Frustration Driven Lattice Distortion in $Y_2Mo_2O_7$

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Outline

- What is frustration? Why is it interesting?
- Why $Y_2Mo_2O_7$?
- Experimental results.
- Computer simulations-no temperature.
- Computer simulations-crystal “melting”.
- Conclusions.
**Geometrical Frustration**

- AF Hamiltonian and triangular geometry- not all near-neighbor spin interactions can be satisfied: FRUSTRATION.

\[ H = \sum_{i,j} J_{ij} S_i \cdot S_j \]
The Heisenberg Hamiltonian

$$H = \sum_{i,j} J_{ij} S_i \cdot S_j = \frac{J}{2} \sum_{\nabla} \left( \sum_{i \in \nabla} S_i \right)^2 - \frac{J}{2} \sum_{\nabla} \sum_{i} (S_i)^2.$$ 

- The only requirement for minimum of energy: $\sum_{i \in \nabla} S_i = 0$.
- The frustration is “shared” among bonds.
Heisenberg Hamiltonian on the Pyrochlore Lattice

- Infinite set of mean field ground states with zero net spin on all tetrahedra.
- Each tetrahedron has an independent degree of freedom in the ground state!
- No barriers between mean field ground states.
- Infinite degeneracy, no single ground state can be selected by Heisenberg Hamiltonian - lower-order terms become significant.
Is Exchange Constant?

\[ H = \sum_{ij} J_{ij} S_i \cdot S_j \]

- \( J_{ij} \) is controlled by higher energy physics that we like to consider irrelevant at low energies.
  - Atomic spacing
  - Orbital overlap
  - Orbital occupancy
  - Localized or itinerant electronic states

- These degrees of freedom can become relevant if \( H \) produces “degenerate” state.
- The lattice might distort, changing the value of the exchange, if the cost in elastic energy is smaller than the gain in magnetic energy.
Example - the kagome lattice
Suggestion for Relief of Degeneracy - Magnetoelastic Distortion

\[ H = \sum_{i,j} \left( (J + J' \delta r_{ij}) S_i \cdot S_j + \frac{k}{2} (\delta r_{ij})^2 \right) \]

- \( k \) models the electrostatic potential near its minimum.
- \( J' \) is the change in the exchange integral with change in interatomic distance.
Theoretical Ground State, $T=0$

$$H = \sum_{i,j} \left( (J + J' \delta r_{ij}) S_i \cdot S_j + \frac{k}{2} (\delta r_{ij})^2 \right)$$

- Find minimal value of normal vibrational coordinates in the presence of magnetoelastic term $J' \delta r_{ij} S_i \cdot S_j$.
- Arrange distorted tetrahedrons on pyrochlore lattice.
- Net zero spin on each tetrahedron.

Tchernyshyov et al., PRB 66 (2002)
The q=0 State

- The minimum energy state for a single tetrahedron can be arranged on the pyrochlore lattice in one of two q=0 configurations.
- The q=0 distortion: tetrahedrons with identical orientation distort the same way.

Tchernyshyov et al., PRB 66 (2002)
The q=0 State- Characteristics

- 2/3 strong (shortened) bonds,
- 1/3 weak (lengthened) bonds,
- collinear spins
- 2/3 bonds with antiparallel spins, 1/3 bonds with parallel spins.
Searching for Frustration Driven Distortion
We chose $Y_2Mo_2O_7$ as a candidate to look for frustration-driven distortion, since it is a spin glass, and we want to understand the origin of the disorder in this material.
Y$_2$Mo$_2$O$_7$ Characteristics

- Cubic pyrochlore $A_2B_2O_7$
- Magnetic ion $Mo^{4+}$, spin 1
- AF interaction, $\theta_{CW} = 200K$, $J = \theta_{CW}/z$ $\sim$ 33K.
- Spin-Glass transition at 22.5K
Experimental Motivation: \( Y_2\text{Mo}_2\text{O}_7 \)

- Booth et al., XAFS: the Mo tetrahedra are in fact disordered from their ideal structure, with a relatively large amount of pair distance disorder, in the Mo-Mo pairs and perpendicular to the Y-Mo pairs (2000).

