Waves in the unseen: theory of spin excitations in a quantum spin-nematic

Andrew Smerald$^{1,2}$ and Nic Shannon$^{1,2}$

$^1$Okinawa Institute of Science and Technology, Onna-son, Okinawa 904-0412, Japan
$^2$H. H. Wills Physics Laboratory, University of Bristol, Tyndall Av, BS8-1TL, UK.

(Dated: May 11, 2014)

The idea that a quantum magnet could act like a liquid crystal, breaking spin-rotation symmetry without breaking time-reversal symmetry, holds an abiding fascination. However, the very fact that spin nematic states do not break time-reversal symmetry renders them “invisible” to the most common probes of magnetism — they do not exhibit magnetic Bragg peaks, a static splitting of lines in NMR spectra, or oscillations in $\mu$SR. Nonetheless, as a consequence of breaking spin-rotation symmetry, spin-nematic states do possess a characteristic spectrum of dispersing excitations which could be observed in experiment. With this in mind, we develop a symmetry-based description of long-wavelength excitations in a spin-nematic state, based on an SU(3) generalisation of the quantum non-linear sigma model. We use this field theory to make explicit predictions for inelastic neutron scattering, and argue that the wave-like excitations it predicts could be used to identify the symmetries broken by the otherwise unseen spin-nematic order.

I. INTRODUCTION

The search for quantum spin liquids, magnets which do not order at any temperature, has become one of the cause célèbre of modern physics. Another equally intriguing possibility is that the spins of a quantum magnet do order, but in a way which does not transform like a spin. Such a state would be almost invisible to the usual probes of magnetism, and could therefore appear as a “hidden order”. A concrete example of this is the quantum spin-nematic — a magnetic analogue of a liquid crystal.

Conventional nematic order is associated with the directional order of rod- or disk-like molecules. Spin-nematic order occurs where the fluctuations of a spin mimic a uniaxial molecule, selecting an axis without selecting a direction along it. For example, a system could exhibit fluctuations such that $\langle (S^x)^2 \rangle = \langle (S^y)^2 \rangle \neq \langle (S^z)^2 \rangle$ while maintaining $\langle S \rangle = 0$. Such a phase would break spin-rotation symmetry without breaking time-reversal symmetry. This particular type of spin-nematic state can be described as “ferro-quadrupolar” (FQ), since the fluctuations form a quadrupole moment of $S$ with a common axis on all sites (for an introduction, see [1]). More generally, quadrupole moments tend to select orthogonal axes. Examples of this kind of “antiferroquadrupolar” (AFQ) order are shown in Fig. 1.

There are now good theoretical reasons to believe that spin-nematic order should occur in a range of low-dimensional and frustrated systems. However, because the spin-nematic state does not break time-reversal symmetry, it is “invisible” to the tests commonly used to discern magnetic order, namely the existence of magnetic Bragg peaks in elastic neutron scattering, the splitting of lines in NMR spectra, or through the asymmetry of oscillations in $\mu$SR spectra. Nevertheless, since excitations of the spin-nematic state induce a fluctuating dipole moment, spin-nematic order can, in principle, be detected by dynamic probes of magnetism, such as inelastic neutron scattering or the NMR $1/T_1$ relaxation rate. This hints at an interesting question — if we can’t measure the symmetry breaking in a spin-nematic state directly, can we infer it from the associated excitations?

In this paper, we set aside all questions of the microscopic origin of spin-nematic order, and attempt to say something about what the excitations of a spin-nematic state would look like, assuming it existed. To this end we develop a phenomenological, symmetry-based description of long-wavelength excitations in AFQ spin-nematic states, based on an SU(3) generalisation of the quantum non-linear sigma model, and use it to make concrete predictions for inelastic neutron scattering and the dynamical quadrupolar susceptibility.

We build on a long history of studying spin-nematic states. In one dimension, theoretical studies support the existence of Luttinger liquids with dominant power-law
correlations of spin-quadrupole moments (and in some cases, higher-order spin-multipoles), in frustrated ferromagnetic spin chains, in spin-1/2 ladders with cyclic exchange, and for spin-1 models with biquadratic interactions.

In two dimensions, theoretical studies suggest the existence of a bond-centred, spin-nematic ground state in models of spin-1/2 frustrated ferromagnets on the square, and the triangular lattices, and of a generalised chiral nematic phase on the square lattice. Similarly, two-dimensional, spin-1 models with biquadratic interactions support $T = 0$ nematic order. Entropy-driven nematic order has also been widely studied in the context of the classical Heisenberg model on the Kagome lattice.

In three dimensions, quantum Monte Carlo calculations find evidence for a spin-1 nematic state in the bilinear-biquadratic model, and classical spin-nematic states have been proposed on various frustrated lattices. Weakly-coupled chains in magnetic field also exhibit long-range spin-nematic order.

Recently, the study of spin-nematic order has been energised by the proposal that it might occur in a number of real materials. The unusual magnetic ground state of the spin-1 layered magnetic insulator NiGa$_2$S$_4$ has been discussed in terms of both FQ and AFQ order, and spin-freezing in the presence of FQ correlations, with the bilinear-biquadratic model on a triangular lattice used as a prototype for calculations. Exact diagonalization of the relevant multiple spin-exchange model suggest that the “spin liquid” ground state of thin films of $^3$He might be associated with a 3-sublattice, bond-centred, AFQ phase, [cf. Fig. 2]. Related calculations suggest that a 2-sublattice, bond-centered, AFQ spin-nematic state [cf. Fig. 3] might also be also be realised in the spin-1/2 frustrated Heisenberg model relevant to a family of square lattice vanadates. And finally, magnetisation measurements on the spin-chain system LiCuVO$_4$ show a phase transition close to saturation, which has been interpreted as the onset of a bond-centred, AFQ state.

In parallel with this new work on magnetic insulators, there has been an explosion of interest in electronic-nematic states in itinerant transition-metal compounds, and a resurgence of interest in the study of multipolar “hidden order” phases in rare-earth materials. Since these systems are typically metallic and/or subject to strong spin-orbit coupling, somewhat different considerations apply, and we will not attempt to review either subject here. We concentrate instead on local moments with a high degree of spin-rotational symmetry.

While this brings some simplifications, the microscopic models needed to describe thin films of $^3$He and LiCuVO$_4$ are already very complex, with dominant nearest-neighbour ferromagnetic interactions frustrated by a large number of competing antiferromagnetic exchange pathways. The complexity of these models points to the need for a phenomenological description of AFQ order which makes explicit the physical nature of its excitations, and parameterizes them in terms of the smallest possible number of experimentally-measurable parameters.

In this article, we develop a symmetry-based description of the long-wavelength excitations of 3-sublattice AFQ order on the triangular lattice. Our approach, based on an SU(3) generalisation of the quantum nonlinear sigma model, could be applied equally to the spin-1 magnet NiGa$_2$S$_4$ or to thin films of $^3$He. With minor modifications, the action we derive also offers a description of the 2-sublattice AFQ order proposed to occur in LiCuVO$_4$, and square lattice frustrated ferromagnets. In fact, it can be modified to de-
scribe any system where spin-quadrupoles display short- or long-range, non-collinear order. The only requirement is that the Hamiltonian either has a continuous symmetry (e.g. SU(2) or U(1)), or is close to having a continuous symmetry.

In order to demonstrate the validity of this approach, we show explicitly how our sigma-model like action can be derived from a microscopic model exhibiting 3-sublattice AFQ order, the spin-1 bilinear-biquadratic (BBQ) model on a triangular lattice. At long wavelength, the resulting continuum theory exactly reproduces published results for “flavour wave” analysis of the lattice model. However, the continuum theory is both independent of the “flavour-wave” theory and far more general, and could equally well be parameterised from experiment, or from analysis of a more-complicated microscopic model where the “flavour-wave” approach is not applicable.

Good reviews exist of “flavour wave” techniques for spin-nematic order, but sigma-model approaches have yet to be reviewed, and have so far been restricted to FQ order. We therefore provide a complete and pedagogical account of the steps needed to derive a non-linear sigma-model description of AFQ order.

The fact that different branches of excitation correspond to different rotations of the order parameter, allows us to assign each branch of excitations a clear physical meaning. In the case of 3-sublattice AFQ order, we identify two, physically-distinct types of magnetic excitation — three degenerate branches of “quadrupole waves”, the gapless, linearly-dispersing Goldstone modes of AFQ order, and three degenerate branches of gapped, high-energy “spin-wave” excitations. The spin-wave excitations have a substantial fluctuating dipole moment, and so should be clearly visible in experiment.

Having constructed a general theory for the long-wavelength excitations of the 3-sublattice AFQ spin-nematic states, we are in position to make explicit predictions for inelastic neutron scattering experiments. An example is given in Fig. 4. Observation of these features in experiment would provide strong evidence for spin-nematic order, and a means of distinguishing between different types of spin-nematic states.

We also show predictions for the dynamic quadrupole susceptibility. This may be measurable using, for example, resonant x-ray scattering.

When calculating the experimental response we neglect interaction between the modes. Since we are primarily interested in the universal, long wavelength features it is expected that this is a good approximation. We will return to the role of interactions in a future publication.

We note that any treatment of the 2-particle continuum excitations must take the role of 3- and 4-particle interactions into account if it is to obey the symmetry-constrained sum rules, and for this reason we do not discuss the continuum in this publication.

The remainder of this article is structured as follows. In Section II we develop a theory of long-wavelength excitations in a 3-sublattice AFQ spin-nematic state. In Section III we explore how the excitations of each of these states would manifest themselves in inelastic neutron scattering experiments. In Section IV we consider the dynamical quadrupolar susceptibility. Finally, in Section V we conclude with a summary of results and discussion of their experimental context. Readers who are already expert in sigma models, or simply uninterested in these technical details, are invited to pass directly to Section VI where all key results are summarised. Results for spin-nematic states in 2-sublattice states, in applied magnetic field and predictions for the NMR 1/T1 relaxation rate, will be presented in a separate publication.


II. CONTINUUM THEORY OF 3-SUBLATTICE AFQ ORDER

A. Minimal microscopic model

To keep our continuum theory grounded in microscopic reality, it is helpful to be able to derive it directly from a concrete lattice model, even though the resulting field theory will have far broader applicability. The simplest microscopic model with an AFQ ground state is the spin-1 bilinear-biquadratic (BBQ) model on a triangular lattice\(^5,39\). This model is defined by

\[
H_{\Delta}^{\text{BBQ}} = \sum_{\langle ij \rangle} J_1 S_i \cdot S_j + J_2 (S_i \cdot S_j)^2, \tag{1}
\]

where the sum on \(\langle ij \rangle\) runs over the nearest-neighbour bonds of a triangular lattice.

The mean-field phase diagram for the spin-1 BBQ model on a triangular lattice\(^5,39\), reproduced in Fig. 5 exhibits an extended region of 3-sublattice AFQ order for \(J_2 > 0\), terminating in a point for \(J_1 = J_2\) where the symmetry of the model is enlarged from SU(2) to SU(3)\(^6\). AFQ order is accompanied by a ferroquadrupolar (FQ) phase for \(J_2 < 0\). Conventional ferromagnetic (FM) and 3-sublattice \(120^\circ\) antiferromagnetic (AFM) phases separate these two spin-nematic states. A very similar phase diagram is found in exact diagonalisation\(^5,39\), and the existence of AFQ and FQ phases for closely related BBQ models has been independently confirmed by density matrix renormalisation group calculations\(^6,60\), and quantum Monte Carlo simulations\(^6,61\).

For \(J_2 > J_1 > 0\), \(H_{\Delta}^{\text{BBQ}}\) [Eq. (1)] favours states in which the quadrupole moments on neighbouring sites take on perpendicular directions. The relative simplicity of this model follows from the fact that each spin-1 can form a quadrupole by itself, and the triangular lattice is tripartite, and so naturally supports a 3-sublattice state in which all quadrupoles are orthogonal to one another. The fact that an approximate ground-state wave function can be written in a site-factorized form\(^5\) makes it possible to calculate physically interesting quantities perturbatively from the Hamiltonian using “flavour-wave” theory\(^5,37,41,67-69\) — the SU(3) generalization of the more usual SU(2) spin-wave theory.

The “flavour-wave” approach does not generalise easily to the complicated spin-1/2 models that are relevant to systems such as \(^3\)He and LiCuVO\(_4\). However it provides an important benchmark for the field-theoretical approach developed in this article. In what follows we briefly review some of the features of the spin-1 BBQ model on a triangular lattice, including a useful mean-field parametrisation in terms of spin coherent states, which makes explicit the director nature of the order parameter\(^5,39,59\).

