The Emergence of Non-coplanar Magnetism in Non-Bravais Lattices

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Abstract

In this paper we consider the problem of finding the magnetic properties of crystalline materials. Starting from a model of two hydrogen atoms, we motivate the Heisenberg Hamiltonian, $H_H = -\sum J_{ij} s_i \cdot s_j$, which is the fundamental equation governing the materials considered here. Essentially, the Heisenberg Hamiltonian says that pairs of spins interact with energy linear in their dot product. These pairings will depend upon the structural properties of the crystal lattice, while the proportionality constant is due to the chemical/electronic properties of the materials. To simplify our analysis, we treat the spins as unit vectors localized to their lattice sites. Given this setup, we search for specific combinations of structures and couplings $J_{ij}$ that will produce non-coplanar magnetic order, where the spins span all directions of spin space.

In this process, we will construct several simple yet novel lattices. These structures are part of a class of lattices known colloquially as non-Bravais lattices, which are structures where the basic translational unit (or unit cell) contains more than one lattice site. To determine the ground state of the Heisenberg Hamiltonian (that is, to minimize it), we use three different techniques, as none are entirely satisfactory when applied to non-Bravais lattices. The Luttinger-Tisza method, which minimizes the Hamiltonian as a function of wavelength (i.e., over the Brillouin zone), can always give an analytic solution for a Bravais lattice. But the solutions it predicts for non-Bravais lattices often fail to satisfy the unit spin constraint. To find the ground state by Luttinger-Tisza analysis, then, we must piece together several different (sub-optimal) solutions by hand. This method is significantly more difficult to apply than our second technique, the iterative minimization algorithm. In this approach, we simulate a lattice with initially random spins and then allow it to relax to its ground state. The relaxation method used here is to randomly set spins anti-parallel to their effective field, $-\sum J_{ij} s_j$. This produces successively more accurate approximations of the ground state. To make these approximations more accurate, we use them as the basis of our final technique - variational optimization. This idealizes the results of the simulations, and then analytically minimizes the Hamiltonian given the idealized structure. In doing so, we also find an analytic parametrization of the spin configuration, thus giving an explicit ground state.

The first structure that we analyze is the octahedral lattice (a three dimensional structure with three lattice sites per unit cell), where we find a number of distinct coplanar states and a handful of non-coplanar ones. Of these non-coplanar states, two are of particular importance, which we call the $\pi/3$ and $2\pi/3$ cuboctahedral states. These states are important because they are special cases
of a more generic, highly degenerate state. As we will argue, viewing a combination of couplings as selecting out a particular state from the highly degenerate set is a good way to construct new non-coplanar states. Also, the cuboctahedral states are constructed solely of wave-vectors related by symmetry. We call such states one-star states, and they are particularly fruitful as a source of non-coplanar states. In particular, the double-twist spiral state, which we could only stabilize on the phase boundary of two coplanar states, is another non-coplanar, one-star ground state. In addition, we also found one non-coplanar ground state of the octahedral lattice that was not a one-star state. This was an asymmetric conic state, which was also only found on the phase boundary of coplanar states. More important, this ground state was a function of only one variable.

Because the asymmetric conic state is a function of one spatial variable and we want to stabilize it outside of the phase boundary motivates us to turn to the chain lattice. This structure is constructed by projecting the octahedral lattice along a vector (it’s one dimensional with two lattice sites per unit cell). In this lattice, we manage to stabilize the asymmetric conic, as well as a new non-coplanar state, the alternating conic. These states are neither part of a highly degenerate family of states or one-star states, so the perspectives that are important to understanding non-coplanar states in the octahedral lattice do not apply here. Instead, we argue that these states should be understood in terms of encompassing parametrizations. An encompassed parametrization is a state that can be represented as a particular case of a more general state, called the encompassing parametrization. This is important encompassing parametrizations allow the ground state to continuously deform between two orthogonal states (as a function of couplings). If the two orthogonal states are coplanar, then the encompassing parametrization will necessarily be non-coplanar, providing us with another means of constructing non-coplanar states

Finally, in addition to the these two techniques for deriving non-coplanar states (degenerate families of states and encompassing parametrizations), which are based off of the results of specific lattices, we consider how the results of the Luttinger-Tisza method could be used to motivate non-coplanar states. This approach is more generic than the other two, but less explicit in its prescriptions.
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References
I. INTRODUCTION

The magnetic properties of materials is a source of seemingly endless variation. Even the simplest types of magnetic order belie the astounding complexity of their physical origin. From these simple types of magnetism, we create simple models of the behavior of magnetic systems, models which abstract away from the underlying physics. And from these simple models there emerges a plethora of different magnetic orders. This proliferation of magnetic orders matches those encountered in nature, at least in most cases.

A. The Physical Origin of Rudimentary Magnetic Interactions

Although magnetic materials is a topic discussed in undergraduate courses on electromagnetism, such a picture barely explains paramagnetism and diamagnetism. To explain even something as common as the household magnet (or ferromagnet), we require a quantum mechanical picture of the electron. To see why this is the case, consider the model of magnetic order as arising from atomic-scale magnetic dipoles. From a classical perspective, this is not a bad picture, as the electron circles about the nucleus fast enough to be considered a current loop. The interaction energy of two dipoles separated by a distance $\vec{r}$ is

$$U_B = \frac{\mu_0}{4\pi} \frac{\vec{m}_1 \cdot \vec{m}_2 - 3(\vec{m}_1 \cdot \hat{r})(\vec{m}_2 \cdot \hat{r})}{|\vec{r}|^3}. \quad (1.1.1)$$

For electrons, this interaction energy is about equal to the thermal energy at 2K [2]. According to a purely classical picture, then, we should not observe any magnetic interaction between atoms at room temperature.

We get a much better explanation of magnetic phenomena when we treat electrons quantum mechanically. In particular, electrons are fermions, so we must properly anti-symmetrize their wave-functions. We must also minimize the Coulomb repulsion between the two electrons, although satisfying the anti-symmetrization constraint takes precedence (the Coulomb repulsion is a monopole-monopole reaction, and therefore significantly stronger than dipole interactions at the atomic scale).

Consider two hydrogen atoms. Let the two nuclei be labeled 1 and 2. The electron spatial eigenfunctions are then $|1\rangle$ and $|2\rangle$, where the number denotes the nucleus the electron is localized around. These wave functions will have an overlap

$$l \equiv |\langle 1|2\rangle|, \quad (1.1.2)$$
which will be non-zero only when the nuclei are sufficiently close and the spatial wave-functions are non-orthogonal. In addition to the spatial eigenfunction, the electrons will also possess a spin state, which will either be the singlet state

$$|00\rangle = \frac{|\uparrow\rangle|\downarrow\rangle - |\downarrow\rangle|\uparrow\rangle}{\sqrt{2}}$$  \hspace{1cm} (1.1.3)$$

or one of the triplet states

$$|11\rangle = |\uparrow\rangle|\uparrow\rangle$$ \hspace{1cm} (1.1.4)$$
$$|10\rangle = \frac{|\uparrow\rangle|\downarrow\rangle + |\downarrow\rangle|\uparrow\rangle}{\sqrt{2}}$$ \hspace{1cm} (1.1.5)$$
$$|1-1\rangle = |\downarrow\rangle|\downarrow\rangle.$$ \hspace{1cm} (1.1.6)$$

In this notation, $|\uparrow\rangle|\downarrow\rangle$ means that the first electron has $\sigma_z = 1/2$, while the second electron has $\sigma_z = -1/2$. Since anti-symmetrization is required for the total wave-function, the spatial symmetry depends on whether the spin state is a singlet (anti-symmetric) or triplet (symmetric). Specifically, the spatial wave-functions are

$$|s\rangle = \frac{|1\rangle|2\rangle + |2\rangle|1\rangle}{\sqrt{2} + 2l^2}$$ \hspace{1cm} (1.1.7)$$
$$|t\rangle = \frac{|1\rangle|2\rangle - |2\rangle|1\rangle}{\sqrt{2} - 2l^2}.$$ \hspace{1cm} (1.1.8)$$

Furthermore, the Hamiltonian for this system is

$$H = \frac{p_1^2}{2m} + \frac{p_2^2}{2m} - \frac{e^2}{|\vec{r}_1 - \vec{R}_1|} - \frac{e^2}{|\vec{r}_2 - \vec{R}_2|}$$
$$+ \frac{e^2}{|\vec{r}_1 - \vec{r}_2|} + \frac{e^2}{|\vec{R}_1 - \vec{R}_2|}$$
$$- \frac{e^2}{|\vec{r}_1 - \vec{R}_2|} - \frac{e^2}{|\vec{r}_2 - \vec{R}_2|}$$ \hspace{1cm} (1.1.9)$$

where $p_i$ is the momentum of the $i^{th}$ electron, $\vec{r}_i$ is the electron’s displacement vector, and $\vec{R}_i$ is the displacement vector of the $i^{th}$ nucleus. We define

$$\langle 1|2\rangle H(|1\rangle|2\rangle) = \langle 2|1\rangle H(|2\rangle|1\rangle) = 2E_0 + U$$ \hspace{1cm} (1.1.10)$$
$$\langle 1\rangle|2\rangle \left( \frac{e^2}{|\vec{r}_1 - \vec{r}_2|} + \frac{e^2}{|\vec{R}_1 - \vec{R}_2|} - \frac{e^2}{|\vec{R}_1 - \vec{R}_2|} - \frac{e^2}{|\vec{r}_2 - \vec{R}_2|} \right) |1\rangle|2\rangle \equiv U$$ \hspace{1cm} (1.1.11)$$
$$\langle 1\rangle|2\rangle H(|2\rangle|1\rangle) = \langle 2|1\rangle H(|1\rangle|2\rangle) = 2E_0l^2 + V$$ \hspace{1cm} (1.1.12)$$
$$\langle 1\rangle|2\rangle \left( \frac{e^2}{|\vec{r}_1 - \vec{r}_2|} + \frac{e^2}{|\vec{R}_1 - \vec{R}_2|} - \frac{e^2}{|\vec{r}_1 - \vec{R}_2|} - \frac{e^2}{|\vec{r}_2 - \vec{R}_2|} \right) |2\rangle|1\rangle \equiv V$$ \hspace{1cm} (1.1.13)$$
to get the symmetric and antisymmetric energies:

\[ E_s = \langle s | H | s \rangle = 2 \frac{2E_0 + U + 2l^2E_0 + V}{2 + 2l^2} = 2E_0 + \frac{U + V}{1 + l^2} \]  
(1.1.14)

\[ E_t = \langle t | H | t \rangle = 2 \frac{2E_0 + U - 2l^2E_0 - V}{2 - 2l^2} = 2E_0 + \frac{U - V}{1 - l^2}. \]  
(1.1.15)

The difference between these energies is

\[ E_t - E_s = 2l^2U - V \frac{1}{1 - l^4} \equiv -J \]  
(1.1.16)

[2]. This difference in energy enforces either the symmetry or anti-symmetry of the electronic spins. For positive \( J \), the triplet is lower energy than the singlet, so the spins tend to be parallel (ferromagnetism). For negative \( J \), the singlet is lower energy than the triplet, so the spins tend to be anti-parallel (anti-ferromagnetism).

**B. Simple Models of Magnetic Interaction**

The Heitler-London calculation of the previous section captures certain salient features of magnetic interactions, but it is only an approximation (especially for macroscopic systems). Treating the wave-functions as linear combinations of atomic orbitals (LCAO) is part of the issue, as is the Coulomb potential, which quickly becomes unwieldy for large numbers of charged particles. In addition, the direct exchange interaction (Coulomb potential) is only part of the picture for macroscopic materials. There is also indirect exchange (where the interaction is mediated by conduction electrons) and superexchange (where the interaction is mediated neutral atoms) [2]. To capture the essential ground state symmetrization requirements while abstracting away from the specific form of subatomic interactions, we replace the spatial Hamiltonian (equation 1.1.9) with one that depends only spin [2]:

\[ H = 2E_0 + \frac{U - V}{1 - l^2} + \left( \frac{1}{4} - \vec{s}_1 \cdot \vec{s}_2 \right) J. \]  
(1.2.1)

This Hamiltonian results in the same predictions (depending on the sign of \( J \)) as Heitler-London model, but only includes two terms: a constant and a term that depends linearly on \( \vec{s}_1 \cdot \vec{s}_2 \). To extend this model to more than two atoms, we get

\[ H = H_0 - J \sum_{n.m.} \vec{s}_i \cdot \vec{s}_j, \]  
(1.2.2)
or the Ising model. Now the Ising model assumes that the only interactions are limited to nearest neighbors (n.n.), but we can just as easily generalize to the Heisenberg Hamiltonian [2, 3]:

\[ H_H = - \sum_{ij} J_{ij} \vec{s}_i \cdot \vec{s}_j \]  

(1.2.3)

(the overall constant is dropped as it is not physically meaningful). This Hamiltonian is basic equation that governs all the results we derive in this paper. In particular, our goal is to minimize this sum, thereby determining the ground state.

C. Building on the Heisenberg Hamiltonian.

For crystalline materials, the couplings between spins \((J_{ij})\) simplifies. Because of translational symmetry, we can group interactions as coupling nearest neighbors \((J_1)\), second nearest neighbors \((J_2)\), etc. In a Bravais lattice with one lattice site per unit cell (i.e. the periodic domain only includes one atom), this classification scheme is sufficient. But in a lattice with a basis (a Bravais lattice with more than lattice site per unit cell, colloquially known as a non-Bravais lattice), there may be exceptions to this rule. In this paper, we will encounter two types of exceptions, depending upon how the sublattices are defined (any non-Bravais lattice can be decomposed into a series of interpenetrating Bravais lattices, or sublattices). In the octahedral lattice (section IV), the couplings in different sublattices are equivalent, but, because the definitions of the sublattices introduces an anisotropy, not all equidistant sites are equivalent. In the chain lattice (section V), there is no such anisotropy, but the couplings of different sublattices are inequivalent. In each of these cases, we distinguish between different couplings of equidistant pairs by using primes (e.g. \(J_2\) vs \(J'_2\)). In defining couplings in this way, we connect the magnetic order with the physical structure.

Since the structures we examine are complicated and we will include many different types of couplings, the problem of minimizing the energy of the Heisenberg Hamiltonian (and thereby determining the ground state) quickly becomes hard. To simplify the problem, it is helpful to turn to a semi-classical model of magnetism. While we still use the Heisenberg Hamiltonian, which comes from a quantum mechanical picture of the electron’s spin, we treat the spin as a classical vector. In particular, we use a hard spin model, where these vectors are constrained to be unit vectors. In this model, all of the details of the magnitude
of the spins are absorbed into the magnitude of $J_{ij}$, so we can concentrate upon the angle between spins $s_i$ and $s_j$.

While applying these approximations to an already simplified Hamiltonian may seem highly artificial, this model is still useful. It is in good agreement with experimental results on crystalline systems. For these materials, the quantum nature of the electron is not integral to explaining its magnetic properties - the hard spin Heisenberg Hamiltonian will produce an identical ground state [2]. As such, our efforts to create novel forms of magnetic ground states applies directly to understanding the magnetic order displayed by real materials [21].

In determining the ground states for some lattice as a function of its couplings $J_{ij}$, we will focus upon finding a particular type of magnetic order: the non-coplanar ground state. A state like a ferromagnet is called collinear, because all the spins lie along a single line in spin space. Similarly, the anti-ferromagnetic state, where spins alternate direction (say, pointing up - pointing down - pointing up, etc) is also collinear. The helimagnetic state, where the spins on different sites trace out a spiral, is a case of coplanar spins. That is, all of the spins in a lattice with helimagnetic order lie within a single plane. All three of these configurations are well-documented and understood, but the same cannot be said about non-coplanar states. When the spins do not all lie within a single plane, but instead fill all three dimensions of spin space, a great many more types of magnetic order become available. The work presented in this paper is the first effort at giving an organized analysis of the non-coplanar states [22]- what types are possible, how they arise from the couplings, and how to predict them.

The analysis of non-coplanar states is interesting both as an unexplored instance of magnetism and emergent order and for its practical applications. Much of modern technology is based upon the understanding and control of magnetic properties (in particular, conventional digital storage technology exploits magnetic order). As we discover new forms of magnetic order, it becomes easier to improve upon existing implementations of magnetic devices. In particular, we will encounter a ground state called the “asymmetric conic,” which has some potential to be the magnetic ordering of next generation multiferroic materials [4–7].
II. METHODS

In the general case, determining the magnetic ground state from the Heisenberg Hamiltonian is quite difficult. To approach this problem, we relied upon several different techniques to find the ground state. In section 1, we outline the Luttinger-Tisza [8, 9] method for determining ground states. This is the most mechanical approach, but is of less utility in non-Bravais lattices. Section 2 focuses on an iterative minimization algorithm, a numerical technique that produces increasingly accurate approximations of the ground state. The states found through this technique are idealized and used to inform a variational approach to the minimum, as discussed in section 3. Variational optimization has the advantage that it relies less heavily upon numerics than the iterative minimization and is more flexible than Luttinger-Tisza, but it is limited by the ingenuity of state to be optimized. To aid in the categorization of states found in the iterative minimization algorithm and prepare them for variational analysis, we created a variety of diagnostic metrics, which are described in section 4.

