Orbital-exchange and fractional spinon excitations in an f-electron metal Yb_2Pt_2Pb

Igor Zaliznyak

BNL CMPMS Neutron Scattering Workshop: Beyond integrability.

Montreal, 2015

BROOKHAVEN NATIONAL LABORATORY

a passion for discovery

Office of Science
U.S. DEPARTMENT OF ENERGY

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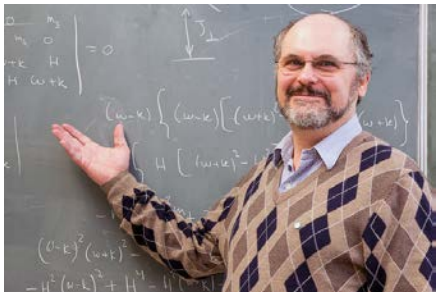

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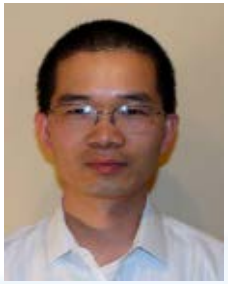

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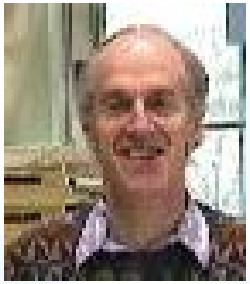

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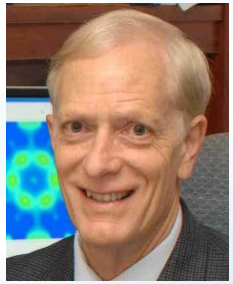

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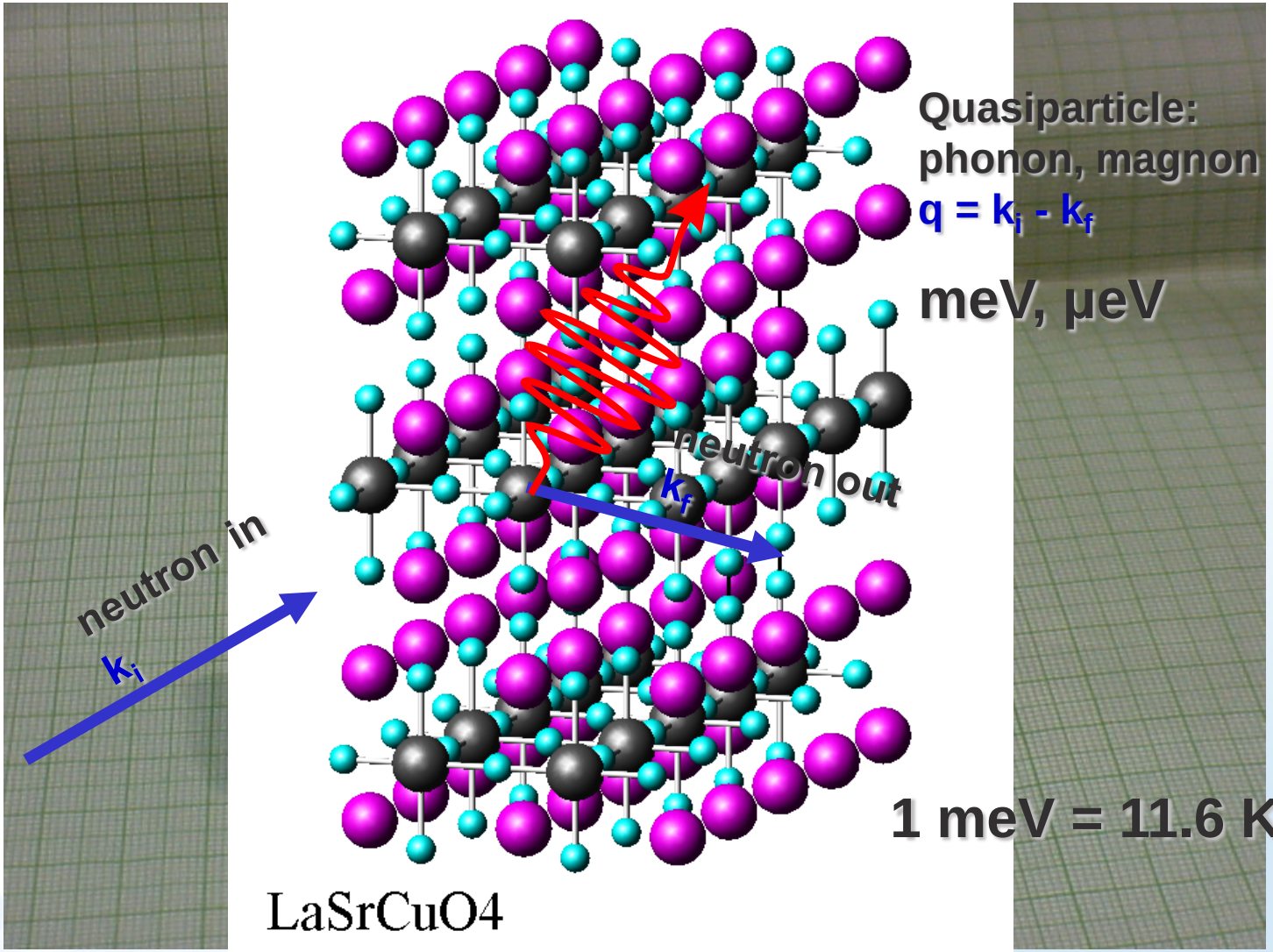
Quasiparticles in condensed matter



The scattering experiment

$$\frac{d^2\sigma(\mathbf{q},E)}{dEd\Omega} = \frac{1}{\Phi_i(\mathbf{k}_i)}\frac{\delta J_f(\mathbf{k}_f)}{dE_f d\Omega_f}$$

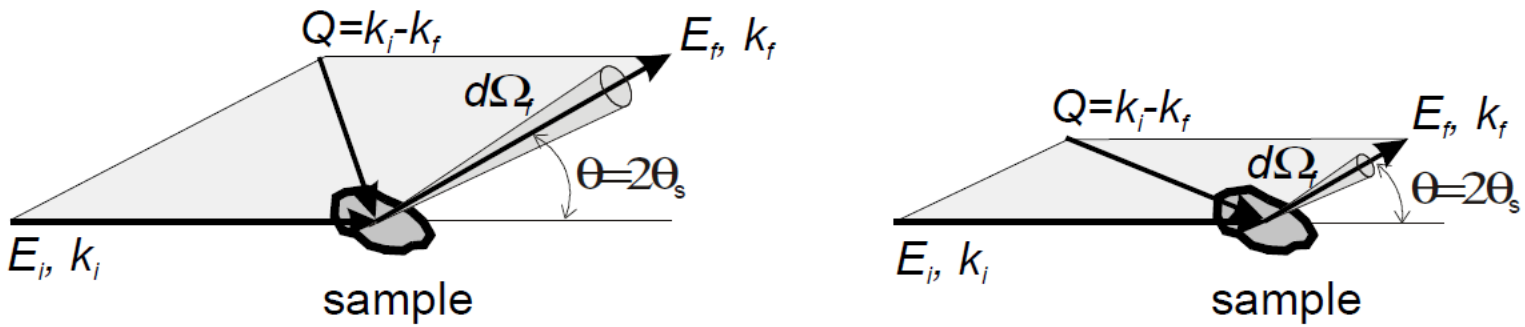


Figure 2-1. Typical geometry of a scattering experiment, (a) elastic, (b) inelastic.

$$\delta J_f(k_f) = \Gamma_{i\to f}\frac{d^3k_f}{(2\pi)^3}$$

$$\Gamma_{i\to f} = \frac{2\pi}{\hbar}\left|\left\langle \mathbf{k}_f, S_f^z, \eta_f \middle| \mathbf{T} \middle| \mathbf{k}_i, S_i^z, \eta_i \right\rangle\right|^2 \delta\left(E_i - E_f + \frac{(\hbar k_i)^2}{2m} - \frac{(\hbar k_f)^2}{2m}\right)$$

$$E = E_f(\eta_f) - E_i(\eta_i) = \frac{(\hbar k_i)^2}{2m} - \frac{(\hbar k_f)^2}{2m}$$

$$\boldsymbol{q} = \boldsymbol{k}_i - \boldsymbol{k}_f$$

Neutron interactions and scattering cross-section: precisely known!

$$\mathbf{T} = \mathbf{V} + \mathbf{TGV}$$

Lippmann&Schwinger, Phys. Rev. 79 (1950)

$$\mathbf{T} = \mathbf{V} + \mathbf{VGV} + \mathbf{VGVGV} + \ldots.$$

$$\mathbf{b} = -\frac{m}{2\pi\hbar^2}\mathbf{T}$$

$$\frac{d^2\sigma(\mathbf{q},E)}{dEd\Omega} = \sum_{i,f}\frac{k_f}{k_i}\left|\left\langle S_f^z,\eta_f\left|\mathbf{b}(-\mathbf{q})\right|S_i^z,\eta_i\right\rangle\right|^2\delta\left(E_i(\eta_i)-E_f(\eta_f)+\frac{(\hbar k_i)^2}{2m}-\frac{(\hbar k_f)^2}{2m}\right)$$

$$\mathbf{b}_N = b_N\delta(\mathbf{r}_n - \mathbf{R})$$

$$\mathbf{V}_N = -\frac{m}{2\pi\hbar^2}b_N\delta(\mathbf{r}_n - \mathbf{R})$$

nuclear scattering length, $b \sim 10^{-13}$ cm

Strong interaction, but extremely short-range: Fermi pseudo-potential, not Born approximation!

$$\mathbf{V}_{mag} = -\left\{\frac{8\pi}{3}(\boldsymbol{\mu}_n\boldsymbol{\mu}_e)\delta(\mathbf{r}) - \frac{(\boldsymbol{\mu}_n\boldsymbol{\mu}_e)}{r^3} + \frac{3(\boldsymbol{\mu}_n\mathbf{r})(\boldsymbol{\mu}_e\mathbf{r})}{r^5}\right\}$$

magnetic scattering length, $r_m = -5.39*10^{-13}$ cm

Weak interaction: Born approximation is OK!

How to measure quasiparticle excitations: Neutron Scattering.

Long-lived quasiparticle ➔ delta function singularity in cross-section

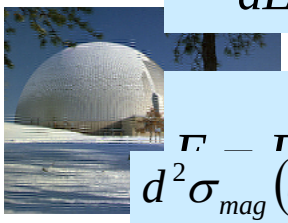
$$\frac{d^2\sigma_{nuc}(\boldsymbol{q},E')}{dEd\Omega} = \frac{k_f}{k_i}\sum_{j,j'}\left(b_j b_{j'}^*\right)\int_{-\infty}^{\infty} e^{-\frac{i}{\hbar}Et}\left\langle e^{-i\boldsymbol{q}\cdot\boldsymbol{R}_{j'}(0)} e^{i\boldsymbol{q}\cdot\boldsymbol{R}_j(t)}\right\rangle \frac{dt}{2\pi\hbar}\left(1+\frac{(\hbar k_i)^2}{2m}-\frac{(\hbar k_f)^2}{2m}\right)$$

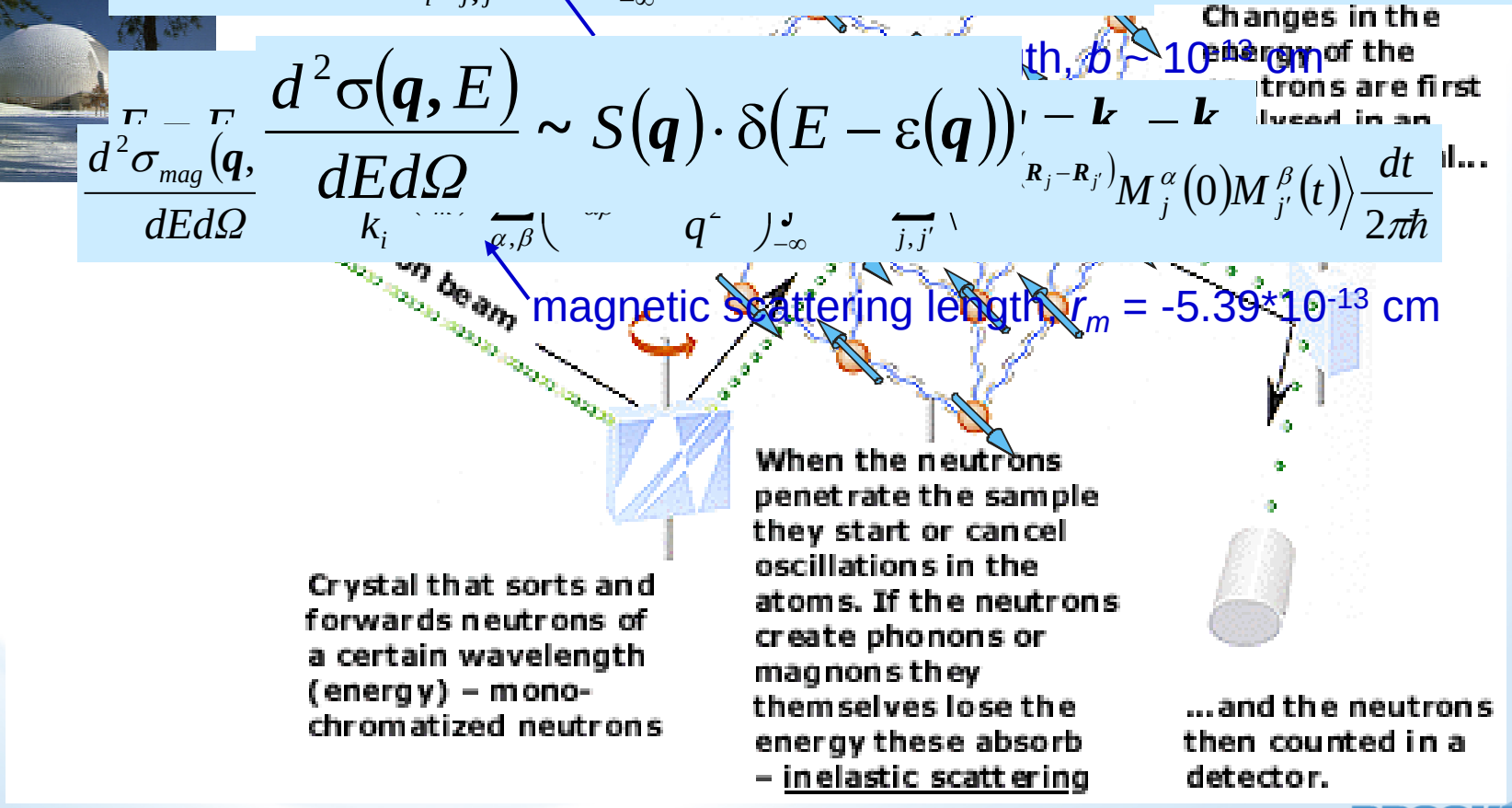
$$\frac{d^2\sigma(\boldsymbol{q},E)}{dEd\Omega} \sim S(\boldsymbol{q})\cdot\delta(E-\varepsilon(\boldsymbol{q}))$$

$$\frac{d^2\sigma_{mag}(\boldsymbol{q},}{dEd\Omega} \quad k_i \quad \sum_{\alpha,\beta} \quad q^{\alpha} \quad \int_{-\infty} \quad \sum_{j,j'} \quad \left(\boldsymbol{R}_j-\boldsymbol{R}_{j'}\right) M_j^{\alpha}(0) M_{j'}^{\beta}(t)\Big\rangle \frac{dt}{2\pi\hbar}$$

