Recent progress on the first-principles analysis in heavy-electron systems

Hiroaki Ikeda (Kyoto University)

1. Hidden order in URu_2Si_2
2. Unconventioncal superconductivity in $CeCu_2Si_2$

Hidden order in URu_2Si_2

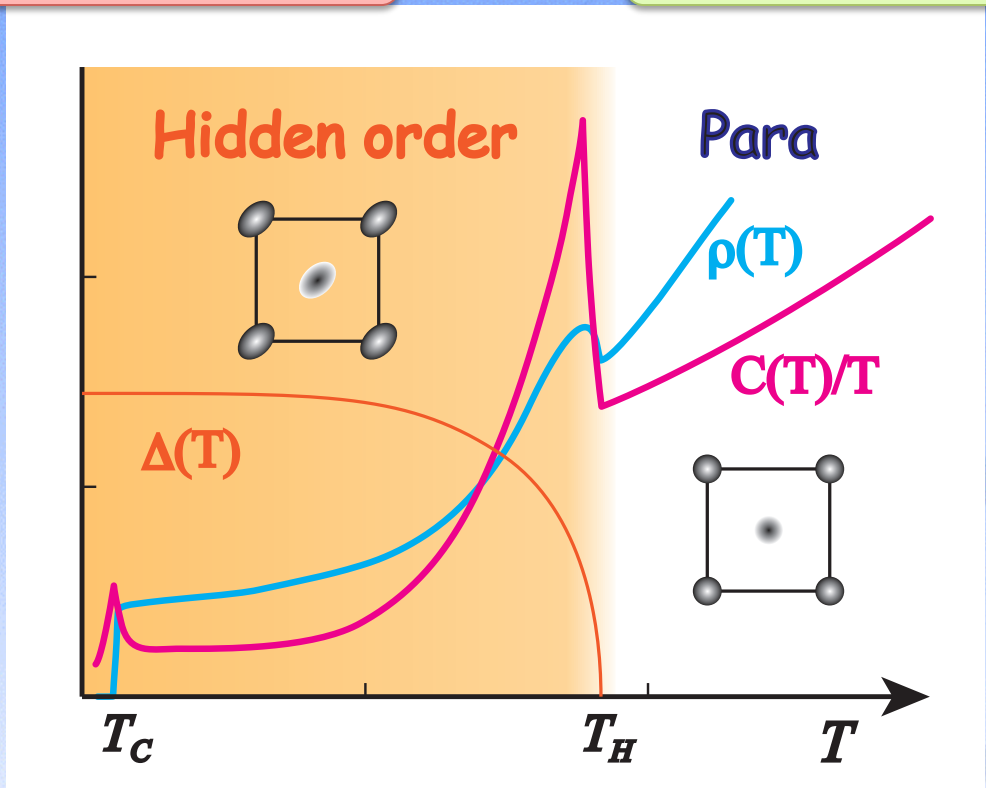
Collaborators

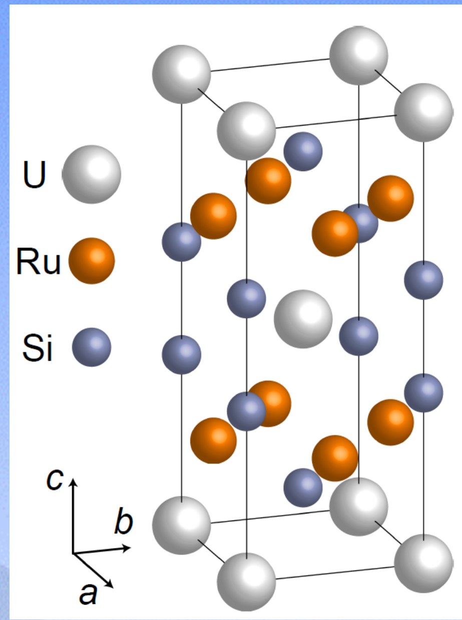
Michi-To Suzuki (CCSE JAEA)
Ryotaro Arita (University of Tokyo)
Testuya Takimoto (APCTP Pohang)
Takasada Shibauchi (Kyoto University)
Yuji Matsuda (Kyoto University)

URu_2Si_2 (Hidden Order)

Specific heat

Resistivity





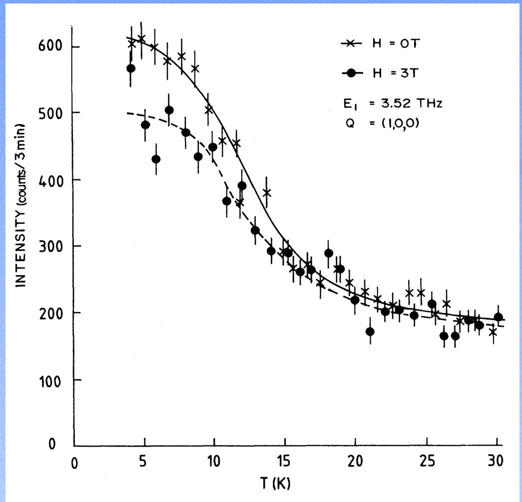
T.T.M Palstra *et al.* PRL (1985)

