Quantum Criticality and Orbital-dependent Renormalization of Quasiparticles in $Ca_{2-x}Sr_xRuO_4$

–Importance of spatial correlation near a magnetic QCP–

Naoya Arakawa

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Acknowledgements: Y. Yanase, T. Kariyado,
H. Kontani, S. Onari, Y. Yamakawa, ISSP Super Comp.

Contents

- Introduction
  - Electronic structure for Ru oxides; e.g., Sr_2RuO_4
  - Importance of Ru t_{2g} orbitals and Octahedral distortions
  - Heavy fermion behavior in $Ca_{2-x}Sr_xRuO_4$ around $x = 0.5$
- Method
  - Effective model in the presence of the octahedral rotation
  - Fluctuation-exchange (FLEX) approximation
- Results: Mag. prop.s and Renormalization of QPs
  - Roles of the octahedral rotation
  - Roles of the van Hove singularity (vHs) for the d_{xy} orbital
- Summary and Message

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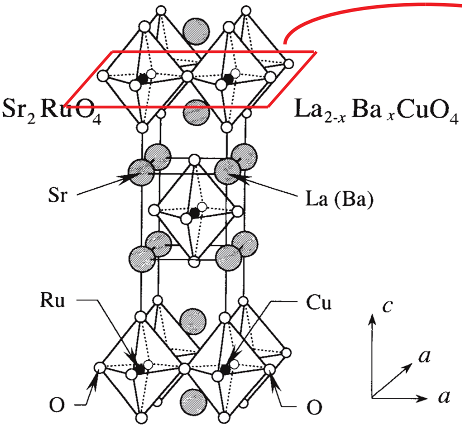
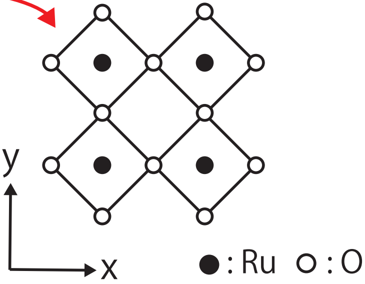
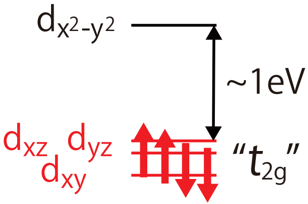
Electronic structure for Ru oxides; e.g., Sr_2RuO_4



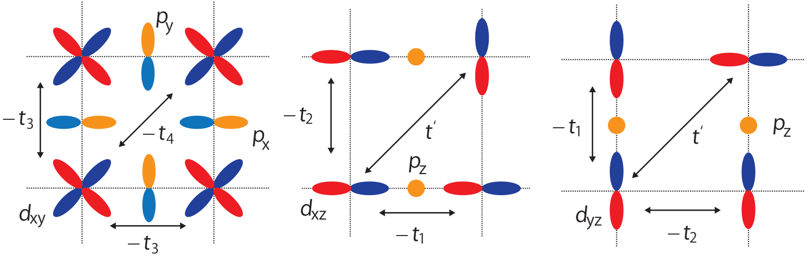




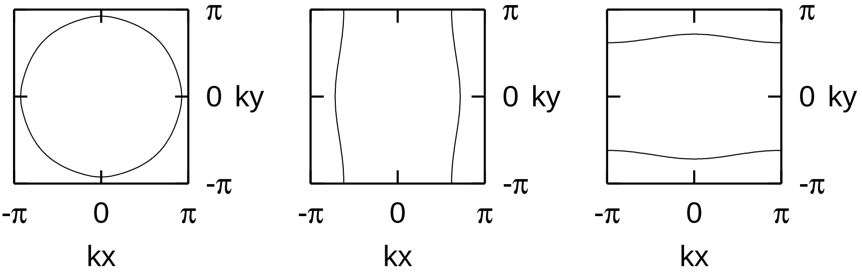
$\rho_{xy}/\rho_z \sim 10^{-3}$ "Quasi 2D"

