# Theoretical Study of Van-Vleck susceptibility in $Sr_2RuO_4$

Masafumi Udagawa[1]    Youichi Yanase[2]

[1] Univ. of Tokyo, School of Engineering, Dept. of Applied Physics
[2] Univ. of Tokyo, School of Science, Dept. of Physics

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# Outline

§ Introduction

- Non-conserved magnetization and its susceptibility
- Experimental relevance
- Purpose of the study

§ Model & Method

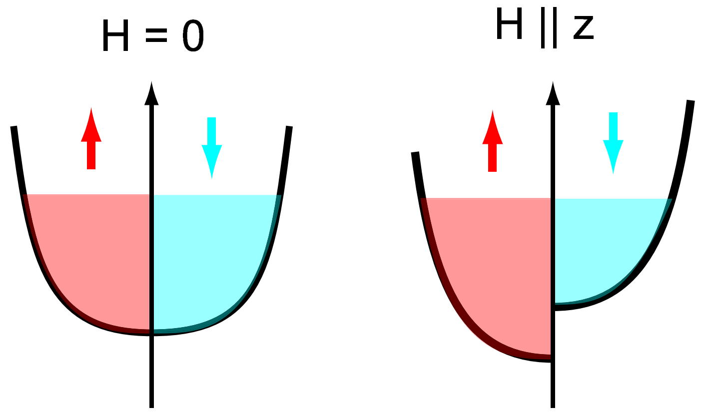
- Multi-orbital Hubbard model
- Dynamical mean-field theory
- Formulation of Van-Vleck susceptibility

§ Results

- Effects of electron correlation of Van-Vleck susceptibility
- Emergence of diagonal components

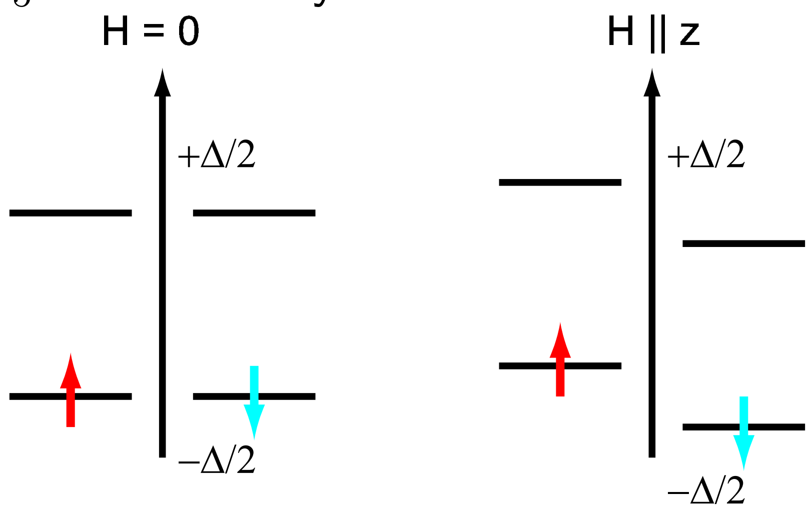
## Pauli and Van-Vleck susceptibility

§ Metallic system (Pauli paramagnetism)