A. The Luttinger-Tisza Method

The Luttinger-Tisza method [8, 9] is essentially a spatial Fourier Transform of the Heisenberg Hamiltonian. Just as for every finite set of basis vectors there is a finite set of one-forms, so for every unit cell there is a finite region of reciprocal space that completely characterizes all possible wave-vectors (up to an alias). This unit cell of reciprocal space is called the Brillouin Zone, and is related to unit cell by a Fourier Transform. In writing wave-vectors in the Brillouin Zone, there will always be a factor of $2\pi/V$, where $V$ is the volume of the unit cell. This factor is normally left implicit. So, if we had a unit cell of volume 5 (in some units) and the wave-vector $(\pi/5, \pi/5, 0)$ would be written $(1/2, 1/2, 0)$.

Examining the possible solutions to the Heisenberg Hamiltonian in the Brillouin Zone, rather than in real space, is useful because the ground state will necessarily display a spatial pattern composed of wave-vectors from the Brillouin Zone. So, in theory, we have reduced the problem of analyzing an infinite system to analyzing a function on a finite domain. Unfortunately, as we will see, there is some subtlety here that complicates the problem.

The Luttinger-Tisza equation is sufficiently simple to derive that we can do it here. We
begin with the Heisenberg Hamiltonian and take Fourier Transforms

\[ H_H = -\sum_{ij} J(\vec{r}_{ij}) \vec{s}(\vec{r}_i) \cdot \vec{s}(\vec{r}_j) \]  

(2.1.1)

\[ = -\sum_{ij} \left( \sum_{q_1} J(q_1^i) e^{-i\vec{q}_1 \cdot \vec{r}_{ij}} \right) \left( \frac{1}{\sqrt{N}} \sum_{q_2} \vec{s}(q_2^i) e^{i\vec{q}_2 \cdot \vec{r}_i} \right) \cdot \left( \frac{1}{\sqrt{N}} \sum_{q_3} \vec{s}(q_3^j) e^{i\vec{q}_3 \cdot \vec{r}_j} \right) \]  

(2.1.2)

\[ = -\frac{1}{N} \sum_{ij} \sum_{q_1} \sum_{q_2} \sum_{q_3} J(q_1^i) \vec{s}(q_2^i) \cdot \vec{s}(q_3^j) e^{i(\vec{q}_2 - \vec{q}_1) \cdot \vec{r}_i} e^{i(\vec{q}_3 + \vec{q}_1) \cdot \vec{r}_j} \]  

(2.1.3)

\[ = -\frac{1}{N} \sum_{q_1} \sum_{q_2} \sum_{q_3} J(q_1^i) \vec{s}(q_2^i) \cdot \vec{s}(q_3^j) N \delta_{q_1^{q_2}} \delta_{q_1^{q_3}} \]  

(2.1.4)

\[ = -\sum_{q} J(q) |\vec{s}(q)|^2 \]  

(2.1.5)

where we used \( \vec{r}_{ij} = \vec{r}_i - \vec{r}_j \) and the orthogonality of the Fourier Transform. Typically, normalization and the structure of the ground state allow us to drop the \( \vec{q} \) dependence of \( |\vec{s}(\vec{q})|^2 \) and pull it out of the sum. In this case, minimizing the Hamiltonian reduces to minimizing \( J(\vec{q}) \). Since a (not-necessarily unique) minimum \( \vec{q}_m \) must exist, we can construct a solution of the form

\[ \vec{s}(\vec{r}_i) = \cos(\vec{q}_m \cdot \vec{r}_i) \hat{n}_1 + \sin(\vec{q}_m \cdot \vec{r}_i) \hat{n}_2 \]  

(2.1.6)

assuming that the crystal displays inversion symmetry (i.e. \( J(-\vec{q}) = J(\vec{q}) \)). If we can construct solutions of this form, then \( |\vec{s}(q)|^2 \) really is a constant (1 because we work with normalized spins), so our approach is justified.

The derivation of the Luttinger-Tisza equation presented here is limited because we implicitly assumed that all sites are equivalent, i.e. that the lattice was a Bravais lattice. For non-Bravais lattices, the solution is actually the generalized Luttinger-Tisza

\[ H_{LT} = -\sum_{q, \alpha \beta} \vec{s}_\alpha(\vec{q}) \cdot J_{\alpha \beta}(\vec{q}) \cdot \vec{s}_\beta(\vec{q}) \]  

(2.1.7)

where we now have a quadratic form, with \( \alpha, \beta \) indexing the sublattices and \( J_{\alpha \beta}(\vec{q}) \) being a Hermitian matrix. It then follows that we can diagonalize \( J_{\alpha \beta}(\vec{q}) \) and minimize the eigenvalues to determine \( \vec{q}_m \). However, it does not follow from this that all of the spins can be represented in the simple helimagnetic form presented above, because the solution is now an eigenvector, and therefore may possess different amplitudes on different sublattices. In particular, it may be impossible to satisfy normalization of all the spins while simultaneously limiting the modes to those degenerate with \( \vec{q}_m \).
So in the generic case for a non-Bravais lattice, the Luttinger-Tisza method will not allow us to explicitly construct the solution. But does it still have some use for non-Bravais lattices? Surprisingly, yes. It gives a lower bound upon the energy, which allows for a check against the solutions determined by other methods. Moreover, we expect that, in some sense, the solution will be “near” to the optimal wave-vector. Although it might include sub-optimal modes, it will not completely neglect the optimal mode. This implies that the phase diagram of the optimal Luttinger-Tisza mode will be similar (but, in the interesting cases, only approximately) to the ground state phase diagram. While such intuitive application of Luttinger-Tisza sounds pleasant, it should only be used with caution in analyzing the generic solutions to the Heisenberg Hamiltonian. Too often, important aspects of ground state are missed with an overly-facile Luttinger-Tisza analysis.

B. Iterative Minimization

The iterative minimization algorithm is a brute force approach to finding the ground state of the Heisenberg Hamiltonian. The theory behind it is simple

\[ H_H = \sum_i H_i \]  \hspace{1cm} (2.2.1)

\[ H_i = \vec{s}_i \cdot \left( -\sum_j J_{ij} \vec{s}_j \right). \]  \hspace{1cm} (2.2.2)

The term \(-\sum_j J_{ij} \vec{s}_j\) is the effective field at \(\vec{s}_i\), the magnetic field produced by the interaction of one site with all other sites. Clearly, to minimize a partial sum, we need to set

\[ \vec{s}_i \rightarrow \frac{\sum_j J_{ij} \vec{s}_j}{\| \sum_j J_{ij} \vec{s}_j \|^2}, \]  \hspace{1cm} (2.2.3)

i.e. anti-parallel to its effective field. Each time we do this we necessarily lower the energy (unless, \(\| \sum_j J_{ij} \vec{s}_j \|^2 = 0\), in which case \(\vec{s}_i \rightarrow \vec{s}_i\) and the energy is unchanged). Therefore, we can minimize the Hamiltonian by starting with the spins in any random configuration, selecting particular spins at random, and setting them anti-parallel to their effective field. Repeatedly doing this should produce successively more accurate approximations of the ground state. Furthermore, because we are dealing with semi-classical spins, there are enough degrees of freedom to the problem that this method will not get stuck at a local minimum, but will approach the true global minimum.
While this sounds good, there are several important caveats. First of all, the randomization algorithm used in this method must be robust. For example, relaxing the spins in a set pattern will produce spurious ground states. What’s more, for real implementations of a lattice, there will be some finite size to the system being modeled. To account for this discrepancy, it is customary to use periodic boundary conditions. In our implementation, we apply both periodic and anti-periodic boundary conditions, as periodic boundary conditions are not always sufficient.

While these boundary conditions will help, they will not completely eliminate the effects of a finite size. In particular, not every possible ground state will fit within an arbitrary sample. For example, a quasi-periodic helimagnet (a spiral where \(|q_m|\) is irrational) will not fit in any choice of sample. In this situation, two solutions are possible. Either (1) the iterative minimization algorithm will reduce the problem to a proximate state (e.g. a periodic helimagnet with \(|\tilde{q}| \approx |q_m|\)) or (2) the ground state will be achieved locally, but there will be a finite number of domain walls within the system. These domain walls will appear as a “buckling” within the spin order, which is easily detectable using the techniques described in section 4. In part because of the relative ease of detecting domain walls versus proximate states, we found that the domain walls were more common in incommensurate systems. Lastly, the convergence of the iterative minimization algorithm is very slow. For long \(q\), the minimization goes like \(|q|^2\), meaning that any real implementation of this algorithm will invite defects. Although these are fairly strong scruples, the iterative minimization algorithm is robust enough that, in practice, it will normally be sufficient to determine the ground state.

C. Variational Minimization

While the iterative minimization algorithm is effective at producing approximations, they remain only approximations. In most cases, we can make the results of an iterative minimization approach more rigorous by turning to better approximation methods. Since we already have some idea of the ground state, the method of choice is a variational optimization. In many cases, the ground state of the iterative minimization will be characterized by a free parameter \(\alpha\) (for example, the polar angle \(q\) in the case of a helimagnet). If we assume that the overall form of the ground state was correct, but the values of the free parameters
were approximations of their true values, then we can attempt to find the precise values. That is, we can say

\[ \hat{s}(\vec{r}) \approx f(\vec{r}; \{\alpha\}) \]  
\[ H = H(f; \{J\}) \]  
\[ \frac{\partial}{\partial \alpha_i} H = \frac{\partial H}{\partial f} \frac{\partial f}{\partial \alpha_i} = 0. \]

Following this procedure for all free parameters \( \alpha_i \) will produce a system of equations with solution(s) \( \{\alpha_m\} \). Calculating \( H(f(\vec{r}, \{\alpha_m\}); \{J\}) \) for all sets \( \{\alpha_m\} \) and comparing the energies will give a more rigorous approximation of the ground state.

There are two principle problems with this approach. The first is that, for systems of sufficient complexity, the determination of \( \{\alpha_m\} \) is highly non-trivial. The second is the possibility that the ground state is actually of the form \( g(\vec{r}, \{\beta_m\}) \), where no \( f(\vec{r}, \{\alpha\}) = g(\vec{r}, \{\beta\}) \), i.e. that there is some other, true ground state that was not considered during the selection of \( f \). For sufficiently clever selections of \( f \), this risk is minimized, but not eliminated. In practice, we take the approach of using the functional forms produced by iterative minimization and assume that, with a sufficiently large sample of parameter space and generality of \( f \), we possess a good approximation of the ground state. For the cases considered here, variational optimization is the most rigorous method of determining the ground state that we could find. Using its results to extrapolate the phase diagram yielded counter-intuitive predictions (in particular, the existence of splayed states in the chain lattice, see section V A 3) which were born out by further iterative minimization.

There are a variety of simple techniques for extending the power and applicability of the variational method to other problems. This is especially true for phase diagrams, as the presence of an analytic expression for the spins will help boot-strap our way to the phase boundaries, even when \( \{\alpha_m\} \) is unknown. There are three ways of approaching this problem of extending the results of variational optimization. The first is to limit our inquiry to functional forms

\[ H(\{\alpha\}, \{J\}) = \sum_i F_i(\{\alpha'_j\}, \{J\})g_i(\alpha_j). \]

In practice, this approach will often allow us to determine \( (\alpha_j)_m = h(\{\alpha'_j\}, \{J\}) \) where \( \{\alpha'_j\} \) is the set of \( \{\alpha\} \)'s excluding \( \alpha_j \). We can then selectively substitute this solution back into
the system of equations and find an expression for another $\alpha$. Repeating this method will often be more effective than blindly optimizing all the $\alpha$’s separately from the beginning.

The second method is closely related to this functional approach. But whereas the functional method treated the couplings $\{J\}$ as unimportant, it is worth recalling that the final solution to a variational optimization will have $(\alpha_i)_m = f_i(\{J\})$. If we can find a particular combination of $J$’s (or $J$’s and $\alpha$’s, if we have not eliminated them) such that varying both will leave the resultant $\alpha_m$ unchanged, then we have found effective couplings, $J_{\text{eff}}$. A clever choice of $J_{\text{eff}}$ will often simplify the algebra of the problem, much as the functional approach outlined above. While this approach is sometimes more effective at simplifying the expressions for $\alpha$, it has the disadvantage of making it harder to compare energies of two different solutions. As a result, this approach was not used extensively.

The final technique is to focus upon local properties. Phase transitions will either be first order or continuous. In the case of first order transitions, the stability of two distant states will be exchanged, resulting in a different global order. These transitions are hard to detect without calculating $H(\{\alpha_m\})$, since there is no local indicator of this change of stability. Continuous transitions, on the other hand, are purely local phenomena. There are two ways that we can exploit this locality. The first is to realize that certain simple states are always going to satisfy $\partial \{\alpha\} H = 0$. For example, a ferromagnetic state will necessarily be stationary point. Now we can Taylor expand about this point to second order in $\alpha_i$, giving

$$H \approx H(\{\alpha_0\}) + \frac{1}{2} \frac{\partial^2}{\partial \alpha_i^2} H(\{\alpha_0\}) \alpha_i^2 \tag{2.3.5}$$

When $\frac{\partial^2}{\partial \alpha_i^2} H(\{\alpha_0\}) < 0$, then the state $H(\{\alpha_0\})$ is unstable, and no longer the ground state. Instead, a nearby state will be the ground state (this is because higher order terms in the Taylor expansion will produce a minima near the old one). So we can find the phase boundary by considering the curves $\frac{\partial^2}{\partial \alpha_i^2} H(\{\alpha_0\}) = 0$.

The second approach to continuous phase transitions is to consider circumstances where $f(\vec{r}; \{\alpha\})$ simplifies to a more convenient expression, for example a helimagnet. We can then determine $\{\alpha_m\}$ for this simpler case, giving a set of possible solutions. For a continuous transition, this set of solutions must become degenerate with another at some particular value of $\{J\}$. If we know the value of one set of solutions (for example, the angles of a ferromagnet are all 0) and an expression for the other set of solutions (for a helimagnet, this would be $F(q_m; \{J\}) = 0$) but not its solution, then we can solve for the $\{J\}$ such that these
solutions sets are equal (i.e. $F(q_m \equiv 0; \{J\}) = 0$) this will give a function in parameter space that is the boundary for the continuous phase transition. While this second approach sounds complicated, it is often surprisingly successful.

D. Diagnostic Tools

Plainly, to properly set up the variational solution and compare it with the approximations of the other methods, we need to have a good understanding of the approximate ground state. But for large, complicated crystal systems with elaborate spin patterns, it is not always easy to just look at the spins on the lattice and find the functional form. We therefore use a variety of specialized techniques to help characterize a state.

1. Fourier Analysis

The first technique used is to take the Fourier Transform of the ground state produced by the iterative minimization algorithm. This method helps analyze the symmetry between different sublattices and has the advantage of connecting the result of the Luttinger-Tisza and iterative minimization algorithms. For more complicated states (in particular, the double twist discussed later), Fourier analysis is the method of choice for analyzing the functional form of the ground state. Moreover, the Fourier analysis that we can carry out numerically is more powerful than the experimental diffraction techniques, which use the same methods of analysis. We can find both the phase and amplitude of the Fourier Transform, whereas diffraction experiments only recover the intensity, so our inverse problem is simpler.

2. Qualitative Techniques: Plotting and Grouping

However, the construction of the functional from the Fourier spectrum is sufficiently complicated that, in most cases it is ancillary. Instead, we use a “common-origin plot.” Rather than drawing each spin as a vector with tail at its location in the lattice, we translate all of the spins so that their tails lie at the origin. This washes out the spatial information of the spin order, but highlights symmetries in the spins. For example, a helimagnetic
state becomes a great circle (see Fig. 3). Common-origin plots are a very powerful tool for the analysis of spin order, often times highlighting information that would be difficult to observe otherwise. For example, the cuboctahedral states are most quickly identified through a common-origin plot.

In the event that a common-origin plot is insufficient, an alternative plotting algorithm exists. As in a random walk, we lay each spin head-to-tail and observe the pattern of the entire set of spins. For the chain lattice, we alternate spins from different sublattices and proceed along the lattice. For the octahedral lattice, we only use a single sublattice and raster-scan through the three-dimensional array of lattice sites. This plotting technique focuses upon the symmetry in the spins, but at the same time allows a partial reconstruction of the real-space order. It is therefore a compromise between the real-space and common origin plotting techniques.

To complement these plotting tools, we also group spins by their orientation. When the dot product of two spins indicates that they are nearly parallel \( \hat{s}_i \cdot \hat{s}_j \geq .99 \), then they are considered to be of the same grouping. The grouped spins are averaged, washing out any noise in the iterative minimization relaxation process. For sufficiently separated spins, this serves as a more analytic approach to the analysis carried out by eye in the plotting techniques.

3. The Spin Inertia Tensor

But not every diagnostic is as qualitative as these. One of the most useful tools in our hunt for non-coplanar spin states is the “spin-inertia” tensor:

\[
M_{ij} = \frac{1}{N} \sum_k s_i(r_k)s_j(r_k)
\]  

(2.4.1)

where \( N \) is the total number of spins, \( i, j \) run over the basis of spin-space, and \( k \) is the site index. That is, we sum over all spins (we do not distinguish between sublattice), and the matrix elements are the elements of self-tensor product of a spin \( \hat{s} \otimes \hat{s} \). Note that \( \text{Trace}(M) = 1 \) and that \( M \) is symmetric. Significantly, the number of non-zero eigenvalues of \( M \) is equal to the dimensionality of the spins, so a set of non-zero eigenvalues implies a non-coplanar state. In addition, the eigenvectors of \( M \) define an orthogonal matrix, which transforms the spins into a canonical set of coordinates. Such a transformation is useful in
representing the ground state from the iterative minimization algorithm in a canonical form. For example, in the conic spirals, the canonical transformation sets the conic axis along the $s_z$-axis.