Following \(^39,41\) the Hamiltonian, Eq. (1), can be rewritten in the form,

\[
H_{\Delta}^{\text{BBQ}} = \sum_{\langle ij \rangle} \left( J_1 - \frac{J_2}{2} \right) S_i \cdot S_j + \frac{J_2}{2} Q_i \cdot Q_j + \frac{J_2}{3} S^2 (S + 1)^2, \tag{2}
\]

where the quadrupole operator \(Q\) is given by,

\[
Q = \begin{pmatrix}
Q_{x^2-y^2} & Q_{xy} & Q_{yz} \\
Q_{xy} & Q_{x^2-y^2} & Q_{xz} \\
Q_{yz} & Q_{xz} & Q_{zz}
\end{pmatrix} = \begin{pmatrix}
(S^x)^2 - (S^y)^2 & (S^x)^2 (S^z)^2 & S^x S^z & S^y S^z & S^x S^z + S^y S^z \\
2(S^z)^2 - (S^x)^2 (S^y)^2 & S^x S^y & S^y S^z & S^z S^x & S^x S^z + S^y S^z \\
S^x S^z + S^y S^z & S^y S^z & S^x S^z + S^y S^z
\end{pmatrix}.
\tag{3}
\]

The operator \(Q\) encodes the 5 linearly independent degrees of freedom contained in the traceless, symmetric tensor,

\[
Q^{\alpha\beta} = -\frac{2}{3} S(S+1) \delta^{\alpha\beta} + S^\alpha S^\beta + S^\beta S^\alpha. \tag{4}
\]

It is common practice to parametrise the two magnetic exchange interactions as,

\[
J_1 = \tilde{J} \cos \theta, \quad J_2 = \tilde{J} \sin \theta, \tag{5}
\]
and to plot phase diagrams on a circle, as in Fig. 5. In this article we concentrate on the AFQ phase, bounded by the SU(3) point at $\theta = \pi/4$.

Since spin-nematic states are time-reversal invariant, it is useful to introduce a set of basis states that respect this symmetry. Following $^{5,9,59}$, we consider the following linear superpositions of the usual spin-1 basis states,

$$|x\rangle = \frac{|1\rangle - |\bar{1}\rangle}{\sqrt{2}}, \quad |y\rangle = \frac{|1\rangle + |\bar{1}\rangle}{\sqrt{2}}, \quad |z\rangle = -i|0\rangle.$$  \hspace{1cm} (6)

A general wavefunction for an spin-1 spin at a site $j$ can then be written in the form,

$$|\psi_{\mathbf{j}}\rangle = d^x_j|x\rangle + d^y_j|y\rangle + d^z_j|z\rangle,$$  \hspace{1cm} (7)

where $d^x_j$, $d^y_j$, $d^z_j$ is a 3 vector of complex numbers. It is sometimes convenient to write this out explicitly in real and imaginary components as,

$$d^x_j = u_j + i v_j.$$  \hspace{1cm} (8)

Requiring the wavefunction to be normalised gives the constraint,

$$d^x_j \cdot d^x_j = 1 \quad \text{or} \quad u_j^2 + v_j^2 = 1,$$  \hspace{1cm} (9)

while the overall phase is set by the equation,

$$d^x_j = d^x_j \quad \text{or} \quad u_j \cdot v_j = 0.$$  \hspace{1cm} (10)

Since the phase does not affect any physical observables, one is free to choose this convenient value. As a consequence of Eq. (9) and Eq. (10), there are 4 degrees of freedom associated with each site.

Within the spin-coherent state framework, the operator products appearing in the Hamiltonian, Eq. (2), can be calculated as

$$S^i S^j = |d^i_j \cdot d^j_j|^2 - |d^i_j \cdot d^j_j|^2$$  
$$Q^i Q^j = |d^i_j \cdot d^j_j|^2 + |d^i_j \cdot d^j_j|^2 - \frac{2}{3}.$$  \hspace{1cm} (11)

where the spin value has been set to spin-1. As a result, the Hamiltonian is,

$$\mathcal{H}^\text{BBQ}_\Delta = \sum_{(ij)} J_1 |d^i_j \cdot d^j_j|^2 + (J_2 - J_1) |d^i_j \cdot d^j_j|^2 + J_2.$$  \hspace{1cm} (12)

By minimising this equation, a mean-field, low temperature phase diagram can be mapped out, as shown in Fig. 5.

Purely real or purely imaginary values of $d$ correspond to static nematic states, in which the quadrupole operators take on finite expectation values, but the spin-dipole operators do not. The associated director is parallel to the “director vector”, $\mathbf{d}$. When $\mathbf{d}$ has both real and imaginary components, this corresponds to mixing in a non-zero, static dipole moment, given within the coherent state representation by,

$$S^i_j = 2u^i_j \times v^i_j.$$  \hspace{1cm} (13)

The largest dipole moment occurs when $u$ and $v$ are equal in magnitude (although even in this state there remain quadrupole operators with non-zero expectation values).

The physical observables in the system are expectation values of the dipole and quadrupole operators, $S$ and $Q$. It is useful to write these in the coherent state representation, terms of the vectors $\mathbf{d}$, $\mathbf{u}$ and $\mathbf{v}$, as,

$$
\begin{pmatrix}
S^x \\
S^y \\
S^z \\
Q^{x^2-y^2} \\
Q^{3x^2-2y^2} \\
Q^{xy} \\
Q^{yz} \\
Q^{zx}
\end{pmatrix} = \begin{pmatrix}
2u^x v^y - v^y u^x \\
2u^x v^x - v^x u^x \\
2u^y v^y - v^y u^y \\
(u^x)^2 + (v^y)^2 - (u^y)^2 - (v^x)^2 \\
2(u^x u^y + v^y v^y) \\
2(u^x u^y + v^y v^y) \\
-2(v^x u^y + v^y u^x)
\end{pmatrix}\sqrt{3}$$  \hspace{1cm} (14)

B. Continuum theory at the SU(3) point

1. Why start here?

For $J_1 = J_2$, the symmetry of the spin-1 BBQ model $\mathcal{H}^\text{BBQ}_\Delta$ [Eq. (1)] is enlarged from SU(2) to SU(3). Exactly at this point, the ground states of $\mathcal{H}^\text{BBQ}_\Delta$ include both the 3-sublattice AFQ state and the 3-sublattice “120” Néel antiferromagnet. Moreover, generic 3-sublattice ground states can be constructed from both dipole and quadrupole moments of spins. These physically distinct building blocks are connected by SU(3) rotations that transform $S$ into $Q$, and vice versa — as well as rotating one spin (or quadrupole) configuration into another. These SU(3) rotations are precisely what is needed to describe the long-wavelength excitations of spin-nematic order, and the SU(3) point $(J_1 = J_2)$ therefore provides a very natural starting point for building a continuum theory of 3-sublattice AFQ order.

In the remainder of Section II B below, we construct a sigma model description of long-wavelength excitations of 3-sublattice AFQ order at the SU(3) point. We arrive at a field theory comprising of six identical, linearly-dispersing Goldstone modes, associated with rotations of a triad of $\mathbf{d}$ vectors. Then, in Section II D we explore the consequence of those terms in the Hamiltonian which break this SU(3) symmetry down to the more generic SU(2), introducing these as perturbations about the SU(3) point. This leads to a completely general theory of long-wavelength excitations in a 3-sublattice
AFQ state, comprising three gapless Goldstone modes and three gapped spin-wave excitations.

The structure of this field theory is completely determined by the symmetries of the order parameter, and therefore independent of its derivation. However starting from the SU(3) point of the spin-1 BBQ model allows us to achieve a controlled derivation of a field theory for a 3-sublattice AFQ state from a microscopic model, in a way which keeps the physical nature of its excitations in view. This approach draws inspiration from earlier work on FQ order in one dimension[29,30], and for the 3-sublattice $120^\circ$ AFM state on the triangular lattice[29,31]. In order to keep the text accessible and reasonably self-contained, the necessary steps are described in some detail below.

2. Brief summary of calculation

Before embarking on the calculation, it is useful to briefly summarise the main steps. We start with a single triangular plaquette, which hosts a triad of orthogonal director vectors, and define matrices that describe all the physically relevant, infinitesimal rotations of this triad in the complex vector space of $d$ (ie. those spanning the coset $\text{SU}(3)/H$, where $H$ defines the isotropy subgroup). By the successive action of these matrices, any physical configuration of the three directors can be accessed. Some of these matrices perform global rotations of the director triad, within its complex vector space, and therefore leave the energy invariant. The remainder perform local rotations of the director configuration and thus change the energy of the configuration [see Fig. 6 and Fig. 7]. In analogy with the collinear antiferromagnet[29,30], which undergoes a local ferromagnetic canting, these matrices can be described as a ‘canting’ of the orthogonal director configuration.

The triangular plaquette acts as the basic unit from which to build the triangular lattice [see Fig. 8]. By defining fields at the centre of plaquettes, it is possible to move from a lattice theory written in terms of a Hamiltonian to a continuum theory in terms of a Lagrangian. The fields inherit the properties of the rotation matrices. As in the case of the collinear antiferromagnet[29,30], in moving from the lattice Hamiltonian to the continuum Lagrangian, it is necessary to introduce a dynamical term, which arises from the quantum mechanical overlap of director configurations.

Since we wish to describe the low temperature excitations of the antiferroquadrupolar state, it is reasonable to assume that the directors are approximately orthogonal to one another on short length-scales. In consequence, the Lagrangian can be expanded in terms of the ‘canting’ fields. These can then be eliminated by a Gaussian integral, and the resulting action is an SU(3) symmetric non-linear sigma model.

One way to gain a better physical understanding of the resulting theory is to linearise the fields. This allows a natural division of the modes into those with predominantly quadrupole-fluctuation character and those with spin-fluctuation character. This forms the starting point for calculations of the experimental signatures that could prove the existence of nematic order [see Section 3].

3. Structure of the ground state manifold

The order parameter for the AFQ phase of $H_{\text{BBQ}}$ [Eq. (1)] can be defined on a triangular plaquette containing a triad of directors [cf. Fig. 6]. These directors, which we will label A, B and C, could in principle be located on the sites of the lattice, as is the case here, or on the bonds, as is the case in multiple spin exchange models relevant to thin films of $^3$He.

At the high symmetry $\text{SU}(3)$ point, $J_1 = J_2 = J$, the Hamiltonian, $H_{\text{BBQ}}$ [Eq. (14)], simplifies to,

$$H_{\text{SU}(3)} = J \left( |d_A \cdot \bar{d}_B|^2 + |d_B \cdot \bar{d}_C|^2 + |d_C \cdot \bar{d}_A|^2 \right) + 3J. \quad (15)$$

This can be minimised by requiring $d_i \cdot \bar{d}_j = 0$ on every bond, resulting in a 3-sublattice order in which neighbouring $d$ vectors are orthogonal. There is no requirement that $d$ should be real (or imaginary) and therefore the ground state manifold includes both quadrupolar, dipolar and mixed phases.

One choice for the ground state of such a system is,

$$d_A^{gs} = (1, 0, 0), \quad d_B^{gs} = (0, 1, 0), \quad d_C^{gs} = (0, 0, 1). \quad (16)$$

This corresponds to an AFQ state in which the three directors lie along the principle axes, $(x, y, z)$, and is illustrated in Fig. 6.

The Hamiltonian, Eq. (15), is invariant under the global rotation $d \rightarrow Ud$, provided that $U^{-1} = U^\dagger$, mak-
cylinders) on the three sites of the plaquette are combined at the plaquette centre. (a) shows the orthogonal ground state given in Eq. (16). (b) shows the result of acting on this particular ground state with plaquette [see Fig. 6], showing the action of global rotations.

FIG. 7: (Color online). Real component of the complex director configurations for antiferroquadrupolar (AFQ) order on a triangular plaquette [see Fig. 8], showing the action of global rotations U(\phi) and local rotations associated with the canting fields I. Directors (red cylinders) on the three sites of the plaquette are combined at the plaquette centre. (a) shows the orthogonal ground state given in Eq. (16). (b) shows the result of acting on this particular ground state with D_{\Delta}(\phi_1, 0, \ldots) [see Eq. (20)]. This performs a global rotation of the directors around the z-axis, and a different orthogonal ground state is generated. (c) shows the result of acting with D_{\Delta}(0, \ldots, l_{xyz}^2, 0, \ldots), which is seen to rotate directors orientated along the x- and y-axes in opposite directions around the z-axis. In consequence the angle between the directors changes, and this costs energy according to the Hamiltonian \langle H_{\Delta} \rangle [Eq. (12)].