$$\chi \longrightarrow \chi_P \ (\neq 0) \text{ as } T \longrightarrow 0$$

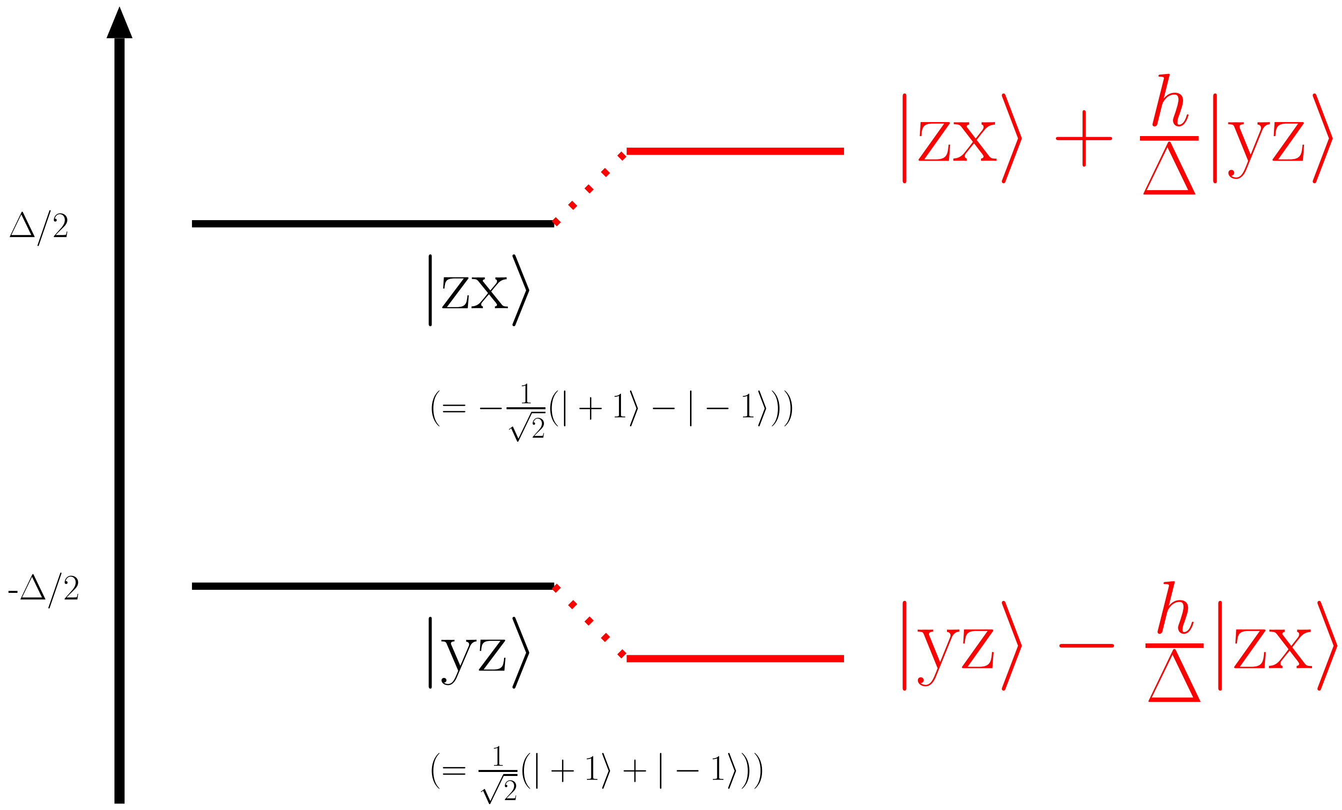
§ Two-level system




$$\chi \longrightarrow 0 \text{ as } T \longrightarrow 0$$

Only if spin (magnetization) is conserved

# Pauli and Van-Vleck susceptibility




$$\mathcal{H} = \begin{bmatrix} +\frac{\Delta}{2} & -h \\ -h & -\frac{\Delta}{2} \end{bmatrix} \qquad M_z = \langle l_z + 2s_z \rangle = \frac{2h}{\Delta}$$

## Non-commutativity of magnetization and Hamiltonian

$M_z \equiv l_z + 2s_z$ does not commute with $\mathcal{H}$: $[M_z, \mathcal{H}] \neq 0$