Y. Maeno *et al.*, Nature(London), **372**, 532 (1994).

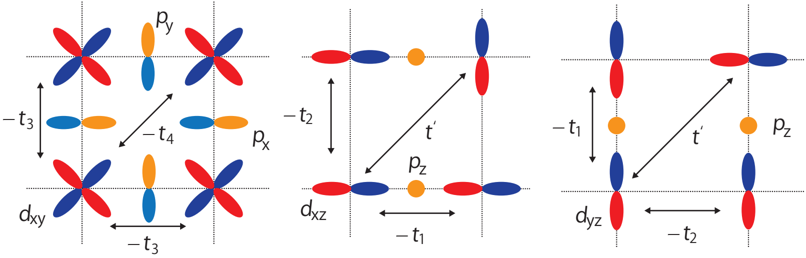
- Cond. bands: Antibonding orbitals between Ru t_{2g} and O $2p$



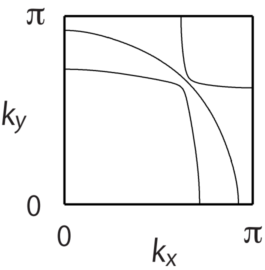
- quasi 2D γ-FS and quasi 1D α/β-FS:

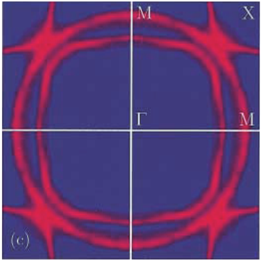


- Cond. bands: Antibonding orbitals between Ru t_{2g} and O $2p$



- quasi 2dim. γ-FS and quasi 1dim. α/β-FS:

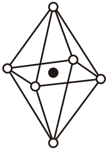
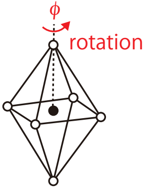
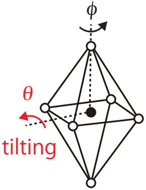
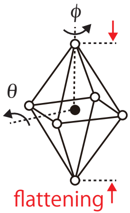




Right fig. (ARPES): A. Damascelli *et al.*, PRL **85**, 5194 (2000).

Importance of Ru t_{2g} orbitals and Octahedral distortions

e.g. $Ca_{2-x}Sr_xRuO_4$

	x = 2	1.5 > x ≧ 0.5	0.5 > x ≧ 0.2	0.2 > x ≧ 0
RuO_6				
Ground states	Spin triplet SC	Paramag. metal x~0.5: Heavy fermion	Paramag. metal with Metamag. trans.	AF insulator

e.g., see S. Nakatsuji *et al.*, PRL **90**, 137202 (2003); O. Friedt *et al.*, PRB **63**, 174432 (2001)

Cf. Wrong proposal by μSR: $m_{\mathrm{AF}} \sim 0.25\mu_{\mathrm{B}}$ at $x = 1.5$

$\rightarrow$ This should be understood as short-range order.

$\because$ Elastic neutron: $m_{\mathrm{AF}} = 0$ at $x = 1.5$

Heavy fermion behavior in $Ca_{2-x}Sr_xRuO_4$ around $x = 0.5$

Q. WHAT is more important in enhancing m^* than the location of the vHs?

e.g. $Ca_{2\text{-}x}Sr_xRuO_4$

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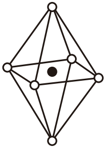
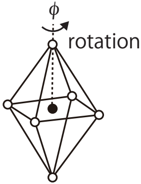
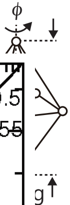
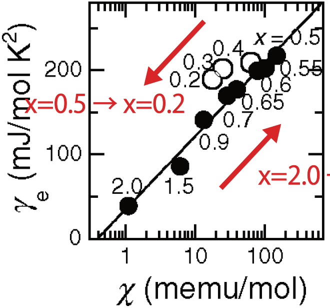
RuO_6				
vHs	60meV	−20meV @x=0.5		
γ_e	37.5 mJ/mol K^2	255 mJ/mol K^2 @x=0.5		

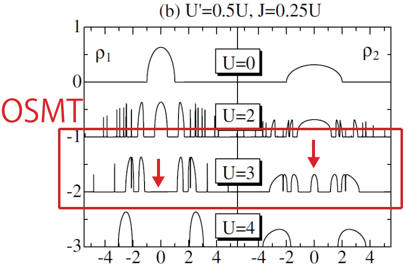


e.g., see S. Nakatsuji *et al.*, PRL **90**, 137202 (2003); O. Friedt *et al.*, PRB **63**, 174432 (2001)

