Linear spin wave theory

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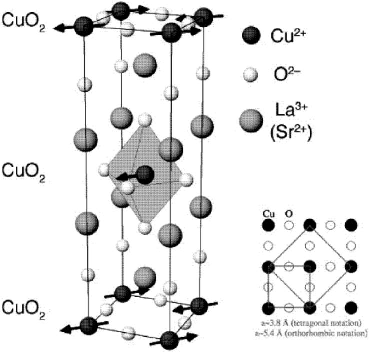
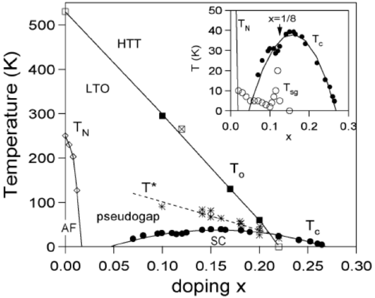
Presentation Outline

Why to calculate spin waves?





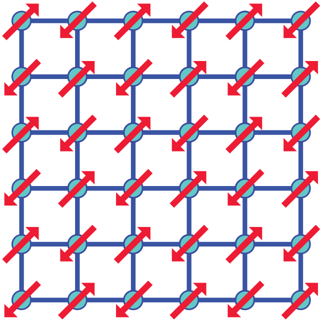
La_2CuO_4 crystal structure.

R. Gilardi, PhD thesis, ETH Zurich (2004).

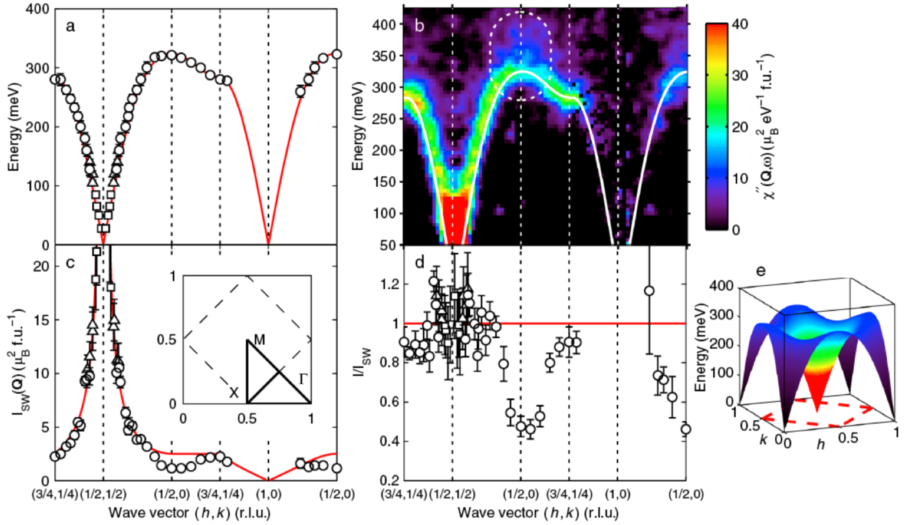
Phase diagram with superconducting phase.

Square lattice antiferromagnet

- Heisenberg spin Hamiltonian: $\mathcal{H} = \sum J_{ij} S_i S_j$
- Antiferromagnetic ground state is not the eigenstate of the Hamiltonian (P. W. Anderson, 1951)
- Resonating Valence Bond states?
- 2 decades of experimental and theoretical work on La_2CuO_4
- Neutron scattering as the main experimental technique
- Understanding the underlying electronic interactions



Square lattice antiferromagnet



N.S. Headings et al., Phys. Rev. Lett. 105, 247001 (2010).

