4th COE symposium, June 28 (2006), Tohoku Univ.

Elementary excitations and optical responses in strongly correlated electron systems

Hiroaki Matsueda

Condensed matter theory group,

Department of Physics, Tohoku University

Collaborators:
Takami Tohyama (*IMR*)
Sadamichi Maekawa (*IMR*, *CREST*)
Nejat Bulut (*IMR*, *CREST*)
Sumio Ishihara (*Dept. of Phys.*, *Tohoku Univ.*)

Strongly correlated electrons

electron: charge $-e$, spin ½ → Coulomb and exchange interactions

materials

Semiconductor
~ 'doped band insulator'

Transition metal oxides ~ 'doped Mott insulator'

$La_{2-x}Sr_xCuO_4$: high-T_c superconductivity

$La_{1-x}Sr_xMnO_3$: colossal magnetoresistance

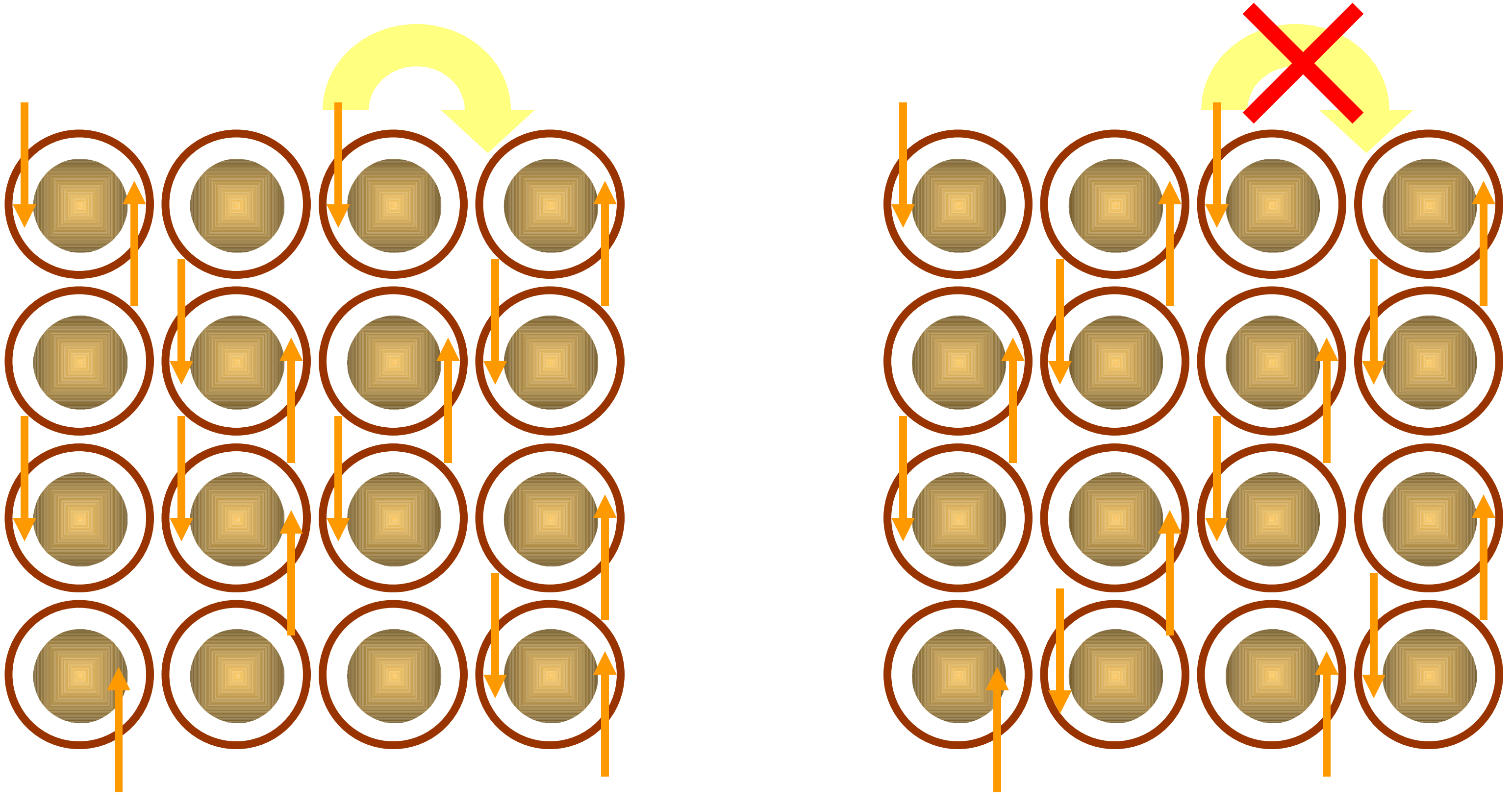
Competition between t (hopping) and U (on-site Coulomb)

Mott insulator

Half-filling: $N_L = N_e$

(a) Band metal ($U<t$)

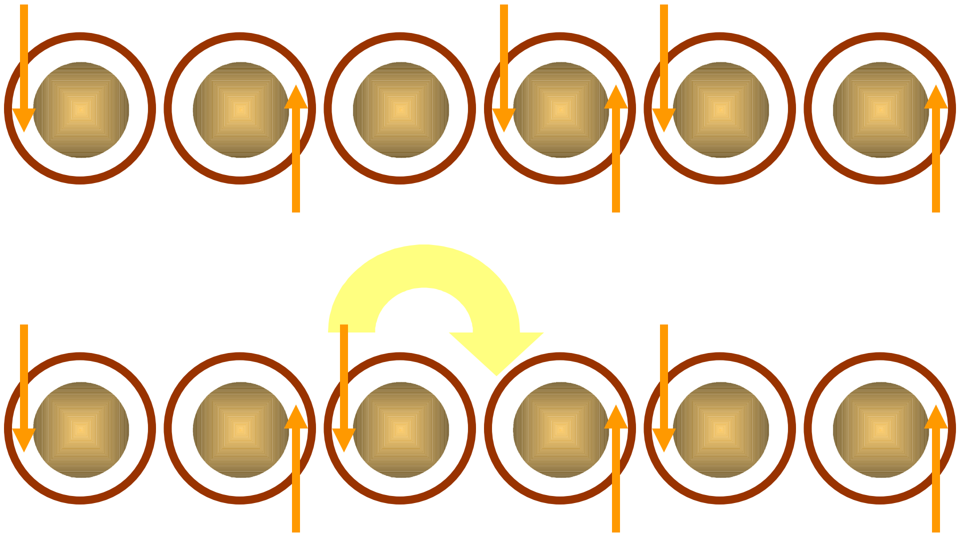
(b) Mott insulator ($U>t$)



Optical gap of Mott insulators

Excited state $E_1 \sim E_0 + U$:

Groud state E_0:



Optical gap $\Delta = E_1 - E_0 \sim U$

$\Leftrightarrow$ (gap of band insulators) ~ (periodic potential of lattice)

‘Gigantic’ third-order nonlinear susceptibility

$$P = \varepsilon_0 \left(\chi^{(1)} E + \chi^{(3)} E^3 + \cdots \right)$$

Electro-modulation spectroscopy:

