Spin Waves in a Honeycomb Dimer Antiferromagnet with XY symmetry

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A recent experiment showed that the strongly spin-orbit coupled quantum dimer magnet Yb2Si2O7 observes four distainct quantum phase under an external magnetic field including an XY symmetry broken phase. Such a phase is associated with the Bose–Einstein condensation of magnons. We attempt to reproduce the phase diagram from a model assuming nearest neighbor Heisenberg interactions using spin wave theory. We describe three distinct quantum phases: a quantum dimer magnet, an XY Néel state, and a fully saturated paramagnetic phase. Our analysis is complemented by results from a numerical Linked Cluster expansion and Density Matrix Renormalization Group simulations. We conclude that XY symmetric interactions are unlikely to contain the full physics of the magnet as such a model can only contain three phases.

I. INTRODUCTION

In the past few years, there has been a surge in interest in the ground states of antiferromagnetic materials due to their potential to display unique quantum properties. The properties of these magnets have been shown to be well described by models of localized spins where the breadth of phenomena is explained by the competition between different interactions in the ground state.

One such phenomenon is the existence of XY symmetry broken phases in otherwise XY symmetric models. This occurs in certain materials where the antiferromagnetic interactions are forced to compete with a large polarizing magnetic field. This phenomenon, associated with the $U(1)$ symmetry breaking of magnetic excitations, has been termed the Bose–Einstein condensation of magnons [1]. For it to occur, a very large applied magnetic field takes the place of a chemical potential and the magnetic moments are forced to cant in a plane orthogonal to the XY plane, breaking $U(1)$ symmetry. There are three quantum phases in this paradigm: a phase dominated by local order contiguous with the zero field ground state at low fields such as a quantum dimer magnet (QDM), a fully polarized phase at high fields, and a Bose condensed phase with in plane canting at medium fields.

Recently, an experimental paper has challenged this paradigm by discovering a fourth quantum phase between the symmetry-broken and polarized phases [2]. A model that strongly breaks XY symmetry is not expected to display any phase between the low and high field limits, however an XY symmetric model is expected to only contain the three phases.

In this work, we examine the extent to which the newly discovered Yb2Si2O7 can be described by nearest neighbor Heisenberg or XXZ interactions. We completed our analysis using spin wave theory, a numerical linked expansion, and a DMRG simulation. We hypothesize that Yb2Si2O7 must contain perturbations to the nearest neighbor interactions of the Heisenberg model that weakly break the XY symmetry.

II. MODEL ANTIFERROMAGNET

Reference [2] describes a QDM in the distorted honeycomb lattice magnet Yb2Si2O7. The 3 dimensional material can be pictured as sheets of two dimensional honeycomb lattices stacked on top of one another. From this picture, it is sufficient to consider a single 2 dimensional lattice to understand the ground state phase diagram.

We keep three experimental considerations in mind for our starting point. In reference [2], it was shown that the zero field specific heat data for the quantum dimer magnet is well described by nearest neighbor Heisenberg interactions. The magnet exhibits a fully polarized phase at high fields. It was also shown that the magnet hosts an approximate Goldstone mode with gap less than 0.037 meV. From this we conclude to start with a nearest neighbor Heisenberg model with dimer interactions, shown in Figure 1.

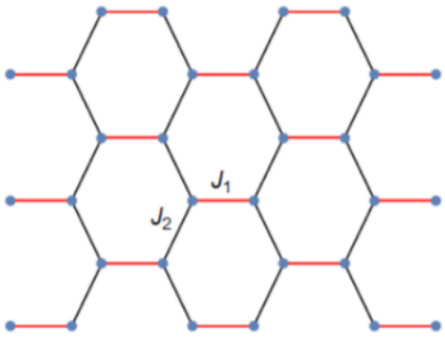

FIG. 1: We assume a model of nearest neighbor Heisenberg interactions. Intradimer coupling is J_1 and interdimer coupling is J_2. Since $J_1 > J_2$, the ground state in zero field is a product state of singlets on the red bonds.

This decision is justified by all three considerations. It makes sense to start from a model that is approximately valid in low fields and exhibits a polarized phase at high fields. Additionally, its XY symmetry makes it a candidate for Bose–Einstein condensation of magnons [1].

