Analytical Linear Spin-wave Theory Analysis of LaMnO3

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1. Introduction.

This tutorial shows one path to analytically study magnon excitations in magnetically ordered quantum spin systems. Here, we use the theoretical framework called linear spin-wave theory (LSWT), for which the book “Spin-wave Theory and its Applications to Neutron Scattering and THz Spectroscopy” is a good reference. [1] Central to our discussion will be the Holstein-Primakoff transformation, a technique that transforms our quantum spin problem into a more tractable bosonic representation. By doing this, we can effectively turn a complex interacting spin problem into a simpler problem of non-interacting bosons.

The steps we'll cover are:

1. Spin Hamiltonian. Representing our spin system via a suitable Hamiltonian. This includes defining the magnetic structure and the local coordinate systems for the spin operators.
2. Bosonic Hamiltonian. Applying the Holstein-Primakoff transformation to rewrite this Hamiltonian in the language of bosonic operators. Linearizing the bosonic Hamiltonian.
3. Fourier transformed Hamiltonian. Using the discrete Fourier transform to move our problem from real space to momentum space, leveraging the translational symmetries of our system.
4. Magnon modes. Diagonalizing our momentum-space Hamiltonian (a generalized Bogoliubov transformation) to a set of magnon bosons, which allows calculation of the magnon dispersion.
5. Additional observables. Using the solution to calculate the LSWT correction to the ordered moment.
6. Spin Hamiltonian.
7. The real-space spin Hamiltonian of LaMnO3

The material chosen is LaMnO3 (LMO), a pseudo-perovskite manganite A-type antiferromagnet. [2] The oxidation states of the ions are La3+Mn3+O2-3, such that the magnetic term of Mn3+ is the Jahn-Teller active 3d4 5Eg and S = 2. The crystallographic lattice in the magnetic state is defined in Table I. The magnetic structure is visualized in Figure 1.

Table I. Crystallographic parameters of LaMnO3 at T = 1.4 K. [3]

|  |  |  |  |
| --- | --- | --- | --- |
| Atom (Wyckoff site) | x | y | z |
| La (4c) | -0.0095(5) | 0.0513(7) | 0.25 |
| Mn (4b) | 0.5 | 0 | 0 |
| O1 (4c) | 0.0777(7) | 0.48493(80) | 0.25 |
| O2 (8d) | 0.7227(5) | 0.3085(5) | 0.0408(4) |
| a = 5.5333 Å, b = 5.7461 Å, and c = 7.6637 Å  Space group = *Pbnm* | | | |

The Heisenberg model Hamiltonian with single-ion anisotropy may be expressed as

|  |  |
| --- | --- |
|  | (1) |

where the notation indicates a sum over nearest neighbors, the sum is over all N sites in the lattice, is the exchange, and is the uniaxial single-ion anisotropy. When dealing with sums in the Hamiltonian, remember the goal is to keep track of all the site energies and pair-wise energies. The simplest form of (1) that stabilizes an A-type antiferromagnet may be expressed as

|  |  |
| --- | --- |
|  | (2) |

where is superexchange within the ab-plane, is superexchange along the c-axis, is a sum over nearest neighbors in the ab-plane, and is a sum over nearest neighbors along the c-axis.

|  |  |
| --- | --- |
|  |  |
| Figure 1. This is the magnetic structure directly from reference Moussa et al. [3] showing only the manganese atoms and their dipole moments. |  |

First, the magnetic structure has two sublattices (denoted and ). And equation 2 is more explicitly

|  |  |
| --- | --- |
|  | (3) |

1. Local coordinate systems

Each sublattice has a local coordinate system for which the quantization axis is along the observed dipole moment direction. Start with a global coordinate system defined as x∥c, y∥a, and z∥b, which may be used for one of the sublattices. Then, the antiparallel sublattice with have a rotated coordinate system. The mapping from the global to local coordinate systems is then

|  |  |
| --- | --- |
| , , | (4) |

and

|  |  |
| --- | --- |
| , , | (5) |

where here the local reference frame is the over-barred operator.