Presentation Outline

Origin of magnetism

Magnetic moments in solids:

- electron spin, $M_e = g\mu_B S$, $g = 2$, $S = 1/2$
- electron orbital angular momentum, $M_L = q\mu_B L$, $g = 1$
- nuclear magnetic moment, $M_n << M_e$

Energy scale:

- $g\mu_B \approx 0.12meV/T$
- 1 meV = 11.6 K = 8.3 T

Atomic magnetism:

- transition metals: Fe(3d), Pd(4d), Pt(5d)
- rare-earth metals (4f) and actinides (5f)
- competition between Hund's coupling, spin orbit coupling and crystal field

Magnetic interactions in solids

Dipole-dipole interaction

- $E_{DD} \sim \left\{ 3(\mathbf{S}_1 \cdot \hat{\mathbf{r}})(\mathbf{S}_2 \cdot \hat{\mathbf{r}}) - \mathbf{S}_1 \cdot \mathbf{S}_2 \right\}$
- for electron spins: $E_{DD} \approx 0.3 \frac{meV}{r_A^3} \quad \rightarrow 1$ K

Interatomic exchange:

- no hopping: Coulomb repulsion favors parallel spin alignment (FM)
- hopping: favors AFM alignment (lower kinetic energy)

Heisenberg interaction:

- two electron wave function: antisymmetric:
    - $S = 0 \quad \Psi_1 = \Psi_S(r, r')\chi_A(\sigma, \sigma')$
    - $S = 1 \quad \Psi_2 = \Psi_A(r, r')\chi_S(\sigma, \sigma')$
- $S_{TOT}^2 = S^2 + S'^2 + 2\mathbf{S} \cdot \mathbf{S}'$
- if $E_1 - E_2 = 2J \quad \rightarrow \quad E = J\mathbf{S} \cdot \mathbf{S}'$

Magnetic interactions in solids

Kanamori-Goodenough rules:

- phenomenological description of superexchange interaction
- interaction between overlapping orbitals: strong AF
- interaction between non-overlapping orbitals: weak FM

Spin-orbit coupling (SOC):

- in $3d$ transition metals weak, gives weak anisotropy and Dzyaloshinskii-Moriya interaction
- crystal field quenches orbital angular momentum
- in rare earth elements, SOC is stronger than crystal field, orbital momentum unquenched
- strong anisotropy

Presentation Outline

Neutron as an ideal probe

Basic properties

- charge: 0
- spin: $S_N = 1/2$
- magnetic moment: $M_N = -1.913\,\mu_N$
- mass: $1.675 \cdot 10^{-27}$ kg

Energy scale:

- $E = \frac{h^2}{2m\lambda^2}$
- $\lambda = 1$ Å$\rightarrow E = 81.8$ meV

Scattering cross section

Definition:

$$\frac{d^2\sigma}{d\Omega dE} = \frac{\text{sc. n. in the } d\Omega \text{ solid angle in the } (\theta\varphi) \text{ direction with energy} (E';E'+dE')}{\Phi d\Omega E'}$$

Nuclear scattering:

- cross section: $\frac{d\sigma}{d\Omega} = b^2$
- b is usually real, and non monotonic of the atomic number unlike X-ray
- typically diffraction and inelastic scattering of phonons

Magnetic scattering:

- general cross section:
  $$\frac{d^2\sigma}{d\Omega dE} = C \cdot F^2(\mathbf{K}) \sum_{\alpha\beta} \left(\delta_{\alpha\beta} - \hat{k}_\alpha \hat{k}_\beta\right) S^{\alpha\beta}(\omega, \mathbf{k})$$
- dynamic spin correlation function:
  $$S^{\alpha\beta}(\omega, \mathbf{k}) = \frac{1}{2\pi\hbar} \int dt e^{-i\omega t} \frac{1}{N} \sum_{i,j} e^{i\mathbf{k}\cdot(r_i - r_j)} \langle S_i^\alpha(0) S_j^\beta(t)\rangle$$

Useful rules

Total scattering

- $\int S^{\alpha\alpha}(\mathbf{Q},\omega)d^3\mathbf{Q}d\omega = S(S+1)$

Energy integrated scattering intensity:

- equal time correlation function $\langle S^\alpha(0)S^\beta(0)\rangle$
- snapshot of the fluctuating magnetic moments

Elastic scattering:

- measures static correlations
- $\langle S^\alpha(0)S^\beta(t\to\infty)\rangle$ limit
- magnetic Bragg scattering

Inelastic scattering with fixed E energy transfer:

- measures time evolution at frequency $\omega = E/\hbar$
- measured dE energy width of resonant excitations gives the quasiparticle lifetime $t = h/dE$

Presentation Outline

Spin waves: definition

Definition:
Spin waves are propagating disturbances of an ordered magnetic lattice.