Superconducting transition

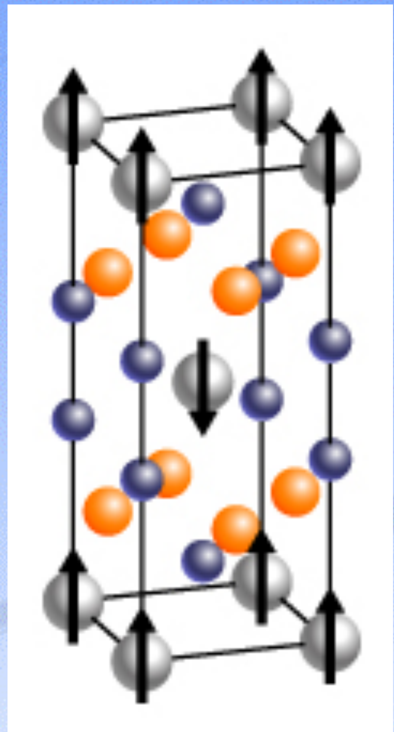
17.5K

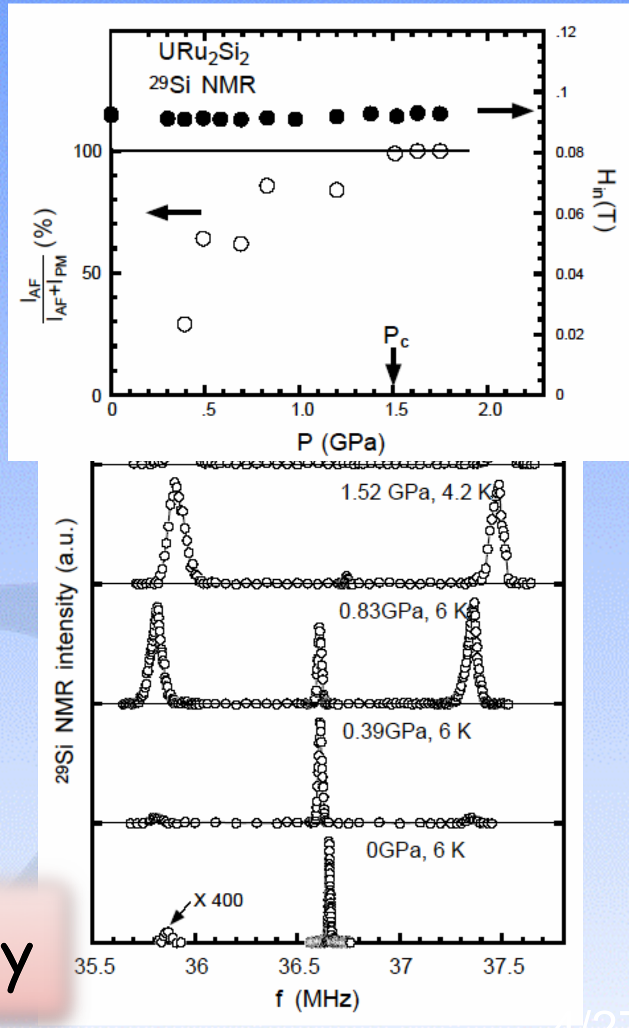
Clear phase transition

Anti-ferromagnetic state？



C. Broholm *et al.* PRB (1991)

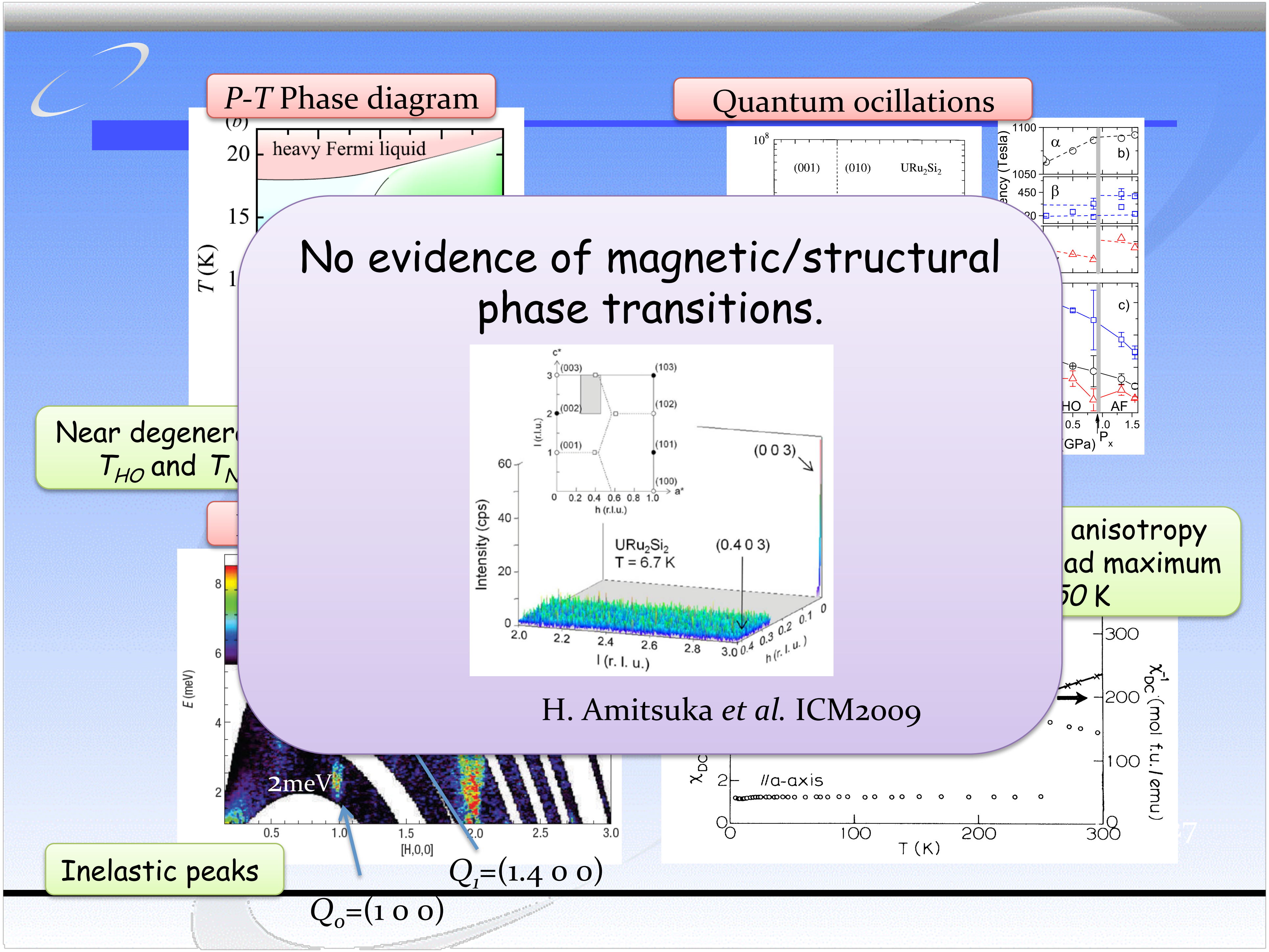




Tiny moment
~$0.03\mu_B$

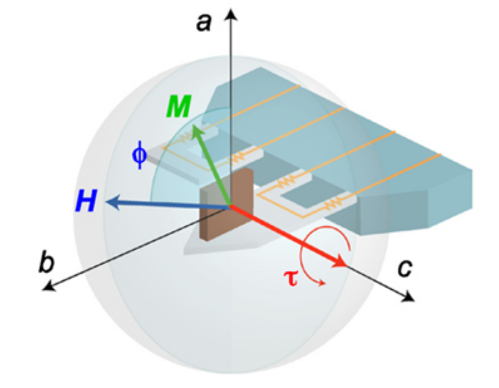
→ inhomogeneity

K. Matsuda *et al.* PRL (2001)

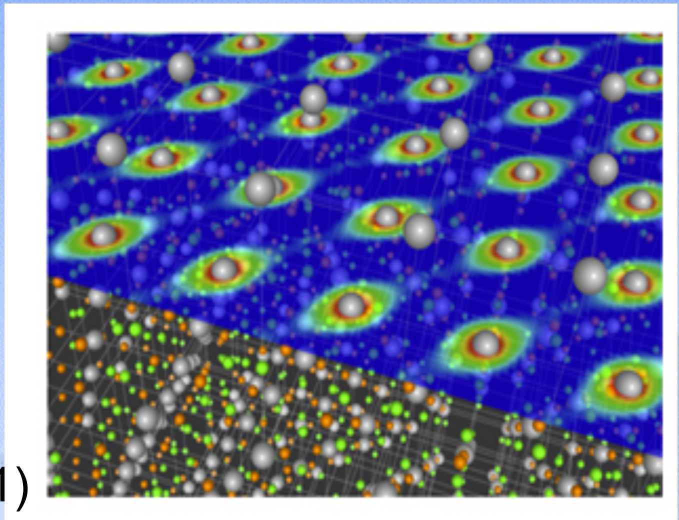

P-T Phase diagram
heavy Fermi liquid
T (K)
Quantum ocillations
(001) (010) URu$_2$Si$_2$
No evidence of magnetic/structural phase transitions.
URu$_2$Si$_2$
T = 6.7 K
(0 0 3)
(0.4 0 3)
Intensity (cps)
l (r. l. u.)
h (r. l. u.)
H. Amitsuka et al. ICM2009
Near degener
T_{HO} and T_N
anisotropy
ad maximum
50 K
//a-axis
T (K)
2meV
Inelastic peaks
Q_1=(1.4 0 0)
Q_0=(1 0 0)

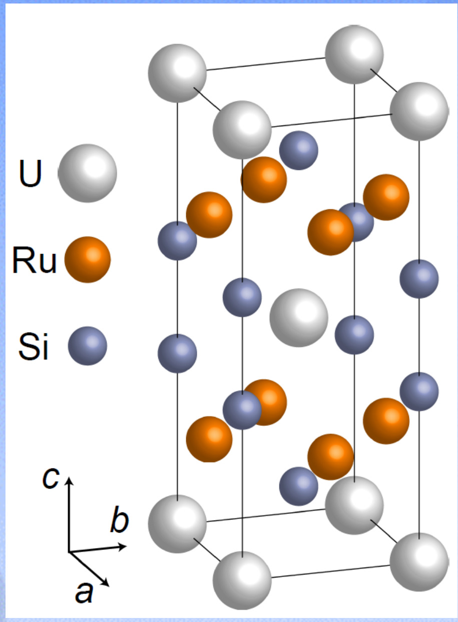
Nematic Electronic State

The magnetic-torque measurement indicates that the in-plane four-fold symmetry is broken.



R.Okazaki et al.
Science 331, 439 (2011)





Cyclotron resonance

S.Tonegawa et al.
PRL 109, 036401(2012)

Possible candidates are restricted.

Thalmeier & Takimoto
PRB 83, 165110(2011)

Proposed theoretical models

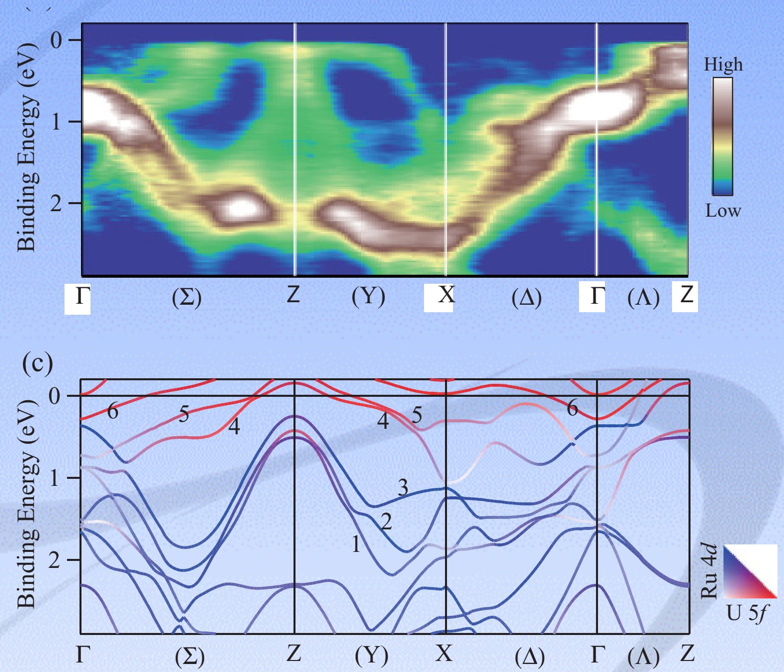
• Double- or triple-spin correlator	V.Barzykin and L.P.Gor'kov, PRL (93)
• Quadrupole (Rank 2)	P.Santini and G.Amoretti, PRL (94)
	H.Harima, K.Miyake, and J.Flouquet, JPSJ (10)
• Octupole (Rank 3)	A.Kiss and P.Fazekas, PRB (05)
• Hexadecapole (Rank 4)	K.Haule and G.Kotliar, Nature Phys. (09)
	H.Kusunose and H.Harima JPSJ(11)
• Dotriacontapole (Rank 5)	F.Cricchio *et al.* PRL (09)
• Spin Density Wave	V.P.Mineev and M.E Zitomirsky, PRB (01)
• Unconventional SDW	H.I and Y.Ohashi, PRL (98)
• d-density wave	A.Virosztek, *et al.* Int. J. Mod. Phys. (02)
• Orbital antiferromagnetism	P.Chandra et al. Nature (02)
• Helicity order	C.M. Varma and L.Zhu, PRL (06)
• Dual model	A.E.Sikkema, *et al.* PRB (96)
	Y.Okuno and K.Miyake, JPSJ (98)
• Spin Nematic	S.Fujimoto PRL (10)
• Hybridization wave	Y.Dubi and A.V.Balatzky PRL (11)
• Modulated spin liquid	C.Pepin et al. PRL (11)
• Hastatic order	P.Chandra, P.Coleman, R.Flint, arXiv (12)

Importance of quantitative analysis

Itinerant f electrons

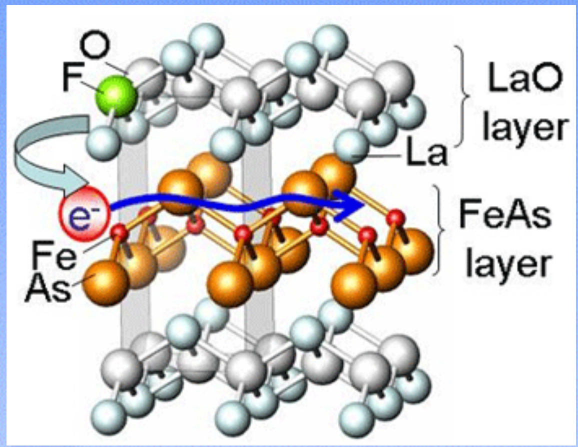
ARPES in paramagnetic phase

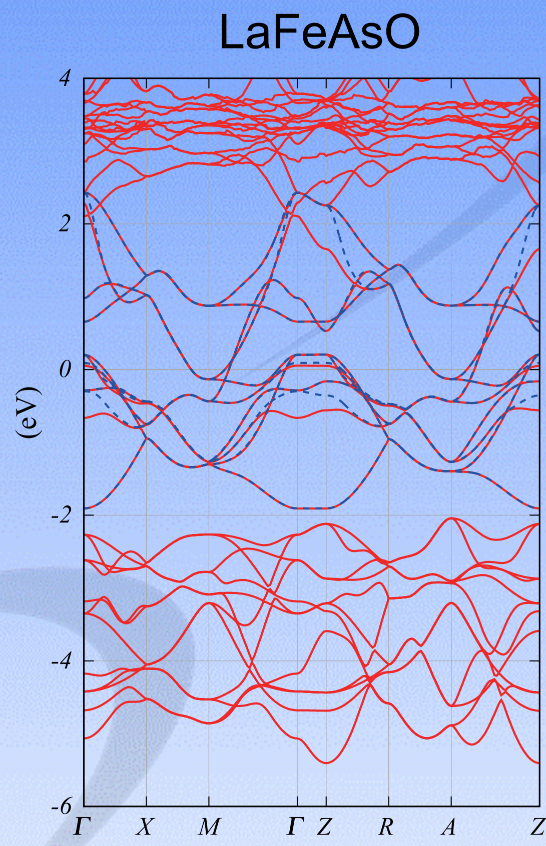
Kawasaki *et al.* PRB (2011)