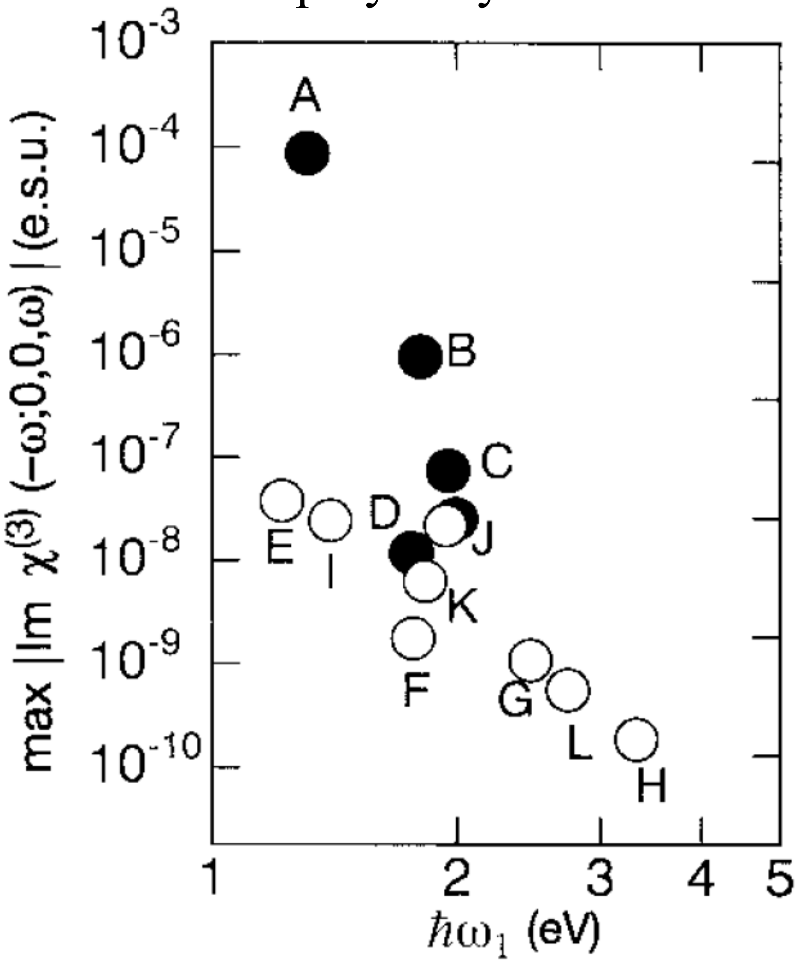
$$\Delta \varepsilon_2(\omega) = 3\,\mathrm{Im}\,\chi^{(3)}(-\omega;0,0,\omega)\,E(0)^2$$
$$\propto \left|\langle 0|x|1\rangle\right|^2 \left|\langle 1|x|2\rangle\right|^2$$

$|2\rangle$ even

$|1\rangle$ odd

$|0\rangle$ even

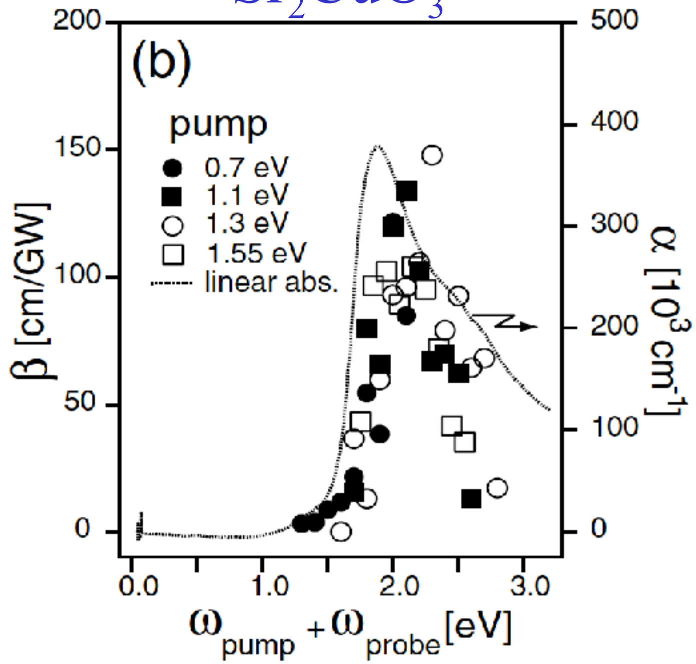
A, B, C : Ni-compounds
D : Sr_2CuO_3
I : polyacetylene



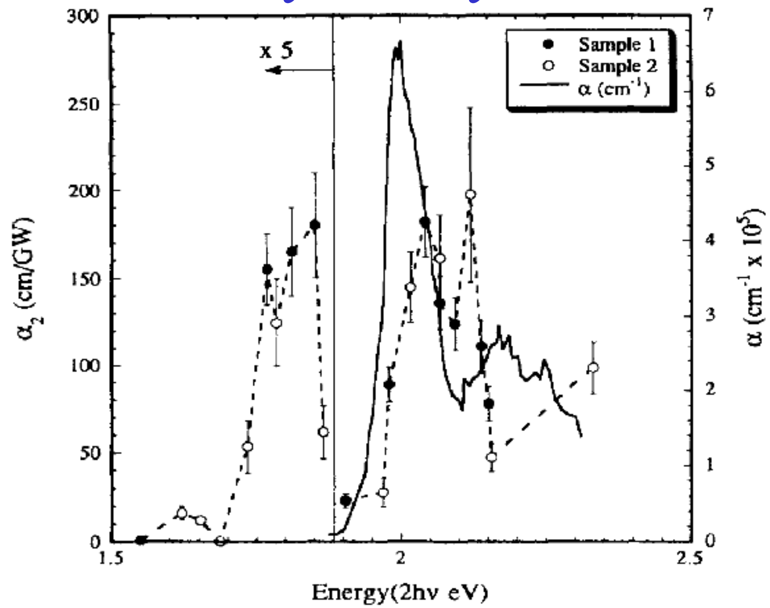
H.Kishida et al., Nature 405, 929 (2000)