Classically:

- spin is a vector with fixed length
- equation of motion (Larmor precession):

$$\frac{d\mathbf{S}}{dt} = \mathbf{S} \times \mathbf{h}$$

- $\mathbf{h}$ field can be exchange (Weiss) field, external field or anisotropy
- general solution:

$$\begin{aligned} dS_i^x =& A_i^x \cos(\omega t + \mathbf{k} \cdot \mathbf{r}_i + \varphi_i^x) \\ dS_i^y =& A_i^y \sin(\omega t + \mathbf{k} \cdot \mathbf{r}_i + \varphi_i^y) \end{aligned}$$

Warm up: classical spin waves in a ferromagnetic chain

Energy:

$$E = \sum_i J\mathbf{S}_i \cdot \mathbf{S}_{i+1}$$

Equation of motion:

$$\dot{\mathbf{S}}_i = J\mathbf{S}_i \times (\mathbf{S}_{i-1} + \mathbf{S}_{i+1})$$

Assuming small deviation from equilibrium:

$$\begin{aligned}
\delta\dot{S}_i^x =& JS(-\delta S_{i-1}^y - \delta S_{i+1}^y + 2\delta S_i^y) \\
\delta\dot{S}_i^y =& JS(\delta S_{i-1}^y + \delta S_{i+1}^y - 2\delta S_i^y)
\end{aligned}$$

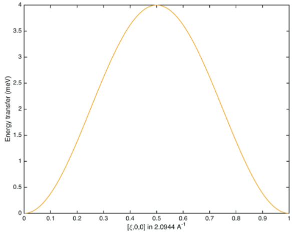
Warm up: classical spin waves in a ferromagnetic chain

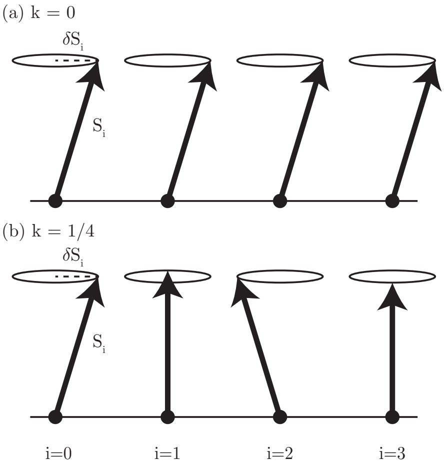
Using the spin precession ansatz:

$$\omega \delta S_i^y = JS(2\delta S_i^y \cos(\mathbf{k} \cdot \mathbf{a}) - 2\delta S_i^y)$$

Spin wave dispersion:

$$\omega = 2JS(\cos(\mathbf{k} \cdot \mathbf{a}) - 1)$$

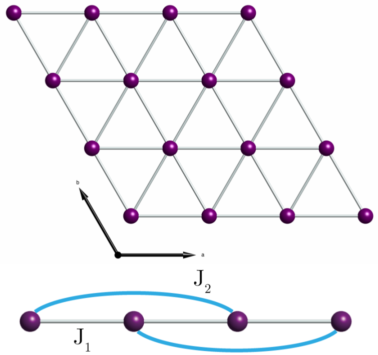




Presentation Outline

Spiral ground state

- Frustration:
  geometrical
  competing interactions
- Degenerate ground states
- Weak interactions (e.g. order by disorder) select state from manifold



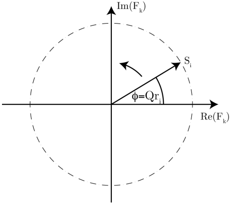
Luttinger-Tisza theory

The classical ground state of a Heisenberg Hamiltonian on the Bravais lattice is always a single-Q spiral.