$\Rightarrow$ The ground state acquires magnetization by modulating wavefunction

§ Pauli and Van Vleck

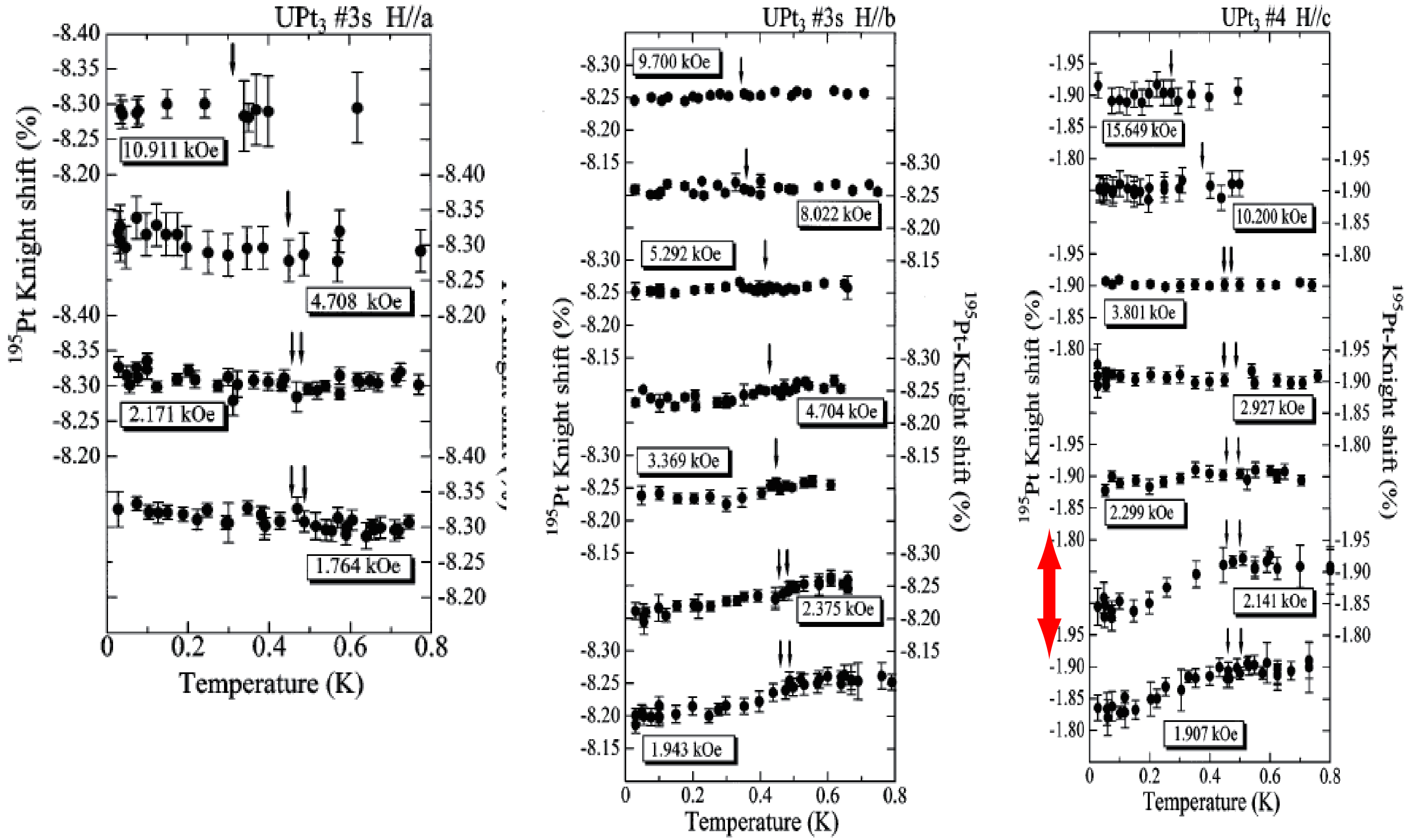
- Pauli ($\chi_P$) $\Leftarrow$ Fermi surface
- Van Vleck ($\chi_{VV}$) $\Leftarrow$ away from Fermi surface

§ Origins of non-commutativity

- orbital magnetic moment
- spin-orbit interaction
- Spontaneous symmetry breaking

# Relevance in Knight shift

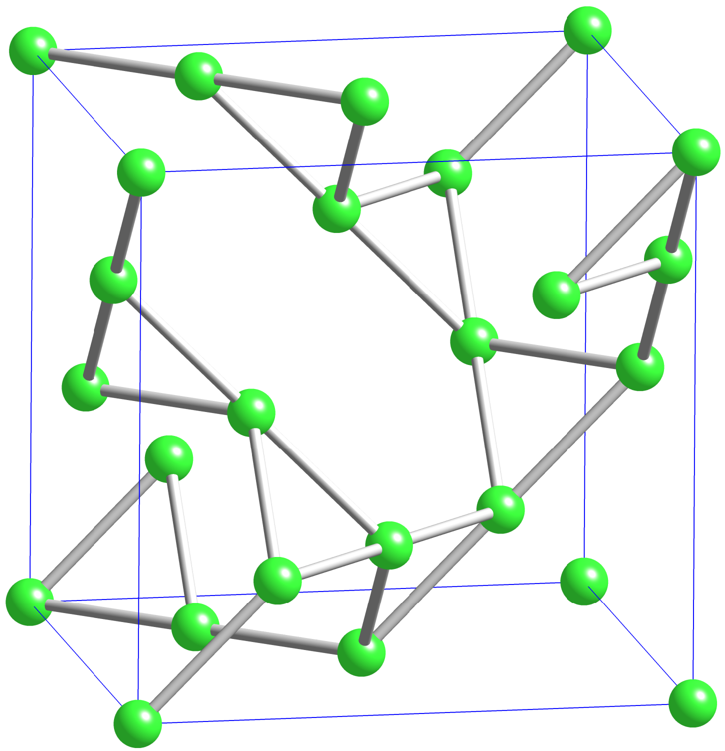
§ $UPt_3$ H. Tou *et al.*, Phys. Rev. Lett.