4. Energy Classification

In addition to the spin-inertia tensor, we also calculate

$$E(J_a) = -\frac{1}{N} \sum_{\{ij\}_a} \vec{s}_i \cdot \vec{s}_j$$

where $\{ij\}_a$ runs over all pairs of spins having interaction $J_a$. Note that this quantity is independent of $J_a$'s magnitude, depending only upon the geometry of the spins and the direction of $\vec{r}(J_a)$. This is useful, for example, in the cuboctahedral states, where the $E(J_a)$ values are independent of $J$, allowing for quicker characterization of the state and revealing that the spins are not a function of $\{J\}$. Complementary to this technique is the use of a normalized energy $H_H/N$, which is useful in giving a measure of the energy per site, thus facilitating comparisons of ground states with the same couplings but different numbers of sites.

5. Local Geometries

The final diagnostic technique is used primarily in the chain lattice, where states with variational parameters are more common. We use local measures to calculate these variational parameters, focusing upon the conic states as they are the most general. For example,

$$\cos 2\psi_i = \vec{s}_\alpha(\vec{r}_i) \cdot \vec{s}_\alpha(\vec{r}_{i+1})$$

where $\alpha$ denotes sublattice and $\psi_i$ the polar angle at site $i$. Similar geometric techniques allow us to find the conic angles and conic axis (for example, summing spins averages out $\psi$ and gives the components along the polar axis). Even more important than the use of these techniques in determining the variational parameters is their use in finding local defects. In the case of domain walls, these measures will deviate from a stable value, allowing us to quickly isolate these defects and focus on the spins outside of them (another quick way to finding domain walls is to calculate $E(\vec{s}(\vec{r})) = H_i$, since that will also show local defects).
III. CONCEPTUAL TOOLS

The methods described in the previous section are principally useful in calculating the ground state for some combination of couplings. But this is not all that we want to know, it is also important to understand how the ground states fit together. In particular, we want to be able to determine when a phase boundary is first order or second order. This is another highly non-trivial problem, so we introduce several conceptual tools to aid the analysis. These tools are fairly specialized, each is to be used to understand the properties of specific types of ground states.

A. One-Star States

The one-star states are conceptually the simplest. When a lattice has more than one spatial dimension, there will be more than one dimension to the Brillouin zone. This will imply that the wave-vectors in the Brillouin zone are truly vectors, rather than scalars. As such, there are more possible symmetries to the wave-vectors, and therefore the Hamiltonian as a function of the wave-vectors. In one dimension, the only symmetries are translational (which allows us to restrict attention to the Brillouin zone) and inversion (which allows us to construct real Luttinger-Tisza eigenstates). But in higher dimensions there are also rotational symmetries.

For an arbitrary, real-valued ground state, we expect the Fourier transform (Luttinger-Tisza analysis) to possess inversion symmetry, but not necessarily rotational symmetry (even when the lattice does possess such symmetry). When the ground state is composed of wave-vectors that are related by rotational symmetry (and inversion symmetry), we call such a configuration a one-star state, as the wave-vectors all lie along the same “star” of the Brillouin zone. One-star states are fairly ubiquitous when there are multiple sublattices related by rotational symmetry (as we will see in the octahedral lattice, presented in section IV). The most interesting scenario for one-star states, though, is when each sublattice contains a one-star state, rather than just the lattice as a whole. States of this form can often be incredibly complicated (see section IV C 2 for an example). Furthermore, they are often related to families of highly degenerate states (see the following section), in which case they may well be intrinsically non-coplanar (this point is developed in section VI C).
B. Families of Highly Degenerate States

As previously mentioned, one-star states are often special cases of a more degenerate family of states (see section VI C for an example). In families of degenerate states, the ground state can be characterized by some free parameters, exactly as in variational optimization of section II C. What distinguishes these families of state, though, is that the free parameter is not fixed by variational concerns (of course, such families of states only exist under specific combinations of couplings, and perturbations typically break the degeneracy). For example, a 1D Bravais lattice (i.e. an infinite line of lattice sites) with only $J_2 = 1$ is minimized by any configuration satisfying $s(z + 2) = s(z)$. In particular, $s(z + 1)$ can be at any arbitrary angle to $s(z)$ without materially effecting the energy, as the lattice has decoupled into two separate ferromagnets. Families of highly degenerate states are likely when the lattice has decoupled or when spin configurations imply perfect cancellations among spins.

C. Encompassing Parametrizations

The final conceptual tool for the analysis of phase boundaries is encompassing parametrization. The full utility of this concept can only be appreciated in light of the ground states of the chain lattice (section V). As such, much of this analysis will refer to ground states presented in that section. In addition, this presentation will be reiterated and expanded there.

We call a parametrization encompassed if it can be expressed by another parametrization with particular values of free parameters. For example, ferromagnetism and anti-ferromagnetism are both encompassed parametrizations of a helimagnetic parametrization ($\psi = 0$ or $\pi$, respectively), while helimagnetism is itself a encompassed parametrization of a conic state ($\alpha, \beta \to 0$). Conversely, helimagnetism is the encompassing parametrization of (anti-)ferromagnetism.

The generality of a state (which is related to the number of free parameters, and therefore the number of distinct states that can be parametrized) is distinct from the concept of encompassing. An encompassing state is necessarily more general than the encompassed state, but a more general state is not necessarily an encompassing one. For example, the asymmetric conic is more general than the alternating conic (in the sense that there are more
distinct asymmetric conic states than there are alternating conic states), but neither conic encompasses the other. This absence of an encompassing parametrization is important. In particular, there is no splayed state (ferromagnetic or ferrimagnetic) that encompasses the generic helimagnet ($\psi \neq 0, \pi$), nor does the generic helimagnet encompass the splayed states. Similarly, the asymmetric conic does not encompass the ferromagnetic splayed state (and vice versa).

There are certain similarities between the concepts of encompassing parametrization and degenerate family of states. Both rely upon the idea that there is a state with some free parameters, and that specific values of these parameters can be used to reproduce a more specialized state. The principle difference between the two concepts is one of stability. Because highly degenerate families are highly degenerate, they are not likely to exist within any finite region of a phase diagram. Instead, they are almost exclusively found on phase boundaries between two non-degenerate states. These non-degenerate states are members of the degenerate family along the phase boundary. Conversely, encompassing parametrizations have their free parameters uniquely determined by the couplings, so degeneracy is not an essential aspect. This means that perturbative couplings do not necessarily destroy the encompassing state. As a result, it is possible to find encompassing parametrizations that are the non-degenerate ground state for a finite regions of parameter space. So perturbations will change the values of the free parameters, but will not necessarily destroy the state.

D. Applications to Phase Boundaries

To categorize phase transitions as first order or second order, we adopt the following criteria. If, on the phase boundary, we can continuously evolve a ground state from one ground state to another via some arbitrary intermediate state, then it is second order. Conversely, when no such transformation is possible, then the transition is first order. This emphasis upon continuous evolution through intermediate states explicitly excludes cases like nucleation and growth or spinodal decomposition, which relate to how the two states coexist in separate domains at the phase boundary. The intermediate state must be truly different than some arbitrary combination of the two ground states.

If there is a family of degenerate states between two ground states, we can continuously evolve through any combination of degenerate states. Therefore, the phase transition is
second order. In the octahedral lattice this scenario is rather common (section IV B), but
the logic for constructing the degenerate families is slightly different in each case.

When a phase transition is between a state and its encompassed parametrization, then
it is (unless otherwise noted) second order. This is because, as the couplings approach the
phase boundary and an encompassed parametrization exists, the free parameters will tend
towards the values of the encompassed parametrization. This situation is most common in
the chain lattice (section V B), but it is also present in the octahedral lattice (section IV B).

IV. THE OCTAHEDRAL LATTICE

The principal system studied in this paper is the octahedral lattice (Figure 1). This lattice
consists of sites lying upon the bond midpoints of a simple cubic lattice (dashed cubes in
Figure 1). It can therefore be seen as three interpenetrating simple cubic lattices, each offset
by half a bond length with respect to the original simple cubic lattice. Alternatively, using
the lattice with a basis perspective, it is a simple cubic lattice composed of corner-sharing
octahedra (and lattice sites at the corners of octahedra) (solid lines in Figure 1). We will use
the first perspective in parametrize states, typically turning to the second to aid in analysis.
An important point about the first perspective is that each sublattice is associated with a
basis vector (the local symmetry axis), which will impact the way we define couplings. These
symmetry axes will be important in the understanding of different ground states, especially
in the anti-ferromagnets.

While this lattice is relatively unexplored, it has been introduced in other contexts. It is,
properly speaking, the three-dimensional analog of the checkerboard lattice, which was itself
introduced as a two-dimensional analog of the pyrochlore lattice [10]. It has also been used
to as a toy model for analysis of the pyrochlore lattice in its own right [11-15]. Moreover,
the octahedral sites are Wyckoff positions (sites of higher symmetry), and therefore likely
candidates for a lattice sites. That is, within a more complicated structure, the magnetic
sites form an octahedral sublattice. A few examples of real materials are known, although
they do not possess any of the more exotic ground states discussed here. First of all, there is
the Cu$_3$Au superstructure of the fcc lattice [11], but all known realizations of the structure
are non-magnetic, except for Mn$_3$Ge, which is ferromagnetic [16]. Second, it is found in
metallic perovskites, such as Mn$_3$SnN, although these are also ferromagnetic [17]. Finally,
Sublattice  $\mathbf{R}(J_2)$  $\mathbf{R}(J'_2)$  $\mathbf{R}(J_4)$  $\mathbf{R}(J'_4)$

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Table I: Anisotropic Couplings of the Octahedral Lattice

it is related to the magnetic lattice found in Ir$_3$Ge$_7$ structures, including Mo$_3$Sb$_7$ [18, 19]. These structures are composed of a simple-cubic lattice with disjoint octahedra. But in the case of strong ferromagnetic couplings between adjacent corners, the spins would necessarily be parallel and we could analyze everything in terms of the octahedral lattice. However, Mo$_3$Sb$_7$ is anti-ferromagnetic, so to give a semi-classical treatment requires us to invert the spins in every other sublattice.

In examining this lattice, we use couplings through the fourth nearest neighbor, where the displacement vectors ($\mathbf{R}$) associated with different couplings ($J_i$) are

\[
\mathbf{R}(J_1) = \frac{1}{2} \begin{pmatrix} 1 \\ 1 \\ 0 \end{pmatrix} \\
\mathbf{R}(J_2) = 100 \\
\mathbf{R}(J_3) = \frac{1}{2} \begin{pmatrix} 1 \\ 1 \\ 2 \end{pmatrix} \\
\mathbf{R}(J_4) = 110
\]

and all permutations. The odd couplings connect sites of different sublattice, while even couplings connect those of the same sublattice. It is important to realize that the even couplings are not all equivalent. For example, the $J_2$ coupling could either link sites in the same octahedron or sites in different octahedra. To differentiate between these different sorts of couplings, we use table I (and all permutations of sign). That is, if the local symmetry axis’ dot product with a coupling is 0, then it is designated as primed, and unprimed if the product is non-zero. To generalize this notation, we sort couplings by the angle they make with the local symmetry axis (the angle between two vectors $a, b$ is the familiar $\cos \theta = a \cdot b / |a||b|$).

An equivalent method of ordering couplings is to count the minimum number of octahedra that must be traversed to link two sites (i.e. $J_2$ connects sites within the same octahedron, while $J'_2$ requires 2 octahedra).
Figure 1: The Octahedral Lattice. Sublattice 001 denoted by circles, 010 by squares, 001 by x’s. Cube shown in dotted lines, octahedron in solid. Couplings shown in dashed vectors.

For the purposes of understanding states in the octahedral lattice, it is preferable to group couplings as follows. $J_1, J_2,$ and $J'_2$ are all “short-range” couplings, and in some sense stronger than the “long-range” couplings $J_3, J_4$ and $J'_4$. As such, we analyze parameter space for all non-zero short range couplings and a single non-zero long-range coupling, necessarily weaker than the largest short-range coupling. What’s more, iterative minimizations with couplings $J_1$ through $J_4$ and $J'_4$ produced the same states found with $J_1$ through $J_3$ only. Therefore, we focus primarily upon the four couplings $(J_1, J_2, J'_2) + J_3$ in examining parameter space. Adopting this method of attack, the phase diagram is surprisingly simple. A total of eight states fill the $(J_1, J_2, J'_2) + J_3$ phase diagram, almost all of them separated by first order transitions. To group these states, the easiest choice of coordinates is to normalize by $|J_1|$ and use $J'_2/|J_1| \times J_2/|J_1|$ coordinates while taking slices for various $J_3/|J_1|$. In doing so, we see that the phase diagram divides up into quadrants, with additional states sometimes appearing near the $J'_2 \approx 0, J_2 \gtrsim 0$ region (Figures 4 through 12, which are examined in more detail through the following four sections).
A. Basic States in the Octahedral Lattice

1. (0,0,0) Modes

Within the first quadrant, the adopted state depends upon the sign of $J_1 + 2J_3$. For positive $J_1 + 2J_3$, the lattice adopts a ferromagnetic ordering, with all of the spins collinear (Figures 4, 6, 7, 8, and 11). Conversely, for $J_1 + 2J_3 < 0$, spins in different sublattices are no longer parallel and the lattice adopts a 3 sublattice 120° state (Figures 5, 9, 10, and 12). Because the combination of $J_2$ and $J'_2$ is primarily ferromagnetic (i.e. $J_2 + 2J'_2 > 0$), the spins within a sublattice maintain their ferromagnetic order - each sublattice can be characterized by a single spin. These three spins are arranged so that each spin is rotated 120° with respect to the other two. Or, to put it another way, any three spins from different sublattices form an equilateral triangle, giving a vector sum of 0. This implies that there is no net magnetic moment in the 3 sublattice 120° state, which is very different from the ferromagnetic state's large magnetic moment. An interesting property of the 3 sublattice 120° state is that the spins on the triangular face of an octahedron necessarily cancel each. This implies that the 3 sublattice 120° state is another way to break the degeneracy of the 120° states, developed in section VIC.

2. Anti-ferromagnetic (1/2 and 0) Modes

The second, third and fourth quadrants each display different states. These three states do share certain qualities, which inspires our grouping them here. First, these states are always present, unlike the first quadrant - where two different states compete with each other. That is, they are found in all the phase diagrams (Figures 4 through 12). Second, all of these states are some form of anti-ferromagnetism. In all of the anti-ferromagnetic states, each sublattice is again characterized by a single spin direction, but now spins will alternate between parallel or anti-parallel to this vector. Because of this alternation and the symmetry of the interactions, the different sublattices decouple. Therefore, there is no relation between the spin directions adopted by each of the sublattices. Now recall that each sublattice is characterized by a local symmetry axis, since each sublattice is offset along a particular axis. Recall also that these local symmetry axes are used to distinguish $J_2$ vs $J'_2$ and $J_4$ vs $J'_4$. The relation between the local symmetry
axis and the alternation of spins within the sublattice defines the modulation of the anti-ferromagnet in exactly the same manner as one uses polarization vectors and wave-vectors to distinguish between transverse and longitudinal waves.

The second quadrant \((J_2'/|J_1| \lesssim 0, J_2/|J_1| > 0)\) these inequalities are made precise in Table III only displays transversely-modulated anti-ferromagnetism (Figures 4 through 12). For transversely-modulated anti-ferromagnet, we can parametrize the state by

\[
s_x = (-1)^{011 \cdot r} \hat{n}_x \quad (4.1.1)
\]

\[
s_y = (-1)^{101 \cdot r} \hat{n}_y \quad (4.1.2)
\]

\[
s_z = (-1)^{110 \cdot r} \hat{n}_z \quad (4.1.3)
\]

where the subscripts denote (decoupled) sublattices, there is no relation between the different \(\hat{n}\) (in particular, they do not necessarily point along 100 and its permutations), and \(r\) is the set of integers nearest to the spin’s location. That is, spins do not alternate in the direction parallel to the local symmetry axis. Instead, all alternation in the spins is confined to the planes perpendicular to the local symmetry axis.

An analogy may make this clearer. Think of a checkerboard, where the red squares represent spins pointing in one direction and the black squares represent spins pointing along the opposite direction. Each plane transverse to the sublattice’s local symmetry axis contains such a checkerboard. These checkerboards are stacked one atop the other in exactly the same pattern (i.e. red placed over red, black over black). Note how this pattern exactly satisfies the couplings. The \(J_2\) coupling is ferromagnetic and links sites along the local symmetry axis, whereas \(J_2'\) is anti-ferromagnetic and links sites transverse to the local symmetry axis. Finally, analyzing this ground state in the Brillouin Zone shows that it is solely composed of \((1/2, 1/2, 0)\) modes (that is, \(\pi(110)\), when we include the assumed prefactors).

