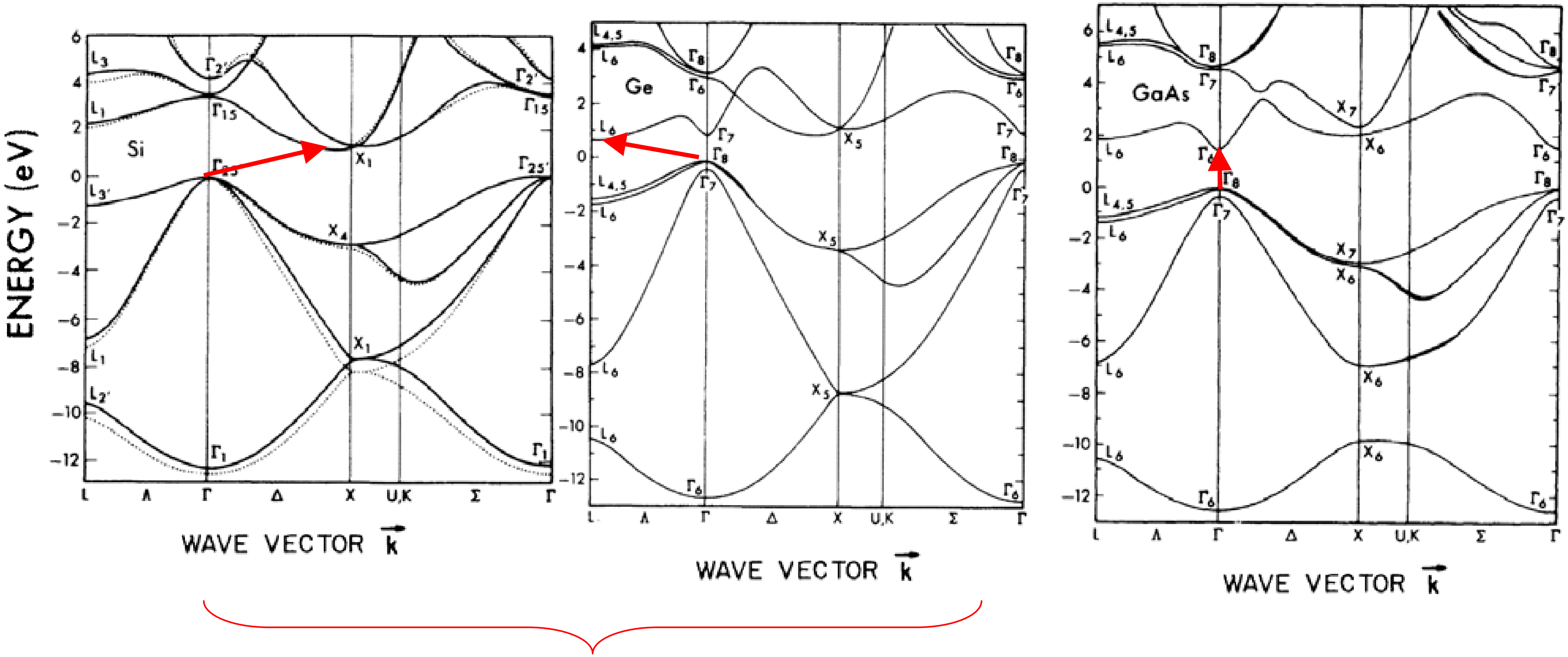




new concept !

Band structure of semiconductors

Si Ge GaAs



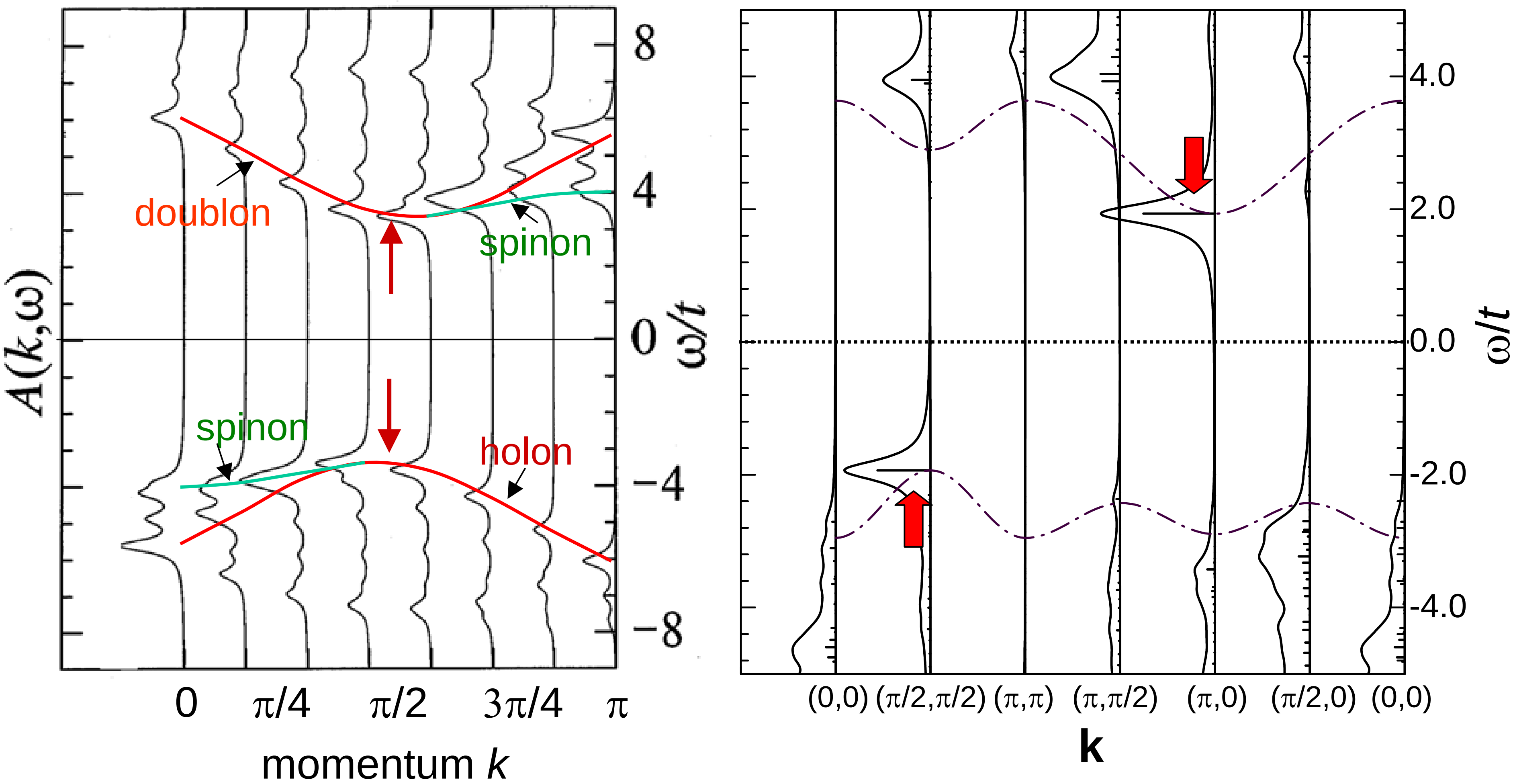
indirect gap

direct gap

[J. R. Chelikowsky and M. L. Cohen: Phys. Rev. B **14**, 556 (1976)]

Band structure of Mott insulators: Theoretical prediction

one dimension (14-site *t*-*U*) two dimension (4X4 *t*-*t'*-*t''*-*U*)



direct gap

indirect gap
because of *t'* and *t''*

[K. Tsutsui, T. T. and S. Maekawa: Phys. Rev. Lett. **83**, 3705 (1999); Phys. Rev. B **61**, 7180 (2000)]

Why is the upper Hubbard band (UHB) important?

UHB characterizes the “particle-hole” excitation.

↓

It is necessary to understand its nature for future application of the Mott insulators.

We propose a method to see the momentum-dependent UHB.