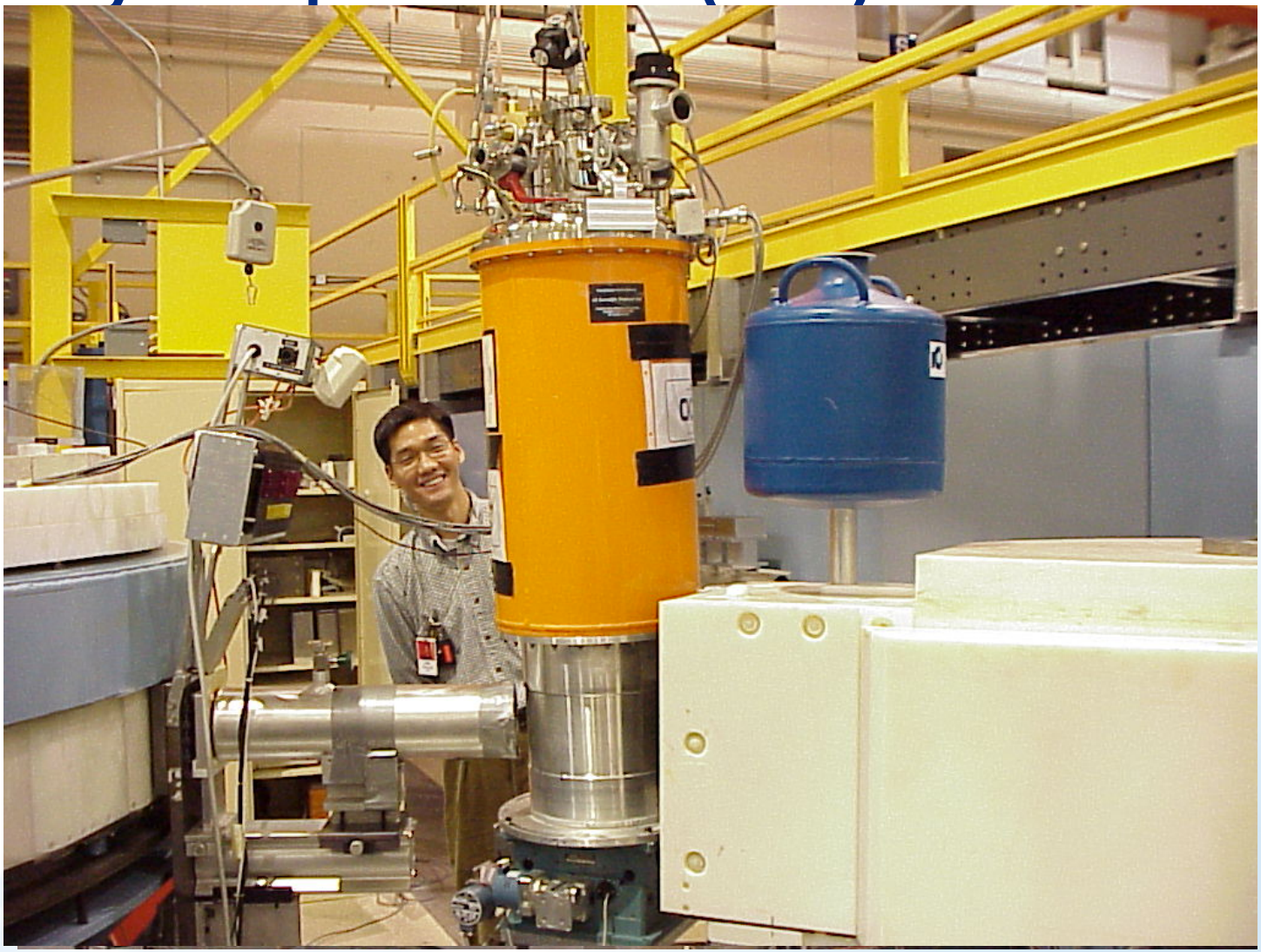
th, b ~ 10^{-13} cm

magnetic scattering length, r_m = -5.39*10^{-13} cm



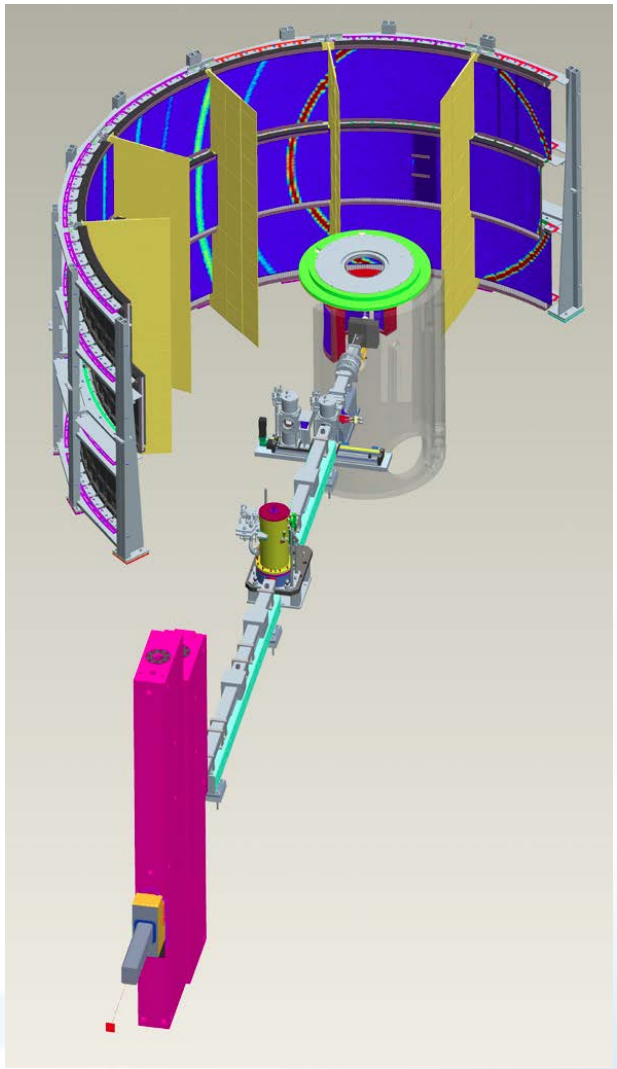


How do neutrons measure excitations: triple-axis crystal spectrometer (TAS)

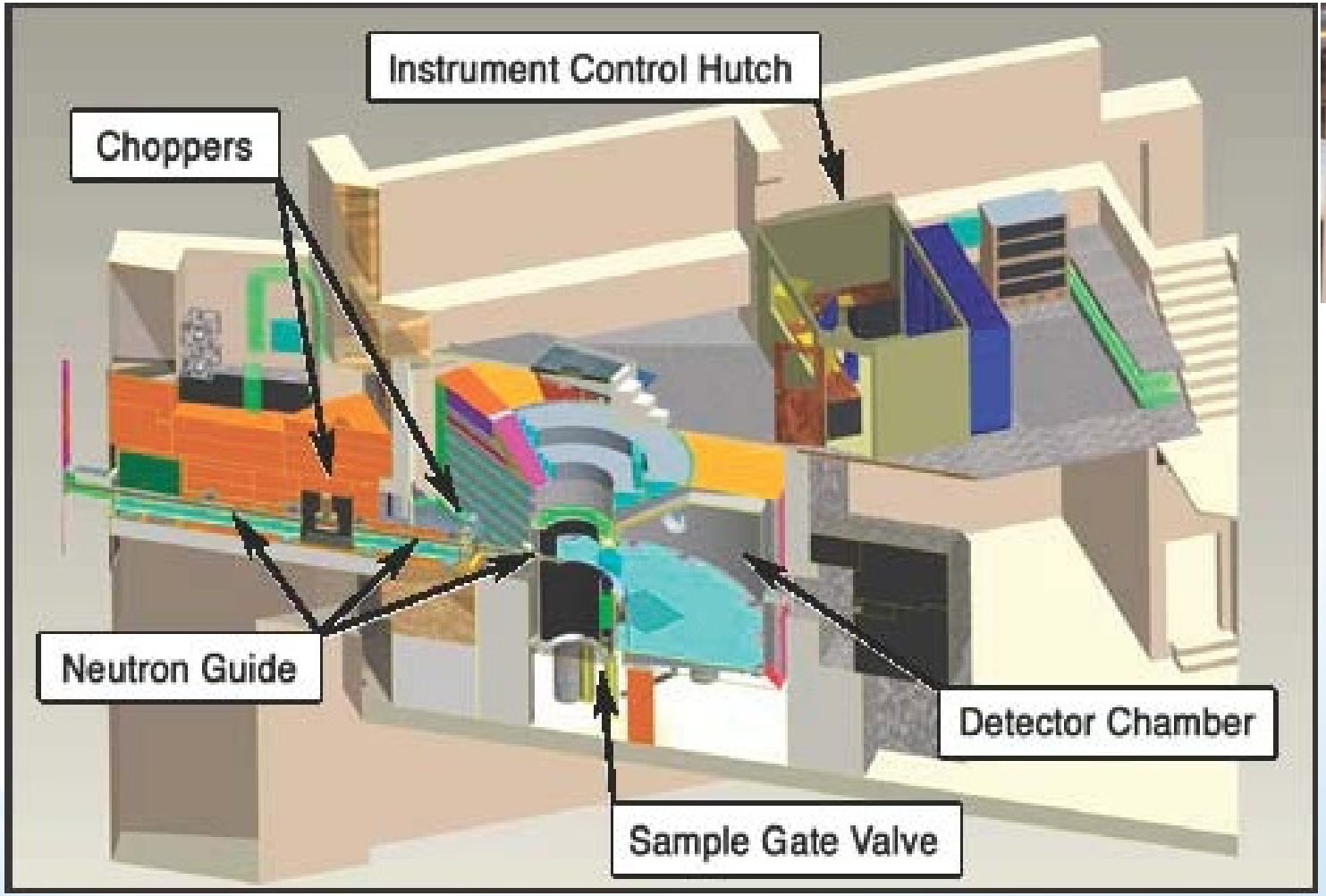


How do neutrons measure excitations: time-of-flight (TOF) direct geometry spectrometer (DGS)

How do neutrons measure excitations TOF DGS @SNS (ARCS)

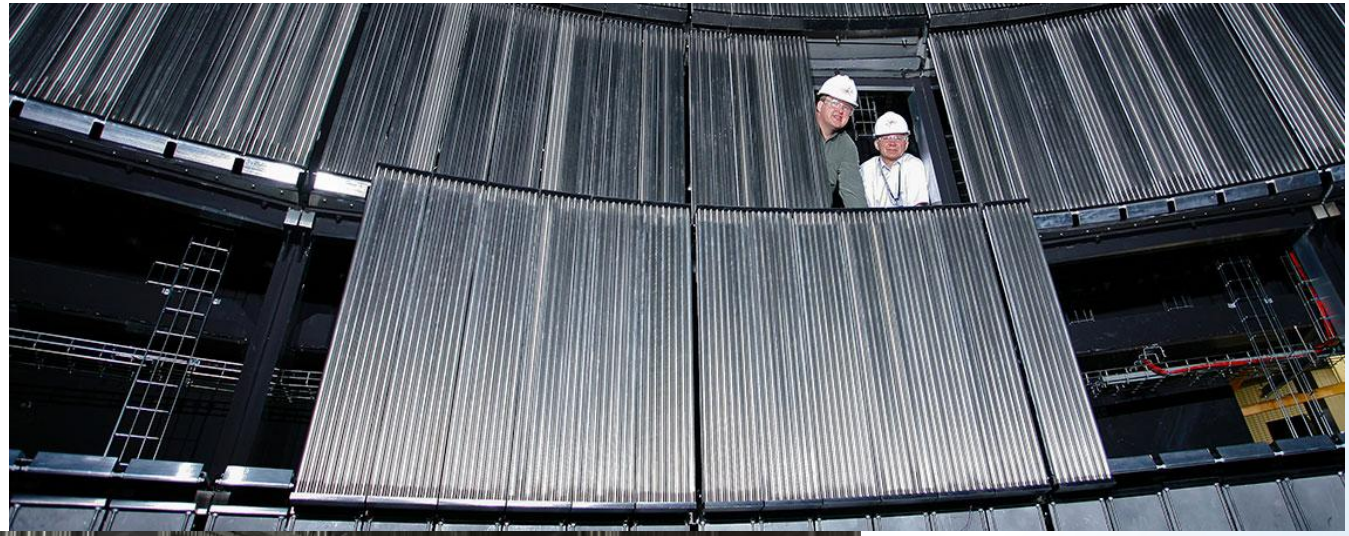


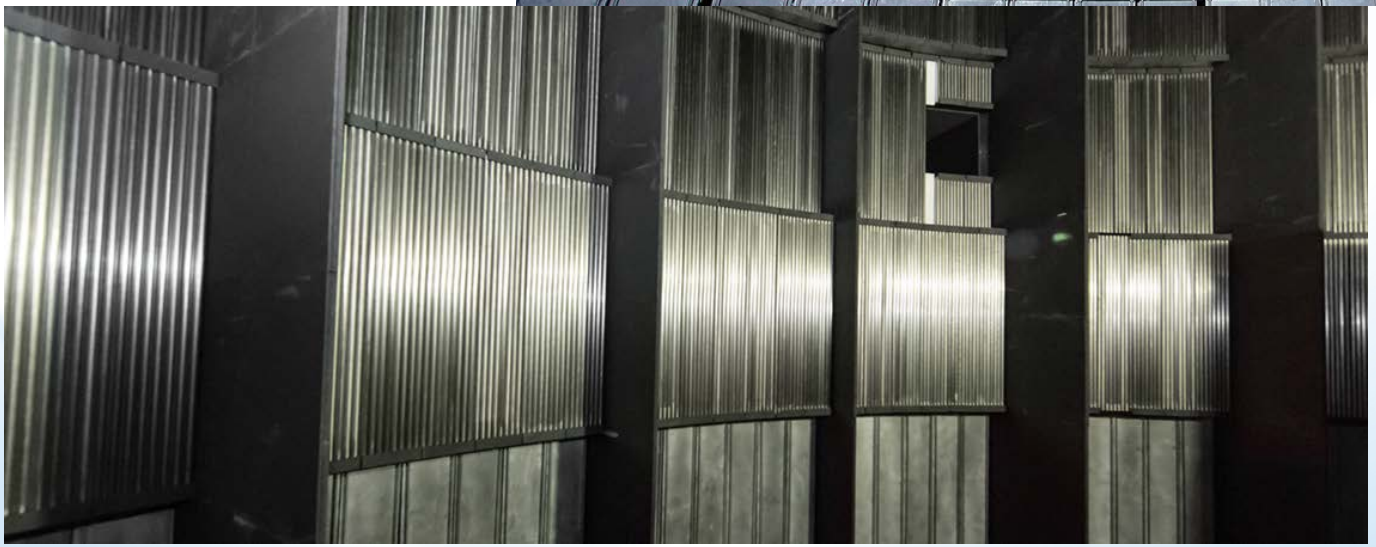
How do neutrons measure excitations TOF DGS ARCS@SNS



How neutrons measure excitations TOF DGS SEQUOIA@SNS

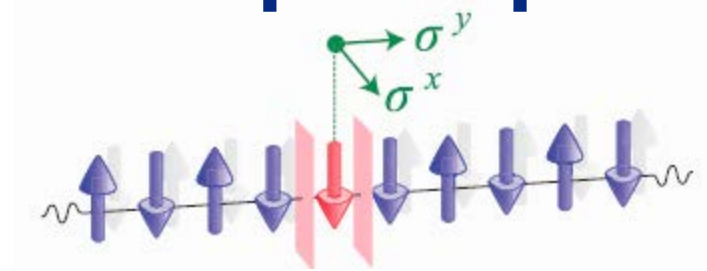
Cylindrical geometry
Sample-to-detector distance
5.5–6.3 m
Incident energy range
4–2000 meV
Resolution (elastic)
1–5% E_i
Vertical detector coverage
-18–18˚
Horizontal detector coverage
-30–60˚

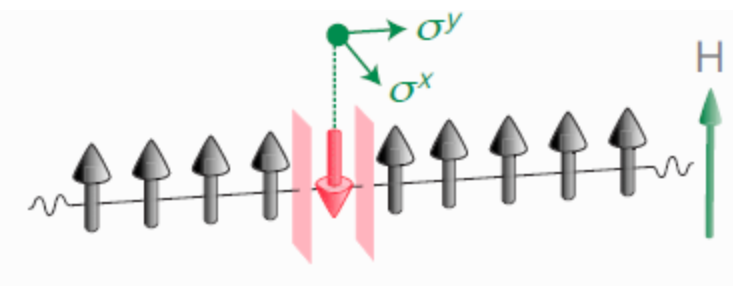


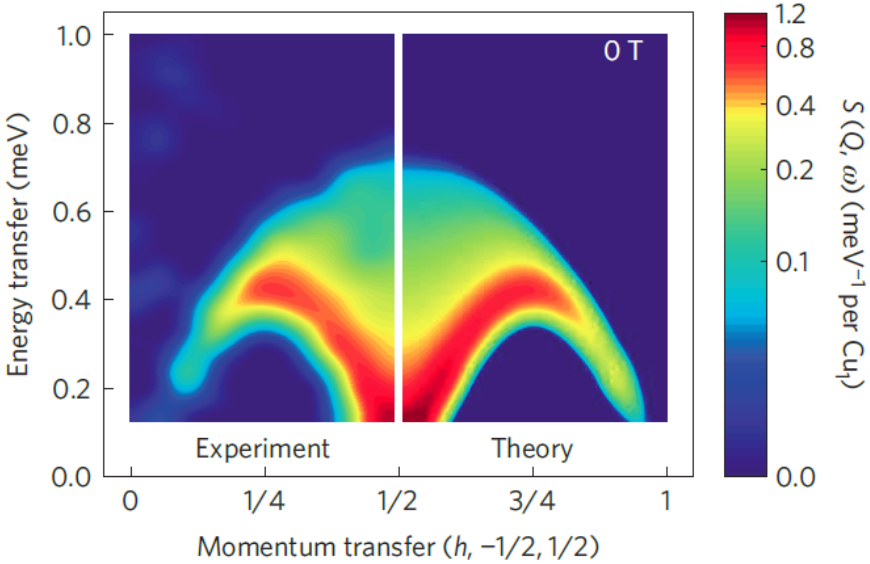


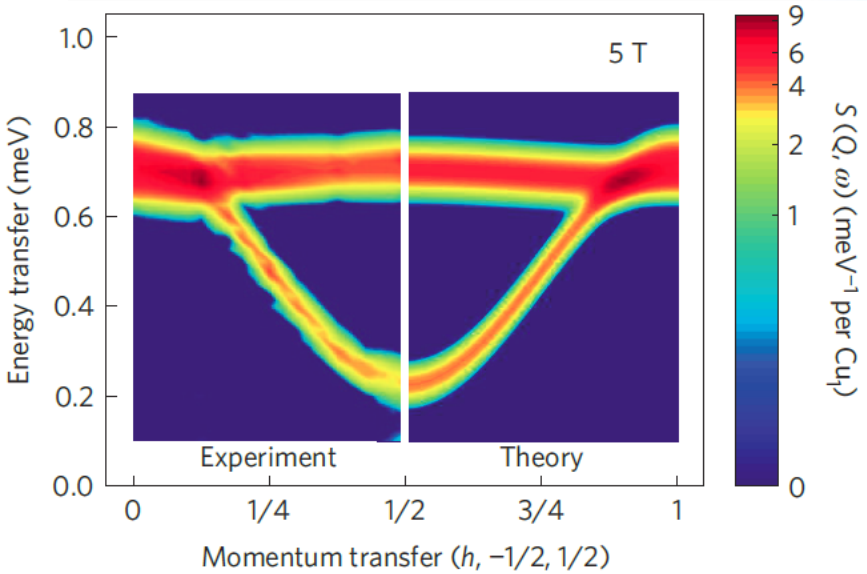
Simply an enormous camera streaming a movie