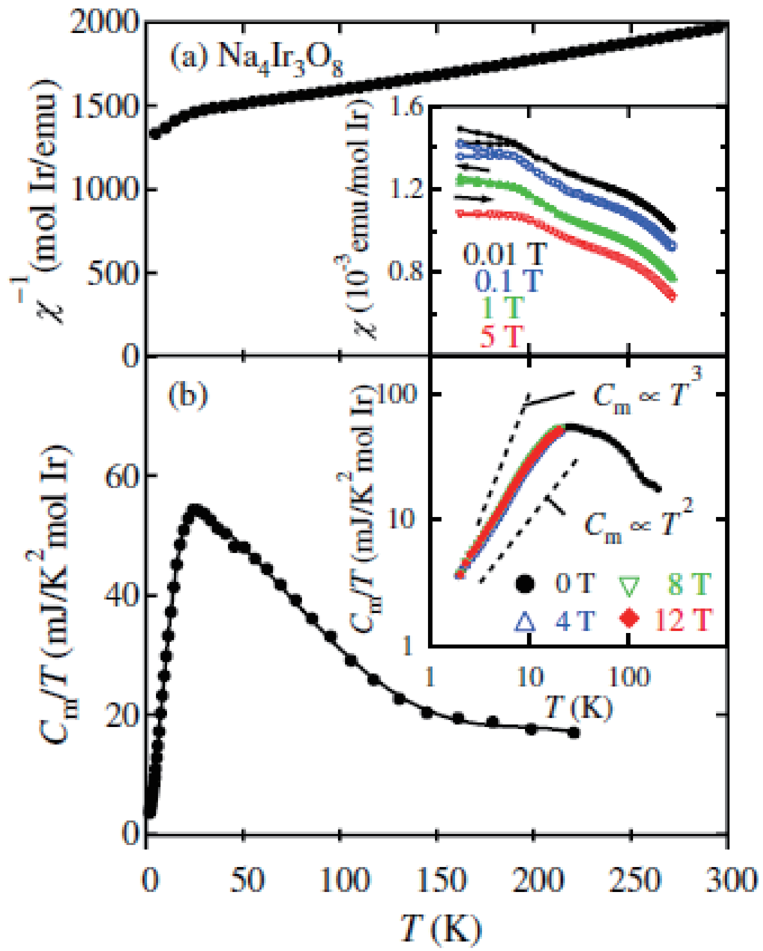



- Only small reduction of Knight shift at $T_c$
- How large $\chi_{VV}$ can be, compared with $\chi_P$ ? $\rightarrow$ relation to electron correlation

# Diverging Wilson ratio

§ “Hyperkagome material” $Na_4Ir_3O_8$






Y. Okamoto *et al.*, Phys. Rev. Lett.

- Wilson ratio $\to \infty$ as $T \to 0$
- Role of large spin-orbit coupling in Ir

## Purpose of our study

How electron correlation affects the Van Vleck susceptibility ?
$\Rightarrow$ study $\chi_{VV}$ of $Sr_2RuO_4$ as a model case

§ Questions

- "Inter-band picture" still valid ?
- What happens in the vicinity of metal-insulator transition ?
- Is it possible that $\chi_{VV} >> \chi_P$ ?

§ Previous studies

Electron correlation is irrelevant for $\chi_{VV}$

- R. Kubo and Y. Obata, J. Phys. Soc. Jpn., **11** 549 (1956)
- Z. Zou and P. W. Anderson, Phys. Rev. Lett., **57** 2073 (1986)

$\chi_{VV}$ is renormalized in the same order as $z^{-1}$

- H. Kontani and K. Yamada, J. Phys. Soc. Jpn., **65** 172 (1995)
- T. Mutou and D. Hirashima, J. Phys. Soc. Jpn., **65** 366 (1996)

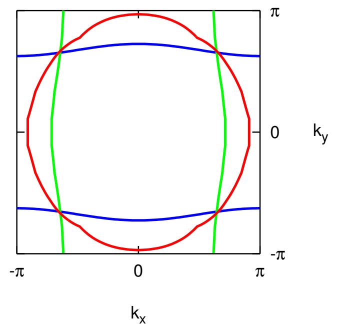
# Model: Multi-orbital Hubbard model

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1 + \mathcal{H}_{ext}.$$

($\mathcal{H}_0$: kinetic energy, $\mathcal{H}_1$: interaction energy, $\mathcal{H}_{ext}$: Zeeman energy)

$$\mathcal{H}_0 = \sum_{\mathbf{k}} \sum_{s=\pm 1} \sum_{a,b} \epsilon_{ab}(\mathbf{k}) c^\dagger_{\mathbf{k}as} c_{\mathbf{k}bs}$$

$$\begin{cases} \epsilon_{00}(\mathbf{k}) = -2t'_{xy} \cos k_x - 2t_{xy} \cos k_y - \mu, \\ \epsilon_{11}(\mathbf{k}) = -2t_{xy} \cos k_x - 2t'_{xy} \cos k_y - \mu, \\ \epsilon_{22}(\mathbf{k}) = -2t_z(\cos k_x + \cos k_y) - 4t'_z \cos k_x \cos k_y - \mu \end{cases}$$