... (16). The global rotations of the order parameter can be split into two categories. In order to see this, it is useful to use the shorthand notation U_1 = U(\phi_1, 0, 0, 0, 0, 0) and similarly for U_2, \ldots, U_6. The matrices U_1, U_2 and U_3 perform rotations of the directors which are real in the sense that, if \mathbf{d} is real [as in Eq. (16)], it will remain so under these transformations. Applied to the AFQ ground state, they act only to rotate the quadrupole moments. However the matrices U_4, U_5 and U_6 transform a real \mathbf{d} vector into a complex one in such a way as to mix a dipolar component into the AFQ ground state. We will return to this point below when classifying spin excitations.

C. Canting of a plaquette

Our ultimate aim is to describe the long-wavelength, director-wave fluctuations about the ‘invisible’ AFQ spin-nematic ground state — the ‘waves in the unseen’. This involves canting of the director triad out of the orthogonal ground state.

A necessary first step, is to construct a matrix, D_{\Delta}, that can be used to access any configuration of three \mathbf{d}-vectors on a triangular plaquette. In order to do this, it is useful to introduce a second set of generators,

\[
\mu_1 = \begin{pmatrix} 0 & -i & 0 \\ -i & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad \mu_2 = \begin{pmatrix} 0 & 0 & -i \\ 0 & 0 & 0 \\ -i & 0 & 0 \end{pmatrix},
\]

\[
\mu_3 = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & -i \\ 0 & -i & 0 \end{pmatrix}, \quad \mu_4 = \begin{pmatrix} 0 & 1 & 0 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \end{pmatrix},
\]

\[
\mu_5 = \begin{pmatrix} 0 & 0 & -1 \\ 0 & -1 & 0 \\ 1 & 0 & 0 \end{pmatrix}, \quad \mu_6 = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & -1 & 0 \end{pmatrix}.
\]

When these act on a triad of \mathbf{d} vectors [see Fig. 7c], they change the angles between the vectors, thus changing the energy, according to Eq. (15) [see Fig. 7c]. Any configuration of the three \mathbf{d} vectors can be accessed from Eq. (16).
using,

\[ D_\Delta(\phi, l) = \exp \left[ i \sum_{p=1}^{6} \lambda_p \phi_p + i \mu_1 l_1^2 + i \mu_2 l_1^4 + i \mu_3 l_1^6 + i \mu_4 l_2^2 + i \mu_5 l_2^4 + i \mu_6 l_2^6 \right], \quad (20) \]

where the vector \( l \) is defined by,

\[ l = \begin{pmatrix} l_x^2 \\ l_y^2 \end{pmatrix} = \begin{pmatrix} l_x^2 + i l_y^2 \\ l_x^2 + i l_y^2 \end{pmatrix}. \quad (21) \]

This notation may appear unnatural at first sight, but will prove convenient for calculation. A completely general configuration of the three \( d \) vectors is thus given by,

\[ d_A = D_\Delta \cdot \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix}, \quad d_B = D_\Delta \cdot \begin{pmatrix} 0 \\ 1 \\ 0 \end{pmatrix}, \quad d_C = D_\Delta \cdot \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix} \quad (22) \]

We now make the assumption that the system has at least short-range order, and thus expand for small canting fields \( l \). Retaining fields up to \( \mathcal{O}(l) \),

\[ d_A = U \cdot \begin{pmatrix} 1 \\ l_x^2 \end{pmatrix}, \quad d_B = U \cdot \begin{pmatrix} l_x^2 \\ 1 \end{pmatrix}, \quad d_C = U \cdot \begin{pmatrix} l_x^2 \\ 1 \end{pmatrix}, \quad (23) \]

and it follows that the length and phase constraints of Eq. (9) and Eq. (10) hold to \( \mathcal{O}(l^2) \).

The eventual aim is to eliminate the canting fields \( l \) from the partition function by integration. What will remain is a theory describing the dynamics of the order parameter matrix, \( U \), in terms of the variables \( \phi \).

1. Continuum limit

We now consider how to pass from a lattice theory to a continuum theory of the AFQ state. The lattice can be partitioned into clusters based on triangular plaquettes, as shown in Fig. 8. The director fields are defined at the centre of these clusters, and the physical location of the directors is taken into account by performing a gradient expansion. The continuum limit involves the assumption that physically interesting variation takes place on a lengthscale much larger than the lattice constant, \( a \), and so gradients within the plaquette are small.

One of the requirements of a continuum field theory is that it should describe the dynamics of both the broken symmetry state and the nearby paramagnetic region, in which the order parameter is assumed to be locally robust but slowly varying over macroscopic length scales. It is therefore necessary to allow the fields to fluctuate in space and time,

\[ d_A(\mathbf{r}, \tau) = U(\mathbf{r}, \tau) \begin{pmatrix} 1 \\ \bar{l}_x(\mathbf{r}, \tau) \\ \bar{l}_y(\mathbf{r}, \tau) \end{pmatrix} + \mathcal{O}(l^2), \]

\[ d_B(\mathbf{r}, \tau) = U(\mathbf{r}, \tau) \begin{pmatrix} \bar{l}_x(\mathbf{r}, \tau) \\ 1 \\ \bar{l}_y(\mathbf{r}, \tau) \end{pmatrix} + \mathcal{O}(l^2), \]

\[ d_C(\mathbf{r}, \tau) = U(\mathbf{r}, \tau) \begin{pmatrix} \bar{l}_y(\mathbf{r}, \tau) \\ \bar{l}_x(\mathbf{r}, \tau) \\ 1 \end{pmatrix} + \mathcal{O}(l^2). \quad (24) \]

A useful parametrisation of the matrix \( U \) is,

\[ U(r, \tau) = \begin{pmatrix} n_A^x(\mathbf{r}, \tau) & n_B^x(\mathbf{r}, \tau) & n_C^x(\mathbf{r}, \tau) \\ n_A^y(\mathbf{r}, \tau) & n_B^y(\mathbf{r}, \tau) & n_C^y(\mathbf{r}, \tau) \\ n_A^z(\mathbf{r}, \tau) & n_B^z(\mathbf{r}, \tau) & n_C^z(\mathbf{r}, \tau) \end{pmatrix}, \quad (25) \]

where the complex fields \( n_i(\mathbf{r}, \tau) \), with \( i = \{A, B, C\} \), inherit the length and phase constraints of the \( d \) vectors (Eq. 9 and Eq. 10),

\[ n_i \cdot \bar{n}_i = 1, \quad n_i^2 = n_i^2 = 0, \quad (26) \]

and are also required to be orthogonal to one another according to,

\[ n_i \cdot \bar{n}_j = 0, \quad i \neq j. \quad (27) \]

The apparent 18 degrees of freedom of the \( n_i \) fields is reduced to 6 by the 12 constraints, as expected. The reason that the parametrisation in terms of \( n_i(\mathbf{r}, \tau) \), is useful is that these fields are mutually orthogonal, of unit length, and of fixed phase, and can therefore be interpreted as a ground-state director configuration. In consequence there are two equivalent formulations of the field theory: in terms of the rotation matrix \( U(\mathbf{r}, \tau) \); or in terms of the fields \( n_i(\mathbf{r}, \tau) \). We will make use of both in what follows.
Differentiating the constraints, Eq. (26) and Eq. (27), leads to the relations,
\[ n_i \cdot \partial_\lambda \bar{n}_i = -\bar{n}_i \partial_\lambda n_i, \quad n_i \cdot \partial_\lambda \bar{n}_j = \bar{n}_j \partial_\lambda n_i, \quad i \neq j, \]
where the partial derivative \( \partial_\lambda \) can be with respect to any space-time variable. These relations prove very useful for simplifying subsequent expressions.

The partition function can be written in terms of a functional integral over all director configurations,
\[ Z_{\Delta}^{SU(3)} = \int \mathcal{D}[d] e^{-S_{\Delta}^{SU(3)}[d]}, \]
where \( S_{\Delta}^{SU(3)}[d] \) is the Euclidean action and the integration measure \( \mathcal{D}[d] \) includes the delta function constraints on the length and phase of the director. The action can be split into Hamiltonian and kinetic terms,
\[ S_{\Delta}^{SU(3)} = S_{\text{kin}} + S_{\mathcal{H}[SU(3)]}, \]
where \( S_{\text{kin}} \) is a dynamic, geometric-phase term and \( S_{\mathcal{H}[SU(3)]} \) accounts for the energy cost of static director configurations at the SU(3) point.

2. The Hamiltonian term

The energy cost of a particular static configuration of directors is given by Eq. (12). In principle, the Hamiltonian term in the action, \( S_{\mathcal{H}} \), takes into account all static configurations of directors. However, we make the approximation that only those with a slow spatial variation are important.

The Hamiltonian term is given by,
\[ S_{\mathcal{H}[SU(3)]} = \int_0^\beta d\tau H_{SU(3)} \]
\[ = \frac{2}{3\sqrt{3}a^2} \int_0^\beta d\tau \int d^2r H_{SU(3),\text{clus}}, \]
where \( H_{SU(3),\text{clus}} \) refers to the Hamiltonian for a single cluster, and the numerical prefactor is related to the area of the cluster.

The gradient expansion of the fields in terms of the small parameter \( a \) is given by,
\[ d_j(r + \epsilon_i, \tau) = d_j(r, \tau) + a(\epsilon_i \cdot \nabla)d_j(r, \tau) + \frac{a^2}{2!}(\epsilon_i \cdot \nabla)^2d_j(r, \tau) + \mathcal{O}(a^3), \]
where \( \epsilon_i \) is the vector connecting the centre of the cluster to the lattice sites within it (cf. Fig. 5).

Expanding the Hamiltonian to second order in the lattice parameter \( a \) gives,
\[ H_{SU(3),\text{clus}} \approx 3J \left[ |d_A(r, \tau) \cdot d_B(r, \tau)|^2 + |d_B(r, \tau) \cdot d_C(r, \tau)|^2 + |d_C(r, \tau) \cdot d_A(r, \tau)|^2 \right] + \frac{3Ja^2}{2} \sum_{\lambda=x,y} \left[ |d_A \cdot \partial_\lambda d_B|^2 + |d_B \cdot \partial_\lambda d_C|^2 + |d_C \cdot \partial_\lambda d_A|^2 \right]. \]

The first term in this expression vanishes if the system is an AFQ ground state. Fluctuations about this can be expanded in terms of the canting field \( \mathbf{l} \) using,
\[ d_A(r, \tau) \cdot d_B(r, \tau) \approx 2l^2(r, \tau) \]
\[ d_B(r, \tau) \cdot d_C(r, \tau) \approx 2l^3(r, \tau) \]
\[ d_C(r, \tau) \cdot d_A(r, \tau) \approx 2l^3(r, \tau). \]

Since the gradient terms are already \( \mathcal{O}(a^2) \), the fields \( d(r, \tau) \) can be replaced by the orthogonal fields \( \mathbf{n}(r, \tau) \), giving the Hamiltonian,
\[ H_{SU(3),\text{clus}} \approx 12Ja^2 \mathbf{l} \cdot \mathbf{I} \]
\[ + \frac{3Ja^2}{2} \sum_{\lambda=x,y} \left[ |\mathbf{n}_A \cdot \partial_\lambda \mathbf{n}_B|^2 + |\mathbf{n}_B \cdot \partial_\lambda \mathbf{n}_C|^2 + |\mathbf{n}_C \cdot \partial_\lambda \mathbf{n}_A|^2 \right]. \]

3. The kinetic term

The action describing long wave-length fluctuations of the AFQ state also contains a kinetic energy term. This is quantum-mechanical in origin, and a consequence of the overcompleteness of the coherent states used to represent spin configurations. At a semiclassical level it describes the rotational motion of the directors, and can therefore be interpreted as a geometrical phase. For a more detailed explanation we refer the interested reader to the chapters on spin path integrals in [72,73].