In a magnetic field, our Hamiltonian is

$$\mathcal{H} = -h\sum_n S^z_n + J_1 \sum_{\{ij\}\in A} S_i \cdot S_j + J_2 \sum_{\{ij\}\notin A} S_i \cdot S_j \quad (1)$$

where we will set the spin to $S = \frac{1}{2}$ only when comparing to data and otherwise leave S abstract to facilitate with spin

wave perturbations. The term h is proportional to the applied magnetic field and physical values for the gyromagnetic factors will be inserted when necessary for making comparisons to experiment. The set of bonds, A refers to intradimer bonds with the complement of A being interdimer bonds. Finally, the distortion is taken into account by the two parameters J_1 and J_2 where $J_1 > J_2$.

III. SPIN WAVE ANALYSIS

A. Dimer Phase

At low fields, the ground state is in the $S^z_{tot} = 0$ sector. Considering J_2 to be much smaller than J_1, this state is dominated by singlets on the dimerized pairs. The energy bands at zero field can be computed perturbatively in J_2 using a Numerical Linked Cluster Expansion [3]. By identifying the zero field energy gap we can directly extrapolate the band crossing from doping the $S^z = -S$ sector with an external magnetic field, since spin quantum number is respected. We find that

$$h_{c1} = S\left(J_1 - J_2 - J_2^2 + \frac{5}{16}J_2^3\right) \tag{2}$$

Inserting the spin, gyromagnetic factor, and experimentally measured couplings from reference [2] returns $h_{c1} = 0.43$ Telsa which is in strong agreement with the experimentally reported value of 0.4 Tesla.

B. Paramagnetic Phase

As our Hamiltonian commutes with the spin-z operator, at high enough fields, the ground state must simply be a pure polarized state. The zero magnon sector hass $S^z_{tot} = 2SN$. We wish to find at what field the gap to the single magnon sector closes, then. We expand our Hamiltonian in the ladder operators and treat the S^- operators as excitations, equivalent to non interacting hard core bosons on a lattice. This Hamiltonian is exactly solvable via Fourier transformation and two by two matrix diagonalization. We find the energy gap to be

$$\Delta E = Sh - 2S^2\left(J_1 + 2J_2\right) \tag{3}$$

yielding an upper critical field of

$$h_{c2} = 2S\left(J_1 + 2J_2\right) \tag{4}$$

Inserting the spin, gyromagnetic factor, and experimentally measured couplings from reference [2] we find the critical field to be 1.42 Tesla, nearly identical to the reported 1.4 Tesla.

Since, in this phase, magnons are non interacting, all sectors of N and fewer magnons all mix with the ground state at the phase transition. This is in perfect analogy with a conventional Bose-Einstein condensation transition in which particle number conservation is violated in the thermodynamic limit [1].

C. XY phase

Our starting point for the spin wave analysis of the XY phase is the ground state of our Hamiltonian for classical spins. Without loss of generality we can assume the spins order in the x-z plane from U(1) symmetry breaking. Letting ϕ be the angle of each spin from the z axis, using equation 1, the energy is

$$E_c(\phi) = -2NSh\cos(\phi) + N\cos(2\phi)(J_1 + 2J_2) \tag{5}$$

and minimizing ϕ, the spins cant with angle

$$\cos\phi = \frac{h}{2S(J_1 + 2J_2)} = \frac{h}{h_{c2}} \tag{6}$$

where we find the magnetization saturates at the upper critical field.

The details of the spin wave calculation are in direct analogy with reference [4] but are summarized in the following paragraphs. To proceed, we upgrade from classical to quantum spins, but in a frame where the local z axis at each site is canted at an angle ϕ. The rotated spin operators may be mapped to bosonic operators via the Holstein–Primakoff transformation. The resulting Hamiltonian is insoluble, however, may be solved perturbatively in powers of $\frac{1}{S}$. The zeroth order Hamiltonian is conveniently quadratic.