$$\mathcal{H}_1 = U \sum_a \sum_{\mathbf{k},\mathbf{k}',\mathbf{q}} c^\dagger_{\mathbf{k}'+\mathbf{q}a\downarrow} c^\dagger_{-\mathbf{k}'a\uparrow} c_{-\mathbf{k}a\uparrow} c_{\mathbf{k}+\mathbf{q}a\downarrow} + (U - 2J) \sum_{a>b} \sum_{ss'} \sum_{\mathbf{k},\mathbf{k}',\mathbf{q}} c^\dagger_{\mathbf{k}'+\mathbf{q}as} c^\dagger_{-\mathbf{k}'bs'} c_{-\mathbf{k}bs'} c_{\mathbf{k}+\mathbf{q}as}$$

$$- J \sum_{a>b} \sum_{ss'} \sum_{\mathbf{k},\mathbf{k}',\mathbf{q}} c^\dagger_{\mathbf{k}'+\mathbf{q}as} c^\dagger_{-\mathbf{k}'bs'} c_{-\mathbf{k}bs} c_{\mathbf{k}+\mathbf{q}as'} + J \sum_{a\neq b} \sum_{\mathbf{k},\mathbf{k}',\mathbf{q}} c^\dagger_{\mathbf{k}'+\mathbf{q}a\downarrow} c^\dagger_{-\mathbf{k}'a\uparrow} c_{-\mathbf{k}b\uparrow} c_{\mathbf{k}+\mathbf{q}b\downarrow}$$

$$\mathcal{H}_{ext} = -h \sum_{\mathbf{k}} \sum_{s=\pm 1} \sum_{a,b} (l_z + 2s_z)_{ab} c^\dagger_{\mathbf{k}as} c_{\mathbf{k}bs}$$

$$= \sum_{\mathbf{k}} \sum_{s=\pm 1} \begin{pmatrix} c^\dagger_{\mathbf{k}0s} & c^\dagger_{\mathbf{k}1s} & c^\dagger_{\mathbf{k}2s} \end{pmatrix} \begin{pmatrix} -sh & h & 0 \\ h & -sh & 0 \\ 0 & 0 & -sh \end{pmatrix} \begin{pmatrix} c_{\mathbf{k}0s} \\ c_{\mathbf{k}1s} \\ c_{\mathbf{k}2s} \end{pmatrix}.$$

- orbital angular momentum only between $d_{yz}$ abd $d_{zx}$

# Magnetic susceptibility

§ Definition

$$\chi^{zz}_{\mathbf{q}}(i\nu_q) = \frac{1}{N}\int_0^\beta d\tau\ e^{i\nu_q\tau}\langle M^z_{-\mathbf{q}}(\tau)M^z_{\mathbf{q}}(0)\rangle, \quad M^z_{\mathbf{q}} = \sum_{\mathbf{k}}\sum_{s}\sum_{a,b}(l^z+2s^z)^{ss}_{ab}c^\dagger_{\mathbf{k}+\mathbf{q}as}c_{\mathbf{k}bs}$$

§ Separation of Pauli ($\chi_P$) and Van-Vleck ($\chi_{VV}$) terms (Kontani and Yamada)

$$\chi_{tot} = \lim_{\mathbf{q}\to 0}\lim_{\omega\to 0}\chi_{\mathbf{q}}(\omega)$$

$$\chi_{VV} = \lim_{\omega\to 0}\lim_{\mathbf{q}\to 0}\chi_{\mathbf{q}}(\omega)$$

$$\chi_P = \chi_{tot} - \chi_{VV}$$

- $\chi_P$ vanishes when the Fermi surface disappears

$c.f.$ Non-commutativity of $q$-limit and $\omega$-limit (Abrikosov, Gorkov, Dzyaloshinski)

$$G(\mathbf{p},\epsilon)G(\mathbf{p}+\mathbf{q},\epsilon+\omega) = 2\pi i Z^2\frac{\mathbf{v}\cdot\mathbf{q}}{\omega-\mathbf{v}\cdot\mathbf{q}}\delta(\epsilon)\delta(\xi_k) + \phi(\mathbf{q},\omega)$$

$\Rightarrow$ Only $\phi(\mathbf{q},\omega)$ (non-singular as to the Fermi surface) contributes to $\chi_{VV}$