The contribution of the kinetic term to the action is,
\[ S_{\text{kin}} \approx \int_0^\beta d\tau \frac{2}{3\sqrt{3}a^2} \int d^2r \sum_i \partial_i \cdot \partial_\tau d_i, \]
where spatial gradient terms have been ignored. To first order in the canting field \( \mathbf{l} \),
\[ \sum_i \partial_i \cdot \partial_\tau d_i \approx \text{Tr}[U^\dagger \cdot \partial_\tau U] + 2 \left[ s \cdot (\mathbf{l} - s) \cdot \mathbf{l} \right], \]
where the complex field \( s(r, \tau) \) is defined as,
\[ s = \begin{pmatrix} (U^\dagger \cdot \partial_\tau U)_{21} \\ (U^\dagger \cdot \partial_\tau U)_{32} \\ (U^\dagger \cdot \partial_\tau U)_{13} \end{pmatrix} = \begin{pmatrix} \mathbf{n}_B \cdot \partial_\lambda \mathbf{n}_A \\ \mathbf{n}_C \cdot \partial_\lambda \mathbf{n}_B \\ \mathbf{n}_A \cdot \partial_\lambda \mathbf{n}_C \end{pmatrix}. \]

The kinetic term gives an imaginary contribution to the Euclidean Lagrangian. Derivatives of the field \( \mathbf{l} \) vanish, since they are total derivatives and can therefore be converted to a vanishing surface integral.
4. Integrating out fluctuations

Having derived an action for long-wavelength fluctuations of the AFQ state, the task which remains is to eliminate the canting fields \( I(r, \tau) \), so as to arrive at an action written entirely in terms of the order parameter \( \mathbf{n}(r, \tau) \). Taking into account both potential and kinetic energy terms in the Hamiltonian, we start from the partition function,

\[
Z_{\Delta}^{SU(3)} \propto \int \prod_{i \neq j} Dn_i D\bar{n}_i D\bar{D} \delta(n_i \cdot \bar{n}_i - 1) \delta(n_i^2 - \bar{n}_i^2) e^{-S_{\Delta}^{SU(3)}[n_A, \bar{n}_A, n_B, \bar{n}_B, n_C, \bar{n}_C, I, l]},
\]

where the action,

\[
S_{\Delta}^{SU(3)}[n_A, \bar{n}_A, n_B, \bar{n}_B, n_C, \bar{n}_C, I, l] = \int_0^\beta d\tau \int d^2r \frac{2}{3\sqrt{3}d^2} \int d^2r L_{\Delta}^{SU(3)},
\]

is written in terms of the Lagrangian,

\[
L_{\Delta}^{SU(3)} \approx Tr[U^\dagger \partial_\tau U] + 2 [s \cdot 1 - \bar{s} \cdot 1] + 12 J \lambda \bar{l} + \frac{3Ja^2}{2} \sum_{\lambda=x,y} \left[ |\bar{n}_\lambda \cdot \partial_\tau n_\lambda|^2 + |n_\lambda \cdot \partial_\tau n_\lambda|^2 + |\bar{n}_\lambda \cdot \partial_\tau n_\lambda|^2 \right].
\]

(41)

The canting fields \( I \) and \( \bar{l} \) enter the Lagrangian at a quadratic level and can therefore be eliminated by a Gaussian integral, or, equivalently, using the steepest-descent approximation. This process is slightly simpler if the two fields are decoupled by the linear transformation,

\[
I = I_1 + iI_2, \quad \bar{l} = l_1 - i l_2,
\]

(42)

where \( I_1 \) and \( I_2 \) are real. Taking functional derivatives with respect to these fields gives,

\[
\frac{\delta L_{\Delta}^{SU(3)}}{\delta I_1} \approx 2(s - \bar{s}) + 24 J I_1 \approx 0
\]

\[
\frac{\delta L_{\Delta}^{SU(3)}}{\delta I_2} \approx 2i(s + \bar{s}) + 24 J I_2 \approx 0
\]

and these equations are resolved as,

\[
I_1 \approx -\frac{1}{12J}(s - \bar{s})
\]

\[
I_2 \approx -\frac{i}{12J}(s + \bar{s}).
\]

(43)

At this point it is helpful to introduce a ‘director stiffness’,

\[
\rho_d = Ja^2,
\]

(45)

describing the energy cost of twisting the order parameter, and the generalised susceptibility,

\[
\chi_\perp = \frac{2}{9J},
\]

(46)

associated with fluctuations of the canting field \( I \).

Substituting the canting fields, Eq. (44), into the Lagrangian, Eq. (41), and using Eq. (38) and Eq. (25) to re-express this in terms of the fields \( \mathbf{n} \), we arrive at

\[
Z_{\Delta}^{SU(3)}[n_A, \bar{n}_A, n_B, \bar{n}_B, n_C, \bar{n}_C] \approx \frac{1}{\sqrt{3\pi^2}} \int_0^\beta d\tau \int d^2r \left\{ 2 \sum \bar{n}_i \cdot \partial_\tau n_i + \chi_\perp \left[ |\bar{n}_A \cdot \partial_\tau n_B|^2 + |\bar{n}_B \cdot \partial_\tau n_C|^2 + |n_C \cdot \partial_\tau n_A|^2 \right] + \rho_d \sum_{\lambda=x,y} \left[ |\bar{n}_\lambda \cdot \partial_\tau n_\lambda|^2 + |n_\lambda \cdot \partial_\tau n_\lambda|^2 + |\bar{n}_\lambda \cdot \partial_\tau n_\lambda|^2 \right] \right\},
\]

(47)

with associated partition function,

\[
Z_{\Delta}^{SU(3)} \propto \int \prod_{i \neq j} Dn_i D\bar{n}_i \delta(n_i \cdot \bar{n}_i - 1) \delta(n_i^2 - \bar{n}_i^2) e^{-S_{\Delta}^{SU(3)}[n_A, \bar{n}_A, n_B, \bar{n}_B, n_C, \bar{n}_C]},
\]

(48)

where the canting fields have been eliminated at a Gaussian level.

Equivalently, Eq. (37) can be used to write the action, Eq. (17), in terms of the unitary matrices, \( \mathbf{U}(r, \tau) \), as,

\[
S_{\Delta}^{SU(3)}[\mathbf{U}] = \frac{1}{2 \sqrt{3\pi^2}} \int_0^\beta d\tau \int d^2r \left\{ \frac{4}{3} Tr[U^\dagger \partial_\tau U] + \chi_\perp \left[ Tr[\partial_\tau U^\dagger \partial_\tau U] - \sum_m |[U^\dagger \partial_\tau U]_{mmm}|^2 \right] + \rho_d \sum_{\lambda=x,y} \left[ Tr[\partial_\tau U^\dagger \partial_\tau U] - \sum_m |[U^\dagger \partial_\tau U]_{mmm}|^2 \right] \right\},
\]

(49)

where \( m = \{1, 2, 3\} \) labels matrix elements. This formulation of the action is further removed from the physical state than Eq. (17), but makes explicit the \( SU(3) \) symmetry of the Hamiltonian.

5. Linearising the order parameter fields

The physical nature of the excitations of the AFQ state — and in particular the division into quadrupole-wave and spin-wave modes — is easier to understand once the action describing them has been linearized. This can be achieved by expanding fluctuations about the AFQ ground state to leading order in \( \phi \). We will consider in detail the interaction of the \( \phi \) fields in a future publication.\[\text{publication}\]
After linearization, the unitary matrix field \( U(r, \tau) \) [Eq. 15] is approximated by,
\[
U(r, \tau) \approx \begin{pmatrix}
1 & \phi_1 + i \phi_4 & -\phi_2 + i \phi_5 \\
-\phi_1 + i \phi_4 & 1 & \phi_3 + i \phi_6 \\
\phi_2 + i \phi_5 & -\phi_3 + i \phi_6 & 1
\end{pmatrix},
\] (50)

where the angular variables \( \phi_p = \phi_p(r, \tau) \) fluctuate in both space and time. It follows from Eq. 25 that the fields \( n_i(r, \tau) \) are given by,
\[
n_A(r, \tau) \approx \begin{pmatrix}
1 & -\phi_1 + i \phi_4 \\
\phi_1 + i \phi_4 & 1 \\
-\phi_4 & 0
\end{pmatrix},
n_B(r, \tau) \approx \begin{pmatrix}
\phi_3 + i \phi_6 \\
\phi_4 & 1 \\
0 & 0
\end{pmatrix},
n_C(r, \tau) \approx \begin{pmatrix}
-\phi_2 + i \phi_5 \\
-\phi_3 + i \phi_6 & 1
\end{pmatrix},
\] (51)

making explicit that the fields \( \phi_p(r, \tau) \) have a simple interpretation in terms of small, local angles of rotation away from the direction of spontaneous symmetry breaking.

Eq. 14 can now be used to reconstruct the fluctuating dipolar and quadrupolar moments on each sublattice. To leading order in \( \phi_p(r, \tau) \), these can be written as,
\[
S_A \approx 2 \begin{pmatrix}
0 \\
-\phi_5 \\
\phi_4
\end{pmatrix}, \quad S_B \approx 2 \begin{pmatrix}
\phi_6 \\
0 \\
-\phi_4
\end{pmatrix}, \quad S_C \approx 2 \begin{pmatrix}
-\phi_6 \\
\phi_5 \\
0
\end{pmatrix},
\] (52)

and
\[
Q_A \approx \begin{pmatrix}
-1/\sqrt{3} \\
-2\phi_1 \\
0
\end{pmatrix}, \quad Q_B \approx \begin{pmatrix}
1/\sqrt{3} \\
2\phi_1 \\
-2\phi_3
\end{pmatrix}, \quad Q_C \approx \begin{pmatrix}
0 \\
-2/\sqrt{3} \\
2\phi_1
\end{pmatrix}.
\] (53)

This shows that the fields \( \phi_1, \phi_2 \) and \( \phi_3 \) are primarily associated with fluctuations of the quadrupole moments, and so justifies the name “quadrupole-waves”. Since the fields \( \phi_4, \phi_5 \) and \( \phi_6 \) are primarily associated with transverse fluctuations of the dipole moments, we refer to them as “spin-waves”. In Section III, we extend this analysis to also include time derivatives of the \( \phi \) fields. The supplemental material contains animations showing the nature of the quadrupole-wave and spin-wave modes that follow from Eq. (53).

Linearizing the action \( S_{\Delta}^{U(3)}[U] \) [Eq. 19] also allows us to eliminate the delta function constraints from the partition function \( Z_{\Delta}^{SU(3)} \) [Eq. 43], to give
\[
Z_{\Delta}^{SU(3)} \propto \int \mathcal{D}\phi e^{-S_{\Delta}^{SU(3)}[\phi]},
\] (54)

where the linearised action is,
\[
S_{\Delta}^{SU(3)}[\phi] \approx \frac{1}{\sqrt{3} \alpha} \int_0^\beta d\tau \int d^2r \sum_{p=1}^6 \left[ \chi_1 (\partial_\tau \phi_p)^2 + \rho_0 \sum_{\lambda=x,y} (\partial_\lambda \phi_p)^2 \right].
\] (55)

At this level of approximation the equations of motion for each field are independent of one another and given by,
\[
\chi_1 (\partial_\tau^2 \phi_p + \rho_0 (\partial_\phi^2 + \rho_0 \partial_\phi^2 \phi_p = 0.
\] (56)

These can be solved by the ansatz,
\[
\phi_p = A_p e^{i \mathbf{r} \cdot \mathbf{q} + \omega \tau},
\] (57)

and in consequence the dispersion (shown in Fig. 11) is,
\[
\omega_q = \sqrt{\frac{\rho_0}{\chi_1}} |\mathbf{q}| = v |\mathbf{q}|,
\] (58)

with the director-wave velocity,
\[
v = \sqrt{\frac{\rho_0}{\chi_1}} = \frac{3J_0}{\sqrt{2}}.
\] (59)

Here the vector \( \mathbf{q} \) measures the distance in reciprocal space from the centre of the magnetic Brillouin zone (mbz), which is centred on the \( \mathbf{k} \) point, \( \mathbf{k}_\parallel = (4\pi/3, 0) \), as shown in Fig. 10.

Thus, at the \( SU(3) \) point, there are 6 gapless excitations, which disperse linearly with the same velocity, regardless of whether they have spin-wave or quadrupole-wave character. This reflects the large ground state manifold at the \( SU(3) \) point, which consists of all 3-sublattice orthogonal arrangements of the \( d \) vectors, and therefore includes both the AFQ and AFM states [cf. Fig. 5].

In Section III we show that, as \( J_2 \) is increased and dipolar order becomes energetically unfavourable, only three linearly dispersing modes remain — the quadrupole-wave modes, which are the Goldstone modes of AFQ order.