The third quadrant \((J_2'/|J_1| < 0, J_2/|J_1| < 0\) except for near the origin, where the ground state will be some non-anti-ferromagnetic ground state, depending on \(J_1\) and \(J_3\)) is again anti-ferromagnetic. This time, however, it is an isotropically-modulated anti-ferromagnet.
(see Figures 4 through 12). The states are simply parametrized by

\[ s_x = (-1)^{111} r \hat{n}_x \]  
\[ s_y = (-1)^{111} r \hat{n}_y \]  
\[ s_z = (-1)^{111} r \hat{n}_z \]  

where the variable definitions are unchanged from equations 4.1.1 to 4.1.3. This time, the spins flip with a translation in any direction (think a three-dimensional checkerboard), meaning that each sublattice forms the standard definition of anti-ferromagnetic order for a simple-cubic lattice. This convergence is to be expected, since this quadrant contains the line \( J_2 = J'_2 \), which is makes the sublattices of the octahedral lattice truly equivalent to the simple-cubic. Finally, analyzing this ground state in the Brillouin Zone shows that it solely composed of (1/2, 1/2, 1/2) modes.

The fourth quadrant (\( J'_2 / |J_1| > 0, J_2 / |J_1| < 0, |J_3| < |J_1| \)) is the converse of the second. This time the spins adopt a longitudinally-modulated anti-ferromagnetism (see Figures 4 through 12). The parametrization follows the same pattern as the previous cases:

\[ s_x = (-1)^{100} r \hat{n}_x \]  
\[ s_y = (-1)^{010} r \hat{n}_y \]  
\[ s_z = (-1)^{001} r \hat{n}_z . \]  

The spins in planes transverse to the local symmetry axis are all parallel, with the only alternation taking place along this local symmetry axis. This configuration perfectly satisfies all the couplings, since the transverse interaction is due to \( J'_2 \), which is ferromagnetic, and the longitudinal coupling is due to \( J_2 \), which is anti-ferromagnetic. Finally, analyzing this ground state in the Brillouin Zone shows that it solely composed of (1/2, 0, 0) modes.

3. Cuboctahedral (1/2, 0, 0) Modes

The next two states are the most interesting. They are only found as ground states in the \( J'_2 / |J_1| \times J_2 / |J_1| \) phase diagrams for certain values of \( J_1 \) and \( J_3 \), but when they are present they lie primarily between the first and second quadrant (\( J'_2 \approx 0, J_2 \gtrsim 0 \)). We call these states cuboctahedral states. The two cuboctahedral states (\( 2\pi/3 \) and \( \pi/3 \), for reasons which
will soon become clear) are separated by the $J_1 = 2J_3$ boundary, with the $2\pi/3$ state for $J_1 > 2J_3$. The $2\pi/3$ cuboctahedral state exists when

$$J_1 < -J'_2 - 6J_3 \quad (4.1.10)$$
$$J_1 < \frac{1}{2} J'_2 \quad (4.1.11)$$
$$J_1 < -J_2 \pm J'_2 + 2J_3 \quad (4.1.12)$$

are all satisfied (see Figures 7, 8, and 12), whereas the $\pi/3$ cuboctahedral state exists when

$$J_1 < -\frac{1}{3}(J'_2 + 2J_3) \quad (4.1.13)$$
$$J'_2 < 4J_3 \quad (4.1.14)$$
$$J_1 < J_2 \pm J'_2 + 2J_3 \quad (4.1.15)$$

are all satisfied (see Figure 5 and 9).

The cuboctahedral states are parametrized by

$$s_x = + \frac{1}{\sqrt{2}}(-1)^y \hat{y} + \frac{1}{\sqrt{2}}(-1)^z \hat{z} \quad (4.1.16)$$
$$s_y = + \frac{1}{\sqrt{2}}(-1)^z \hat{z} \pm \frac{1}{\sqrt{2}}(-1)^y \hat{y} \quad (4.1.17)$$
$$s_z = \pm \frac{1}{\sqrt{2}}(-1)^x \hat{x} \pm \frac{1}{\sqrt{2}}(-1)^y \hat{y} \quad (4.1.18)$$

where we have specialized to canonical coordinates and the $\pm$ differentiates between the $2\pi/3$ and $\pi/3$ cuboctahedral states (recall that canonical coordinates have the basis of the spin-inertia tensor’s eigenvectors, see section II D 3). In each sublattice of the cuboctahedral states, the spins are invariant along their local symmetry axis (as expected for $J_2 > 0$). Planes orthogonal to the local symmetry axis contain 4 spin directions. These spins make two anti-ferromagnetic pairs, which are orthogonal to each other (think a cross in the common-origin plot, or a square when laid end to end). This is not simply helimagnetism, though, because is no single wave-vector that characterizes this variation in the spins. Instead, it is a superposition of two different permutations of the $(1/2, 0, 0)$ wave vector (the permutations being orthogonal to the local symmetry axis). This superposition of wave-vectors is to be expected when the modes are degenerate, as they clearly are here.

The two cuboctahedral states are differentiated by their correlations between sublattices. In both cases, spins of differing sublattices define an angle of $60^\circ$ or $120^\circ$. The spins within a
Rearranging spins to show symmetry:

Figure 2: The $\pi/3$ cuboctahedral state in real space (left), spin space (tip-to-tail) (center), and spin space (common-origin) (right). Sublattice 100 in red, 010 in black, 001 in blue. The $2\pi/3$ cuboctahedral plots are identical, except for the real space plot, where all spins point inwards or outwards from the cube.

The sublattice are coplanar, but the three planes are orthogonal (that is, their normal vectors are orthogonal). Thinking about the spins in terms of the common origin plot gives a star, spins pointing along all 12 permutations/sign changes of 110. If we use the tip-to-tail plotting, where each sublattice makes a square, and translate spins appropriately, then we find an octahedron. Alternatively, in the Luttinger-Tisza approach, the two cuboctahedral states use different eigenvectors of the $1/2, 0, 0$ eigenmode ($011$ for $2\pi/3$ and $0,1,-1$ for $\pi/3$).

In the $2\pi/3$ cuboctahedral state, this octahedron lies upon the real space cube (recall that the octahedral lattice has sites on the bond mid-points of a simple-cubic lattice), with the spins pointing outward from the cube (very similar to Figure 2, which is for the $\pi/3$ cuboctahedral state). In the $\pi/3$ cuboctahedral state, the spins no longer point outward from the cube (see Figure 2). Half of the spins point outwards and half inwards, but the spins still define an octahedron in the same sense as in the $2\pi/3$ cuboctahedral state. Finally, for the $2\pi/3$ cuboctahedral state, existence requires $J_3 < 0 \leq J_1$, meaning that it requires more long-range couplings than the $\pi/3$ cuboctahedral state, which requires $J_3 > J_1/6$ and $J_1 < 0$ (in particular, there exist $J_1, J_2$, and $J'_2$ such that it is the ground state even when $J_3 = 0$).

It should be noted that, when $J_4, J'_4 \neq 0$, what appears to be a third type of cubocta-
hedral state is produced [23]. Because of the couplings used in creating this state, iterative minimization is less effective at removing numerical artifacts from this state. It is encountered when $J_1 = \pm 1, J_4 = J'_4 = -1.1$ and when $J_3 = -1, J_4 = J'_4 = -1.1$, so it also goes beyond our $J_3, J_4, J'_4$ small approximation.

In this “third type” of cuboctahedral state, there are again two different spin directions per sublattice and each spin direction will have an anti-parallel counterpart (giving a total of four spin directions per sublattice). The spatial pattern of the spins is also identical to the previous cuboctahedral states’, including the alternation between parallel and anti-parallel spins along the real-space diagonals. However, numerical simulations lack the other symmetries of the standard cuboctahedral states. The pairs of spins in the same sublattice are no longer orthogonal, nor is the $60^\circ - 120^\circ$ correlations between sublattices preserved.

The optimal Luttinger-Tisza eigenmode and eigenvector in this state are $(1/2, 0, 0)$ and $(011)$, exactly as they were in the standard $2\pi/3$ cuboctahedral states. The degeneracy of Luttinger-Tisza modes, combined with spatial order observed by iterative minimization, suggests that this “third type” of cuboctahedral state is actually spurious - and that the ground state is actually a $2\pi/3$ cuboctahedral. The observed deviations from the standard cuboctahedral angles is because the $J_4$’s couple sites in a sublattice less strongly than the $J_2$’s. Specifically, there is no combination of $R(J_4)$ or $R(J'_4)$ that can produce $R(J_2)$ or $R(J'_2)$ (the converse, on the other hand, is possible). Therefore, sites related by $R(J_2)$ or $R(J'_2)$ are almost decoupled. Their connection is mediated by the other sublattices (as combinations of $R(J_1)$ or $R(J_3)$ can produce $R(J_2)$ or $R(J'_2)$). This helps to explain why, for $J_3 = -1, J_4 = J'_4 = -1.1$, we observed both this form of cuboctahedral state and the $2\pi/3$ cuboctahedral state; this is merely a region of parameter space where numerical artifacts are slow to die out as we approach the cuboctahedral state.
Finally, for a small range within

\[ 0 < J_1 < \frac{-1}{2} J_1 < J_3 < 0 \]

\[ -2[J_1 + J_2' + 2(3 - 2\sqrt{2})J_3] < J_2' \lesssim 0 \]

\[ -J_1 - 2J_3(7 - 4\sqrt{2}) < J_2' < 8J_3(1 - \sqrt{2}) \]

a helimagnetic state (or incommensurate spiral) is possible (see Figures 7 and 8). In this state, all of the sublattices are correlated and spins rotate about a common axis - 111. To fully analyze this state we will require the stacking vector formalism introduced below (section IV C 1). For now, we focus on the phase boundaries. The helimagnetic state will transition to one of three states - 2\pi/3 cuboctahedral, isotropic anti-ferromagnet, longitudinal anti-ferromagnet, or (for sufficiently small magnitudes of \( J_3 \)) ferromagnetism. What’s more, helimagnetism is the only configuration (at least without \( J_4 \)) that fills a finite area of parameter space. The fact that helimagnetism exists in such a small range of parameters explains why we fail to observe it in iterative minimization, the chance of hitting upon the right combination of couplings is minute. However, since we can justify its existence analytically, the absence of any simulations should not detract from the validity of the results.
### Table II: Ground States of the Octahedral Lattice

<table>
<thead>
<tr>
<th>State (wave-vector)</th>
<th>Energy</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ferromagnetic (000)</td>
<td>$-4J_1 - J_2 - 2J'_2 - 8J_3 - 4J_4 - 2J'_4$</td>
</tr>
<tr>
<td>3 sublattice - 120° (000)</td>
<td>$2J_1 - J_2 - 2J'_2 + 4J_3 - 4J_4 - 2J'_4$</td>
</tr>
<tr>
<td>Transverse AFM ($\frac{1}{2} \frac{1}{2} 0$)</td>
<td>$-J_2 + 2J'_2 - 4J_4 + 2J'_4$</td>
</tr>
<tr>
<td>Isotropic AFM ($\frac{1}{2} \frac{1}{2} 2$)</td>
<td>$J_2 + 2J'_2 - 4J_4 - 2J'_4$</td>
</tr>
<tr>
<td>Longitudinal AFM ($\frac{1}{2} 0 0$)</td>
<td>$J_2 - 2J'_2 - 4J_4 + 2J'_4$</td>
</tr>
<tr>
<td>Cuboctahedral -2π/3 ($\frac{1}{4} 00$)</td>
<td>$-2J_1 - J_2 + 4J_3 + 4J_4$</td>
</tr>
<tr>
<td>Cuboctahedral -π/3 ($\frac{1}{2} 00$)</td>
<td>$2J_1 - J_2 - 4J_3 + 4J_4$</td>
</tr>
<tr>
<td>Helimagnetism (qqq)</td>
<td>$-2J_1 - \frac{(2J_1 + J_2 + 2J'_2 + 4J_3)^2}{8(2J_3 + 2J'_2 + J'_4)}$</td>
</tr>
</tbody>
</table>

#### B. Summary and Phase diagrams of the Octahedral Lattice

There are still a few more states to be discussed, but these were only observed upon phase boundaries in $J_1, J_2, J'_2, J_3$-space. So it is meet that, having understood the different states within the phase diagram, we finally delineate the phase diagram precisely. The energies of the states in the octahedral lattice are given by Table II.

Specializing to $J_4, J'_4 = 0$, we find the phase boundaries by setting the difference in energies to 0. This gives Table III.

Except for helimagnetism, none of these configurations has a variational form that is functionally dependent upon the couplings. This distinction aids us in determining the order of the phase transitions. For transitions with variational parameters, the transition is second order if there exists a encompassing parametrization (section III C) i.e. helimagnetism for isotropically modulated anti-ferromagnetism and ferromagnetism.

For states without encompassing parametrizations, the situation is more complicated. In general, the transition will be first order, unless there is some form of decoupling or special degeneracy. That is, if there is a family of degenerate states (section III B) between two
<table>
<thead>
<tr>
<th>Basic States</th>
<th>FM</th>
<th>3S-120°</th>
<th>AFM-TM</th>
<th>AFM-IM</th>
<th>AFM-LM</th>
</tr>
</thead>
<tbody>
<tr>
<td>FM</td>
<td>-</td>
<td>“</td>
<td>“</td>
<td>“</td>
<td>“</td>
</tr>
<tr>
<td>3S-120°</td>
<td>$J_1 + 2J_3$</td>
<td>-</td>
<td>“</td>
<td>“</td>
<td>“</td>
</tr>
<tr>
<td>AFM-TM</td>
<td>$J_1 + J'_2 + 2J_3$</td>
<td>$J_1 - 2J'_2 + 2J_3$</td>
<td>-</td>
<td>“</td>
<td>“</td>
</tr>
<tr>
<td>AFM-IM</td>
<td>$2J_1 + J_2 + 2J'_2 + 4J_3$</td>
<td>$J_1 - J_2 - 2J'_2 + 2J_3$</td>
<td>$J_2$</td>
<td>-</td>
<td>“</td>
</tr>
<tr>
<td>AFM-LM</td>
<td>$2J_1 + J_2 + 4J_3$</td>
<td>$J_1 - J_2 + 2J_3$</td>
<td>impossible</td>
<td>“</td>
<td>-</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Exotic States</th>
<th>Cuboc-2π/3</th>
<th>Cuboc-π/3</th>
<th>Helimagnetic</th>
</tr>
</thead>
<tbody>
<tr>
<td>FM</td>
<td>$J_1 + J'_2 + 6J_3$</td>
<td>$3J_1 + J'_2 + 2J_3$</td>
<td>$\frac{-2J_1 + J_2 + 2J'_2 + 4J_3}{8J_3} = \sqrt{2} - 1$</td>
</tr>
<tr>
<td>3S-120°</td>
<td>$2J_1 - J'_2$</td>
<td>$J'_2 - 4J_3$</td>
<td>impossible</td>
</tr>
<tr>
<td>AFM-TM</td>
<td>$J_1 + J'_2 - 2J_3$</td>
<td>$J_1 - J'_2 - 2J_3$</td>
<td>$\frac{-2J_1 + J_2 + 2J'_2 + 4J_3}{8J_3} = -1 + \frac{-J_1}{J_3}$</td>
</tr>
<tr>
<td>AFM-IM</td>
<td>$J_1 + J_2 + J'_2 - 2J_3$</td>
<td>$J_1 - J_2 - J'_2 - 2J_3$</td>
<td>$\frac{-2J_1 + J_2 + 2J'_2 + 4J_3}{8J_3} = 1 - \sqrt{2}$</td>
</tr>
<tr>
<td>AFM-LM</td>
<td>$J_1 + J_2 - J'_2 - 2J_3$</td>
<td>$J_1 - J_2 + J'_2 - 2J_3$</td>
<td>$\frac{-2J_1 + J_2 + 2J'_2 + 4J_3}{8J_3} = 1 \pm \frac{(2J_1 + J'_2)^{1/2}}{J_3}$</td>
</tr>
<tr>
<td>Cuboc-2π/3</td>
<td>$J_1 - 2J_3$</td>
<td>$\frac{-2J_1 + J_2 + 2J'_2 + 4J_3}{8J_3} = -1 \pm \frac{-J_1 + J_2 + 2J'_2 + 4J_3}{8J_3}^{1/2}$</td>
<td></td>
</tr>
<tr>
<td>Cuboc-π/3</td>
<td>impossible</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table III: Phase Boundaries of the Octahedral Lattice. Equations = 0 unless otherwise noted

ground states, then the phase transition is second order. Several transitions possess these families of degenerate states.

Transitions between anti-ferromagnetic states occur when $J'_2 = 0$, a implying that planes/rows within a sublattice are decoupled. In these cases, the ground state along the boundary could be any member of a whole family of states. These degenerate states create a second order transition. A second example of this phenomena is the 120° state (found when $J_i = -\delta_{1i}$, we analyze this state in section VI C), which governs the transition between the cuboctahedral states and the (000) modes.

A third example is the boundary of the cuboctahedral states and the longitudinally-modulated anti-ferromagnet. All of these states are 1/2 0 0 modes. At the phase boundaries, two of the modes become degenerate. This means that the eigenvectors we previously used are no longer unique. Within the degenerate sub-space, we can use any two non-trivial modes as eigenmodes. As such, any linear combination of the degenerate modes is a ground state and we again have a degenerate family. This is distinct from the formation of magnetic domains, as the combination of the two modes can exist throughout the lattice.

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In this first phase diagram (Figure 4) we see the quadrant structure outlined in section IV A. It is clear, however, that the division is not perfect, as not all the phase boundaries lie upon the axes. Notice that only the transition to ferromagnetism is first order.

