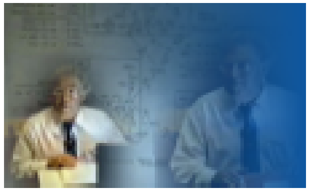
Alba Nova and Nordita Colloquium



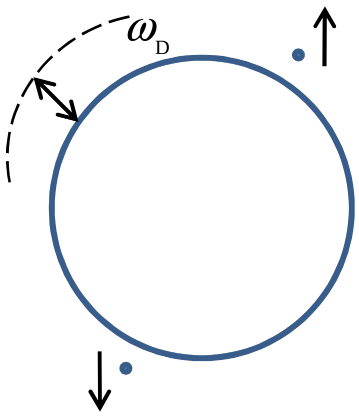
Stockholm - May 7, 2009

Superconductivity through intra-atomic excitations

Collaborations: J. Chang, I. Eremin P. McHale
P. Thalmeier, G. Zwicknagl

Superconductivity

instability of the normal state $\Longrightarrow$ Cooper-pair formation



Schrödinger equat. for 2 electrons with attrat. $V\left(\mathbf{r}_1-\mathbf{r}_2\right)$

$$\Longrightarrow \quad \Delta E=-2\omega_D e^{-2/\left(N(0)V\right)}$$

creation operat. for single pair : $\phi_0^+=\sum_{\mathbf{k}} g(\mathbf{k})\, c_{\mathbf{k}\uparrow}^+ c_{-\mathbf{k}\downarrow}^+$

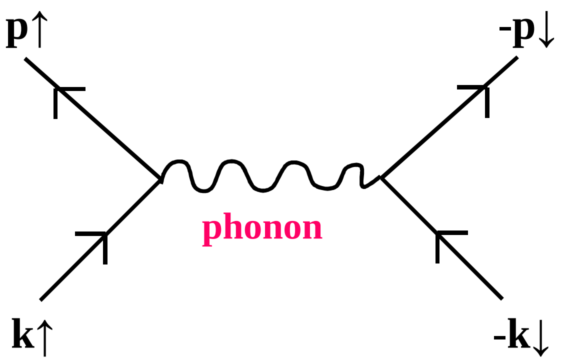
$$\phi\left(\mathbf{r}_1-\mathbf{r}_2\right)=\sum_{\mathbf{k}} g(\mathbf{k})\, e^{i\mathbf{k}\left(\mathbf{r}_1-\mathbf{r}_2\right)}$$

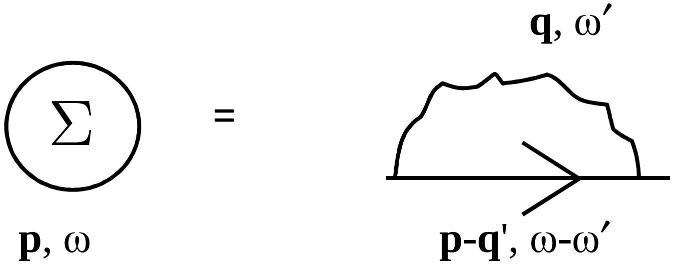
BCS ground state :

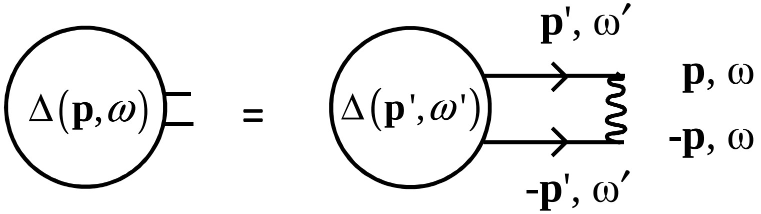
coherent state : $\left|\psi_0\right\rangle=e^{\phi_0^+}|0\rangle=\exp\left(\sum_{\mathbf{k}} g(\mathbf{k})\, c_{\mathbf{k}\uparrow}^+ c_{-\mathbf{k}\downarrow}^+\right)|0\rangle$

pairing in time reversed states !