Cf. Case of La-doped Sr_2RuO_4: $\gamma_e \approx 50\text{mJ/mol K}^2$

N. Kikugawa *et al.*, PRB **70**, 060508(R) (2004)

- Proposal of Orbital-selective MT for $d_{xz/yz}$ around $x = 0.5$



V. I. Anisimov *et al.*, Eur. Phys. J. B (2002); A. Koga *et al.*, PRL (2004)

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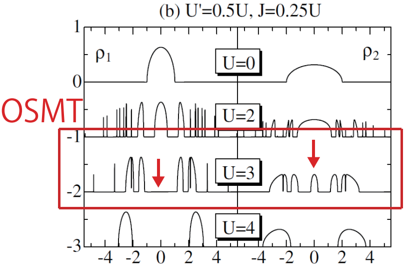
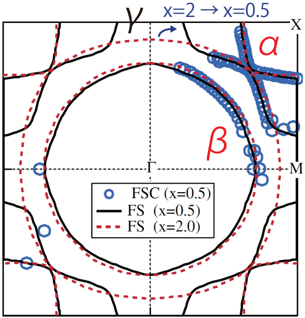
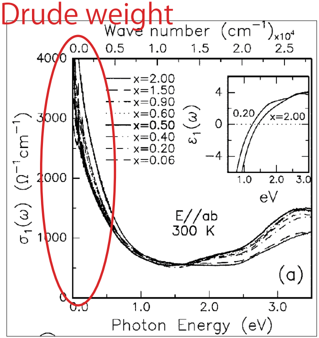


V. I. Anisimov *et al.*, Eur. Phys. J. B (2002); A. Koga *et al.*, PRL (2004)

- Its inconsistency with ARPES and Optical measurements

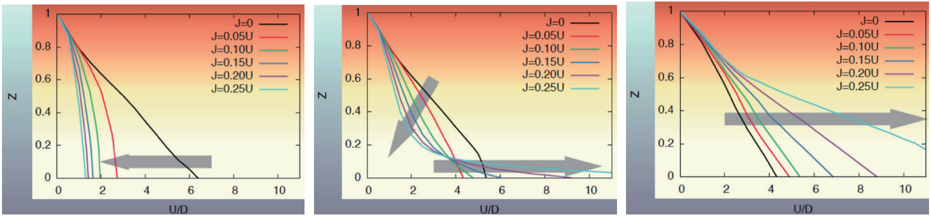




S.-C. Wang *et al.*, PRL (2004); J. S. Lee *et al.*, PRL (2003)

- "Hund's metal": Metal, extended by $J \nearrow$, with small Z

i) N=3 in 3 orbitals/site ii) N=2 in 3 orbitals/site iii) N=1 in 3 orbitals/site



L. de' Medici *et al.*, PRL (2011)

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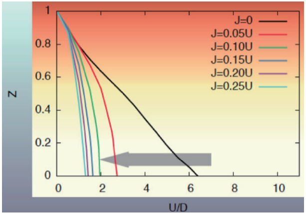
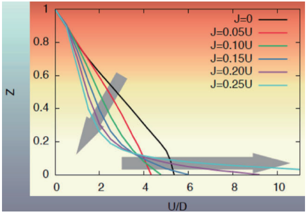


ii) N=2 in 3 orbitals/site



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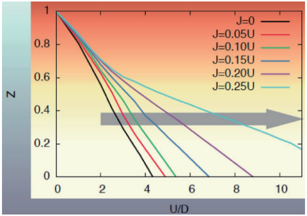

L. de' Medici *et al.*, PRL (2011)

- Proposal of "Hund's metal" in Sr_2RuO_4; $U = 2.3$eV

J [eV]	$m^*/m_{\mathrm{LDA}}\vert_{xy}$	$m^*/m_{\mathrm{LDA}}\vert_{xz}$	T^*_{xy} [K]	T^*_{xz} [K]
0.0, 0.1	1.7	1.7	>1000	>1000
0.2	2.3	2.0	300	800
0.3	3.2	2.4	100	300
0.4	4.5	3.3	60	150

J. Mravlje *et al.*, PRL (2011)

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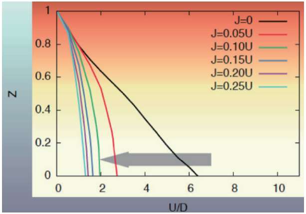

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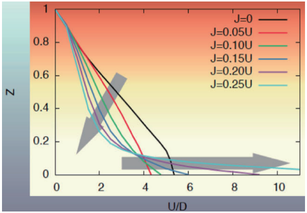