One- and two-photon absorption spectra

Sr_2CuO_3



Polydiacetylene



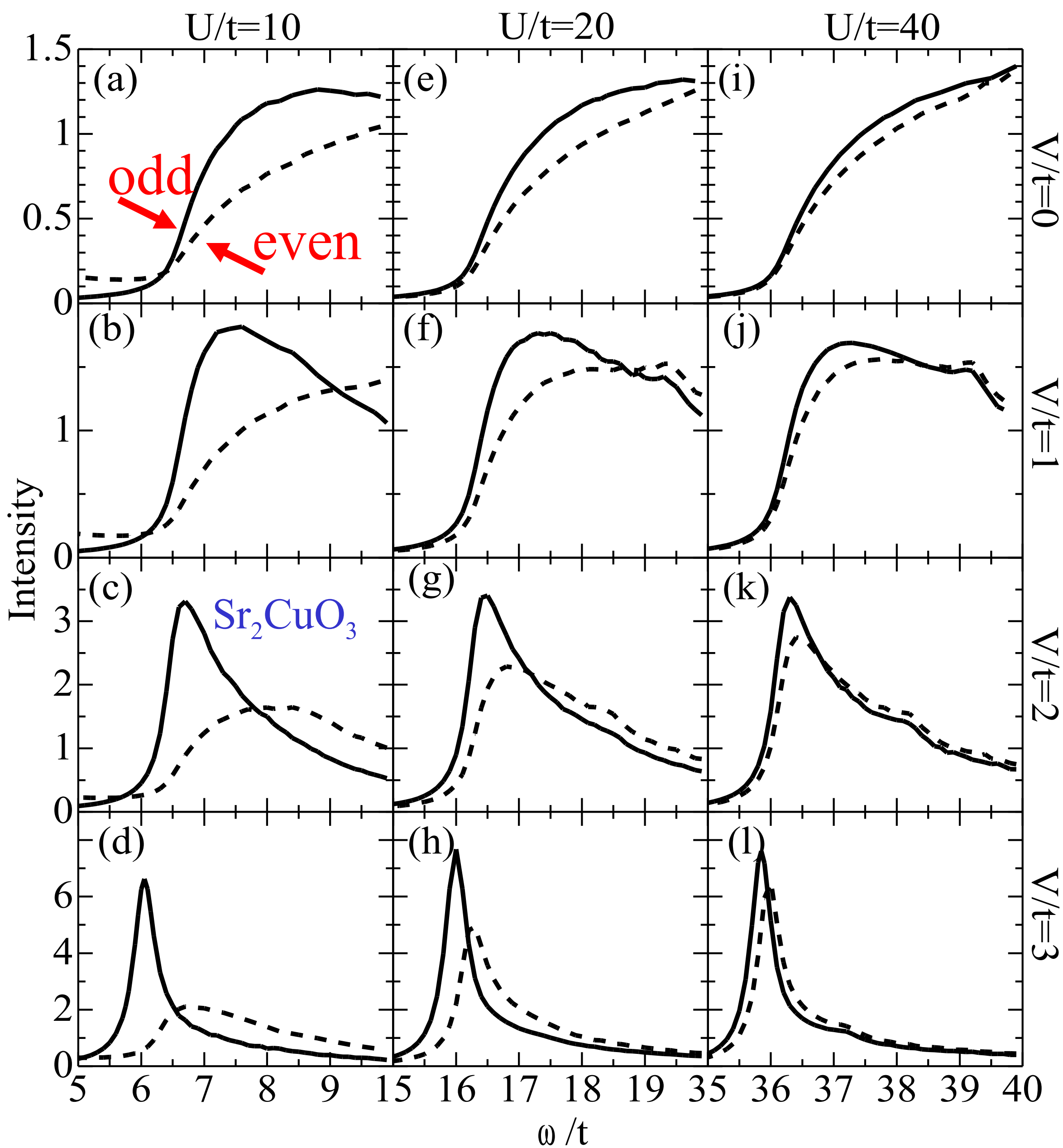
T. Ogasawara, et al., PRL85, 2204 (2000)

B. Lawrence, et al., PRL73, 597 (1994)

Purpose

Gigantic $\chi^{(3)} \Leftrightarrow$ degeneracy of photoexcited states

$$H = -t\sum_{i,\sigma}(c^{+}_{i,\sigma}c_{i+1,\sigma} + H.c.) + U\sum_{i} n_{i,\uparrow}n_{i,\downarrow} + V\sum_{i} n_i n_{i+1}$$

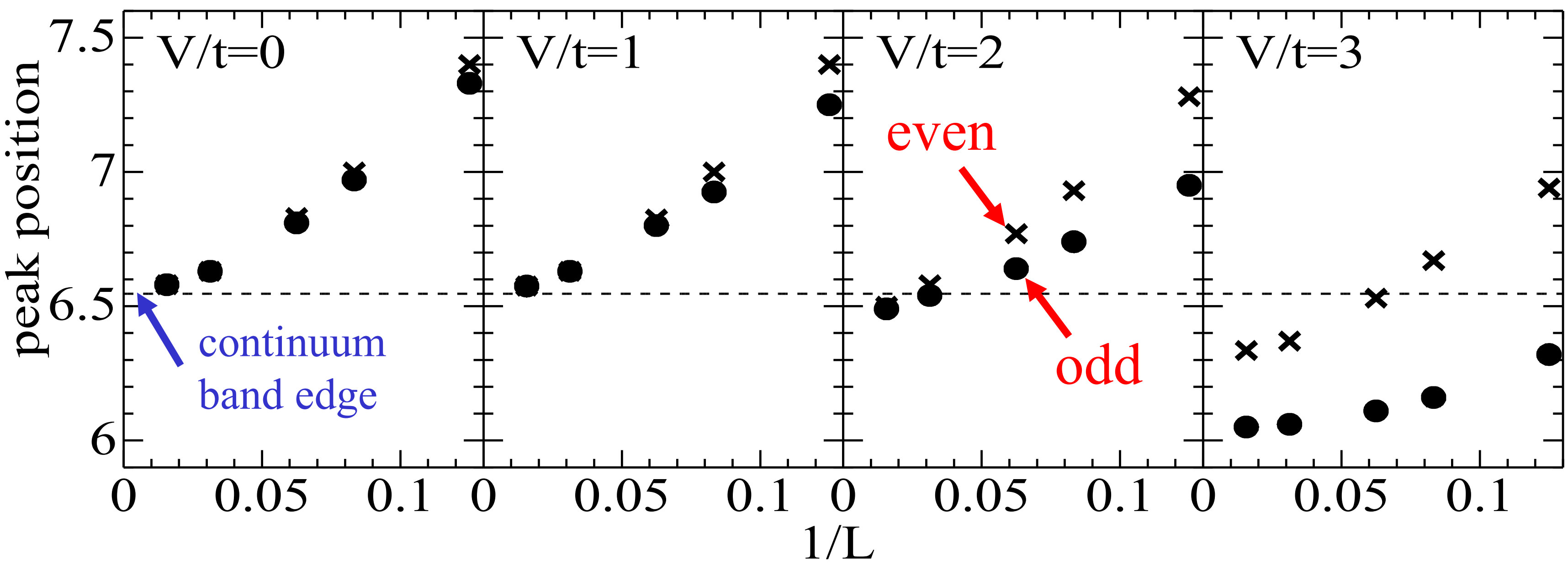


$V_c/t \sim 2$

H. Matsueda, et al., PRB70, 033102 (2004)

Scaling for the lowest energy states

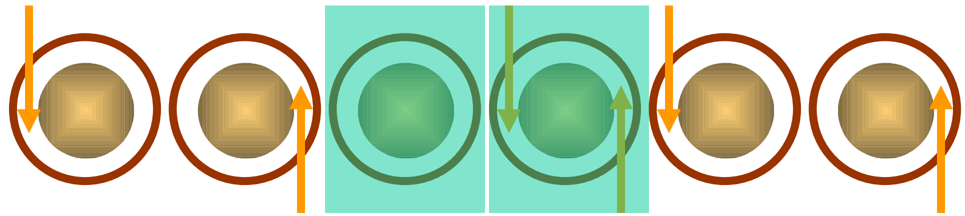
U/t=10



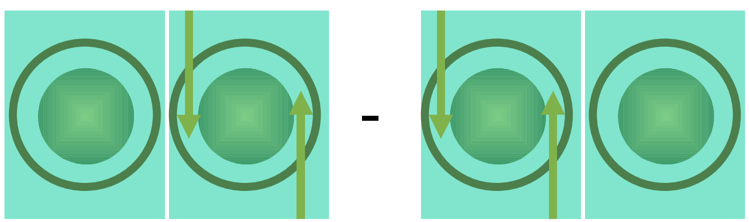
H. Matsueda, et al., PRB70, 033102 (2004)

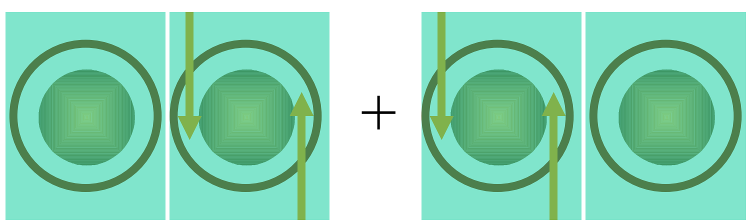
Interpretation: 'spin-charge separation' and excitations