Angle-resolved two-photon photoemission

(AR-2PPES)

cf. Inverse photoemission — low-energy resolution

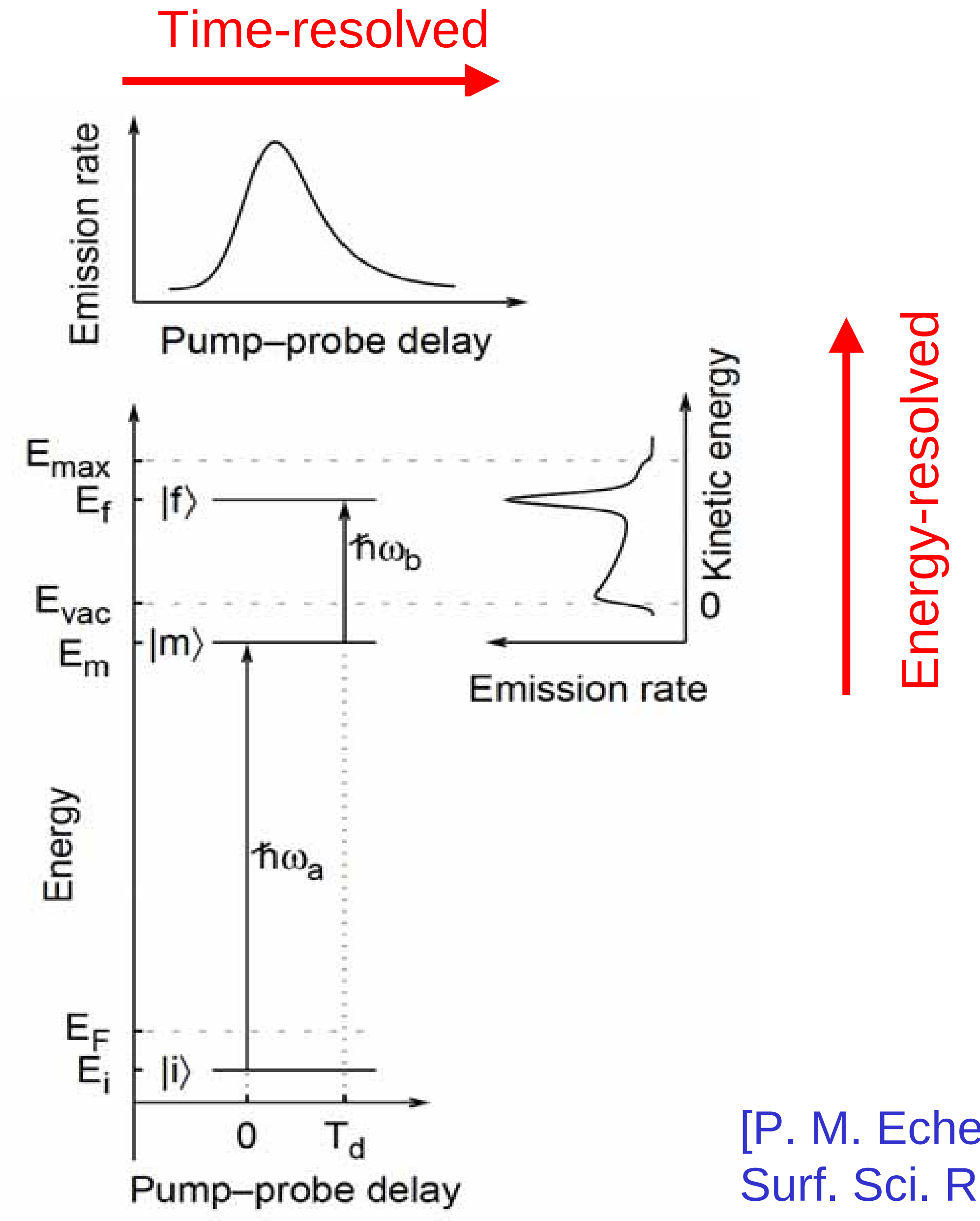
Electron-energy loss spectroscopy (EELS)

Resonant inelastic X-ray scattering (RIXS)

Indirect measurements

[2D cuprates: PRL**83**, 3705(1999); Science**288**, 1811(2000)]

Energy diagram for 2PPES



[P. M. Echenique *et al*.,
Surf. Sci. Rep.**52**, 219 (2004)]

Two-photon photoemission

- Image-potential states at metal surfaces and their decay

Time, energy- and angle-resolved modes

Reviews: M. Weinelt, J. Phys. Condens. Matter **14**, R1099 (2002)

P. M. Echenique *et al*., Surf. Sci. Rep.**52**, 219 (2004)

- High-Tc cuprate superconductor: $Bi_2Sr_2CaCu_2O_{8+\delta}$

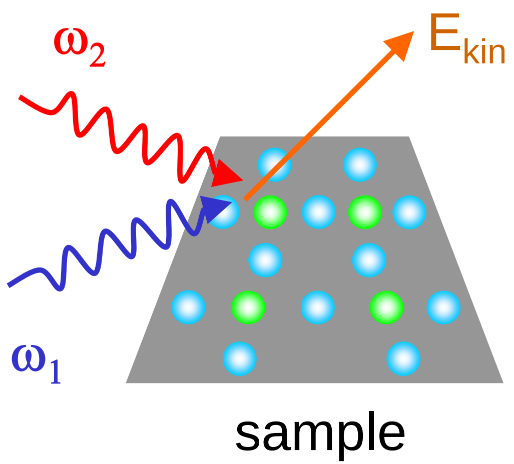
Time-resolved mode

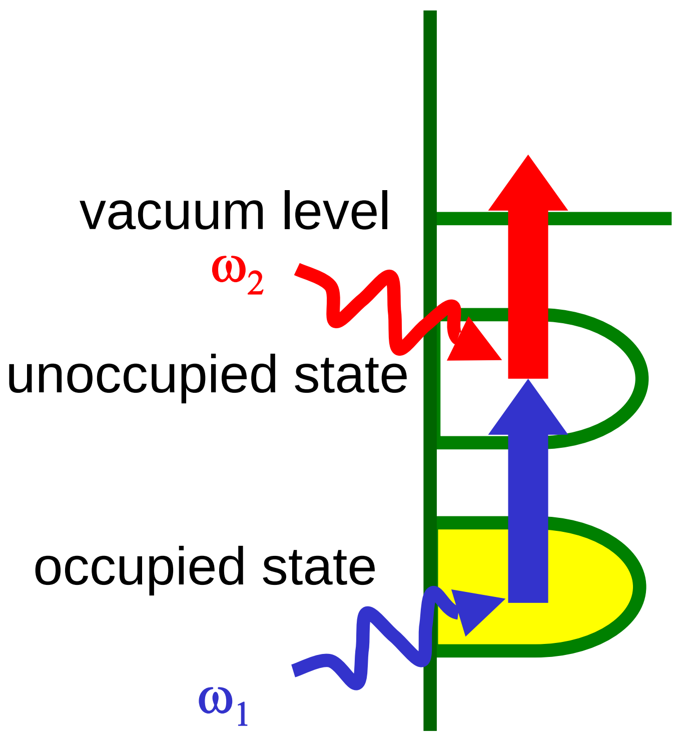
W. Nessler *et al*., Phys. Rev. Lett. **81**, 4480 (1998)

Energy-resolved mode

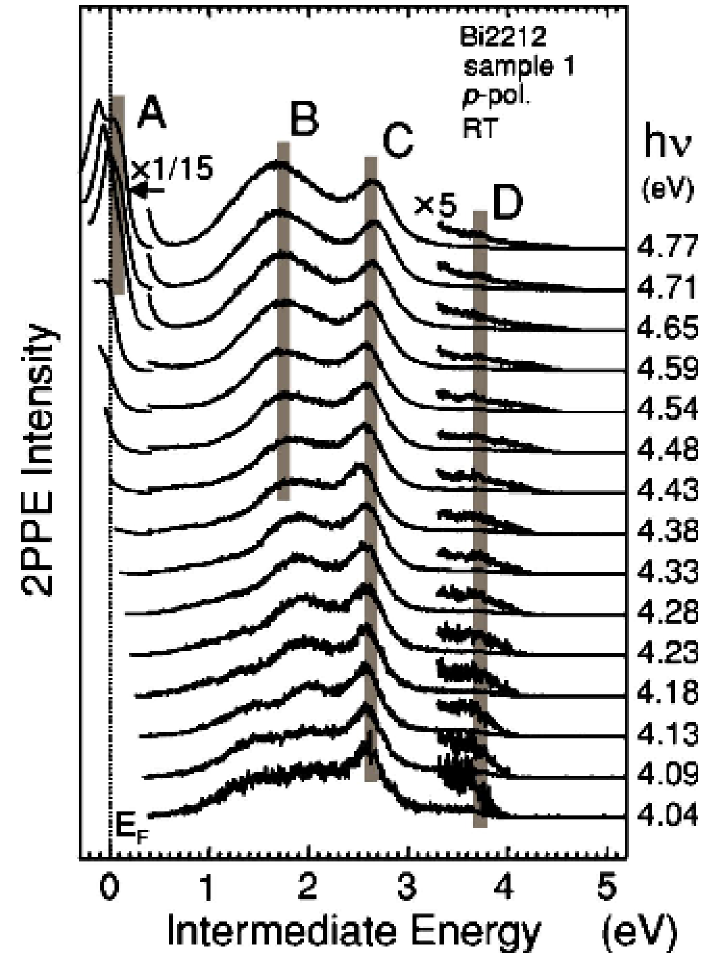
Y. Sonoda and T. Munakata, Phys. Rev. B **70**, 134517 (2004)

No experimental works in Mott insulators!



