The 2nd Asia Pacific Workshop on Quantum Magnetism

Multipolar Order and Superconductivity in $Pr(TM)_2(Al,Zn)_{20}$ Kondo Materials

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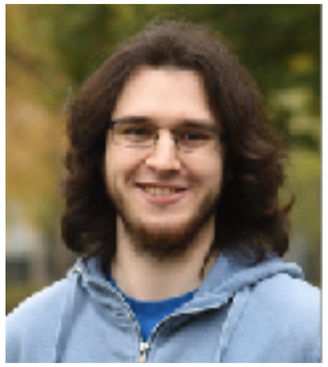

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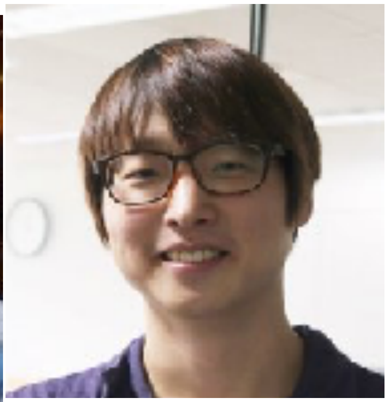

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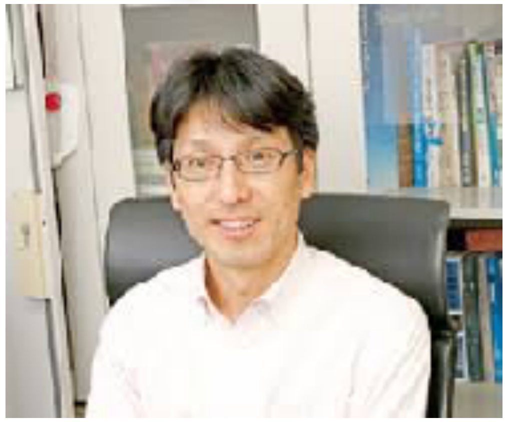

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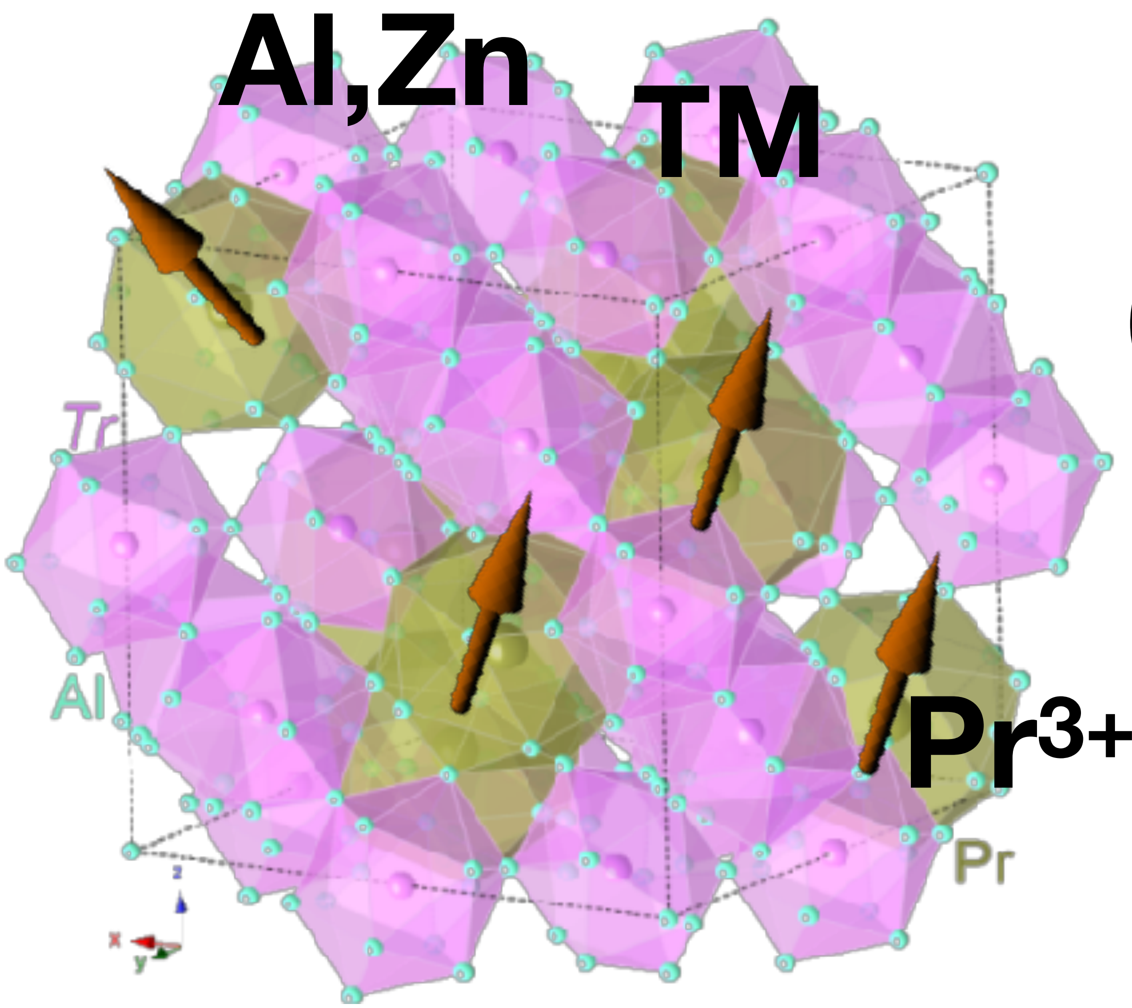
Introduction - $Pr(TM)_2(Al,Zn)_{20}$ Materials

$Pr(TM)_2(Al,Zn)_{20}$

All interesting phenomena coexist!



Pr^{3+} **Quadrupole-Octupolar ordering**

(TM)+ Al,Zn **Itinerant electrons**
Fermi pockets at k=0

Kondo coupling

Pr^{3+} + (TM)+ Al,Zn

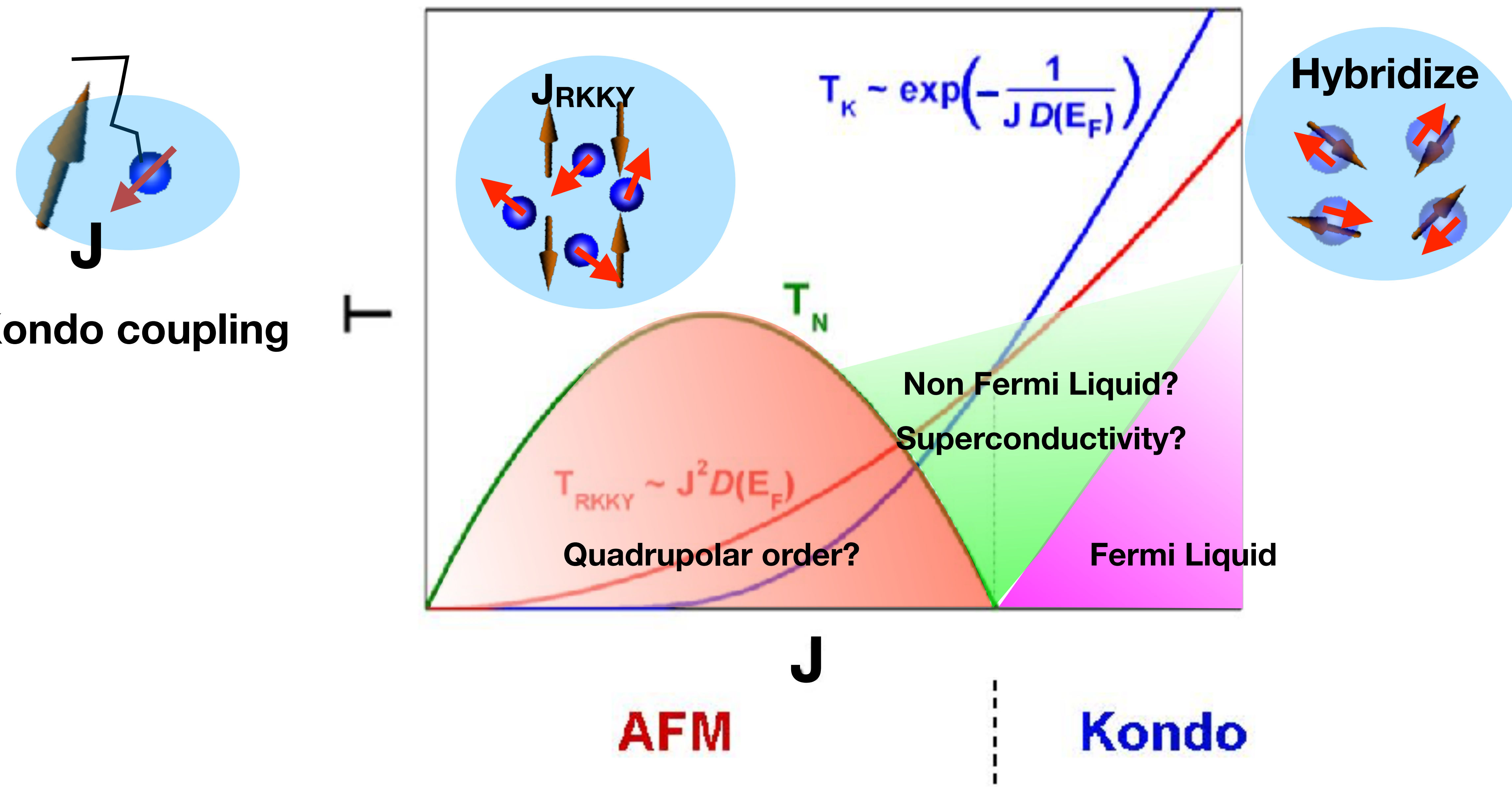
superconductivity

Q) How can we understand them?

Introduction — Doniach Phase Diagram

Doniach phase diagram

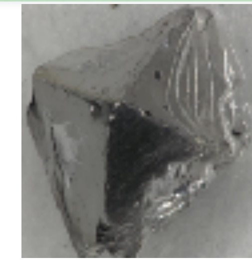
S. Doniach, Physica B **91**, 231 (1977)

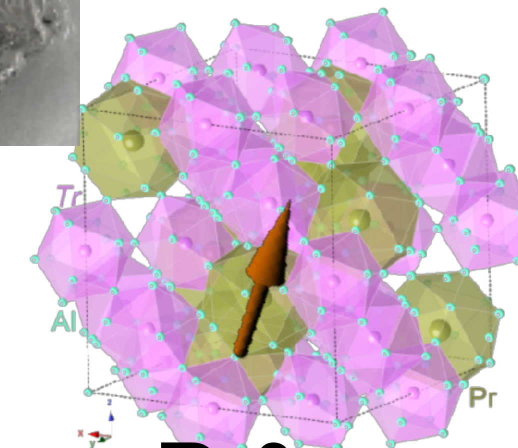


Q) Search for new Doniach phase diagram with multipolar order ?

Multipolar order in $Pr(TM)_2(Al,Zn)_{20}$

Pr^{3+} $4f^2$ non Kramers Γ_3 doublets





Pr^{3+}

Spin orbit coupling
+ Crystalline Electric Field

$$|\Gamma_1\rangle = \frac{1}{2}\sqrt{\frac{5}{6}}|+4\rangle + \frac{1}{2}\sqrt{\frac{7}{3}}|0\rangle + \frac{1}{2}\sqrt{\frac{5}{6}}|-4\rangle$$

$$|\Gamma_{5\pm}^{(2)}\rangle = \frac{1}{2}\sqrt{\frac{7}{2}}|\pm 3\rangle - \frac{1}{2}\sqrt{\frac{1}{2}}|\mp 1\rangle$$

$$|\Gamma_5^{(2)}\rangle = \sqrt{\frac{1}{2}}|+2\rangle - \sqrt{\frac{1}{2}}|-2\rangle$$

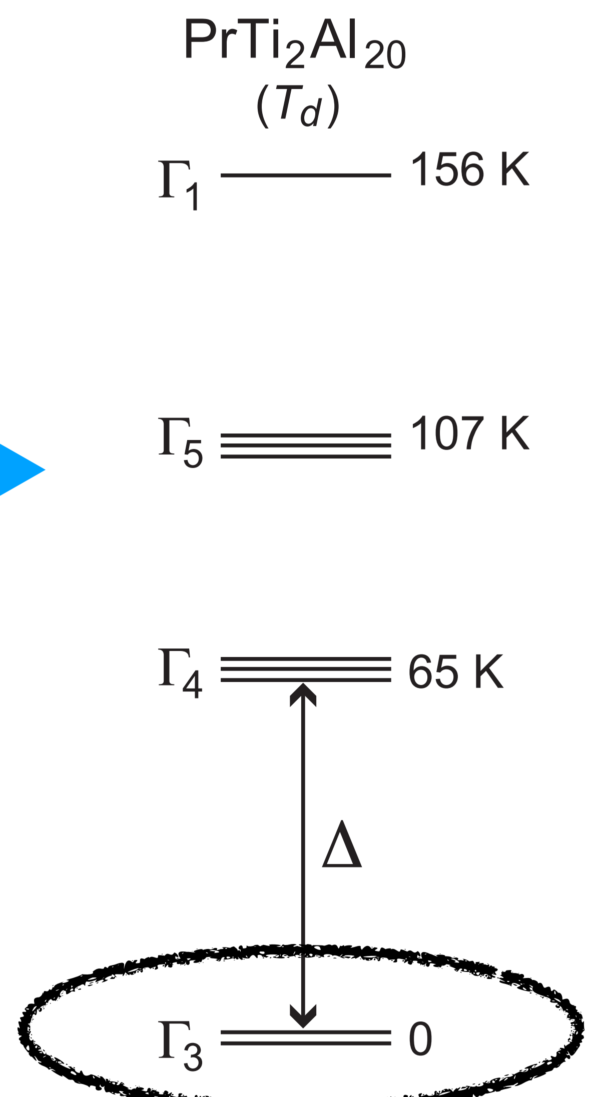
$$|\Gamma_{4\pm}^{(1)}\rangle = \frac{1}{2}\sqrt{\frac{1}{2}}|\mp 3\rangle + \frac{1}{2}\sqrt{\frac{7}{2}}|\pm 1\rangle$$

$$|\Gamma_4^{(2)}\rangle = \sqrt{\frac{1}{2}}|+4\rangle - \sqrt{\frac{1}{2}}|-4\rangle$$

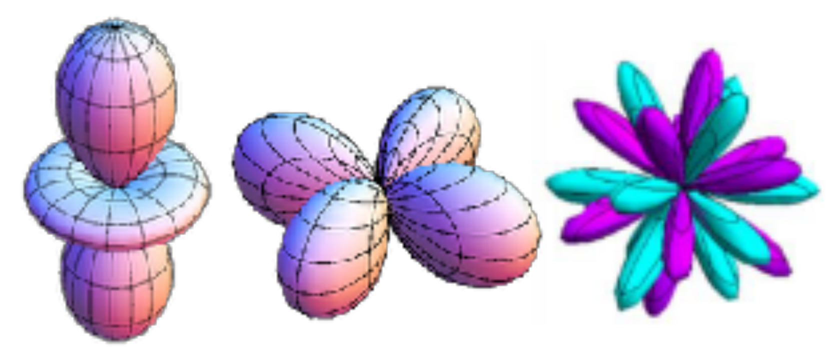
$$|\Gamma_3^{(1)}\rangle = \frac{1}{2}\sqrt{\frac{7}{6}}|+4\rangle - \frac{1}{2}\sqrt{\frac{5}{3}}|0\rangle + \frac{1}{2}\sqrt{\frac{7}{6}}|-4\rangle$$

$$|\Gamma_3^{(2)}\rangle = \sqrt{\frac{1}{2}}|+2\rangle + \sqrt{\frac{1}{2}}|-2\rangle$$

TJ Sato et al PRB 86, 184419 (2012)