1D → spin-charge separation: $\psi=\psi_{spin}\times\psi_{charge}$

Excited state $E_1 \sim E_0+U$: 

'holon' 'doublon'

odd parity: 

even parity: 

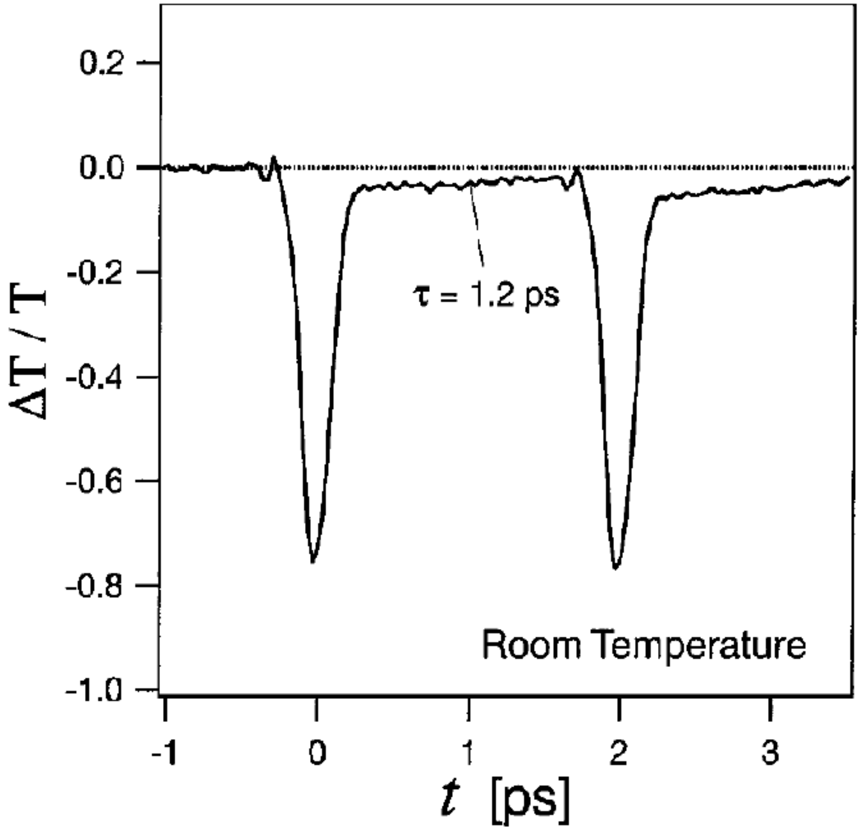
$U \rightarrow \infty$:

Prohibition of exchange

↓

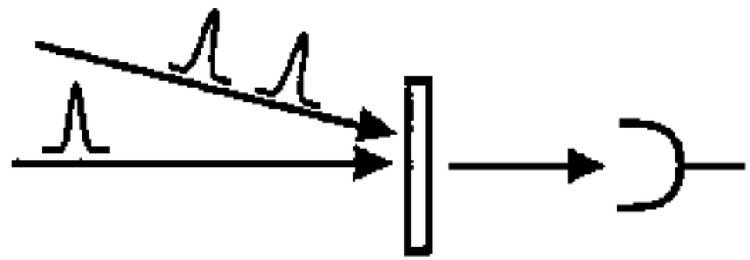
degeneracy

Ultra-fast relaxation



Optical gap ～ 2 eV

Pump photon: 0.88 eV



Probe photon: 1.03 eV

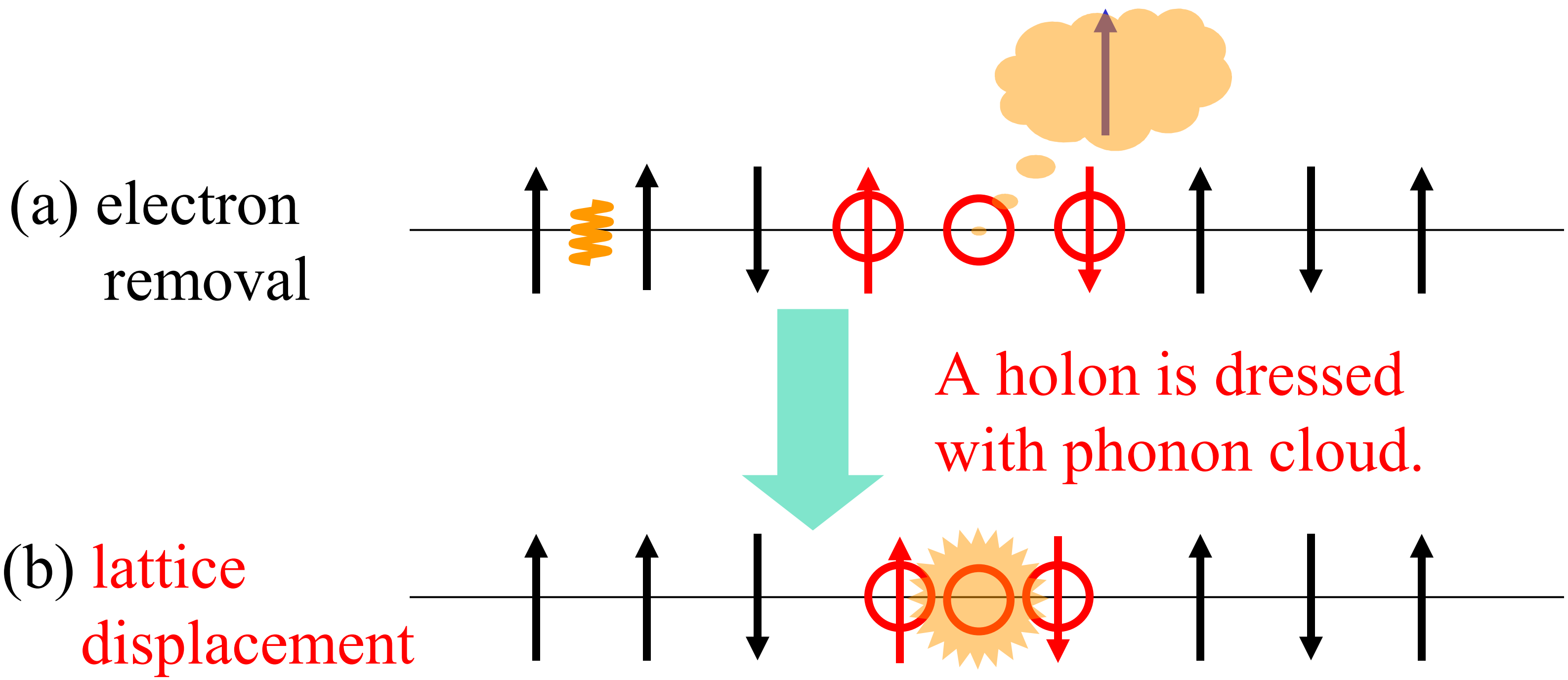
Possible phenomena of all-optical switching devices

