High Field ESR on Materials with Magnetic Correlations

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Motivation: Heisenberg model on a cubic lattice

$$\mathcal{H} = J \sum \mathbf{S}_i \mathbf{S}_j$$

J<0: ferromagnet; in ground state all spins are parallel
Macroscopic spin: $S_{tot}=NS$
classical ($S=\infty$) ↔ quantum ($S=\frac{1}{2}$) SAME

J>0: antiferromagnet ($J \sim t^2/U$); Néel state; bi-partite with sublattices A and B
Sublattice magnetization: $S^A_{tot}=(N/2)S$
classical: OK ↔ quantum NO ; S^A_{tot} does not commute with $\mathcal{H}$

Elementary excitations: Spin waves (classical) magnons (quantum) $\omega(q)=JS\sqrt{(1-\cos^2(qa))} \sim q$ for small q
Néel may be unstable against quantum fluctuations: $S^A_{tot}=(N/2)S - \int g(q)/\omega(q)\, d^D q$ (D dimensional lattice)

Other choices: combination of singlet states (bond order, RVB, ...) with no magnetic order, possibly no symmetry breaking at all (e.g. in 1 dimension: no Néel order, no magnons, excitations are spinons)

Paramaters

Spin magnitude

Topology (bi-partite, triangular, kagome, other frustrated...)
Range of coupling beyond n.n. $J \rightarrow J(\mathbf{r})$

Coupling to lattice (spin orbit coupling)

- single ion anisotropy $A_x S_x^{\,2} + A_y S_y^{\,2} + A_z S_z^{\,2}$
- exchange anisotropy (extreme case: Ising model $\mathcal{H} = J\Sigma\, \mathbf{S}_i^z \mathbf{S}_j^z$)
- Dzyaloshinskii-Moriya coupling: $\mathbf{D}(\mathbf{S}_1 \times \mathbf{S}_2)$ when crystal is not symmetric

Coupling to lattice will introduce gap in the magnon spectrum, and may stabilize Néel state

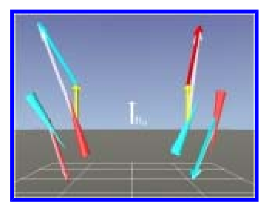
Antiferromagnetic resonance: ESR frequency at zero field: *q*=0 magnon energy

Examples: Classical solution in molecular field approximation; uniaxial anisotropy: $A_x = A_y = 0$;

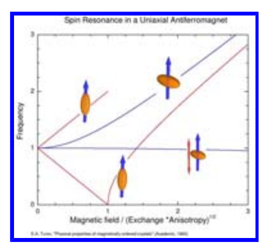
For $A_z < 0$, easy axis

For $A_z > 0$, easy plane

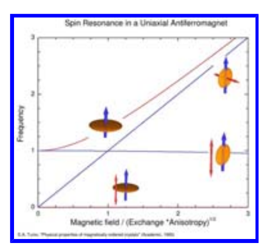
- Exchange field: $H_E = SJ/\mu_B$
- Anisotropy field: $H_A = SA/\mu_B$



- Find the ground state: Fourier transform $J(\mathbf{r})$; system becomes unstable where $J(\mathbf{q})$ is min.
  - Simple nearest neighbor models: $J(q)=J\cos(qa)$ -> leads to AFM or FM.
  - Frustration: Complex lattice or competing second, third etc. neighbor interactions -> leads to incommensurate **q** vector (helical solution).
  - D-M interaction: makes AF into canted or helical.



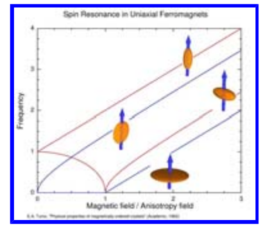
Large body of work in '50s, theory by Keffer, Kittel and others; Experiments: Richards, Tinkham, Foner



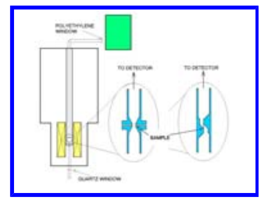
Results: Zero Field AF Frequencies for Easy Axis, Easy Plane, Dzyaloshinskii-Moriya, and field dependance

- Easy axis: $\hbar\omega_0 = 2S\sqrt{(A^2 + 2JA)}$ $\omega_0 = g\sqrt{(H_A^{\,2} + 2H_A H_E)}$
- Easy plane: $\hbar\omega_0 = 2S\sqrt{(2JA)}$ $\omega_0^{\,2} = g\sqrt{(2H_A H_E)}$

- D-M: $\hbar\omega_0=2SD\sqrt{[2/(1+D^2/J^2)]}$
- Easy axis and D-M: Degeneracy of two modes is removed: $\omega_1^2=g^2(H_A^2+2H_AH_E)$ $\omega_2^2=g^2(H_A^2+2H_AH_E+H_D^2)$