- Keren & Gardner, NMR: many nonequivalent \(^{89}\text{Y}\) sites, possibly stemming from a lattice distortion (2001).
Experimental Data

- DC magnetization.
- $\mu$SR.
- High resolution neutron diffraction.
DC magnetization

- Measure sample magnetization with moving sample magnetometer.
- Observe phase transition to spin-glass.
What is µSR?

- 100% spin polarized muons.
- Muon life time: 2.2 μsec.
- Positron emitted preferentially in the muon spin direction.
- Collect positrons, obtain distribution of muon spin orientations.
\[ N(t) = Bg + N_0 e^{-t/\tau_\mu} \left[ 1 + A_0 P(t) \right] \]
Muon Relaxation Mechanisms

- Relaxation caused by dynamical field fluctuations, consists of both longitudinal relaxation caused by fluctuations in the xy plane, and dynamical transverse relaxation caused by fluctuations in the z direction.

- Static relaxation, which is reversible. It is caused by field inhomogeneities in the sample $\Delta B$ which are responsible for dephasing in the xy plane.
The $\mu$SR Experiment

- TF $\mu$SR: measure both static and dynamic relaxation.
- LF $\mu$SR: measure dynamic relaxation.
- Simultaneous TF and LF measurements, $H=6000\,\text{G}$, $20^0\text{K}<T<240^0\text{K}$.

- Subtract LF relaxation from TF relaxation- obtain relaxation from static fields only $\rightarrow$ compare to magnetization.
\( \mu \text{SR Data} \)

\[
A(t) = A_0 \exp\left(-\left(R_{LF}t\right)^{1/2}\right) + Bg
\]

- Relaxation increases as temperature is decreased.
- TF data displayed in rotating-reference-frame, \( H = 5600 \text{G} \).
μSR Data

\[ P_{\text{static}}(t) = P_0 e^{-\frac{(\Delta t)^1}{2}} \cos(\omega t) \]

\[ \Delta = \left( R_{TF}^{1/2} - R_{LF}^{1/2} \right)^2 \]

\[ \omega = \gamma \mu H_{TF} \]

- \( \Delta \) increases exponentially fast with increasing \( \chi \).
What Does it Mean?

- The muon’s Hamiltonian:
  \[ H = \gamma_\mu I \cdot (H_{TF} + H_{int}) \]
  \[ H_{int} = A(r)S \]

- Mean field:
  \[ \langle S \rangle = M = \chi H \]

- Relaxation function measured by \( \mu \)SR:
  \[ P(t) = \int P_0 \cos[\gamma_\mu (1 + A\chi)H_{TF}] \rho(A) dA \]

  Evolution of polarization of a single muon
  Averaging over different muons

A - magnetic coupling
I - muon spin
S - electronic spin
We want the relation between what we measure in μSR and what happens in matter:

\[ P(t) = P_0 e^{-\frac{(\Delta t)^{1/2}}{\nu}} \cos(\omega t) \]

\[ e^{-\frac{(\Delta t)^{1/2}}{\nu}} = \int \cos(A \chi \gamma_\mu H_{TF} t) \rho(A) dA \]

\[ \delta A = \left| \frac{\Delta}{\chi \gamma_\mu H_{TF}} \right| \]

\[ \rho(A) = \frac{1}{|A|} f \left( \frac{\delta A}{|A|} \right) \]

\( \delta A \) represents the width of the distribution.

As the temperature is lowered, the ratio \( \frac{\Delta}{\chi} \) and therefore \( \delta A \), grows, and the distribution widens.
Conclusions from Magnetic Measurements:

⇒ The change in the muon environment indicates that atoms shift!