T. Ogasawara, et al., PRL85, 2204 (2000)

Electron-phonon coupling in spin-charge separated systems

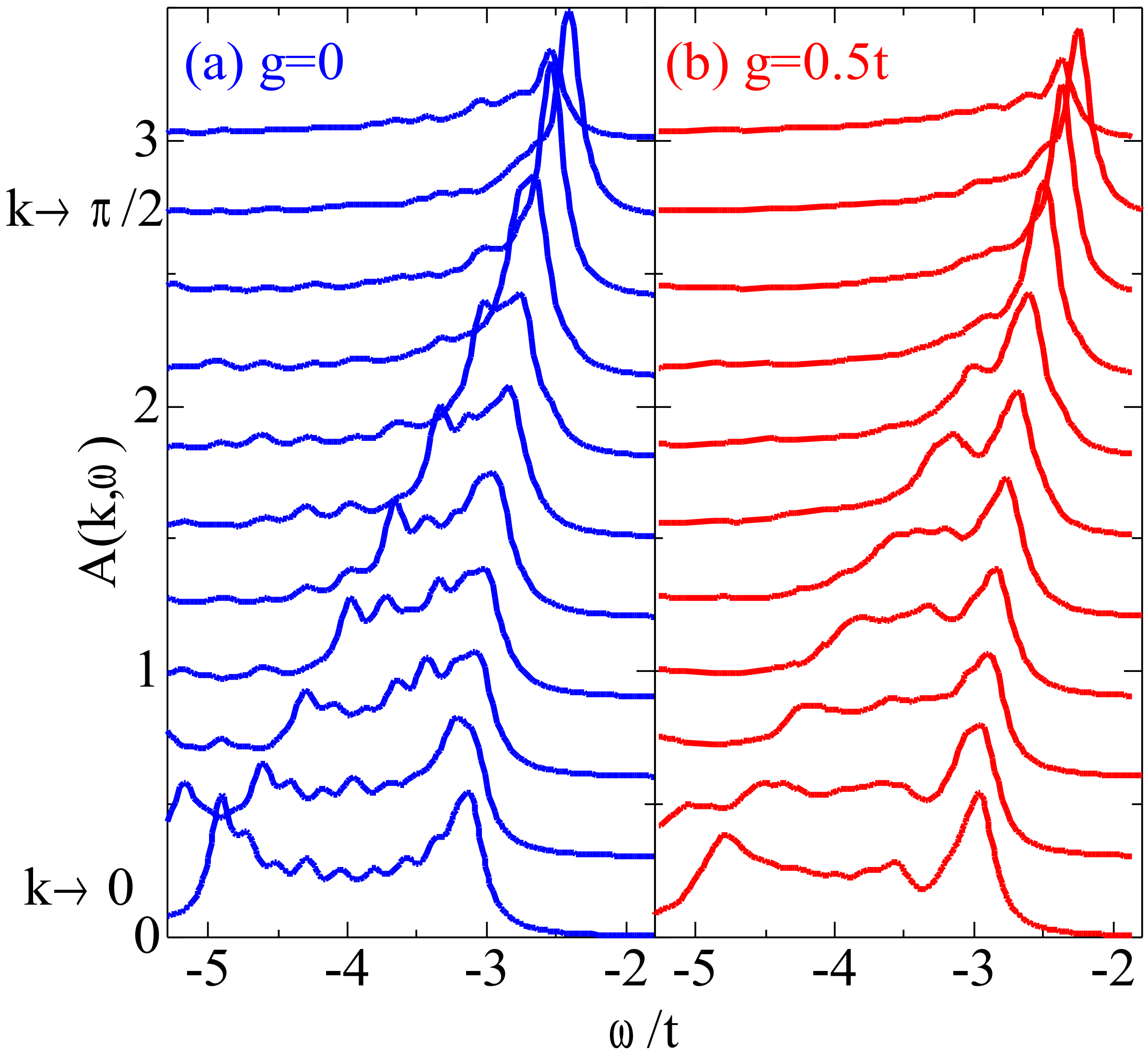
The ground state (uniform distribution of charge) → no phonons

How about the excited states?



$$H = -t\sum_{i,\sigma}(c_{i,\sigma}^{+}c_{i+1,\sigma} + H.c.) + U\sum_{i} n_{i,\uparrow}n_{i,\downarrow} + \omega_0\sum_{i} b_i^{+}b_i - g\sum_{i}(b_i^{+} + b_i)n_i$$

Phonon effects on one-electron spectral function



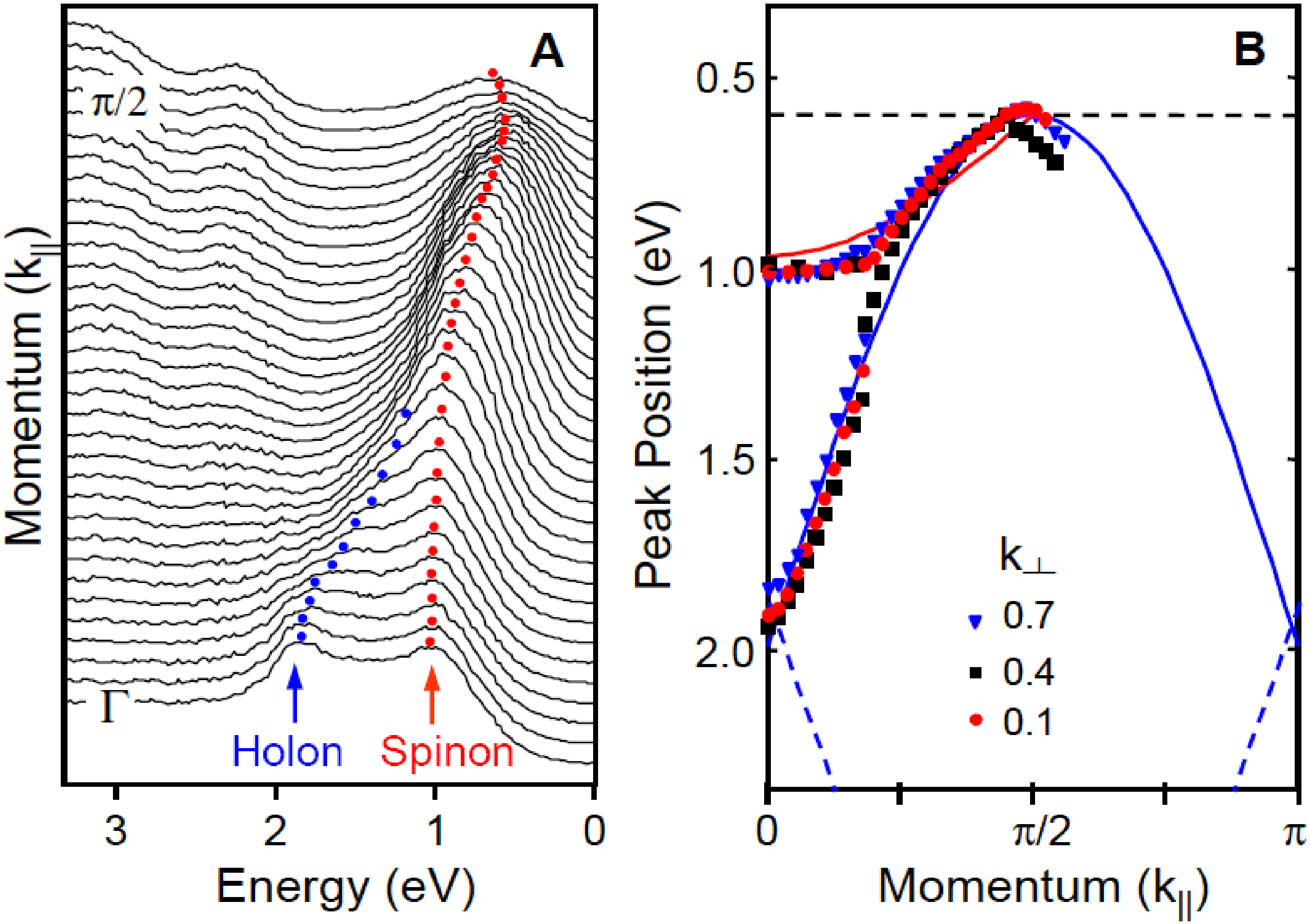
U=8t
ω_0=0.5t
N_L=N_e=20

H. Matsueda, et al.

broad holon branch: spinon and holon behave differently.