# Method: dynamical mean-field theory

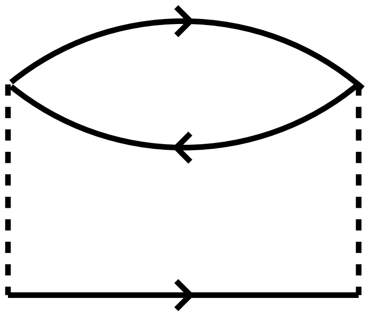
§ Effective action of impurity model: Repeat (1) and (2) until convergence

$$S_{eff} = \int_0^\beta d\tau_1 \int_0^\beta d\tau_2 \sum_{a,b,\sigma} c_{a\sigma}(\tau_1) g^{-1}_{a,b,\sigma}(\tau_1 - \tau_2) c_{b\sigma}(\tau_2) - U \int_0^\beta d\tau \sum_\gamma n_{\gamma\uparrow}(\tau) n_{\gamma\downarrow}(\tau)$$

(1) Impurity solver – Iterative perturbation theory

$$\Sigma_{as}(\tau) = -\big[U^2 G_a(\tau)^2 G_a(-\tau) + 2(U'^2 + U' J_H + J_H^2) G_a(\tau) \sum_{b\neq a} G_b(\tau) G_b(-\tau)$$

$$+ J^2 G_a(-\tau) \sum_{b\neq a} G_b(\tau)^2\big] + \text{(Hartree-type terms)}$$

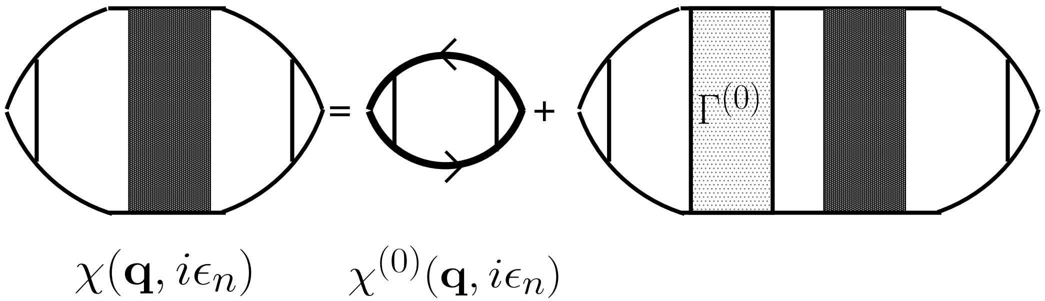


(2) Self-consistent determination of Weiss field $g_{a,b,\sigma}(\tau)$

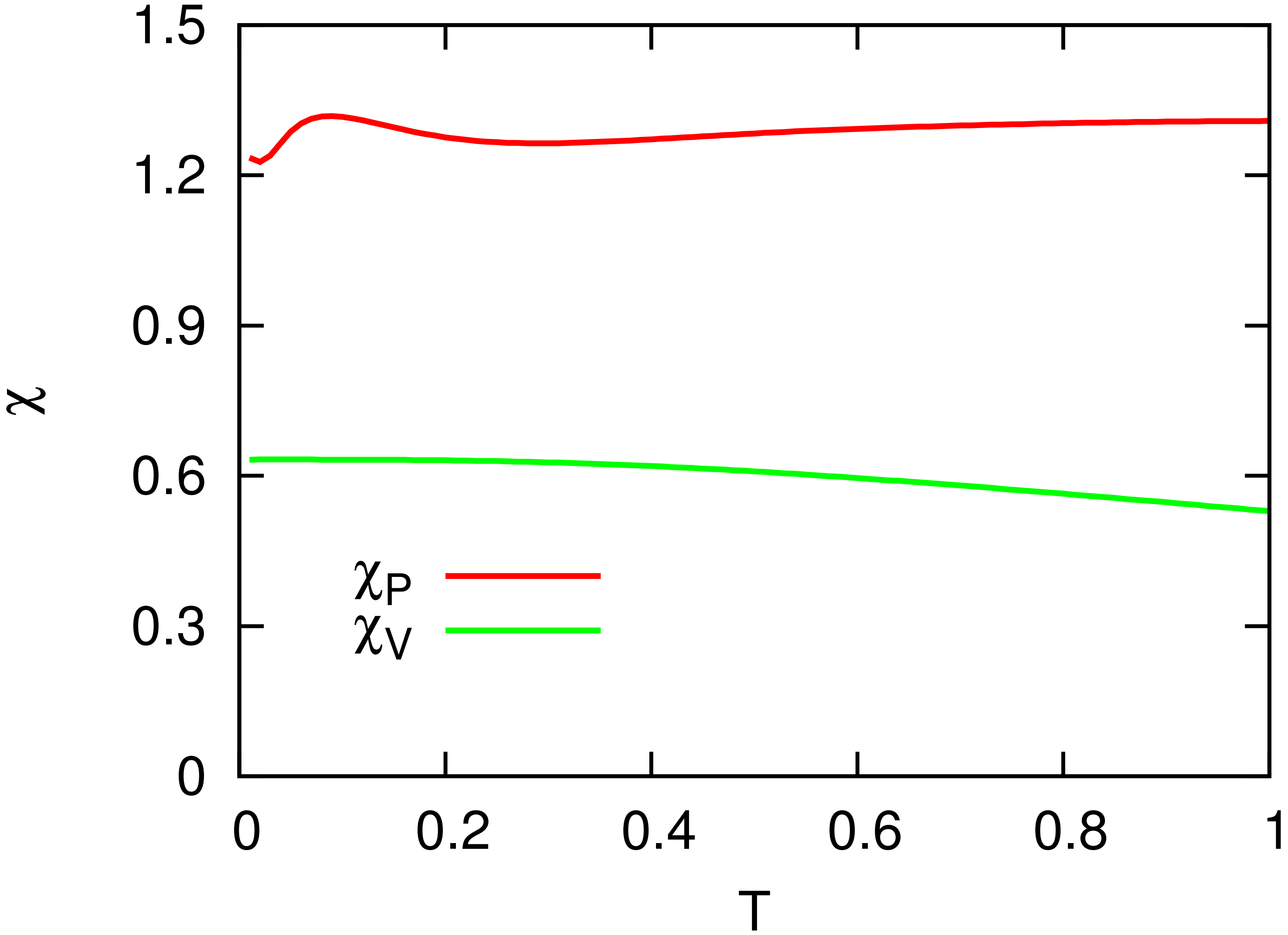
$$g^{-1}_{a,b,\sigma}(i\epsilon_n) = \Big[\frac{1}{N}\sum_{\mathbf{k}} G_{a,b,\sigma}(\mathbf{k}, i\epsilon_n)\Big]^{-1} + \Sigma_{a,b,\sigma}(i\epsilon_n)$$