Importance of study based on the first-principles approach

Application to iron-based superconductors





WIEN2k+wien2wannier+wannier90

P.Blaha *et al. WIEN2k* (01)

J.Kunes, *et al.* Comput. Phys. Commun. (10)

N.Marzari and D.Vanderbilt, PRB (97), *ibid.* (01)

The obtained model Hamiltonian gives us a good starting point, and can describe material dependence of physical properties

nature physics

LETTERS

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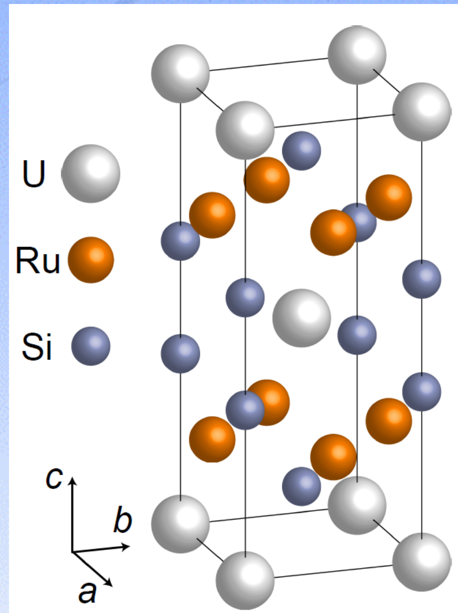
Nature Physics 8, 528 (2012)

Emergent rank-5 nematic order in URu_2Si_2

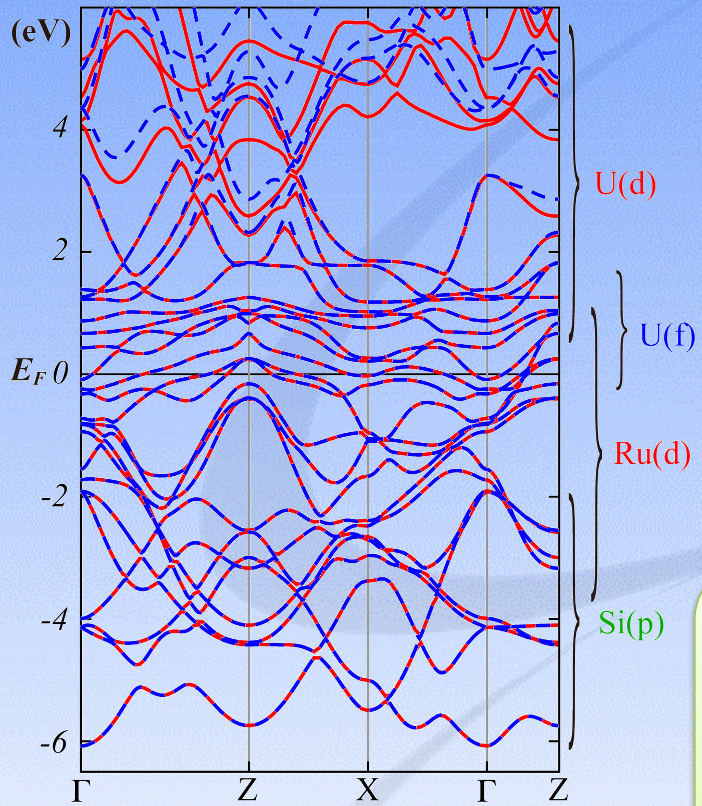
Hiroaki Ikeda[1]*, Michi-To Suzuki[2], Ryotaro Arita[3], Tetsuya Takimoto[4], Takasada Shibauchi[1] and Yuji Matsuda[1]

- ✓ Construction of realistic itinerant model in URu2Si2 based on the first-principles calculations
- ✓ The first report of a complete set of multipole density wave correlations
- ✓ Hidden Order parameter : AF Rank-5 (dotriacontapole) state with E^- irreducible representation (breaking fourfold symmetry and time-reveral symmetry)



Model Hamiltonian in URu_2Si_2

Band structure



$H=H_0+H'$

H is 56 band Anderson lattice model including spin-orbit coupling.

H' is the on-site Coulomb repulsions in the LS basis.

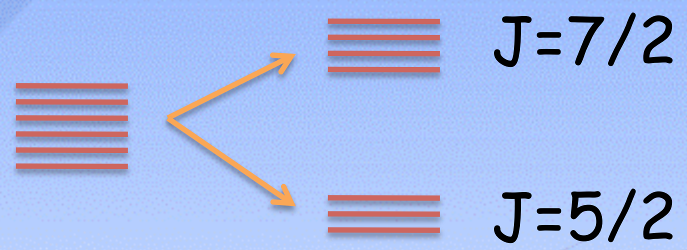
$$
\begin{aligned}
H' = &\frac{U}{2}\sum_{i\ell}\sum_{\sigma} f^{\dagger}_{i\ell\sigma} f^{\dagger}_{i\ell\bar{\sigma}} f_{i\ell\bar{\sigma}} f_{i\ell\sigma} \\
&+ \frac{U'}{2}\sum_{i\ell\neq m}\sum_{\sigma\sigma'} f^{\dagger}_{i\ell\sigma} f^{\dagger}_{im\sigma'} f_{im\sigma'} f_{i\ell\sigma} \\
&+ \frac{J}{2}\sum_{i\ell\neq m}\sum_{\sigma\sigma'} f^{\dagger}_{i\ell\sigma} f^{\dagger}_{im\sigma'} f_{i\ell\sigma'} f_{im\sigma} \\
&+ \frac{J'}{2}\sum_{i\ell\neq m}\sum_{\sigma} f^{\dagger}_{i\ell\sigma} f^{\dagger}_{i\ell\bar{\sigma}} f_{im\bar{\sigma}} f_{im\sigma},
\end{aligned}
$$

We unveil the missing link beyond simple consideration of band structure in URu_2Si_2, based on RPA analysis in the itinerant picture and beyond.