1. Bosonic Hamiltonian.
2. Holstein-Primakoff transformation

Now, a Holstein-Primakoff (HP) transformation can be introduced to change from spin operators to HP boson operators

|  |  |
| --- | --- |
|  | (6) |
|  |  |
|  |  |

and

|  |  |
| --- | --- |
|  | (7) |
|  |
|  |

where and are the two different bosons, and is either a site index or an imaginary number depending upon context. The plus and minus operators have been linearized in preparation for solving, while the z operator is bilinear.

Plugging equations 6 and 7 into the Hamiltonian yields (remembering )

|  |  |
| --- | --- |
|  | (8) |

1. Linearized bosonic Hamiltonian

To get a system of non-interacting bosons, equation 8 must be further (bi)linearized for the z operators, and this is the point where terms like uniaxial single-ion anisotropy (remember up until here z operators were exact) will incorrectly capture the energy spectrum of the local units but may be renormalized.

|  |  |
| --- | --- |
|  | (9) |

Now there is a site independent “zero point energy” from the terms

|  |  |
| --- | --- |
|  | (10) |

and the direct space Hamiltonian of non-interacting bosons of order

|  |  |
| --- | --- |
|  | (11) |

1. Fourier transformed Hamiltonian.
2. Fourier transformed HP operators

The next step in finding a solution is to introduce Fourier transformed HP boson operators ()

|  |  |  |
| --- | --- | --- |
|  |  | (12) |
|  |  |

and

|  |  |  |
| --- | --- | --- |
|  |  | (13) |
|  |  |

where the BZ denotes that momentum sums are over the Brillouin zone (BZ); be careful, as the BZ is for the magnetic cell. Formally, the momenta in these transforms are quantized, so for a 1-d case with one spin per unit cell , where , and is the lattice spacing.

1. A pair-wise portion of the HP Hamiltonian

Now, begin Fourier transforming the operator containing portion of the first part of

|  |  |
| --- | --- |
|  | (14) |

and plugging in the Fourier transformed operators yields

|  |  |
| --- | --- |
|  | (15) |

To simplify the summations, we will make use of the orthogonality theorem (the Kronecker delta function)

|  |  |
| --- | --- |
|  | (16) |

1. Deriving the Fourier transform of a site operator

Consider first the term, which can be cast as a sum over site indices, recognizing there are two neighbors in the plane per site when tiling

|  |  |
| --- | --- |
|  | (17) |

Expanding and re-ordering the sums

|  |  |
| --- | --- |
|  | (18) |

And invoking the orthogonality theorem

|  |  |
| --- | --- |
|  | (19) |

1. Deriving the Fourier transform of a pair-wise operator

Now let’s look at the term

|  |  |
| --- | --- |
|  | (20) |

At this point, we can’t cheat as if it was an on-site term and we must explicitly parameterize the abstract nearest neighbor definition to arrive at a quantitative and specific solution. Denote the site by coordinates and the site by coordinates . The nearest neighbors of site are and . Using the global cartesian coordinate system above (, , ) and the definition of the Mn positions from Table I

|  |  |
| --- | --- |
|  | (21) |

And the Fourier transform relations as a function of are

|  |  |
| --- | --- |
|  | (22) |
|  |

and then (introduce a ½ as here bonds are double counted by going both positive and negative direction neighbors)

|  |  |
| --- | --- |
|  | (23) |

Plugging in the Fourier transformed operators

|  |  |
| --- | --- |
|  | (24) |

Then, each term may be collapsed by a Kronecker delta, but with an added phase factor

|  |  |
| --- | --- |
|  | (25) |

This further simplifies to

|  |  |
| --- | --- |
|  | (26) |

1. The full Fourier transformed Hamiltonian

Analogous calculations can be done for the other terms, yielding

|  |  |
| --- | --- |
|  | (27) |

And commuting the first operator term

|  |  |
| --- | --- |
|  | (28) |

For the magnon dispersion relation, this constant offset from commuting the operators has no effect, as transitions are from the ground state to the first excited state, however, one must keep track of the constant term as it applies to the energy, which can be used to look at how the LSWT correction to the system effects other observables (such as isothermal, high-field magnetization because ). Looking forward, there will be commuting and grouping of operators depending upon the operation being considered, so book-keeping is important.