When we change the sign of $J_1$ from Figure 4 to produce Figure 5, we find several important changes. First, the phase boundaries shift and the 3-sublattice 120° state becomes preferable to the ferromagnetic state. What’s more interesting, though, is the presence of the $\pi/3$ cuboctahedral state, the first instance of a non-coplanar state (also, one which only requires 2 non-zero couplings). One similarity with Figure 4 that is rather unexpected is the
Figure 5: Octahedral Phase Diagram for $J_1 = -1, J_3 = 0$. First instance of a non-coplanar state (the $\pi/3$ cuboctahedral state). Circled point is the $120^\circ$ degree state (introduced in section VI C)

location of the first order phase transitions, which have retained their topology.

As we turn on a ferromagnetic $J_3$ from Figure 4 (giving Figure 6), we strengthen the $J_{\text{eff}}$ effective coupling between sublattices (the $J_{\text{eff}}$ concept is introduced in section II C). This has the effect of increasing the size of the ferromagnetic domain.

Conversely, creating an anti-ferromagnetic $J_3$ (giving Figure 7) results in drastic changes to the phase diagram. We now find two new phases: the helimagnetic and $2\pi/3$ cuboctahedral states. That is, as $J_3$ becomes more negative, the region of parameter space where ferromagnetism is stable shrinks. A portion of this area goes to longitudinally-modulated anti-ferromagnetism, but none to the other forms of anti-ferromagnetism. Instead, these regions
Figure 6: Octahedral Phase Diagram for $J_1 = 1$, $J_3 = 1/3$. Note similarity to Figure 4

become filled by the $2\pi/3$ cuboctahedral state. Simultaneous with this effect, we find that helimagnetism is becoming stable around what would have been the triple point of the $2\pi/3$ cuboctahedral state, isotropic anti-ferromagnetism, and longitudinal anti-ferromagnetism. Figure 7 actually shows the end of this phenomena as a function of decreasing $J_3$ (Figure 8 shows the earlier stages). As $J_3$ decreases the continuous transitions of the helimagnetic state (that is, its boundaries with ferromagnetism and isotropically-modulated anti-ferromagnetism) spread apart. However, this effect is countered by a simultaneous narrowing due to the first order transitions (the phase boundaries with $2\pi/3$ cuboctahedral state and longitudinally-modulated anti-ferromagnetism approach each other). As $J_3 \rightarrow -\frac{1}{2}J_1 < 0$, this narrowing is complete and helimagnetism ceases to be a possible ground state.
Figure 7: Octahedral Phase Diagram for $J_1 = 1, J_3 = -1/3$. First instance of helimagnetism and $2\pi/3$ cuboctahedral.

Further tuning $J_3 < 0$ (giving Figure 8) sheds further light upon the shifting (as a function of $J_3$ in $J'_2/|J_1| \times J_2/|J_1|$) of the first and second order transitions of the helimagnetic state (i.e. why the helimagnetic state is confined to a finite area). We see here that the helimagnetic state is still increasing its area as a function of decreasing $J_3$ (this is principally evidenced by the continued presence of the helimagnetic - ferromagnetic transition, which disappears by the time we reach Figure 7), but we already have the first order transitions starting to impinge upon this region where helimagnetism is possible.

When $J_1$ is already anti-ferromagnetic, increasing ferromagnetic $J_3$ (giving Figure 9) is not the same as tuning $J_{1\text{eff}}$, as it was in Figure 6. Instead, the effect is close to Figure 7.
Figure 8: Octahedral Phase Diagram for $J_1 = 1, J_3 = -1/10$. Note difference in helimagnetic phase boundaries in Figure 7.

(where $J_1$ and $J_3$ have opposite signs to those they possess in Figure 9). In this case, though, we do not have any helimagnetic ground state. Instead, we find that the $\pi/3$ cuboctahedral state and the longitudinally-modulated anti-ferromagnet continue to expand, now at the expense of the 3-sublattice 120° state. This similarity with Figure 7 is quite surprising, since we would expect a change of sign in the couplings completely change the ground states and their topology.

Even when $J_1$ and $J_3$ are both anti-ferromagnetic (as in Figure 10), we do not find a symmetry with the $J_1$ anti-ferromagnetic, $J_3 = 0$ states (Figure 5). While we might expect $J_1$ and $J_3$ to combine and become part of a $J_{1\text{eff}}$ (this occurred between Figures 4 and 6,
$J_1 = -1, J_3 = 1/3$

![Octahedral Phase Diagram for $J_1 = -1, J_3 = 1/3$.](image)

Figure 9: Octahedral Phase Diagram for $J_1 = -1, J_3 = 1/3$. Circed point is the double-twist state.

where the couplings were ferromagnetic). In particular, we find the $\pi/3$ cuboctahedral state in Figure 5, but this state is nowhere to be found in Figure 10.

This presence of the $\pi/3$ cuboctahedral state actually implies that there cannot such an effective coupling description. Every state found in Figures 4, 5, 6, and 10 has an energy that is linear in the couplings, so that the $J_1, J_3$ contribution is of the form $aJ_1 + bJ_3$. Furthermore, all of these states except the $\pi/3$ cuboctahedral (i.e. ferromagnetism, 3 sublattice 120° state, and anti-ferromagnetism) have sign($a$) = sign($b$). But this is not case for the cuboctahedral states (see Table II for the exact values of $a$ and $b$). Finally, the $J_{eff}$ perspective concerns specific states (or parametrizations of states), and are not necessarily global. That is, the
$J_1 = -1, J_3 = -1/3$

Figure 10: Octahedral Phase Diagram for $J_1 = -1, J_3 = -1/3$. Note the difference from Figure 5, unlike the Figure 4, Figure 6 symmetry.

$J_{\text{eff}}$ for the $\pi/3$ cuboctahedral state would be different from that of the 3 sublattice 120° state. This is why we cannot explain the relation of Figure 5 to Figure 10 in terms of effective couplings.

To approach the phase diagram given in Figure 11, it is best to take two steps. First, from Figure 4, we increase $J_3$ to 1/3, giving us Figure 6. After that, we decrease $J_1$ to 0, giving us Figure 11. This second step is somewhat subtle, though, since we had previously normalized all of our couplings to $J_1$, which plainly fails here. Instead, we normalize with respect to $|J_3|$. The process of reducing $J_1$ also has the effect of shrinking the ferromagnetic domain, which is entirely expected.
Unlike the approach to $J_1 = 0, J_3 = 1/3$ that we took in Figure 11, the approach to $J_1 = 0, J_3 = -1/3$ (Figure 12) is surprising. We again require two steps, first decreasing $J_3$ from Figure 5 to Figure 10, then increasing $J_1$ to Figure 12 (we must again renormalize appropriately). This process has the effect of shifting the boundary of the cuboctahedral states in some rather surprising ways. Increasing $J_1$ (decreasing $J_3$) reduces the domain of stability of the $\pi/3$ cuboctahedral while increasing the domain of the the $2\pi/3$ cuboctahedral
Figure 12: Octahedral Phase Diagram for $J_1 = 0, J_3 = -1/3$. Note that $2\pi/3$ cuboctahedral persists, making this slice of parameter space similar to that of Figure 5. Circled point is asymmetric conic (stacking vector 100).

state. When $J_1 = 2J_3$, the trade-off is complete and the two cuboctahedral states are degenerate. Furthermore, while the route we illustrate here (using slices of parameter space for various $J_1, J_3$) for the $2\pi/3 - \pi/3$ cuboctahedral transition passes through a region where neither cuboctahedral state exists (as functions of $J_2, J'_2$), there are other routes where both cuboctahedral states coexist (in practice, this will likely produce a large but measure-zero region where the $120^\circ$ state dominates, see section VI C for details of this state).
C. Beyond the Basic States

1. Conic States, Stacking Vectors, and the Chain Lattice

As previously mentioned, there were a couple of states that do not fit into this framework even when \( J_4 \neq 0 \neq J'_4 \). The first of these is the asymmetric conic state. In canonical coordinates, this takes the form:

\[
\begin{align*}
    s_x &= (\hat{n}_1 \cos \psi z + \hat{n}_2 \sin \psi z) \cos \alpha + \sin \alpha \hat{n}_3 \quad (4.3.1) \\
    s_y &= s_x \quad (4.3.2) \\
    s_z &= (\hat{n}_1 \cos \psi z + \hat{n}_2 \sin \psi z) \cos \beta - \sin \beta \hat{n}_3 \quad (4.3.3)
\end{align*}
\]

where \( \hat{n}_i \) is a Cartesian basis vector in spin space, \( \psi \) is a helimagnetic (or polar) angle between 0 and \( \pi \), and \( \alpha \) and \( \beta \) are conic (or azimuthal) angles between 0 and \( \pi/2 \). This state was found for \( J_1, J'_2, J_4, J'_4 = 0 > J_2, J_3 \), which is precisely the phase boundary between isotropic and longitudinal anti-ferromagnetism (see Figure 12) [24]. It is possible, however, that the inclusion of \( J_4 \) terms will stabilize this state (or, more likely, \( J_5 \)). This is partially supported by the observed results for the chain lattice, which was motivated by the asymmetric conic state in the octahedral lattice.

There are two particular important properties to the asymmetric conic state in the octahedral lattice. First, the spins are solely a function of one variable - \( z \) in this case. Second, \( s_x = s_y \). This implies that all spins located on the plane \( z = \text{constant} \) are equivalent. In that case, it is perfectly reasonable to treat the entire lattice as two sublattices, rather than three.

We can go further, though, by noting how all the properties of this lattice are preserved when projecting the lattice onto the 001 axis. This reduces a three dimensional lattice with three sublattices to a one dimensional lattice with two sublattices while simultaneously enforcing the \( s_x = s_y \) requirement. In this context, the 001 vector is called a stacking vector \( \vec{n} \), and the transformation between the three dimensional and one dimensional lattices is

\[
z = \vec{n} \cdot \vec{a} \quad (4.3.4)
\]

where \( \vec{a} \) is a position vector \( \vec{r}(s) \) or a coupling vector \( \mathbf{R}(J) \). Notice that, in this new lattice, the lattice sites are located at integer or half-integer \( z \). We cannot reduce this lattice to
a one dimensional Bravais lattice with cell length $1/2$, because the half-integer and integer sites are not equivalent. While the octahedral lattice had each sublattice interchangeable, the spins located at integer sites correspond to twice as many spins in the octahedral lattice as spins located on half-integer sites. In particular, this implies that coupling constants are different for each sublattice.

In the chain lattice, we adopt the following convention for labeling couplings. Once again, we order couplings be first-nearest neighbor, second nearest, etc. Odd numbered couplings link sites in different sublattices, and so are the same for both sublattices. Even couplings link sites within a sublattice, and are inequivalent. For integer sites, we use an unprimed $j$ (we reserve $J$ for the couplings of the octahedral lattice), whereas half-integer sites have primed $j$. With this convention, the specific $J \rightarrow j$ transformation from the octahedral to the chain lattice with $\vec{n} = 001$ is

$$
\begin{pmatrix}
    j_0 \\
    j_1 \\
    j_2 \\
    j'_2
\end{pmatrix} =
\begin{pmatrix}
    4 & 2 & 4 & 4 & 2 \\
    8 & 16 \\
    2 & 8 & 4 & 4 \\
    1 & 4
\end{pmatrix}
\begin{pmatrix}
    J_1 \\
    J_2 \\
    J_3 \\
    J_4 \\
    J'_4
\end{pmatrix}
$$

while $\vec{n} = 110$ gives

$$
\begin{pmatrix}
    j_0 \\
    j_1 \\
    j_2 \\
    j'_2 \\
    j_3 \\
    j_4 \\
    j'_4
\end{pmatrix} =
\begin{pmatrix}
    2 & 1 & 2 & 4 & 2 & 1 \\
    8 & 8 \\
    2 & 2 & 2 & 4 & 4 & 4 \\
    2 & 4 \\
    8 \\
    2 \\
    1
\end{pmatrix}
\begin{pmatrix}
    J_1 \\
    J_2 \\
    J_3 \\
    J_4 \\
    J'_4
\end{pmatrix}
$$

Interestingly, these transformations explain why we do not observe the conic states in the octahedral lattice except on phase boundaries. For the conic to be stable in the octahedral
Figure 13: The transformation from the Octahedral to the Chain lattice for \( n = 001 \). Integer sites (100 and 010) are circles, half-integer sites (001) are squares. Dotted lines show structure, dashed lines show mapping, and solid lines show couplings.

lattice, it must be stable in the chain lattice. But conics in the chain lattice require at least \( J_3 \) (see section V A 4). There are only two ways that this requirement of the chain lattice’s couplings can be met by the octahedral lattice’s couplings. Either the ground state in the octahedral lattice uses a stacking vector other than 100 (or 111, see section IV C 3 for details) and our previous range of couplings (i.e. \( J_1 \) through \( J_4 \)) or the ground state uses a 100 stacking and at least \( J_5 \) (or \( J_6, J'_6 \), depending on the type of conic). Since most real materials have negligible \( J_4 \), the utility of even considering \( J_{5+} \) couplings is questionable. Therefore, it was preferable to search for states with stacking vectors other than 100, in the hopes that these states would be conics.
2. Double Twist \((Q, Q, 0)\) State and the \((110)\) Stacking Vector

In principle, any other stacking vector is possible and gives interesting results for the \(J \rightarrow j\) transformation. For example, the \(\vec{n} = 211\) stacking vector maps \(J_1\) through \(J_4\) to \(j_1\) through \(j_6\). The possibility of couplings in the chain lattice being longer-range than those of the octahedral lattice is part of the reason that we focus more upon the effects of \(j_4, j'_4\) in the chain lattice. A stacking vector of particular interest is the 011 vector, since it is the next longest after 001 and gives further generality to the octahedral-chain transformation.

In an effort to locate such a stacking vector, we relied upon Luttinger-Tisza analysis. The logic here is relatively straightforward. The Luttinger-Tisza wave-vectors \(q\) denote the directions of spatial variation, so we would expect the direction of variation to be parallel to the stacking vector (that is, the stacking vector is a direction of variation). Minimizing the eigenvalues of the octahedral Luttinger-Tisza \(\tilde{J}(q)\) matrix as a function of \(J\)'s does give wave-vectors along 110 and its permutations. For the purpose of simulations with iterative minimization, it was preferable to tune \(q_m(\{J\})\) to be a rational number, and therefore commensurate with periodic boundary conditions.

For a period \(3/8\), \(q_m \propto 110\), we found the couplings \(J_1 = -2, J_2 = -3.771657\) and \(J_3 = 1\) (all others 0, note that this is along the phase boundary of longitudinally-modulated anti-ferromagnetism and isotropically-modulated anti-ferromagnetism) (see Figure 9). Plugging these couplings into the iterative minimization algorithm, however, failed to yield any stacking vectors. Instead, it produced a configuration dubbed the double twist state. This was, by far, the most complicated state encountered, and the idealizations discussed in the following paragraph are very tenuous. The difficulty in idealizing this configuration was exacerbated by the need to maintain a commensurate \(q_m\) to remove defects in the iterative minimization, coupled with the extremely narrow range of parameter space where this state could exist (i.e. only on the transition between longitudinally-modulated anti-ferromagnetism and isotropically-modulated anti-ferromagnetism - and then only for a small range of couplings). As such, we were unable to find any other combination of couplings that would produce a double twist state with commensurate \(q_m\).
The idealized configuration for the double twist state is approximately:

\[ s_\alpha = \Gamma U_\alpha^{(x)} \cos(Qy - \phi_y) \cos(Qz - \phi_z) \vec{A}_0 \]
\[ + U_\alpha^{(y)} \sin(Qz - \phi_z) \vec{B}(x) \]
\[ + U_\alpha^{(z)} \sin(Qy - \phi_y) \vec{C}(x) \]

where \( \alpha \) is the sublattice index and \( Q, \Gamma, \phi_y, \phi_z \), and \( U \) are all variational parameters. The presence of so many variational parameters (6 in total) is a testament to the general uncertainty clouding this state. We do know that

\[ \vec{B}(x) = \vec{B}_0 \cos Qx + \vec{C}_0 \sin Qx \]
\[ \vec{C}(x) = -\vec{B}_0 \sin Qx + \vec{C}_0 \cos Qx \]

where \( \vec{A}_0, \vec{B}_0, \vec{C}_0 \) define an orthonormal basis. That is, two of the axes spiral around the third (\( \vec{A} \)) as a function of \( x \) (of course, there is nothing particular unique about \( x \). This was merely the observed case. If we permute \( \alpha = 1, 2, 3 \) and \( i = x, y, z \) in the same manner we will define the same state). \( \Gamma \) is a numerical factor between 1 and 1.5 (found \( \Gamma = 1.36 \) through iterative minimization, but that is only a rough guide here). \( \Gamma \) is introduced because the amplitudes along \( \vec{A}, \vec{B}, \vec{C} \) for different sublattices have a very symmetric structure (corresponding to the Luttinger-Tisza eigenvectors), which we make precise in \( U_\alpha^{(i)} \). However, this symmetry in amplitudes is only preserved for ratios of amplitudes, the components along \( \vec{A} \) appear larger than the values given by \( U_\alpha^{(x)} \) by some constant factor - which we call \( \Gamma \). \( U_\alpha^{(i)} \) gives the sublattice-dependent amplitude for each term, with \( (i) \) denoting the preferred direction of the term. If we treat \( U_\alpha^{(i)} \) as a matrix, then we find

\[ U = \begin{bmatrix} \pm a & b & b \\ b & -a & b \\ b & b & -a \end{bmatrix} \]

where the exact values of \( a \) and \( b \) correspond to Luttinger-Tisza eigenvector amplitudes (for the \( \pm \) + was observed numerically, but we should expect – for the \( a \)'s and \( b \)'s to truly to a Luttinger-Tisza eigenmode). \( Q \) is, of course, the angle of rotation between adjacent spins, while \( \phi_i \) is a phase factor, which is to be determined variationally.