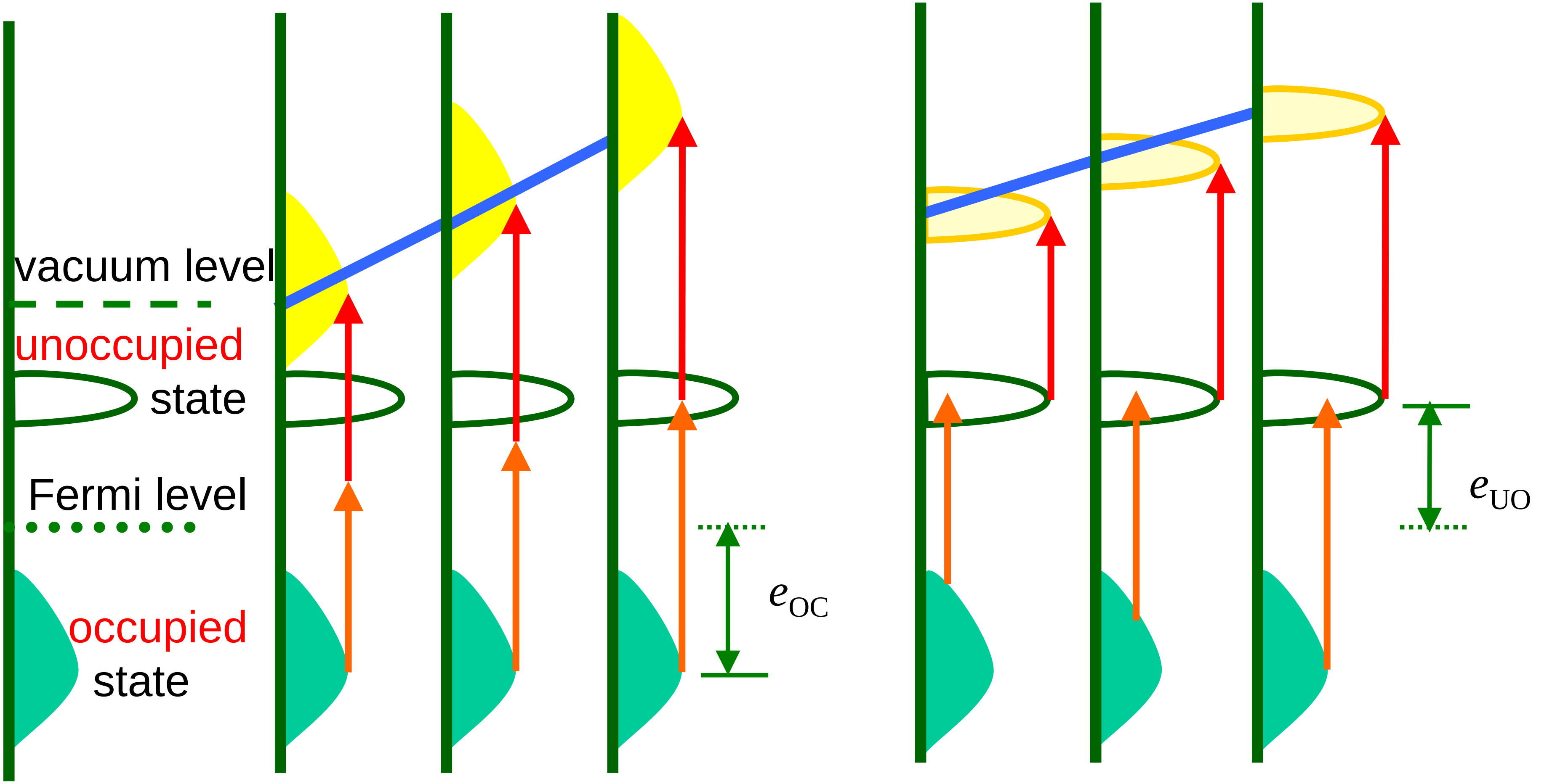


Y. Sonoda and T. Munakata
Phys. Rev. B **70**, 134517 (2004)



A:excitations from lower-Hubbard band
B:excitations from upper Hubbard band
C,D: excitations perpendicular to CuO_2

The "2ω" and "ω" processes



The "2ω" process

Information on occupied state

$E_{kin} = -e_{OC} + 2\omega$

The "ω" process

Information on unoccupied state

$E_{kin} = e_{UO} + \omega$

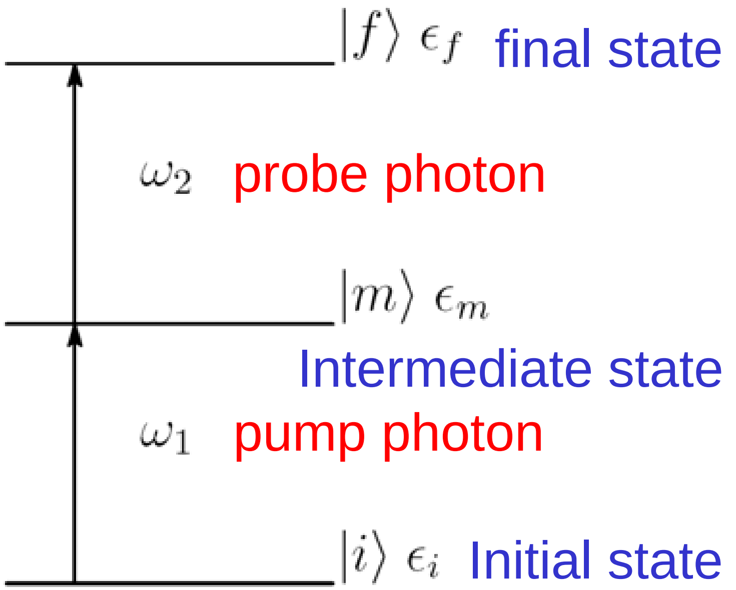
Possible to separate the two states from the ω dependence

Formulation for two-photon photoemission

Three-level model

$$H = \sum_{\alpha=i,m,f} \epsilon_\alpha |\alpha\rangle\langle\alpha| - \mu \mathbf{E}(t)$$

$$\mu = V_{mi}|m\rangle\langle i| + V_{mf}|f\rangle\langle m| + \mathrm{H.c.}$$



Optical Bloch equation

$$\frac{d}{dt}\rho_{\alpha\beta} = -i\left[H, \rho\right]_{\alpha\beta} - \Gamma_{\alpha\beta}\rho_{\alpha\beta}$$

$$\rho = \begin{pmatrix} \rho_{ii}^{(0)} & \rho_{im}^{(1)} & \rho_{if}^{(2)} \\ \rho_{mi}^{(1)} & \rho_{mm}^{(2)} & \rho_{mf}^{(3)} \\ \rho_{fi}^{(2)} & \rho_{fm}^{(3)} & \rho_{ff}^{(4)} \end{pmatrix} \qquad \Gamma = \begin{pmatrix} 0 & \Gamma_{im} & \Gamma_{if} \\ \Gamma_{im} & \Gamma_{mm} & \Gamma_{mf} \\ \Gamma_{if} & \Gamma_{mf} & \Gamma_{ff} \end{pmatrix}$$

ρ: density operator, where the superscript (#) denotes the order of the electric field.