$$\Big(G_{a,b,\sigma}(\mathbf{k}, i\epsilon_n) = \frac{1}{i\epsilon_n + \mu - t_{ab}(\mathbf{k}) - \Sigma_{a,b,\sigma}(i\epsilon_n)}\Big)$$

§ Magnetic susceptibility – Irreducible vertex $\Gamma^{(0)}$ expanded up to $O(H_1^1)$ (only RPA term)



**Results:** $\chi_P$ **and** $\chi_{VV}$ **@** $U = U' = J = 0$



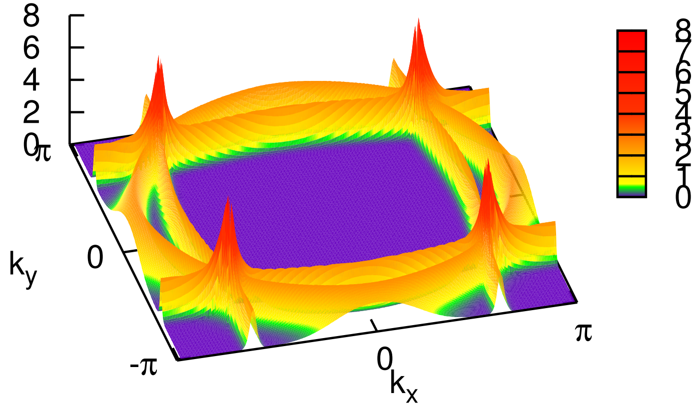

- $\chi_{VV} \simeq \frac{1}{2}\chi_P$

## Results: Momentum dependence @ $U = U' = J = 0$

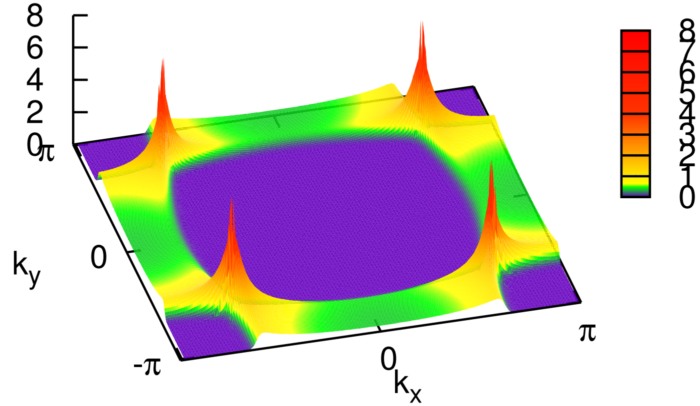
$$\chi^{zz}_{\mathbf{q}=0}(i\nu_q) = \frac{1}{N}\sum_{\mathbf{k}} \chi^{zz}_{\mathbf{q}=0}(i\nu_q, \mathbf{k}): \qquad \chi^{zz}_{\mathbf{q}=0}(i\nu_q, \mathbf{k}) = \sum_{ab} \chi^{zz}_{\mathbf{q}=0ab}(i\nu_q, \mathbf{k})$$