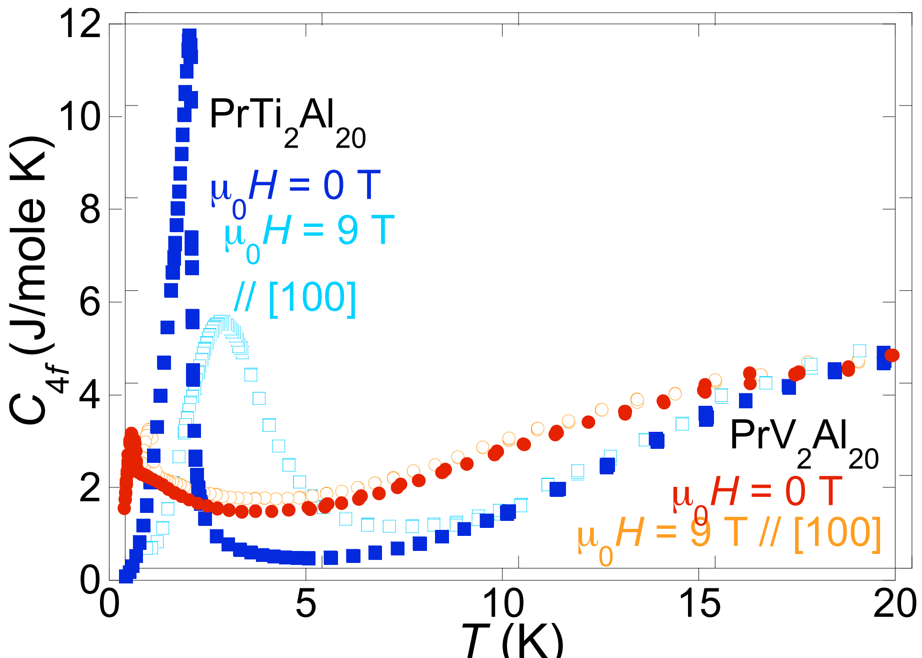
pseudospin-1/2 with
Γ_3 doublets describes



$3J_z^2 - J^2$ **$J_x^2 - J_y^2$** **$J_xJ_yJ_z$**

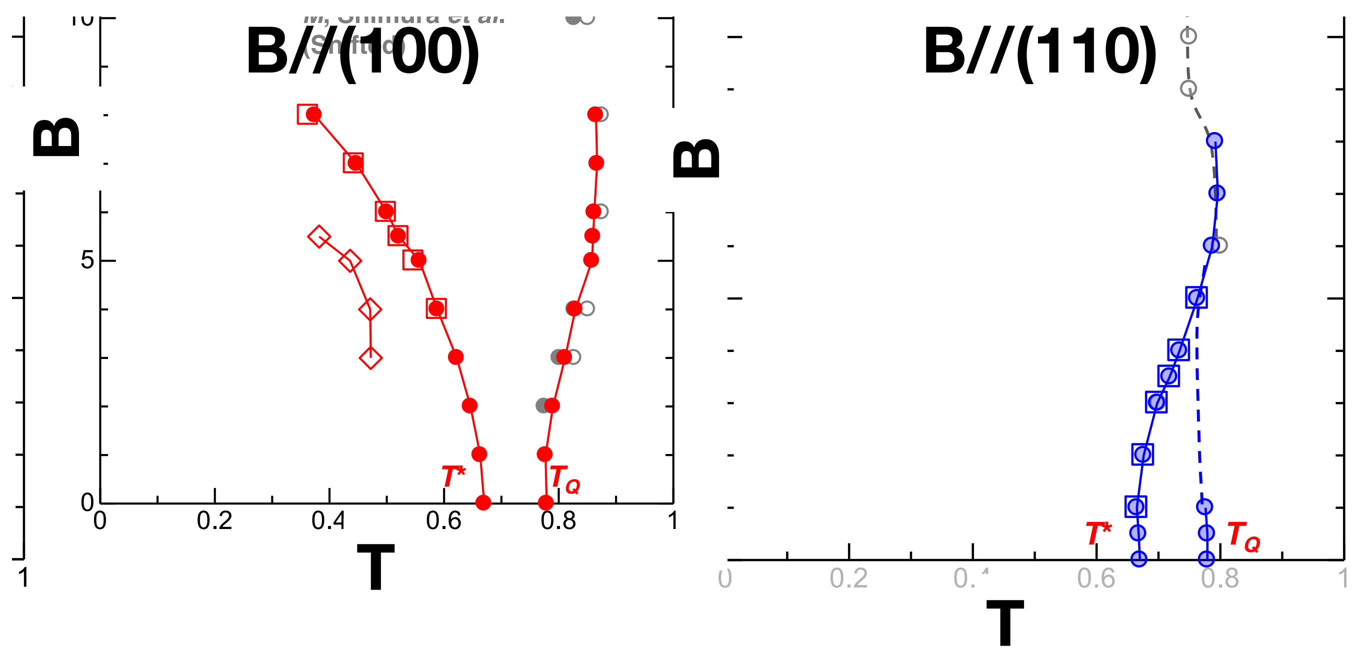
Quadrupole **Octupole**

Multipolar order and field effect

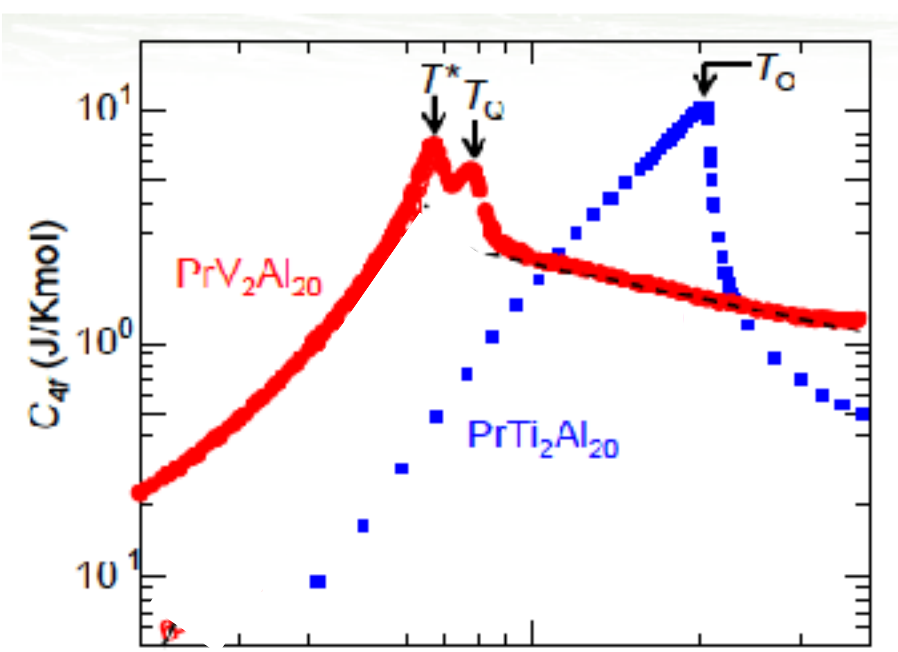


A. Sakai et al, J. Phys. Soc. Jpn . **80** (2011) 063701

Field dependence PrV_2Al_{20}



Y. Shimura et al, Phys. Rev. B 91 241102 (R) (2015)



M. Tsujimoto et al, Phys. Rev. Lett 113 267001 (2014)

$PrTi_2Al_{20}$ **Ferro-quadrupolar order**

T_Q = 2K

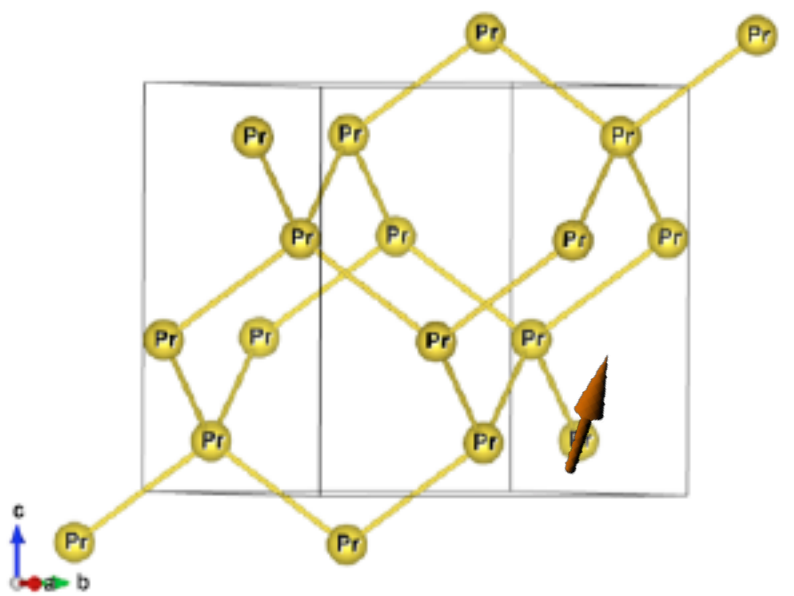
PrV_2Al_{20} **Antiferro-quadrupolar order**

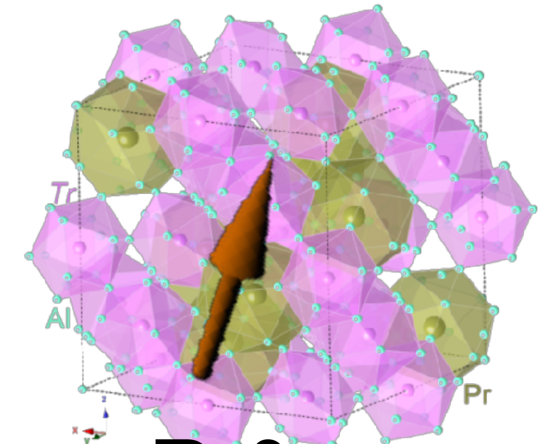
T_Q =0.75K, T*=0.65K

Anisotropic field effect

Multipolar order and finite T transitions

Pr^{3+} ions form a diamond lattice

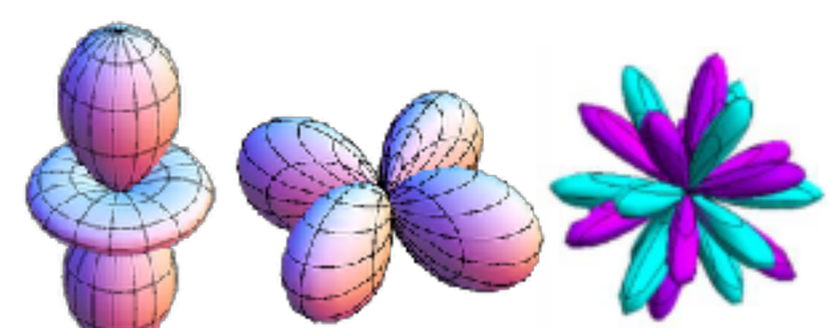




Pr^{3+}

Kondo coupling with itinerant electrons
—> multiple spin interactions

pseudospin-1/2 with Γ_3 doublets describes



$3J_z^2 - J^2$	$J_x^2 - J_y^2$	$J_xJ_yJ_z$
τ_x	τ_y	τ_z
Quadrupole		**Octupole**

Quadrupolar moments - TR even

$$H = \frac{1}{2}\sum_{i,j} J_{ij}(\vec{\tau}_i^{\perp}\cdot\vec{\tau}_j^{\perp} + \lambda\tau_i^z\tau_j^z) - K\sum_{\langle\langle ij\rangle\langle km\rangle\rangle}\vec{\tau}_i^{\perp}\cdot\vec{\tau}_j^{\perp}\tau_k^z\tau_m^z\,.$$

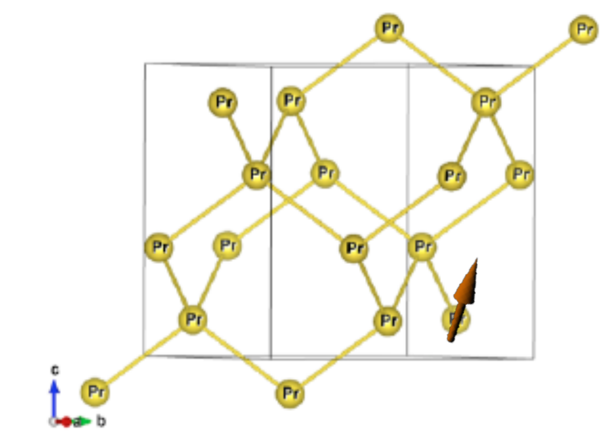
Octupolar moments -TR odd

Q) Possible phases and finite T transitions ?

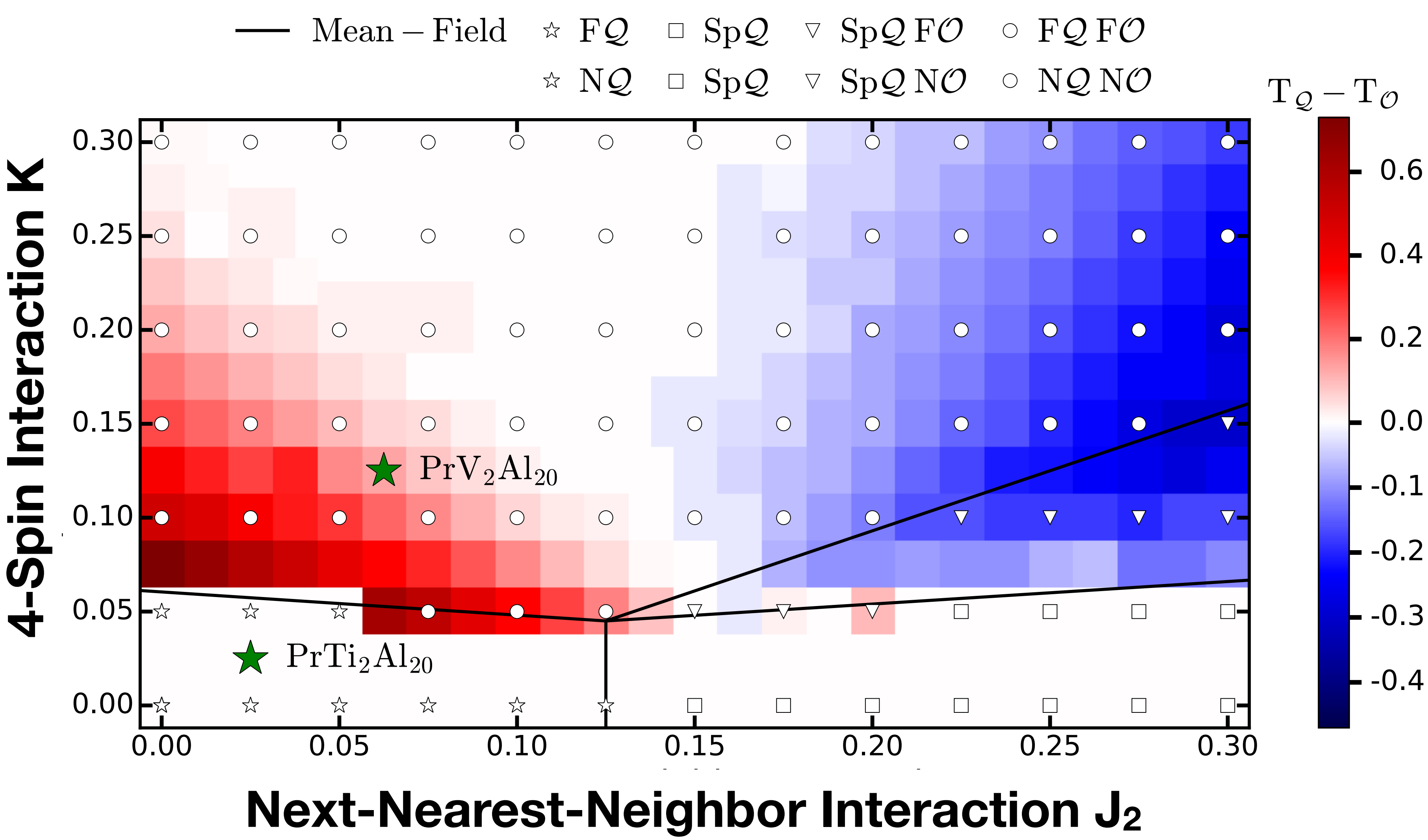
Multipolar order and finite T transitions