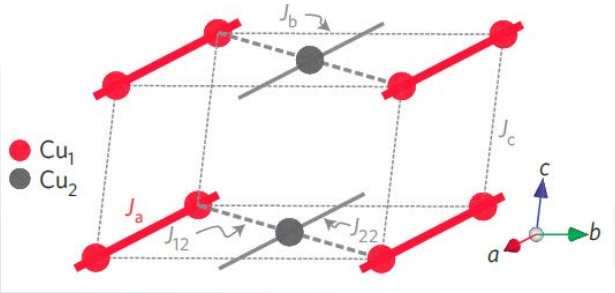
Example: spinons in S=1/2 Cu chains





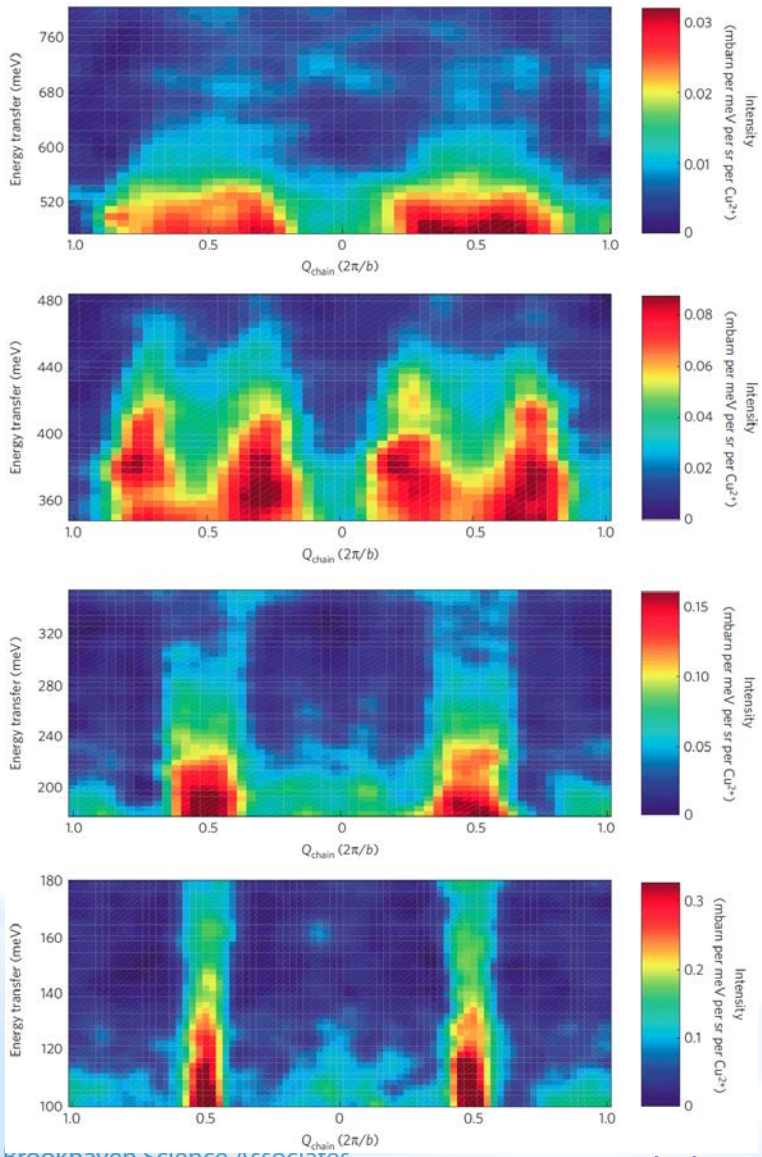




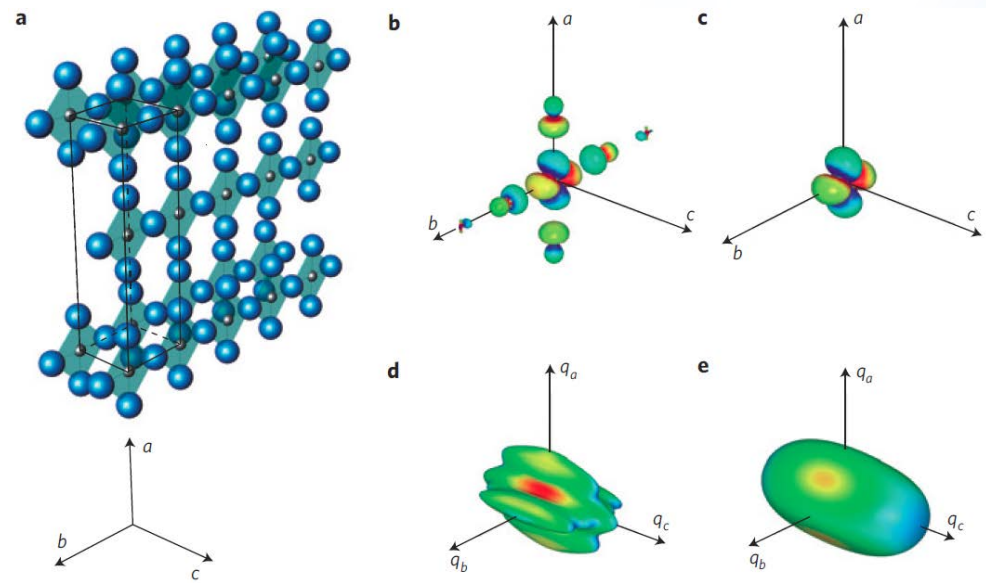


Multi-spinon (B = 0) and magnon (B = 5T) excitations in copper sulphate pentahydrate
Mourigal, et al. Nature Physics (2013)

Example: spinons in S=1/2 Cu chains



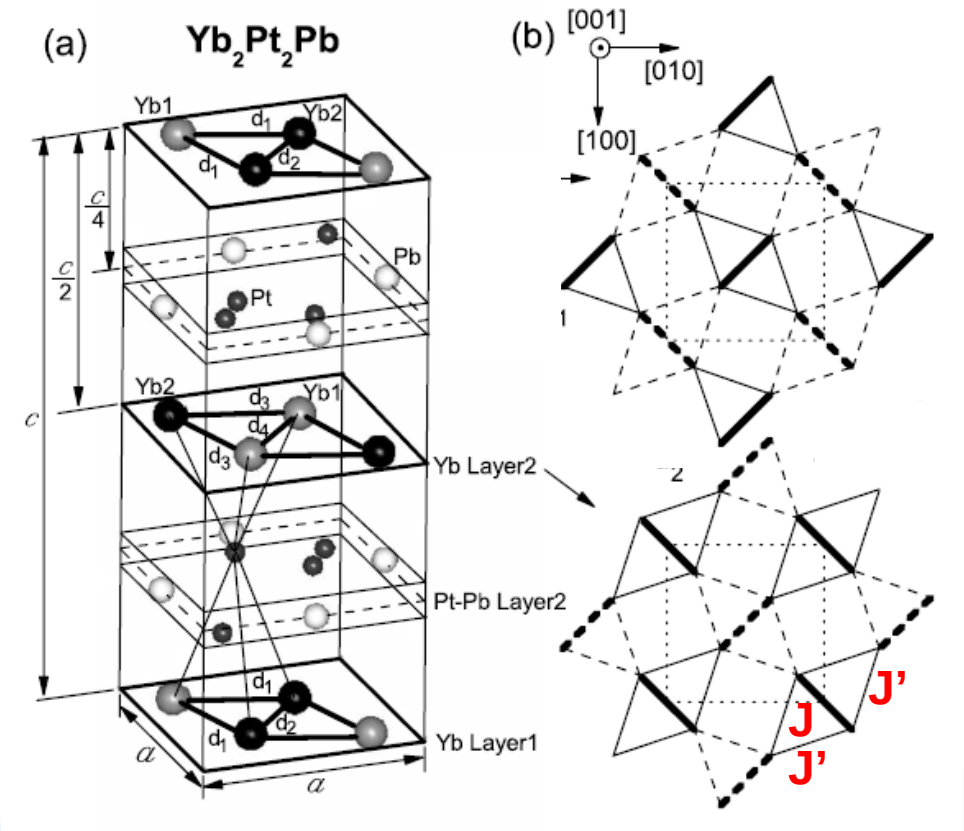
Chains are formed by particular orbital overlap patterns, which define the superexchange.



Multi-spinon excitations in Sr_2CuO_3
Walters, Perring, Caux, Savici, Gu, Lee, Ku, Zaliznyak, Nature Physics (2009)

Yb_2Pt_2Pb: f-electron Ising dimers on a Shastry-Sutherland lattice?

Kim, Benett, Aronson, PRB (2008); PRL (2013)



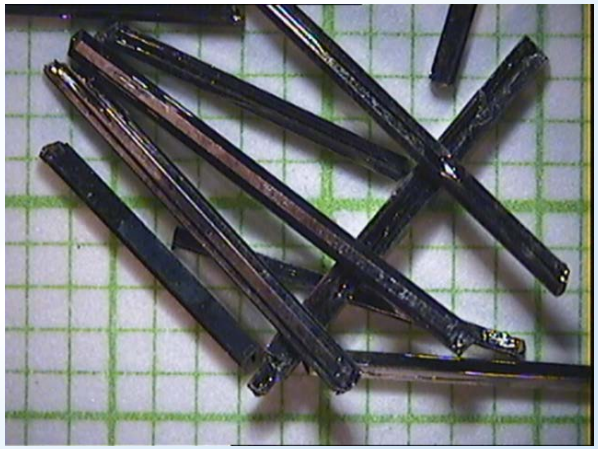
Dimers on the Shastry-Sutherland lattice

J=intradimer coupling,
J'=interdimer coupling

Dimer Liquid ★ Magnetic Order

J'/J

QCP

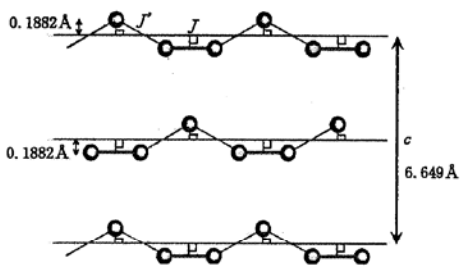


- Tetragonal $P4_2/mmm$: U_2Pt_2Sn type (Pöttgen 1999)
- Other Shastry – Sutherland Lattice Systems:
  - RB_4 (R=Pr,Gd,Tb,Dy,Ho,Er, Tm): magnetically ordered metals, Ising moments.
  - $SrCu_2(BO_3)_2$: Insulator, Cu S=1/2, spin-liquid (B=0), AF order (B>34 T).

Excitations of a S=1/2 Shastry-Sutherland dimers: $SrCu_2(BO_3)_2$

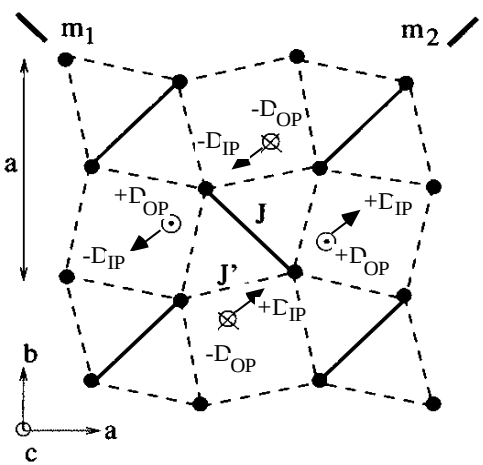
Kakurai, et. al, Prog. Theor. Phys. Suppl. (2005)

a)

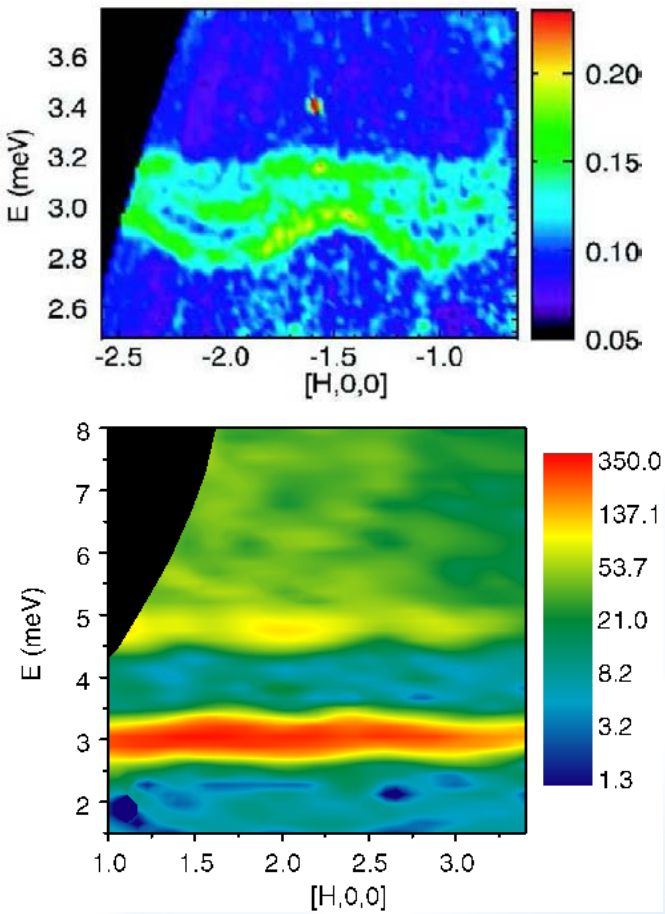


$J'/J \approx 0.63 < (J'/J)_c$

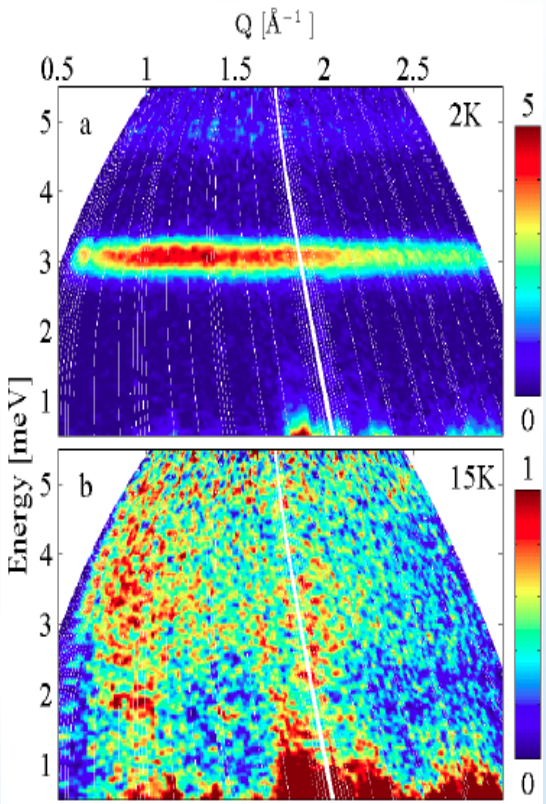
b)



Gaulin, et. al, PRL (2004)



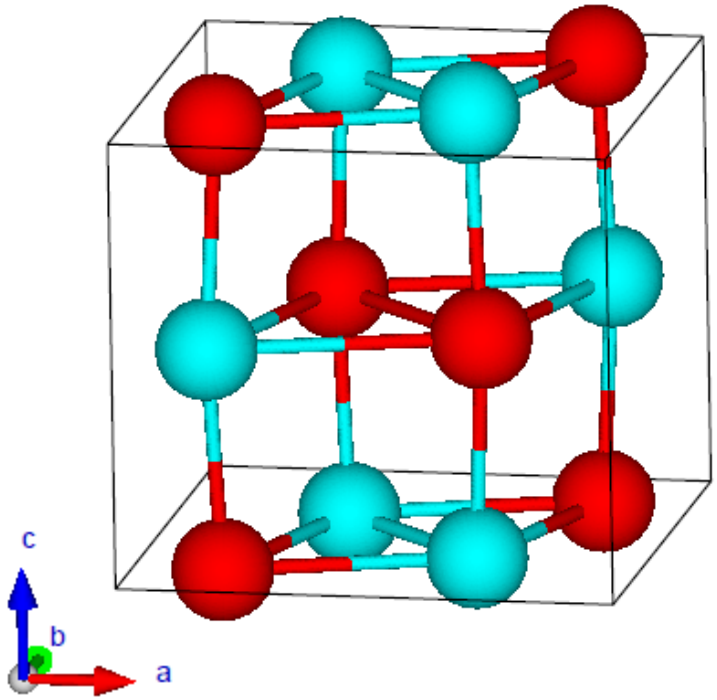
Zayed, et. al, PRL (2012)

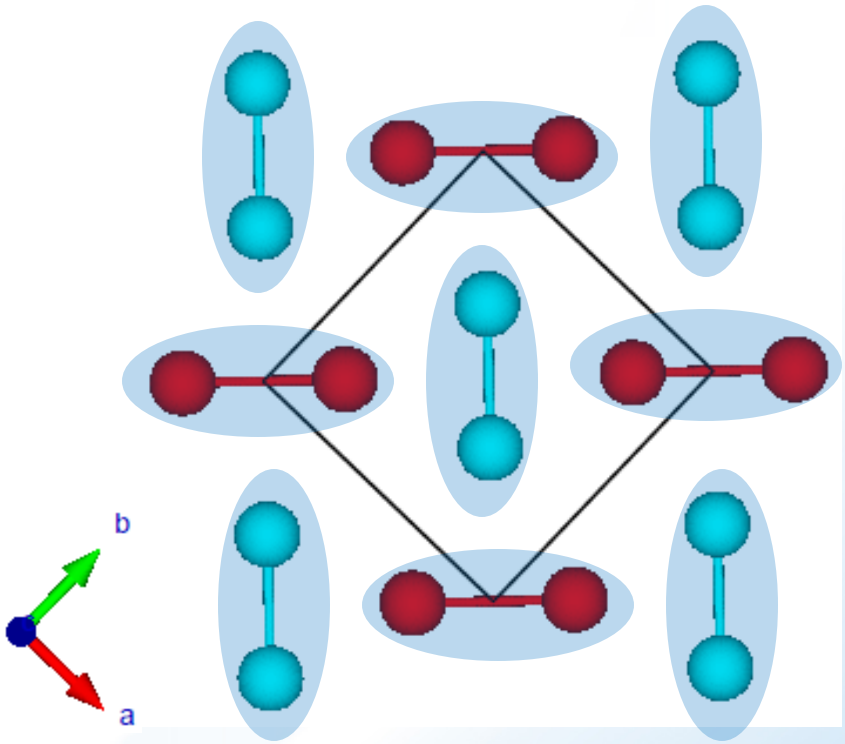


- gapped triplet magnon with flat dispersion – propagates only in high order of perturbation in J'
- fine energy-structure from low-symmetry perturbations – DM, staggering, …

Yb_2Pt_2Pb crystal structure: f-electron Ising dimers on a Shastry-Sutherland lattice?