'Spinon' and 'Holon' dispersions in photoemission spectrum

Sample: $SrCuO_2$, photon beam: 85 eV, T=300 K

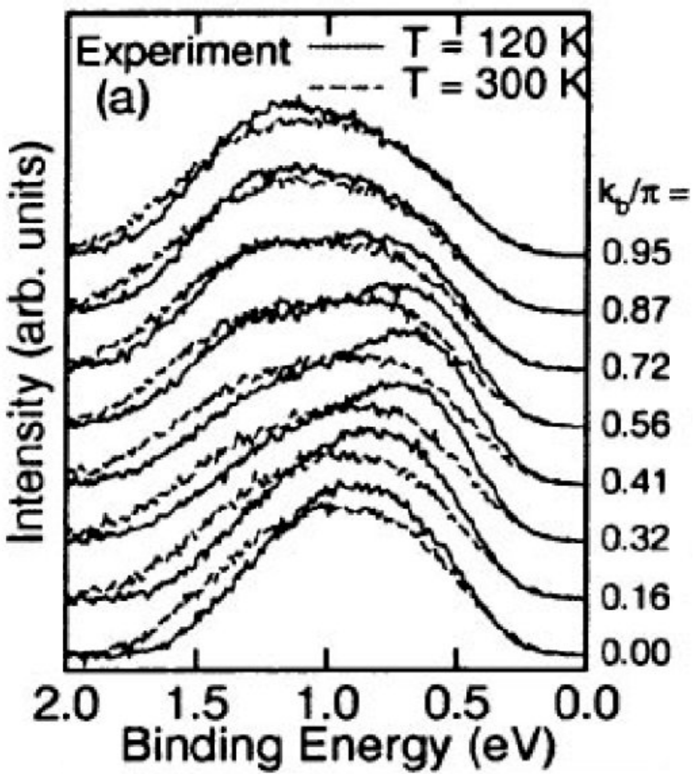


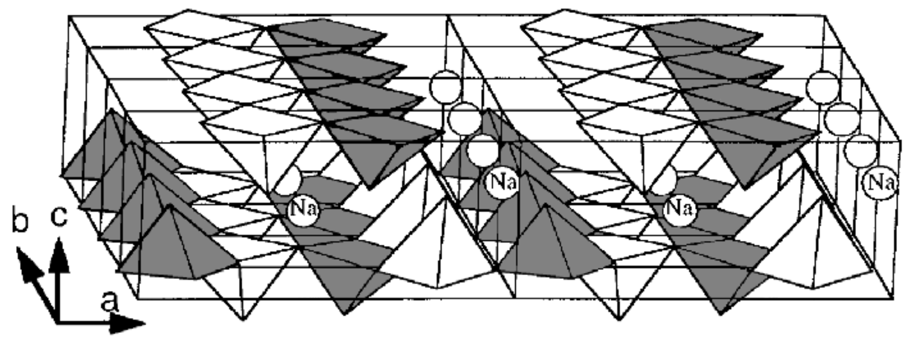
t=0.65 eV, J=4t^2/U=0.27 eV

B. J. Kim, et al., Nature Physics 2, 397 (2006)

T-dependent ARPES measurement on $Na_{0.96}V_2O_5$

Weight shift $\gg \Delta T$





α′-NaV_2O_5

Mixed valence:

V^{4+} (d^1, S=1/2) / V^{5+} (d^0)

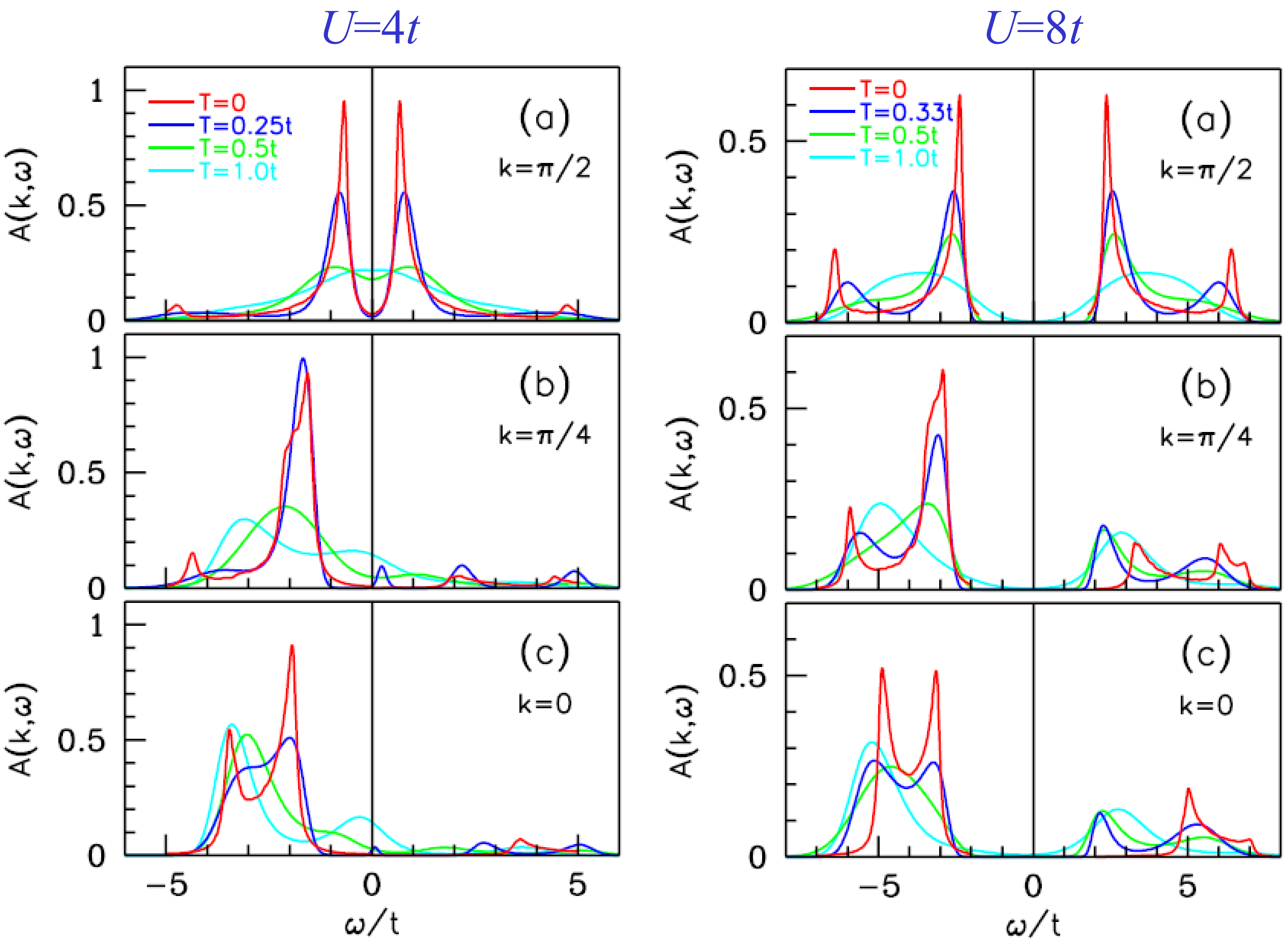
K. Kobayashi et. al., PRL82, 803 (1999)