To solve the Hamiltonian to quadratic order, we force the first order term to vanish by setting the canting of the axis to be the classical canting angle of the spins. After Fourier and Bogoliubov transformations, we find the energy bands to be

$$\epsilon(k) = \sqrt{\left(\frac{h_{c2}}{2} \pm |\gamma| S\frac{h^2}{h_{c2}^2}\right)^2 - S^2|\gamma|^2\left(1 - \frac{h^2}{h_{c2}^2}\right)} \tag{7}$$

where γ is the lattice harmonic

$$\gamma(k) = J_1 e^{ik\cdot\delta_1} + J_2\left(e^{ik\cdot\delta_2} + e^{ik\cdot\delta_2}\right) \tag{8}$$

The band structure is shown in Figure 2 for simple hexagonal symmetry. A few things can be observed from this band structure. As expected from the U(1) symmetry breaking, there is a Goldstone mode. Additionally, there are band crossings at the edges of the Brillouin zone that are not linear along all paths, thus are not Dirac points. The band structure is in strong numerical agreement with the results from the neutron scattering experiment in reference [2] when h is close to h_{c2}. Agreement is weaker close to h_{c1}. This should be expected as the classical model from which we are expanding is not aware of the dimer phase for low fields as we have explicitly broken XY symmetry in the canting.

1. Comparison to Neutron Scattering

Reference [2] measures the energy bands using inelastic neutron scattering. Directly comparing the energy bands to

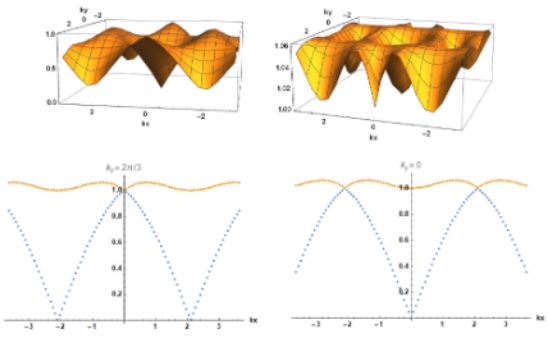

FIG. 2: Energy bands in the XY phase. There is a Golstone mode at the origin and a pair of band crossings at the vertices of the Brillouin zone.

the results of linear spin wave theory within the dome, provides a good agreement between theory and experiment. Additionally, however, the scattering rate is proportional to the dynamic structure constant. This quantity, defined as the space time Fourier transform of a generic two point function,

$$S^{\alpha\beta}(k,\omega) = \sum_{ij} \int_{-\infty}^{\infty} dt \left\langle S_i^{\alpha}(0) S_j^{\beta}(t) \right\rangle e^{i(\omega t - k\cdot(r_i - r_j))} \tag{9}$$

can be calculated directly from linear spin wave theory, where the expectation value is taken with respect to the band that is being scattered off. The result depends on the lattice geometry as well as the hyperbolic trigonometry of the Bogoliubov rotations used to solve the model. In general we found good agreement between linear spin wave theory and experiment.

2. *Anisotropic Coupling*

One potential question to ask is the role of anisotropic couplings on the spin wave analysis. There is only one anisotropy that doesn't break the symmetries of the Hamiltonian so we chose to consider an XXZ coupling of the form

$$\mathcal{H}_{ij} \propto \lambda S_i^z S_j^z + S_i^x S_j^x + S_i^y S_j^y \tag{10}$$

where $\lambda = 1$ reduces to the Heisenberg case. Since the symmetries were not changed, the qualitative aspects of the results were not different. However the critical field

$$h_{c2} = (1+\lambda) S \left(J_1 + 2J_2\right) \tag{11}$$

and energy bands

$$\epsilon(k) = \left(\left(\frac{h_{c2}}{1+\lambda} \pm |\gamma| S \left(1 - \frac{1+\lambda}{2} \left(1 - \frac{h^2}{h_{c2}^2} \right) \right) \right)^2 - \frac{(1+\lambda)^2}{4} S^2 |\gamma|^2 \left(1 - \frac{h^2}{h_{c2}^2} \right) \right)^{\frac{1}{2}} \tag{12}$$

are renormalized. Tuning λ, however, does not significantly improve the agreement with experiment in either the phase transitions or the scattering calculation. Thus we assume that there is little to no XXZ anisotropy within the material.