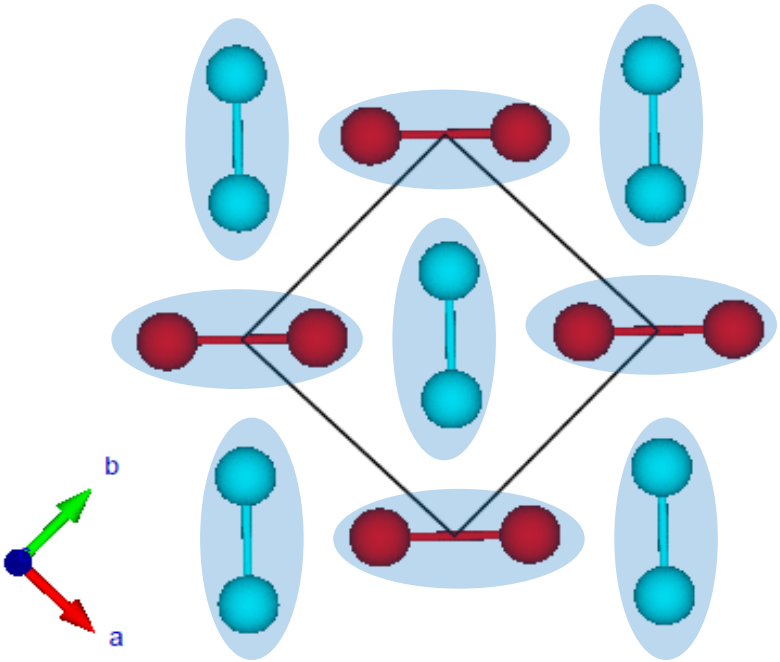
M.S. Kim et al., 2008, 2012

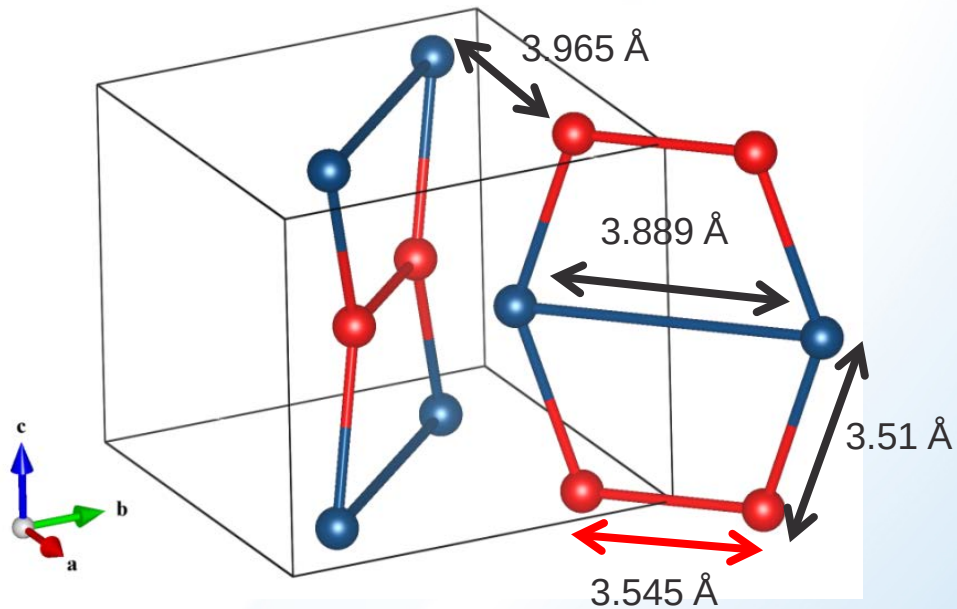




- Two Yb sites form orthogonal pairs => SSL
- 2D SSL physics: dimers, singlet ground states?
- Neutron scattering: singlet ground state - triplet magnon excitation?

Yb_2Pt_2Pb: f-electron Ising dimers on a Shastry-Sutherland lattice?





- Weakly coupled layers with Shastry-Sutherland lattice ? Two dimensional magnetic system?
- Spin ladders with rungs in Shastry-Sutherland lattice layers? One dimensional magnetic system?

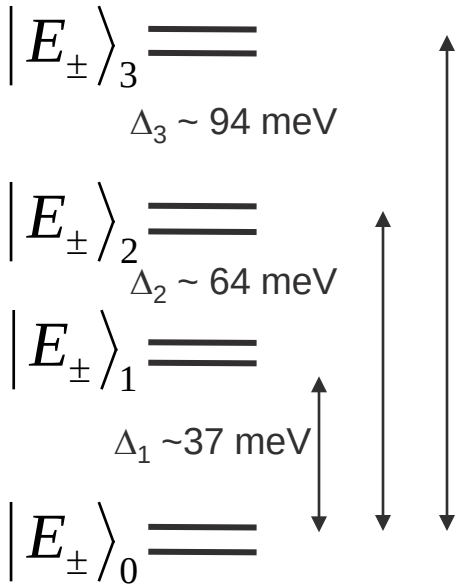
$d_{chain}<d_{rung}(red)<d_{rung(}blue)<d_{inter\text{-}ladder}$

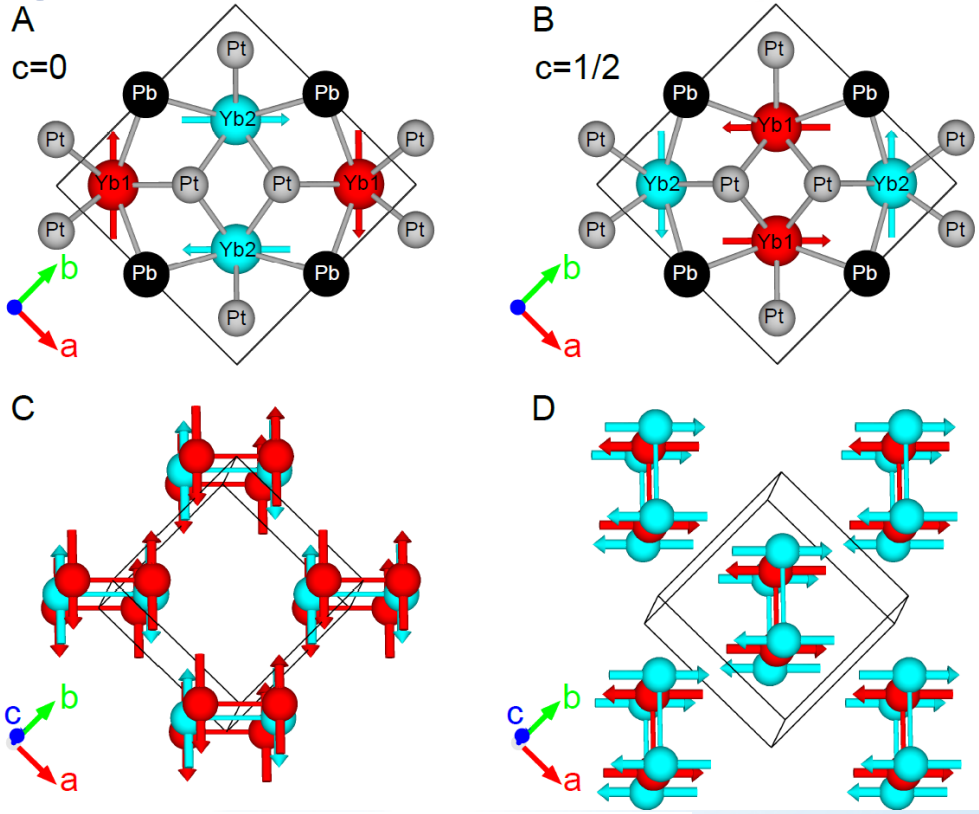
$J_{chain}>J_{rung}>J_{inter\_ladder}$

} **Weakly coupled spin ladders?**

Yb_2Pt_2Pb CEF point charge model calculation: Ising moments

Yb^{3+}: CEF Levels





$$|E_\pm\rangle_0 = 0.992|\pm 7/2\rangle + 0.100|\pm 3/2\rangle + 0.064|\mp 1/2\rangle + 0.032|\mp 5/2\rangle,$$
$$|E_\pm\rangle_1 = -0.082|\pm 7/2\rangle + 0.300|\pm 3/2\rangle + 0.355|\mp 1/2\rangle + 0.881|\mp 5/2\rangle$$
$$|E_\pm\rangle_2 = 0.092|\pm 7/2\rangle + 0.785|\pm 3/2\rangle + 0.420|\mp 1/2\rangle - 0.446|\mp 5/2\rangle,$$
$$|E_\pm\rangle_3 = -0.153|\pm 7/2\rangle + 0.832|\pm 3/2\rangle - 0.533|\mp 1/2\rangle + 0.005|\mp 5/2\rangle$$

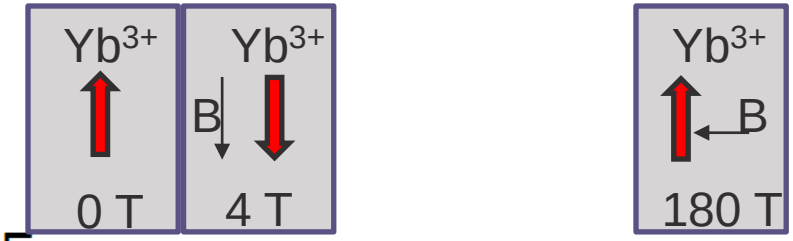
Wu, et. al, unpublished (2015)

Yb_2Pt_2Pb: effective spin-1/2 description of the G. S. doublet

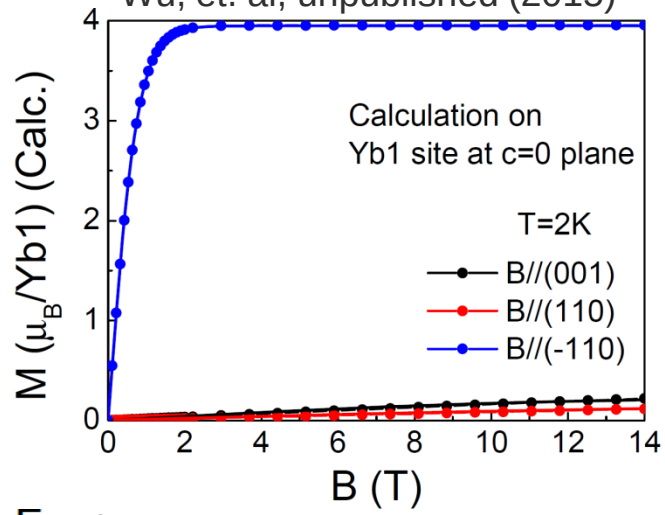
$$M^{\alpha} = g_{eff}^{\alpha}\mu_B S^{\alpha}, \quad \left(S^{\alpha}\right)^2 = \frac{1}{4}$$

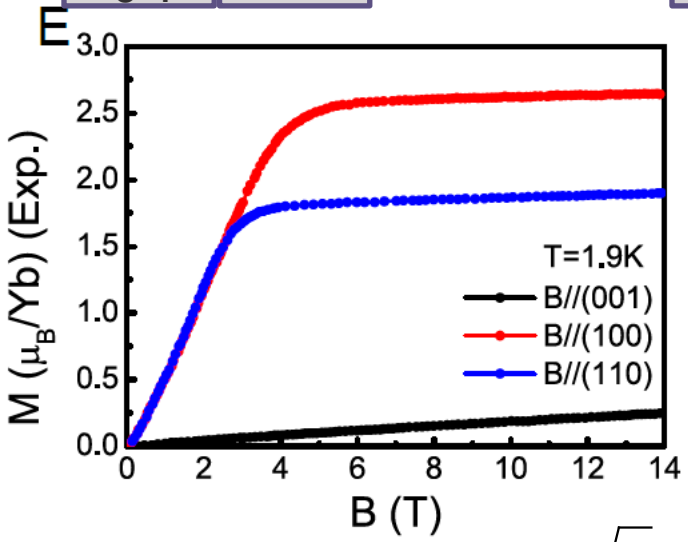
$$g_{eff}^{z} = 7.9 = g_{eff}^{(110)}(Yb1) = g_{eff}^{(-110)}(Yb2)$$

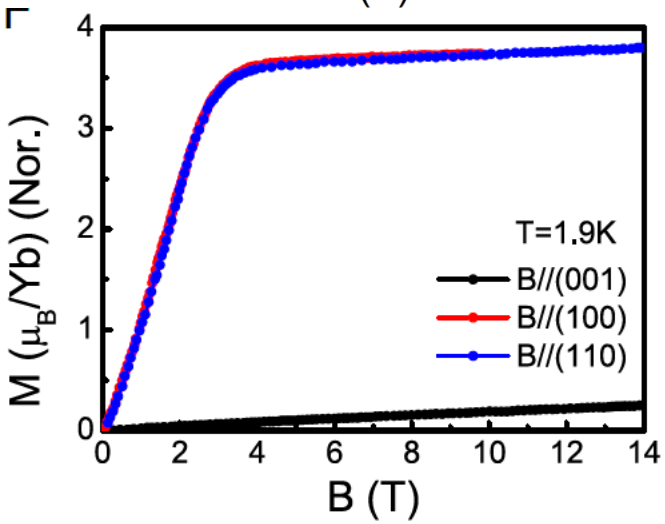
$$g_{eff}^{x,y} \le 0.8$$



Wu, et. al, unpublished (2015)







$$M_{100} \approx 2.6\mu_B, \quad M_{110} \approx 3.6\mu_B, \quad M_{100} \approx \sqrt{2}M_{110} >> M_{001}$$

Yb_2Pt_2Pb: effective spin-1/2 description of the G. S. doublet

General projected Hamiltonian:

$$H = \sum_{i \neq j, \alpha, \beta} J_{\alpha\beta} S_i^{\alpha} S_j^{\beta} - \mu_B \sum_{i, \alpha, \beta} H_{\alpha} g^{\alpha\beta} S_i^{\beta}, \quad \mathbf{S}^2 = \frac{3}{4}$$

e. g. Ross, et. al, PRX (2011)

Neutron Scattering intensity:

$$I^{\alpha\alpha} \sim \left(1 - \frac{q_\alpha^2}{q^2}\right) \int_{-\infty}^{\infty} e^{-\frac{i}{\hbar}Et} \left\langle M_{\mathbf{q}}^{\alpha} M_{-\mathbf{q}}^{\alpha}(t) \right\rangle \frac{dt}{2\pi\hbar} \sim g_\alpha^2 \left(1 - \frac{q_\alpha^2}{q^2}\right) \int_{-\infty}^{\infty} e^{-\frac{i}{\hbar}Et} \left\langle S_{\mathbf{q}}^{\alpha} S_{-\mathbf{q}}^{\alpha}(t) \right\rangle \frac{dt}{2\pi\hbar}$$

Sensitive to the longitudinal sector only:

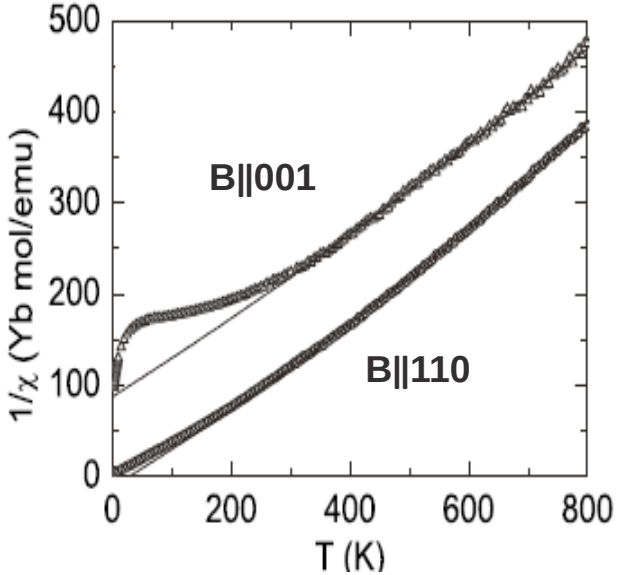
$$I^{zz} / I^{xx} \sim \left(g^{zz} / g^{xx}\right)^2 > 100$$

Sum rule for spin-1/2:

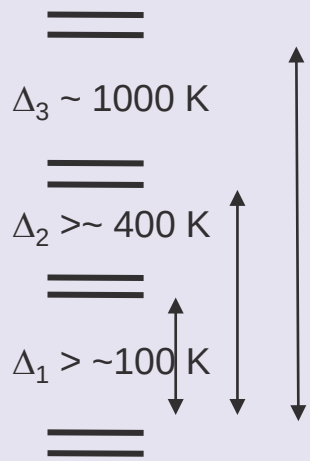
$$\int dE \int \frac{d^n q}{(2\pi)^n} S^{\alpha\alpha}(q, E) = \frac{1}{4}$$

Wu, et. al, unpublished (2015)

Strongly Anisotropic Yb^{3+} moments: Isolated Doublet Ground State



Yb^{3+}: CEF Levels



Yb^{3+}: L=3,S=1/2, J=7/2
Ground State doublet:
$|\pm\rangle=\alpha|\pm 7/2\rangle + \beta|\pm 1/2\rangle + \delta|-+5/2\rangle$

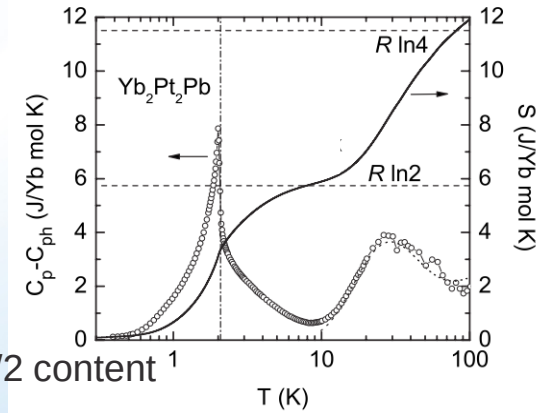
$\langle\pm|J_z|\pm\rangle=\pm\frac{1}{2}(7\alpha^2+\beta^2-5\delta^2) = \pm\frac{1}{2} j_1$
$\langle\pm|J_x|\mp\rangle= \sqrt{7}\,\alpha\delta + 2\beta^2 = \frac{1}{2} j_2$

Strong anisotropy: $\alpha \gg \beta,\delta$ $\quad j_1 \gg j_2$

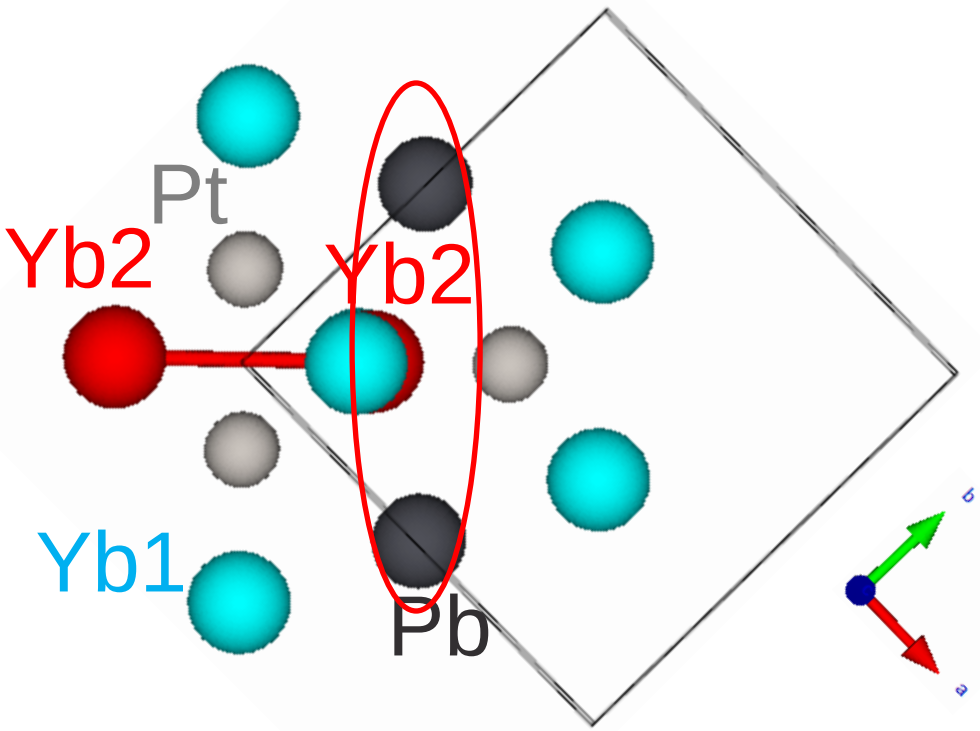
- Curie Weiss Susceptibility T>300 K $\chi=\chi_o + C/(T-\theta)$
  $\mu_{CW}=4.54\ \mu_B/Yb$ (B‖ 001, 110) $\quad \theta_{001}=-217$ K $\quad \theta_{110}=28$ K

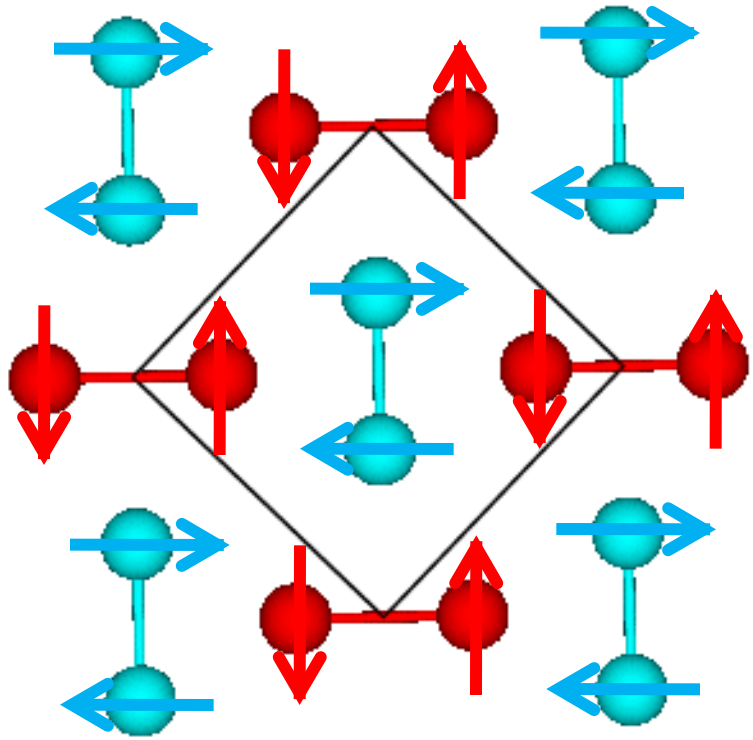
- Specific heat and inelastic neutron scattering confirm CEF level spacings:
  - Doublet ground state is energetically isolated (T < ~100 K).