Spiral ground state

Heisenberg Hamiltonian (Bravais lattice):

$$\mathcal{H} = \sum_{ij} J_{ij} S_i S_j$$



Magnetic structure:

$$\mathbf{S}_i = \sum_{\mathbf{k}} \mathbf{F}_{\mathbf{k}} e^{-i\mathbf{k}\cdot\mathbf{r}_i}$$

$$\mathbf{F}_{\mathbf{k}} = \overline{\mathbf{F}}_{-\mathbf{k}}$$

Spiral ansatz: there is only a single k vector (Q)

$$\mathbf{S}_i = S\left(\mathbf{u}\cos(\mathbf{Q}\cdot\mathbf{r}_i) + \mathbf{v}\sin(\mathbf{Q}\cdot\mathbf{r}_i)\right)$$

Spiral ground state

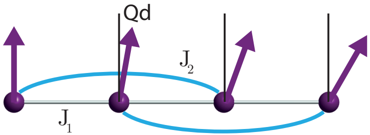
Classical energy:

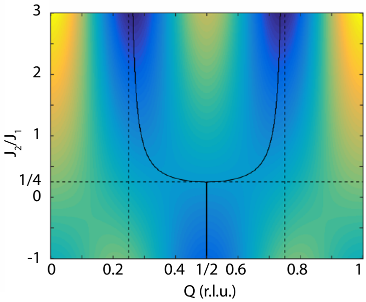
$$E = S^2 \min(J(\mathbf{k})) \rightarrow \mathbf{Q}$$

where:

$$J(\mathbf{k}) = \sum_{\mathbf{d}} J(\mathbf{d}) e^{i\mathbf{k}\cdot\mathbf{d}}$$

d in l.u.	exchange
(1,0,0)	J_1
(-1,0,0)	J_1
(2,0,0)	J_2
(-2,0,0)	J_2





Quantum mechanical treatment of spin waves

- Same ground state and dispersion as for the classical model
- Excitations are quasiparticles: magnons
  - bosons
  - commutator : $[a, a^{+}] = 1$
  - Bose-Einstein statistics: $n(\omega) = \frac{1}{\exp(\hbar\omega/k_B T)-1}$
- Equivalent to coupled quantum harmonic oscillators:
  - zero point energy

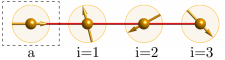
  $\rightarrow$ reduction of magnetic order

  $\rightarrow$ quantum order by disorder
- Strong magnon-magnon interaction in non-collinear magnets

Rotating coordinate system

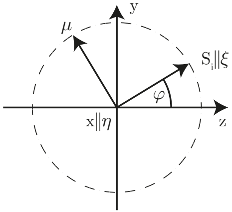
Coordinate transformation:

$$
\begin{aligned}
S_i^x &= S_i^\eta \\
S_i^y &= S_i^\mu \cos(\varphi_i) + S_i^\xi \sin(\varphi_i) \\
S_i^z &= -S_i^\mu \sin(\varphi_i) + S_i^\xi \cos(\varphi_i)
\end{aligned}
$$



Rotation angle:

$$\varphi_i = \mathbf{Q} \cdot \mathbf{r}_i$$



In the rotating coordinate system: ferromagnet

Hamiltonian

Heisenberg Hamiltonian on Bravais lattice:

$$\mathcal{H} = \sum_{i,d} J(d)\mathbf{S}_i \cdot \mathbf{S}_{i+d}$$

After substitution and a bit of trigonometry:

$$\mathcal{H} = \sum_{i,j} J(\mathbf{d}_{ij}) \left(S_i^\eta S_j^\eta + \sin(\mathbf{Q}\cdot\mathbf{d}_{ij})(S_i^\mu S_j^\xi - S_i^\xi S_j^\eta) \right) +$$
$$+ \cos(\mathbf{Q}\cdot\mathbf{d}_{ij}) \left(S_i^\mu S_j^\mu + S_i^\xi S_j^\xi \right)$$