3. *Nonlinear Spin Waves*

To go beyond zeroth order in the $\frac{1}{S}$ expansion we need to include cubic and quartic terms from the Hamiltonian. This can be done within the Holstein–Primakoff formulation via a Hartree–Fock decoupling of the fluctuating pieces of higher order terms. Ultimately, a complete calculation of the first order correction proved too difficult and likely irrelevant. However, to report one result from the first order correction, the canting angle is renormalized as

$$\cos(\phi) = \frac{h}{h_{c2}} \left(1 + \frac{n - m - \Delta}{S h_{c2}} \right) \tag{13}$$

where n is the average Bose occupation, m is the average hopping parameter and Δ is the average annihilation parameter of the Holstein–Primakoff bosons. These parameters must be calculated self consistently from quadratic and quartic pieces of the Hamiltonian. However, one may note that as h approaches h_{c2}, these parameters should be expected to go to zero as the model fully polarizes and no excitations exist. This would hold true to all orders in the $\frac{1}{S}$ expansion as the Hartree–Fock calculation can only return corrections in terms of these averages. Thus we conclude that the Heisenberg (or XXZ) model only contains three quantum phases and does not capture the full physics of the material.

D. Comparison to DMRG

In order to test the accuracy of the zeroth order $\frac{1}{S}$ expansion, we compare our magnetization results from linear spin wave theory magnetization to a DMRG simulation of 55 electrons for the Heisenberg model. DMRG simulations can converge very quickly for calculating one point functions like magnetization [5]. However, a complete analysis would also study the two point functions in order to compare more deeply with experimental results. The magnetization results are shown in Figure 3.

Since linear spin wave theory assumes a symmetry broken state, it has no predictive validity in either the polarized phase

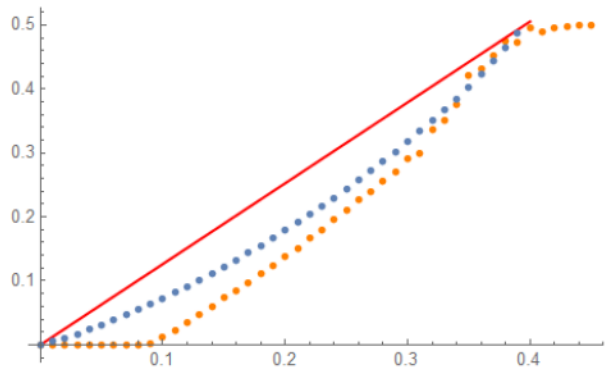

FIG. 3: Magnetization ($\langle S^z \rangle$) vs applied field (h) for the model in equation 1. Orange dots: DMRG data, blue dots: linear spin wave theory, red line: classical spins. The DMRG result shows all three phases predicted. The linear spin wave theory occurs in good agreement with the DMRG result near the upper phase transition but not near the lower one as it misses the dimer phase entirely.

or the dimer phase. However, the DMRG simulation shows excellent agreement with theoretical calculations for the location of both transitions with no magnetization below h_{c1} and full polarization above h_{c2}. Between these two phase transitions linear spin wave theory shows a large range of validity close to h_{c2} and breaking down near h_{c1}. Ultimately, both techniques agree on the location of h_{c2} thus are in general agreement.

IV. CONCLUSION

Through several theoretical techniques, we have described the ground state phase structure of the dimerized quantum antiferromagnet with XY symmetry on a honeycomb lattice in a magnetic field. Since our results only contain three phases, we must conclude that an XY symmetric model cannot capture the full physics of the material. However, the zero field ground state of the magnet is a quantum dimer magnet, thus the XY breaking terms must be small enough not to break this structure. Additionally, since reference [2] observed a Goldstone mode to within 0.037 meV, any breaking of XY symmetry must not induce a gap greater than this value. This is further compounded by the high degree of accuracy in predicting the location of the h_{c1} and h_{c2} phase transitions. If confirmed by future work, the ability of such a small perturbation to dramatically enhance the phase structure of a model would be extremely fascinating and a compelling avenue for further theoretical and experimental work.

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