- Point charge model:
  - a doublet ground state for Yb^{3+} with dominantly (~98%) $J_Z=\pm 7/2$ content
  - an anisotropic Lande g-factor: $g_{eff}^Z = 7.9$, $g_{eff}^x=0.5$
  - for $S_x=S_y=S_z$: $M_{S(110)}=g_{eff}^Z\mu_B S_Z=\pm 4\ \mu_B$ $\quad M_{S(001)}=g_{eff}^x\mu_B S_x=0.25\ \mu_B$



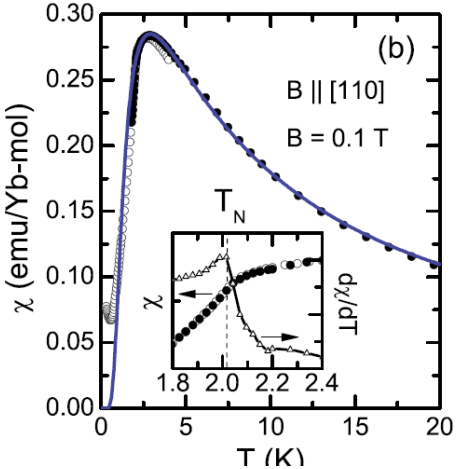
Yb_2Pt_2Pb: Ising dimers on a Shastry-Sutherland lattice?



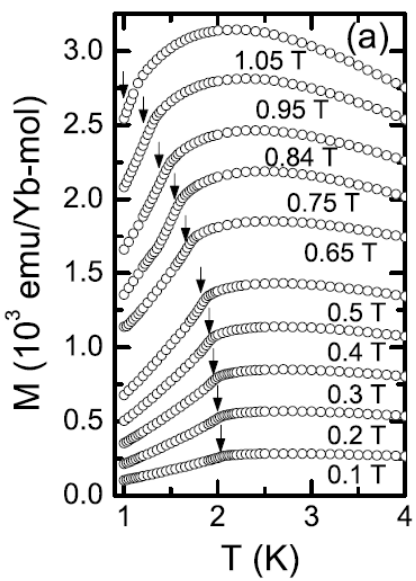


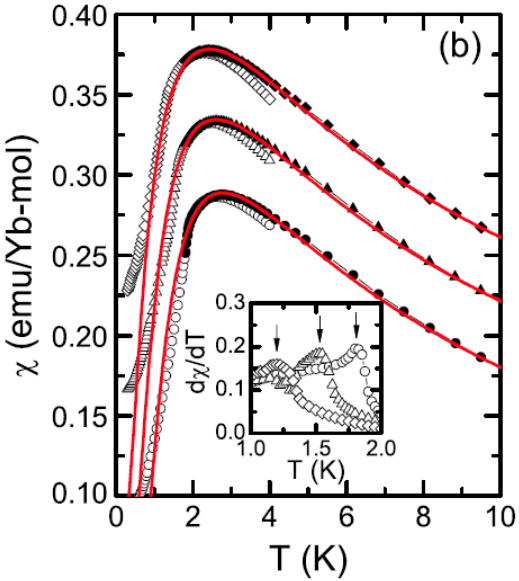
- Strong Spin-Orbit coupling + Crystal Electric field => well-separated ***J***-doublet Ground State: $\left|G.S.\right\rangle = 0.99\left|\pm 7/2\right\rangle + \ldots$
- Yb 4f magnetic moments, $\boldsymbol{\mu}_{Yb} = g_J \mu_B \boldsymbol{J}$ (g_J=8/7) point along (-1, 1, 0) or (1, 1, 0)
- Very strong **Ising** anisotropy, >30meV (> ~300 K, > ~300 T)

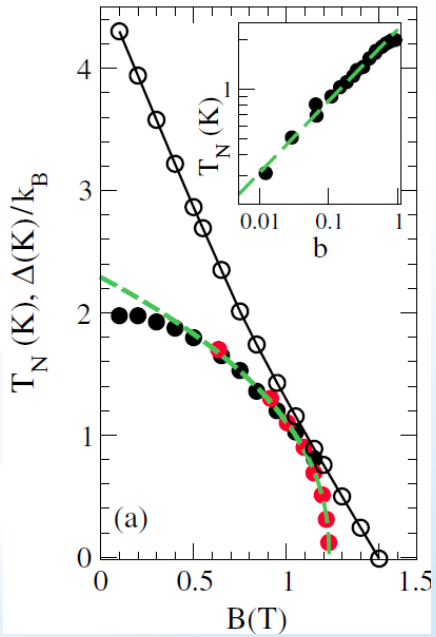
Yb_2Pt_2Pb: Ising dimers on a Shastry-Sutherland lattice?



- Broad peak in χ(T) interpreted as resulting from a gap over the non-magnetic G. S.
  - Order from from a paramagnetic liquid of Yb dimers, having a gap Δ ≈ 4.3 K
  - Gap Δ(T) decreases with the increasing T
- Mean-field-RPA expression for the χ(T) of coupled dimers:
  - J ≈ -2.3 K, J' ≈ -1.95 K, J'/J ≈ 0.85, g ≈ 5.43

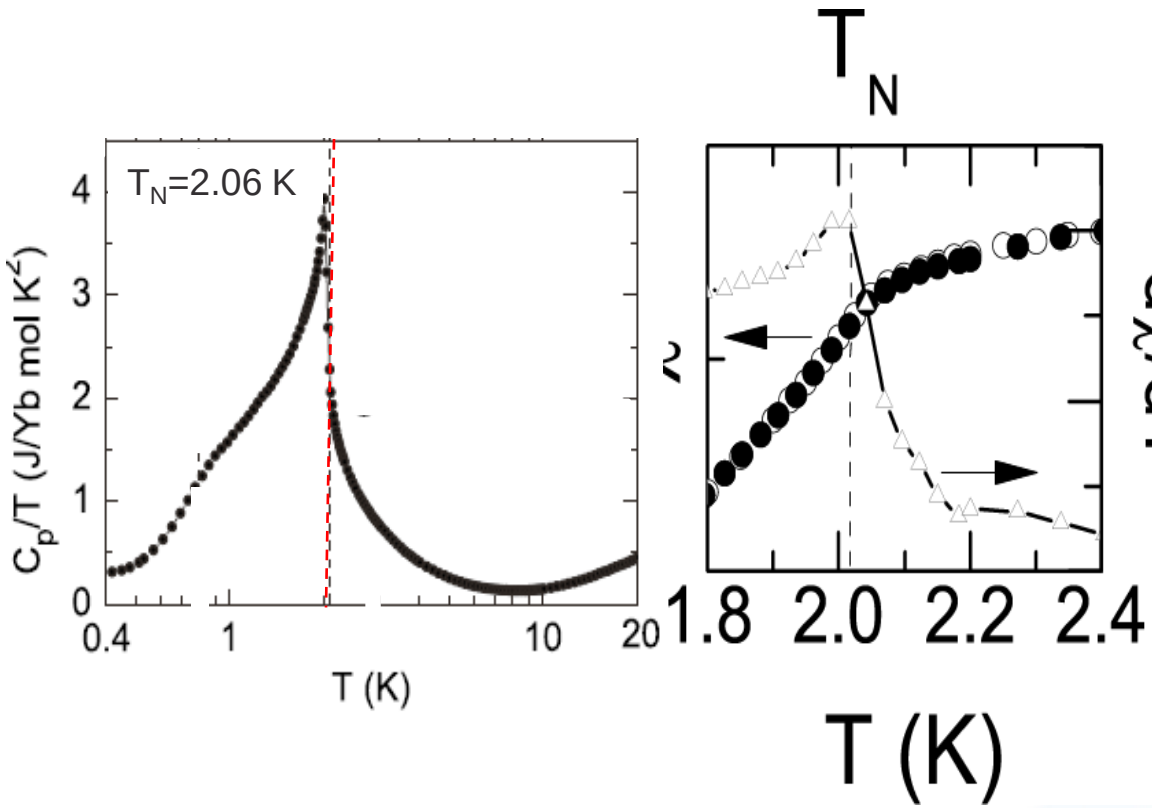


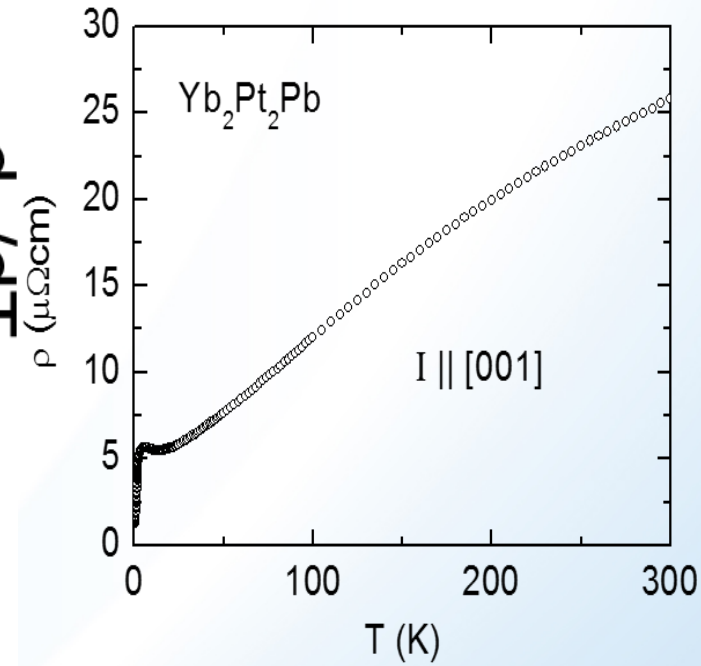




Kim, Aronson, PRL (2013)

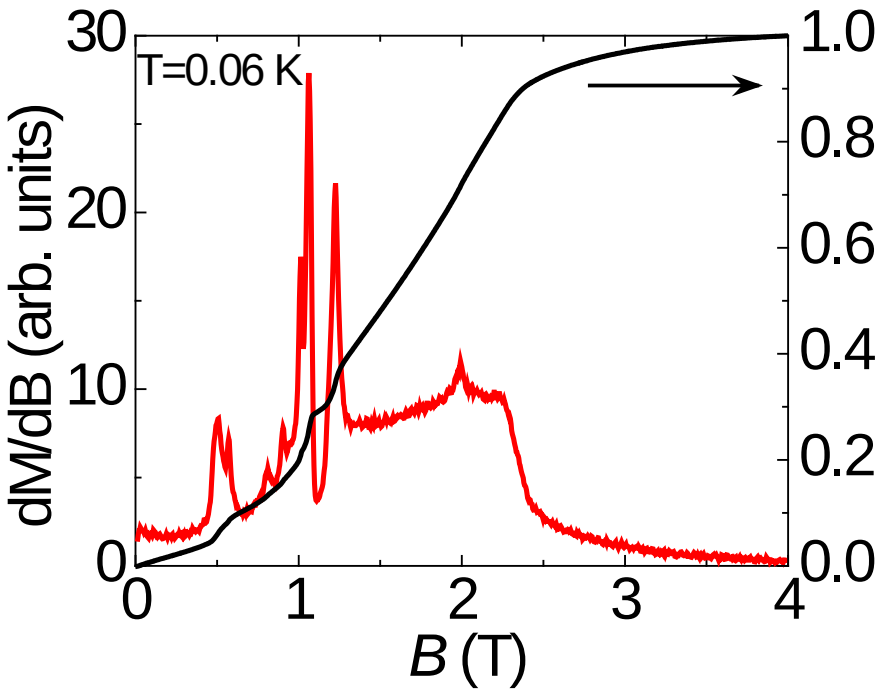
Antiferromagnetic Order in Yb_2Pt_2Pb

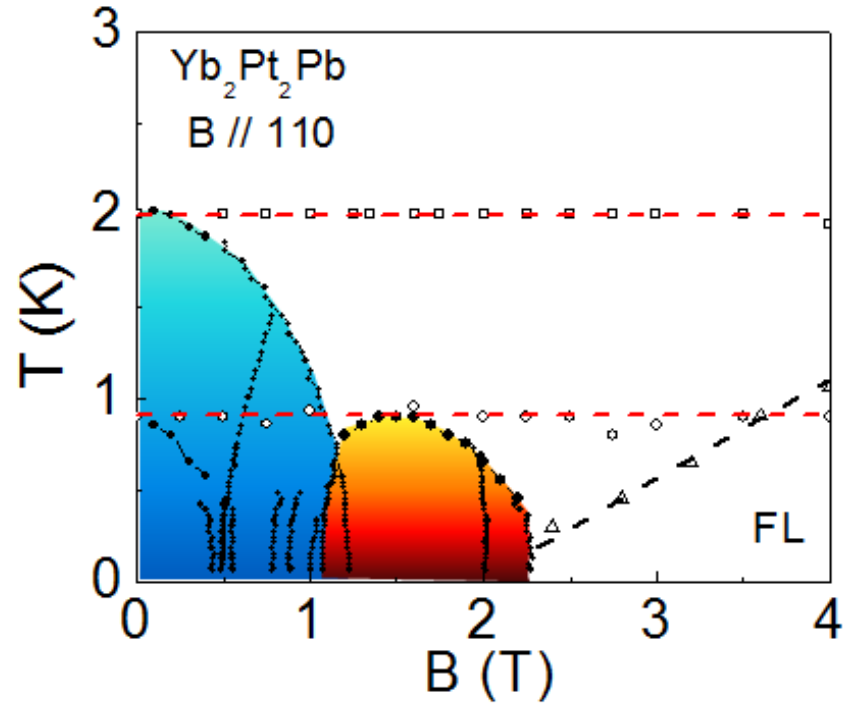




- Antiferromagnetic Order in Yb_2Pt_2Pb: $T_N \approx 2.07$ K
  - Sharp cusp in specific heat C/T, superimposed on broad background.
  - Entropy loss ~0.58Rln2.
  - Peak in $d\chi/dT$.
  - Suppression of spin disorder scattering in resistivity $\rho(T_N)$: $\rho(T=0)$~1.5 $\mu\Omega$-cm

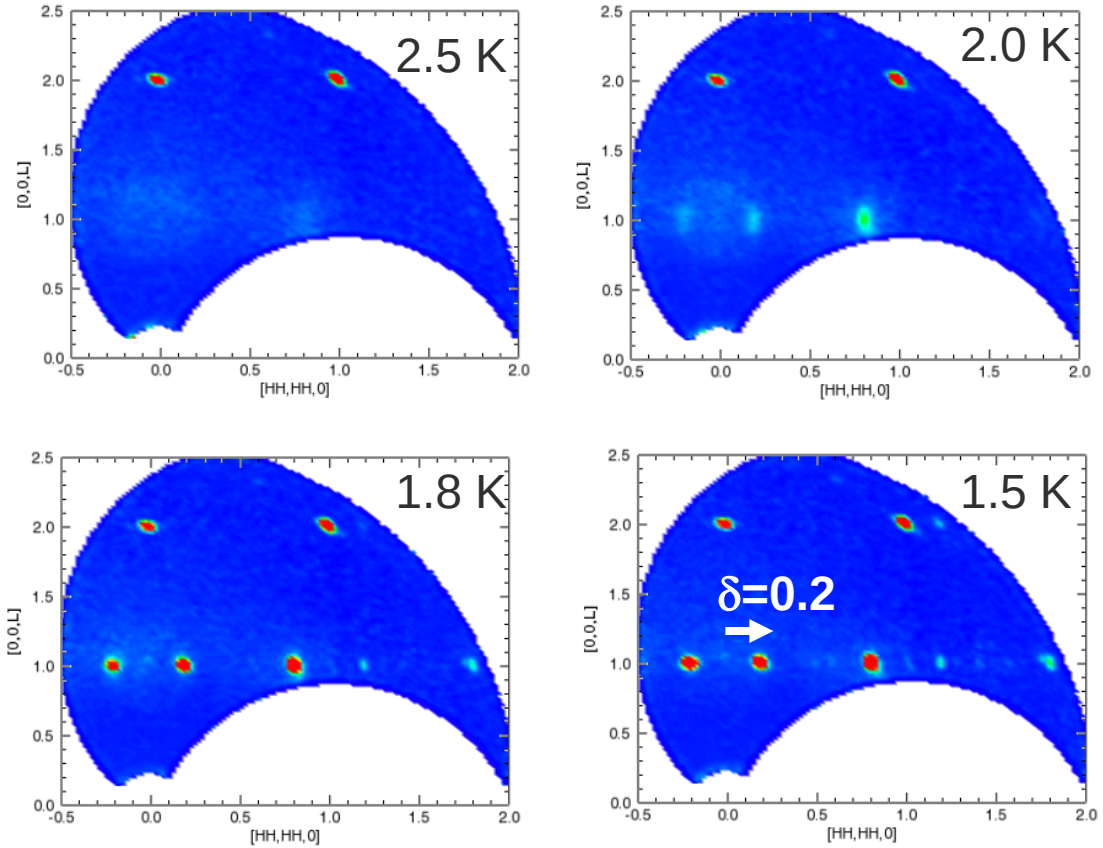
Yb_2Pt_2Pb: Magnetization and phase diagram