Γ: retaliation rate

“simultaneous” process

Virtual excitation to $|m\rangle$

$$\rho_{ii} \to \rho_{im} \to \rho_{if} \to \rho_{mf} \to \rho_{ff} \Rightarrow I^{(s)}$$

$$I^{(s)}_{2PPE}(E,\omega) = 2\pi \sum_{i,f} \rho^{(0)}_{ii} \left| \sum_m \frac{V_{im}V_{mf}}{\epsilon_m - \epsilon_i - \omega_1 - i\Gamma_{im}} \right|^2 \delta(\epsilon_f - \epsilon_i - \omega_1 - \omega_2)\delta(E - \epsilon_f)$$

Γ_{im} : phase-relaxation rate

“sequence” or “cascade” process

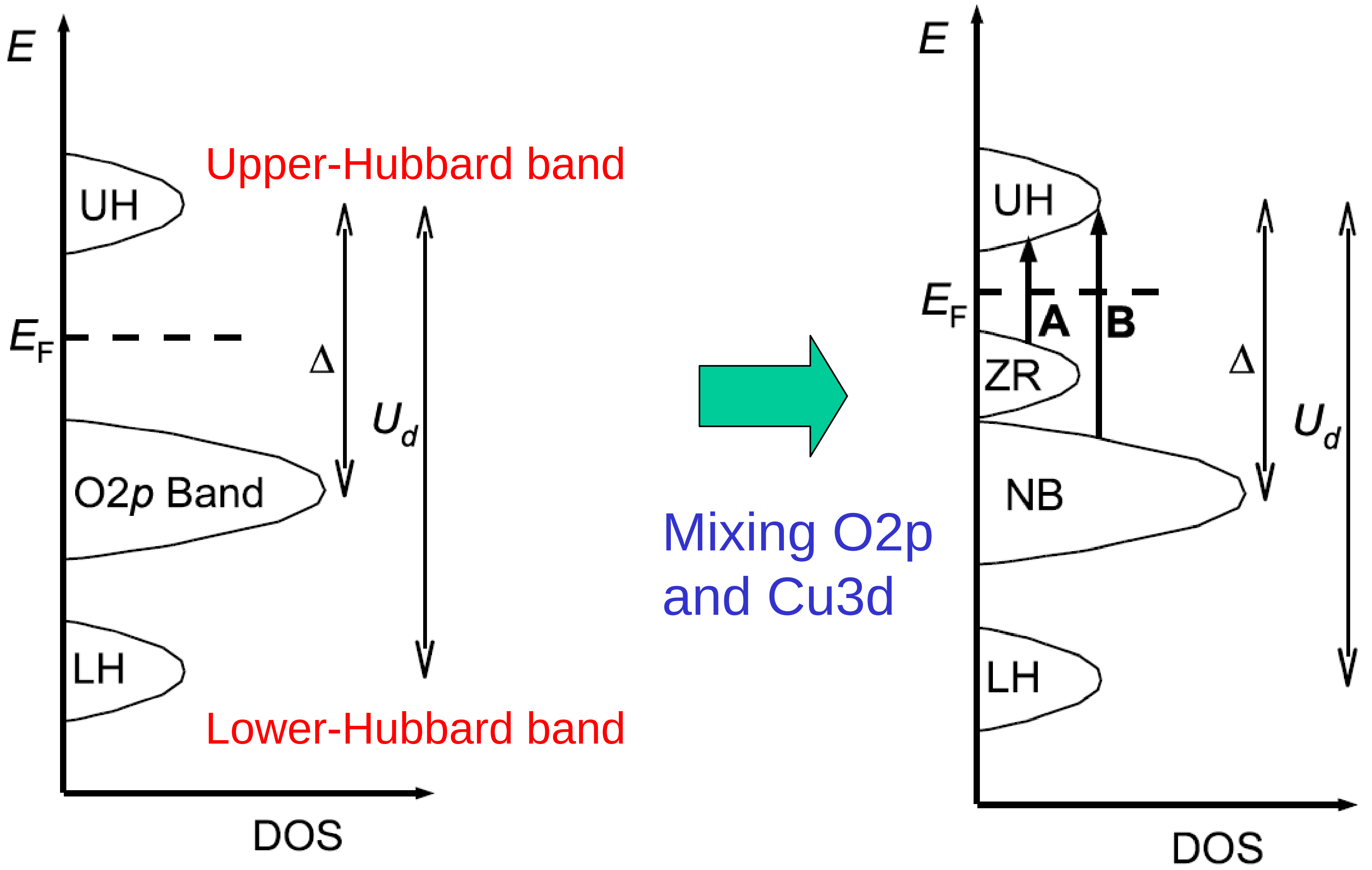
Real excitation to $|m\rangle$

$$\rho_{ii} \to \rho_{im} \to \rho_{mm} \to \rho_{mf} \to \rho_{ff} \Rightarrow I^{(c)}$$

$$I^{(c)}_{2PPE}(E,\omega) = (2\pi)^2 \sum_{imf} \rho^{(0)}_{ii} \frac{|V_{im}|^2 \, |V_{mf}|^2}{\Gamma_{mm}} \delta(\epsilon_m - \epsilon_i - \omega_1)\delta(\epsilon_f - \epsilon_m - \omega_2)\delta(E - \epsilon_f)$$

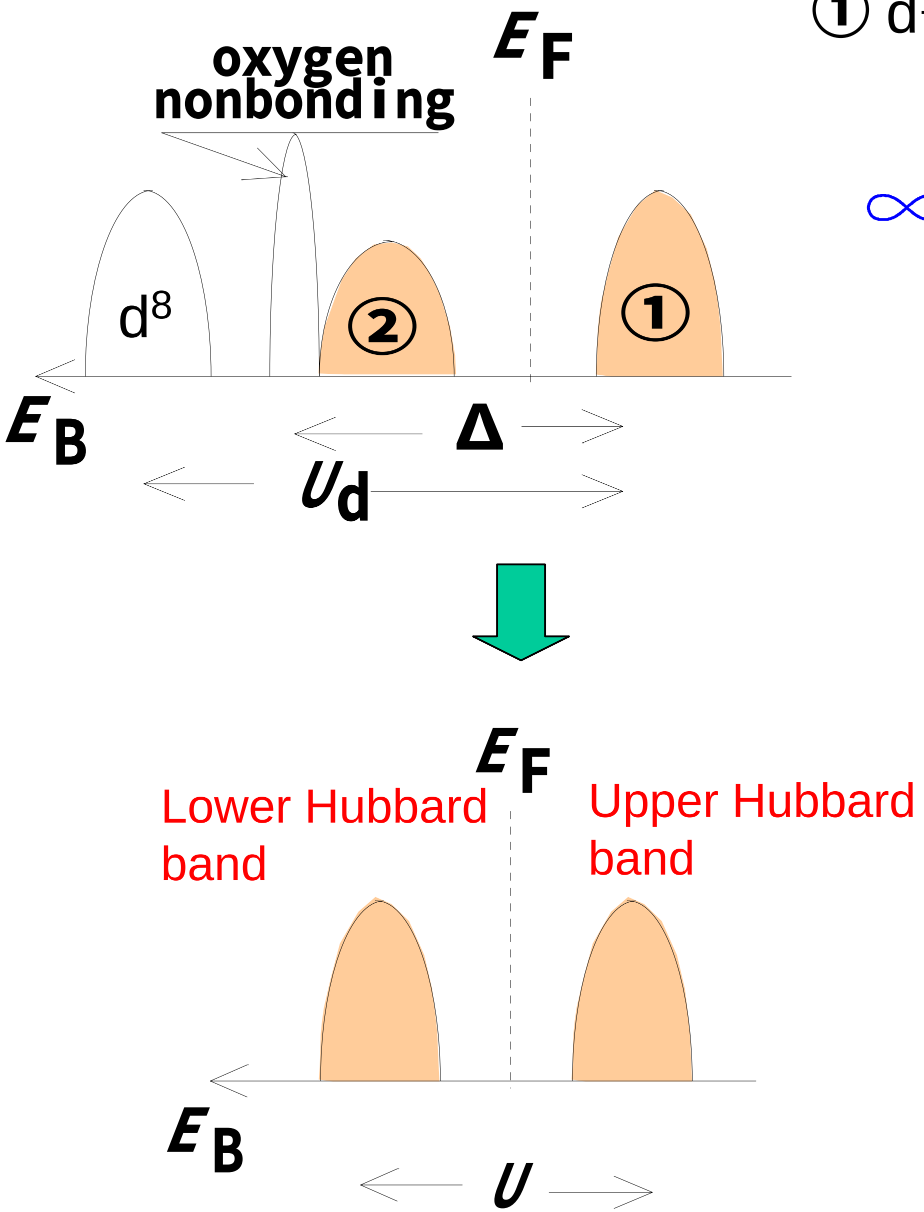
Γ_{mm} : energy-relaxation rat

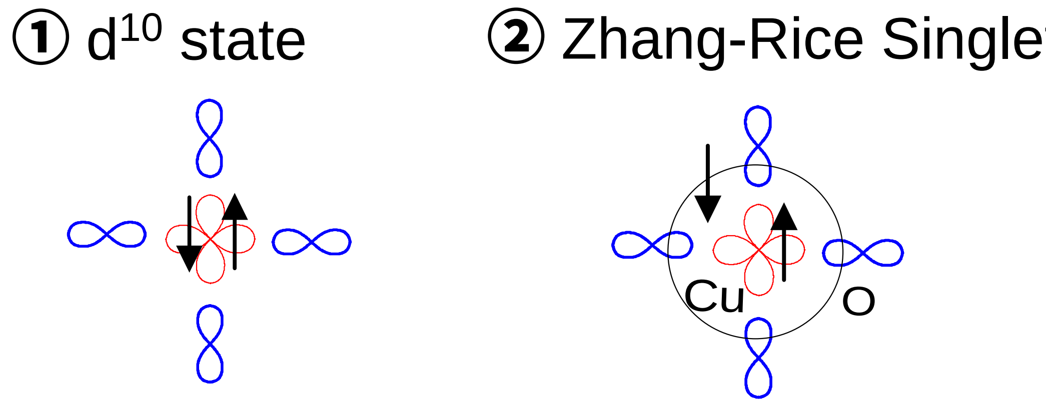
Density of states for insulating cuprates