We note that Tsunetsugu and Arikawa have previously determined the dispersion of Eq. 10 in the AFQ phase using a linearised “flavour-wave” theory. At the high symmetry \( SU(3) \) point they find,
\[
\omega_\kappa = 3J \sqrt{1 - |\gamma_\kappa|^2},
\] (60)

where,
\[
\gamma_\kappa = \frac{1}{3} \left( e^{ik \cdot a} + 2 e^{-ik \cdot a} \cos \frac{3k \cdot a}{2} \right).
\] (61)

As \( k \to 0 \) the limiting value of Eq. 60 is \( \omega_\kappa \approx v |\kappa| \), where the velocity \( v = 3J_0/\sqrt{2} \) is identical to the one predicted by the field theory [Eq. 59].
FIG. 9: (Color online). The dispersion of magnetic excitations at the SU(3) point. (a) prediction of the continuum field theory $S_{\Delta}^{\rm SU(3)}$ [Eq. (55)]. (b) prediction of the spin-1 bilinear-biquadratic (BBQ) model on a triangular lattice, $H_{\Delta}^\text{BBQ}$ [Eq. (1)] for $J_1=J_2$. Approximating the ordering vector $\mathbf{k} = \mathbf{k}_M (q = 0)$, the continuum theory and the lattice theory match exactly. At this high-symmetry point there is a 6-fold degenerate branch of linearly-dispersive, gapless excitations. These can be split into 3 modes that primarily describe fluctuations of quadrupole moments (quadrupole waves) and 3 that primarily describe fluctuations of dynamically generated dipole moments (spin waves). These 3 spin-wave fields become gapped on entering the antiferroquadrupolar (AFQ) phase bordering the SU(3) point.

D. Continuum theory away from the SU(3) point

1. Symmetry breaking terms

The SU(3) point of $H_{\Delta}^\text{BBQ}$ [Eq. (1)], $J_2 = J_1 = J$, has an artificially high symmetry. For $J_2 > J_1$ the symmetry of $H_{\Delta}^\text{BBQ}$ is reduced to SU(2), with important implications for the excitations of the AFQ state. In what follows, we construct a continuum field theory for the AFQ phase by perturbing away from the SU(3) point. The most significant change, required for the stability of AFQ order, is the opening of a gap to the 3 spin-wave modes.

$H_{\Delta}^\text{BBQ}$ [Eq. (1)] can be written

$$H_{\Delta}^\text{BBQ} = H_{\text{SU}(3)} + \Delta H_{\text{SU}(2)}$$

where

$$\Delta H_{\text{SU}(2)} = (J_2 - J_1) \sum_{(j)} |d_i \cdot d_j|^2,$$

and $H_{\text{SU}(3)}$ is defined by Eq. (13). In order to develop a perturbative expansion around the high-symmetry SU(3) point, we make the assumption that $J_2 - J_1 \ll J_1, J_2$. This assumption breaks down for $\theta \to \pi/2$, and places a limit on the range of wavelengths for which the sigma-model description developed in this Section is a valid description of $H_{\Delta}^\text{BBQ}$ [Eq. (1)].

The kinetic term in the action $S_{\text{kin}}$ [Eq. (31)] is unchanged since it is a property of the coherent state representation of the spin states, not of the Hamiltonian. The change to the Hamiltonian term in the action $S_{\Delta}$ for a 3-sublattice AFQ state can be calculated by performing a gradient expansion for the 3-site, 9-bond cluster shown in Fig. 8 following the example of Eq. (32). This gives

$$\Delta H_{\text{SU}(2), \text{clus}} \approx 3(J_2 - J_1) \left( (d_A (r, \tau) \cdot d_B (r, \tau))^2 + |d_B (r, \tau) \cdot d_C (r, \tau)|^2 + |d_C (r, \tau) \cdot d_A (r, \tau)|^2 \right)$$

+ $3(J_2 - J_1) \frac{a^2}{2} \sum_{\lambda = x,y} \left[ |d_A \cdot \partial_\lambda d_B|^2 + |d_B \cdot \partial_\lambda d_C|^2 \right]$

- $3(J_2 - J_1) \frac{a^2}{4} \sum_{\lambda = x,y} \left[ (d_A \cdot \partial_\lambda d_A \cdot d_B) + (d_A \cdot d_B \cdot \partial_\lambda d_A) + (d_B \cdot d_C \cdot \partial_\lambda d_A) + (d_C \cdot d_A \cdot \partial_\lambda d_A) \right],$

Following the notation of Section IIB, the Hamiltonian

FIG. 10: (Color online). The full Brillouin zone (fbz) of the triangular lattice, together with the reduced magnetic Brillouin zone (mbz) for 3-sublattice order. Important symmetry points are labelled $\Gamma [k_F = (0,0)]$, $M [k_M = (2\pi/3, \pi/\sqrt{3})]$, $K [k_K = (4\pi/3, 0)]$ and $K' [k_{K'} = -k_K]$. In the field theory for the 3-sublattice antiferroquadrupolar (AFQ) state, the $\Gamma$ and $K'$ points are folded onto the $K$ point, and the wavevector $q$ measures the deviation from this point. The circuit in reciprocal space $\Gamma$-$K$-$M$-$\Gamma$ followed when plotting the inelastic neutron scattering intensity in Fig. 12 is indicated in red.
where the expansion has been truncated at second order in $a$.

Consider the product,

$$d_A(r, \tau) \cdot d_B(r, \tau) \approx (1, \tilde{r}, r') \cdot U^T U \cdot \left( \begin{array}{c} l_x \\ 1 \\ l_x \end{array} \right),$$  \hspace{1cm} (65)

where the matrices can be expressed as,

$$U^T U = \left( \begin{array}{ccc} n_A^2 & n_A \cdot n_B & n_C \cdot n_A \\ n_A \cdot n_B & n_B^2 & n_B \cdot n_C \\ n_C \cdot n_A & n_B \cdot n_C & n_C^2 \end{array} \right).$$  \hspace{1cm} (66)

The ground state of the system involves purely real (or purely imaginary) $d$ vectors, and therefore at low $T$ it is reasonable to approximate,

$$n_i^2 \approx 1,$$  \hspace{1cm} (67)

and,

$$n_i \cdot n_j \ll 1, \quad i \neq j.$$  \hspace{1cm} (68)

It follows that,

$$n_i \cdot n_j \approx -n_i \cdot n_j,$$  \hspace{1cm} (69)

and therefore,

$$d_A(r, \tau) \cdot d_B(r, \tau) \approx l_x + \tilde{r} + n_A \cdot n_B$$

$$d_B(r, \tau) \cdot d_C(r, \tau) \approx l_x + \tilde{r} + n_B \cdot n_C$$

$$d_C(r, \tau) \cdot d_A(r, \tau) \approx l_x + \tilde{r} + n_C \cdot n_A.$$  \hspace{1cm} (70)

Using these approximations, the first term in Eq. (64) can be re-expressed as,

$$|d_A(r, \tau) \cdot d_B(r, \tau)|^2 + |d_B(r, \tau) \cdot d_C(r, \tau)|^2 + |d_C(r, \tau) \cdot d_A(r, \tau)|^2 \approx$$

$$(l_x + \tilde{r})^2 + (l_x + \tilde{r})^2 + (l_x + \tilde{r})^2$$

$$+ |n_A \cdot n_B|^2 + |n_B \cdot n_C|^2 + |n_C \cdot n_A|^2.$$  \hspace{1cm} (71)

Following the same procedure as in Section 11.3 results in the Lagrangian,

$$L_{\Delta}^{SU(2)} = \text{Tr} \left[ U^T \cdot \partial_{\tau} U + 2 \left[ (s - \bar{s}) \cdot l_1 + i(s + \bar{s}) \cdot l_2 \right] 
+ 12J_1 l_1 l_1 + 12J_1 l_2 l_2 
+ 3(J_2 - J_1) (|n_A \cdot n_B|^2 + |n_B \cdot n_C|^2 + |n_C \cdot n_A|^2) + \text{gradient terms}. \right.$$  \hspace{1cm} (72)

The canting fields $\mathbf{I}$ can once again be eliminated within a saddle-point approximation. Performing the necessary functional derivative, and using Eq. (55) to write the result in terms of $n$, we find

$$I_1 \approx -\frac{1}{12J_2} \left( \begin{array}{c} \bar{n}_B \cdot \partial_{\tau} n_A - n_B \cdot \partial_{\tau} \bar{n}_A \\ \bar{n}_C \cdot \partial_{\tau} n_B - n_C \cdot \partial_{\tau} \bar{n}_B \\ n_A \cdot \partial_{\tau} n_C - n_A \cdot \partial_{\tau} \bar{n}_C \end{array} \right)$$

$$I_2 \approx \frac{i}{12J_1} \left( \begin{array}{c} \bar{n}_B \cdot \partial_{\tau} n_B + n_B \cdot \partial_{\tau} \bar{n}_B \\ \bar{n}_C \cdot \partial_{\tau} n_B + n_C \cdot \partial_{\tau} \bar{n}_B \\ n_A \cdot \partial_{\tau} n_C + n_A \cdot \partial_{\tau} \bar{n}_C \end{array} \right).$$  \hspace{1cm} (73)

These two canting fields correspond to physically distinct spin-- and quadrupole wave excitations. These are no longer degenerate once the SU(3) symmetry is broken, and to parameterise them, we need to introduce two distinct susceptibilities,

$$\chi_\perp^Q = \frac{2}{9J_1}, \quad \chi_\perp^S = \frac{2}{9J_2}.$$  \hspace{1cm} (74)

and two distinct director stiffnesses (which for this particular model, happen to be equal),

$$\rho_d^Q = \rho_d^S = J_2 a^2.$$  \hspace{1cm} (75)

It also proves convenient to reparameterize the term in $L_{\Delta}^{SU(2)}$ which breaks SU(3) symmetry in terms of a gap to spin wave excitations, i.e.,

$$\delta L_{\Delta}^{SU(2)} = \frac{3}{8} \chi_\perp^Q \Delta^2 (|n_A \cdot n_B|^2 + |n_B \cdot n_C|^2 + |n_C \cdot n_A|^2)$$  \hspace{1cm} (76)

where

$$\Delta = \sqrt{36J_2(J_2 - J_1)}.$$  \hspace{1cm} (77)

Collecting these facts together, the action describing long-wavelength excitations of $\mathbf{3}$-sublattice AFQ order is,

$$S_{\Delta}^{SU(2)}[n_A, n_B, n_C]$$

$$= \frac{1}{4\sqrt{3}} \int_0^3 \int d\tau d^2 r \left( \frac{8}{3} \sum_i \bar{n}_i \cdot \partial_{\tau} n_i + \chi_\perp^Q \left[ \left( \bar{n}_A \cdot \partial_{\tau} n_B + n_A \cdot \partial_{\tau} \bar{n}_B \right)^2 
+ \left( \bar{n}_B \cdot \partial_{\tau} n_C + n_B \cdot \partial_{\tau} \bar{n}_C \right)^2 
+ \left( \bar{n}_C \cdot \partial_{\tau} n_A + n_C \cdot \partial_{\tau} \bar{n}_A \right)^2 \right] \right.$$ 

$$- \chi_\perp^S \left[ \left( \bar{n}_A \cdot \partial_{\tau} n_B - n_A \cdot \partial_{\tau} \bar{n}_B \right)^2 
+ \left( \bar{n}_B \cdot \partial_{\tau} n_C - n_B \cdot \partial_{\tau} \bar{n}_C \right)^2 
+ \left( \bar{n}_C \cdot \partial_{\tau} n_A - n_C \cdot \partial_{\tau} \bar{n}_A \right)^2 \right] \right.$$ 

$$+ \rho_d^Q \sum_{\lambda=x,y} \left[ \left( \bar{n}_A \partial_{\lambda} n_B + n_A \partial_{\lambda} \bar{n}_B \right)^2 
+ \left( \bar{n}_B \partial_{\lambda} n_C + n_B \partial_{\lambda} \bar{n}_C \right)^2 
+ \left( \bar{n}_C \partial_{\lambda} n_A + n_C \partial_{\lambda} \bar{n}_A \right)^2 \right]$$ 

$$- \rho_d^S \sum_{\lambda=x,y} \left[ \left( \bar{n}_A \partial_{\lambda} n_B - n_A \partial_{\lambda} \bar{n}_B \right)^2 
+ \left( \bar{n}_B \partial_{\lambda} n_C - n_B \partial_{\lambda} \bar{n}_C \right)^2 
+ \left( \bar{n}_C \partial_{\lambda} n_A - n_C \partial_{\lambda} \bar{n}_A \right)^2 \right]$$ 

$$+ \chi_\perp^S \Delta^2 (|n_A \cdot n_B|^2 + |n_B \cdot n_C|^2 + |n_C \cdot n_A|^2) \right\}.$$  \hspace{1.5cm} (78)

where the relevant parameters for the microscopic model $H_{\Delta}^{BBQ}$ [Eq. (1)] are given in Table II and the partition function is defined as in Eq. (45).
we use Eq. (51) to expand small fluctuations about the lattice. (a) prediction of the continuum field theory $S_{\Delta}^{SU(2)}[\phi]$ [Eq. (80)], with dispersion given by $\omega_1^Q$ [Eq. (81)] and $\omega_2^S$ [Eq. (82)]. (b) prediction of the microscopic model $H_{\Delta}^{BBQ}$ [Eq. (1)] in the magnetic Brillouin zone (mbz) [see Fig. 11], for parameters $J_1 = 1$ and $J_2 = 1.22$. The dispersion is given by $\omega_1^Q$ [Eq. (83)]. In both cases a three-fold degenerate branch of gapless, quadrupole-wave excitations, are centered on the ordering vector $k = k_1$ [i.e. $\mathbf{q} = 0$]. These are the Goldstone modes of the AFQ order. They are accompanied by a three-fold degenerate branch of gapped, spin-wave excitations. Approaching the centre of the mbz, $\mathbf{q} → 0$, the continuum theory and the lattice theory match exactly.