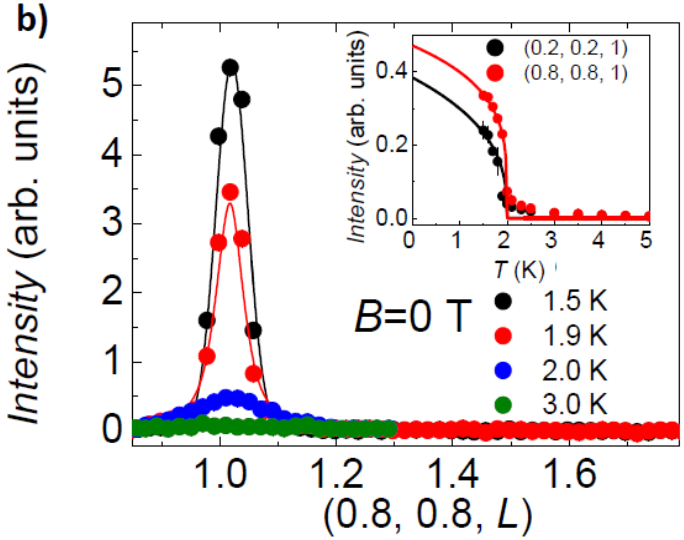




- Sequence of Magnetic Plateaux phases at low temperatures
- Linear-resistivity "dome" phase in-between $B_{c1} \approx 1.25T$ and $B_{c2} \approx 2.3T$
- Horizontal field-independent phase lines at $T_{N1} \approx 2.07$ K and $T_{N2} \approx 0.8$ K

Antiferromagnetic Order in Yb_2Pt_2Pb



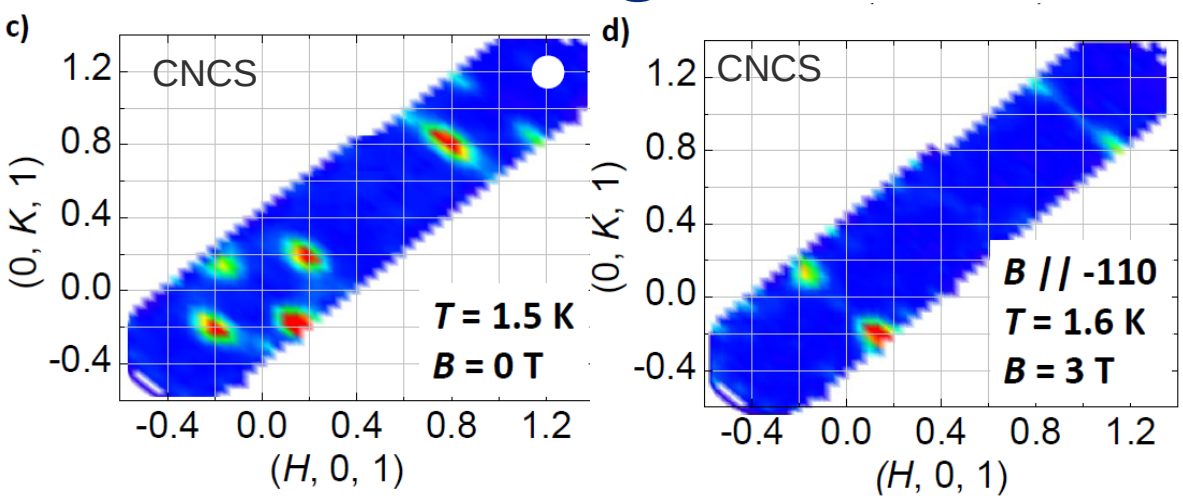


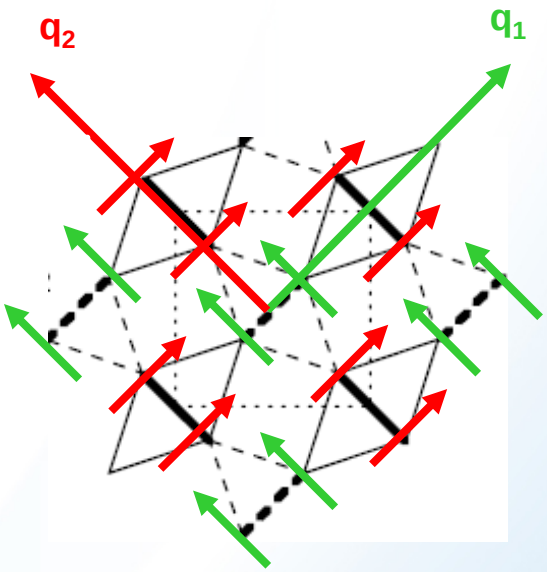
7 g array of ~400 aligned crystals.

- $T<T_N=2$ K: AF supercell 5x5x1 $\mathbf{q_1}$ = (0.2,0.2,1) rlu
- Order parameter: 2D Ising universality class (β=1/8)

$$I \propto (T - T_N)^{2\beta} \square \beta = 0.12 \qquad I \propto (B - B_{QCP})^{2\beta}, \beta = 0.15$$

Antiferromagnetic Order in Yb_2Pt_2Pb

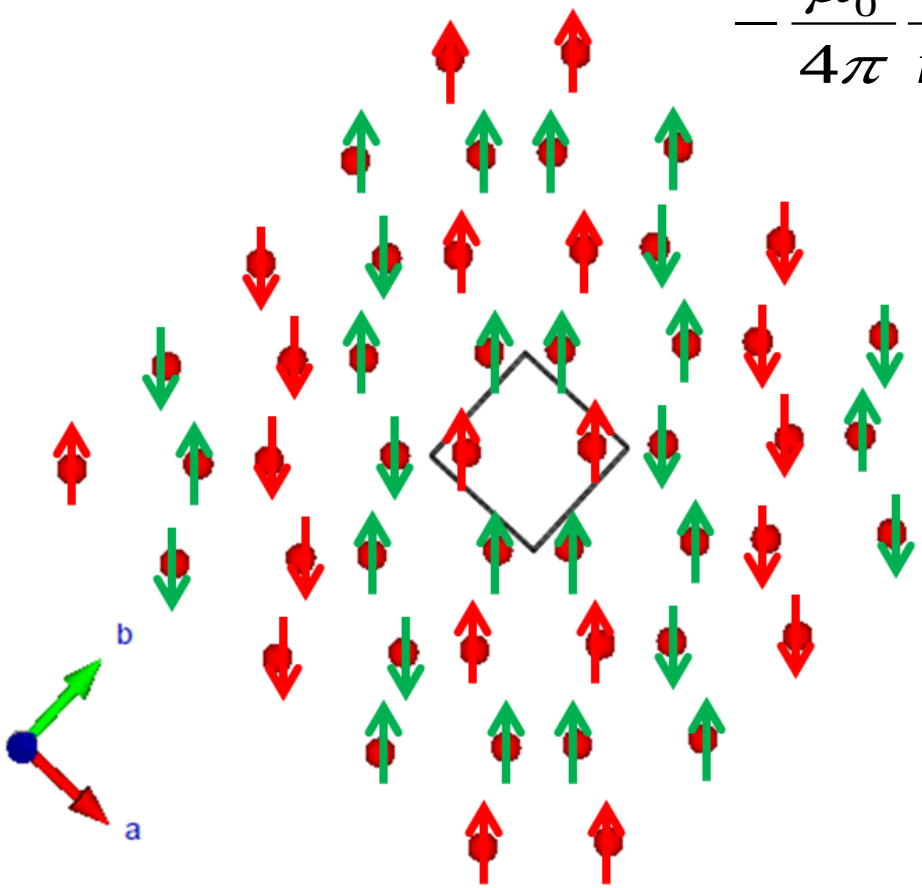


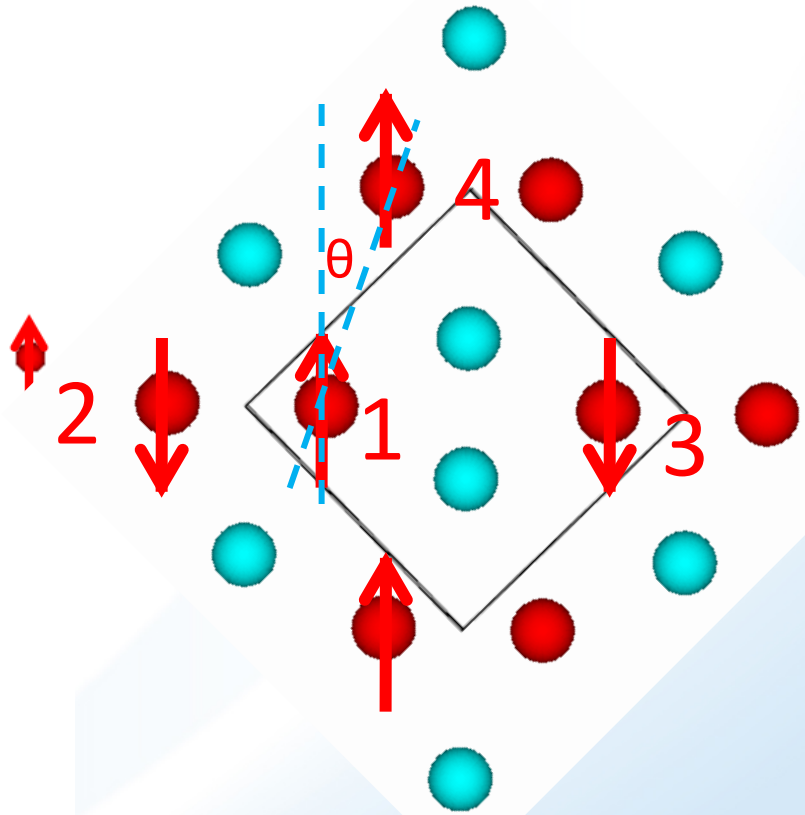


- Yb_2Pt_2Pb: B=0, two orthogonal AF propagation wave vectors: $\mathbf{q}_{1,2}=(\pm 0.2,0.2,0)$ rlu in *ab*- (SSL) plane
- B‖ (-110): only the $\mathbf{q}_2$=(-0.2,0.2,0) sublattice survives, $\mathbf{q}_1$=(0.2,0.2,0) sublattice is fully polarized.
- Ising character: Yb moments can be either parallel or perpendicular to (110),(-110) dimer bonds but magnetic structure has moments perpendicular to dimers.
  - -Sublattice $\mathbf{q}_1$= (0.2,0.2,0): dimer bonds ‖ (110), moments along (-110)
  - -Sublattice $\mathbf{q}_2$= (-0.2,0.2,0): dimer bonds ‖ (-110), moments along (110)

Antiferromagnetic Order in Yb_2Pt_2Pb

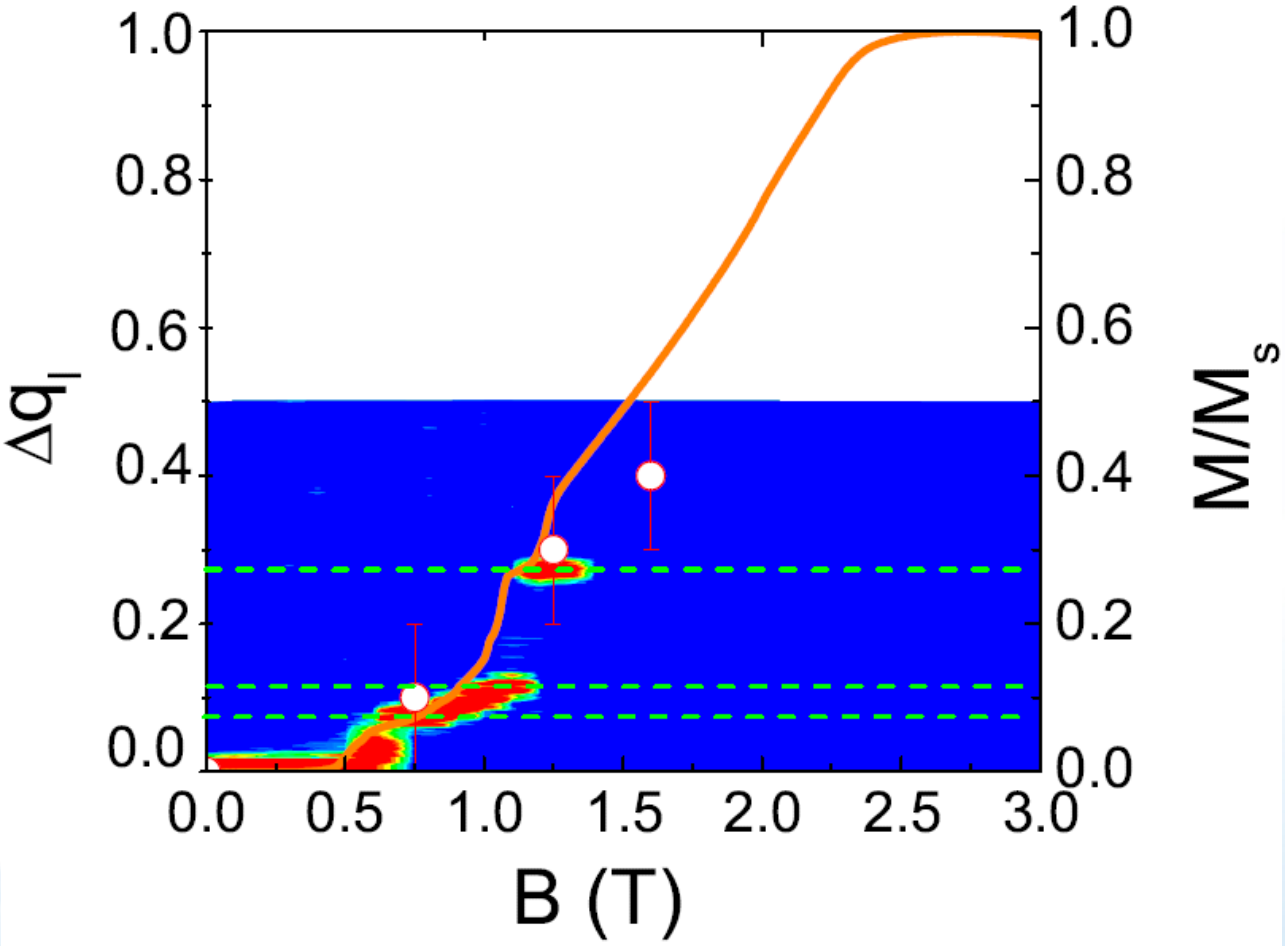
$$-\frac{\mu_0}{4\pi}\frac{1}{r^3}(3(m_1\cdot r)(m_2\cdot r)-(m_1\cdot m_2))$$





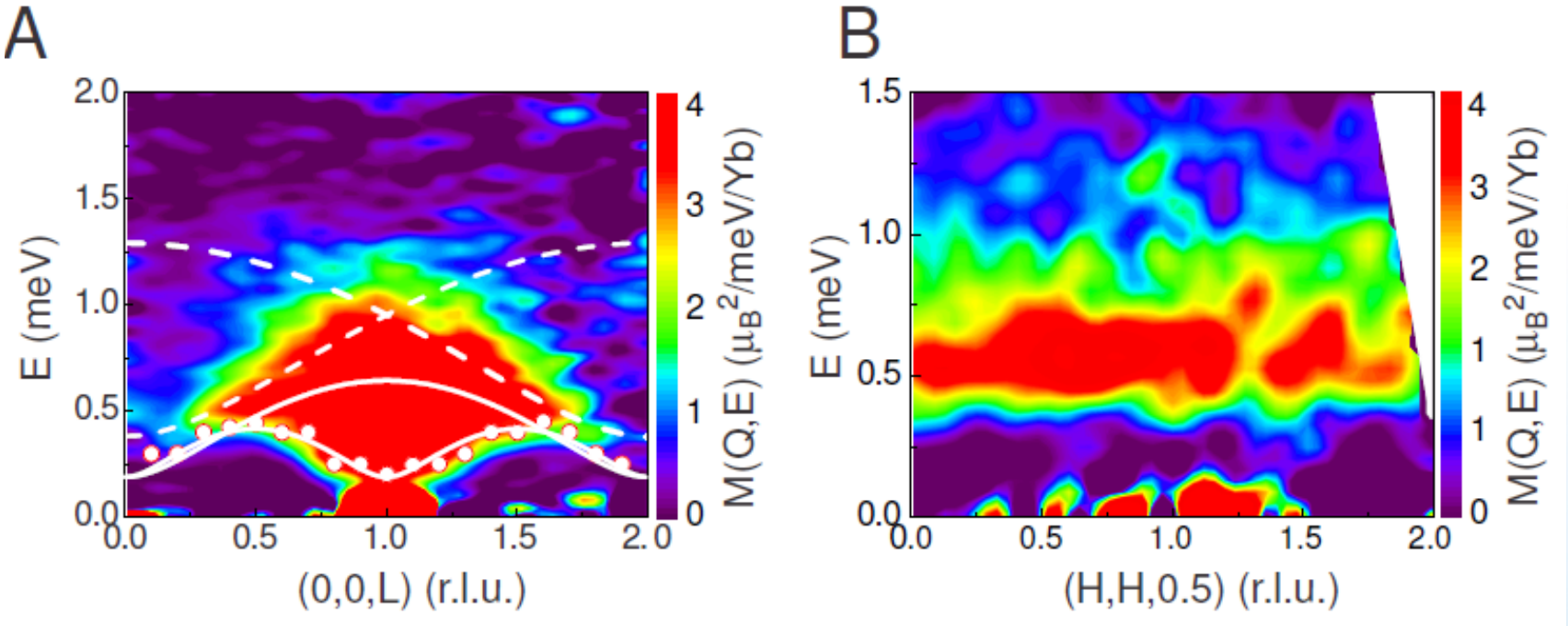
- $\mathbf{q_1}$ = (0.2,0.2) 5x5 ordering in the a-b plane minimizes the energy of dipole-dipole interactions!
  - Yb(1) and Yb(2) dimer sublattices are decoupled

Magnetic order in Yb_2Pt_2Pb in magnetic feld: magnetization and incommensurability