Quadrupolar, Octupolar orderings

$$H=\frac{1}{2}\sum_{i,j}J_{ij}(\vec{\tau}_i^{\perp}\cdot\vec{\tau}_j^{\perp}+\lambda\tau_i^z\tau_j^z)-K\sum_{\langle\langle ij\rangle\langle km\rangle\rangle}\vec{\tau}_i^{\perp}\cdot\vec{\tau}_j^{\perp}\tau_k^z\tau_m^z\,.$$



Monte-Carlo Results



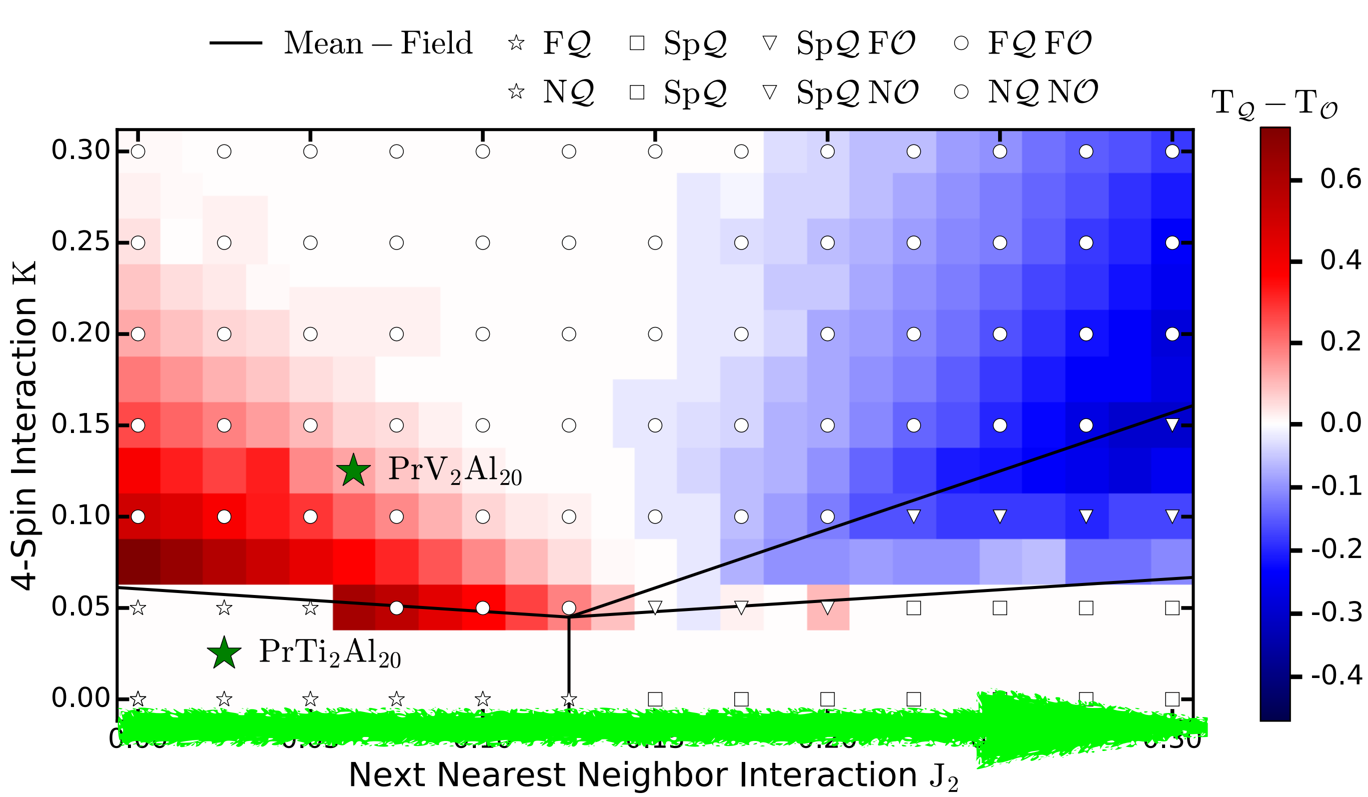
multispin interactions + frustration

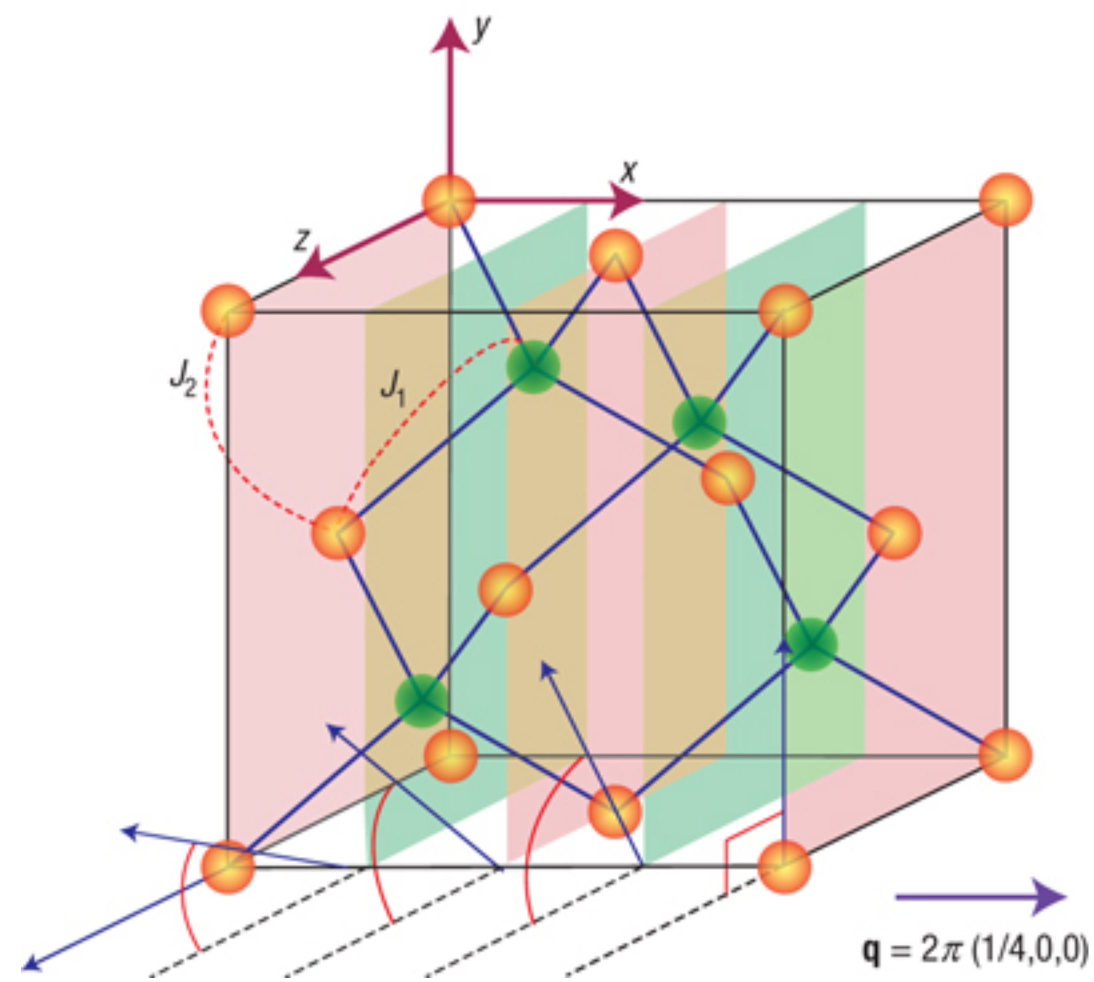
PRB 97, 115111 (2018)

Multipolar order and finite T transitions

Quadrupolar, Octupolar orderings

Phase diagram based on MC

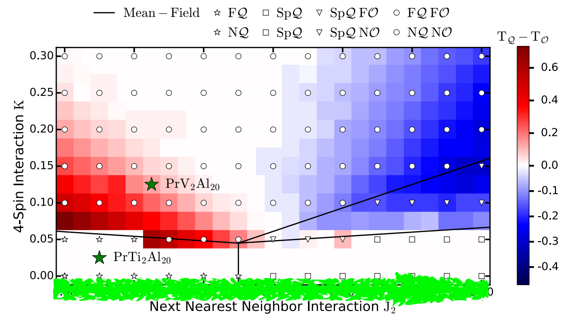




Quadrupolar order : (Anti-) Ferro → spiral order with finite-Q

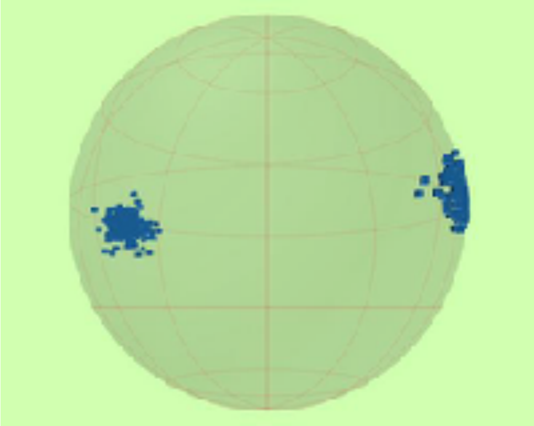
Multipolar order and finite T transitions

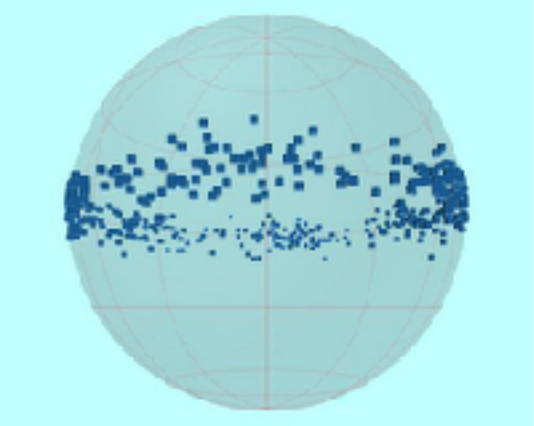
Quadrupolar, Octupolar orderings

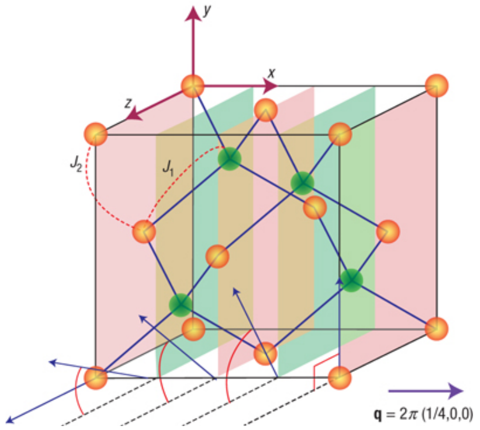


Quadrupolar order : (Anti-) Ferro → **spiral order with finite-Q**

Common origin plots of τ







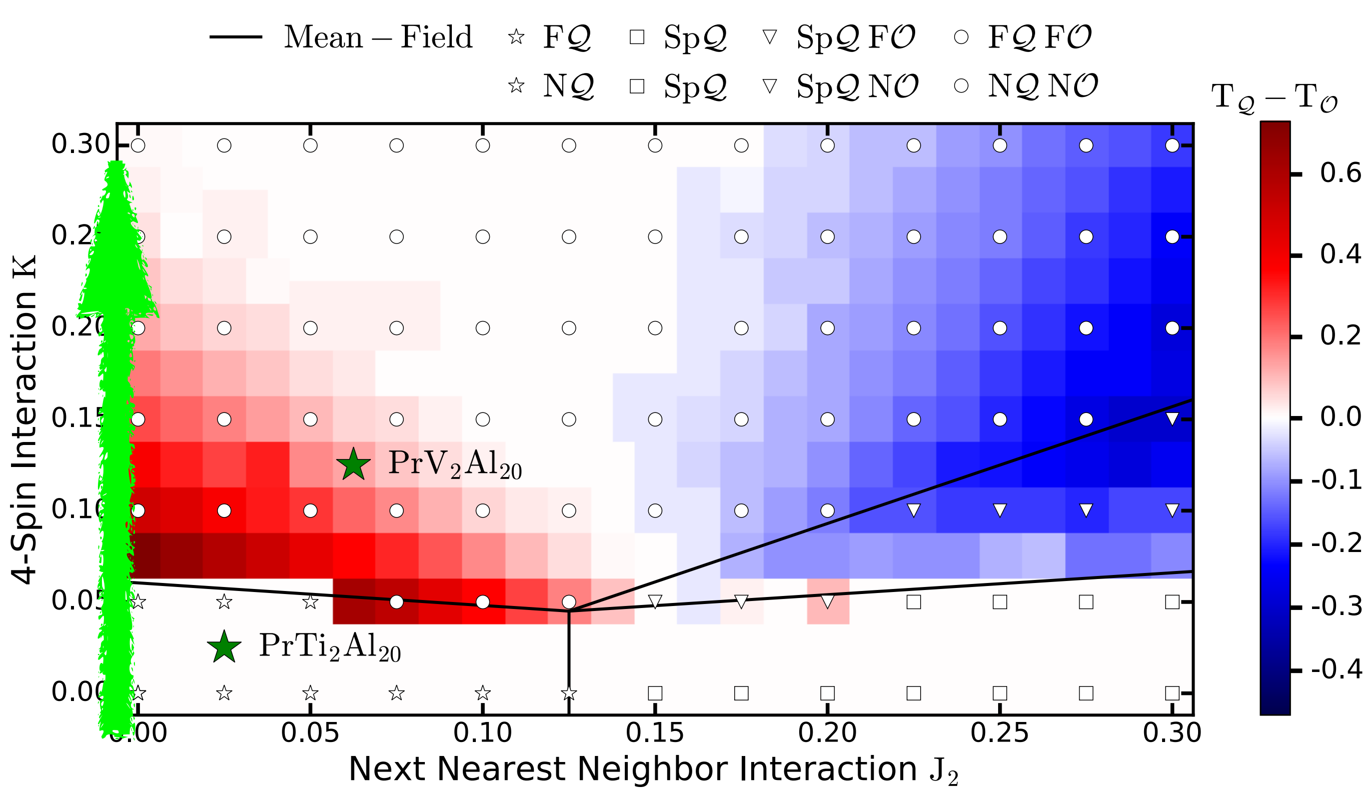
J_2

with increasing next-nearest neighbor J_2

Multipolar order and finite T transitions

Quadrupolar, Octupolar orderings

Phase diagram based on MC



$T_Q > T_O$

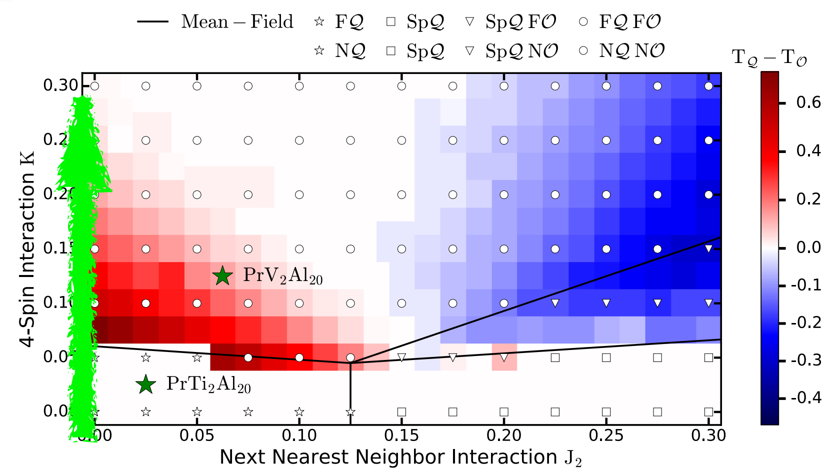
$T_Q < T_O$

Pure (anti-) ferro quadrupolar order

quadrupolar + octupolar coexisting order with $T_Q > T_O$

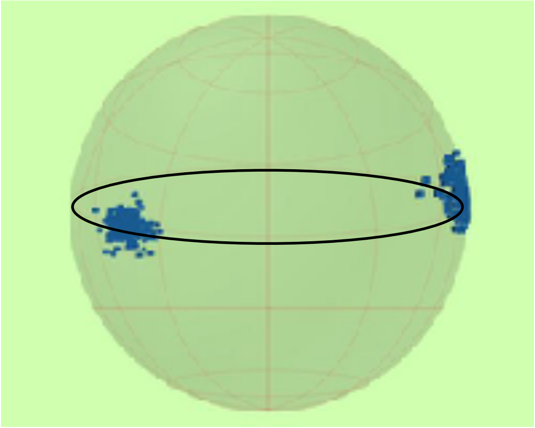
Multipolar order and finite T transitions