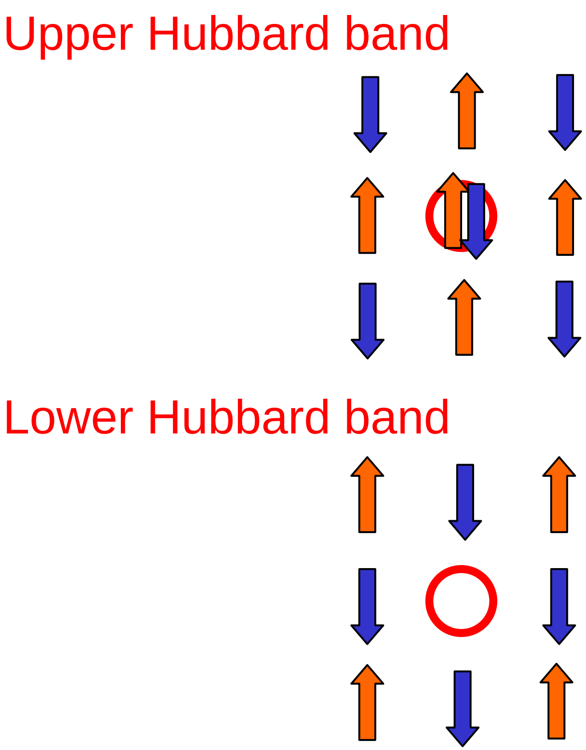


Emergence of a bound state:
Zhang-Rice singlet band

Mapping from three-band model to single-band model

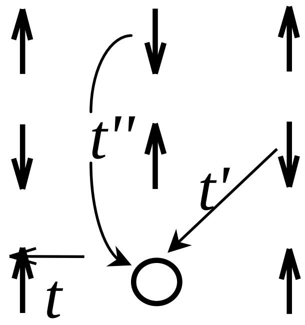






t-t'-t''-U Hubbard model

$$H = -t\sum_{\langle ij\rangle,\sigma}\left(c^{+}_{i,\sigma}c_{j,\sigma}+\mathrm{h.c.}\right) - t'\sum_{\langle ij\rangle',\sigma}\left(c^{+}_{i,\sigma}c_{j,\sigma}+\mathrm{h.c.}\right)$$
$$-t''\sum_{\langle ij\rangle'',\sigma}\left(c^{+}_{i,\sigma}c_{j,\sigma}+\mathrm{h.c.}\right) + U\sum_{i} n_{i,\uparrow}n_{i,\downarrow}$$



$U/t = 10$

Cuprates in 2D: $Ca_2CuO_2Cl_2$

t = 0.35 eV, t' = - 0.12 eV, t'' = 0.08 eV

Angle-resolved 2PPES for cuprates

"simultaneous" process

$$I^{(s)}(\omega_1,\omega_2,E_{\rm kin},\mathbf{k}) = \sum_F \left| \sum_M \frac{\langle F|c_{\mathbf{k}}|M\rangle\langle M|j_x|I\rangle}{E_M - E_I - \omega_1 - i\Gamma_{IM}} \right|^2 \delta(\omega_1+\omega_2+E_I-E_F-E_{\rm kin})$$

"sequence" or "cascade" process

$$I^{(c)}(\omega_1,\omega_2,E_{\rm kin},\mathbf{k}) = \sum_F \sum_M \frac{\left|\langle F|c_{\mathbf{k}}|M\rangle\right|^2 \left|\langle M|j_x|I\rangle\right|^2}{\Gamma_{MM}} \delta\left(E_M - E_I - \omega_1\right)\times\delta\left(\omega_1+\omega_2+E_I-E_F-E_{\rm kin}\right)$$

The spectra are calculated for finite-size clusters such as a 4x4 lattice.

Initial state $|I\rangle$: the standard Lanczos method

$I^{(s)}, I^{(c)}$: the correction-vector method based on the conjugate gradient technique

$\Gamma_{MM} = 2\Gamma_{IM} = 0.4t$, neglecting the pure dephasing

Calculation of spectrum in the second-order process

Correction-vector method

$$I^{(s)}(\omega_1, \omega_2, E_{\mathrm{kin}}, \mathbf{k}) = \sum_F \left| \sum_M \frac{\langle F | c_{\mathbf{k}} | M \rangle \langle M | j_x | I \rangle}{E_M - E_I - \omega_1 - i\Gamma_{\mathrm{IM}}} \right|^2 \delta(\omega_1 + \omega_2 + E_I - E_F - E_{\mathrm{kin}})$$

Correction vector:

$$|\phi\rangle = \frac{1}{H - E_I - \omega_1 - i\Gamma_{\mathrm{IM}}} j_x |I\rangle \quad \Longrightarrow \quad (H - E_I - \omega_1 - i\Gamma_{\mathrm{IM}})|\phi\rangle = j_x |I\rangle$$

This is evaluated by the conjugate-gradient method.

$$I^{(s)}(\omega_1, \omega_2, E_{\mathrm{kin}}, \mathbf{k}) = \sum_F \left| \langle F | c_{\mathbf{k}} | \phi \rangle \right|^2 \delta(\omega_1 + \omega_2 + E_I - E_F - E_{\mathrm{kin}})$$

This is easily calculated by the standard Lanczos algorithm (recursion method).

Calculation of spectrum in the second-order process

$$I^{(c)}(\omega_1, \omega_2, E_{\rm kin}, \mathbf{k}) = \sum_F \sum_M \frac{\left|\langle F | c_{\mathbf{k}} | M \rangle\right|^2 \left|\langle M | j_x | I \rangle\right|^2}{\Gamma_{MM}} \delta\left(E_M - E_I - \omega_1\right) \times \delta\left(\omega_1 + \omega_2 + E_I - E_F - E_{\rm kin}\right)$$

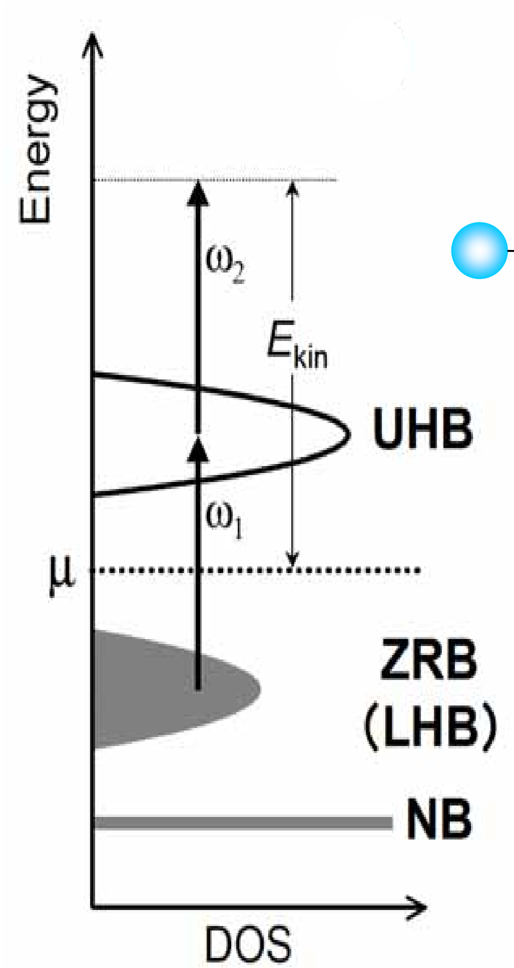
$$\approx \sum_F \sum_{M'=1}^{M'_{\rm max}} \frac{\left|\langle F | c_{\mathbf{k}} | M' \rangle\right|^2 \left|\langle M' | j_x | I \rangle\right|^2}{\Gamma_{MM}} \delta\left(E_{M'} - E_I - \omega_1\right) \times \delta\left(\omega_1 + \omega_2 + E_I - E_F - E_{\rm kin}\right)$$

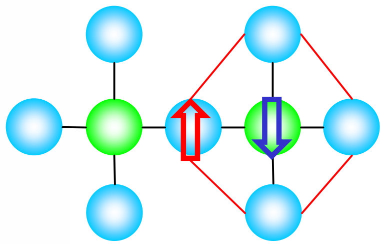
$|M'\rangle$: approximate eigenstates with large value of $|\langle M' | j_x | I \rangle|^2$, which are evaluated by the standard Lanczos algorithm $M'_{\rm max} \sim 20$

For each $|M'\rangle$, we perform the Lanczos process.