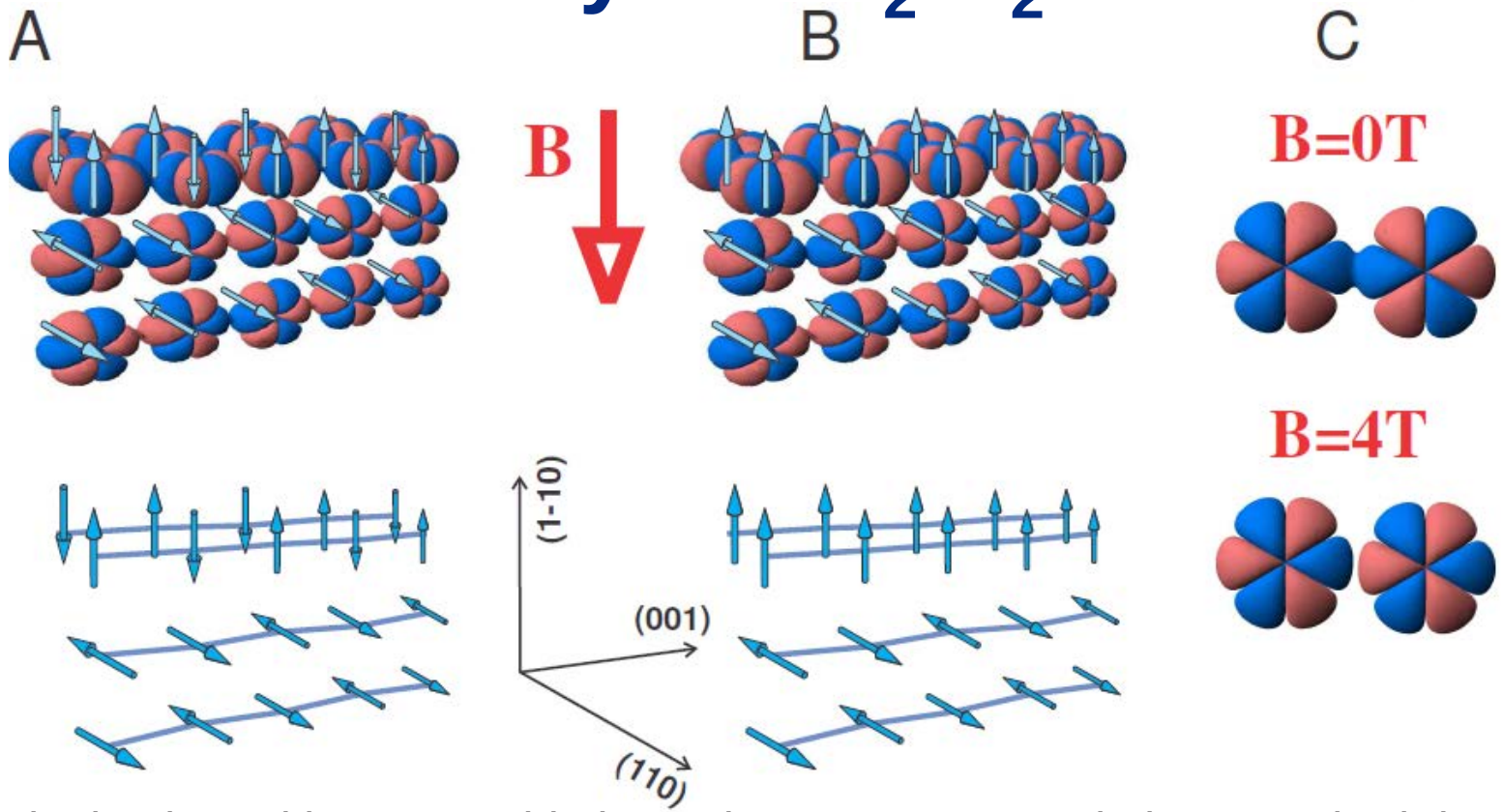
$\Delta q = m$: magnetization in spin-1/2 1D chain?

Yb_2Pt_2Pb: magnetic excitations



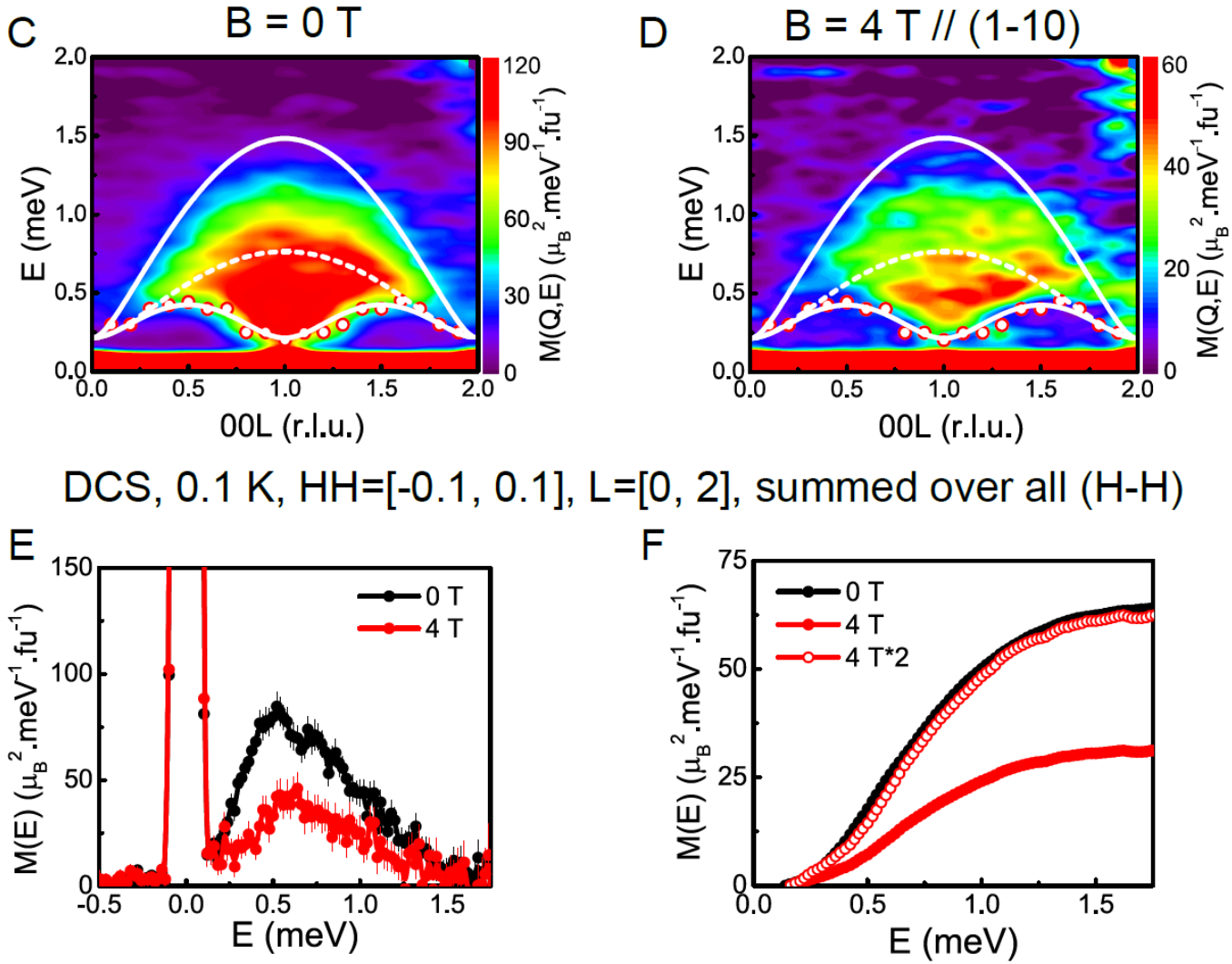
- Dispersing excitations along (00L)
  - continuum of fractional quantum spinons
- Gapped, dispersion-less excitations in the HHL (SSL) plane
  - excitations are one-dimensional, propagation confined to c-axis

Orbitals and emerging one-dimensionality in Yb_2Pt_2Pb



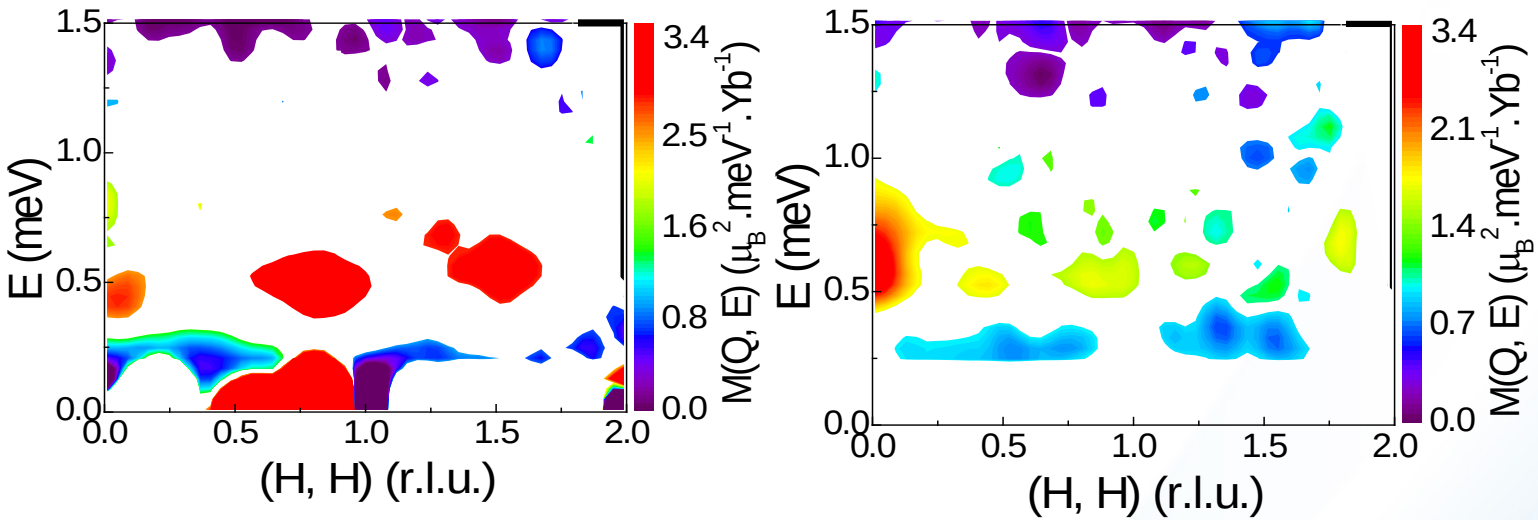
- J is dominated by L = 3 orbital angular momentum, spin is “quenched” by spin-orbit
  - orbital overlaps generate one-dimensional pattern
  - six-fold symmetry of the f-orbital makes ladder legs and rungs inequivalent
  - chains, not ladders
  - orbital spinons, charge-orbital separation, …

Magnetic excitations in Yb_2Pt_2Pb: suppression by magnetic field



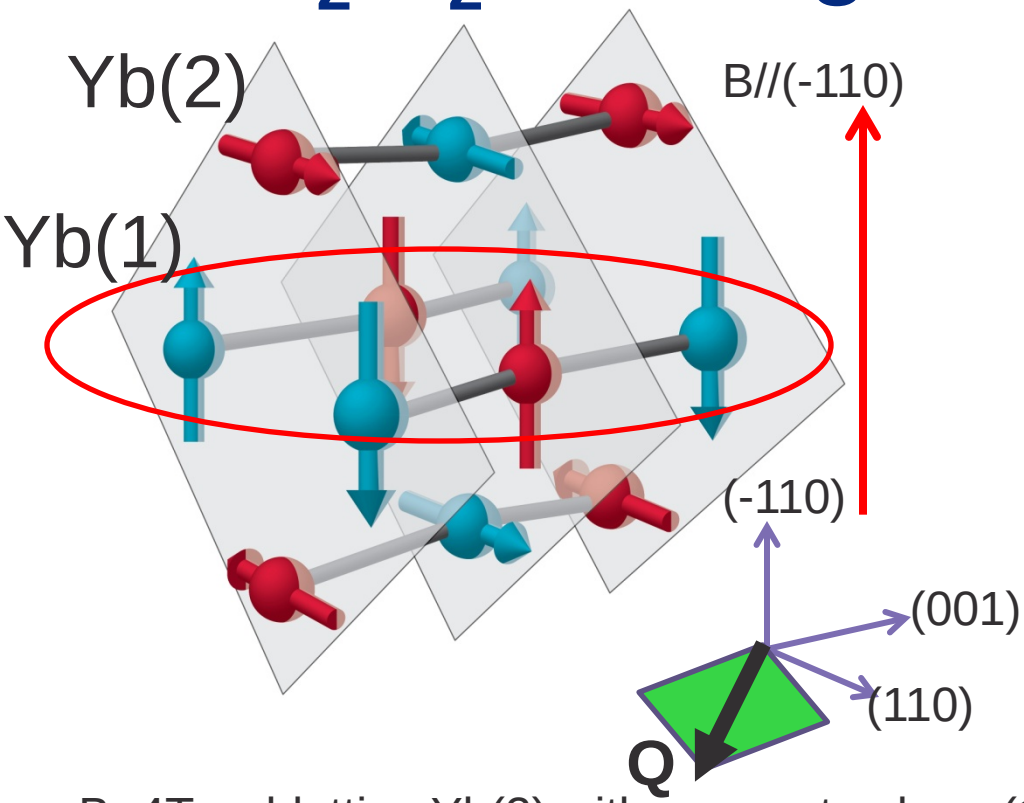
- At B = 4T one sublattice is fully polarized; the other is not affected
  - the energy integrated spectral weight is half that for B = 0
  - no coherent magnon/spin wave!
  - seeing the longitudinal sector only: magnon is dark matter!

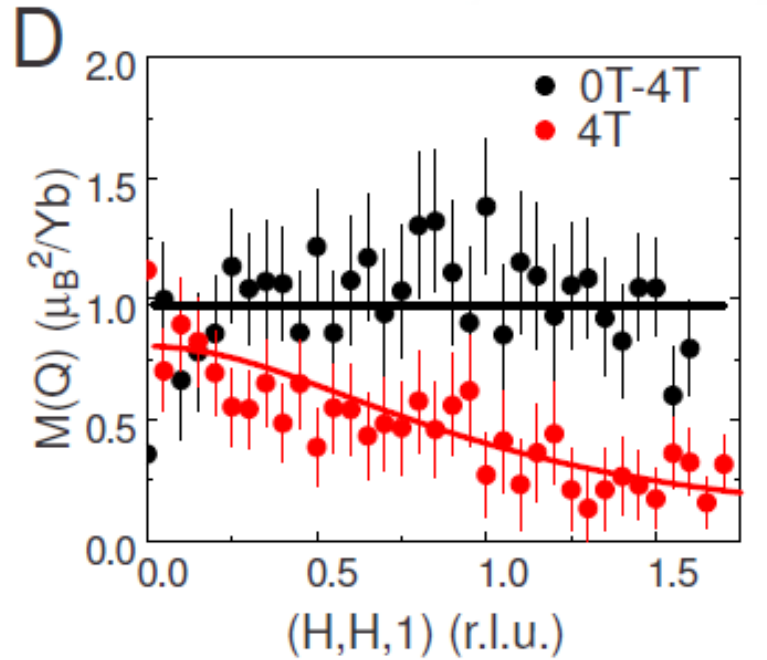
Polarization of magnetic excitations in Yb_2Pt_2Pb: longitudinal dynamics



- Sensitive to longitudinal fluctuations only
  - no transverse components are observable
  - no spin waves seen

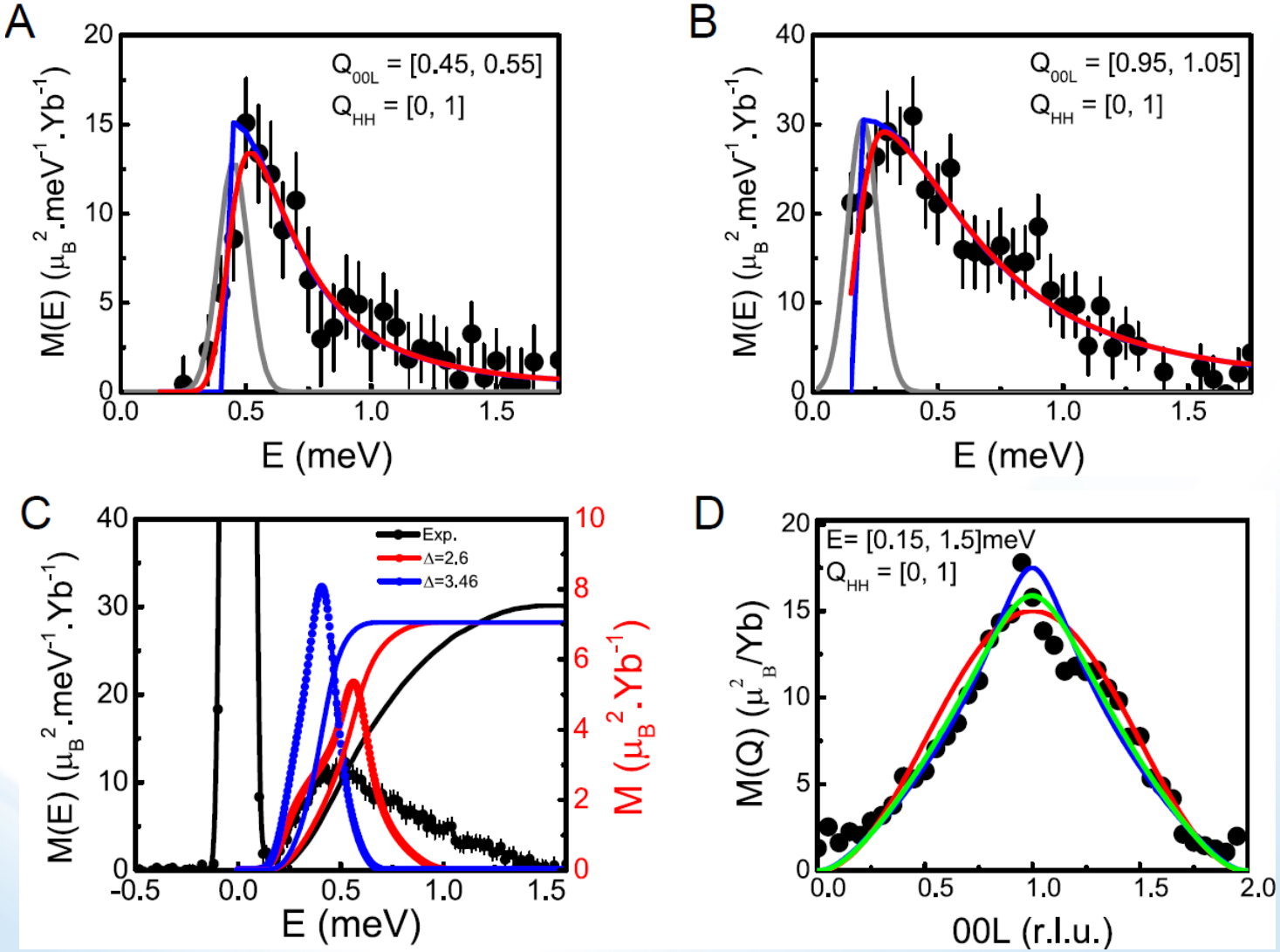
Polarization of magnetic excitations in Yb_2Pt_2Pb: longitudinal dynamics