One major problem with this parametrization of the ground state is that it is not normalized. This lack of normalization is due to our method of deriving the parametrization.
After optimizing the ground state through iterative minimization and taking the Fourier Transform, we found that the majority of the weighting lay upon the \((QQ0)\) modes. However, there were small components in other modes. We set these components to 0 and then attempted to parametrize the resultant state. It is unclear whether the ground state should include these other modes, although it is clear that the existing parametrization needs to be modified in some way (for example, making \(\phi_i\) become a function of position). Were it possible to modify the parametrization while preserving the exclusive use of \((QQ0)\) modes, so much the better (especially because we should then be certain that this was the ground state).

Having defined the double twist state, we are compelled to ask what exactly is going on. As previously mentioned, the spins all spiral around \(\vec{A}_0\) as \(\vec{B}\) and \(\vec{C}\) rotate. This rotation is conic rather than helimagnetic, with a (generically) non-zero component of the spins pointing along \(\vec{A}\). What distinguishes this state from the conics explored in the chain lattice (see section V A 4) is the azimuthal angle and the polar angle both vary as functions of position. This option is unavailable in the chain lattice, where there is only one spatial component. That is, as we move between spins in an \(x=\text{constant}\) plane, the conic angle will oscillate between 0 and \(\pi\).

The double-twist state bears more resemblance to the cuboctahedral states than to any conic, in that the ground state is a composition of different wave-vectors all related by symmetry. While it is true that the cuboctahedral state is composed of \(\frac{1}{2}00\) modes, not \(QQ0\) modes, this merely implies that their exact spatial patterns will be distinct [25]. We can still group them within the same class of states, which we dub one-star states (see section III A), since all of the wave-vectors come from the same star of symmetry-related \(q\)'s.

Clearly, the double twist state cannot reasonably be considered an instance of a stacking along 110. The failure of these couplings to produce a stacked state underscores the incompleteness of the Luttinger-Tisza method. Whenever the ground state is constructed out of modes unrelated by symmetry, Luttinger-Tisza gives little insight into its spatial order. The situation is particularly acute in the case of conic spirals, which also necessarily combine multiple modes. These modes are not necessarily the optimum modes, but are typically within a neighborhood of the optimal modes.

As a further example, consider the phase diagram of optimal modes as a function of couplings. When the ground states are composed of single modes, then the phase boundaries
of the ground states will necessarily include the phase boundaries of the Luttinger-Tisza optimal modes. But there will be additional phase boundaries that a Luttinger-Tisza analysis will miss. And when ground states become multi-mode (or mixed states), then the correspondence between the Luttinger-Tisza phase boundaries and the ground state phase boundaries becomes approximate. These concepts are further developed in section VI A.

3. Existence of Helimagnetism

We are now in a position to justify the existence of helimagnetism in the octahedral lattice, despite some rather stringent constraints on the form of helimagnetism adopted. As a result of this analysis, we will also be able to justify the absence of any “classical” anti-ferromagnetism, which would be the isotropically-modulated anti-ferromagnetic state with $\hat{n}_i = \hat{n}$. This is useful, as an intuitive approach would suggest that, for ferromagnetic interactions between sublattices, this would decrease the energy at no cost.

First, consider the parametrization of a helimagnetic state. Such a state will require correlations between sublattices, so, without loss of generality, the spins must all be coplanar and sublattices can differ by no more than a phase. For this to be true, there must be some wave-vector such that all spins can be written in the form

$$s_\alpha(\vec{r}) = \cos(\vec{k} \cdot \vec{r} + \phi_\alpha)\hat{n}_1 + \sin(\vec{k} \cdot \vec{r} + \phi_\alpha)\hat{n}_2$$

(4.3.11)

where $\hat{n}_i$ is a basis vector in spin space. Notice now that $s_i(\vec{r} + \vec{r}_\perp) = s_i(\vec{r})$ where $\vec{r}_\perp \cdot \vec{k} \equiv 0$. Now if this is true, then it follows that all variation is along one dimension and $\vec{k}$ is also a stacking vector (even if $\vec{k}$ is incommensurate). But using stacking vectors to transform the octahedral lattice to a one-dimensional chain will necessarily produce a non-Bravais lattice unless the stacking vector is (111) (or a permutation of sign). And any helimagnetic mode in the one-dimensional non-Bravais lattice will break normalization in the octahedral lattice, since some spin directions would be represented more than others (this was allowable for conics because they mixed multiple modes, but helimagnets are explicitly single mode). Therefore, the only allowable stacking vector (and by implication, wave-vector) is (111).

The exception is for the case of a stacking vector along one of the 4 non-trivial alterations in sign of the 111 vector ($-\hat{n}$ is the same as $\hat{n}$). These stacking vectors will treat all the sublattices equally, producing a one dimensional Bravais, not a chain lattice. The
transformation, through $J_4$, is

$$\begin{pmatrix} j_0 \\ j_1 \\ j_2 \end{pmatrix} = 3 \begin{pmatrix} 2 & 2 & 1 \\ 2 & 1 & 2 \\ 2 & 2 & 1 \end{pmatrix} \begin{pmatrix} J_1 \\ J_2 \\ J_3 \\ J_4 \end{pmatrix}$$

To account for a single spin in the octahedral lattice, the energy for a helimagnet is:

$$H = \frac{1}{3}(-j_0 - j_1 \cos \psi - j_2 \cos 2\psi).$$

Which we can minimize to find that the optimal angle is:

$$\cos \psi^* = \frac{-j_1}{4J_2}$$

and

$$= \frac{- (2J_1 + J_2 + 2J'_2 + 4J_3)}{4(2J_3 + 2J_4 + J'_4)}$$

assuming that this is between -1 and 1 (this immediately excludes helimagnetism without long-range couplings, i.e. those beyond $J_2$). Notice that all of the phase transitions for helimagnetism are expressed as functions of $\psi$. Performing a comparison of energies within the various domains of parameter space reveals that, without $J_4, J'_4$, the only possible range of couplings where helimagnetism exists is within $J_1 > 0, -\frac{1}{2}J_1 < J_3 < 0$.

Finally, we use this mapping to eliminate the possibility of anti-ferromagnetism with coupled sublattices. When we plug in $\psi = \pi$ to the helimagnet, we get an anti-ferromagnetic state with the spins of different sublattices pointing along the same spin direction, but we find the energy to be

$$H = \frac{1}{3}(-j_0 + j_1 - j_2)$$

$$= J_2 + 2J'_2 - 4J_4 - 2J'_4$$

precisely what we find in Table II for the decoupled isotropic anti-ferromagnet (at least to $J_4$). This justifies our previous claim for ignoring sublattice-coupled anti-ferromagnets - namely that all the couplings between sublattices really do cancel.

These results illustrate the power of the stacking vector formalism. It transformed a seemingly intractable problem on a three-dimensional lattice to the optimization of a quadratic form for a one-dimensional lattice.
V. THE CHAIN LATTICE

While the stacking vector approach does not work all the time, it clearly does work some of the time. And when this is the case, it is much easier to analyze the ground states in the one-dimensional lattice than in the octahedral lattice. For most stacking vectors, the one-dimensional lattice has a motif with two sites (integer and half-integer), which we call the chain lattice. The ease of analysis in the chain lattice is partially because it is easier to use iterative minimization to simulate large lattices when the site scaling is $O(L)$ instead of $O(L^3)$, especially as all of calculations scale with the number of sites. But there is also reason to study the chain lattice in its own right. It is the simplest possible non-Bravais lattice, but we will find considerable diversity in its ground states (even greater than in the octahedral lattice).

As we would expect from the octahedral-chain transformation, there is some resemblance between the phase diagrams of the two lattices. However, the resemblance is primarily qualitative, in particular the division of phase diagram into quadrants (this time we will use $j_2/|j_1| \times j'_2/|j_1|$ coordinates - the reflection in our conventions is relatively unimportant as the meaning of primed and unprimed couplings is different in the two lattices). A property of the chain lattice that is particularly useful in understanding its phase diagram is the equivalence of two sublattices under exchange of couplings. That is, if we make the reflection in parameter space $j_2 \rightarrow j'_2, j'_2 \rightarrow j_2$ and $j_4 \rightarrow j'_4, j'_4 \rightarrow j_4$ (reflections about $j_2 = j'_2$ and $j_4 = j'_4$, respectively), then we have merely changed the conventions between integer and half-integer sites. Therefore, the phase diagram must reflect this symmetry.

Moreover, along $j_2 = j'_2, j_4 = j'_4$, the two sublattices are exactly equivalent and the chain lattice degenerates to a one dimensional Bravais lattice with half the cell size. And by Luttinger-Tisza analysis, we know that such states must be described by some form of helimagnetism. This will provide a convenient sanity check of the analysis of the phase diagram. In performing this analysis, we again assume that $j_1, j_2,$ and $j'_2$ are in some sense stronger than $j_3, j_4,$ and $j'_4$. What's more, we require that no more than one of couplings $(j_3, j_4, j'_4)$ is non-zero. This distinction between $(j_1, j_2, j'_2)$ and $(j_3, j_4, j'_4)$ preserves our division between short and long-range couplings.
A. The States of the Chain Lattice

1. **Collinear (0) States**

Just as we saw in the octahedral lattice (section IV A 1), there are two states possible in the first quadrant \((j_2/|j_1|, j'_2/|j_1| > 0)\). For \(j_1 + j_3 > 0\), all the couplings are positive and the ground state is ferromagnetic (see Figures 17, 19, 20, 24 and 25). For \(j_1 + j_3 < 0\), the ground state is anti-ferromagnetic (see Figures 18, 21, 22, 26, and 27). Since each sublattice has ferromagnetic couplings \((j_2, j'_2 > 0)\), there is only one spin direction per sublattice. But because the net interaction between sublattices is anti-ferromagnetic, the spins in different sublattices are anti-parallel. This is, in fact, the standard anti-ferromagnetic state for a one dimensional Bravais lattice. Note that its analog in the octahedral lattice is the 3 sublattice 120° state, which is like an anti-ferromagnetic arrangement with three sublattices.

2. **Helimagnetic (q) States**

For the third quadrant \((j_2/|j_1|, j'_2/|j_1| < 0)\), there is only one ground state. In this region, the ground state is helimagnetic (see Figures 17 through 27). Note that, as longer-range \(j\)'s are introduced, the optimal angle becomes a higher order function of \(j\)'s (the solution to an \(n^{th}\) degree polynomial for non-zero \(j_{n+1}\)). This limits our ability to explicitly find the phase boundaries in the more interesting cases (we can use computer algebra systems or numerical optimization for specific couplings, but neither solution is ideal). In particular, it prevents an explicit calculation of any first-order phase transitions that may be present. This issue is even more problematic when we turn to conic spiral states, forcing us to rely upon numerical detection of the phase boundaries.

3. **Splayed (0, 1/2) States**

In the second and fourth quadrants \((j_2j'_2/j_1^2 < 0)\), there are now two different ground states possible (they’re the same in both quadrants). For \((j_1 + j_3) > 0\), the ground state is a ferromagnetic splayed state (see Figures 17, 19, 20, 24, and 25). For \(j_1 + j_3 < 0\), the ground state is a ferrimagnetic [26] splayed state (note that this phase boundary is identical to the ferromagnetic-anti-ferromagnetic one for the first quadrant) (see Figures 18, 21, 22, 23, 26,
and 27). These states are most easily understood using the alternating conic formalism (we will discuss alternating conics in more detail in section V A 4):

\[ s_A = (\hat{n}_1 \cos 2\psi z + \hat{n}_2 \sin 2\psi z) \cos \alpha + (-1)^2 \sin \alpha \hat{n}_3 \]  
\[ s_B = \hat{n}_1 \cos 2\psi z + \hat{n}_2 \sin 2\psi z \]  

where \( j_2(s_A) < 0, j_2(s_B) > 0, \psi \in [0, \pi] \) (the 2 is conventional so that the angle between half-integer steps is \( \psi \)), \( \alpha \in [0, \pi/2] \) and \( \hat{n}_i \cdot \hat{n}_j = \delta_{ij} \). In the case of splayed states, \( \psi = 0 \) (ferromagnetism) or \( \psi = \pi \) (ferrimagnetism). It is interesting to note that while the splayed state is coplanar, it cannot be explained by a single Luttinger-Tisza eigenmode or as a helimagnet. That is, the ground state must be composed of both \( q = 0 \) and \( q = 1/2 \) modes.

4. Alternating (1/2, q) and Asymmetric (0,q) Conics

When \( j_3, j_4 \) or \( j'_4 \) are non-zero, then the conic states are stabilized near the previous phase transitions (primarily the helimagnet - ferromagnetic splayed transition or the helimagnet - ferrimagnetic splayed transition, although numerical optimization of the energy suggest that there might be narrower regions of conic stability around the old conic - linear and (even narrower) the helimagnetic - linear transitions). The phase boundaries of the conic states cannot be determined explicitly. The conics can be grouped into two classes: alternating (see Figures 22, 23, 25, and 27) and asymmetric conics (see Figures 21, 26, and 27).

The alternating conic is parametrized by a formalism introduced in discussing the splayed states (see equations 5.1.1 and 5.1.2 of this section). It typically requires a (ferromagnetic)
Figure 15: The Alternating Conic. Left sublattice (red) is helimagnetic. Right sublattice (blue) is conic - traces out two cones along the 001-axis in spin space.

$j_4$ or $j'_4$ to stabilize, which makes sense as the sublattices are treated asymmetrically in this state. It is also stabilized with $j_3$, but for a much smaller range of parameters (i.e. the volume of parameter space where the alternating conic is stable is smaller for $j_3$ than for $j_4$ or $j'_4$). The alternating conic state allows for a smooth transition between the helimagnetic and splayed states, as it can parametrize both.

The asymmetric conic, on the other hand, is a more complex state. It typically requires a $j_4$ coupling to stabilize, but it is also stable for $j_3$. It is parametrized by

\[ s_A = (\hat{n}_1 \cos 2\psi z + \hat{n}_2 \sin 2\psi z) \cos \alpha + \sin \alpha \hat{n}_3 \]  
\[ s_B = (\hat{n}_1 \cos 2\psi z + \hat{n}_2 \sin 2\psi z) \cos \beta - \sin \beta \hat{n}_3 \]  

where $\psi \in [0, \pi], \alpha, \beta \in [0, \pi/2]$. In this formalism, an ferrimagnetic splayed state is parametrized by $\psi/2 = \pi/2 = \alpha$ or $\beta$ while there is no parametrization for the ferromagnetic splayed state. This provides greater support to our observation that the asymmetric conic does not have a transition to the ferromagnetic splayed state. This makes sense, as the opposite signs of $\alpha$ and $\beta$ clearly imply that the net interaction between sublattices is anti-ferromagnetic.
To determine if a phase transition is first or second order, it is helpful to consider the concept of encompassing parametrization (introduced in section III C and recapitulated here). We call a parametrization encompassed if it can be expressed by another parametrization with particular values of free parameters. For example, ferromagnetism and antiferromagnetism are both encompassed parametrizations of a helimagnetic parametrization ($\psi = 0$ or $\pi$, respectively), while helimagnetism is itself a encompassed parametrization of a conic state ($\alpha, \beta \rightarrow 0$). Conversely, helimagnetism is the encompassing parametrization of (anti-)ferromagnetism. The generality of a state (which is related to the number of free parameters, and therefore the number of distinct states that can be parametrized) is distinct from the concept of encompassing. An encompassing state is necessarily more general than the encompassed state, but a more general state is not necessarily an encompassing one. For example, the asymmetric conic is more general than the alternating conic (in the sense that there are more distinct asymmetric conic states than there are alternating conic states), but...
neither conic encompasses the other. This absence of an encompassing parametrization is important. In particular, there is no splayed state (ferromagnetic or ferrimagnetic) that encompasses the the generic helimagnet ($\psi \neq 0, \pi$), nor does the generic helimagnet encompass the splayed states. Similarly, the asymmetric conic does not encompass the ferromagnetic splayed state (and vice versa).

When a phase transition is between a state and its encompassed parametrization, then it is (unless otherwise noted) second order. Conversely, if neither state is an encompassing parametrization of the other, then the transition is always first order. This is because, as the couplings approach the phase boundary and an encompassed parametrization exists, the free parameters will tend towards the values of the encompassed parametrization. This limit is not always intuitive when examined numerically, particularly for the conic states, as they may possess multiple parametrization of the same state (for example, both $\alpha, \beta \to \pi/2; \psi \to 0$ and $\psi \to \pi; \alpha, \beta \to 0$ are anti-ferromagnetic; the asymmetric conic parametrization of the ferrimagnetic splayed state is even more complex).