The Key Ingredients

entangled spin and f-orbital degrees of freedom

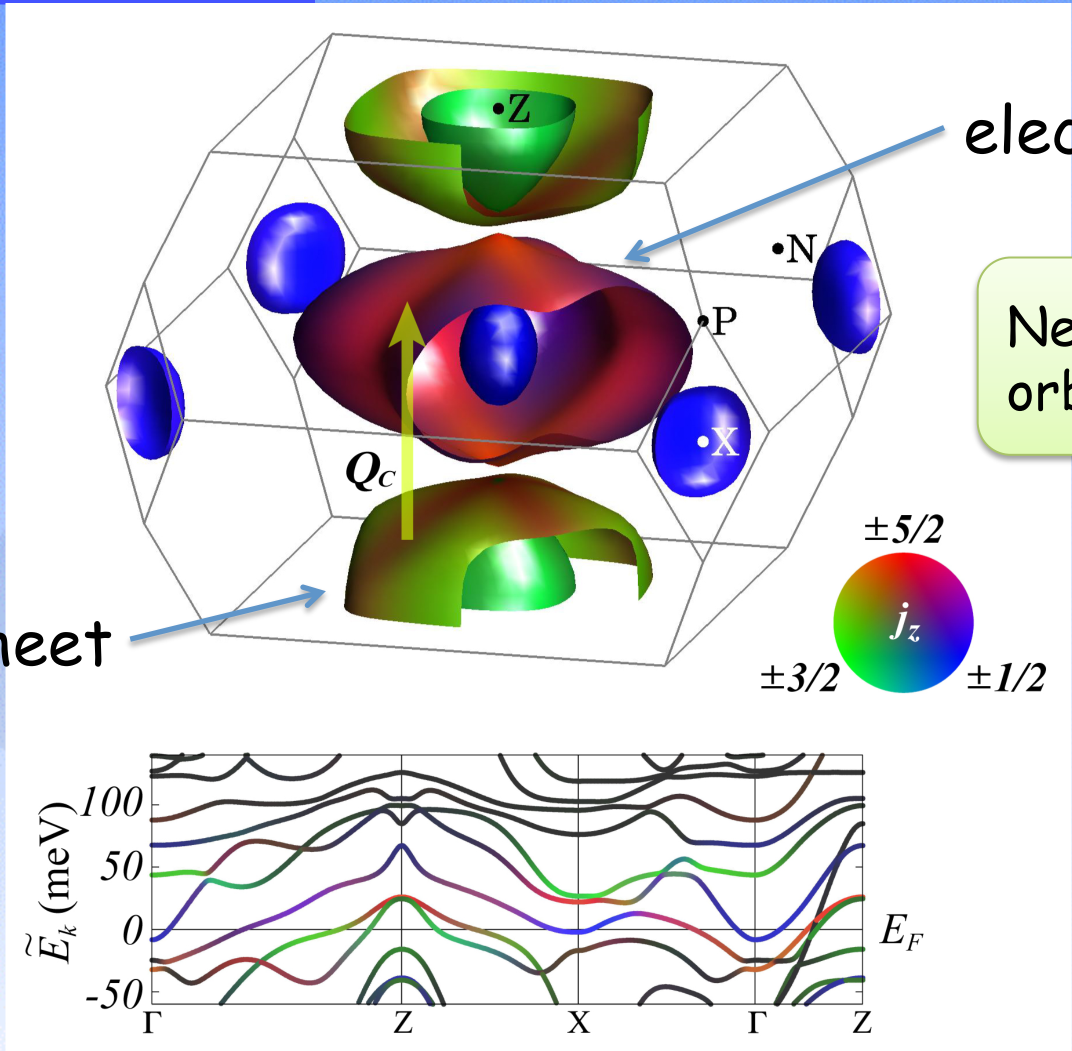
S=1/2 and L=3 → J=L+S



f^1 in Ce $\quad\quad$ f^2 or f^3 in U

Near the Fermi level is dominated by this J=5/2 components

J-Resolved Fermi surface and Nesting



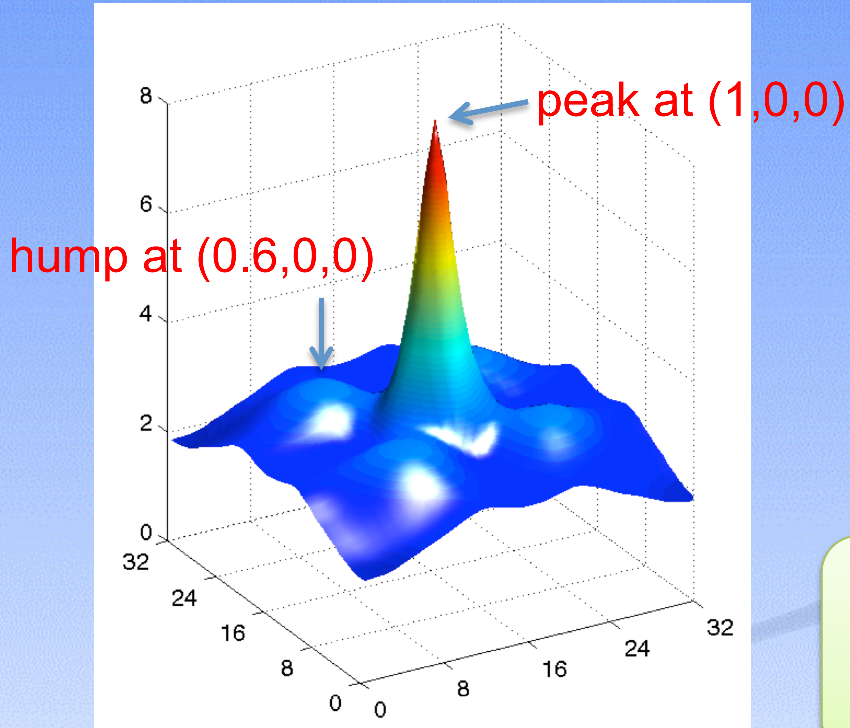
electron sheet

hole sheet

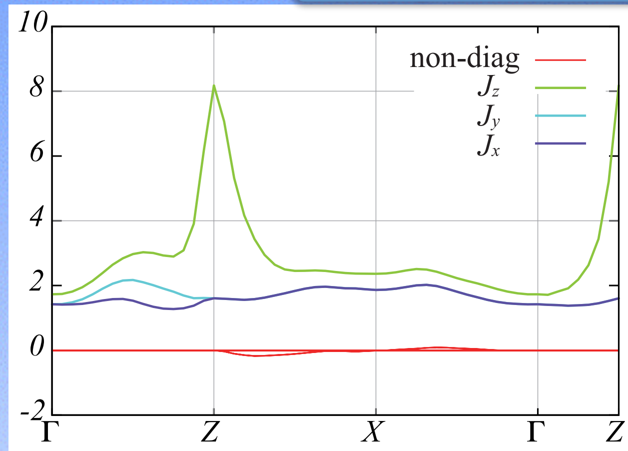
Nesting of jz=±5/2 orbitals

Z (1 0 0) and (0 0 1) : equivalent points in the body-centered tetragonal structures.

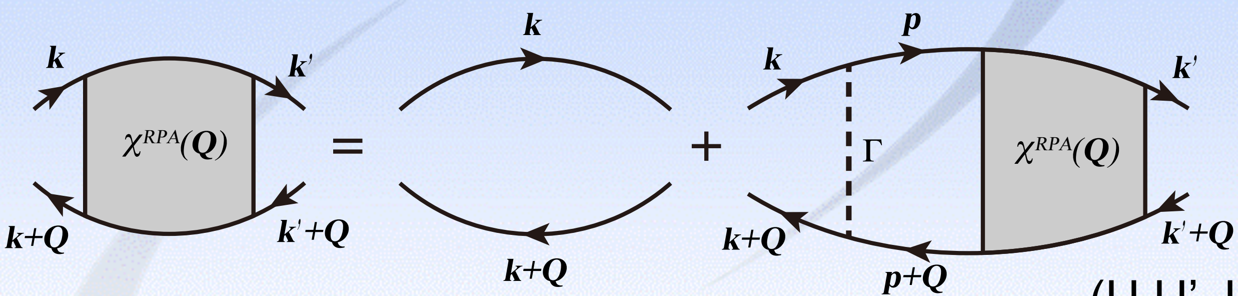
Magnetic correlations



Ising anisotropy



Peak at (1 0 0) and hump at (0.6 0 0) in $\chi_{zz}(Q)$ can be explained by the nesting property.



(U,U',J,J')=(0.5,0.5,0,0)

Possible order parameters

$$\hat{Q} = f^{\dagger}_{i\ell} Q_{\ell m} f_{im}$$

m=5/2,3/2,1/2,-1/2,-3/2,-5/2

Multipole degrees of freedom

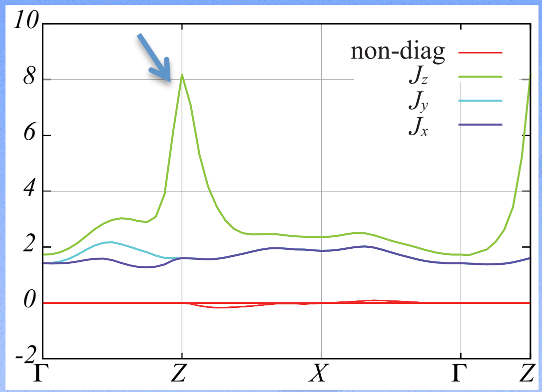
6x6=36 components

Group theory → 36=1+3+5+7+9+11 (rank 0 – 5)

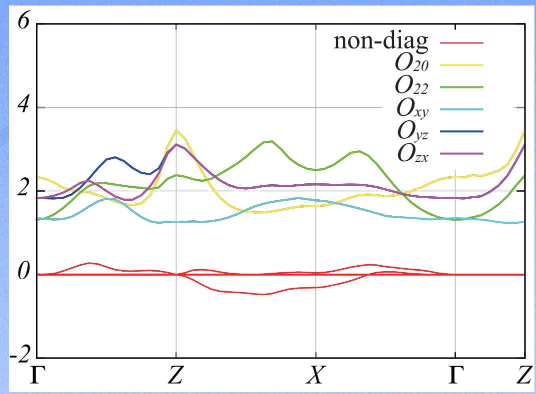
Jx	---	dipole (rank 1)
JxJy	---	quadrupole (rank 2)
JxJyJz	---	octupole (rank 3)

Multipolar correlations

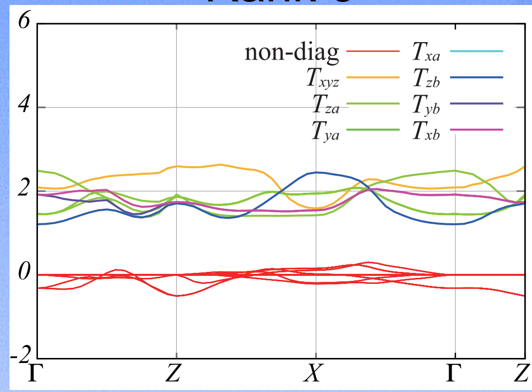
Rank 1



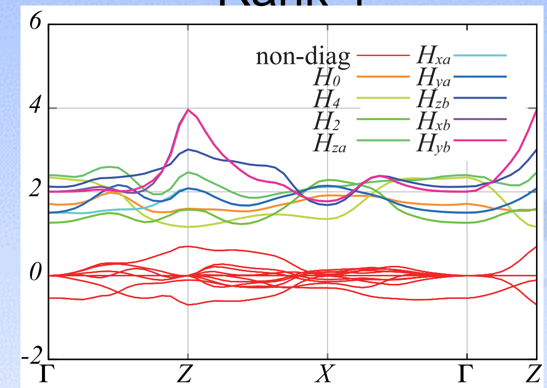
Rank 2



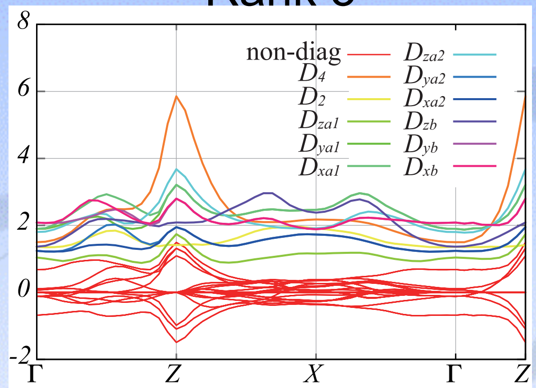
Rank 3



Rank 4

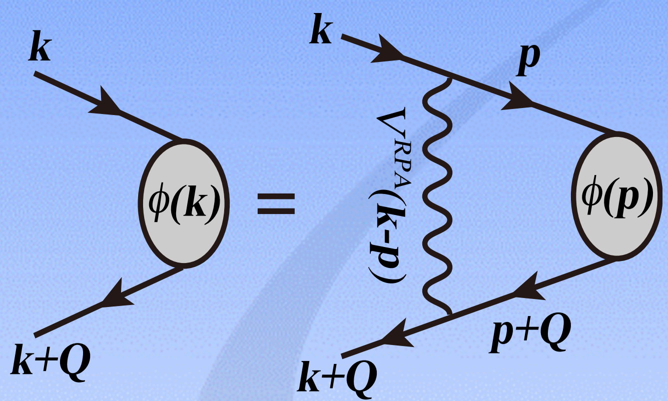


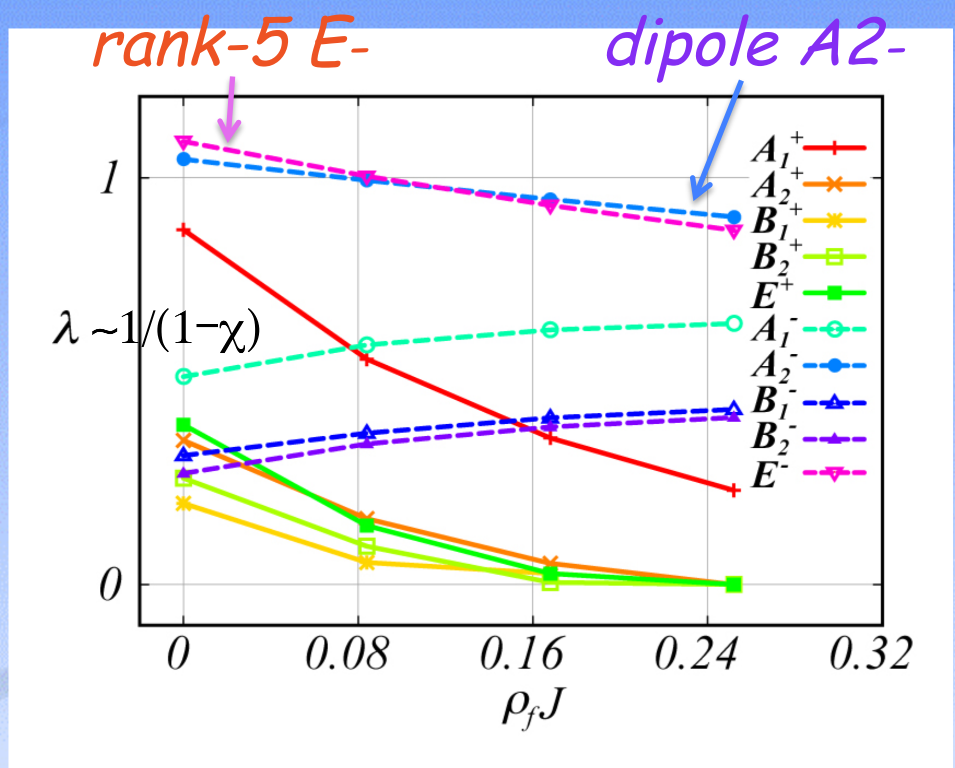
Rank 5



- Peak-hump structure at Q_C and Q_{IC}
- Some peaks except for Rank 1 correspond to candidates for the HO parameter