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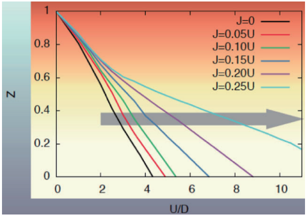

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J. Mravlje *et al.*, PRL (2011)

- Its inconsistency with experimental results
  - Exp. for $Ca_{2-x}Sr_xRuO_4 (x < 2)$: $(m^*_{xy}/m_{xy}) \gg (m^*_{xz/yz}/m_{xz/yz})$

    e.g., J. S. Lee *et al.*, PRL (2003)
  - Gutzwiller analysis for $x = 0.5$: $(m^*_{xy}/m_{xy}) < (m^*_{xz/yz}/m_{xz/yz})$

Gutzwiller analysis: N. Arakawa and M. Ogata, PRB **86**, 125126 (2012)

Heavy fermion behavior in $Ca_{2-x}Sr_xRuO_4$ around $x = 0.5$

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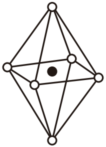
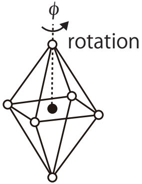
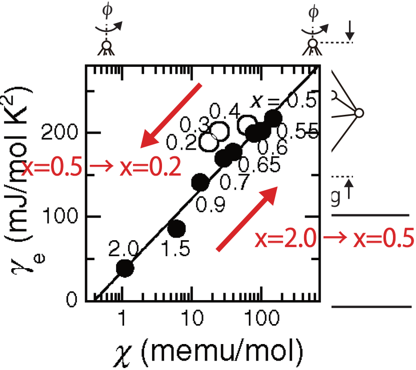
RuO_6				
vHs	60meV	−20meV @x=0.5		
γ_e	37.5 $mJ/mol\,K^2$	255 $mJ/mol\,K^2$ @x=0.5		



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Cf. Case of La-doped Sr_2RuO_4: $\gamma_e \approx 50 mJ/mol\ K^2$

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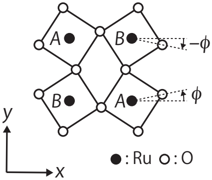
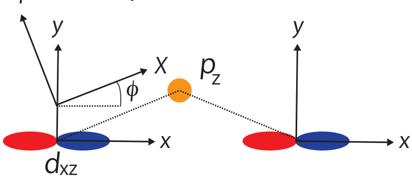
Effective model in the presence of the octahedral rotation

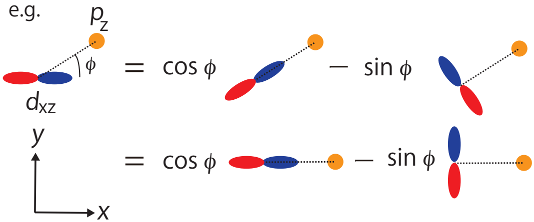
$$\hat{H} = \sum_{\boldsymbol{k}}^{\prime} \sum_{a,b=1}^{3} \sum_{l,l'=A,B} \epsilon_{ab}^{ll'}(\boldsymbol{k},\phi)\hat{c}^{\dagger}_{\boldsymbol{k}al}\hat{c}_{\boldsymbol{k}bl'} + U\sum_{i,a}\hat{n}_{ia\uparrow}\hat{n}_{ia\downarrow} + U'\sum_{i}\sum_{a>b}\hat{n}_{ia}\hat{n}_{ib}$$

$$- J_{\mathrm{H}}\sum_{i}\sum_{a>b}\left(2\hat{s}_{ia}\cdot\hat{s}_{ib} + \frac{1}{2}\hat{n}_{ia}\hat{n}_{ib}\right) + J'\sum_{i}\sum_{a>b}\hat{c}^{\dagger}_{ia\uparrow}\hat{c}^{\dagger}_{ia\downarrow}\hat{c}_{ib\downarrow}\hat{c}_{ib\uparrow}$$

- Change of $V_{ab}(\phi)$; $t_{aa'}^{AB}(\phi) = \sum_{b=p_x,p_y,p_z} \dfrac{V_{ab}(\phi)V_{ba'}(-\phi)}{E_a(0^\circ) - E_b(\phi)}$