Possible excitation due to the pump photon ω_1

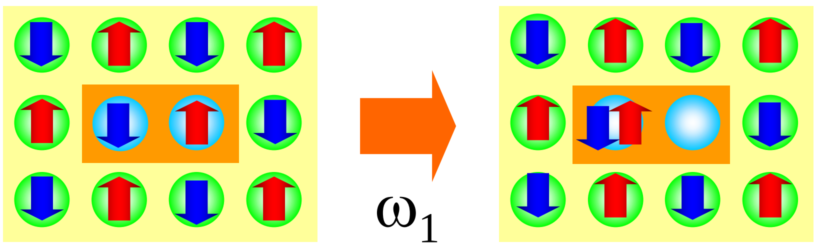
Excitation from Zhang-Rice singlet band





$\omega_1 \sim$ 2-3eV

Single-band picture



Current operator in the Hubbard model

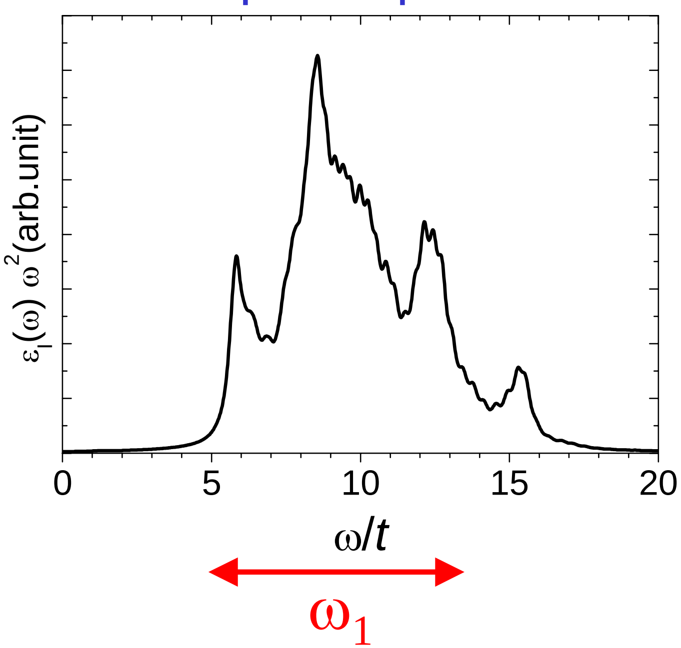
$$j_x = i \sum_{\mathbf{k},\sigma} \alpha_{\mathbf{k}} c^{\dagger}_{\mathbf{k},\sigma} c_{\mathbf{k},\sigma}$$

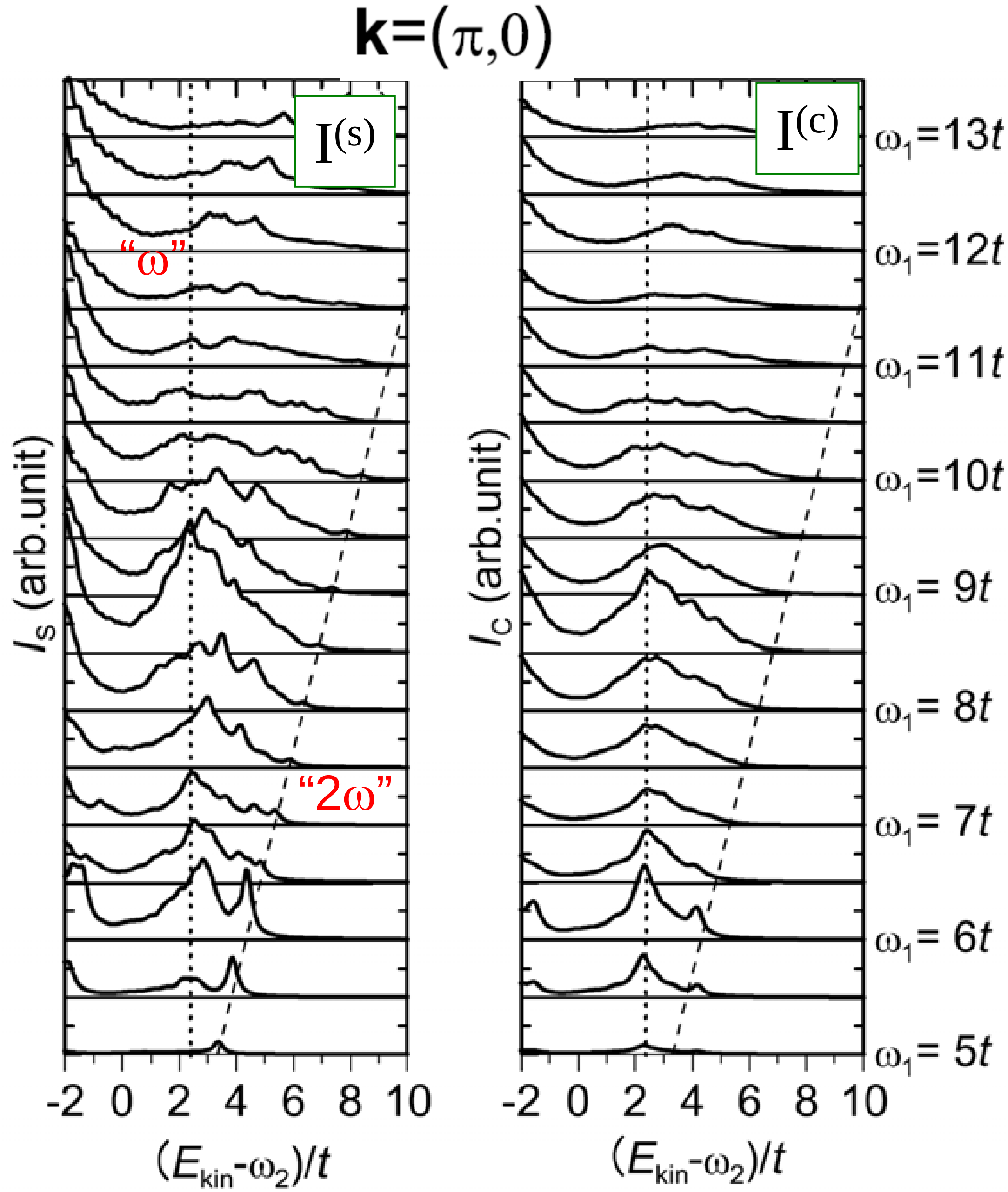
$$\alpha_{\mathbf{k}} = -2t \sin k_x - 4t' \sin k_x \sin k_y + 2t'' \sin 2k_x$$

Excitation from Zhang-Rice band

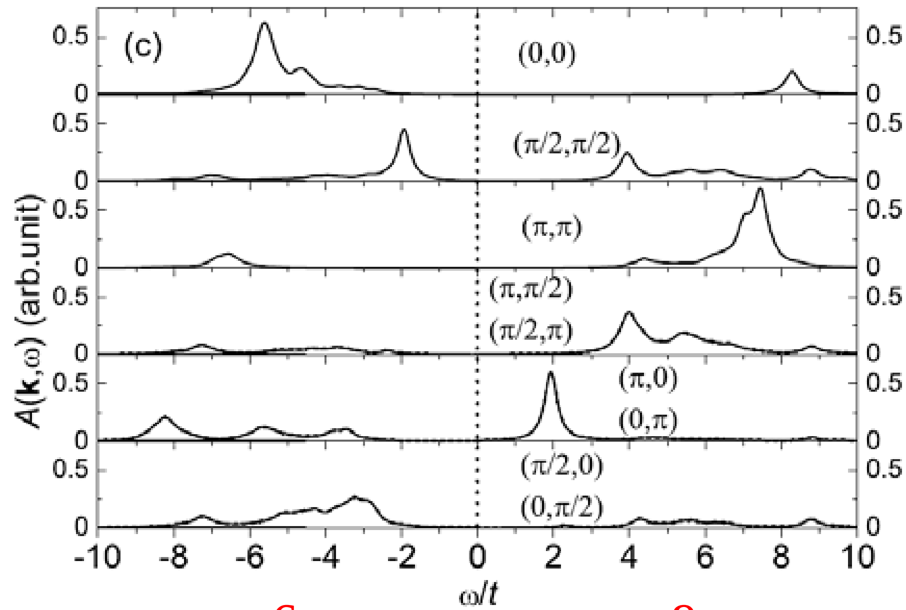
4x4 Hubbard model
$U=10t$
$t'=-0.343t$
$t''=0.229t$