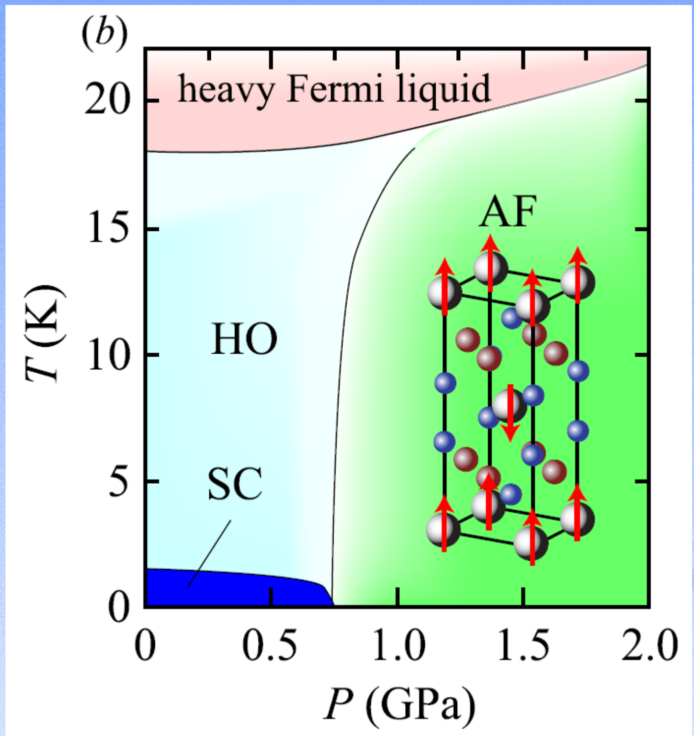
Beyond RPA

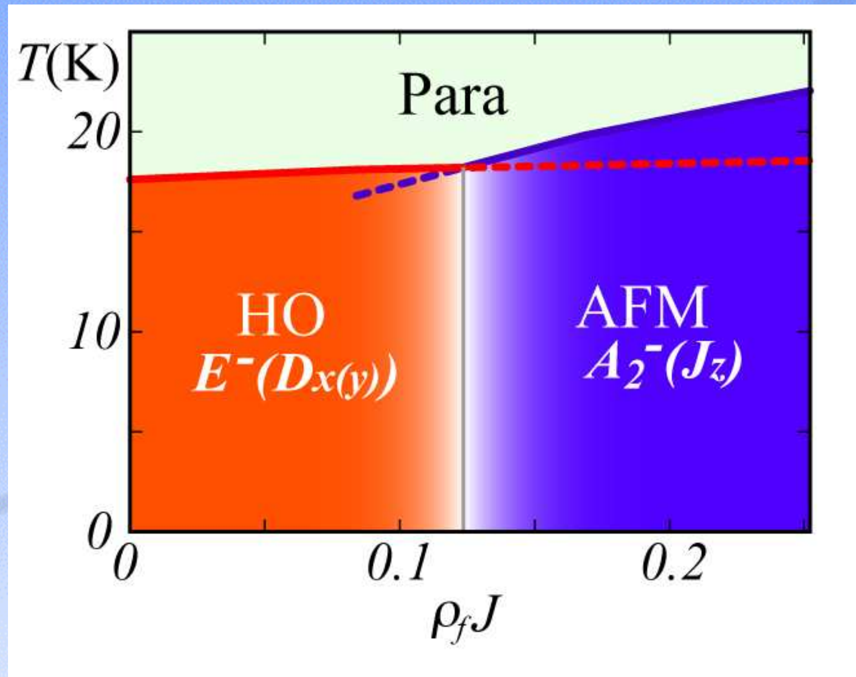




Staggered electron-hole pairing mediated by the RPA multipole fluctuations
→ E- and A2- states are ***nearly degenerate***.

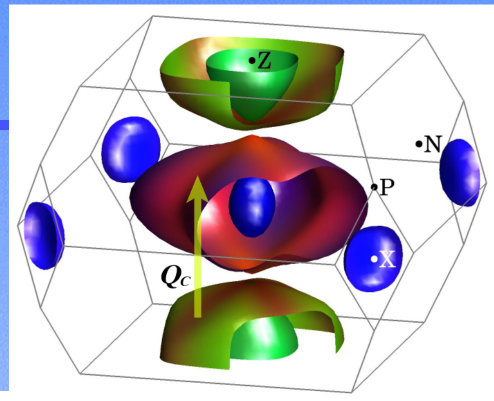
Phase Diagrams





The hidden order parameter is the ***rank-5 E-***, which is compatible with the ***nematicity***.

What is Rank-5 E^- ?



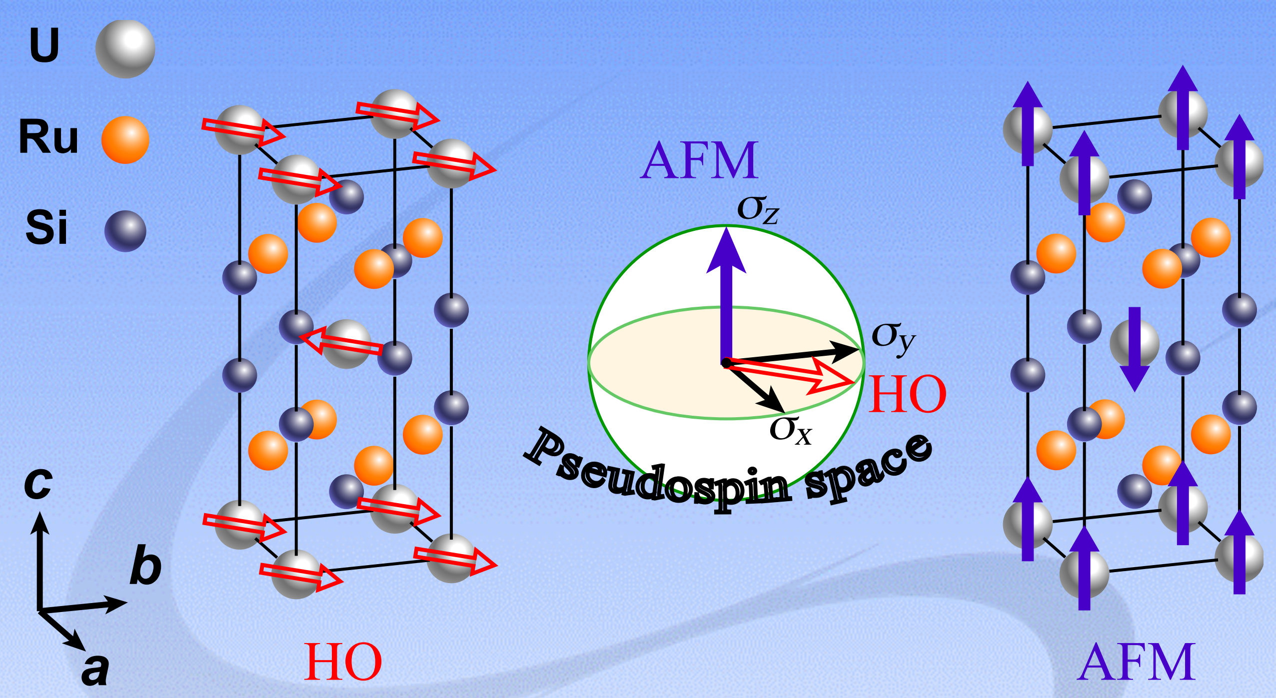
Crucially impotant is the nesting of ±5/2 components !
Consider ±5/2 as pseudospins (↑ , ↓)

$$D_x = \begin{pmatrix} & 0.02 & & 0.11 & & 0.65 \\ 0.02 & & -0.08 & & -0.08 & \\ & -0.08 & & 0.11 & & 0.11 \\ 0.11 & & 0.11 & & -0.08 & \\ & -0.18 & & -0.08 & & 0.02 \\ 0.65 & & 0.11 & & 0.02 & \end{pmatrix} \qquad J_z = \begin{pmatrix} \frac{5}{2} & & & & & \\ & \frac{3}{2} & & & & \\ & & \frac{1}{2} & & & \\ & & & -\frac{1}{2} & & \\ & & & & -\frac{3}{2} & \\ & & & & & -\frac{5}{2} \end{pmatrix}.$$

$$D_y = i\begin{pmatrix} & -0.02 & & 0.11 & & -0.65 \\ 0.02 & & 0.08 & & -0.08 & \\ & -0.08 & & -0.11 & & 0.11 \\ -0.11 & & 0.11 & & 0.08 & \\ & 0.18 & & -0.08 & & -0.02 \\ 0.65 & & -0.11 & & 0.02 & \end{pmatrix}$$