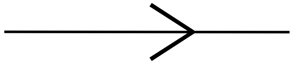
Taking retardation into account: Eliashberg equations for T_c:







$$= K(\mathbf{p}-\mathbf{p}', \omega-\omega')$$



$$= G(\mathbf{p}', \omega')$$

filled skutterudites

$\text{LaOs}_4\text{Sb}_{12}$ $T_c = 0.74\ \text{K}$

$\text{PrOs}_4\text{Sb}_{12}$ $T_c = 1.85\ \text{K}$

$T_\hbar$ symmetry , presumable isotropic s-wave superconductivity

large jump in specific heat : $\Delta C/T_c \approx 500\text{mJ}\big/\big(\text{mol}\cdot\text{K}^2\big)$

⟶ strong coupling

only difference : La^{3+} has $4f^0$ while Pr^{3+} has $4f^2$ electrons

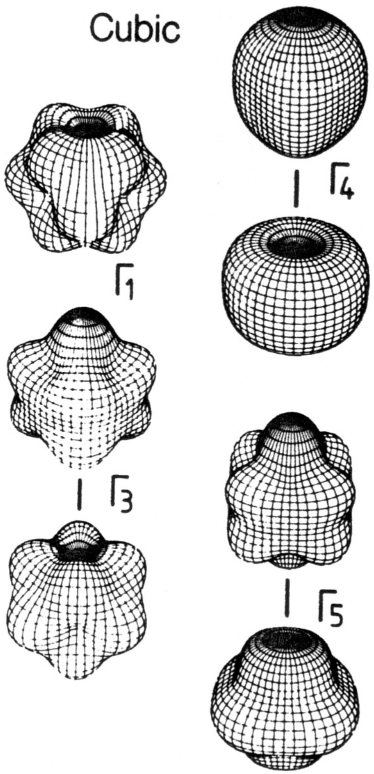
phonons are practically the same in both cases

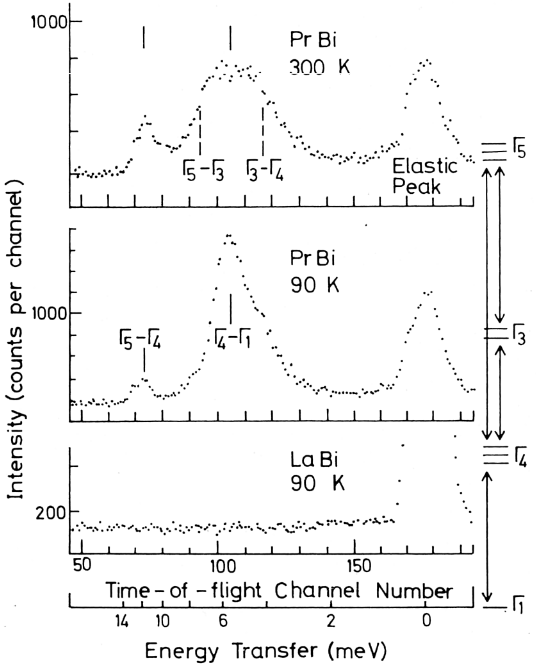
RE ions : incomplete 4f shell, **LS** coupling

Hund's rules → total J

Pr^{3+} → J = 4 9-fold degeneracy

split by CEF of neighborhood





effect of CEF excitations on superconductivity

(P.F., L. Hirst, A. Luther, Z. Phys. 1970)

interactions with cond. electrons

isotropic exchange:
$$H_{\mathrm{ex}} = -2(g-1)J_{\mathrm{ex}} \sum_{\mathbf{kk}'} c^{+}_{\mathbf{k}'\alpha} \boldsymbol{\sigma}_{\alpha\beta} c_{\mathbf{k}\beta} \mathbf{J}$$

anisotr. Coulomb scatt.:
$$H_{\mathrm{AC}} = \sum_{\mathbf{kk}'\sigma} \sum_{m=-2}^{2} I_2 \cdot \left(\frac{5}{4\pi}\right)^{1/2} Q_2 \left\{ y_2^m(\mathbf{J}) c^{+}_{\mathrm{k's}\sigma} c_{\mathrm{kdm}\sigma} + \mathrm{h.c.} \right\}$$

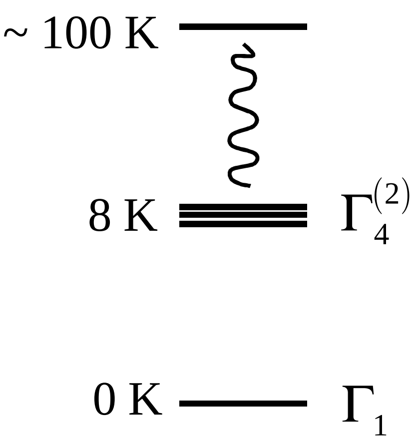
time reversal symmetry:

H_{ex} no ⟹ pair breaking

H_{AC} yes ⟹ pair forming

there is also anisotropic exchange, total of 2(2L+1) interactions; usually H_{ex} dominant

$PrOs_4Sb_{12}$: from inelast neutron scatt. experiments :



$$\left|\Gamma_4^{(2)}(m)\right\rangle = \sqrt{1-d^2}\left|\Gamma_5(m)\right\rangle + d\left|\Gamma_4(m)\right\rangle$$

cubic symm.