Particular care should be taken in the examination of “pinning” transitions, where the free parameter becomes “pinned” to a particular value. For example, in the splayed state (using alternating conic parametrization, the optimal angles are

$$\psi = 0$$

$$|\cos \alpha| = \frac{|j_1 + j_3|}{2j_2^{(i)}}$$

(5.2.1)

(5.2.2)

(where the $j_i^{(t)}$ denotes the coupling appropriate to the sublattice - i.e. whichever is anti-ferromagnetic, as this state only exists within the second and fourth quadrants) Since $0 \leq |\cos \alpha| \leq 1$, the case of $|j_1 + j_3| > |2j_2^{(i)}|$ is outside of the domain of $\exists \alpha : \cos \alpha = |j_1 + j_3|/|2j_2^{(i)}|$. In this case, the angle $\alpha$ becomes pinned to the value that most nearly approaches the minimum - either 0 or $\pi/2$.

To summarize, we present the exact relations for the chain lattice in table IV.

In general, the equations for the phase boundaries may be found by minimizing the energy of each ground state separately and setting the difference of the two minima to 0. Since this problem often has no closed-form solution, we rely upon the numerical simulations as a guide. However, some of the phase boundaries can be found explicitly, and we present those below (note that, for specific choices of couplings, we can simplify the equations and thereby find explicit expressions for phase boundaries, but these specific cases are rather rare).
Examining the simplest phase diagrams for both the octahedral and chain lattices (Figures 4 and 5 and 17 and 18, respectively) reveals several striking aspects of this mapping principle. First of all, the topology of the phase boundaries are identical, even down to their linearity as functions of $j_2, j'_2$. In addition, the phase in the first quadrant is ferromagnetic for both. However, similarity beyond this is rather lacking. The other three quadrants are completely different. In particular, the ground states of octahedral lattice do not have stacking vectors for these quadrants, so there is no mapping that perfectly preserves the state. The splayed states do have certain limited analogies to the transverse- and longitudinally-modulated antiferromagnets, but there does not appear to be any commonality between the helimagnetic

<table>
<thead>
<tr>
<th>State</th>
<th>Spins</th>
<th>Energy</th>
</tr>
</thead>
<tbody>
<tr>
<td>FM</td>
<td>$s_A = \hat{n} = s_B$</td>
<td>$E = -2j_1 - j_2 - j'_2 - 2j_3 - j_4 - j'_4$</td>
</tr>
<tr>
<td>AFM</td>
<td>$s_A = \hat{n} = -s_B$</td>
<td>$E = 2j_1 - j_2 - j'_2 + 2j_3 - j_4 - j'_4$</td>
</tr>
</tbody>
</table>
| HM    | $s_A = \hat{n}_1 \cos 2\psi z + \hat{n}_2 \sin 2\psi z$ | $E = -2j_1 \cos \psi - (j_2 + j'_2) \cos 2\psi$
$\quad s_B = s_B$ | $-2j_3 \cos 3\psi - (j_4 + j'_4) \cos 4\psi$
| SFe   | $s_A = \hat{n}_1 \cos \alpha$ | $E = -2(j_1 + j_3) \cos \alpha - j_2 \cos 2\alpha - j'_2 - j_4 - j'_4$
$\quad +( -1)^2 \sin \alpha \hat{n}_3$ | $\cos \alpha = -(j_1 + j_3)/2j_2$
$\quad s_B = \hat{n}_1$ | $E_{\text{min}} = (j_1 + j_3)^2/2j_2 + j_2 - j'_2 - j_4 - j'_4$
| SFi   | $s_A = \hat{n}_1 \cos \alpha$ | $E = 2(j_1 + j_3) \cos \alpha - j_2 \cos 2\alpha - j'_2 - j_4 - j'_4$
$\quad +( -1)^2 \sin \alpha \hat{n}_3$ | $\cos \alpha = (j_1 + j_3)/2j_2$
$\quad s_B = -\hat{n}_1$ | $E_{\text{min}} = (j_1 + j_3)^2/2j_2 + j_2 - j'_2 - j_4 - j'_4$
| AIC   | $s_A = (\hat{n}_1 \cos 2\psi z + \hat{n}_2 \sin 2\psi z) \cos \alpha$ | $E = -2(j_1 \cos \psi + j_3 \cos 3\psi) \cos \alpha$
$\quad +( -1)^2 \sin \alpha \hat{n}_3$ | $+ j_2 - j_4 - j'_2 \cos 2\psi - j'_4 \cos 4\psi$
$\quad s_B = \hat{n}_1 \cos 2\psi z + \hat{n}_2 \sin 2\psi z$ | $+ -(j_2 \cos 2\psi + 1) + j_4 \cos 4\psi - 1 \) \cos^2 \alpha$
| AsC   | $s_A = (\hat{n}_1 \cos 2\psi z + \hat{n}_2 \sin 2\psi z) \cos \alpha$ | $E = -2(j_1 \cos \psi + j_3 \cos 3\psi) \cos \alpha \cos \beta$
$\quad + \sin \alpha \hat{n}_3$ | $+ 2(j_1 + j_3) \sin \alpha \sin \beta - j_2 - j'_2 - j_4 - j'_4$
$\quad s_B = (\hat{n}_1 \cos 2\psi z + \hat{n}_2 \sin 2\psi z) \cos \beta$ | $+ [-j_2 \cos^2 \alpha + j'_2 \cos^2 \beta] \cos 2\psi - 1)$
$\quad - \sin \beta \hat{n}_3$ | $+ [-j_4 \cos^2 \alpha + j'_4 \cos^2 \beta] \cos 4\psi - 1)$

Table IV: Parametrizations and Energies of the Chain Lattice States

2. Initial Comparisons to the Octahedral Lattice Phase Diagrams

Examining the simplest phase diagrams for both the octahedral and chain lattices (Figures 4 and 5 and 17 and 18, respectively) reveals several striking aspects of this mapping principle. First of all, the topology of the phase boundaries are identical, even down to their linearity as functions of $j_2, j'_2$. In addition, the phase in the first quadrant is ferromagnetic for both. However, similarity beyond this is rather lacking. The other three quadrants are completely different. In particular, the ground states of octahedral lattice do not have stacking vectors for these quadrants, so there is no mapping that perfectly preserves the state. The splayed states do have certain limited analogies to the transverse- and longitudinally-modulated antiferromagnets, but there does not appear to be any commonality between the helimagnetic
and isotropically-modulated anti-ferromagnet.

Moreover, the order of the phase boundaries is reversed in these two lattices (at least for these pairs of phase diagrams - Figures 4 and 5 vs 17 and 18). Where we had previously encountered first order transitions in the octahedral lattice, we find second order transitions in the chain lattice (and vice versa). To some extent, this can be expected by the additional degrees of freedom in the octahedral lattice (the ground states are functions of three spatial variables, not one). This introduces the possibility that the ground state cannot be preserved in transforming to the chain lattice (i.e. no stacking vectors exist), a property that is fairly common among the one-star states. When the couplings are frustrated, the ground state will either require additional variational parameters (introducing the possibility of encompassing parametrizations) or it will exploit the additional degrees of freedom presented by three spatial variables. As the number of spatial variables increases, so too does the likelihood that the ground state can be constructed without introducing free parameters. So we should expect fewer states with free parameters in the octahedral lattice.

However, this cannot be the whole story. Exploiting additional spatial degrees of freedom is an alternative to constructing the ground state by introducing free parameters, but the addition of spatial variables also allows for additional degenerate states at the phase boundaries (these states are first discussed in section III B, and will be analyzed further in section VIC). Since the presence of encompassing parametrization (which require free parameters or degenerate intermediate states) can produce second order transitions, we can clearly see why the second order transitions are in their particular location for each lattice. It is surprising, though, that each lattice has different reasons (free parameters vs degeneracy) for having second order transitions and that the locations of these second order transitions are

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<td>HM</td>
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Table V: Explicit Phase Boundaries of the Chain Lattice.
Figure 17: Chain Lattice Phase Diagram for $j_1 = 1, j_3 = 0, j_4 = 0, j'_4 = 0$. Note similarity of phase boundaries to Figure 4, despite the lack of other similarity.

opposites of each other.

When we introduce a ferromagnetic $j_3$ to Figure 17 (giving Figure 19), we see a similar effect as our transition from Figure 4 to Figure 6. But at this point, the chain lattice and the octahedral lattice clearly part ways. While both lattices have the ferromagnetic phase expand into the adjacent states, the chain lattice has several additional features not shared by the octahedral lattice. First of all, the ferromagnetic splayed phase also expands into the helimagnetic phase as $j_3$ increases, even while the ferromagnetic phase expands into it. Moreover, as the ferromagnetic splayed - helimagnetic phase boundary moves, it ceases to be a straight line and gains some curvature. This is because the ground state energy of each phase is no longer linear in $j_2^{(i)}$. This effect has no analogue in the octahedral lattice, where the ground states in the second and fourth quadrants are anti-ferromagnets, and therefore
Figure 18: Chain Lattice Phase Diagram for $j_1 = -1, j_3 = 0, j_4 = 0, j_4' = 0$. Note similarity of phase boundaries, not to Figure 5, but to Figure 10. 

Conversely, decreasing $j_3$ from Figure 17 (giving Figure 20) increases the prevalence of the helimagnetic state. The ferromagnetic splayed phases also expand into the ferromagnetic phase even while losing ground to the helimagnetic phase. One interesting effect, though, is to observe the reversal of curvature for the helimagnetic - ferromagnetic splayed phase boundary in Figures 19 and 20. This effect can be partially explained by examining the optimal angles of each phase:

$$\cos \alpha = \frac{(j_1 + j_3)}{2j_2}$$  \hspace{1cm} (5.2.3)

$$\cos \psi = \frac{j_2 + j_2'}{12j_3} \pm \sqrt{\left(\frac{j_2 + j_2'}{12j_3}\right)^2 - \frac{j_1 - 3j_3}{12j_3}}$$  \hspace{1cm} (5.2.4)

for ferromagnetic splayed and helimagnetic, respectively. For small $j_3$, $\cos \alpha$ does not change.
Figure 19: Chain Lattice Phase Diagram for $j_1 = 1$, $j_3 = 0.28$, $j_4 = 0$, $j'_4 = 0$. Solution no longer explicit, but determined through numerical optimization. Points denote location and phase of numerical optimization. Open circles denote helimagnetic phase, light grey circles splayed ferromagnetic, and dark grey ferromagnetic.

very much, whereas $\cos \psi$ changes a great deal (for fixed $j_1, j_3$, the graph of $-\sgn(b)x \pm \sqrt{x^2 - \sgn(b)a}$ defines a hyperbola, with $\sgn(j_3)$ determining the slope of the asymptotes). So for any given $j_1, j_2, j'_2$, the helimagnetic ground state depends strongly upon the sign of $j_3$.

Note also that, for $j_1 \cdot j_3 > 0$, a helimagnetic state requires a minimum magnitude of

$$|j_2 + j'_2| = 12j_3(j_1 - 3j_3)$$

which thereby defines a helimagnetic - ferromagnetic phase boundary (this is a pinning transition, as previously alluded to). This helps explain why we see a ferromagnetic ground
state near the origin in Figure 19. On the other hand, for anti-ferromagnetic $j_1 \cdot j_3 < 0$, helimagnetism is stable near $j_2, j'_2 \approx 0$ as there are possible values of $\psi$ for the corresponding $\cos \psi$. This dependence of the ground state about $j_2, j'_2 = 0$ upon the sign of $j_1 \cdot j_3$ is also found in Figures 21 and 22, even though the transition is to anti-ferromagnetism there.

3. Non-Coplanar States in the Chain Lattice Phase Diagram

Moving from Figure 18 to Figure 21 by adding a ferromagnetic $j_3$ shifts the phase boundaries in the same manner as in the move from Figure 17 to Figures 19 and 20. This includes the curvature of the ferrimagnetic splayed - helimagnetic phase boundary and the shrinking of the anti-ferromagnetic state. But this region of parameter space is not simply a repetition of previous phenomena. In much the same way that tuning $j_3$ in the octahedral lattice...
Figure 21: Chain Lattice Phase Diagram for $j_1 = -1, j_3 = 0.28, j_4 = 0, j'_4 = 0$. First state with asymmetric conic, and therefore first instance of non-coplanar ground states in the chain lattice. Open circles denote helimagnetic phase, light grey circles splayed ferrimagnetic, dark grey anti-ferromagnetic, and black asymmetric conic.

produced helimagnetic states around the phase boundaries of the more common states, so in the chain lattice do we find that the asymmetric conic state becomes stabilized around what would be the anti-ferromagnetic, ferrimagnetic splayed, helimagnetic triple point. And unlike the helimagnetic state in the octahedral lattice, which had both first order and second order transitions, the asymmetric conic has only second order transitions (this is because it is an encompassing parametrization of every other state in Figure 21).

A property of the phase diagram in Figure 21 that is not found in the previous diagrams is that the asymmetric conic - anti-ferromagnetic phase boundary has some curvature...
(as a function of $j_2, j'_2$). Previously, the phase boundaries of the “first quadrant” states (anti-ferromagnetism and ferromagnetism in the chain lattice, 3-sublattice 120° state and ferromagnetism in the octahedral lattice) were strictly linear (as functions of $j_2, j'_2$). This can be partially explained by the fact that the asymmetric conic includes both helimagnetic and ferromagnetic modes, and that changing the ratio of the two (i.e. changing the conic angles $\alpha, \beta$) allows the conic state to approach the anti-ferromagnetic state in manner that does not depend linearly on the couplings. This explanation is given further weight by noting that the asymmetric conic is the only state where the boundary with the ferromagnetic and anti-ferromagnetic states has some definite curvature (again, as a function of $j_2, j'_2$).

It is true that similar arguments could be constructed for the alternating conic state or the splayed states and the ferromagnetic or anti-ferromagnetic states. However, we can analytically solve for the phase boundaries between the splayed and (anti-)ferromagnetic states, which ends up being a perfect square\footnote{This is derived from a variational principle and requires solving a transcendental equation. Details are provided in Table V.}: \((2j_2^{(0)} \pm |j_1 + j_3|)^2/2j_2 = 0\), see Table V). So, amazingly, these phase boundaries are strictly linear in the couplings, even though we’ve introduced a variational parameter.

Lastly, as to why we do not find curved phase boundaries between the alternating conic and ferromagnetic or anti-ferromagnetic ground states, the answer is fairly simple. The fact is that, properly speaking, the alternating conic shares no phase boundaries with the ferromagnetic or anti-ferromagnetic, so the question of the boundary’s curvature does not arise. The closest thing that we find to an alternating conic - (anti-)ferromagnetic phase boundary is the quadruple point (alternating conic, ferromagnetic splayed, helimagnetic, and ferromagnetic coexistence) (see Figures 25 and 27), where the properties of the phase boundary can be accounted for without explicit reference to the alternating conic (i.e. by treating it as a triple point).

When we add an anti-ferromagnetic $j_3$ to an anti-ferromagnetic $j_1$ (moving from Figure 18 to Figure 22), it initially seems like nothing of interest occurs. The shifting phase boundaries look rather similar to those of Figure 18, at least around the origin. But moving along the helimagnetic - ferrimagnetic splayed phase boundary, we suddenly encounter the alternating conic state (see boxed inset, expanded in Figure 23). The alternating conic fills a triangular region of parameter space. Surprisingly, two sides of the triangular phase boundaries are shared with helimagnetism (note the corner without a triple point). This is rather unusual. It is likely due to one side of the triangle being a first-order transition to helimagnetism.
Figure 22: Chain Lattice Phase Diagram for $j_1 = -1, j_3 = -0.28, j_4 = 0, j'_4 = 0$. First instance of the alternating conic (sharing precedence as the first instance of a non-coplanar state in the chain lattice with the asymmetric conic state in Figure 21). Grey lines indicate range of iterative minimization calculation. Open circles denote helimagnetic phase, light grey circles splayed ferrimagnetic, dark grey anti-ferromagnetic, and crosses alternating conic.

(degeneracy, but not a continuous transition). As such, it does not seem to be related to what we observe in Figures 25 and 27, where the phase boundary of helimagnetism and alternating conics curves around and ends in a quadruple point. That curvature is likely an extension of the curvature observed in other phase boundaries with multiple degrees of freedom, whereas we see a definite discontinuity here.

With a ferromagnetic $j_1$, adding a ferromagnetic $j'_4$ (giving Figure 24) is surprisingly tame for the chain lattice. All that we observe is that the phase boundaries move - they
Figure 23: Chain Lattice Phase Diagram for $j_1 = -1$, $j_3 = -0.28$, $j_4 = 0$, $j'_4 = 0$, focusing in upon range of non-coplanar ground states near $j_2 = -3$ (boxed region in Figure 22).

When we add an anti-ferromagnetic $j_4^{(t)}$ (giving Figure 25), on the other hand, we do see new phenomena. The alternating conic is stabilized for a rather large region of parameter space (at least compared to other regions of non-coplanar states). This is likely due to the strong dependence of the conic sublattice of the alternating conic on the couplings within the sublattice, which will be highly nonlinear (again, recall that the $j_4^{(t)}$ term of the Hamiltonian is proportional to $\cos^2 \alpha (\cos 4\psi - 1)$, which is highly nonlinear in $\cos \alpha$ and $\cos \psi$, themselves extremely nonlinear functions of the couplings). It is also interesting to note that the phase boundary of the alternating conic and ferromagnetic splayed states is linear, at least as a function of $j_2, j'_2$. Given the strongly non-linear nature of the alternating conic, but then this boundary is just a slice of a surface in $j_2, j'_2, j_4^{(t)}$ space, one which almost certainly possesses some curvature.