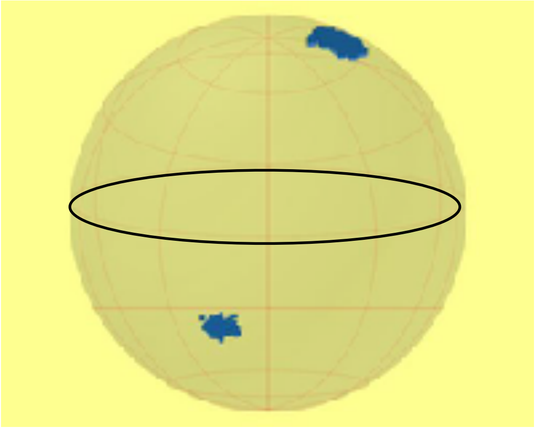
Quadrupolar, Octupolar orderings



pure quadrupolar order → **quadrupolar + octupolar coexisting order**

Common origin plots of τ





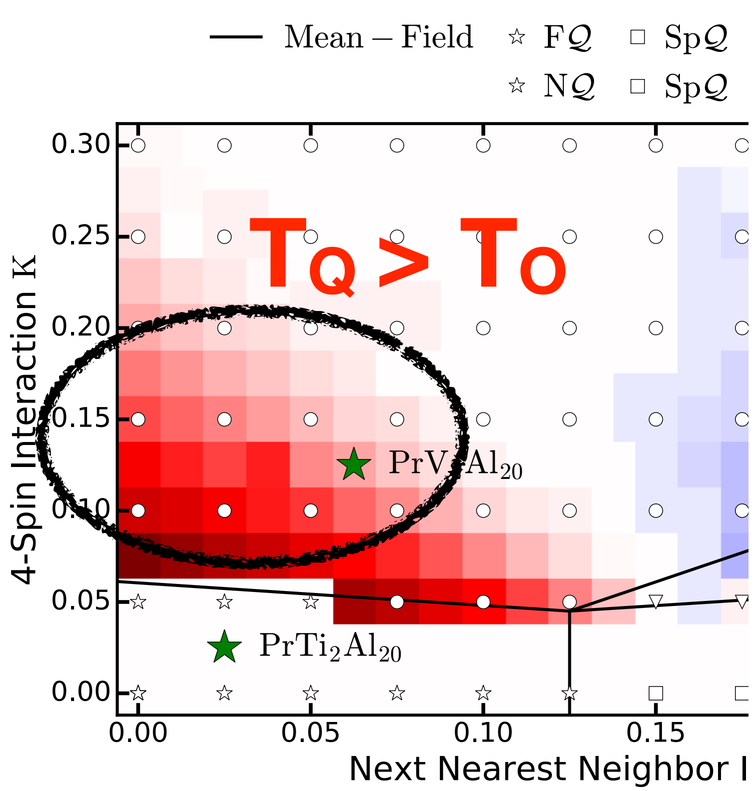
K

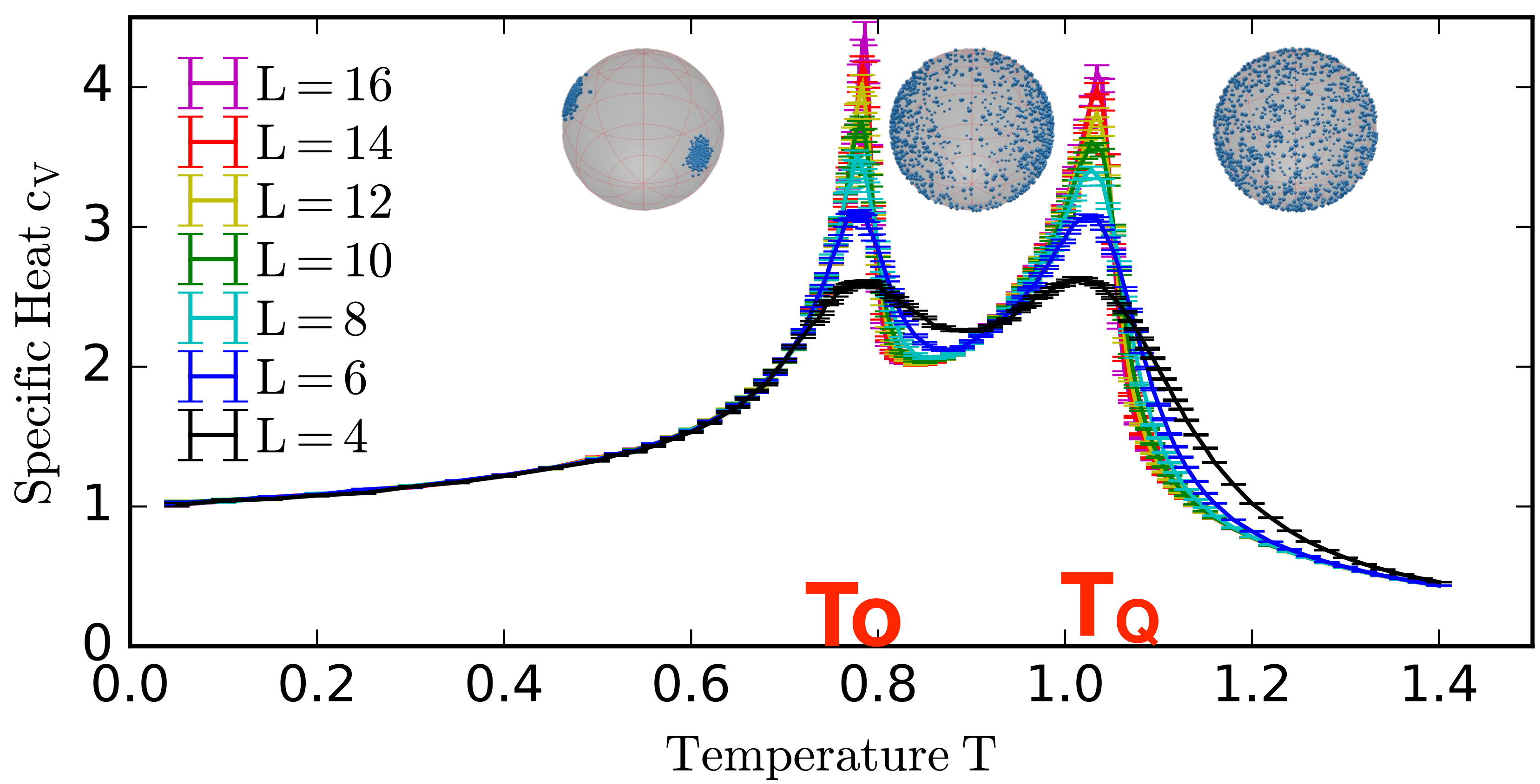
with increasing four spin interaction K

Multipolar order and finite T transitions

Quadrupolar, Octupolar orderings

quadrupolar + octupolar coexisting order at finite T ?