$D_x \rightarrow \sigma_x$
$D_y \rightarrow \sigma_y$
$J_z \rightarrow \sigma_z$

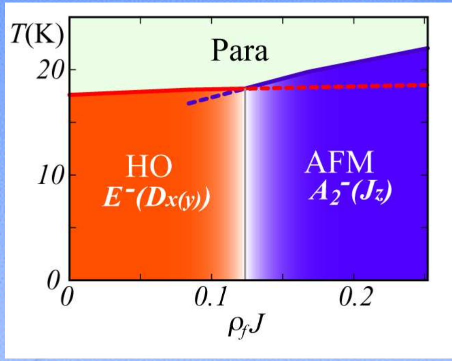
Flip Transition

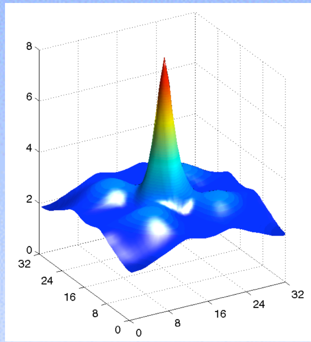


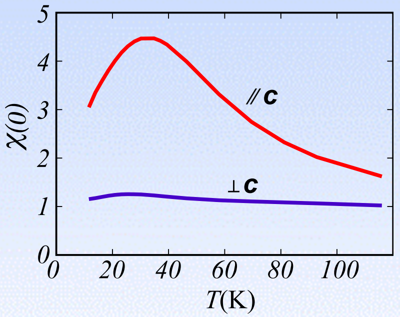
HO → AFM : *the in-plane → [001] Neel ordering in the pseudospin space*

Concluding Remarks

- Near-degenerate T_{HO} and T_N ?
  Yes!
- Ising anisotropy ?
  Yes!
- Inelastic magnetic excitations at Q_0=(1 0 0) and Q_1=(1.4 0 0) ?
  Yes!
- No evidence of drastic change in the Fermi surface ?
  Yes!
- No evidence of low-rank multipole order ?
  Yes! Rank 5
- Nematic behavior in in-plane magnetic susceptibility?
  Yes! E-





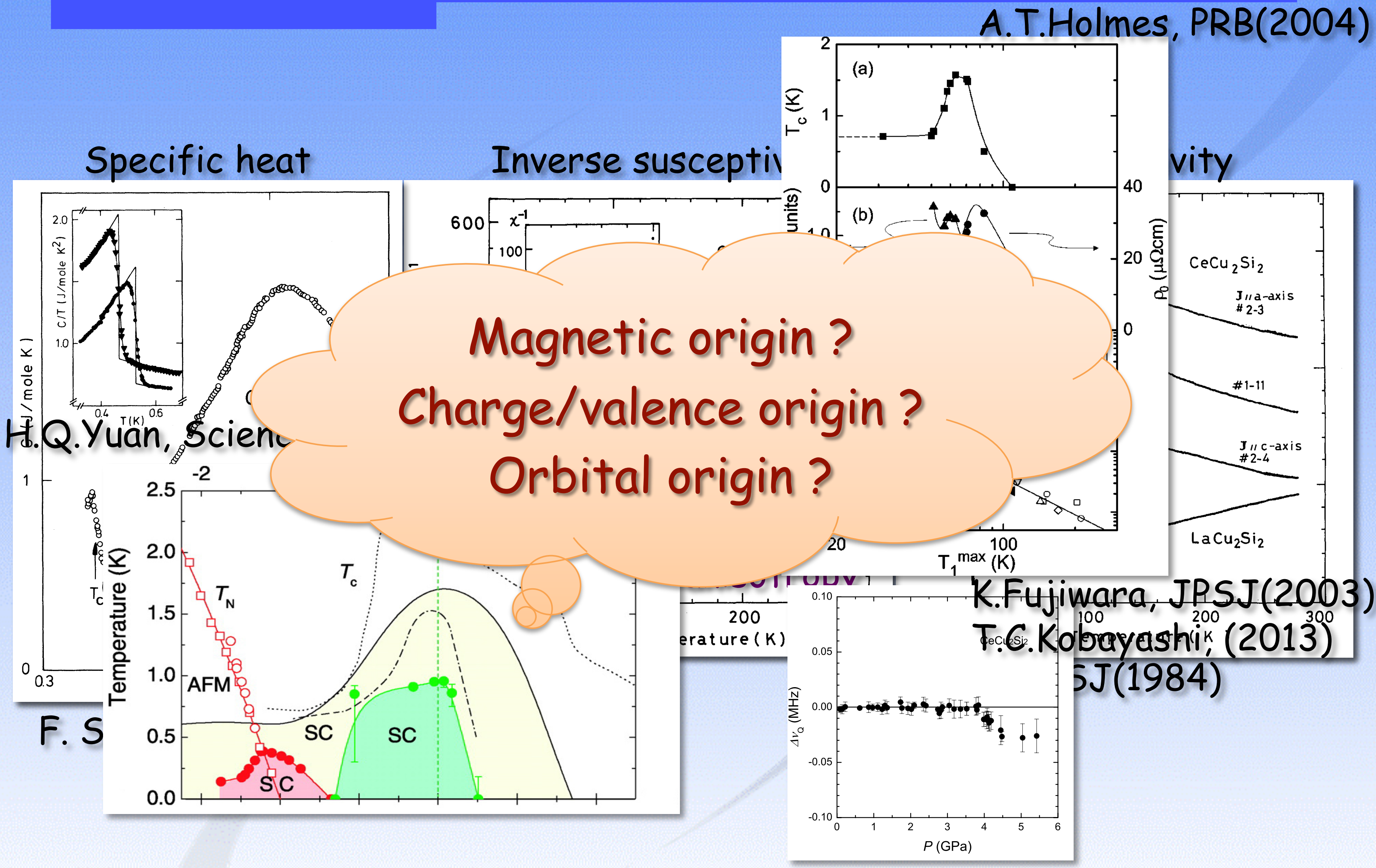


Unconventional superconductivity in $CeCu_2Si_2$

Collaborators

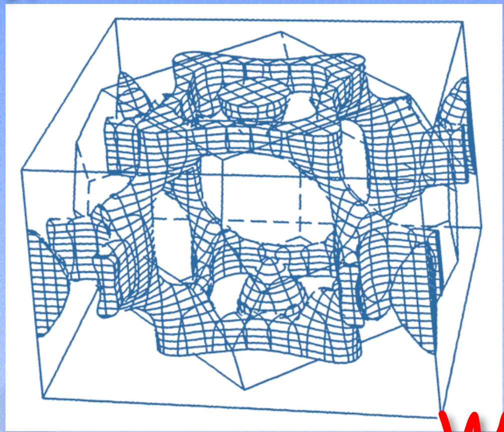
Michi-To Suzuki (CCSE JAEA)
Ryotaro Arita (University of Tokyo)

Introduction



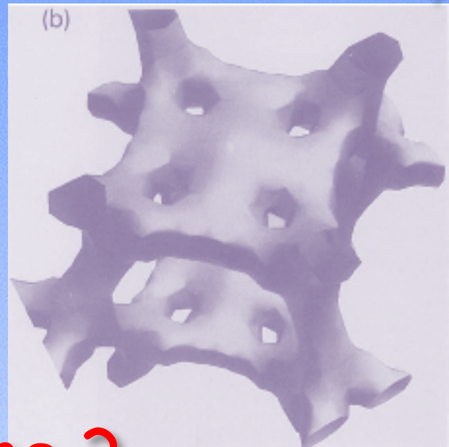
Fermi surface

H.Harima, JPSJ(1991)

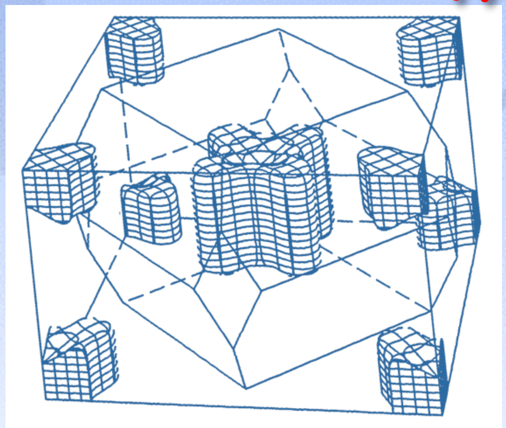


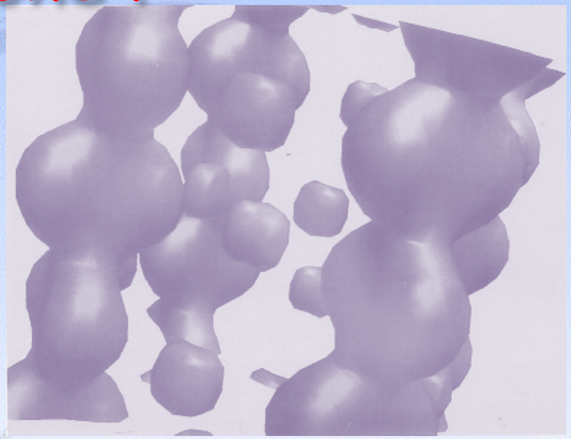
v.s.

G.Zwicknagl, Physica B(1993)