Eq. (78) can be used to re-express this action in terms of the unitary matrix field, $U(r, r)$, as,

$$
S_{\Delta}^{SU(2)}[U] = \frac{1}{8\sqrt{3}a^2} \int_{0}^{\beta} d\tau \int d^2 r \left\{ \frac{16}{3} Tr[U^\dagger \cdot \partial_\tau U] + \chi_1^Q \left[ Tr(U^\dagger \cdot \partial_\tau U + U^T \cdot \partial_\tau U^T) (U^\dagger \cdot \partial_\tau U + U^T \cdot \partial_\tau U^T) \right] + \chi_1^S \left[ Tr(U^\dagger \cdot \partial_\tau U - U^T \cdot \partial_\tau U^T) (U^\dagger \cdot \partial_\tau U - U^T \cdot \partial_\tau U^T) \right] - 4 \sum_m m \left[ ||U^\dagger \cdot \partial_\tau U||_{mm}^2 \right] \right\}.
$$

This reduces to Eq. (79) when $\chi_1^Q = \chi_1^S$ and $\Delta = 0$ (i.e. $J_1 = J_2$), as required.

2. Linearising the order parameter fields

The physical content of the action $S_{\Delta}^{SU(2)}[\mathbf{U}]$ [Eq. (78)], becomes clear on linearisation of the fields. Once again, we use Eq. (61) to expand small fluctuations about the

$$
S_{\Delta}^{SU(2)}[\phi] \approx \frac{1}{\sqrt{3}a^2} \int_{0}^{\beta} d\tau \int d^2 r \left\{ \sum_{p=1…3} \left[ \chi_1^Q (\partial_\tau \phi_p)^2 + \rho_4^Q \sum_{\lambda=xy} (\partial_\lambda \phi_p)^2 \right] \right\} + \sum_{p=4…6} \left[ \chi_1^S (\partial_\tau \phi_p)^2 + \rho_4^S \sum_{\lambda=xy} (\partial_\lambda \phi_p)^2 + \chi_1^S (\partial_\tau \phi_p)^2 \right]
$$

TABLE I: Dictionary for translating between the parameters of the continuum field theory for 3-sublattice AFQ order, $S_{\Delta}^{SU(2)}[\mathbf{U}]$ [Eq. (79)], and the parameters of the relevant microscopic model $H_{\Delta}^{BBQ}$ [Eq. (1)], in the vicinity of the SU(3) point $J_1 = J_2$.

ground state in terms of $\phi$. This leads to the action
We immediately see that there are three gapless, quadrupole-wave modes, $\phi_1$, $\phi_2$ and $\phi_3$, with dispersion,

$$\omega^Q_q \approx v_q |q|, \quad v_q = \sqrt{\frac{\rho_d^Q}{\chi^Q}} = 3 \sqrt{\frac{J_1 J_2}{2} a},$$

and three gapped, spin-wave modes $\phi_4$, $\phi_5$ and $\phi_6$, with dispersion

$$\omega^S_q \approx \sqrt{\Delta^2 + v_S^2 q^2}, \quad v_S = \sqrt{\frac{\rho_d^S}{\chi^S}} = 3 \frac{J_2 a}{\sqrt{2}}.$$  

These are shown in Fig. 11. The Goldstone modes correspond to real rotations of the order parameter fields, while the gapped modes (gap $\Delta$) correspond to rotations into complex space.

The microscopic ‘flavour-wave’ theory developed by Tsunetsugu and Arikawa [40,41] predicts a dispersion,

$$\omega^\pm_q = 3J_2 \sqrt{\left(1 \pm |\gamma_k|\right) \left(1 \pm \left(1 - \frac{2J_1}{J_2}\right) |\gamma_k|\right)} ,$$

where $\gamma_k$ is given by Eq. (61). This is shown in Fig. 11. In the long wavelength limit, and for small $J_2 - J_1$, the dispersion reduces to Eq. (81) and Eq. (82).

We re-emphasise that the validity of the continuum theory breaks down approaching the FM phase for $\theta \to \pi/2$ ($J_1 \to 0$, $J_2 > 0$). Crossing the AFQ phase, there is a progressive reduction in the area of reciprocal space over which the quadrupole-wave dispersion, $\omega^Q_k$, is linear. This is also a feature of the lattice theory — exactly at the phase boundary with the ferromagnet ($J_1 = 0$, $J_2 > 0$) the dispersion, $\omega^Q_k$ [Eq. (83)], becomes quadratic even for $|k| \to 0$. This signals that it is no longer appropriate to describe the system in terms of the quantum non-linear sigma model, $S^{SU(2)}_\Delta [U]$, [Eq. (79)], with canting field,

$$I \approx iI_2 \approx \frac{3}{4} \frac{\rho_d^Q}{\chi^Q} \left( n_A \cdot \partial_r n_A \right) n_A \cdot \partial_r n_A$$

and the partition function is,

$$Z^{SO(3)}_\Delta \propto \int \prod_{i \neq j} \mathcal{D}n_i \delta(n_i^2 - 1) \delta(n_i \cdot n_j) e^{-S^{SO(3)}_\Delta [n_A, n_B, n_C]}.$$  

This is an $SO(3)$ symmetric non-linear sigma model[26], a fact which is clearer if the action is written in matrix form,

$$S^{SO(3)}_\Delta [R] \approx \frac{1}{2 \sqrt{3} a^2} \int_0^\beta d\tau \int d^2 r \left\{ \chi^Q \sum_{\lambda=x,y} \text{Tr} \left[ \partial_\tau R^T \cdot \partial_\tau R \right] + \rho_d^Q \sum_{\lambda=x,y} \text{Tr} \left[ \partial_\lambda R^T \cdot \partial_\lambda R \right] \right\},$$

where $R$ is a real-valued rotation matrix given by,

$$R(r, \tau) = \begin{pmatrix} n^x_A(r, \tau) & n^y_B(r, \tau) & n^z_C(r, \tau) \\ n^z_A(r, \tau) & n^x_B(r, \tau) & n^y_C(r, \tau) \\ n^y_A(r, \tau) & n^z_B(r, \tau) & n^x_C(r, \tau) \end{pmatrix}.$$  

The simplified action, Eq. (84), describes the 3 quadrupole-wave modes shown in Fig. 11 but ignores the 3 spin-wave modes which dominate experimental responses at higher energy.

E. The low temperature, low energy limit

For temperature and energy scales lower than the spin wave gap, $\Delta$, the high-energy, spin-wave modes can be neglected. This considerably simplifies the action, $S^{SU(2)}_\Delta [n]$ [Eq. (79)], and is a useful approximation when considering low temperature thermodynamic properties.

Neglection of the spin-wave modes is equivalent to making the assumption that the fields $n_i$, are real. The simplified action is then given by,

$$S^{SO(3)}_\Delta [n_A, n_B, n_C] \approx \frac{1}{2 \sqrt{3} a^2} \int_0^\beta d\tau \int d^2 r \left\{ \chi^Q \left[ (\partial_\tau n_A)^2 + (\partial_\tau n_B)^2 + (\partial_\tau n_C)^2 \right] 

+ \rho_d^Q \sum_{\lambda=x,y} \left[ (\partial_\lambda n_A)^2 + (\partial_\lambda n_B)^2 + (\partial_\lambda n_C)^2 \right] \right\} ,$$

with

F. Comparison with other forms of magnetic order

It is interesting to compare the continuum theory of long-wavelength excitations in a 3-sublattice AFQ state, $S^{SU(2)}_\Delta [U]$ [Eq. (79)], with sigma-model approaches to other forms of magnetic order. Perhaps the most widely known example is the sigma-model treatment of the collinear antiferromagnet (AFM) [72,73]. The collinear nature of this state means that it does not break the full $SU(2)$ spin-rotation symmetry, but instead $SU(2)/U(1)$. As a consequence the resulting sigma model describes only two, degenerate, linearly-dispersing Goldstone modes, both with the character of spin-wave excitations. The only gapped excitation possible at long wavelength is a longitudinal fluctuation of the order parameter, explicitly absent from the sigma model. Collinear-
modes associated with the breaking of spin-symmetry, and three degenerate, gapped “spin-wave” modes, associated with dipolar excitations of the underlying quadrupolar order. The two actions therefore differ in both the number and the character of the modes they describe. It is also worth noting that, the structure of the interactions between these excitations (not described in this article) is profoundly different, and includes vertices with an odd number of excitations. This topic will be explored further elsewhere.

The action $\mathcal{S}_{\text{SU}(2)}^\text{SU(2)}(\mathbf{U})$ [Eq. (79)] finds more parallels with non-collinear magnetic ordering. A good example of this is the 120° state on the triangular lattice. This fully breaks the SU(2) symmetry, and therefore has three, linearly-dispersing Goldstone modes, all with the character of spin waves. Interactions between odd numbers of spin excitations are also now permitted by symmetry. However, as with the collinear antiferromagnet, the 120° state has no low-energy gapped modes at long-wavelength. Also, the coplanar nature of this state means that the spin stiffness’ associated with the three Goldstone modes are not all equal, and only two of the three Goldstone modes are degenerate.

Finally it is interesting to compare $\mathcal{S}_{\text{SU}(2)}^\text{SU(2)}(\mathbf{U})$ [Eq. (79)] with field theories describing FQ order. As with the collinear AFM, FQ states have only two Goldstone modes. These are degenerate, linearly-dispersing, and have the character of quadrupole waves at long wavelength. Only interactions between even numbers of spin excitations are permitted by symmetry. Both of these points clearly distinguish the present theory of AFQ order from the earlier work on FQ order.

In fact the theory derived in Ref. [50] has the same action as the collinear AFM, albeit with a different physical interpretation. However, in reducing the action to this form, imaginary fluctuations of the director $\mathbf{d}$ have been explicitly integrated out, eliminating much of the information concerning excitations with “spin-wave” character. An important feature of the SU(3)-derived approach developed in this article is its ability to describe gapped excitations with dipolar character, such as the “spin-wave” modes of AFQ order, which cannot be accessed in the SO(3) approach of Ref. [50]. Such modes are particularly interesting since they will be the easiest to observe in, e.g., inelastic neutron scattering.