⇒ However…
High Resolution Neutron Diffraction

- No evidence for periodic rearrangement of the atoms, from μSR or neutrons.
- Neutron scattering data for Y$_2$Mo$_2$O$_7$ show uniform shrinking of the unit cell with decreasing temperature.
Is something wrong with theory?

- Valid only for $T=0$; we’re not there yet...
- Only first order distortional terms were taken into account.
- Assumption of zero net spin on each tetrahedron; not necessarily true in the presence of a magnetoelastic distortion.
- $q=0$ is guessed to be the ground state; the guess might be wrong...
Investigating Further- Computer Simulations

- Energy minimization at $T=0$.
- Periodic boundary conditions for the spins, open for the coordinates, to allow for non-volume-preserving change of the unit cell.
- Structure inspection by Fourier transform and virtual neutron scattering.
- Slow temperature increase from $T=0$ to inspect structure of excited states.
Structure investigation

- Fourier transform:

\[ S(q) = \left| \sum_j e^{iq \cdot R_j} \right| \]

- Magnetic neutron scattering:

\[ S(q) = \left| \sum_{\alpha,\beta} (\delta_{\alpha\beta} - q_\alpha q_\beta) \sum_{i,j} S^i_{\alpha} S^j_{\beta} \cos(q(R_i - R_j)) \right| \]
Finite Size Effects

- Simulation lattice size ~ 10000 atoms << $10^{23}$ atoms in real crystals.
- Examine how characteristic output values of the simulation are affected by lattice size.
- Determine simulation error from finite size effects.
Initial Conditions

- $q=0$ state- is it stable against energy minimization, or can a lower energy state be found?
- Undistorted lattice, random spin arrangement- what minimum energy state will be achieved?
Simulation Results

- The computer could not find a state with lower energy than the $q=0$ state.
- The divergence from linearity stems from non-harmonic effects.

\[
E_{\text{theory}} = -\left( J + \frac{3}{2} \frac{J^2}{k} \right)
\]
Magnetic and non-Magnetic Scattering

Conclusions

- The $q=0$ initial and final states exhibit scattering peaks which are shifted relative to the undistorted lattice peaks; this indicates a shrinking of the entire lattice.

- In the $q=0$ final state, we see a split in the peak corresponding to the kagome-triangular interplane distance, which shows that atoms have moved in and out of planes.

- The final state obtained from a random initial state does not exhibit long range spin correlations, as can be seen from the absence of magnetic scattering peaks.
The near-neighbor spin-spin correlations are similar to those characterizing the $q=0$ state.

Zero net spin on each tetrahedron.
Temperature

- Temperature was increased slowly from $T=0.000001\,\text{J}$ to $T=0.1\,\text{J}$, starting from the $q=0$ initial state.

- Magnetoelastic term $\rightarrow$ long range spin-spin correlations, lattice distortion.

- At $T=0.001\,\text{J}$, splitting is no longer distinguishable, whereas magnetic correlations persist.

- Magnetic probes such as $\mu$SR and NMR are expected to be more sensitive to the presence of the magnetoelastic term than nonmagnetic probes.
Lattice distortion

Spin-spin correlations

Fourier Transform

Magnetic Neutron Scattering
Conclusions

- We looked for the ground state of the pyrochlore lattice with magnetoelastic Hamiltonian, with the aid of computer simulations. We could not find a state with lower energy than the $q=0$ state, for $J'/k<<1$.

- The computer simulation showed that for $J'/k<<1$ the theoretical assumptions hold: zero net spin on each tetrahedron, $2/3$ strong (shortened) bonds, $1/3$ weak (lengthened) bonds, $2/3$ bonds with antiparallel spins, $1/3$ bonds with parallel spins.

- The simulation shows that the $q=0$ state is not distinguishable with non-magnetic probes above $T=0.001J$. For $Y_2Mo_2O_7$ this means $T\sim0.03K$. 