§ Analysis of $\chi^{zz}_{\mathbf{q}=0}(i\nu_q, \mathbf{k})$

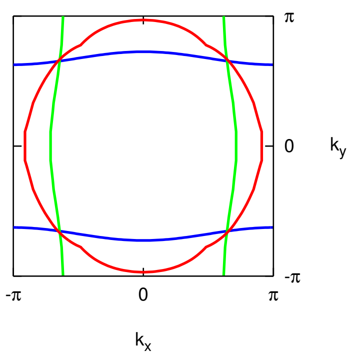
- $\chi_P$ (from $a = b$)




- $\chi_{VV}$ (from $a \neq b$)




§ Fermi surface ($d_{xy}$, $d_{zx}$, $d_{yz}$)




- diagonal part ($a = b$) $\to \chi_P$, offdiagonal part ($a \neq b$) $\to \chi_{VV}$
- $\chi_P$ enhanced near Fermi surface
- $\chi_{VV}$ enhanced near intersecting points of $d_{yz}$ and $d_{zx}$ bands

## Results: Effects of electron correlation @ $T = 0.01$, $U' = U - 2J$

§ $\chi_P$

§ $\chi_{VV}$

- $\chi_{VV}$ and $\chi_P$ are enhanced as increasing $U$
- $\chi_{VV}$ is enhanced as increasing $U'$, while $\chi_P$ is suppressed.

## Results: Bare susceptibility @ $T = 0.01$, $U' = U - 2J$

§ $\chi_P^{(0)}$

§ $\chi_{VV}^{(0)}$

§ $\mathrm{Im}\Sigma(\epsilon + i\delta)(d_{yz,zx})$ @ $U' = 0.6U$

- $\chi_P^{(0)}$ is suppressed as increasing $U'$
- $\chi_{VV}^{(0)}$ is enhanced as increasing $U'$

in spite of the larger self-energy effect

How $\chi_{VV}^{(0)}$ becomes enhanced ?

## Results: Momentum dependence of $\chi^{(0)}_{VV}$ @ $(U' = 0.6U, J = 0.2U)$

- $U = 0.0$
- $U = 1.0$
- $U = 2.0$
- $U = 3.0$

## Results: Origin of the contribution from diagonal part

$$\lim_{\omega\to 0}\lim_{\mathbf{q}\to 0}\chi^{zz(0)}_{\mathbf{q}aa}(\omega+i\delta,\mathbf{k})$$

$$= -\int_{-\infty}^{\infty} d\epsilon\, \frac{f(\epsilon)}{\pi}\, \frac{-\mathrm{Im}\Sigma_a(\epsilon+i\delta)}{(\epsilon-\xi_a(\mathbf{k})-\mathrm{Re}\Sigma_a(\epsilon+i\delta))^2+(\mathrm{Im}\Sigma_a(\epsilon+i\delta))^2}$$

$$\times \frac{\epsilon-\xi_a(\mathbf{k})-\mathrm{Re}\Sigma_a(\epsilon+i\delta)}{(\epsilon-\xi_a(\mathbf{k})-\mathrm{Re}\Sigma_a(\epsilon+i\delta))^2+(\mathrm{Im}\Sigma_a(\epsilon+i\delta))^2}$$

- The integrand has a broad ($\simeq$ Im$\Sigma$) peak away from Fermi level
- Relevant in superconducting phase ?
- Temperature dependence ?

Large contribution not from the Fermi level, but from its neighborhood

## Summary

§ We have studied the Pauli and Van-Vleck susceptibility of $Sr_2RuO_4$ by applying dynamical mean-field theory to the multi-orbital Hubbard model.

§ We found that the bare Van-Vleck term is enhanced, as increasing electron correlation, in contrast to the Pauli term.

§ The growth of Van-Vleck term can be attributed to the diagonal part of bare susceptibility, which comes from the momentum and energy region slightly away from Fermi level.

### Future problems

§ Sophistication of calculational methods (Quantum Monte Carlo method, 2nd-order correction in irreducible vertex)

$\Rightarrow$ Detailed comparison between $\chi_P$, $\chi_{VV}$ and $Z^{-1}$

§ Temperature dependence of $\chi_{VV}$

§ Behavior in superconducting phase

§ Other systems