purpose

Temperature dependence of spinon and holon branches

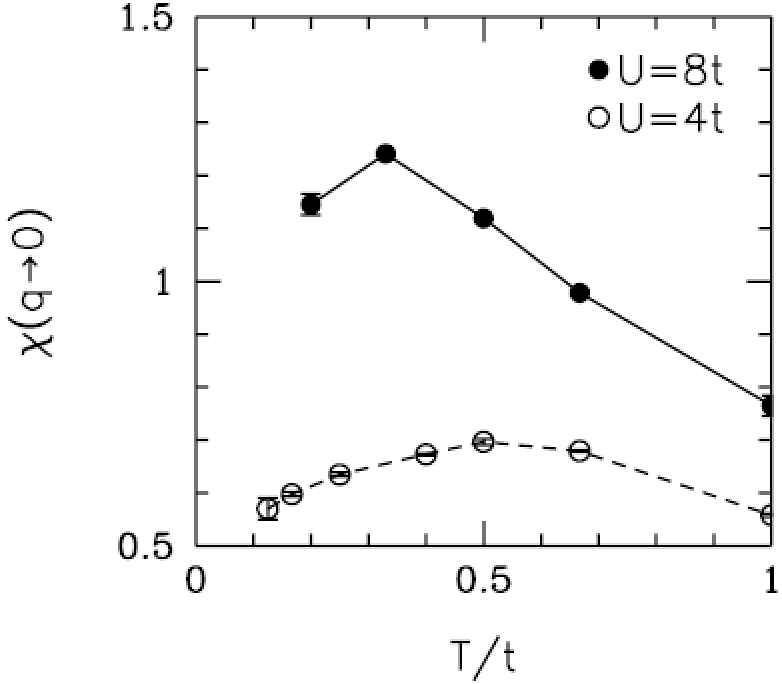
T-dependence of A(k,ω)

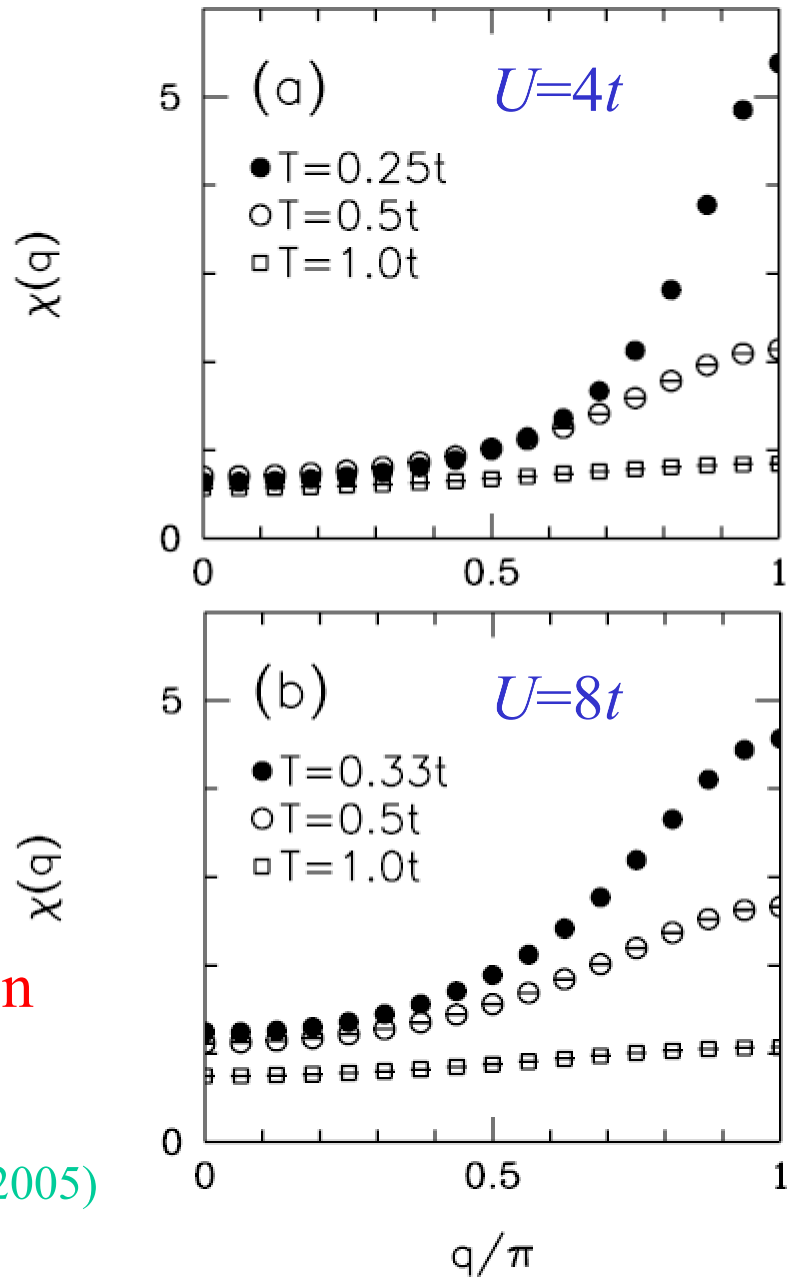
H.Matsueda, et al., PRB72, 075136 (2005)



Static magnetic susceptibility $\chi(q,\omega\rightarrow 0)$

Uniform ($q\rightarrow 0$) component





Enhancement of the AF correlation
→ The spinon branch develops.

H.Matsueda, et al., PRB72, 075136 (2005)

Summary

(1) Novel optical properties in 1D strongly correlated systems

Spin-charge separation → 'holon' and 'doublon'
Degeneracy of even- and odd-parity states ~ gigantic $\chi^{(3)}$

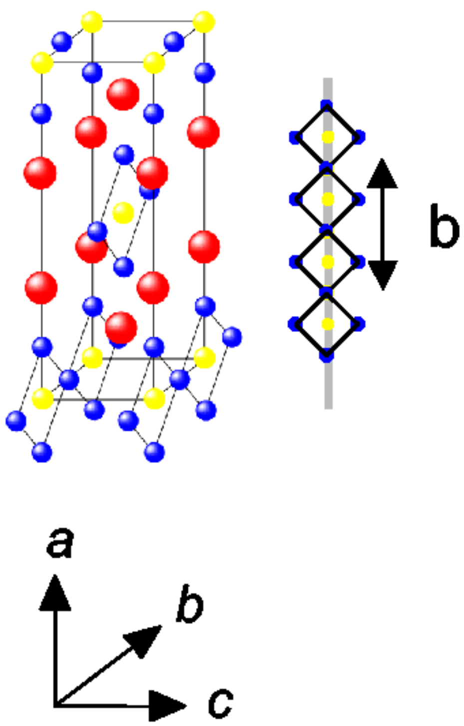
(2) one-electron excitation spectrum

Spin-charge separation → 'spinon' and 'holon'
Electron-phonon interaction ~ holon part is affected.
Finite temperature ~ spinon part is affected.