Absorption spectrum



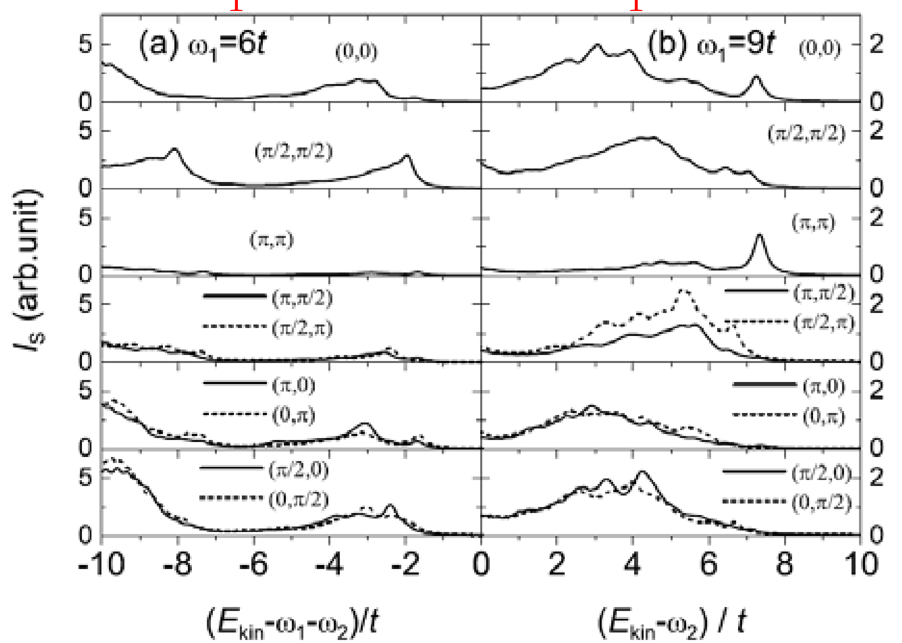


Single-particle excitation $A(\mathbf{k},\omega)$ at half filling



Momentum dependence of $I^{(s)}$ at the two photon energies

$\omega_1=6t$ $\omega_1=9t$



$\omega_1=6t$

"2ω"

Global feature is similar to that of LHB.

$\omega_1=9t$

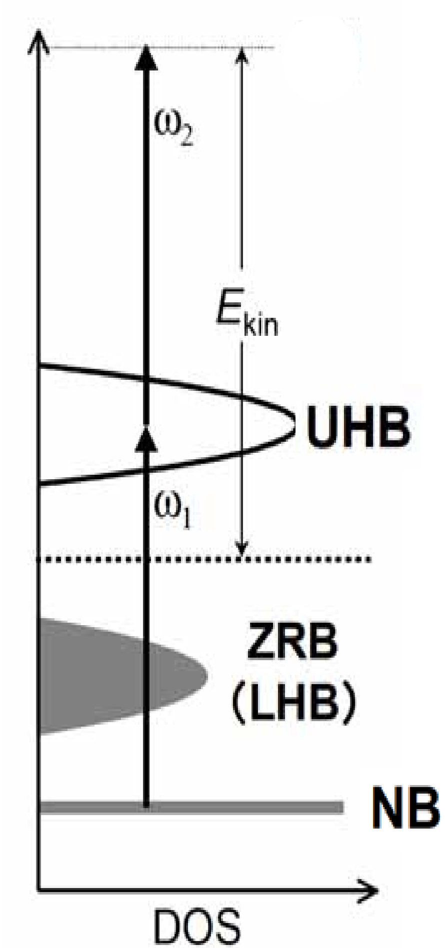
"ω"

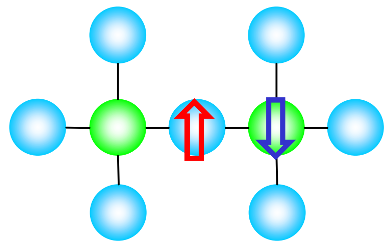
Global feature is similar to that of UHB, but it is difficult to identify the bottom of UHB due to diffusive features.

Possible excitation due to the pump photon ω_1

Excitation from non-bonding band

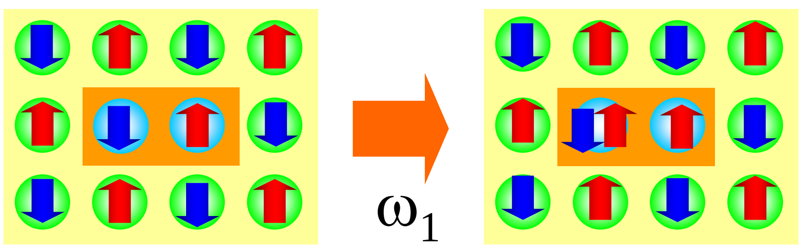
We assume no dispersion of NB and no interactions with other bands.





ω_1 ~ 4-5eV

Single-band picture



Electron-addition operator in the Hubbard model

$$j_x = \sum_{\mathbf{k},\sigma} \beta_{\mathbf{k}} c^{\dagger}_{\mathbf{k},\sigma} b_{\mathbf{k},\sigma}$$

$$\beta_{\mathbf{k}} = C \cos(k_x/2) \sin(k_y/2)/\sqrt{1-(\cos k_x + \cos k_y)/2}$$

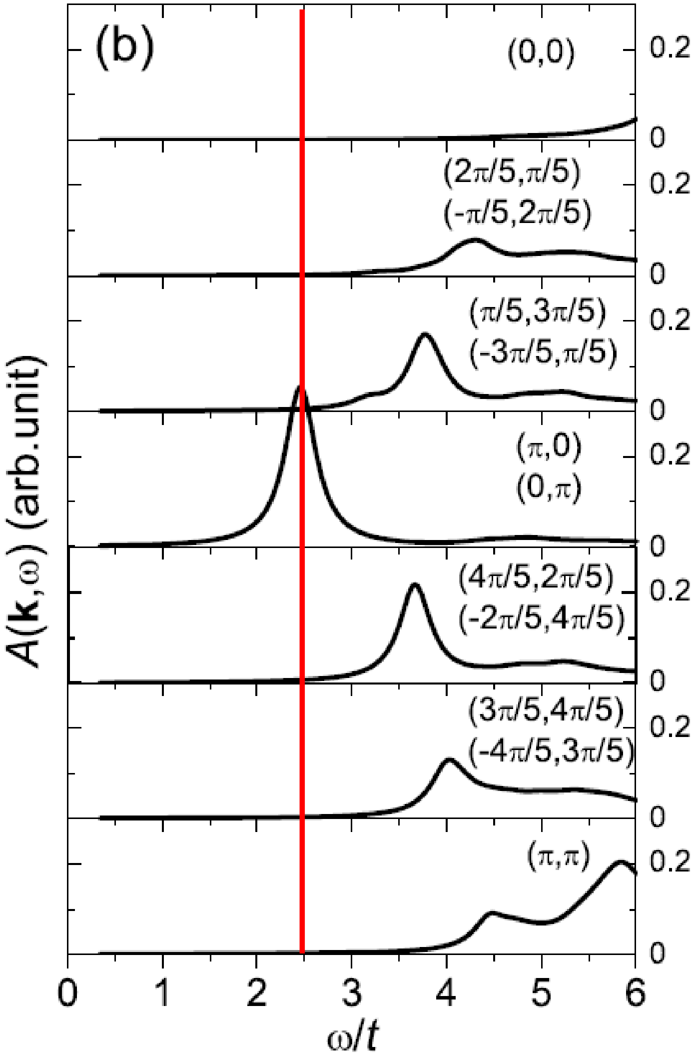
$$C = 2 d_{\mathrm{Cu-O}} T_{pd}$$

Momentum dependence comes from the construction of Wannier orbital, following by Zhang and Rice.