Spin algebra

Spin state (in small field along z-axis):

$$|n\rangle \equiv |S, m = S - n\rangle$$

Ladder operators for spin:

$$S^-|n\rangle = \sqrt{2S}\sqrt{1 - \frac{n}{2S}}\sqrt{n+1}|n+1\rangle$$
$$S^+|n\rangle = \sqrt{2S}\sqrt{1 - \frac{n-1}{2S}}\sqrt{n}|n-1\rangle$$

Ladder operators for harmonic oscillator:

$$a^+|n\rangle = \sqrt{n+1}|n+1\rangle$$
$$a|n\rangle = \sqrt{n}|n-1\rangle$$

$$[a, a^+] = 1$$

Holstein-Primakoff transformation

Ladder operators for spin:

$$S^- = \sqrt{2S} a^+ \hat{f}$$
$$S^+ = \sqrt{2S} \hat{f} a$$
$$\hat{f} = \sqrt{1 - \frac{n}{2S}}$$

Spin wave approximation

Assuming $\langle n \rangle << S$ the coupled harmonic oscillator is a good model. The excited states of the harmonic oscillator are magnons.

$$S_i^\eta = 1/2(S^+ + S^-) = \sqrt{S/2}(a_i^+ + a_i)$$
$$S_i^\mu = -i/2(S^+ - S^-) = \sqrt{S/2}(a_i^+ - a_i)$$
$$S_i^\xi = S - n_i = S - a_i^+ a_i$$

Quadratic form

After substitution into the spin Hamiltonian, such as:

$$S_i^\eta S_j^\eta = S/2(a_i^+ + a_i)(a_j^+ + a_j)$$

$$\dots$$

The terms can be expanded in the number of boson operators, each consecutive term gain a constant factor of $1/\sqrt{S}$.
Constant term:

$$E = S^2 \sum_{i,\mathbf{d}} J(\mathbf{d}) \cos(\mathbf{Q} \cdot \mathbf{d})$$

1 operator term:

$$\mathcal{H}_1 = S^{3/2} \sum_{i,j} \frac{i}{2} J(\mathbf{d}_{ij}) \sin(\mathbf{Q} \cdot \mathbf{d}_{ij})(a_i^+ - a_i + a_j - a_j^+)$$

Quadratic form

2 operator term:

$$\mathcal{H}_2 = S\sum_{ij}\frac{1}{2}J(\mathbf{d}_{ij})\Big\{(1-\cos(\mathbf{Q}\cdot\mathbf{d}_{ij}))(a_i a_j + a_i a_j) +$$
$$+ (1+\cos(\mathbf{Q}\cdot\mathbf{d}_{ij}))(a_i a_j^+ - a_i^+ a_j) - 2\cos(\mathbf{Q}\cdot\mathbf{d})(a_i^+ a_i + a_j^+ a_j)\Big\}$$

3 operator term:

- non-zero in non-collinear structures
- changes the ground state

4 operator term:

- renormalizes the magnon dispersion
- gives finite magnon lifetime

Diagonalization of the quadratic form

To separate the mixing of the operators on different sites, we introduce a Fourier transformation:

$$a_i = \frac{1}{\sqrt{L}} \sum_{\mathbf{k} \in B.Z.} a(\mathbf{k}) e^{-i\mathbf{k}\cdot\mathbf{r}_i}$$

$$a(\mathbf{k}) = \frac{1}{\sqrt{L}} \sum_i a_i e^{i\mathbf{k}\cdot\mathbf{r}_i}$$

After substitution:

$$\mathcal{H}_2 = \frac{S}{2} \sum_{\mathbf{k}} \left(J(\mathbf{k}) - \frac{J(\mathbf{k}+\mathbf{Q}) + J(\mathbf{k}-\mathbf{Q})}{2} \right) \left(a_{\mathbf{k}} a_{-\mathbf{k}} + a^+_{\mathbf{k}} a^+_{-\mathbf{k}} \right) + \\ + \left(J(\mathbf{k}) + \frac{J(\mathbf{k}+\mathbf{Q}) + J(\mathbf{k}-\mathbf{Q})}{2} \right) \left(a_{\mathbf{k}} a^+_{\mathbf{k}} + a^+_{\mathbf{k}} a_{\mathbf{k}} \right) - 4J(\mathbf{Q}) a^+_{\mathbf{k}} a_{\mathbf{k}}$$