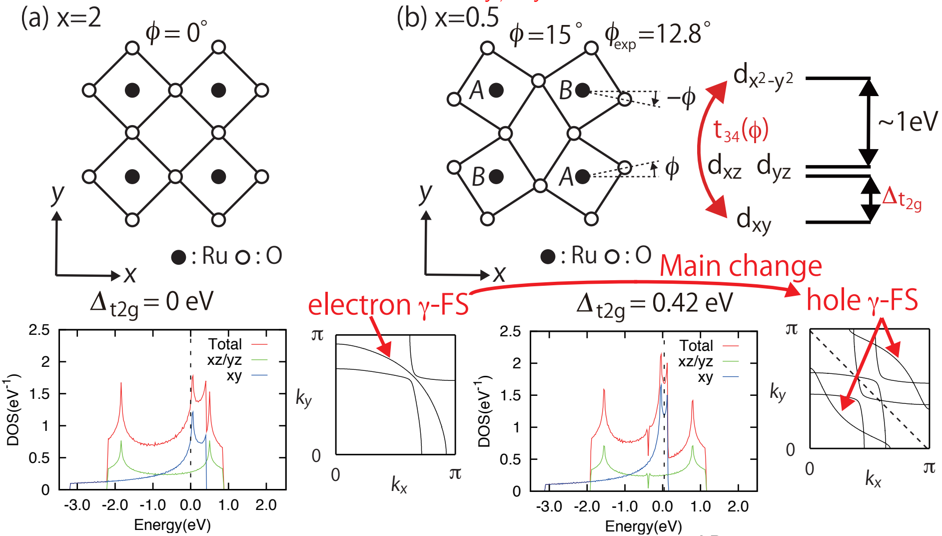






Details→ N. Arakawa and M. Ogata, PRB **86**, 125126 (2012)

- Main changes induced by the octahedral rotation:
  - Reduction of the NN hopping int.s (mainly for d_{xy})
  - Downwards shift for d_{xy} due to $t^{AB}_{xy,x^2\text{-}y^2}(\phi)$



Cf. Difference with LSDA: Small FS $\boldsymbol{k} \approx (0,0)$ due to $\epsilon^{AB}_{xy,x^2\text{-}y^2}(\boldsymbol{k},\phi)$

LDA: Z. Fang and K. Terakura, PRB **64**, 020509(R) (2001), LSDA: T. Oguchi, JPSJ **78**, 044702 (2009)

Treatment of electron correlation: FLEX approximation

$$\hat{H} = \sum_{\boldsymbol{k}}' \sum_{a,b=1}^{3} \sum_{l,l'=A,B} \epsilon_{ab}^{ll'}(\boldsymbol{k},\phi)\hat{c}_{\boldsymbol{k}al}^{\dagger}\hat{c}_{\boldsymbol{k}bl'} + U\sum_{\boldsymbol{i},a}\hat{n}_{\boldsymbol{i}a\uparrow}\hat{n}_{\boldsymbol{i}a\downarrow} + U'\sum_{\boldsymbol{i}}\sum_{a>b}\hat{n}_{\boldsymbol{i}a}\hat{n}_{\boldsymbol{i}b}$$

$$-J_{\mathrm{H}}\sum_{\boldsymbol{i}}\sum_{a>b}(2\hat{\boldsymbol{s}}_{\boldsymbol{i}a}\cdot\hat{\boldsymbol{s}}_{\boldsymbol{i}b}+\frac{1}{2}\hat{n}_{\boldsymbol{i}a}\hat{n}_{\boldsymbol{i}b}) + J'\sum_{\boldsymbol{i}}\sum_{a>b}\hat{c}_{\boldsymbol{i}a\uparrow}^{\dagger}\hat{c}_{\boldsymbol{i}a\downarrow}^{\dagger}\hat{c}_{\boldsymbol{i}b\downarrow}\hat{c}_{\boldsymbol{i}b\uparrow}$$

- Merits of the FLEX approx.:
  - To partially take account of the mode-mode coupling
  - To satisfy several conservation laws automatically

  $\rightarrow$ Possible to discuss properties at low T near a QCP!