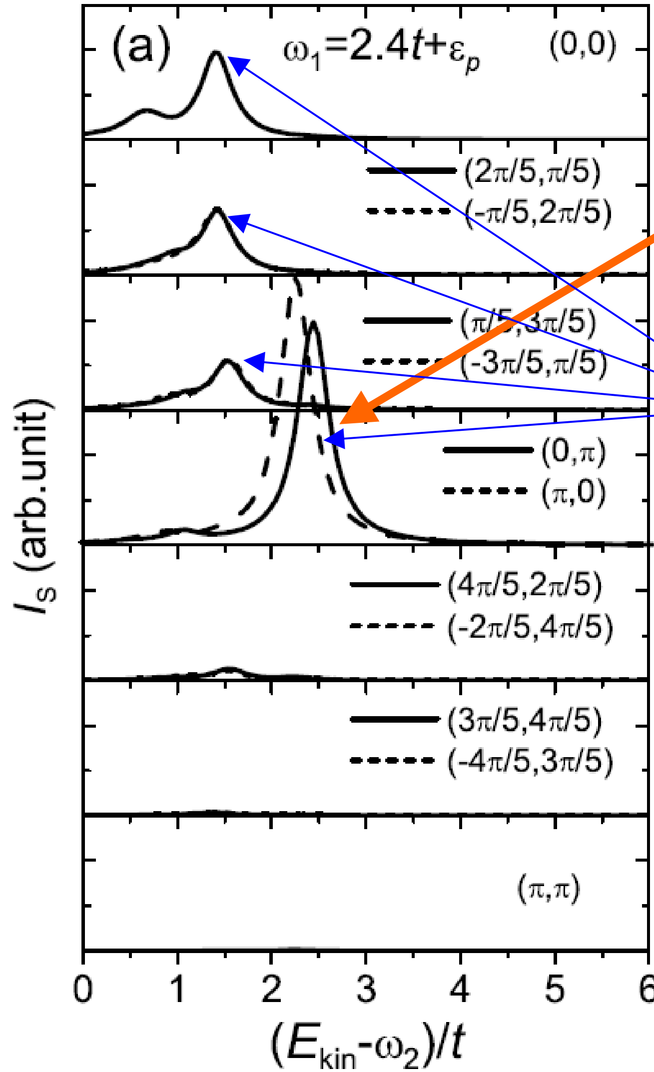
Excitation from non-bonding band

20-site *t-t'-t''-J* model
$U=10t$, $t'=-0.343t$, $t''=0.229t$

$A(\mathbf{k},\omega)$ in UHB
$\omega_1=2.4t+\varepsilon_p$



AR-2PPES: $I^{(s)}$



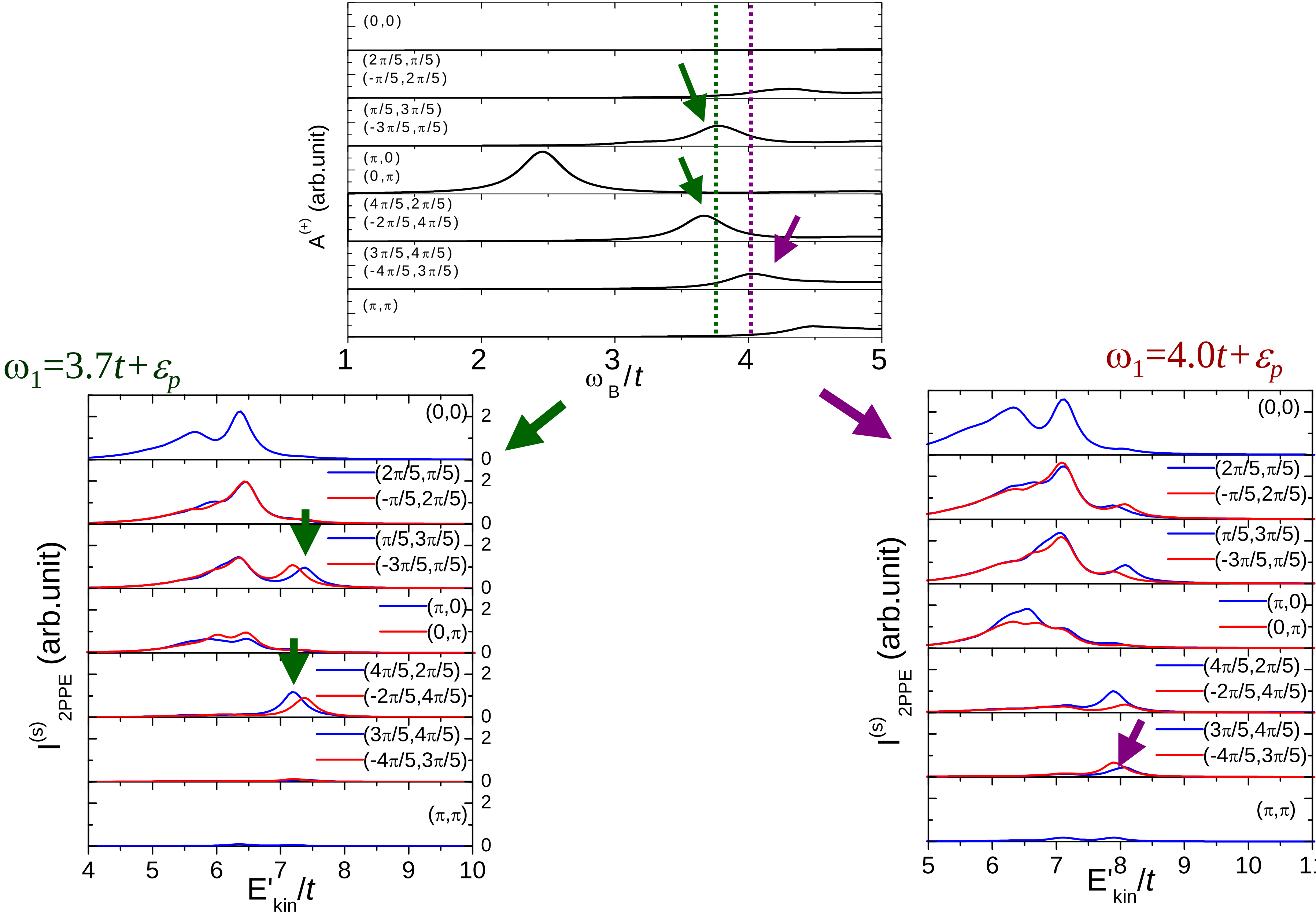
Emission from the (0,π) state.

additional structures

"spin-wave" excitation

Absence in the spectral function

$|I\rangle$: N (half-filling)
$|M\rangle$: N+1
$|F\rangle$: N



With increasing ω_1, the highest-energy position follows UHB dispersion, but accompanied with the spin-related excitation.

Experimental conditions to detect the bottom of UHB at (π,0)

Maximum kinetic energy of photoelectron from the bottom of UHB

$$E_{\mathrm{kin}}^{\mathrm{max}} = E_{\mathrm{gap}}/2+\omega_2 \qquad E_{\mathrm{gap}} = 4.8t \sim 2\mathrm{eV}$$

(i) From Zhang-Rice band → Max. intensity appears at $\omega_1 \sim 9t \sim 3$ eV.

If $\omega_1=\omega_2$, then $E_{\mathrm{kin}}^{\mathrm{max}} \sim 4$ eV .

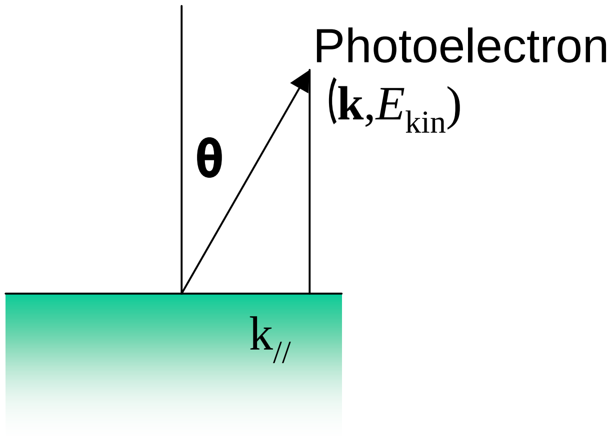
(ii) From non-bonding band → $\omega_1=2.4t+\varepsilon_p \sim 5$ eV.

If $\omega_1=\omega_2$, then $E_{\mathrm{kin}}^{\mathrm{max}} \sim 6$ eV .

Minimum kinetic energy necessary to reach (π,0)

From $\frac{\hbar^2}{2m}k_{//}^{\ 2} = (E_{\mathrm{kin}} - E_{\mathrm{vac}})\sin\theta$, $E_{\mathrm{kin}}^{\mathrm{min}} \sim 6$ eV

using $d_{\text{Cu-Cu}} \sim 0.4$ nm and $E_{\mathrm{vac}} \sim 4$ eV.



The case (ii) critically satisfies this condition.

If the condition $\omega_1 < \omega_2$ is used, it is easy to observe the bottom of UHB.

Summary

We proposed angle-resolved two-photon photoemission spectroscopy (AR-2PPES) as a new technique to detect the location of the bottom of the upper Hubbard band (UHB) in two-dimensional insulating cuprates.

When the pump photon excites an electron from the Zhang-Rice singlet band, the bottom of UHB is less clear, because of diffusive features in the spectra.

When the pump photon excites an electron from the non-bonding band, the bottom of UHB can be clearly identified.

In addition to information of UHB, additional excitations related to spin degree of freedom emerge in the spectrum, which are characteristic of strongly correlated system,

To detect the bottom of UHB at (π,0), it may be necessary to use either the excitation from non-bonding band or two photons with different energies.