When we add a ferromagnetic $j_4^{(t)}$ to an anti-ferromagnetic $j_1$ (giving Figure 26), we stabilize a region of asymmetric conic near what would be the ferrimagnetic splayed, helimagnetic, anti-ferromagnetic triple point. This is very similar to Figure 21, except that the shape of the phase is different. The phase diagram still possesses symmetry about the primed-unprimed couplings, and so we should not read too much into the difference in shape.
\[ j_1 = 1, j_3 = 0, j_4 = 0.28, j'_4 = 0 \]

Figure 24: Chain Lattice Phase Diagram for \( j_1 = 1, j_3 = 0, j_4 = 0.28, j'_4 = 0 \). Compare to Figure 17. Black squares denote iterative minimization sampling.

of the asymmetric conic region in these two Figures. It does reflect the way that the two couplings enter into the ground state, but it does not seem to say anything beyond that.

Lastly, for anti-ferromagnetic \( j_1 \) and \( j_4^{(i)} \) (Figure 27), we find that the phase diagram possesses significant regions of both types of conics. The shape of the asymmetric conic phase is now similar to Figure 21, while the shape of the alternating conic phase is similar to Figure 25. This suggests that the phenomena present here are all very familiar. What’s important here, rather, is that the regions of non-coplanar ground states are so much larger than others encountered in the chain lattice. Moreover, this is the only time when both types of conic coexisted for a given slice of \( j_1, j_3, j_4, j'_4 \).
VI. ORIGINS OF NON-COPLANAR STATES

In this section we synthesize the results of the previous two sections, and use them to develop three different perspectives on the origins of non-coplanar states. These perspectives differ both in how prescriptive they are (that is, how much of a "recipe" they give for making non-coplanar states) as well as the circumstances when they are preferable. First, we focus upon a Luttinger-Tisza perspective, which examines how the ground state is created when it is not characterized by a single wave-vector. This is a highly general method, and gives good intuition about how the ground state should behave. Its major disadvantage is that it does not give specific instructions on applying this intuition.

Second, we build upon the concept of encompassing parametrization, introduced in section III C and V B, to find regions of parameter space where a hunt for non-coplanar states
could be fruitful. The exact manner of this hunting is left vague, since it depends upon
the states and lattice considered. The salient feature of this perspective is that we do not
concentrate upon the dimensionality of the state we’re hunting for, but only that it is an en-
ccompassing state. Focusing in upon encompassing parametrizations is a useful perspective,
especially in the chain lattice. When we succeed in finding encompassing parametrizations,
there is a good chance that we will also have found a non-coplanar state.

Finally, we examine the role of degeneracy in constructing ground states, particularly in
the octahedral lattice. In this perspective, two types of degeneracy are considered. First, we
consider situations when many families of unrelated states are degenerate, such as occurs
when planes of spins decouple from each other. Second, we focus upon individual states with
multiple wave-vectors (this shares some elements with our Luttinger-Tisza perspective),

Figure 26: Chain Lattice Phase Diagram for $j_1 = -1, j_3 = 0, j_4 = 0.28, j'_4 = 0$. Dashed lines
denote regions sampled by iterative minimization.
Figure 27: Chain Lattice Phase Diagram for \( j_1 = -1, j_3 = 0, j_4 = -0.28, j'_4 = 0 \). Both types of conic exist for this range.

which we call one-star states. In this perspective, the general goal is to go from finding instances of the first type of degeneracy to building states that display the second type of degeneracy. This is useful in that one-star states are often non-coplanar, so searching for them increases our odds.

A. Luttinger-Tisza Mixtures

Perhaps the easiest “recipe” for the creation of non-coplanar states comes from examining such states in a Luttinger-Tisza perspective. It is already known that a ground state within a Bravais lattice can be characterized by single Luttinger-Tisza eigenmode (at least in the general, non-degenerate case) [27]. But such solutions are not necessarily possible
in the non-Bravais lattice. There are two options for non-Bravais lattices when a single optimal Luttinger-Tisza mode is excluded, the selection depending upon the degeneracy of the optimal Luttinger-Tisza mode. In the case of cuboctahedral states, for example, the states only contain linear combinations of \((1/2, 0, 0)\)-modes, as they are all degenerate in Luttinger-Tisza (in particular, they are degenerate under rotation, and so contain spatial variations in several independent directions). Conversely, the conics necessarily contain two wave-vectors unrelated by symmetry [28].

An intuitive approach to constructing the ground state would be to order the energy of the eigenvectors and find a linear combination of (say) the two lowest energy states that satisfies the unit spin constraint. This works in the case of the cuboctahedral state, but fails in the conic states. The ground state in the case of conics is not necessarily made of the minimum energy states, since there is not always a way to satisfy unit spin constraints without simultaneously introducing much higher energy wave-vectors. Instead, it is preferable to make a combination of state using wave-vectors within a neighborhood of the minimum energy states (we call such states adjacent, as they are near the minimum).

The inclusion of adjacent states, rather than optimal states, gives sufficient freedom to produce the ground state, and highlights some surprising properties of the generalized Luttinger-Tisza method. Consider the conics. They contain some state of arbitrary \(\psi\) (it’s a one-dimensional mode because the lattice is one-dimensional, see equations 5.1.1 through 5.1.4) along with either 0 or 1/2. If the ordered set of eigenmodes is \(\{q_m, \Omega(q_m), 0\}\) (for asymmetric conics) or \(\{q_m, \Omega(q_m), 1/2\}\) (for alternating conics), with \(\Omega(q_m)\) being a neighborhood of the optimal mode \(q_m\) that includes \(\psi\), it is natural to ask why we only require a neighborhood about \(q_m\), rather than 0 or 1/2.

Simply put, the points 0 and 1/2 (and similarly (000), (0, 0, 1/2), (0, 1/2, 1/2), etc. in the octahedral lattice) are special because they lie upon the boundaries of the Brillouin zone. Recall that, when optimizing a function of one variable, it is necessary to check both the critical points \((q_m)\) and the edges (0 and 1/2). This means that 0 and 1/2 might be global or local minima (of the eigenvalues of the Luttinger-Tisza matrix as a function of \(q\)) without having gradient of 0 (as a function of \(q\)). As such, the local minimum Luttinger-Tisza eigenvalue grows linearly with small displacements, rather than quadratically, as it does for displacements about \(q_m\). Hence, modes in the neighborhood of 0 and 1/2 are higher energy than those in the neighborhood of \(q_m\).
B. Phase Transitions and Encompassing Parametrizations

While the Luttinger-Tisza mixture perspective is useful in its generality, it is perhaps of limited applicability. When multiple wave-vectors are required for the ground state, then perspective developed in the previous section is more useful for intuiting the solution than giving an analytic prescription of determining the ground state. To develop a more prescriptive approach, it is preferable to use a different perspective, especially for the chain lattice. In particular, we focus upon the effect of adding a new coupling to an existing set, or to varying the values of couplings already present. These changes will have a tendency to shift phase boundaries. Specifically, we search for triple points, especially when one state is an encompassing parametrization of the other two states (that is, two of the states are special cases of the third - such as ferromagnetism and anti-ferromagnetism with helimagnetism, see section III C). This approach works best if neither of the latter two states is an encompassing parametrization of the other. While this is not a strict requirement, it does simplify the analysis.

So we take a pair of states with a first order transition between them and search for a new, encompassing state (as is the case with the introduction of conics between helimagnetism and splayed states). In moving through parameter space in this manner, we replace the first order transition with a pair of second order transitions. When this occurs, then tuning couplings allows us to continuously transform from one ground state to the other by passing through the encompassing state. Note that while we call the set of couplings where the encompassing state becomes possible a triple point, it is actually a higher dimensional structure (dimensionality is the number of couplings −2).

This introduction of a new state between two others is very specific. It is not the same as the introduction of the cuboctahedral state in the octahedral lattice, since that is not an encompassing parametrization of any other ground state in the lattice. What’s important in this context is the encompassing parametrization hierarchy. By introducing a encompassing parametrization, the spins can vary continuously (as a function of j’s) between two states when previously this was impossible.

But if such a transition was previously impossible, then this immediately implies that the new state has more degrees of freedom than the old one, which (for collinear or coplanar spin states) immediately implies that the encompassing parametrization fills at least one more
dimension of spin space than the encompassed states did. For example, the lowest dimension encompassing parametrization that covers both ferromagnetism and anti-ferromagnetism is helimagnetism \[29\]. Of course, such scaling will break down in three dimensions, when the parametrization becomes more complicated without requiring any more dimensions of spin space (but if the state already fills three dimensions then we have achieved our goal of finding a non-coplanar state). For example, we could parametrize both types of conic by allowing the conic angle of both sublattices to vary as a function of position. Such a state would still be three dimensional, just frustratingly difficult to work with.

The fact that the encompassing parametrization will (in all interesting cases) have a higher dimensionality than its specialized parametrizations makes this approach to finding non-coplanar states especially fruitful. We can effectively bootstrap our way from two unrelated coplanar configurations to a non-coplanar state. It the suffices to find the appropriate combination of couplings to stabilize this new state, a task that (at least in theory) may be accomplished through variational means.

C. Degeneracy and One-Star States

The final known method of tailoring a non-coplanar state occurs in the stabilization of the cuboctahedral state. Consider the family of degenerate states (see section III B) (called the \(120^\circ\) state, see Figure 5):

\[
\begin{bmatrix}
 s_x \\
 s_y \\
 s_z \\
\end{bmatrix} = \frac{1}{\sqrt{2}} \begin{bmatrix}
 0 & (-1)^b(y) & (-1)^c(z) \\
 -(-1)^a(x) & 0 & (-1)^c(z) \\
 -(-1)^a(x) & -(-1)^b(y) & 0 \\
\end{bmatrix} = \frac{1}{\sqrt{2}} \begin{bmatrix}
 0 & 1 & -1 \\
 -1 & 0 & 1 \\
 1 & -1 & 0 \\
\end{bmatrix} \cdot \begin{bmatrix}
 (-1)^a(x) \\
 (-1)^b(y) \\
 (-1)^c(z) \\
\end{bmatrix}.
\]

(6.3.1)

For \(J_1 < 0\), all other couplings 0, this configuration will be the ground state for any selection of \(a, b, c : \mathbb{Z} \rightarrow \mathbb{Z}\) that simultaneously satisfies the unit spin condition. This state can be understood be examining the octahedra on the lattice. The sum of spins on the octahedron necessarily vanishes (in fact, the sum of spins on the triangular faces of the octahedron also necessarily vanishes). Clearly, this state is highly degenerate and includes quite a few non-coplanar states. By introducing a higher order coupling, such as \(J_2\) (or any other combination of couplings which produces a cuboctahedral state) we break the degeneracy of the \(120^\circ\) state on the octahedron and select out a specific ground state, in our case the cuboctahedral state.
The stabilization of a non-coplanar state from a larger family of degenerate states is conceptually the converse of designing non-coplanar states by encompassing parametrizations. In that case, we had a small collection of coplanar states and wanted to boot-strap our way to an encompassing class of non-coplanar states by tuning degeneracies. In this case, we already have a large class of degenerate, non-coplanar states and we want to select out the correct ground state by finding the right combination of couplings to break the degeneracy. Depending upon what states in the lattice are already known, one approach may be preferable to another, but they are of similar levels of applicability.

Analyzing the cuboctahedral states in the Brillouin Zone allows us to understand them in terms of a more general framework, which we call one-star states (see section IIIA). In the cuboctahedral lattices, the ground state is composed entirely out of the wave-vector $00\frac{1}{2}$, including all three permutations and both signs. These modes are clearly degenerate, so any combination of them will also be degenerate. Each sublattice uses four of the six wave-vectors, with no sublattices sharing the same set of wave-vectors. This produces a ground state that spans all directions in spin-space and in dual space, meaning that it shares more similarity to the double twist state than to the conic spirals. This similarity is important, because it contradicts our intuition that any one-star state (that is, all of the wave-vectors come from the same star of symmetry-related $q$’s) is necessarily highly symmetric and therefore conceptually simple. In theory, however, it should be possible to construct non-coplanar states in other lattices using this same method of analysis. For this to work, the number of site classes should be a multiple of the number of spin components, i.e. three. Therefore, this method might be effective in the half-garnet lattice, where there are six site classes associated with the (110) directions.

VII. CONCLUSIONS

A. Improvements on Existing Methodology

The conceptual techniques developed in the determination of non-coplanar states in these lattices are quite powerful, their utility is not limitless. There is a question of degree of rigor we require to accept a ground state as “real.” The techniques we develop here are sufficient if the standard is self-consistency of the ground states produced by different methods (no
small order, as they approach the ground state in very different manners), but not to prove conclusively that observed ground state is the true one. The latter problem is difficult and largely ancillary. To give a rigorous proof that a ground state is the true one requires knowledge of the ground state, which we find in seeking self-consistency. Hence, it is preferable to concentrate upon self-consistency.

More rigorously enforcing self-consistency can be accomplished either by creating new methods to calculate the ground state or by improving existing methods. Improvements probably do not exist for Luttinger-Tisza or for variational optimization (there’s only so much that can be done with the Fourier transformed couplings \( \tilde{J}(q) \) or variational parametrizations of states, a limit that these techniques reach), but there are several promising alternatives to the iterative minimization technique used here. The most common alternative is the method of conjugate gradients, which would improve the speed of the iterative convergence. A newer, and more promising approach would be to use the “difference map” algorithm developed by V. Elser [20], which searches out the solution to a problem with two simultaneous constraints. This algorithm has significant potential for the numerical approximation of ground states, as we can easily find the two constraints - unit spin and the minimum energy no smaller than the Luttinger-Tisza minimum energy.

B. Future Directions and the Prediction of Non-coplanar States

The techniques that we have developed in this paper, both for conceptually understanding ground states and analyzing them quantitatively, are quite powerful. While there are constraints on how these techniques are used, we can overcome them. Their principle limitation is in extrapolating their results to new conditions. This is because we rely upon a library of stable (or potentially stable) states. Such a list is hardly exhaustive, so we may encounter new states when we examine new lattices.

The way to overcome the limitations of using a list of known states is simply to expand the list. As we add more and more states, the likelihood that any new lattice will present unknown ground states is reduced. Moreover, since previously unencountered states are likely to appear near the phase boundaries of known states, we have a good idea of where to look to expand our library of states. As such, we have a general approach to finding non-coplanar states. While this approach does not determine the ground state for an arbitrary
lattice and couplings, it does make such a problem tractable.

VIII. ACKNOWLEDGMENTS

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[21] We are assuming that the electrons are localized, as they are for most materials. This allows us to confine the spins to particular lattice sites. For most materials, this is not a problem, but this is not a good model for the transition metals. In transition metals, the conduction electrons are highly non-localized. The magnetic properties of transition metals fall under a separate class of magnetic order - itinerant magnetism - which we do not consider here.

[22] Note that, when we discuss non-coplanar spin states, we limit our attention to non-trivial cases. When any three (or more) sets of spins have vanishing couplings with each other (either because they cancel or because the couplings are all 0), a large number of degenerate ground states exist. These degenerate states will necessarily include non-coplanar cases, but this is not a set of couplings that generates non-coplanar states. In addition, including anisotropic couplings between spins is explicitly excluded as a mechanism for generating non-coplanar states.

[23] It is worth reiterating that this was the only new state encountered in \((J_1, J_2, J'_2) + J_4\) or \((J_1, J_2, J'_2) + J'_4\), or indeed \((J_1, J_2, J'_2, J_3, J_4, J'_4)\). This absence of any other new states was not for want of trying either, \(J_4\) and \(J'_4\) just do not seem to add anything else of interest.

[24] This is not to say that the conic states are numerical artifacts. They were observed for multiple values of \(J_2\) and \(J_3\) and different sized lattices. Such stability implies that this state must be seriously considered as a valid ground state, even though it is never the non-degenerate ground state for our choice of couplings.

[25] It is very likely that there is another cuboctahedral state, composed of \(\frac{1}{2} + 10\) modes, which would add further weight to our grouping the double-twist state with the cuboctahedral states. This state was not observed in iterative minimization, and we did not find any region of parameter space where Luttinger-Tisza would predict it, so it was not included in the discussion of states.

[26] ferrimagnetic: a state with anti-ferromagnetic order - \(s(z) \propto (-1)^{\hat{n}}\) - where there is still a net moment. This is typically because the spins pointing different directions have different magnitudes, but a similar effect is obtained here by the “splay” of one sublattice.

[27] For a ground state with literally only one optimal mode, the eigenvector is complex-valued.
For a lattice with inversion symmetry, $q$ and $-q$ are degenerate, and there exists a real-valued eigenvector. Although this ground state includes two eigenmodes, this type of degeneracy is rarely important. Instead, it is degeneracy under rotations that are important, since they define different directions of spatial variation.

[28] While the wave-vectors are unrelated by symmetry, they do not necessarily have multiple, independent directions of spatial variation. In the chain lattice, for example, there can only be one direction of spatial variation, so all wave-vectors are parallel.

[29] We assume here that there is no decoupling. In the ferromagnet - anti-ferromagnet transition, another intermediate state would be spins pointing up or down with random order. This state would only be preferable if the ferromagnet and anti-ferromagnet were both degenerate, which implies decoupling. This scenario is better handled in terms of the section VI C.