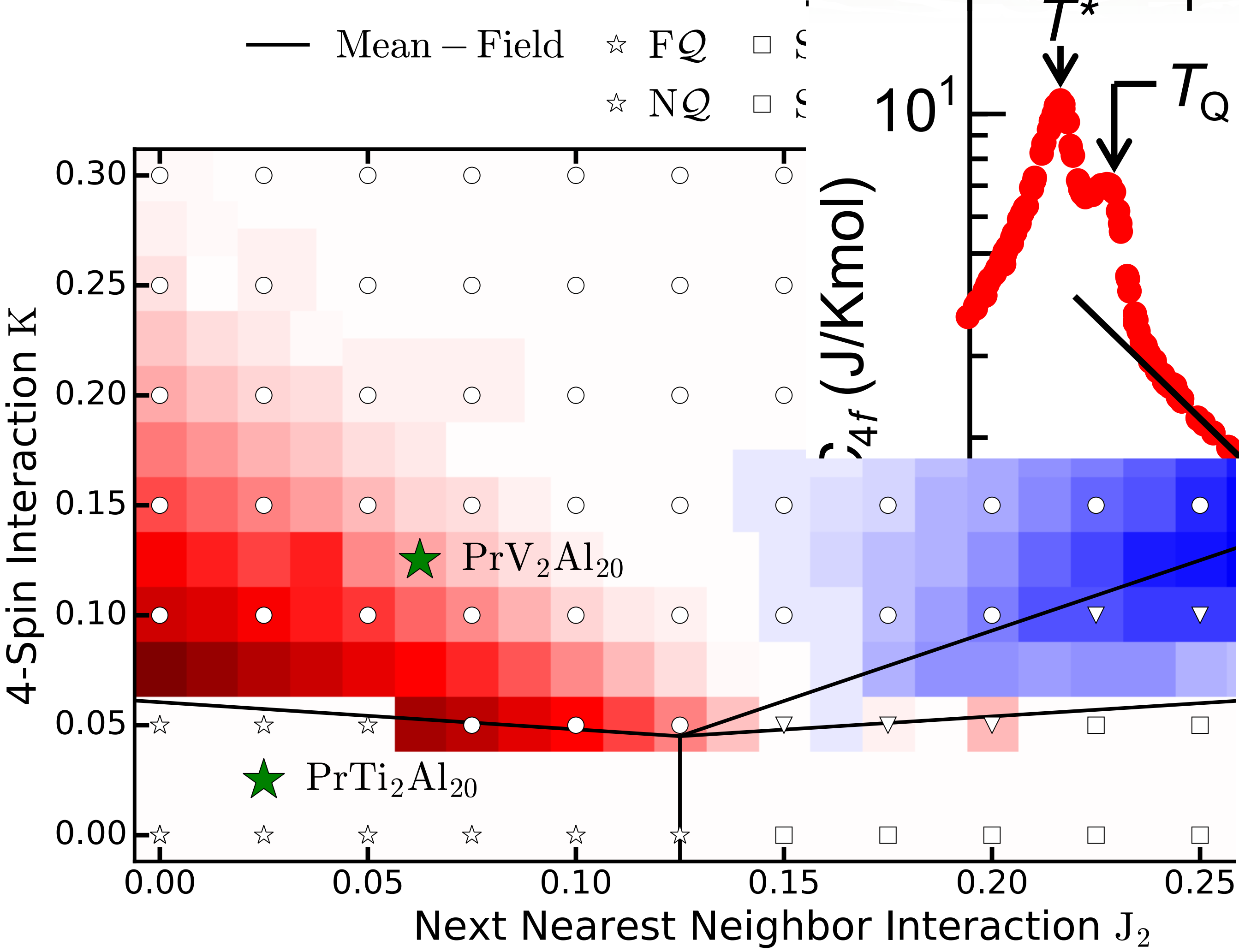


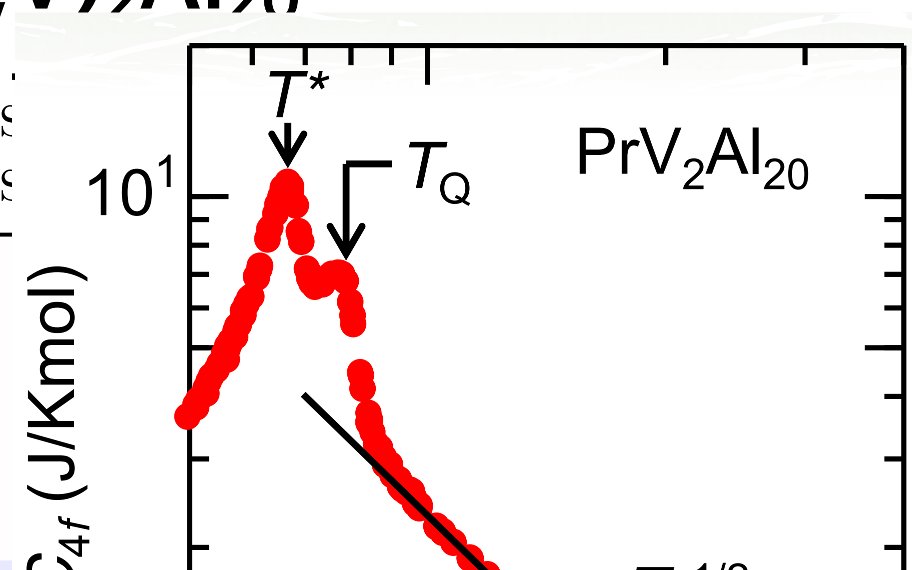


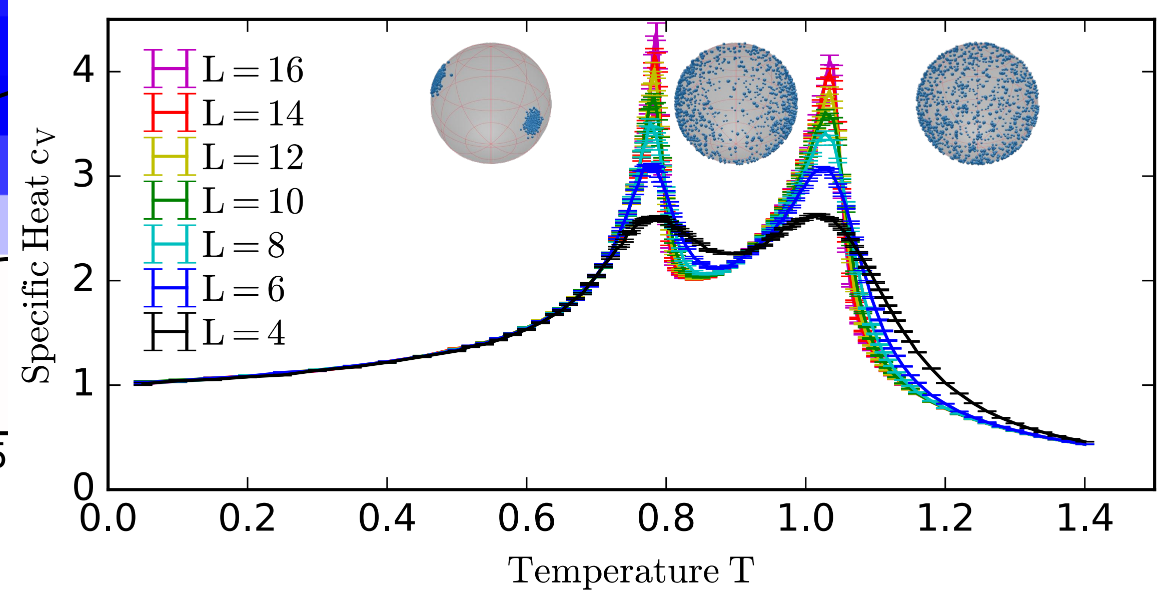
double transitions

Multipolar order and finite T transitions

Comparison with Pr(Ti,V)$_2$Al$_{20}$







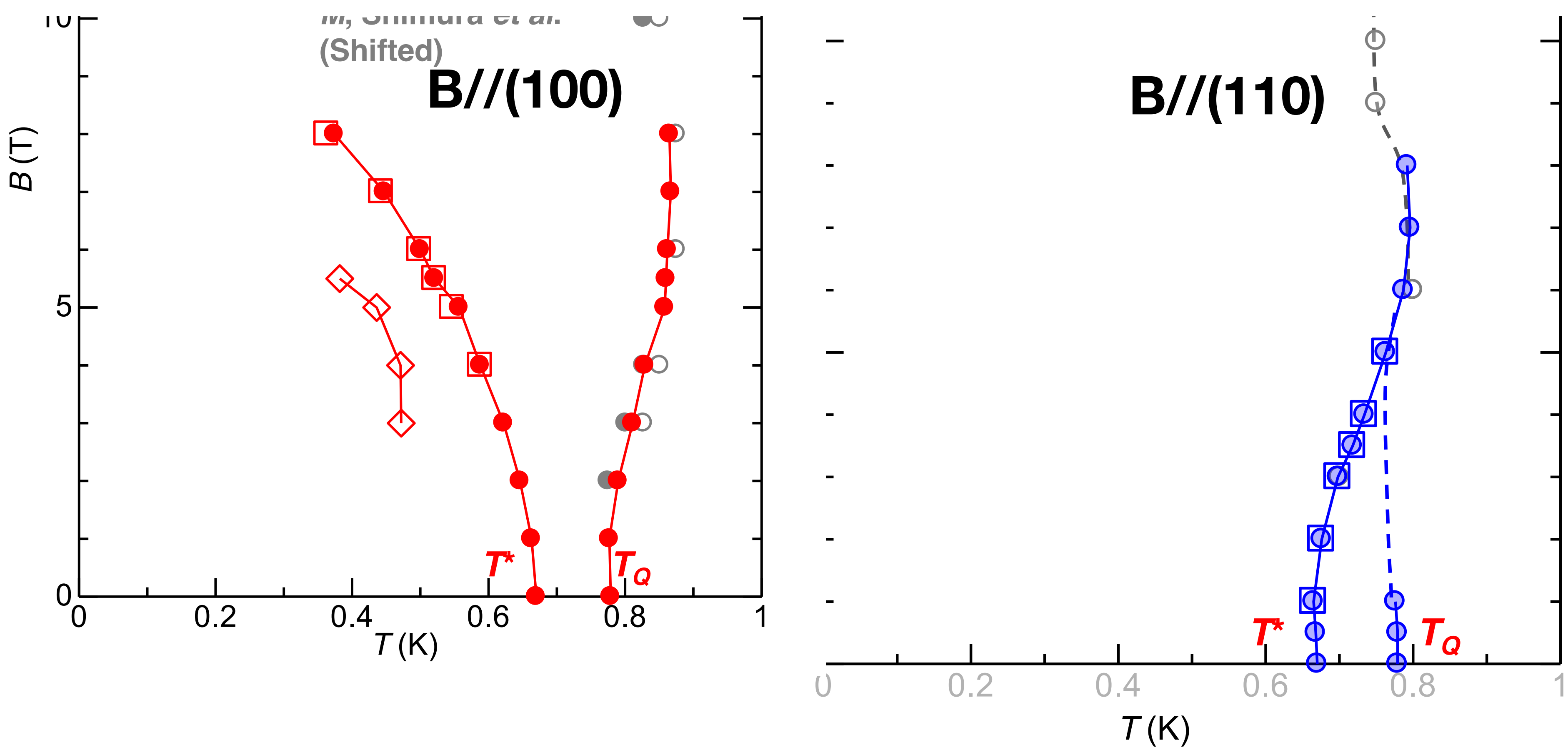
PrTi$_2$Al$_{20}$ → **PrV$_2$Al$_{20}$**

stronger hybridization leads to larger K, J

Quadrupole (T_Q)-octupole (T_O) double transitions

Multipolar order in magnetic fields

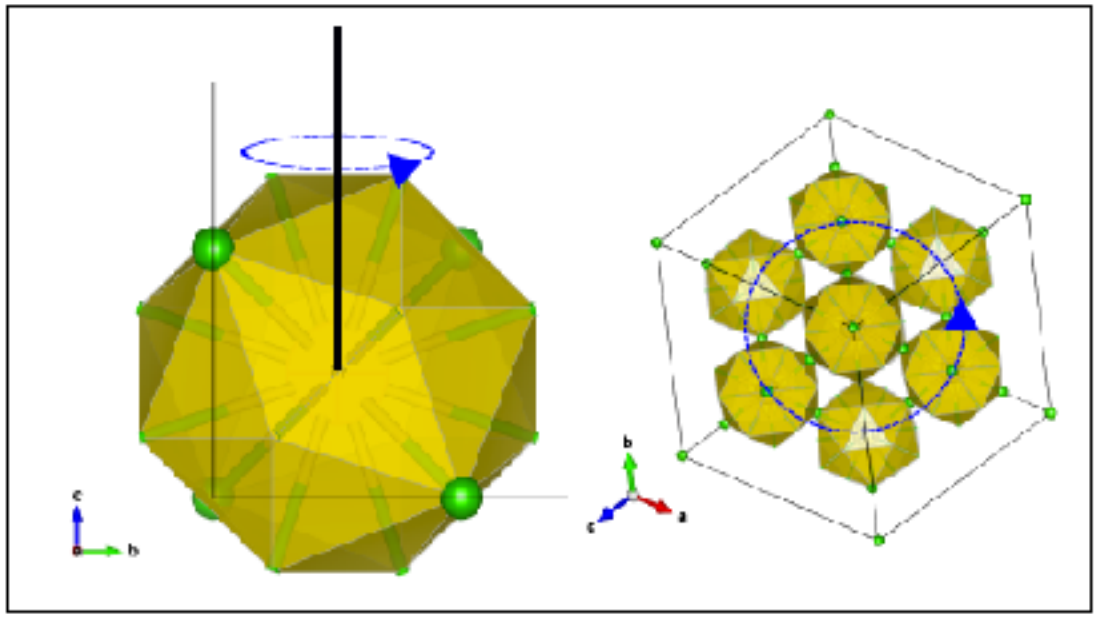
PrV_2Al_{20}



Anisotropy in fields with double transitions?

Multipolar order in magnetic fields

Local symmetry



$$\Theta: \quad \tau^z_{A/B} \to -\tau^z_{A/B},$$
$$\mathcal{I}: \quad \vec{\tau}_A \leftrightarrow \vec{\tau}_B,$$
$$\mathcal{S}_{4z}: \quad \tau^{\pm}_{A/B} \to -\tau^{\mp}_{A/B}; \ \tau^z_{A/B} \to -\tau^z_{A/B}$$
$$\sigma_{d1}: \quad \tau^{\pm}_{A/B} \to -\tau^{\mp}_{A/B}; \ \tau^z_{A/B} \to -\tau^z_{A/B}$$
$$\mathcal{C}_{31}: \quad \tau^{\pm}_{\mu} \to e^{\pm i2\pi/3}\tau^{\pm}_{\mu}$$

symmetry analysis with (anti)ferroquadrupolar order parameter ϕ and octupolar order parameter m

ferro- case ϕ_u, m_u **antiferro- case ϕ_s, m_s**

$$\phi_{u,s} \equiv \langle \tau^+_A \rangle \pm \langle \tau^+_B \rangle$$
$$m_{u,s} \equiv \langle \tau^z_A \rangle \pm \langle \tau^z_B \rangle$$

(u)niform / (s)taggered

Multipolar order in zero magnetic fields

Order Parameters

$$\phi_{u,s} \equiv \langle \tau_A^+ \rangle \pm \langle \tau_B^+ \rangle$$
$$m_{u,s} \equiv \langle \tau_A^z \rangle \pm \langle \tau_B^z \rangle$$

Quadrupole, Octupole

Within Quadrupolar ordering

(i) Pure Ferro-Quadrupolar order **$PrTi_2Al_{20}$**

(ii) Antiferro-Quadrupole with "Parasitic" Ferro Quadrupole

PrV_2Al_{20}

$$\mathcal{F}_{\text{int}}^{(3)} = i\lambda(\phi_s^2\phi_u - \phi_s^{*2}\phi_u^*).$$

Landau Theory of Quadrupole and Octupoles

No magnetic field (B=0)

ϕ_u Ferro-Quadrupole (FQ)
ϕ_s Antiferro-Quadrupole (AFQ)
m_u Ferro-Octupole (FO)

symmetry allows	free energy	properties
cubic (FQ) vs sixth order (AFQ) of anisotropic term	$iv_u(\phi_u^3 - \phi_u^{*3})$ $w_s(\phi_s^6 + \phi_s^{*6})$	phase locking 3-state degeneracy 6-state degeneracy Domains, Field directional anisotropy
coupling between FQ and AFQ	$i\lambda(\phi_u\phi_s^2 - \phi_u^*\phi_s^2)$	Coexisting AFQ and FQ Pure FQ Pure AFQ (X), AFQ+FQ
interaction between order parameters FQ,AFQ and FO	$c_{us}^{\phi\phi}\|\phi_u\|^2\|\phi_s\|^2 + c_{uu}^{\phi m}\|\phi_u\|^2 m_u^2 + c_{su}^{\phi m}\|\phi_s\|^2 m_u^2$	Competition between AFQ and FQ AFQ, FQ induced FO Double transitions : PM-> AFQ+FQ -> AFQ+FQ+FO

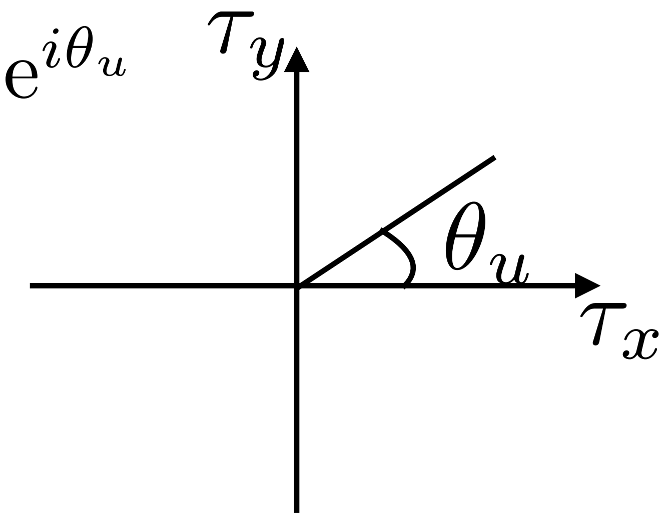
Multipolar order in zero magnetic fields

Order Parameters

$$\phi_{u,s} \equiv \langle \tau_A^+ \rangle \pm \langle \tau_B^+ \rangle$$
$$m_{u,s} \equiv \langle \tau_A^z \rangle \pm \langle \tau_B^z \rangle$$

Quadrupole, Octupole

$$\phi_u \equiv |\phi_u| e^{i\theta_u}$$



Ferro-Quadrupolar phase

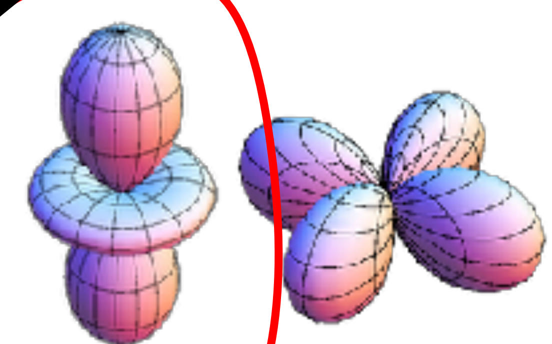
$$\mathcal{F}_{\phi u} = r_{u\phi}|\phi_u|^2 + iv(\phi_u^3 - \phi_u^{*3}) + g_{u\phi}|\phi_u|^4 + \ldots$$

Z_3 clock model : 1st order

cubic anisotropy locks the phase θ_u

Quadrupole

$PrTi_2Al_{20}$



$3J_z^2 - J^2$ **$J_x^2 - J_y^2$**

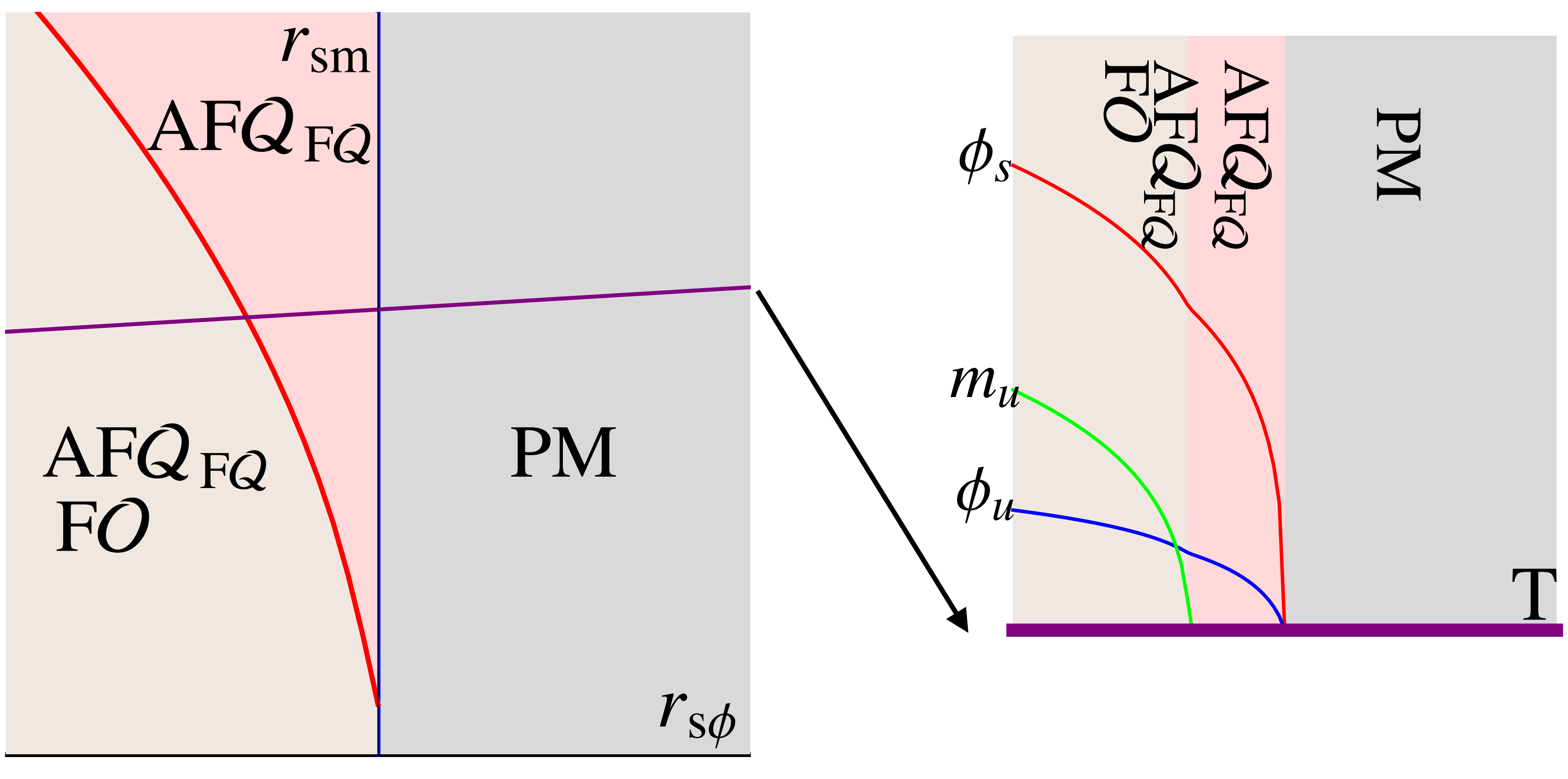
Multipolar order in zero magnetic fields