Treatment of electron correlation: FLEX approximation

$$\hat{H} = \sum_{\boldsymbol{k}}^{\prime} \sum_{a,b=1}^{3} \sum_{l,l'=A,B} \epsilon_{ab}^{ll'}(\boldsymbol{k}, \phi)\hat{c}_{\boldsymbol{k}al}^{\dagger}\hat{c}_{\boldsymbol{k}bl'} + U\sum_{i,a}\hat{n}_{ia\uparrow}\hat{n}_{ia\downarrow} + U'\sum_{i}\sum_{a>b}\hat{n}_{ia}\hat{n}_{ib}$$

$$-J_{\mathrm{H}}\sum_{i}\sum_{a>b}(2\hat{\boldsymbol{s}}_{ia}\cdot\hat{\boldsymbol{s}}_{ib} + \tfrac{1}{2}\hat{n}_{ia}\hat{n}_{ib}) + J'\sum_{i}\sum_{a>b}\hat{c}_{ia\uparrow}^{\dagger}\hat{c}_{ia\downarrow}^{\dagger}\hat{c}_{ib\downarrow}\hat{c}_{ib\uparrow}$$

- Merits of the FLEX approx.:
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  → Possible to discuss properties at low T near a QCP!
- FLEX approx. for $\Phi_{\mathrm{LW}}[\hat{G}]$ consisting of el-h bubbles and ladders
  1. $\hat{G}(k) = \hat{G}^0(k) + \hat{G}^0(k)\hat{\Sigma}(k)\hat{G}(k)$
  2. $\hat{\chi}(q) = -\frac{T}{N}\sum_{k}\hat{G}(k)\hat{G}(k+q)$
  3. $\hat{\chi}^{\mathrm{S}}(q) = (\hat{1} - \hat{\chi}(q)\hat{\Gamma}^{\mathrm{S}})^{-1}\hat{\chi}(q),\ \hat{\chi}^{\mathrm{C}}(q) = (\hat{1} - \hat{\chi}(q)\hat{\Gamma}^{\mathrm{C}})^{-1}\hat{\chi}(q)$
  4. $\hat{V}(q) = \frac{3}{2}\hat{\Gamma}^{\mathrm{S}}\hat{\chi}^{\mathrm{S}}(q)\hat{\Gamma}^{\mathrm{S}} + \frac{1}{2}\hat{\Gamma}^{\mathrm{C}}\hat{\chi}^{\mathrm{C}}(q)\hat{\Gamma}^{\mathrm{C}} + (\frac{3}{2}\hat{\Gamma}^{\mathrm{S}} + \frac{1}{2}\hat{\Gamma}^{\mathrm{C}}) - \hat{\Gamma}^{\uparrow\downarrow}\hat{\chi}(q)\hat{\Gamma}^{\uparrow\downarrow}$
  5. $\hat{\Sigma}(k) = \frac{T}{N}\sum_{q}\hat{V}(q)\hat{G}(k-q)$

T. Takimoto *et al.*, PRB (2004); H. Ikeda *et al.*, PRB (2010)

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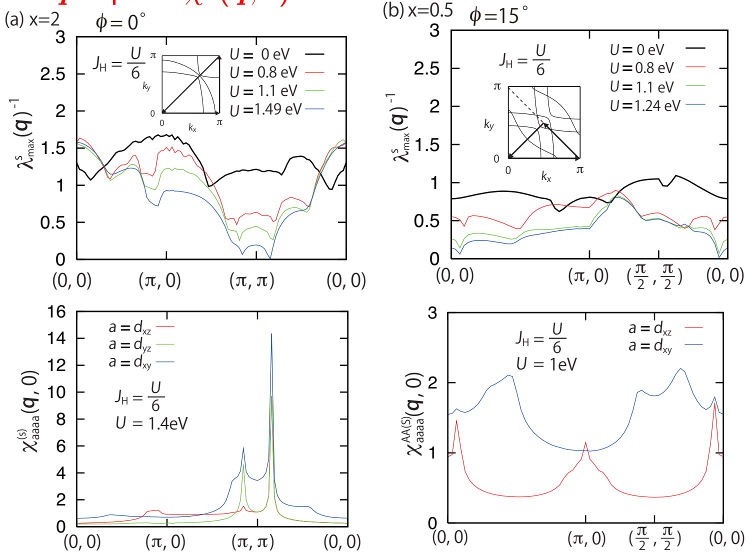
Roles of the rotation in Mag. prop.s

- IC FM spin fluc.s $\nearrow$, related with two dominant fluc.s for $x = 2$
- Flat $\boldsymbol{q}$ dep. of $\hat{\chi}^{\mathrm{S}}(\boldsymbol{q}, 0)$