Diagonalization of the quadratic form

In matrix form:

$$\mathcal{H}_2 = \sum_{\mathbf{k}} \mathbf{x}^\dagger \mathrm{H} \mathbf{x}$$

Vector of boson operators:

$$\mathbf{x} = \begin{bmatrix} a_{\mathbf{k}} \\ a^{+}_{-\mathbf{k}} \end{bmatrix}$$

Matrix of the Hamiltonian:

$$\mathrm{H} = \begin{bmatrix} A & B \\ B & A \end{bmatrix}$$

$$A = J(\mathbf{k}) + J(\mathbf{k}+\mathbf{Q})/2 + J(\mathbf{k}-\mathbf{Q})/2 - 2J(\mathbf{Q})$$

$$B = J(\mathbf{k}) - J(\mathbf{k}+\mathbf{Q})/2 - J(\mathbf{k}-\mathbf{Q})/2$$

Diagonalization of the quadratic form

New operator b with the following transformation:

$$\begin{aligned} b =& ua + va^+ \\ b^+ =& ua^+ + va \end{aligned}$$

The new operator has to fulfill the commutation relations: $[b, b^+] = 1$
$\rightarrow u^2 + v^2 = 1$
With the right parameter choice:

$$\mathcal{H}_2 = \sum_{\mathbf{k}} \omega_{\mathbf{k}} \left(b^+ b + \frac{1}{2} \right)$$

And the spin wave dispersion:

$$\omega_{\mathbf{k}} = \sqrt{A^2 - B^2}$$

The above calculation is equivalent to solving the eigenvalue problem of gH, where $g = [\mathbf{x}, \mathbf{x}^\dagger]$ commutator matrix.

Spin-spin correlation function

Neutron scattering cross section of interacting spin systems:

$$\frac{d^2\sigma}{d\Omega dE} = C \cdot F^2(\mathbf{k}) \sum_{\alpha\beta} \left(\delta_{\alpha\beta} - \hat{k}_\alpha \hat{k}_\beta\right) S^{\alpha\beta}(\omega, \mathbf{k})$$

Correlation function:

$$S^{\alpha\beta}(\omega, \mathbf{k}) = \int_{-\infty}^{\infty} \sum_{i,j} dt \exp(-i\omega t) \langle S_i^\alpha S_j^\beta(t) \rangle$$

Out-of-plane correlation function

Correlation function:

$$\begin{aligned} S^{xx}(t,\mathbf{k}) =& \frac{1}{L^2}\sum_{i,j}\langle S_i^x S_j^x(t)\rangle e^{i\mathbf{k}\cdot\mathbf{d}_{ij}} \\ =& \frac{S}{2}\sum_{\mathbf{k}}\langle a_{-\mathbf{k}}^+ a_{\mathbf{k}}^+(t) + a_{-\mathbf{k}}a_{\mathbf{k}}(t) + a_{\mathbf{k}}^+ a_{\mathbf{k}}(t) + a_{\mathbf{k}}a_{\mathbf{k}}^+(t)\rangle \end{aligned}$$

After conversion into b and b^+ operators and Fourier transformation in time:

$$S^{xx}(\omega,\mathbf{k}) = S\frac{A_{\mathbf{k}} - B_{\mathbf{k}}}{\omega_{\mathbf{k}}}\cdot\delta(\omega-\omega_{\mathbf{k}})\cdot(n_\omega+1)$$

using:

$$\langle b_{\mathbf{k}}^+ b_{\mathbf{k}}(t)\rangle = n_\omega e^{-i\omega_{\mathbf{k}}t}$$

In-plane correlation function

Correlation function:

$$S_i^y S_j^y(t) = -\frac{S}{2}\sum_{i,j}\cos(\mathbf{Q}\cdot\mathbf{d}_{ij})\langle a_i^+ a_j^+(t) + a_i a_j(t) - a_i a_j^+(t) - a_i^+ a_j(t)\rangle$$