$m = +, 0, -$ $d = 0.26$

$\Gamma_1 \rightarrow \Gamma_5$ = quadrupol. (weak dispersion)

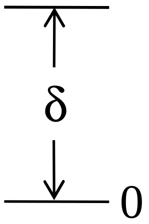
$\Gamma_1 \rightarrow \Gamma_4$ = magnetic

⟹ quadrup. scatt. dominant ⟹ pair formation

bosonic propagator: quadrupolar susceptibility

for a two-level system

$$K(\omega) = |M|^2 \frac{2\delta \tanh \delta/2T}{\delta^2 - \omega^2}$$



solution of Eliashberg equations:

$LaOs_4Sb_{12}$: phonon $\omega_E = 26$ meV , $\mu^* = 0.1$ $\lambda_{ph} = 0.33$ ⟹ $T_c = 0.74$ K

$$\lambda = \lambda_{ph} + \lambda_Q \pm \lambda_M$$

adjusted: $\Delta T_c = 1.11$ K and $\frac{m^*}{m_b} \simeq 2.5$ dHvA

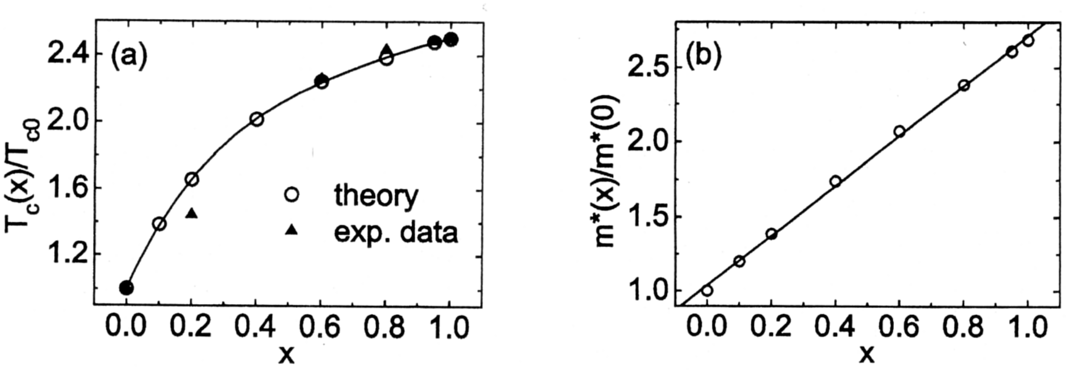
coupling constants agree with the ones estimated from the dispersion of the magnetic and quadrupolar excitons

explained: nonlinear $T_c(x)$ behaviour

linear $m^*(x)/m^*(o)$ relation

$2\Delta_0/k_B T_c \simeq 5.4$ (strong coupl.)

$La_{1-x} Pr_x Os_4 Sb_{12}$:



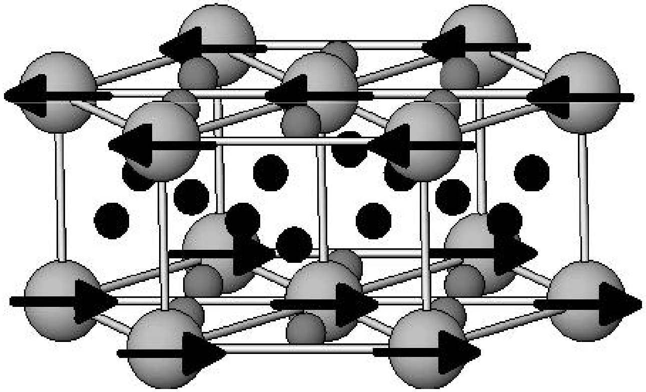
$Pr (Os_{1-x} Ru_x)_4 Sb_{12}$: CEF splitting increases with x