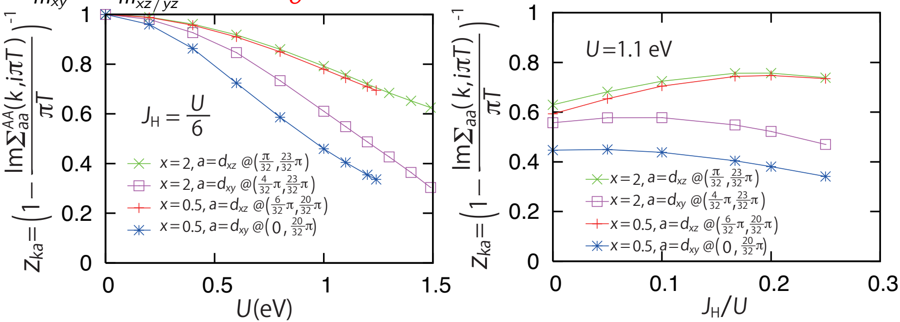


BZ= 64×64, 1024 Matsubara freq., $T = 0.007$eV

Cf. RPA for $x = 2$: T. Nomura and K. Yamada, JPSJ (2000); FLEX for $x = 2$: Y. Yanase and M. Ogata, JPSJ (2003)

Roles of the rotation in Renormalization of QPs

- Enhancement of $\frac{m^*_{xy}}{m_{xy}} (\sim z^{-1}_{\boldsymbol{k}d_{xy}})$ and $\frac{m^*_{xz/yz}}{m_{xz/yz}} (\sim z^{-1}_{\boldsymbol{k}d_{xz/yz}})$
- Enhancement of $\frac{m^*_{xy}}{m^*_{xz/yz}}$
- $\frac{m^*_{xy}}{m_{xy}} > \frac{m^*_{xz/yz}}{m_{xz/yz}}$ for all $\frac{J_{\mathrm{H}}}{U}$



Cf. DMFT for $x = 2$ with $U = 2.1$eV;

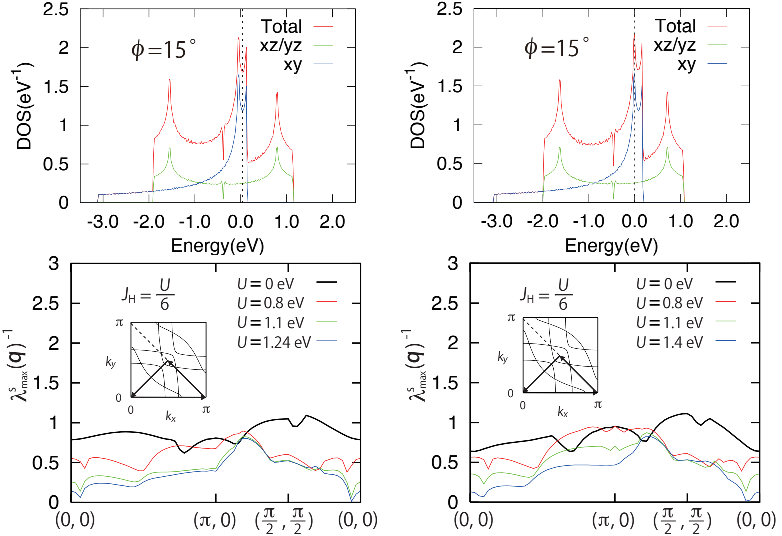
J [eV]	$m^*/m_{\mathrm{LDA}}\|_{xy}$	$m^*/m_{\mathrm{LDA}}\|_{xz}$	T^*_{xy} [K]	T^*_{xz} [K]
0.0, 0.1	1.7	1.7	>1000	>1000
0.2	2.3	2.0	300	800
0.3	3.2	2.4	100	300
0.4	4.5	3.3	60	150

J. Mravlje *et al.*, PRL (2011)

Roles of the vHs in Mag. prop.s

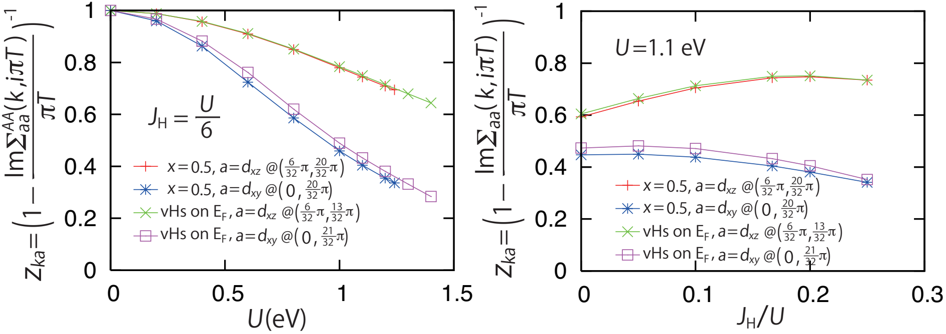
- IC FM spin fluc.s: dominant
- Location of the vHs $\leftrightarrow$ Flat $\boldsymbol{q}$ dep. of $\hat{\chi}^{S}(\boldsymbol{q}, 0)$

ii) $x = 0.5$ (vHs below E_F) $\Delta_{t2g} = 0.39$ eV　　iii) vHs on E_F $\Delta_{t2g} = 0.27$ eV