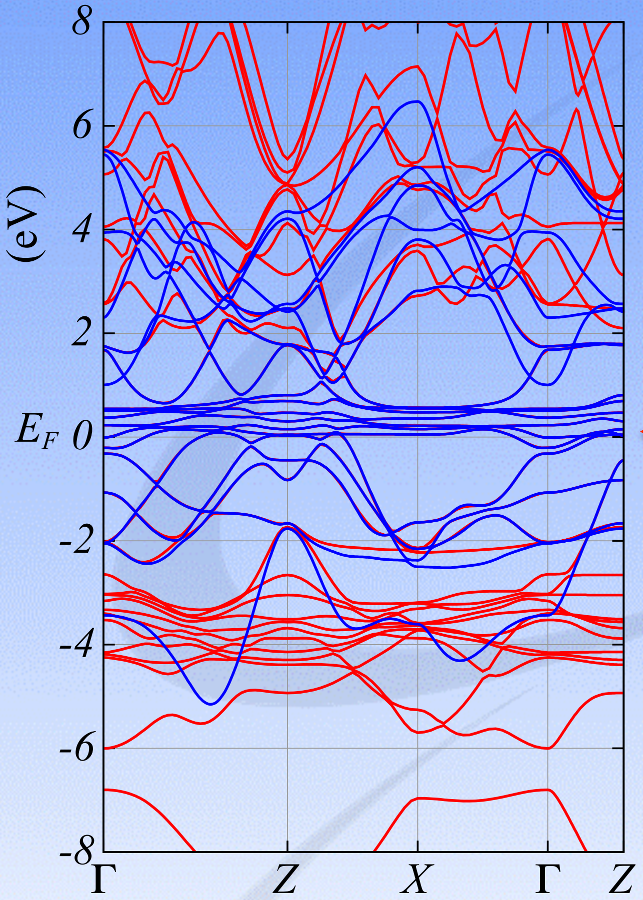


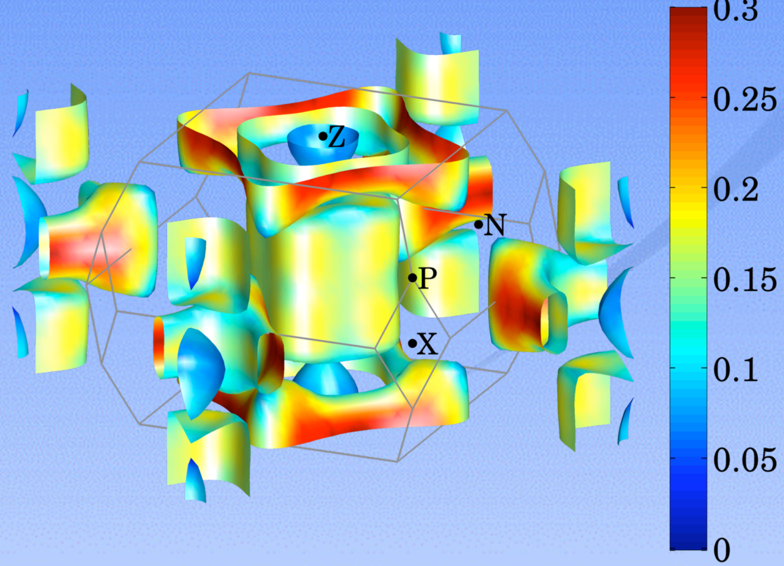
Which one ?





Construction of itinerant model

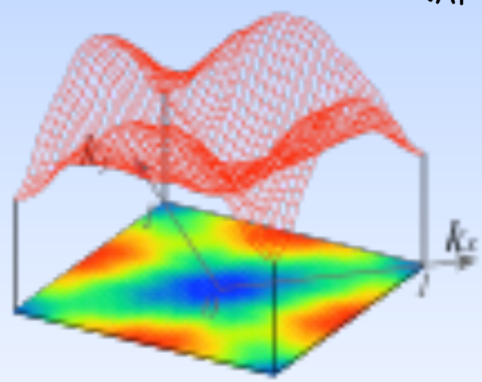




$H = H_0 + H'$

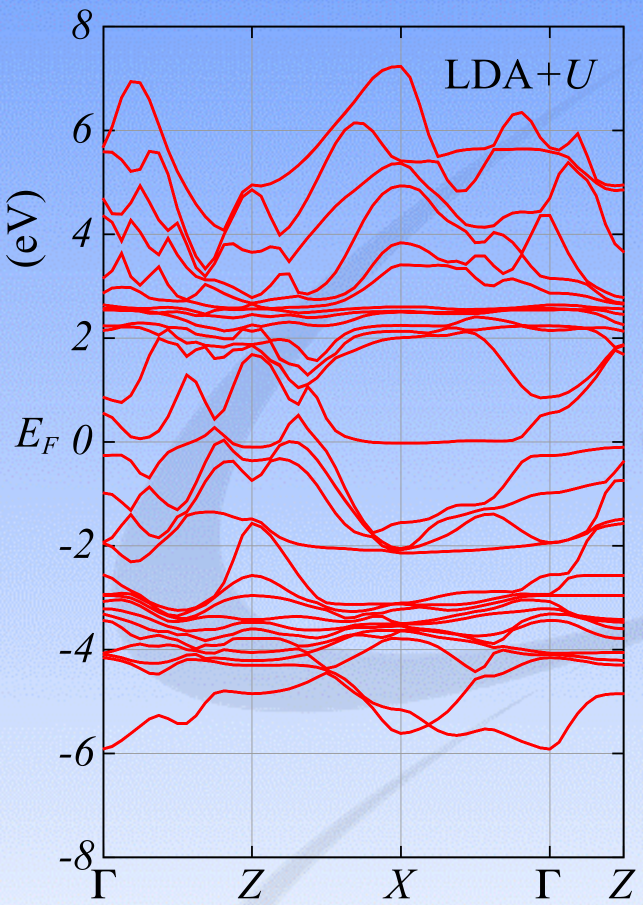
H is 36 band Anderson lattice model including spin-orbit coupling. (Ce 4f, Ce 5d, Si 3p)