- B=4T, sublattice Yb(2) with moments along (1, 1, 0)
  - Polarization factor for Yb(2) sublattice: $P = \frac{Q_l^2}{Q_{hh}^2 + Q_l^2}$
- B=0-4T, sublattice Yb(1) with moments along (-1, 1, 0)
  - Polarization factor for Yb(1) sublattice is constant: $P = 1$

Magnetic excitations in Yb_2Pt_2Pb: onset of the contiuum

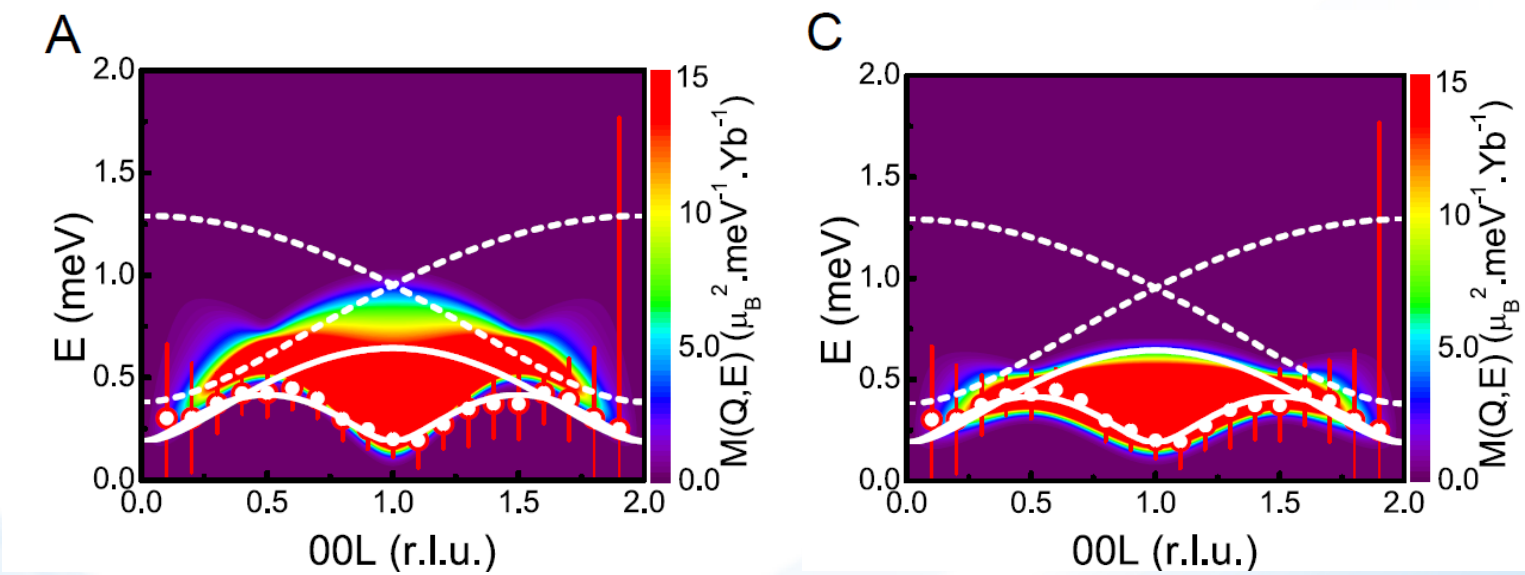


Magnetic excitations in Yb_2Pt_2Pb: comparison with theory (JSC)

Effective spin-1/2 Hamiltonian:

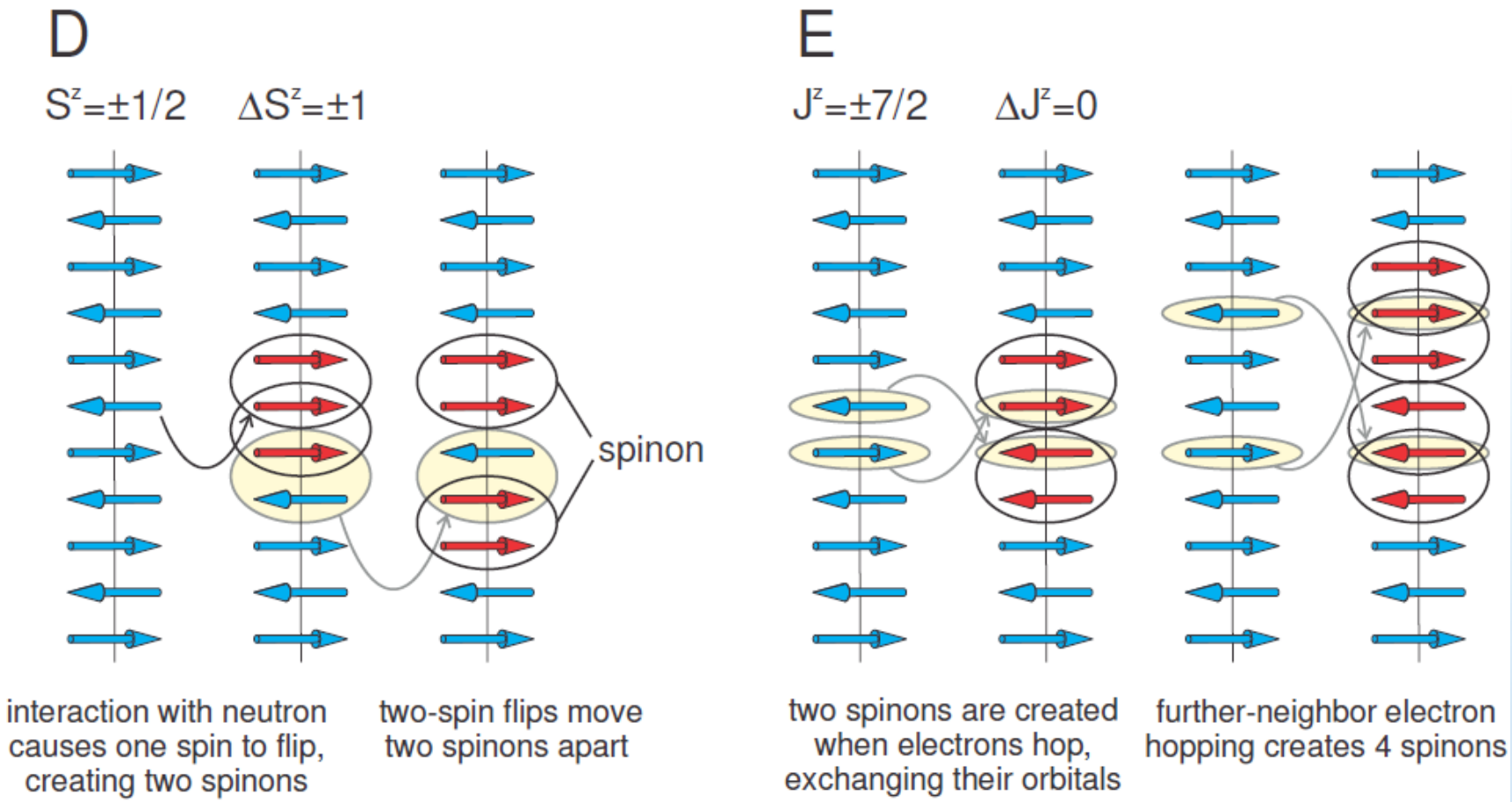
$$H_{XXZ} = J_c \sum_n \left(S_n^x S_{n+1}^x + S_n^y S_{n+1}^y + \Delta S_n^z S_{n+1}^z \right)$$

- Best match of the spectral weight:
  - J ≈ 0.2 meV, Δ ≈ 2.6
- Best fit of the lower boundary:
  - J ≈ 0.1 meV, Δ ≈ 3.46

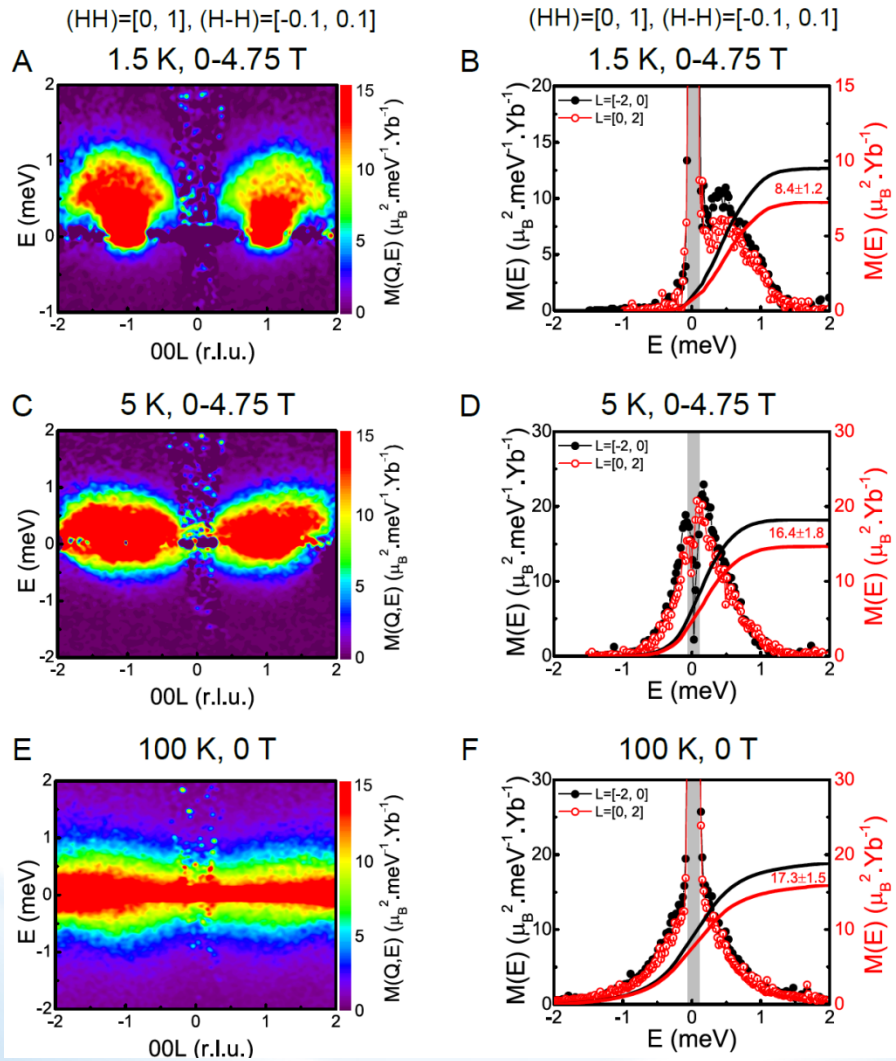


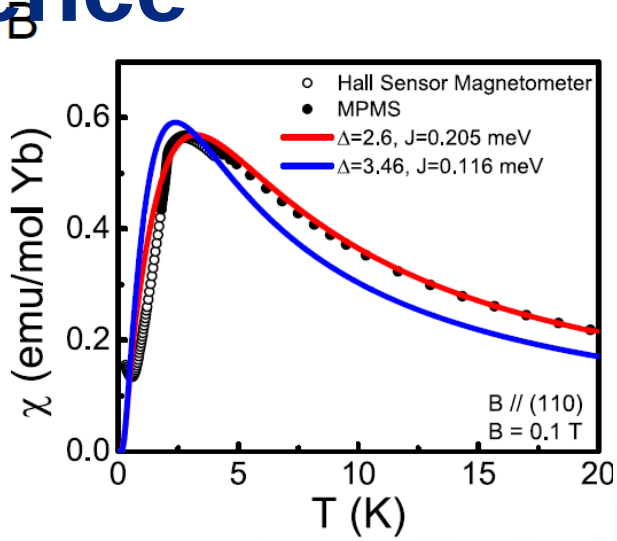
- Large spectral weight at high energy, above 2-spinon continuum
  - 4-spinon continuum enhanced

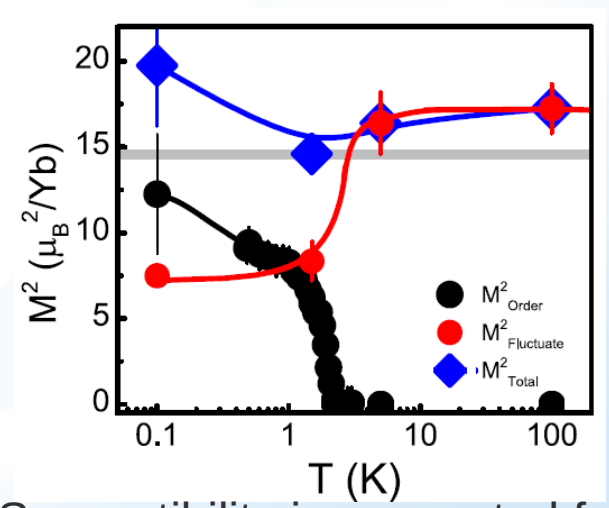
Orbital-exchange and four-spinon excitations



Magnetic excitations in Yb_2Pt_2Pb: temperature dependence



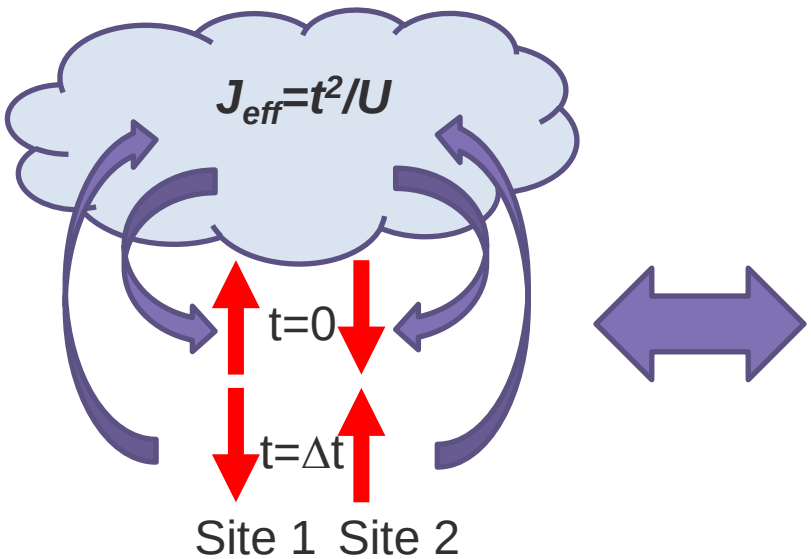


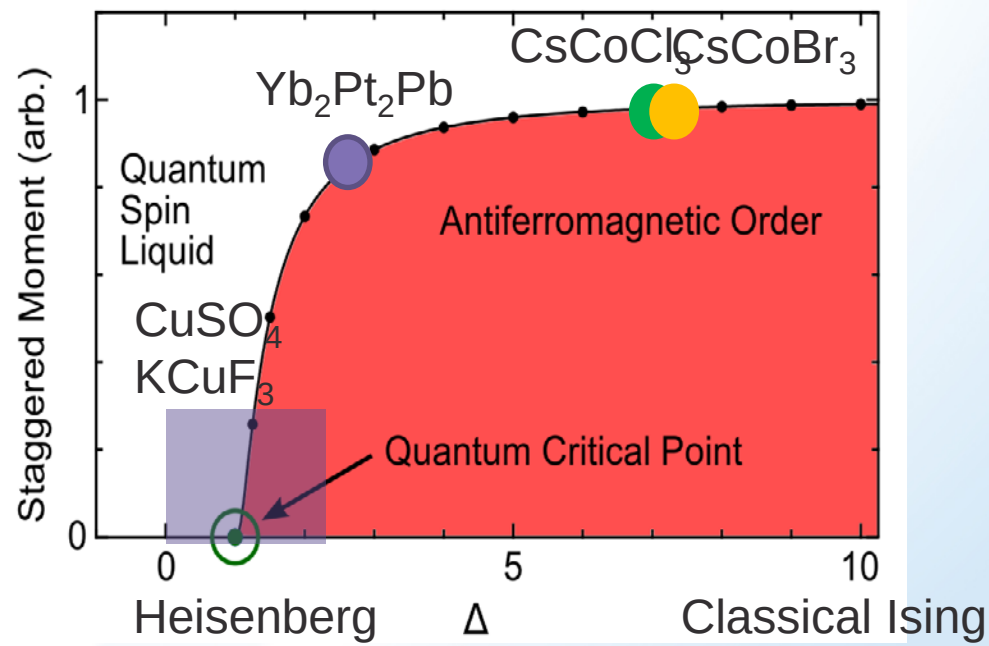


- Susceptibility is accounted for
- Sum rule holds

Emergent dynamics of $J_Z=\pm 7/2$ doublets manifold

$$\mathrm{H} = -\sum_{i,j}\sum_{m=-L}^{L}\sum_{\sigma=\pm 1/2} t(m)\, f^{\dagger}_{im\sigma} f_{jm\sigma} + H_{corr} \qquad \mathrm{H}_{XXZ} = J_c \sum_n \left(\Delta S^z_n S^z_{n+1} + S^x_n S^x_{n+1} + S^y_n S^y_{n+1} \right)$$





- Virtual CEF excitations (ex. $Li_{1-x}Ho_xF_4$, RTi_2O_7): ground state doublet is nearly pure $J_Z=\pm 7/2$
- Correlated hopping via conduction electrons: nearly isotropic coupling described by XXZ Hamiltonian (Uimin 2000)

Summary

- Quantum spin-1/2 dynamics emerges in a system of mainly orbital magnetic moments in Yb_2Pt_2Pb
  - dynamics rendered by electron hopping – orbital exchange mechanism: $S^+S^- \sim \left(J^+\right)^7\left(J^-\right)^7$
  - orbital spinons, charge-orbital separation
  - large J is not classical
- Transverse fluctuations of the effective spin-1/2 are unobservable – “hidden”
  - unique probe of the longitudinal dynamics of the effective model
  - mechanism for hidden orders – URu_2Si_2, …
  - dark matter (hidden magnons), dark energy
- 4-spinon states result from long-range interactions (hopping)
  - Theory?