Finally, this recipe yields a Fourier space Hamiltonian

|  |  |
| --- | --- |
|  | (29) |

where

|  |  |
| --- | --- |
|  | (30) |
|  | (31) |

1. Magnon modes.

From here, we perform a para-unitary transformation to a diagonal basis. Classic papers to consider are those of Colpa [4,5] and White, Sparks, and Ortenburger. [6] Aside from these and the aforementioned book, there is a practical presentation for a bi-partite antiferromagnet in Reference [7]. Keep in mind that it is not explicitly stated in the formulation of Reference [7] that the their Hamiltonian terms are real, which can be violated for a non-Bravais lattice like the honeycomb.

The calculation is organized by first representing as matrix.

|  |  |
| --- | --- |
|  | (32) |

where

|  |  |
| --- | --- |
| , | (33) |

So, in order to represent the Hamiltonian for LaMnO3 as a matrix, we symmetrize to include terms with , such that

|  |  |
| --- | --- |
|  | (34) |

to yield

|  |  |
| --- | --- |
|  | (35) |

This sets the stage to introduce a new set of operators by a linear (Bogoliubov) transformation

|  |  |
| --- | --- |
|  | (36) |

where is diagonal in the new basis. And

|  |  |
| --- | --- |
| , | (37) |

There is an additional constraint on the transformation, which is to preserve the bosonic commutation relations. This is achieved by multiplication by

|  |  |
| --- | --- |
| , such that and | (38) |

to yield the non-Hermitian dynamical matrix

|  |  |
| --- | --- |
|  | (39) |

This matrix is diagonalizable

|  |  |
| --- | --- |
|  | (40) |

The is diagonal with real eigenvalues and , and the matrix formulation can be written out by commuting and terms to yield

|  |  |
| --- | --- |
|  | (41) |

which is the form of the familiar simple harmonic oscillator that can be solved using the number basis. In the low-temperature limit, the magnon population can be taken as a Bose-Einstein distribution. Finally, there is a doubly degenerate magnon mode (one for each sublattice) that corresponds to the transition from the ground state to the first excited state.

|  |  |
| --- | --- |
|  | (42) |
|  | (43) |

Having arrived at a solution, it is important to apply a sanity check. If the anisotropy is zero, then there should be a Goldstone mode at the ordering wave vector.

Note that the LaMnO3 reference has a factor of 2 different in their definition of the exchange energies.

1. Additional Observables

Recall that in the HP transformation

|  |  |
| --- | --- |
|  | (45) |

This means that we can compute the LSWT correction to the ordered moment of the ground state by

|  |  |
| --- | --- |
|  | (46) |

To evaluate this sum, the HP bosons must be transformed to magnon operators using . From the diagonalization process, it can be shown that

|  |  |
| --- | --- |
|  | (41) |

|  |  |
| --- | --- |
|  | (42) |

Then the transformation is possible between magnons and HP bosons.

|  |  |
| --- | --- |
|  | (47) |

writing this out

|  |  |
| --- | --- |
|  | (48) |

Remember that we are taking the expectation value in the ground state, so the correction to the ordered moment is then

|  |  |
| --- | --- |
|  | (49) |
|  | (50) |

Evaluating the sum (in the limit ) yields , so or , which is comparable to the experimentally determined . [3]

REFERENCES:

[1] R. S. Fishman, J. A. Fernandez-Baca, and T. Rõõm‬, *Spin-Wave Theory and Its Applications to Neutron Scattering and THz Spectroscopy* (Morgan and Claypool, San Rafael, CA, 2018).

[2] E. O. Wollan and W. C. Koehler, Phys. Rev. **100**, 545 (1955).

[3] F. Moussa, M. Hennion, J. Rodriguez-Carvajal, H. Moudden, L. Pinsard, and A. Revcolevschi, Phys. Rev. B **54**, 15149 (1996).

[4] J. H. P. Colpa, Phys. A Stat. Mech. Its Appl. **134**, 377 (1986).

[5] J. H. P. Colpa, Phys. A Stat. Mech. Its Appl. **134**, 417 (1986).

[6] R. M. White, M. Sparks, and I. Ortenburger, Phys. Rev. **139**, (1965).

[7] S. M. Rezende, A. Azevedo, and R. L. Rodríguez-Suárez, J. Appl. Phys. **126**, (2019).