Coexisting Antiferro-Quadrupole and octupole

$$\mathcal{F}_{\phi u} = r_{u\phi}|\phi_u|^2 + iv(\phi_u^3 - \phi_u^{*3}) + g_{u\phi}|\phi_u|^4 + \ldots$$

$$\mathcal{F}_{\phi s} = r_{s\phi}|\phi_s|^2 + g_{s\phi}|\phi_s|^4 + w(\phi_s^6 + \phi_s^{*6}) + \ldots$$

$$\mathcal{F}_{mu} = r_{um}m_u^2 + g_{um}m_u^4 + \ldots$$



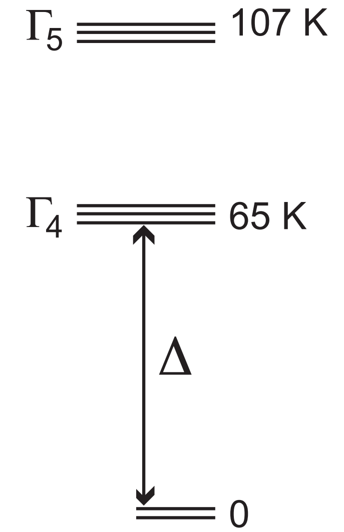
PRB 98 134447 (2018)

Multipolar order in magnetic fields

Quadrupole couples quadratic in fields

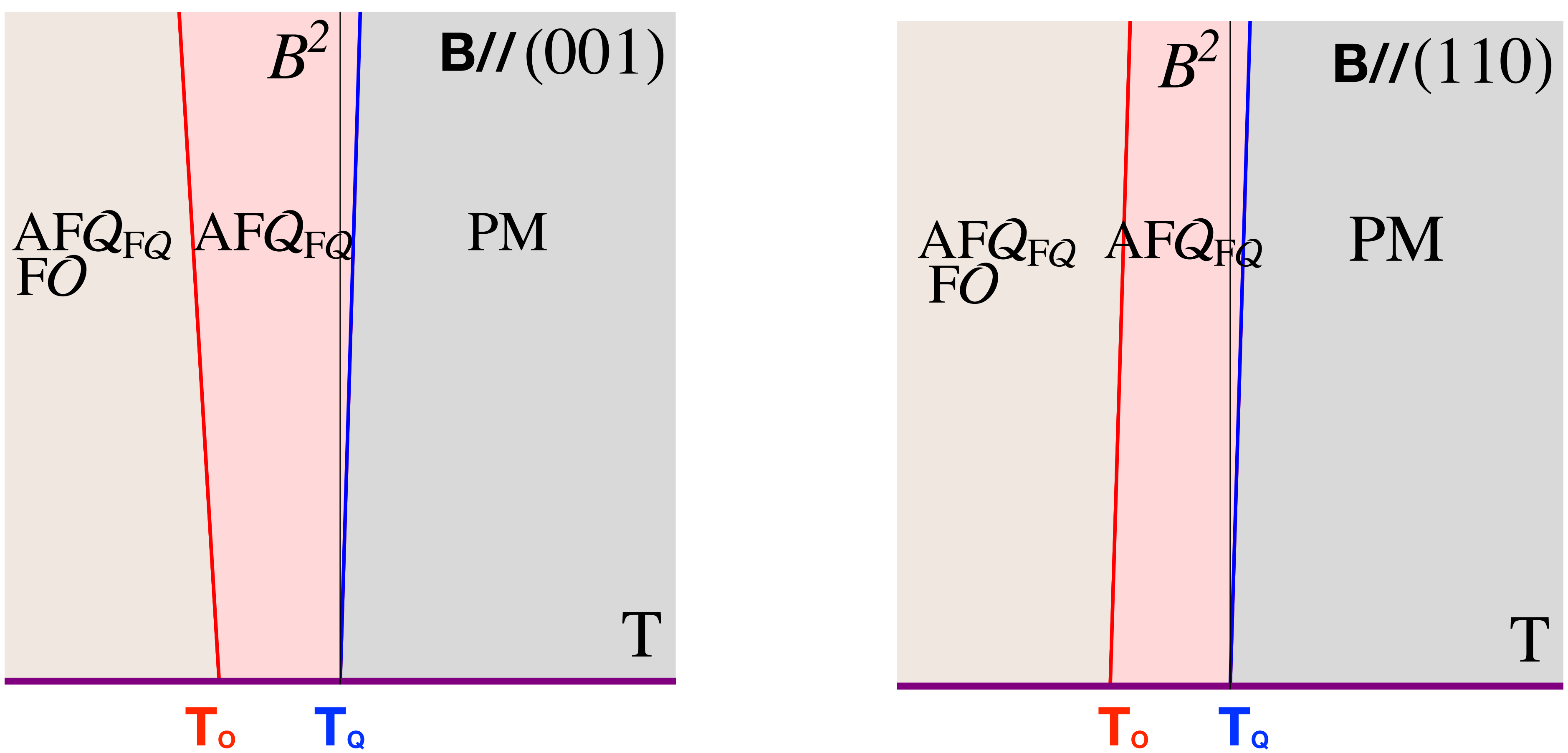
$$H_{\text{field}} = \gamma B^2 (b_1 \tau^x + b_2 \tau^y)$$

$$\gamma \propto \left(-\frac{14}{3\Delta(\Gamma_4)} + \frac{2}{\Delta(\Gamma_5)} \right),$$



an **Field effect based on Landau theory**

wi

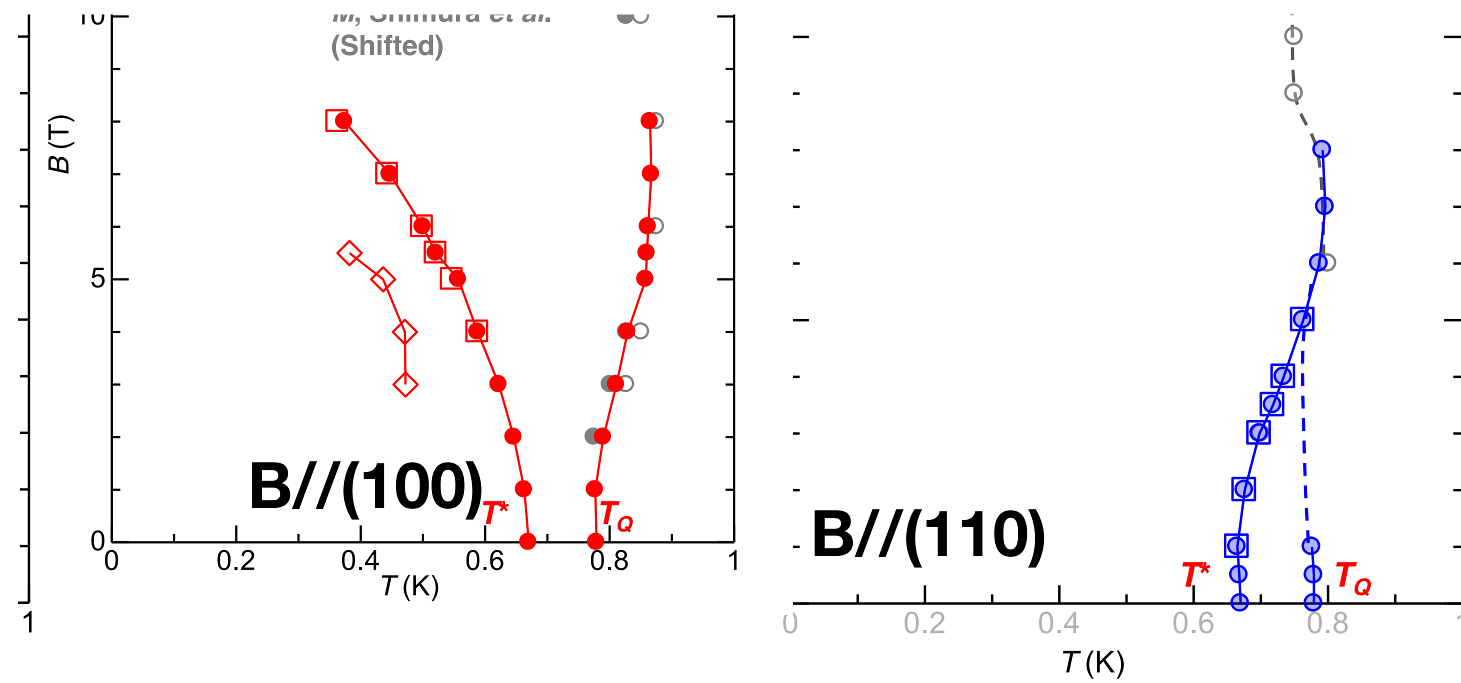


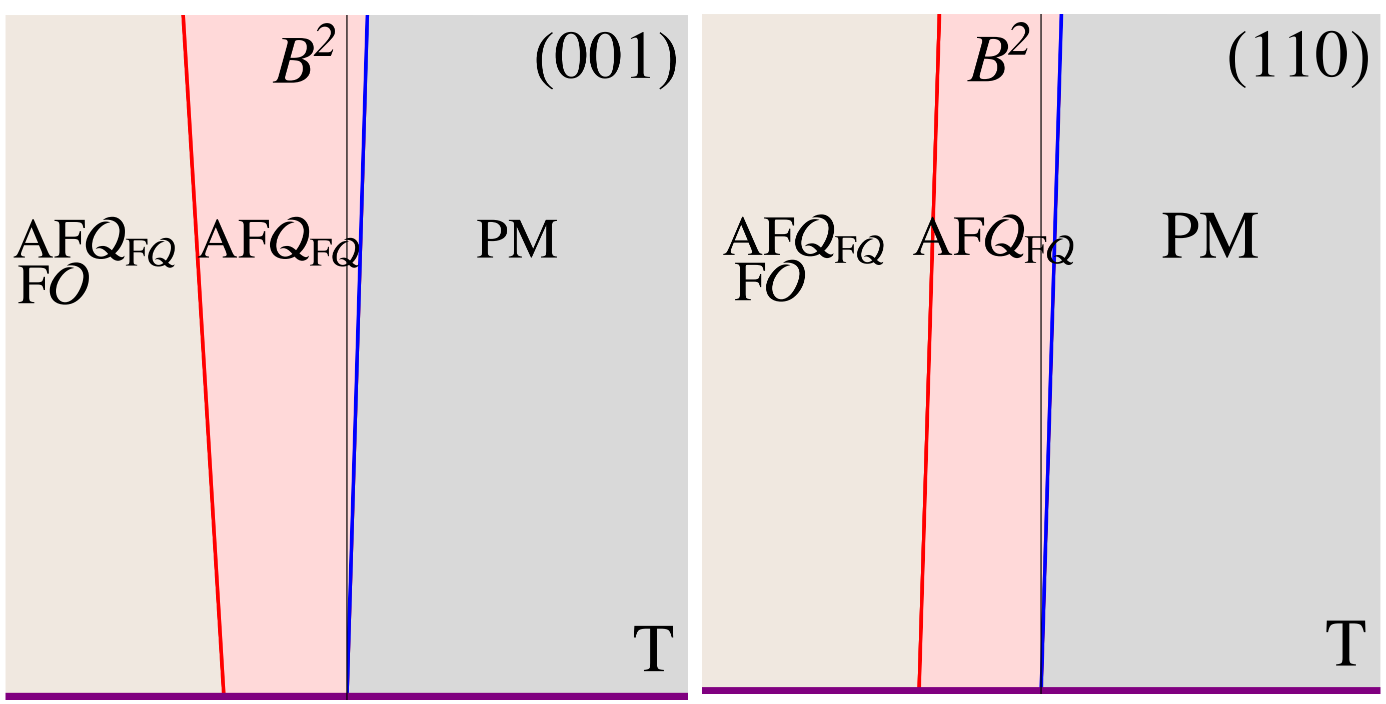
octupolar order transition temperature T_O is very sensitive to B direction

Multipolar order in magnetic fields

Landau Theory Analysis with fields

Comparison with PrV_2Al_{20}





Quadrupolar and octupolar double transition in fields

—> Field direction matters. anisotropic (100) vs (110)

Multipolar order and Superconductivity

Superconductivity in $Pr(TM)_2(Al,Zn)_{20}$

$PrTi_2Al_{20}$ — T_c = 0.2 K

PrV_2Al_{20} — T_c = 0.05 K

Compounds	Δ (K)	T_c (K)	T_Q (K)
$PrIr_2Zn_{20}$	27.6	0.05	0.11 (AFQ)
$PrRh_2Zn_{20}$	31.2	0.06	0.06 (AFQ)
$PrRu_2Zn_{20}$	37.0	— (0.04)	—
$PrOs_2Zn_{20}$		— (0.4)	—
PrV_2Al_{20}	40	0.05	0.6 (AFQ)
$PrTi_2Al_{20}$	65.6	0.2	2.0 (FQ)
$PrNb_2Al_{20}$	21.3	— (0.1)	—
$PrCr_2Al_{20}$		— (0.4)	—
$PrNi_2Cd_{20}$	12	— (1.2)	—
$PrPd_2Cd_{20}$	11	— (1.2)	—

T.Onimaru and H. Kusunose JPSJ 85, 082002 (2016)

Correlation between magnetic quadrupolar order and superconductivity?

Exotic scenario …

Multipolar order and Superconductivity

$Pr(TM)_2(Al,Zn)_{20}$

SOC + cubic symmetry

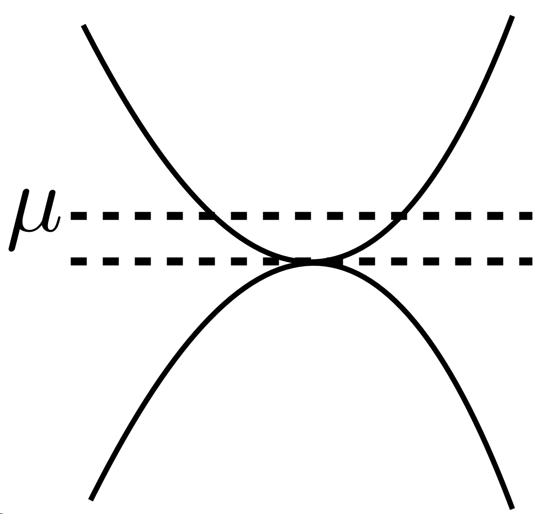
Quadratic

Fermi Surface with SO(3) symmetry

not?