G. Machinery for calculating correlation functions

In order to make predictions for inelastic neutron scattering and for the dynamical quadrupole susceptibility, it is necessary to translate the continuum field theory, $\mathcal{S}_{\text{SU}(2)}^\text{SU(2)}(\mathbf{U})$ [Eq. (79)] — which is written in terms of rotations of directors — back into the language of spins and quadrupoles. Following Eq. (21) and Eq. (50), the directors on the three sublattices can be approximated as,

$$\mathbf{d}_A \approx \begin{pmatrix} 1 \\ -\phi_1 + i\phi_4 + \bar{\Gamma} \\ \phi_2 + i\phi_5 + \bar{r} \end{pmatrix}, \quad \mathbf{d}_B \approx \begin{pmatrix} -\phi_3 + i\phi_6 + \bar{r} \\ \phi_1 + i\phi_4 + \bar{\Gamma} \\ 1 \end{pmatrix}, \quad \mathbf{d}_C \approx \begin{pmatrix} -\phi_2 + i\phi_5 + \bar{r} \\ \phi_3 + i\phi_6 + \bar{r} \\ 1 \end{pmatrix},$$

with the canting fields,

$$\mathbf{l}_1 \approx -\frac{3}{4} \mathbf{\chi}^\perp \begin{pmatrix} \partial_t \phi_4 \\ \partial_t \phi_6 \\ \partial_t \phi_5 \end{pmatrix}, \quad \mathbf{l}_2 \approx \frac{3}{4} \mathbf{\chi}^\perp \begin{pmatrix} \partial_t \phi_1 \\ \partial_t \phi_3 \\ \partial_t \phi_2 \end{pmatrix},$$

where the real time $t = -it$ has been used. It follows that the $\mathbf{d}$ vectors are,

$$\mathbf{d}_A \approx \begin{pmatrix} 1 \\ -\phi_1 + i\phi_4 - \frac{3}{4} \mathbf{\chi}^\perp \partial_t \phi_4 - i\frac{3}{4} \mathbf{\chi}^Q \partial_t \phi_1 \\ \phi_2 + i\phi_5 - \frac{3}{4} \mathbf{\chi}^\perp \partial_t \phi_5 + i\frac{3}{4} \mathbf{\chi}^Q \partial_t \phi_2 \end{pmatrix}, \quad \mathbf{d}_B \approx \begin{pmatrix} -\phi_3 + i\phi_6 - \frac{3}{4} \mathbf{\chi}^\perp \partial_t \phi_6 - i\frac{3}{4} \mathbf{\chi}^Q \partial_t \phi_3 \\ \phi_1 + i\phi_4 - \frac{3}{4} \mathbf{\chi}^\perp \partial_t \phi_4 + i\frac{3}{4} \mathbf{\chi}^Q \partial_t \phi_1 \\ 1 \end{pmatrix}, \quad \mathbf{d}_C \approx \begin{pmatrix} -\phi_2 + i\phi_5 - \frac{3}{4} \mathbf{\chi}^\perp \partial_t \phi_5 - i\frac{3}{4} \mathbf{\chi}^Q \partial_t \phi_2 \\ \phi_3 + i\phi_6 - \frac{3}{4} \mathbf{\chi}^\perp \partial_t \phi_6 + i\frac{3}{4} \mathbf{\chi}^Q \partial_t \phi_3 \\ 1 \end{pmatrix},$$

Substituting these expressions into Eq. (51) leads to the fluctuating dipole moments,

$$\mathbf{S}_A \approx \begin{pmatrix} 0 \\ -2\phi_5 - \frac{3}{2} \mathbf{\chi}^Q \partial_t \phi_2 \\ 2\phi_4 - \frac{1}{2} \mathbf{\chi}^\perp \partial_t \phi_1 \end{pmatrix}, \quad \mathbf{S}_B \approx \begin{pmatrix} 2\phi_6 - \frac{3}{2} \mathbf{\chi}^\perp \partial_t \phi_3 \\ 0 \\ -2\phi_4 - \frac{3}{2} \mathbf{\chi}^Q \partial_t \phi_1 \end{pmatrix}, \quad \mathbf{S}_C \approx \begin{pmatrix} -2\phi_6 - \frac{3}{2} \mathbf{\chi}^Q \partial_t \phi_3 \\ 2\phi_5 - \frac{1}{2} \mathbf{\chi}^\perp \partial_t \phi_2 \\ 0 \end{pmatrix},$$

where terms linear in the $\phi$ fields have been retained. Eq. (92) provides the starting point for the theory of inelastic neutron scattering developed in Section III of this paper.
The supplemental material contains animations showing the nature of the quadrupole-wave excitations and spin-wave excitations.

III. PREDICTIONS FOR INELASTIC NEUTRON SCATTERING

A. General considerations: waves in the unseen

Since each “spin” in a quantum magnet possesses a magnetic dipole, conventional dipolar magnetic order gives rise to a static internal magnetic field. Neutrons, which also possess a dipole moment, diffract from this static field to give magnetic Bragg peaks. As in conventional crystallography, the form of magnetic order present is encoded in the wave number and intensity of these magnetic Bragg peaks. However, since spin-nematic order corresponds to a quadrupolar order of spins, it does not break time-reversal symmetry and cannot give rise to static magnetic fields. For this reason, it does not manifest itself through magnetic Bragg peaks in elastic neutron scattering.

An elegant solution to this problem, in the presence of an anisotropy that breaks SU(2) symmetry, was proposed by Barzykin and Gorkov, who suggested using an external magnetic field to break time-reversal symmetry. In the presence of magnetic anisotropy, applying a uniform magnetic field to an AFQ state induces a small, staggered, dipole moment which can, in principle, be observed in elastic neutron scattering. Resonant magnetic X-ray scattering, which is sensitive to quadrupole moments of spins, has also been used to identify AFQ order in the rare-earth magnet UPd$_2$Sn. However, a very direct and appealing route to identifying spin-nematic order, even in the absence of magnetic anisotropy, would be to map out its magnetic excitations using inelastic neutron scattering.

Since spin-nematic order breaks spin-rotation symmetry it must possess Goldstone modes. The long-wavelength excitations are generated by real SU(2) rotations of the underlying quadrupolar order parameter, and so can best be thought of as “quadrupole waves”. Quadrupole waves possess a small fluctuating dipole moment, and will reveal themselves as linearly-dispersing excitations — visible waves in the unseen spin-nematic order. As we will see in what follows, the size of this dipole moment is directly proportional to the speed at which the quadrupoles rotate, and so the intensity of scattering from a quadrupole wave vanishes linearly with its energy.

However, precisely because the building blocks of spin-nematic order are quadrupole moments of spins, these Goldstone modes do not exhaust the possible excitations of a spin-nematic state. Neutrons can also drive transitions between different triplet states, which mix a strong spin-dipole into the underlying quadrupole moment. In AFQ spin-nematic states, this leads to a second, distinct, type of long-wavelength excitation, with a gapped spectrum and a pronounced intensity in inelastic neutron scattering. Identifying this gapped excitation in experiment, together with the appropriate set of gapless Goldstone modes, would provide strong evidence for the existence of spin-nematic order.

In Section III of this paper we have developed the tools needed to make distinctive, quantitative predictions for both types of excitation of a spin-nematic — a continuum field-theory of the excitations of AFQ order based on the symmetries of the underlying order parameter. This SU(3) “sigma-model” approach offers a quantitative description of excitations — in terms of the minimum set of physically meaningful parameters — without the need to specify a microscopic model.

In what follows we use this continuum theory to make predictions for inelastic neutron scattering carried out on a 3-sublattice AFQ state. These predictions are exact at long wavelength, and fully constrain the symmetries broken by the AFQ state. We make explicit comparison with the predictions of a microscopic, spin-1 lattice model that realises the same ordered state. In order to keep the discussion reasonably self-contained, key results from Section III are quoted in the text.

B. Sum rules and correlation functions

Inelastic neutron scattering measures the imaginary part of the dynamical spin susceptibility,

$$\Im m\{\chi^\alpha_\omega(k, \omega)\} = (g\mu_B)^2 \Im m\{i \int_0^\infty dt e^{i\omega t} <\delta S^\alpha(k,t)\delta S^\beta(-k,0)>\}$$

(94)
where \( \alpha, \beta = x, y, z \) label spin components. In the case of the 3-sublattice AFQ state described in Section [II] this tensor is diagonal, and fluctuations are isotropic in spin space, i.e.

\[
\Im \{ \chi^\beta (k, \omega) \} = \Im \{ \chi^\beta (k, \omega) \} = \Im \{ \chi^\beta (k, \omega) \}.
\]

An important check on any calculation of the dynamical susceptibility is that it obeys the relevant sum rules. For any theory with SU(2) spin symmetry, as is the case for the 3-sublattice AFQ state, it is required that, in perturbation theory. For single particle excitations must vanish for all \( \omega \neq 0 \). The sum rule is related to a Ward-Takahashi identity, and thus holds at each order in perturbation theory. For single particle excitations it is sufficient to consider the non-interacting theory described by \( \mathcal{S}^{SU(2)}[\phi] \) [Eq. (80)]. However, in order to understand the 2-particle continuum it is necessary to take three and four field interactions into account and form a Dyson equation for the self energy. Since this is an involved process, we postpone discussion until a future publication. We note that the linear flavour wave analysis of Tsunetsugu and Arikawa [10] obeys the sum rule, Eq. (87), at leading order, but has finite weight at \( k = 0 \) and \( \omega = 0 \) arising from the 2-particle continuum.

In Section [IV] we also consider the dynamical quadrupole susceptibility. This is given by,

\[
\Im \{ \chi^\alpha \chi^\beta (k, \omega) \} = (g_{\mu B})^4 \Im \{ i \int_0^\infty dt e^{i\omega t} \Delta Q^\alpha (k, t) \Delta Q^\beta (-k, 0) \}. \tag{96}
\]

In the general case of SU(2) spin symmetry, there are no analogous sum rule to Eq. (97), and one expects to find finite weight at \( k = 0 \) and \( \omega = 0 \). However, exactly at the SU(3) point the expanded symmetry leads to the quadrupolar sum rule,

\[
\lim_{q \to 0} \int d\omega e^{i\omega t} \omega \chi^\alpha \chi^\beta (k, \omega) = 0 \quad [SU(3) \text{ point}]. \tag{97}
\]

C. Neutron scattering in a 3-sublattice AFQ

1. Spin excitations in a 3-sublattice AFQ state

Predictions for inelastic neutron scattering from a 3-sublattice AFQ state have previously been published by Tsunetsugu and Arikawa [10,41], based on flavour-wave calculations for the spin-1 bilinear-biquadratic (BBQ) model on the triangular lattice \( \mathcal{H}_\Delta^{BBQ} \) [Eq. (1)]. In what follows we show how the universal, long-wavelength features of these results are completely described by the field theory developed in Section [III] of this paper. The tools needed to calculate \( \Im \{ \chi^\alpha (k, \omega) \} \) — namely a theory of long-wavelength spin excitations in a spin-nematic state — were developed in Section [III] of this paper. Here we briefly reprise the most relevant results.

Small fluctuations about the 3-sublattice AFQ ordered state can be described by the linearized action, \( \mathcal{S}^{SU(2)}[\phi] \) [Eq. (83)], viz :

\[
\mathcal{S}^{SU(2)}[\phi] \approx \frac{1}{\sqrt{3} a^2} \int_0^\beta d\tau \int d^2r \sum_{p=1,3} \left[ \lambda_Q \left( \partial_r \phi_p \right)^2 + \rho_Q \sum_{\lambda=x,y} \left( \partial_\lambda \phi_p \right)^2 \right] + \sum_{p=4,6} \left[ \lambda_\perp \left( \partial_r \phi_p \right)^2 + \rho_\perp \sum_{\lambda=x,y} \left( \partial_\lambda \phi_p \right)^2 + \chi_\perp \Delta^2 \phi_p^2 \right].
\]

The long-wavelength properties of the 3-sublattice AFQ state are completely characterised by the four parameters \( \lambda_Q, \rho_Q = \rho_\perp \) and \( \Delta \). Table [I] in Section [I.D] provides a “dictionary” for converting between the parameters of the continuum theory, and the parameters of the minimal microscopic model \( \mathcal{H}_\Delta^{BBQ} \) [Eq. (1)].

The dispersion of the spin excitations of this spin-nematic state then follow from the usual Euler-Lagrange equations. The three fields \( \phi_1, \phi_2 \) and \( \phi_3 \) describe Goldstone modes with linear dispersion \( \omega^Q_q \) [Eq. (51)], viz :

\[
\omega^Q_q \approx \nu_Q |q|, \quad \nu_Q = \sqrt{\frac{\rho_Q}{\chi_\perp}}.
\]

while the three fields \( \phi_4, \phi_5 \) and \( \phi_6 \), describe gapped excitations with dispersion \( \omega_\perp_q \) [Eq. (52)], viz :

\[
\omega_\perp_q \approx \sqrt{\Delta^2 + \nu_\perp^2 q^2}, \quad \nu_\perp = \sqrt{\frac{\rho_\perp}{\chi_\perp}}.
\]

The remaining challenge is to correctly reference the continuum theory back to the lattice, and to calculate the intensities associated with each branch of excitation. To do this it is necessary to transcribe the spin degrees of freedom \( (S^x, S^y, S^z) \) in terms of the fields \( \phi \) and then decompose spin-spin correlations \( \langle S^\alpha S^\beta \rangle \) as contractions of the \( \phi \) fields. These can contain contributions from more than one kind of excitation. A worked example of this type of calculation is given in Appendix A of Ref. [80].