Instrument at U12IR, National Synchrotron Light Source



Two major components: Spectrometer and magnet

- Magnet: **Oxford Instruments**, 16Tesla, max 37 mm sample size
- Temperature: 1.3K-300K
- Spectrometer: **Bruker 125 HR**, 8cm^{-1} to 10000cm^{-1} , 0.0001cm^{-1} resolution, works with internal and external sources

ESR: magnetic field dependent optical absorption

- 1meV=8cm^{-1} -- 10K=7cm^{-1} -- 1T=1cm^{-1}

Measure absorption as a function of frequency at many fields; convert to map of H - ω plane

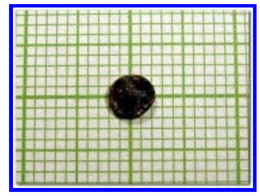
Samples and collaborations

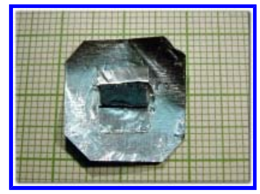
- $CuSO_4$: "paramagnetic" standard; with and without H_2O (Mihaly)
- $LaMnO_3$: canted antiferromagnet T_N=145K (Jianshi

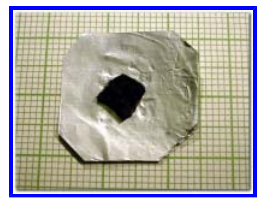
Zhu)

- $NaNiO_2$: triangular spin array (DeBrion)
- $LiCu_2O_2$: spin ½ incommensurate chain T_c=22K (Berger/Forró)
- $Ni_5(TeO_3)_4Cl_2$ spin 1 planar structure T_c=23K (Fehér/Berger/Forró)
- $Cu_2Te_2O_5Cl_2$: tetrahedral spins T_c=18.2K (Fehér/Berger/Forró)
- $BaCu_2Si_2O_7$, $BaCu_2Ge_2O_7$ and $BaCu_2SiGeO_7$: T_c~9K (Zheludev)
- Mn_{12}-acetate: effective spin 10, large anisotropy (Tu, Sarachik)

- Dimitri Basov (UC San Diego), Andrei Sirenko (NJIT), Adrain Gozar (BNL)

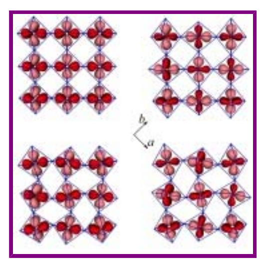


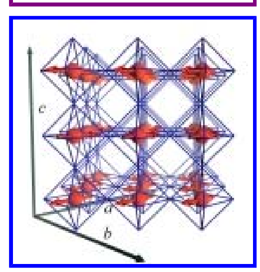




Material: $LaMnO_3$

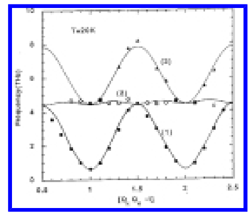
- Parent compound of CMR materials
- Two structural transitions: rotation of octahedra and Jahn-Teller distortion
- Mn sites have localized spins, undergoes antiferromagnetic ordering at 145K
- Spins are ferromagnetic in *ab* plane, AF in *c* direction
- Staggered anisotropy: anisotropy axis points along Mn orbital; orbital is tilted.
- Results in ferromagnetic moment in *c* direction
- Staggered anisotropy: Tilt angle of anisotropy axis, ϕ alternates between planes
- Dzyaloshinskii-Moriya coupling: $\mathbf{D}\,(\mathbf{S}_1 \times \mathbf{S}_2)$

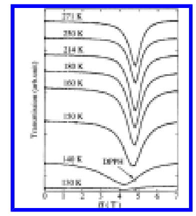


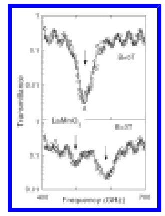


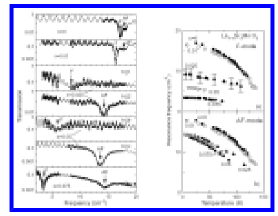
Staggered anisotropy and D-M coupling cause ferromagnetic moment. Which one is more important?