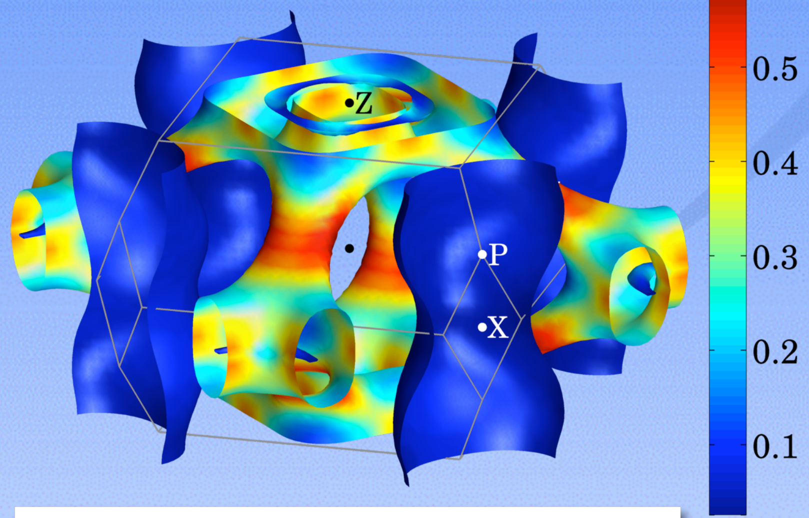
Lack of the observed Q_{AF}

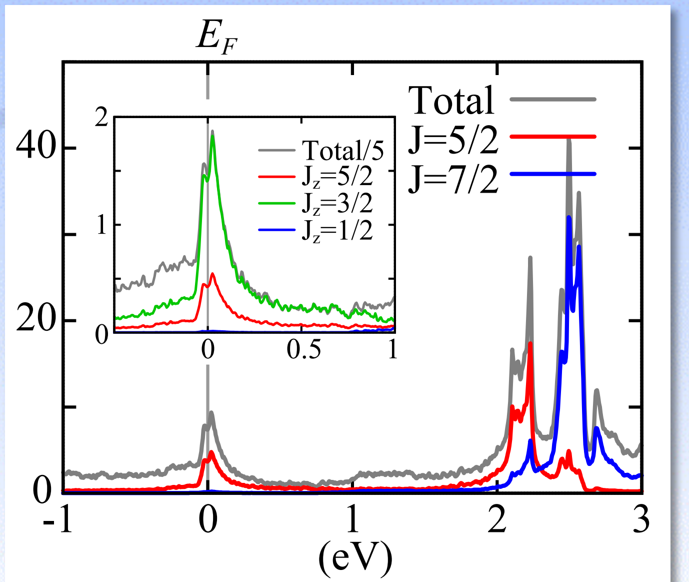


WIEN2k+Wien2Wannier+Wannier90

LDA+U





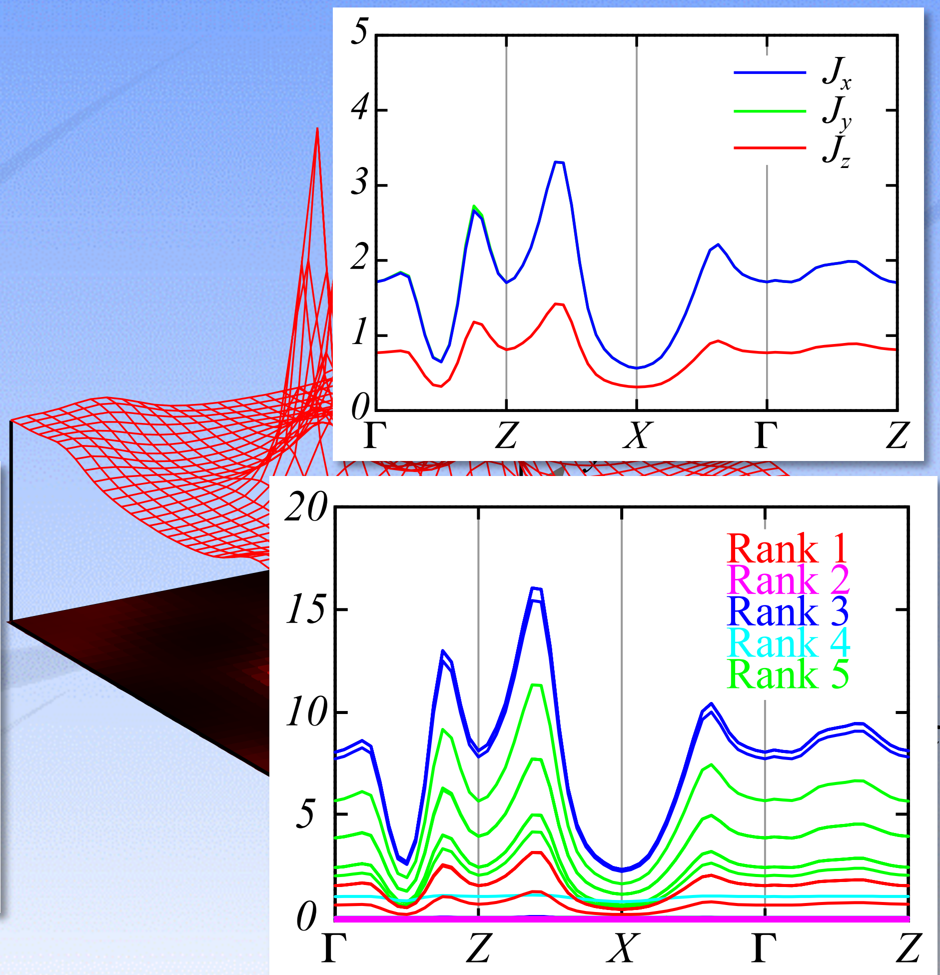


Dominant $J_z=\pm 3/2$

RPA analysis for LDA+U

Weak magnetic anisotropy

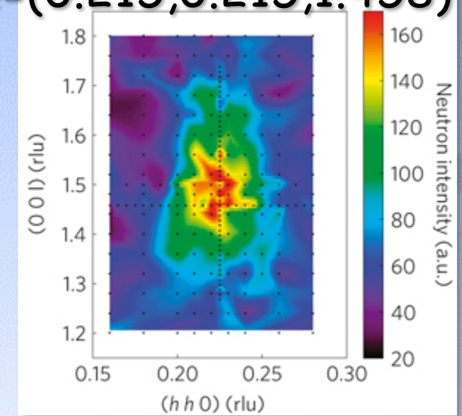
U=U'=0.3
J=J'=0



U=U'=0.23
J=J'=0

Dominant
Octupole
fluctuations

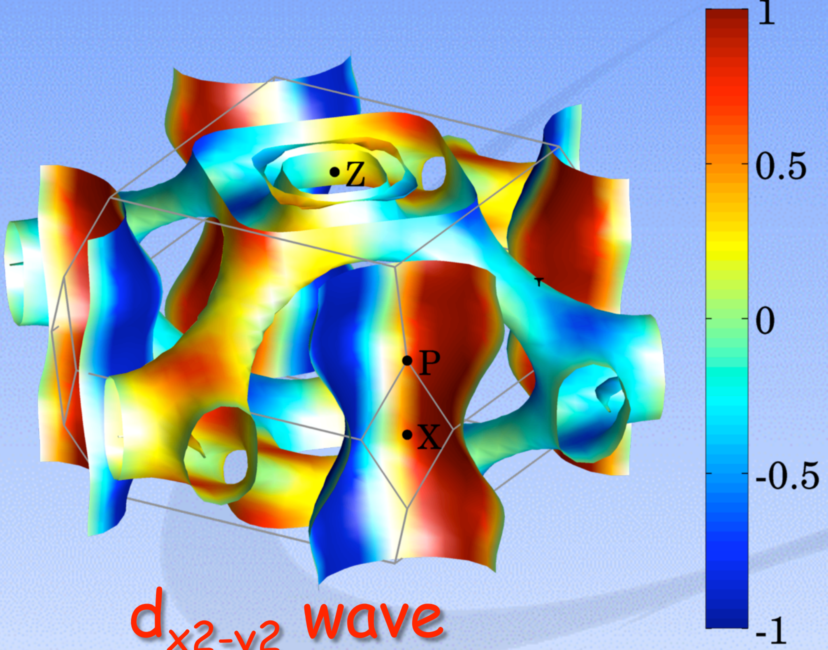
Q_{AF}=(0.215,0.215,1.458)



O.Stockert,
Nature Physics(2010)

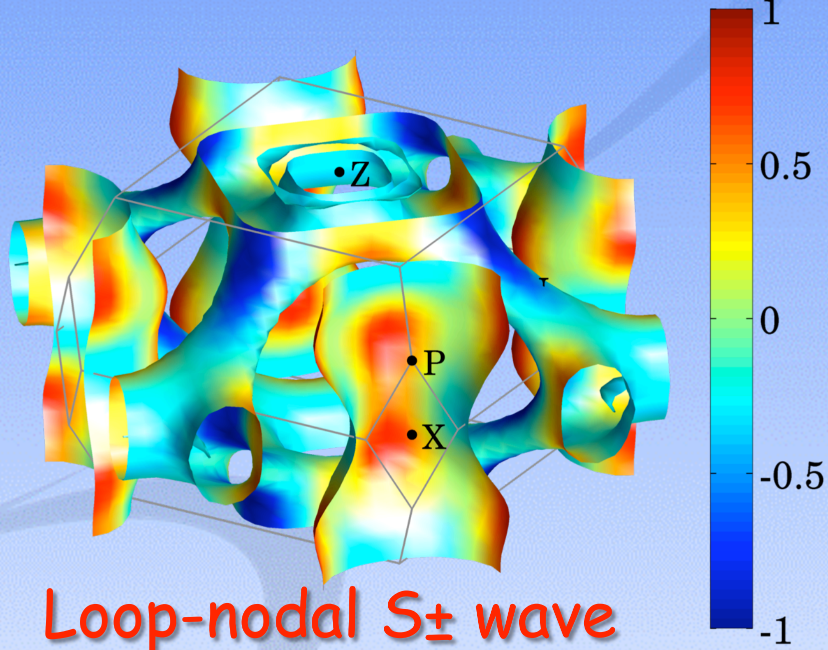
Unconventional Superconductivity

mediated by octupole fluctuations



d_{x2-y2} wave

λ=1.197



Loop-nodal S± wave

λ=1.012

2nd order for U=U'=1.0, J=J'=0

λ=0.787

λ=0.949

Conclusions

- The Fermi surface in A-type materials is consistent with that in LDA+U or the renormalized band by Zwicknagl
- Incommensurate spin fluctuations can be explained by the nesting between the heavy-electron sheets
- The dominant octupole fluctuations can drive d_{x2-y2}-wave or loop-nodal s-wave superconductivity

Multipoles in J=5/2 space

rank	O	D_4	basis(O)
k=1	T_1^-	A_2^-	$J_0^{(1)}=J^z$
		E^-	$\tilde{J}_{[1]+}^{(1)}=J^y$
			$\tilde{J}_{[1]-}^{(1)}=J^x$
k=2	E^+	A_1^+	$J_0^{(2)}=O_2^0$
		B_1^+	$\tilde{J}_{[2]+}^{(2)}=O_2^2$
	T_2^+	B_2^+	$\tilde{J}_{[2]-}^{(2)}=O_{xy}$
		E^+	$\tilde{J}_{[1]+}^{(2)}=O_{yz}$
			$\tilde{J}_{[1]-}^{(2)}=O_{zx}$
k=3	A_2^-	B_1^-	$\tilde{J}_{[2]-}^{(3)}=T_{xyz}$
	T_1^-	A_2^-	$J_0^{(3)}=T_z^\alpha$
		aE^-	$-\frac{1}{2\sqrt{2}}(\sqrt{5}\tilde{J}_{[3]+}^{(3)}+\sqrt{3}\tilde{J}_{[1]+}^{(3)})=T_y^\alpha$
			$\frac{1}{2\sqrt{2}}(\sqrt{5}\tilde{J}_{[3]-}^{(3)}-\sqrt{3}\tilde{J}_{[1]-}^{(3)})=T_x^\alpha$
	T_2^-	B_2^-	$\tilde{J}_{[2]+}^{(3)}=T_z^\beta$
		bE^-	$-\frac{1}{2\sqrt{2}}(\sqrt{3}\tilde{J}_{[3]+}^{(3)}-\sqrt{5}\tilde{J}_{[1]+}^{(3)})=T_y^\beta$
			$-\frac{1}{2\sqrt{2}}(\sqrt{3}\tilde{J}_{[3]-}^{(3)}+\sqrt{5}\tilde{J}_{[1]-}^{(3)})=T_x^\beta$
k=4	A_1^+	aA_1^+	$\frac{1}{2\sqrt{3}}(\sqrt{7}J_0^{(4)}+\sqrt{5}\tilde{J}_{[4]+}^{(4)})=H_0$
	E^+	bA_1^+	$-\frac{1}{2\sqrt{3}}(-\sqrt{5}J_0^{(4)}+\sqrt{7}\tilde{J}_{[4]+}^{(4)})=H_4$
		B_1^+	$-\tilde{J}_{[2]+}^{(4)}=H_2$
	T_1^+	A_2^+	$\tilde{J}_{[4]-}^{(4)}=H_z^\alpha$
		aE^+	$-\frac{1}{2\sqrt{2}}(\tilde{J}_{[3]+}^{(4)}+\sqrt{7}\tilde{J}_{[1]+}^{(4)})=H_x^\alpha$
			$-\frac{1}{2\sqrt{2}}(\tilde{J}_{[3]-}^{(4)}-\sqrt{7}\tilde{J}_{[1]-}^{(4)})=H_y^\alpha$
	T_2^+	B_2^+	$\tilde{J}_{[2]-}^{(4)}=H_z^\beta$
		bE^+	$\frac{1}{2\sqrt{2}}(\sqrt{7}\tilde{J}_{[3]+}^{(4)}-\tilde{J}_{[1]+}^{(4)})=H_x^\beta$
			$-\frac{1}{2\sqrt{2}}(\sqrt{7}\tilde{J}_{[3]-}^{(4)}+\tilde{J}_{[1]-}^{(4)})=H_y^\beta$
k=5	E^-	A_1^-	$\tilde{J}_{[4]-}^{(5)}=D_4$
		B_1^-	$-\tilde{J}_{[2]-}^{(5)}=D_2$
	aT_1^-	aA_2^-	$J_0^{(5)}=D_z^{\alpha 1}$
		aE^-	$\frac{1}{8\sqrt{2}}(3\sqrt{7}\tilde{J}_{[5]+}^{(5)}+\sqrt{35}\tilde{J}_{[3]+}^{(5)}+\sqrt{30}\tilde{J}_{[1]+}^{(5)})=D_y^{\alpha 1}$
			$\frac{1}{8\sqrt{2}}(3\sqrt{7}\tilde{J}_{[5]-}^{(5)}-\sqrt{35}\tilde{J}_{[3]-}^{(5)}+\sqrt{30}\tilde{J}_{[1]-}^{(5)})=D_x^{\alpha 1}$
	bT_1^-	bA_2^-	$\tilde{J}_{[4]+}^{(5)}=D_z^{\alpha 2}$
		bE^-	$\frac{1}{16}(\sqrt{10}\tilde{J}_{[5]+}^{(5)}-9\sqrt{2}\tilde{J}_{[3]+}^{(5)}+2\sqrt{21}\tilde{J}_{[1]+}^{(5)})=D_y^{\alpha 2}$
			$\frac{1}{16}(\sqrt{10}\tilde{J}_{[5]-}^{(5)}+9\sqrt{2}\tilde{J}_{[3]-}^{(5)}+2\sqrt{21}\tilde{J}_{[1]-}^{(5)})=D_x^{\alpha 2}$
	T_2^-	B_2^-	$\tilde{J}_{[2]+}^{(5)}=D_z^\beta$
		cE^-	$\frac{1}{4\sqrt{2}}(\sqrt{15}\tilde{J}_{[5]+}^{(5)}-\sqrt{3}\tilde{J}_{[3]+}^{(5)}-\sqrt{14}\tilde{J}_{[1]+}^{(5)})=D_y^\beta$
			$-\frac{1}{4\sqrt{2}}(\sqrt{15}\tilde{J}_{[5]-}^{(5)}+\sqrt{3}\tilde{J}_{[3]-}^{(5)}-\sqrt{14}\tilde{J}_{[1]-}^{(5)})=D_x^\beta$

$$\hat{Q}=f_{i\ell}^{\dagger}Q_{\ell m}f_{im}$$

36 multipoles up to rank 5.