⟹ T_c decreases in agreement with experiment

UPd_2Al_3

SC with $T_c = 1.8$ K AF with $T_N = 14.3$ K $\mathbf{Q} = \left(0, 0, \frac{\pi}{c}\right)$

heavy quasiparticles : $C = \gamma T$; $\gamma = 0.12$ J/mol K^2



from LDA : $n_{5f} \cong 2.5$ $\Longrightarrow$ 50 % $5f^2$ and 50 % $5f^3$

Dual Model of 5f electrons

(G. Zwicknagl, A. Yaresko, P.F., 2002)

consider $5f^2$: $j = 5/2$ combined to $J = 4, 2, 0$

$U_{J=2} – U_{J=4} = 1.1$ eV $U_{J=0} - U_{J=4} = 2.8$ eV

→ $\Delta U > t$

$5f^3$ → $5f^2$: f^2 must be able to form $J = 4$ (Hund's rule) state

→ some of the hybridization matrix element renormal. to zero

→ electrons in some orbitals remain localized

can be studied by microscopic model calculations

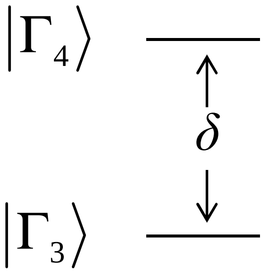
Multiplet structure of $5f^2$ system:

only electrons in $|j_z = \pm 3/2\rangle$ are delocalized; $t_{5/2}$ and $t_{1/2}$ renormalized to zero

6 states from $|j_z = \pm 5/2\rangle$ and $|j_z = \pm 1/2\rangle$

6 x 6 matrix: doubly degenerate ground state with $J_z = \pm 3, \quad J = 4$

CEF lifts degeneracy ⟶ $|\Gamma_3\rangle$ and $|\Gamma_4\rangle$



$$|\Gamma_4\rangle = \frac{1}{\sqrt{2}}\left(|+3\rangle + |-3\rangle\right)$$

$$|\Gamma_3\rangle = \frac{1}{\sqrt{2}}\left(|+3\rangle - |-3\rangle\right)$$

coupling of $|\Gamma_3\rangle$ ⟶ $|\Gamma_4\rangle$ excitations via conduction electrons

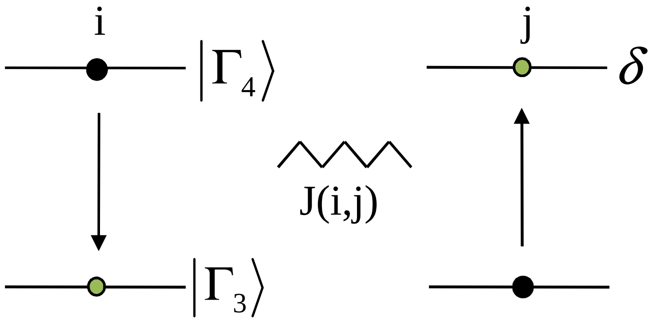
⟶ magnetic excitons

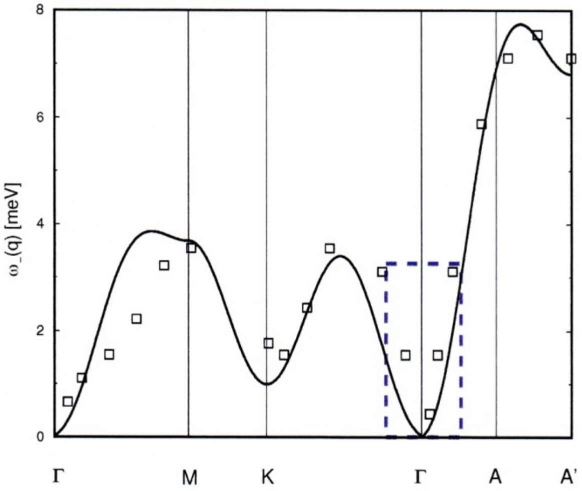
Magnetic excitations:

$H_{CF} = \delta \sum_i |\Gamma_4\rangle\langle\Gamma_4|_i$ (intra-atomic excit.)

plus intersite interaction $J(ij)\, J_i\, J_j$

plus coupling to conduction electrons: RKKY





induced AF: $\mathbf{Q} = (0, 0, 1/2)$

approxim.: $\omega_E(\mathbf{q}) = \omega_{ex}\,[1 + \beta \cos(cq_z)]$

$\omega_{ex} = 5\text{meV}$, $\beta = 0.8$

when averaged over q_x, q_y

exp: A. Mason + G. Aeppli
theory: P. Thalmeier

superconductivity due to intra-atomic excitations

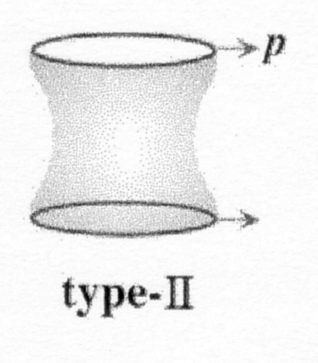
boson : $$K(q_z,\omega) = \frac{I^2}{2}\frac{\omega_{ex}}{\omega_{\mathbf{q}}^2 - \omega^2}$$ I = coupl. const. cond. electrons with CEF states

search for solutions $\Delta(q_z)$

$$\omega_{\mathbf{q}} = \omega_{ex}\left[1 + \beta \cos cq_z\right]$$

solution of Eliashberg equations

$$\Delta(\mathbf{p}) = \Delta\cos(cp_z) \qquad \text{or} \qquad \Delta(\mathbf{p}) = \underset{\substack{\uparrow \\ \text{discard}}}{\Delta}\sin(cp_z)$$



anisotropic thermal conduct. in applied field

→ nodal structure

(Watanabe et al.)