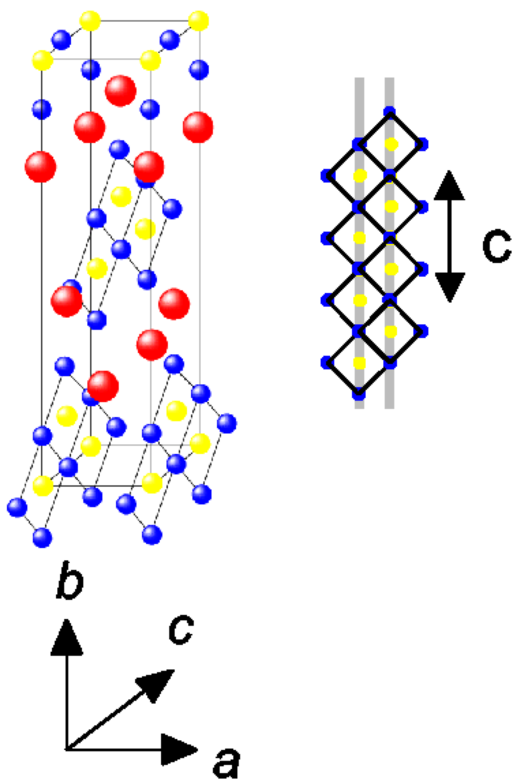
* ultra-fast relaxation, photoinduced phenomena

1D Mott insulators

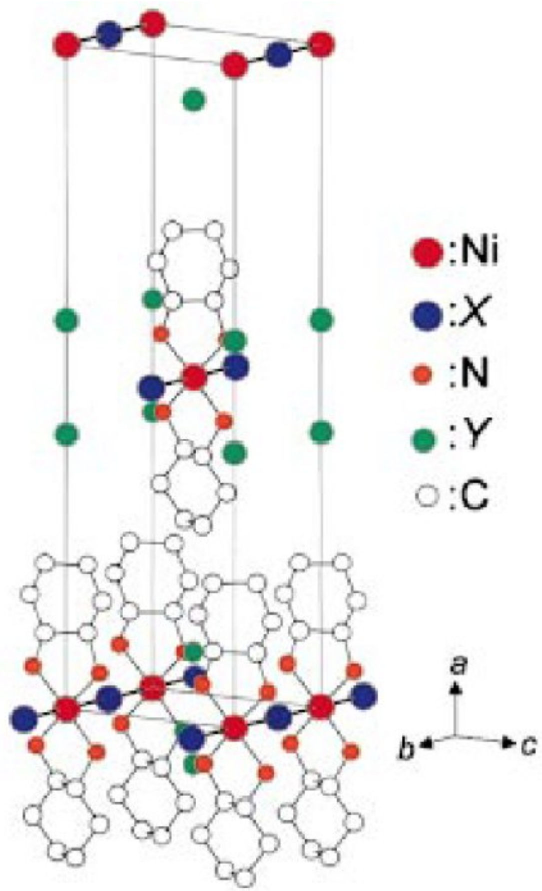
Sr_2CuO_3

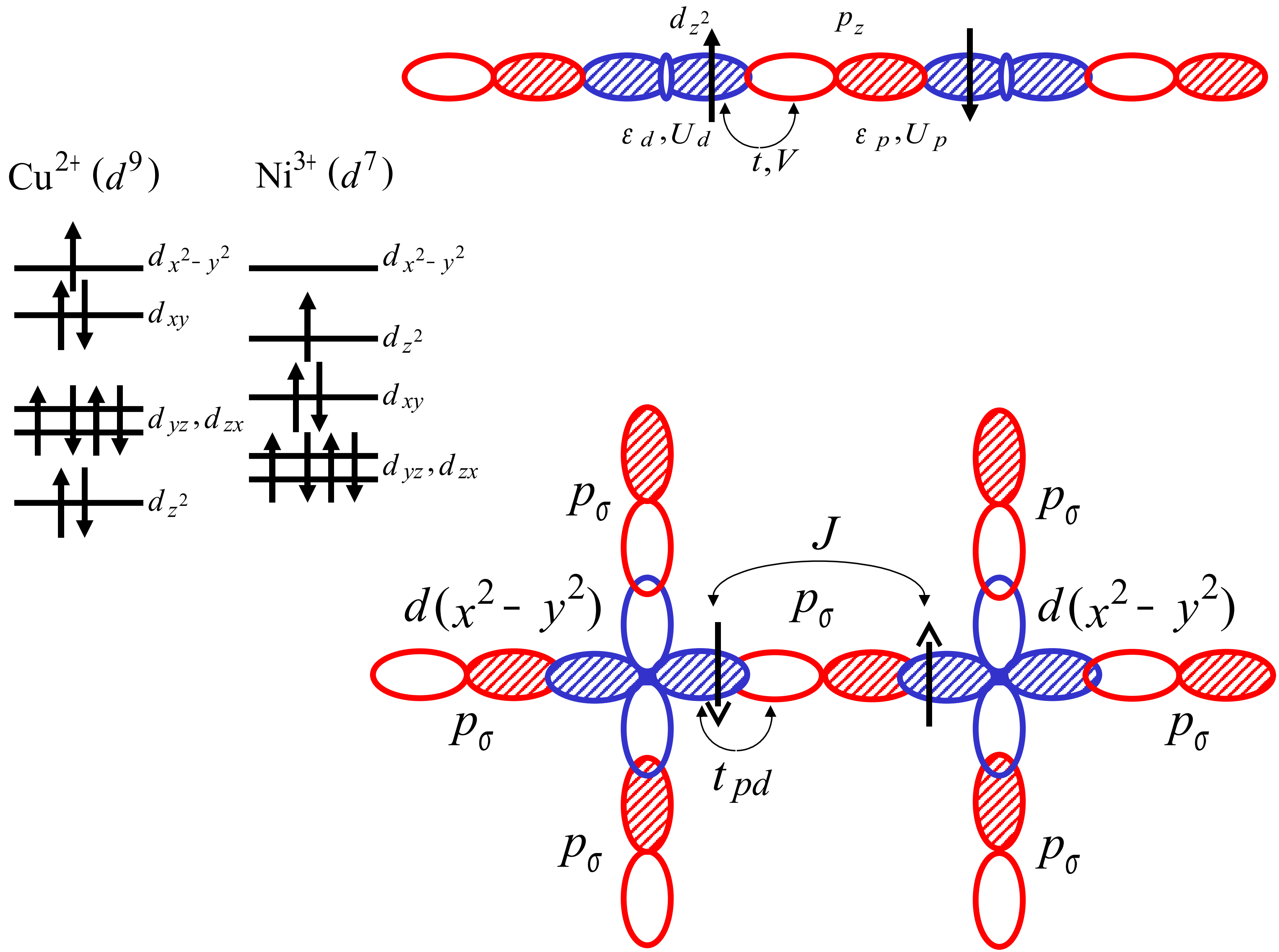


$SrCuO_2$



Halogen-bridged Ni-compound





d_{z^2}
p_z
ε_d, U_d
t, V
ε_p, U_p
Cu^{2+} (d^9)
Ni^{3+} (d^7)
$d_{x^2-y^2}$
d_{xy}
d_{yz}, d_{zx}
d_{z^2}
$d_{x^2-y^2}$
d_{z^2}
d_{xy}
d_{yz}, d_{zx}
p_σ
p_σ
J
$d(x^2-y^2)$
p_σ
$d(x^2-y^2)$
p_σ
t_{pd}
p_σ
p_σ
p_σ