After Fourier transform the $\cos(\mathbf{Q}\cdot\mathbf{d}_{ij})$ brings in a $\pm\mathbf{Q}$ shift in reciprocal space:

$$S^{yy}(\omega,\mathbf{k}) = S^{zz}(\omega,\mathbf{k}) = \frac{1}{4}\Big(S^{\xi\xi}(\omega,\mathbf{k}-\mathbf{Q}) + S^{\xi\xi}(\omega,\mathbf{k}+\mathbf{Q})\Big)$$

Where the correlation function in the rotating frame:

$$S^{\xi\xi}(\omega,\mathbf{k}) = S\frac{A_{\mathbf{k}} + B_{\mathbf{k}}}{\omega_{\mathbf{k}}}\cdot\delta(\omega-\omega_{\mathbf{k}})\cdot(n_\omega+1)$$

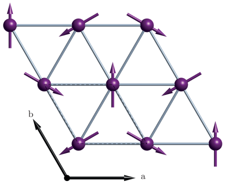
Triangular lattice antiferromagnet

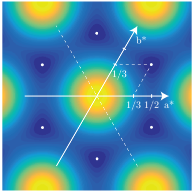
Exchange couplings:

$$J(\mathbf{k}) = \sum_{i=1}^{3} J \cos(\mathbf{k} \cdot \mathbf{e}_i)$$

The three bond vectors:

$$\begin{aligned} \mathbf{e}_1 &= (1, 0) \\ \mathbf{e}_2 &= (-1/2, \sqrt{3}/2) \\ \mathbf{e}_3 &= (-1/2, -\sqrt{3}/2) \end{aligned}$$

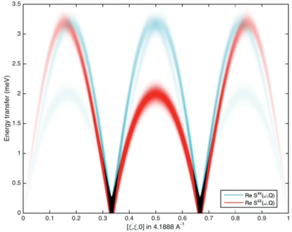




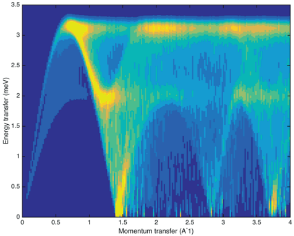
Classical ground state: $\mathbf{Q} = (1/3, 1/3)$

Triangular lattice antiferromagnet

Spin-spin correlation function



Powder spectrum



red: out-of-plane correlations (phason mode)

blue: in-plane correlations

Goldstone modes at $\pm\mathbf{Q}$

- due to spontaneous breaking of continuous symmetry

Presentation Outline

General spin Hamiltonian

General spin Hamiltonian:

$$\mathcal{H} = \sum_{ni,mj} \mathbf{S}_{ni}^{\intercal} \mathrm{J}_{ni,mj} \mathbf{S}_{mj} + \sum_{ni} \mathbf{S}_{ni}^{\intercal} \mathrm{A}_{ni} \mathbf{S}_{ni} + \mu_B \mathbf{H}^{\intercal} \sum_{ni} \mathrm{g}_{ni} \mathbf{S}_{ni}$$

Anisotropic and antisymmetric (Dzyaloshinskii-Moriya) exchange interactions:

$$\mathrm{J}_H = \begin{bmatrix} J & 0 & 0 \\ 0 & J & 0 \\ 0 & 0 & J \end{bmatrix}; \quad \mathrm{J}_{DM} = \begin{bmatrix} 0 & D^z & -D^y \\ -D^z & 0 & D^x \\ D^y & -D^x & 0 \end{bmatrix}$$

General spin Hamiltonian

Non-Bravais lattice:

- additional rotation on every site within unit cell

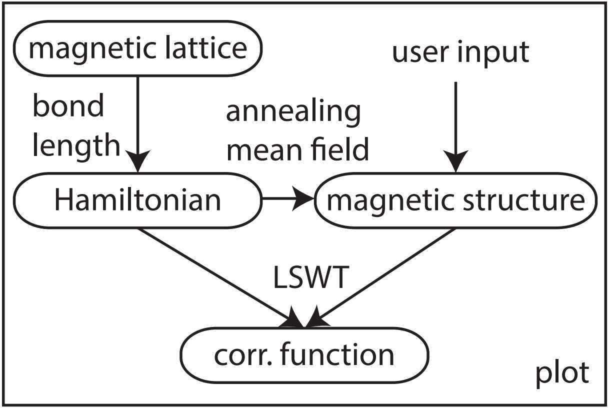
General interactions:

- multi-q magnetic ground states are possible

SpinW

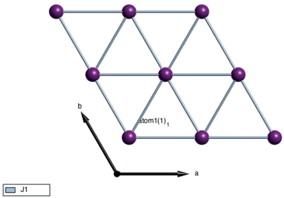
- solves the general spin Hamiltonian
- calculates spin-spin correlation function
- numerical and symbolical
- can apply crystal symmetry operators on the Hamiltonian
- solves single-q magnetic structures
- solves multi-q magnetic structures on a magnetic supercell
- open source, runs on Matlab
- download: `http://www.psi.ch/spinw`

SpinW workflow

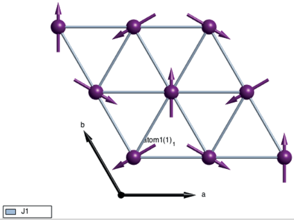


SpinW: triangular lattice AF

```
tri = sw;
tri.genlattice('lat_const',[3 3 4],'angled',[90 90 120])
tri.addatom('r',[0 0 0],'S',1)
```

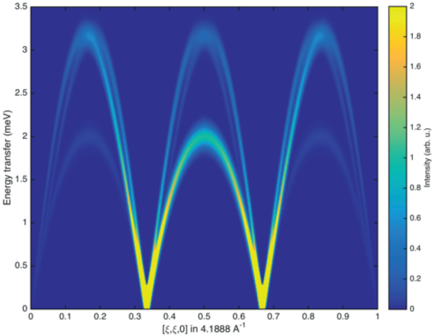



```
tri.addmatrix('value',1,'label','J')
tri.addmatrix('value',diag([0 0 0.2]),'label','D')
tri.gencoupling
tri.addcoupling('J',1)
tri.addaniso('D')
tri.genmagstr('mode','helical','S',[0; 1; 0],...
'k',[1/3 1/3 0],'n', [0 0 1])
```



SpinW: triangular lattice AF

```
spec = tri.spinwave({[0 0 0] [1 1 0] 500});
spec = sw_neutron(triSpec);
spec = sw_egrid(spec);
```



Presentation Outline

What comes...

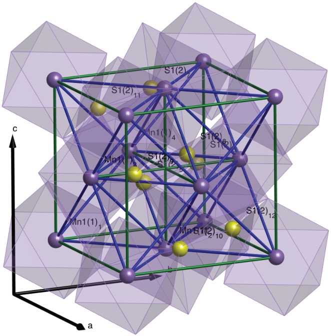
After the break:

- hands on SpinW
- analyze the magnetic order and spin waves on the FCC lattice
- preparation to the experiment on TASP

On the weekend (for those who registered):

- neutron scattering on MnS_2 FFC antiferromagnet
- elastic magnetic scattering
- we will measure spin wave dispersion, maybe diffuse scattering
- improve on this old data:
  T. Chattopadhyay, et al., Phys. B Cond. Mat. 156, 241 (1989).

MnS_2



MnS_2 crystal structure with spin interactions

Single crystal of MnS_2 (hauerite).

Presentation Outline

Summary

- Origin of atomic magnetism and magnetic interactions in solids
- Classically there is always a spin wave solution
- Example: ferromagnetic chain
- Quantum mechanically:
- Equivalence with coupled quantum harmonic oscillators
- Frustration leads to spiral ground state
- Calculation of spin wave dispersion of a single-Q spiral
- Introduction to SpinW
- References:

R. Skomski: Simple models of magnetism
N. Majlis: The quantum theory of magnetism
G. Squires: Introduction to the theory of thermal neutron scattering
ST and Bella Lake: Linear spin wave theory for single-Q magnetic structures, JPCM, 16, 166002 (2015), ArXiv:1402.6069

Thank you!