Roles of the vHs in Renormalization of QPs

- Slight deviation of vHs → Enhancement of $\frac{m^*_{xy}}{m_{xy}}$
- $\frac{m^*_{xy}}{m_{xy}} > \frac{m^*_{xz/yz}}{m_{xz/yz}}$ for all $\frac{J_{\mathrm{H}}}{U}$



Q. WHAT is more important in enhancing m^* than the location of the vHs?
A. Flat $\boldsymbol{q}$ dep. of $\hat{\chi}^{\mathrm{S}}(\boldsymbol{q},0)$.

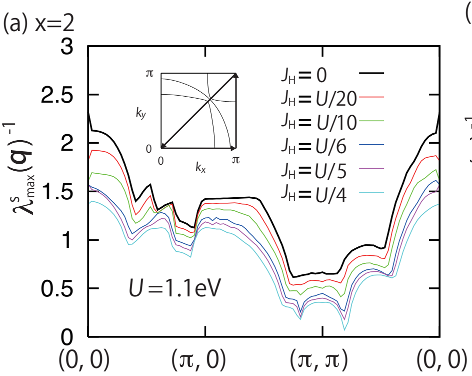
Summary and Message

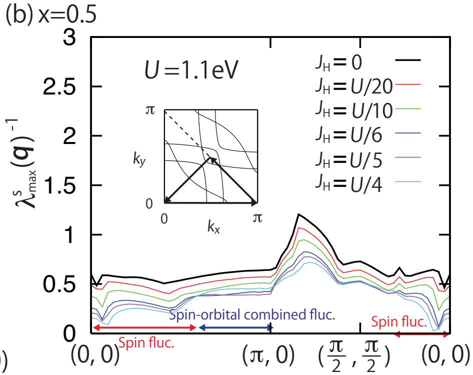
- Roles of the octahedral rotation:
  - IC FM spin fluc.s$\nearrow$, related with dominant fluc.s for $x = 2$
  - Flat $\boldsymbol{q}$ dep. of $\hat{\chi}^{\mathrm{S}}(\boldsymbol{q}, 0)$
  - Enhancement of $\frac{m^*_{xy}}{m_{xy}}$ and $\frac{m^*_{xz/yz}}{m_{xz/yz}}$
  - Enhancement of $\frac{m^*_{xy}}{m^*_{xz/yz}}$
  - $\frac{m^*_{xy}}{m_{xy}} > \frac{m^*_{xz/yz}}{m_{xz/yz}}$ for all $\frac{J_{\mathrm{H}}}{U}$
- Roles of the vHs for the d_{xy} orbital:
  - IC FM spin fluc.s: dominant
  - Flat $\boldsymbol{q}$ dep. of $\hat{\chi}^{\mathrm{S}}(\boldsymbol{q}, 0)$ due to the slight deviation of vHs
  - $\frac{m^*_{xy}}{m_{xy}} \nearrow$ due to the slight deviation of vHs
  - $\frac{m^*_{xy}}{m_{xy}} > \frac{m^*_{xz/yz}}{m_{xz/yz}}$ for all $\frac{J_{\mathrm{H}}}{U}$

Q. WHAT is more important in enhancing m^* than the location of the vHs?
A. Flat $\boldsymbol{q}$ dep. of $\hat{\chi}^{\mathrm{S}}(\boldsymbol{q}, 0)$.

Effects of J_{H} on Mag. prop.s

- $x = 2$: Monotonic enhancement, i.e. enhancement of spin fluc.s
- $x = 0.5$: Monotonic enhancement around $\boldsymbol{q} = (0, 0)$ and
  Nonmonotonic enhancement around $\boldsymbol{q} = (\pi, 0)$





Cf. For spin-orbital combined fluc., see Y. Yamashita and K. Ueda, PRB (2003).