with $I = 0.16\ \mathrm{eV}$ and $N(E_F) \cong 1 \frac{\mathrm{state}}{\mathrm{eV} \cdot \mathrm{uc}}$ → $T_c = 3\mathrm{K}$

scattering of conduct. electrons: no time reversal symmetry

→ no s-wave superconduct.

but : if $\Delta(\mathbf{p})$ changes sign pairing is possible

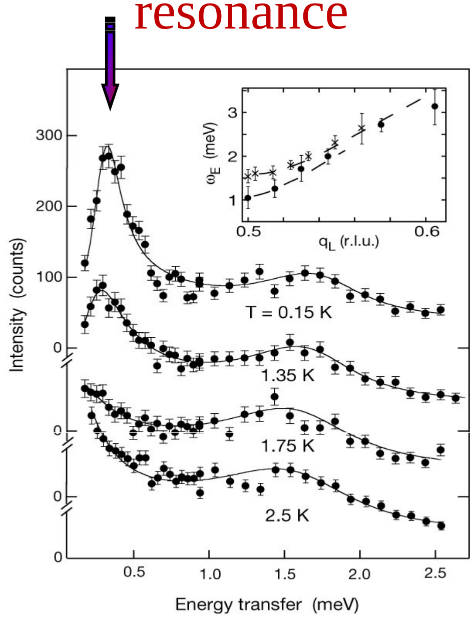
at the same time: $m^* / m_b \cong 10$ from $\frac{m^*}{m_b} = 1 - \frac{\partial \Sigma}{\partial \omega}\Big|_{\omega=0}$

	m* (exp)	**m* (theory)**
ζ	65	59.6
γ	33	31.9
β	19	25.1
ε2	18	17.4
ε3	12	13.4
α	5.7	9.6

no adjustable parameter

theory: G. Zwicknagl exp.: Inada et al. (1999)

Magnetic resonance can be understood too!

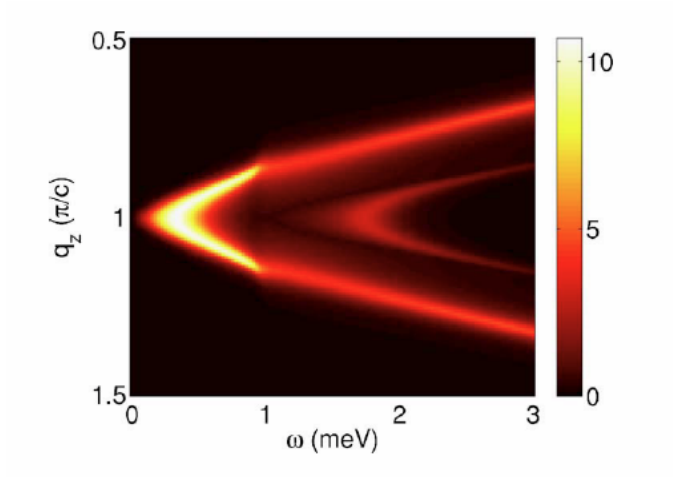

resonance

$$K(\mathbf{q},\omega) = \frac{I^2}{2}\frac{\omega_{ex}}{\omega^2 - \omega_{\mathbf{q}}^2 + 2g^2\Delta_{CEF}\,\mathrm{Re}\,\chi_0(\mathbf{q},\omega)}$$

$$\rightarrow \quad \omega^2 = \omega_{\mathbf{q}}^2 - 2g^2\Delta_{CEF}\,\mathrm{Re}\,\chi_0(\mathbf{q},\omega)$$

new pole due to sc. $\chi_0(\mathbf{q},\omega)$

$\mathrm{Im}\,K(q_z,\omega)$ as measured by INS



Conclusions

- intra-atomic low energy excitations (CEF) can result in Cooper pairing
- in the filled skutterudite $PrOs_4Sb_{12}$ quadrupolar excitations contribute more than 50 % to Cooper-pair formation
- in UPd_2Al_3 superconductivity, the magnet. resonance below T_c and the strong anisotropic mass enhancements can be explained well within the Dual Model