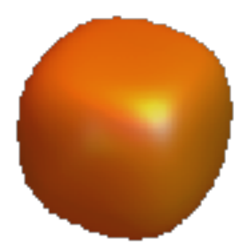
$$d_1 = \sqrt{3} k_y k_z,$$
$$d_2 = \sqrt{3} k_x k_z,$$
$$d_3 = \sqrt{3} k_x k_y,$$
$$_4 = \frac{\sqrt{3}}{2}(k_x^2 - k_y^2),$$
$$d_5 = \frac{1}{2}(3k_z^2 - \mathbf{k}^2)$$

Luttinger Hamiltonian with J=3/2



Quadratic band touching with cubic symmetry

Fermi Surface with cubic symmetry



ices for J=3/2

$\psi_{\mathbf{k}}$

$i=1$

t_{2g} & e_g in k

Multipolar order and Superconductivity

Quadratic band touching at k=0 point + interaction

$$\mathcal{H}_0(\mathbf{k}) = \psi^\dagger_{\mathbf{k}}\Big(c_0\mathbf{k}^2 - \mu + \sum_{i=1}^{5} c_i\ d_i(\mathbf{k})\gamma_i\Big)\psi_{\mathbf{k}}$$

$$d_1 = \sqrt{3}k_y k_z,$$
$$d_2 = \sqrt{3}k_x k_z,$$
$$d_3 = \sqrt{3}k_x k_y,$$
$$d_4 = \frac{\sqrt{3}}{2}(k_x^2 - k_y^2),$$
$$d_5 = \frac{1}{2}(3k_z^2 - \mathbf{k}^2)$$

t_{2g} & e_g in k

+

$$\mathcal{H}_{int}(\mathbf{k}) = g_0(\psi^\dagger\psi)^2 + \sum_{a=1}^{5} g_i(\psi^\dagger\gamma_a\psi)^2$$

Fierz Identity $\Delta_i \equiv \langle \sum_{\mathbf{k}} \psi^T_{-\mathbf{k}}\gamma_{13}\gamma_i\psi_{\mathbf{k}}\rangle$

exactly decoupled into s and d wave pairing channels

→ **open attractive d wave pairing channels**

e.g. $g=g_i$ case $(g_0+5g)\ \Delta^+_s\Delta_s + (g_0-3g)\ \Delta^+_d\Delta_d$

J=3/2 Luttinger model and Superconductivity

Invariant Theory with SO(3) symmetry

$\vec{\Delta} = (\underbrace{\Delta_1, \Delta_2, \Delta_3}_{t_{2g}}, \underbrace{\Delta_4, \Delta_5}_{e_g})$ **complex tensor order parameters of d-wave**

$$I_1 = \mathrm{tr}(\phi^\dagger \phi), I_2 = \mathrm{tr}(\phi^2), I_3 = \mathrm{tr}(\phi^{\dagger 2}), I_4 = \mathrm{tr}(\phi^3),$$
$$I_5 = \mathrm{tr}(\phi^{\dagger 3}), I_6 = \mathrm{tr}(\phi^2 \phi^\dagger), I_7 = \mathrm{tr}(\phi^{\dagger 2}\phi), I_8 = \mathrm{tr}(\phi^\dagger \phi \phi^\dagger \phi).$$

$$\phi_{ij} = \Delta_a \Lambda^a_{ij},$$

$$\Lambda^1 \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \Lambda^2 \frac{1}{\sqrt{3}} \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 2 \end{pmatrix},$$
$$\Lambda^3 = \begin{pmatrix} 0 & 0 & 1 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \end{pmatrix}, \Lambda^4 = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix}, \Lambda^5 = \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$$

e.g) $I_1 = 2|\vec{\Delta}|^2, \; I_2 I_3 = 4(\vec{\Delta}^2)(\vec{\Delta}^2)^*$

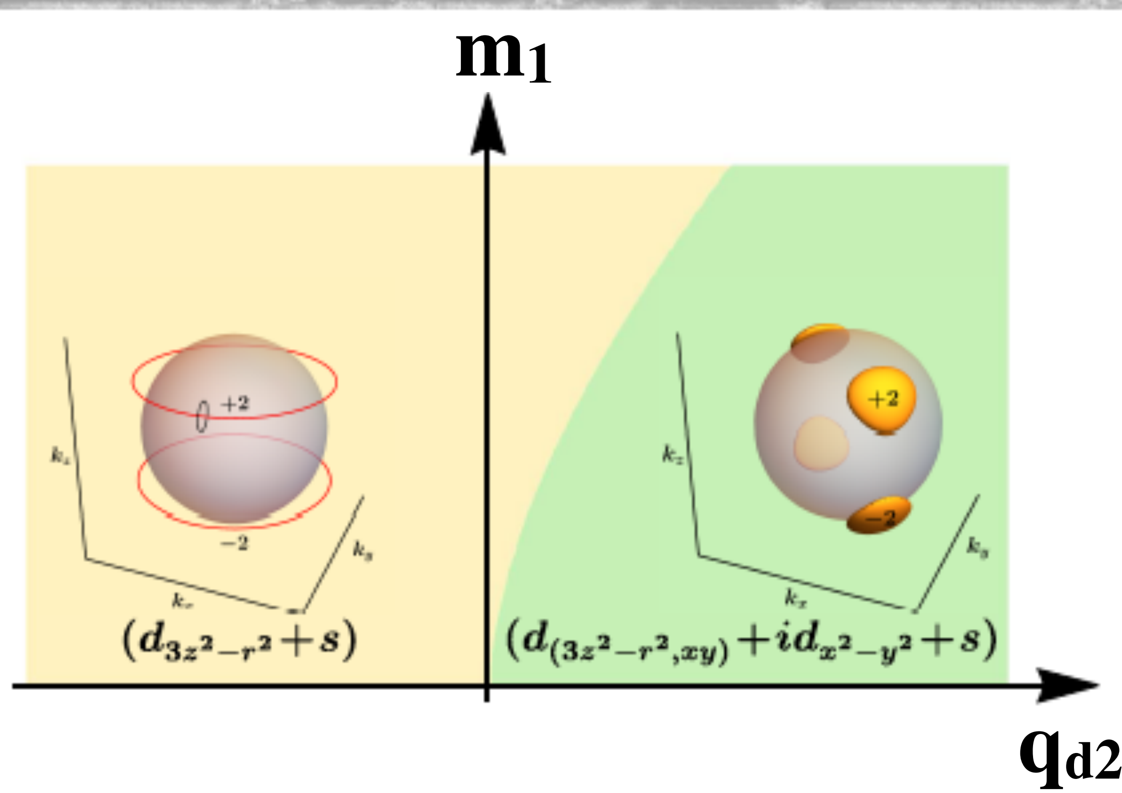
A. J. M. Spencer and R. S. Rivlin, Arch. Rational Mech Anal. 2, 309 (1958)
M. Artin, Journal of Algebra 11, 532 (1969)
C. Procesi, Advances in Mathematics 19, 306 (1976)
B. Igor and I.F. Herbut Phys Rev Letters 120.5 057002 (2018)

J=3/2 Luttinger model and Superconductivity

Invariant Theory with SO(3) symmetry

$\vec{\Delta} = (\underbrace{\Delta_1, \Delta_2, \Delta_3}_{t_{2g}}, \underbrace{\Delta_4, \Delta_5}_{e_g}) + \Delta_s$ complex tensor order parameters of d-wave

$$F = r_d|\vec{\Delta}|^2 + r_s|\Delta_s|^2 + q_{d_1}|\vec{\Delta}|^4 + q_{d_2}|\vec{\Delta}^2|^2 + q_s|\Delta_s|^4 + m_2(|\vec{\Delta}|^2|\Delta_s|^2)$$
$$+ m_3(\vec{\Delta}^2(\Delta_s^*)^2 + c.c.) + q_{d_3}\mathrm{tr}((\phi^\dagger\phi)^2) + m_1(\mathrm{tr}(\phi^2\phi^\dagger)\Delta_s^* + c.c.)$$



$d_{3z2\text{-}r2}$ + s

selective d wave pairing with parasitic s wave

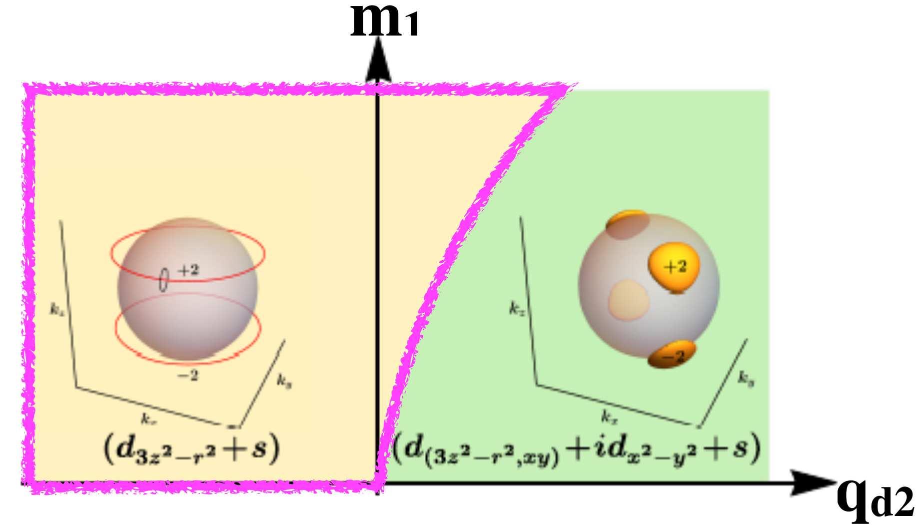
applicable to YPtBi (half heusler)

arXiv:1811.04046 (2018)

J=3/2 Luttinger model and Superconductivity

Landau Free Energy $q_{d_2}|\vec{\Delta}^2|^2 + m_1(\mathrm{tr}(\phi^2\phi^\dagger)\Delta_s^* + c.c.)$

(i) $q_{d2} < 0$: real Δ

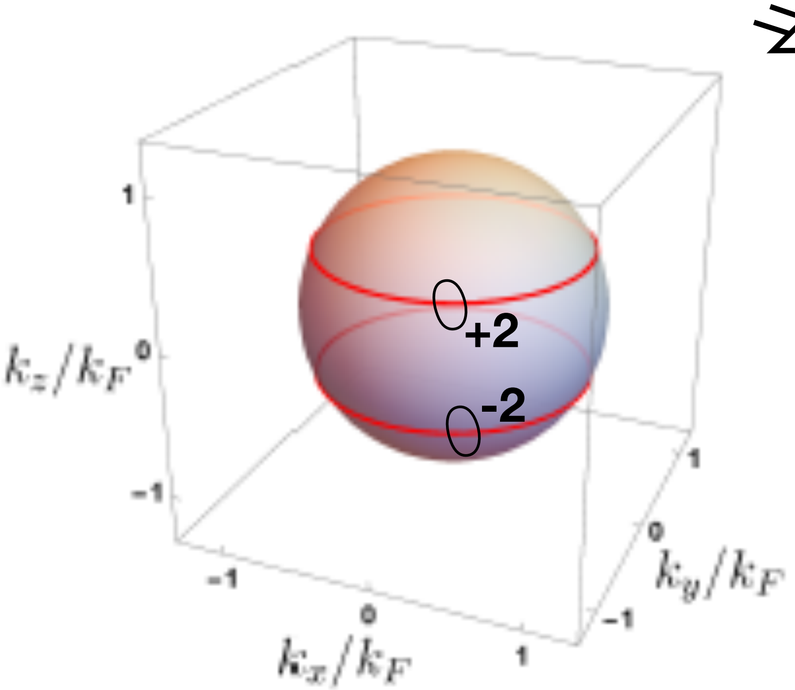


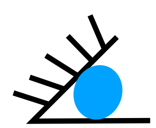
m_1 favors $d_{3z^2-r^2}$ wave with parasitic s wave

$\Delta_{3z^2-r^2} + \Delta_s$

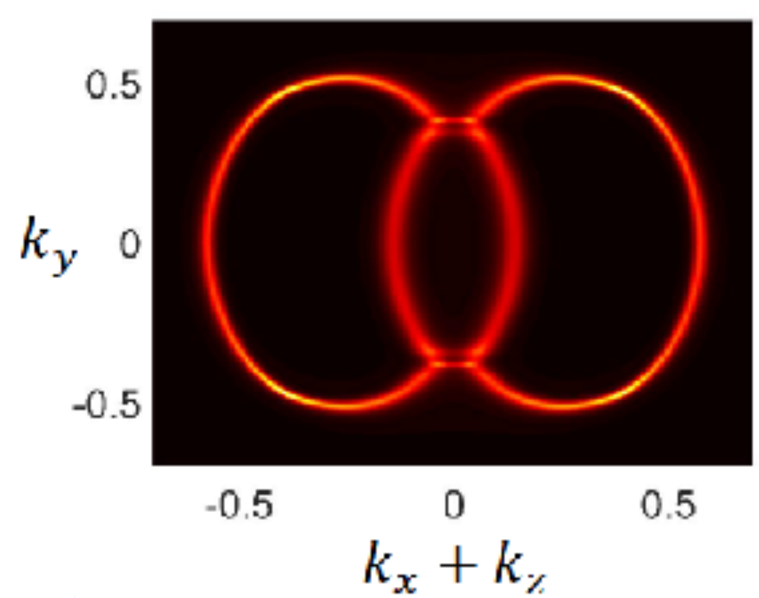
(10$\bar{1}$) plane

gap structure

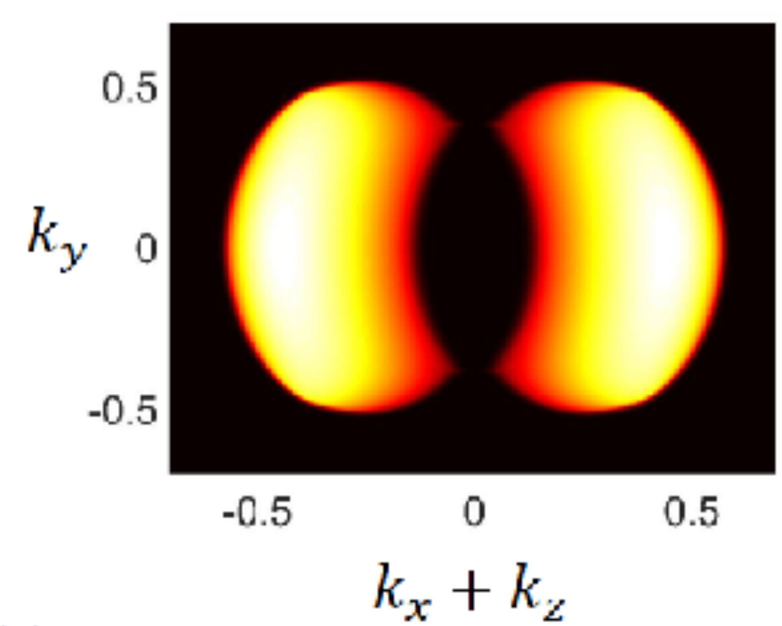




Bulk



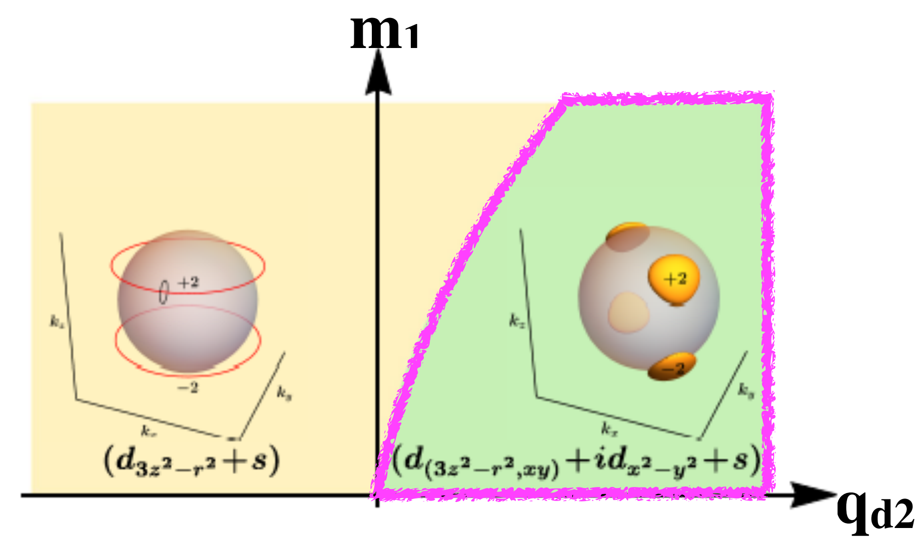
drumhead Surface



J=3/2 Luttinger model and Superconductivity

Landau Free Energy $$q_{d_2}|\vec{\Delta}^2|^2 + m_1(\mathrm{tr}(\phi^2\phi^\dagger)\Delta_s^* + c.c.)$$