Other works



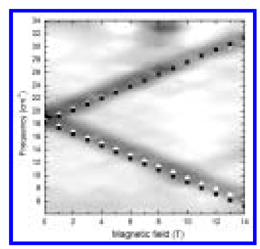




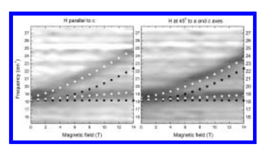


- The resonance line in "pure" $LaMnO_3$ is too broad for Q-band. In doped samples spin diffusion leads to motional narrowing, yields narrower line. Paramagnetic state studied in great detail by Oseroff, Muller and others.
- Neutron scattering (magnons) by Moussa et al.
- High field ESR: Mitsudo, Pimenov's group
- Interpretation by D-M interaction

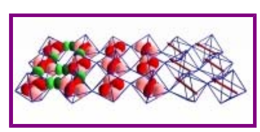
Results: Complete absorption map on the field - frequency plane



- Field parallel to spins (*b* direction): simple uniaxial anisotropy (Kittel) seems to work
- Field perpendicular to spins (*a* and *c* directions): must include D-M coupling
- Dzyaloshinskii-Moriya coupling turns out to be relatively small; H_A=5.3T H_D=1.36T



- Canting angle of spins can be calculated, yields 2.5° (Agrees well with static magnetization measurements.)
- The tilt angle of the anisotropy is 18°



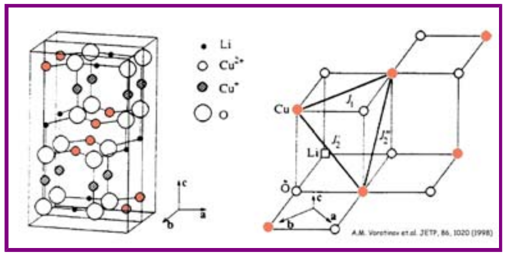
  - Rotation of octahedra (by 15°) explains 10° tilt
  - Rest is non-trivial, qualitative: Mn orbits pulled by O orbits

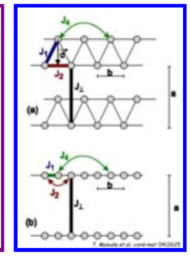
See more here: D. Talbayev, L. Mihaly, J. Zhu, Phys. Rev. Letters, **93** 017202, 2004

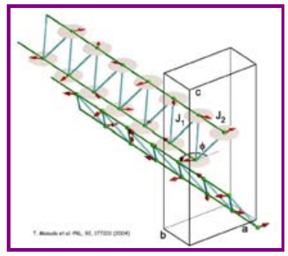
Material: $LiCu_2O_2$

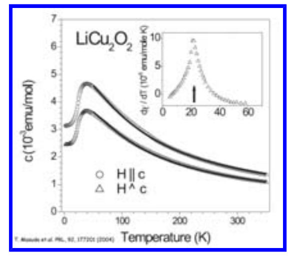
- Two copper ions, one has spin
- Spin 1/2 frustrated ladder

- Spiral order at 22K
- AF resonance at 30GHz (1cm^{-1}) (A.M. Vorotynov et al. JETP **86**, 1020 (1988))
- ESR linewidth diverges as *T* approaches transition temp. (S. Zvyagin et al. PRB **66**, 064434 (2002))
- Coupling strengths, from magnon dispersion (T. Matsuda et al. cond-mat/0412625)
  - J_1=6.4meV,
  - J_2=-11.9meV (ferromagn.),
  - J_4= 7.4meV,
  - J_{perp}=1.8meV





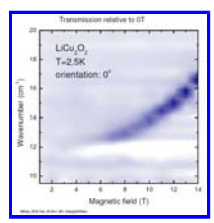


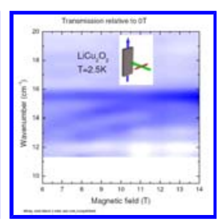


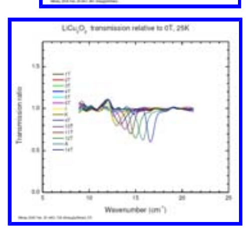
What pins the direction of the spins to the lattice? (S=1/2 -> no single ion anisotropy)

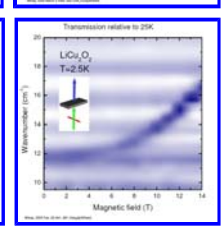
Results

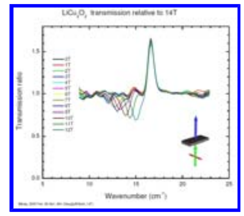
- Finite "AF resonance" frequency: 11.8cm^{-1}
- Intensity **strongly** depends on field; signal disappears at H=0
- Interpretation: Anisotropy is entirely due to D-M interaction.
- D-M field: ~12cm^{-1}~1.5meV
- Note: D-M interaction can lead to spiral structure; it was totally disregarded in earlier models.
- Mystery result: H in ab plane

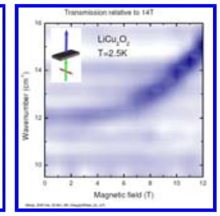












L.Mihály, D. Talbayev, L.F. Kiss, J. Zhu, T. Fehér and A. Jánossy, PRB 69 024414 (2004)

D. Talbayev, L. Miháy, J. Zhu, Phys. Rev. Letters, 93 017202, 2004