(ii) $q_{d2} < 0$: complex Δ

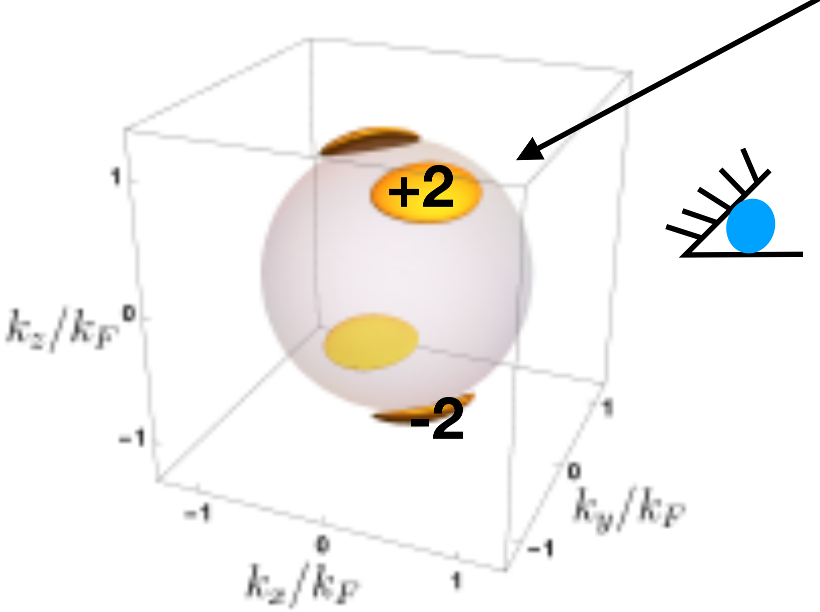


m_1 favors $d_{3z^2-r^2} + i\, d_{x^2-y^2}$ wave
with parasitic s wave with

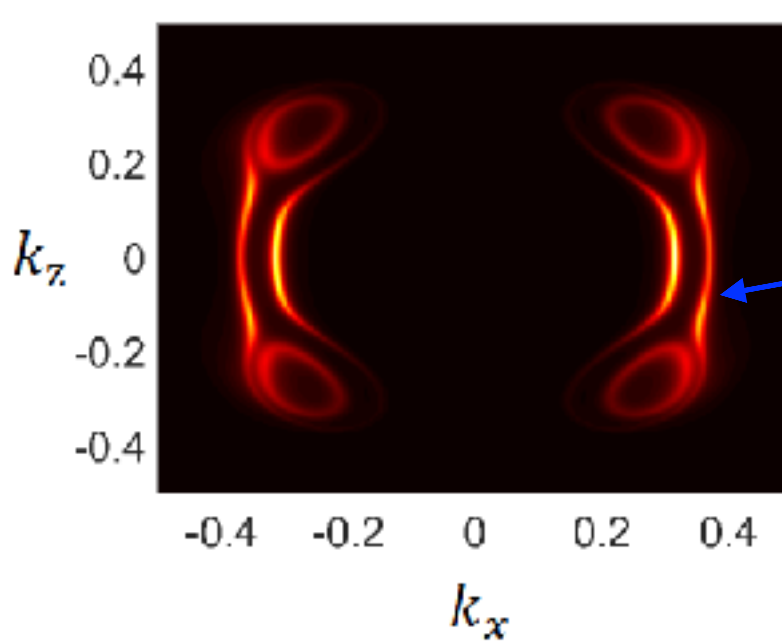
$$\Delta_{3z^2-r^2} + i\,\Delta_{x^2-y^2} + \Delta_s$$

Bogoliubov Quasiparticles form
Fermi Pocket having Chern number

gap structure



(010) surface plane

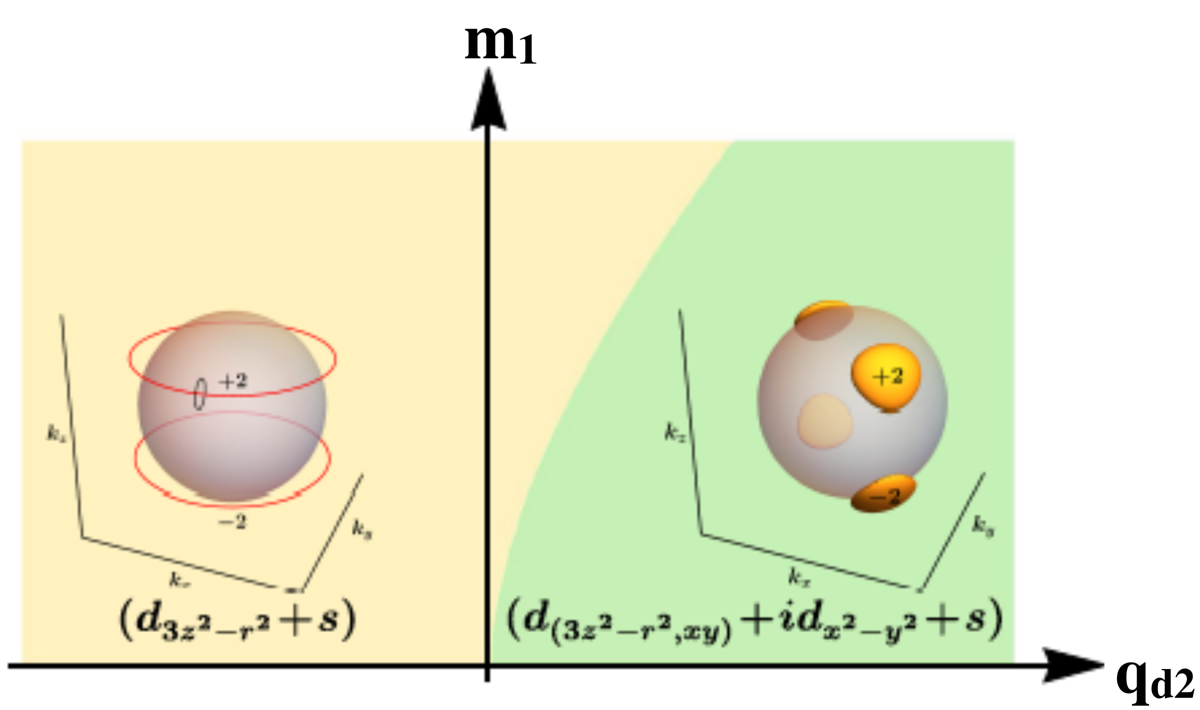


Fermi Arc

Multipolar order and Superconductivity

Attractive d wave pairing channel (with parasitic s wave)

Topological d+s superconductor

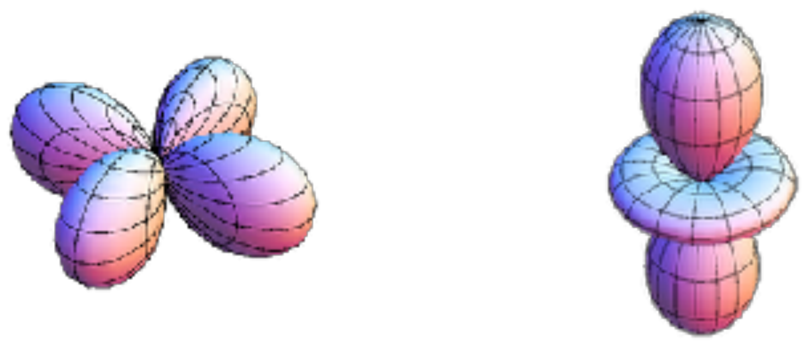


How does the quadrupolar order (e_g type) affect to superconductivity?

Quadrupolar order -> Fermi surface distortion

Multipolar order and Superconductivity

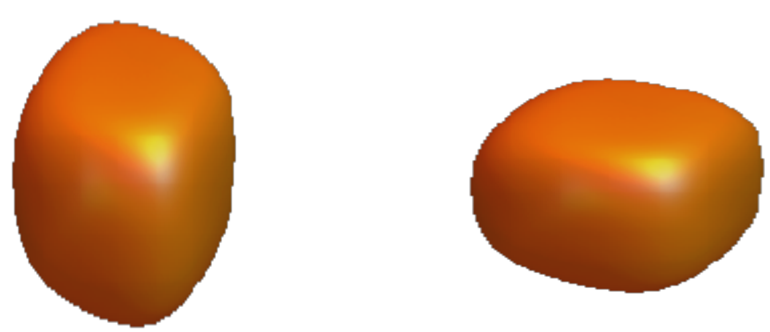
Ferro Quadrupolar order



$J_x^2 - J_y^2$ τ_x

$3J_z^2 - J^2$ τ_y

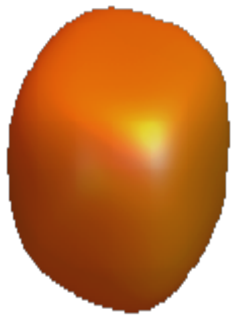
Fermi Surface Distortion



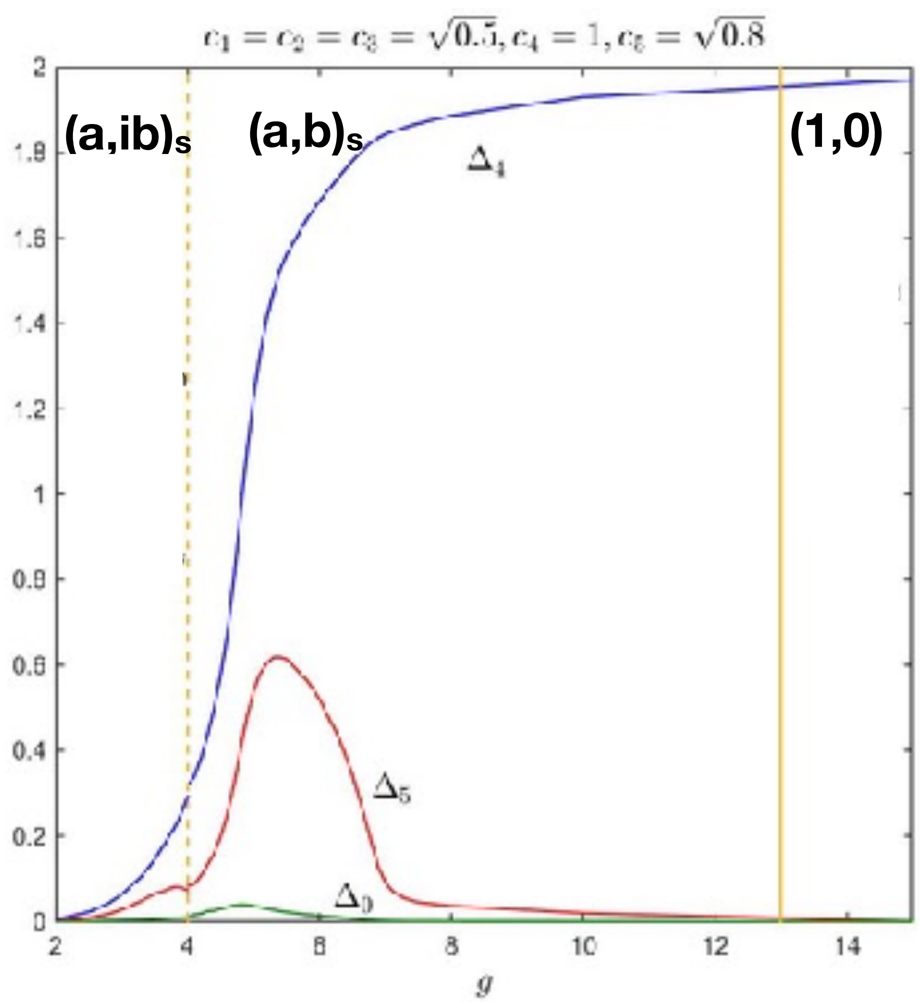
(under preparation)

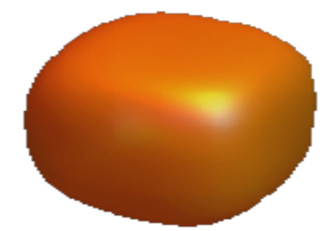
Multipolar order and Superconductivity

Fermi Surface Distortion and evolution of superconductivity

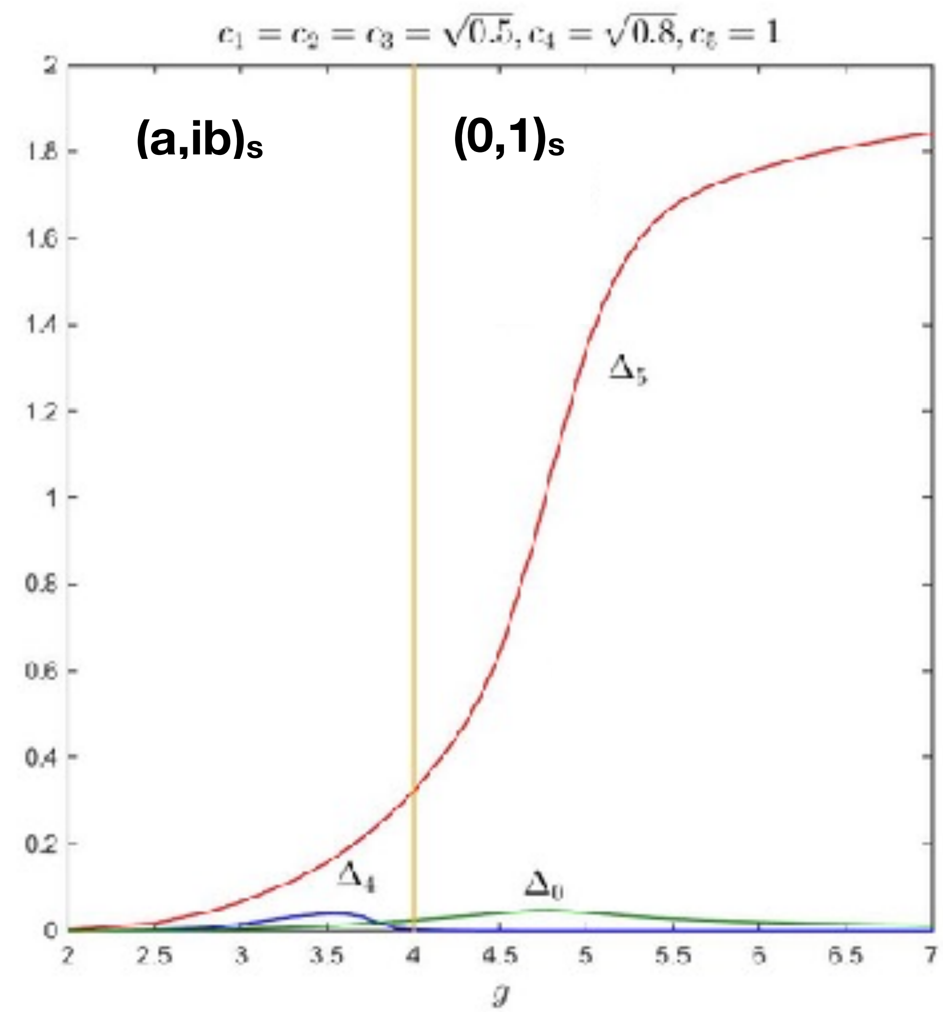


$(\Delta_{x^2-y^2}, \Delta_{3z^2-r^2})$

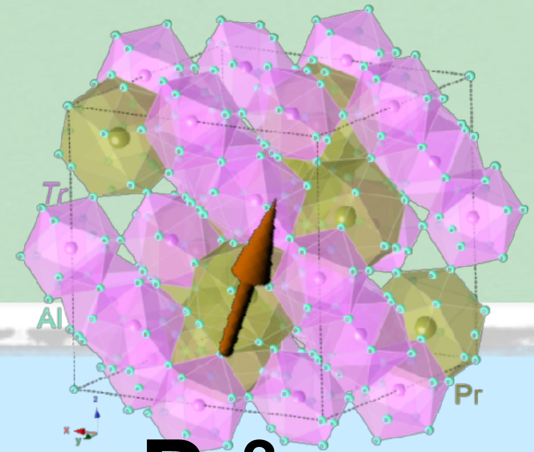




$(\Delta_{x^2-y^2}, \Delta_{3z^2-r^2})$



Summary



Pr^{3+}

Pr^{3+} : Quadrupolar, octupolar order

(i) Frustration & Multiple spin interactions

— double transitions of quadrupole - octupole orderings.

(ii) Magnetic field dependence

— Quadrupole couples quadratic in fields

Competition between quadrupolar anisotropic term vs field coupling term leads to very field directional dependence.

Superconductivity and ferroquadrupolar order

(i) Luttinger model with interaction

— d wave topological superconductivity is favored with parasitic s wave.

(ii) Ferroquadrupolar order distorts Fermi surface

— different types of d wave topological superconducting transition occurs.

Thank you!