It follows from Eq. (52) [Section I.E] that, to leading
order in $\phi$, 

$$\delta S(r, t) \approx \frac{-\chi^Q}{2} \left( \left( 2 - e^{ik_{\phi, r}} - e^{-ik_{\phi, r}} \right) \partial_i \phi_3 ight)$$

$$- \frac{2}{3} \left( \frac{e^{ik_{\phi, r}} - e^{-ik_{\phi, r}}}{e^{ik_{\phi, r}} - e^{-ik_{\phi, r}}} \right) \left( \left( 1 + e^{i\frac{2\pi}{3}} \right) e^{ik_{\phi, r}} + \left( 1 - e^{-i\frac{2\pi}{3}} \right) e^{-ik_{\phi, r}} \right)$$

$$+ \frac{2}{3} \left( \left( 1 + e^{i\frac{2\pi}{3}} \right) e^{ik_{\phi, r}} + \left( 1 - e^{-i\frac{2\pi}{3}} \right) e^{-ik_{\phi, r}} \right) \phi_3$$

(98)

From Eq. (98), we can immediately identify the three fields $\phi_1$, $\phi_2$ and $\phi_3$ with quadrupole waves whose contribution to scattering vanishes as $\chi^Q \propto e^{-i\omega t}$. Meanwhile, the three fields $\phi_4$, $\phi_5$ and $\phi_6$ are spin waves with a robust dipole moment.

2. Single particle scattering near $k = k_\mathrm{K}$

Let us consider first scattering involving a single excitation near to the ordering vector, $k = k_\mathrm{K}$. Here the field theory predicts a gapless Goldstone mode with dispersion $\omega^Q_k$ [Eq. (51)], for small $q = k - k_\mathrm{K}$. This is accompanied by a gapped spin-wave excitation with dispersion $\omega^S_k$ [Eq. (52)]. The associated single-particle contribution to the dynamical susceptibility is,

$$\Im \{ \chi^S(k_\mathrm{K} + q, \omega) \} \approx \frac{\pi}{2} (g \mu_B)^2 \chi^Q_{\omega q} \delta(\omega - \omega^Q_k)$$

$$+ \frac{2\pi}{3} (g \mu_B)^2 \frac{1}{\chi^S_{\omega q}} \delta(\omega - \omega^S_k), \quad (99)$$

where $q \approx 0$. Scattering close to the $K'$ point is exactly equivalent. From Eq. (99) we see that the intensity of scattering from the quadrupole wave vanishes as $\chi^Q_{\omega q} \propto 1/|q|$ for $q \to 0$. Meanwhile the scattering from the spin-wave excitation is enhanced as $1/(\chi^S_{\omega q}) \sim 1/(\Delta \chi^S_k)$ in the same limit. The spin-wave excitation will therefore dominate the response seen in experiment. These features are illustrated in Fig. [12]

Exactly the same quadrupole and spin wave excitations are found in flavour-wave calculations for the 3-sublattice AFQ phase of the spin-1 BBQ model on the triangular lattice $H_{\mathrm{BBQ}}$ [Eq. (1)]. These predict a 1-particle contribution to the dynamical susceptibility which behaves as,

$$\Im \{ \chi^S(k, \omega) \} \approx$$

$$\pi (1 + \cos \theta_k)(g \mu_B)^2 J_2(1 - |\gamma_k|) \omega_k^- \delta(\omega - \omega_k^-)$$

$$+ \pi (1 - \cos \theta_k)(g \mu_B)^2 J_2(1 + |\gamma_k|) \omega_k^+ \delta(\omega - \omega_k^+),$$

(100)

where $\omega_k^\pm$ is given in Eq. (83), $\gamma_k$ in Eq. (61) and,

$$e^{i\theta_k} = \frac{\gamma_k}{|\gamma_k|}.$$
FIG. 12: (Color online). Prediction for inelastic neutron scattering from a state with 3-sublattice antiferroquadrupolar (AFQ) spin nematic order of the type shown in Fig. 6. Animations showing the nature of the spin-dipole fluctuations associated with the gapless expansion, the only non-zero entries in the susceptibility in this basis, and at leading order in the perturbation $H_gapped$ have been convoluted with a gaussian of FWHM 0.042$\Delta$ to mimic experimental resolution. The circuit $\Gamma-K-M-\Gamma$ in reciprocal space is shown in Fig. 10.

A. Quadrupolar excitations in a 3-sublattice AFQ state

It follows from Eq. (103) [Section II C] that, to linear order in $\phi$,

\[
\delta Q(r, t) \approx \begin{pmatrix}
0 \\
\frac{2}{3} \\
\frac{\sqrt{3}}{2}
\end{pmatrix} \\
\begin{pmatrix}
0 \\
(1 - e^{i2\Delta \phi})e^{i\mathbf{k}_3 \cdot \mathbf{r}} + [1 - e^{-i2\Delta \phi}]e^{-i\mathbf{k}_3 \cdot \mathbf{r}} \phi_1 \\
(e^{i2\Delta \phi} - e^{-i2\Delta \phi})e^{i\mathbf{k}_6 \cdot \mathbf{r}} - e^{-i\mathbf{k}_6 \cdot \mathbf{r}} \phi_3 \\
(1 - e^{-i2\Delta \phi})e^{i\mathbf{k}_3 \cdot \mathbf{r}} + [1 - e^{i2\Delta \phi}]e^{-i\mathbf{k}_3 \cdot \mathbf{r}} \phi_2 \\
\end{pmatrix}
\]

and those related by the symmetry of the $Q^{\alpha \beta}$ tensor.

From Eq. (103) we can see that the $\phi_1$, $\phi_2$ and $\phi_3$ Goldstone mode fields give a diverging contribution to the quadrupolar susceptibility approaching the Bragg peak at $k = k_K$. Conversely, the quadrupole fluctuations induced dynamically by the gapped, spin wave modes are small as $\chi^3_0 \partial_\phi \sim \omega^3_0$.

1. Single particle scattering near to $k = k_K$

The dynamical quadrupolar susceptibility can be determined in an analogous manner to the spin susceptibility [see Section III C]. Close to $k = k_K$ the field theory predicts,

\[
\Im \{\chi^{xy}_{\mathbf{Q}}(\mathbf{k} + \mathbf{q}, \omega)\} \approx \frac{2\pi}{3}(g\mu_B)^4 \frac{1}{\chi_1^0 \omega_0^0} \delta(\omega - \omega^0_\mathbf{q}) + \frac{\pi}{8}(g\mu_B)^4 \chi^3_0 \omega^3_0 \delta(\omega - \omega^3_\mathbf{q}),
\]

In this basis, and at leading order in the perturbation expansion, the only non-zero entries in the susceptibility tensor are,

\[
\Im \{\chi^{xy}_{\mathbf{Q}}(\mathbf{k}, \omega)\} = \Im \{\chi^{zyz}_{\mathbf{Q}}(\mathbf{k}, \omega)\} = \Im \{\chi^{xzz}_{\mathbf{Q}}(\mathbf{k}, \omega)\},
\]

(104)
Fig. 13: (Color online). Prediction for the dynamical quadrupolar susceptibility, \( \Im m \{ \chi^{\text{xyxy}}_Q(k, \omega) \} \), for a state with 3-sublattice antiferroquadrupolar (AFQ) spin nematic order of the type shown in Fig. 6. Animations showing the nature of the spin-dipole fluctuations associated with the gapless and gapped excitations are shown in the supplemental material. (a) prediction of the microscopic ‘flavour wave’ theory, as calculated from \( \mathcal{H}_{\text{BBQ}} \) [Eq. (1)] for \( J_1 = 1, J_2 = 1.22 \). The dashed white lines show the one-particle dispersion relations \( \omega_k^\pm \) [Eq. (81)], where the gap to spin-wave excitations is \( \Delta = 6 \sqrt{J_2(J_2 - J_1)} \). (b) prediction of the continuum theory \( \mathcal{S}_{\text{SU}(2)} \Delta [\phi] \) [Eq. (80)] for the same set of parameters. The dashed white lines show the one-particle dispersion relations \( \omega^Q_k \) [Eq. (81)] and \( \omega^S_k \) [Eq. (82)]. The dominant feature is a diverging ‘Bragg peak’ associated with the gapless, quadrupole-wave mode in the vicinity of the 3-sublattice AFQ ordering vector \( k = (4\pi/3, 0) \). All predictions have been convoluted with a gaussian of FWHM 0.042\( \Delta \) to mimic experimental resolution.

where \( q \approx 0 \). Scattering close to the \( K' \) point is exactly equivalent. Eq. (105) shows that the intensity of scattering due to the quadrupolar modes diverges as \( 1/(\chi^Q_{\perp} \omega^Q_q) \sim 1/|q| \) for \( q \to 0 \). Thus there is a ‘Bragg peak’ in the quadrupolar susceptibility, as one expects for quadrupolar order. The gapped spin-wave modes induce a small quadrupole fluctuation, and this gives only a weak contribution to the susceptibility.

Linear flavour wave theory for the spin-1 BBQ model on the triangular lattice, \( \mathcal{H}_{\text{BBQ}} \) [Eq. (1)], predicts,

\[
\Im m \{ \chi^{\text{xyxy}}_Q(k, \omega) \} \approx \frac{\pi}{2} (g \mu_B)^4 \chi^S_\perp \omega^S_q \delta(\omega - \omega^S_q).
\]

and this is quantitative agreement with the field theory, Eq. (106), approaching the high symmetry points.

2. Single particle scattering near \( k = 0 \)

Close to the \( \Gamma \) point, we find a one-particle contribution to the dynamical quadrupolar susceptibility,

\[
\Im m \{ \chi^{\text{xyxy}}_Q(q, \omega) \} \approx \frac{\pi}{2} (g \mu_B)^4 \chi^S_\perp \omega^S_q \delta(\omega - \omega^S_q).
\]

The quadrupole fluctuations induced dynamically by the gapped, spin-wave modes are suppressed by a factor \( \chi^S_\perp \omega^S_q \) and have low intensity compared to the diverging Goldstone mode at the \( \Gamma \) point. One interesting feature is that the gapless quadrupole mode at the \( \Gamma \) point does not appear in the field theory calculation of the susceptibility, due to the fact that neighbouring quadrupoles beat in antiphase [see Eq. (93)]. This is in agreement with the flavour wave theory, Eq. (106), where the susceptibility turns on very slowly as \( \Im m \{ \chi^{\text{xyxy}}_Q(q, \omega) \} \sim q^5 \).

3. Adding it all up

Fig. 13 shows the result of summing all the 1-particle contributions to the \( T = 0 \) dynamic quadrupolar susceptibility, to give an overall prediction for scattering from a 3-sublattice AFQ state. The dominant feature is the
presence of ‘Bragg peaks’ at the K- and K’-points. There is also a faint band where the gapped, spin-wave excitations dynamically induce a small, fluctuating quadrupole moment.

V. DISCUSSION AND CONCLUSIONS

Spin-nematic order remains an enigma. First proposed almost 40 years ago, and now studied in a wide range of theoretical models, it has never yet been unambiguously observed in experiment. Much of the difficulty in identifying a spin-nematic state arises from the fact that spin-nematic order does not break time-reversal symmetry. As a consequence, it cannot give rise to the internal magnetic fields measured by the common probes of static magnetic order — neutron scattering, NMR and muon spin rotation. In this respect, spin-nematic order has much in common with multipolar ‘hidden order’ phases in rare earth magnets. In principle, spin nematic order could be probed through its excitations. However, because of the complexity of the problem, these remain relatively poorly understood.

In this paper we have attempted to narrow the gap between theory and experiment, by constructing a continuum field theory of a three-sublattice antiferroquadrupolar (AFQ) spin-nematic state. This field theory offers a ‘model-independent’ approach to interpreting experiment, and can be used to explore the physical nature of the magnetic excitations of AFQ states. In the absence of magnetic field, we find that the long-wavelength excitations of AFQ states naturally divide into a set of three gapless, quadrupole-wave modes, — the Goldstone modes — together with three gapped excitations with a strong spin-dipole character.

This field theory can also be used to make concrete predictions for the fluctuating spin-dipole fields associated with each type of excitation, and its associated signature in experiment. In this paper we have focused on the most direct probe of spin-dipole fluctuations — inelastic neutron scattering. We find that quadrupole waves couple only weakly with neutrons, with the intensity of scattering vanishing linearly at low energies. However the gapped modes possess a substantial dipole moment and couple strongly to neutrons. The observation of this gapped excitation, together with a set of ghostly low-energy Goldstone modes, in the absence of magnetic Bragg peaks, would constitute strong evidence for AFQ spin-nematic order.

Finally we make predictions for the dynamical quadrupole susceptibility. This exhibits diverging Bragg-peak like intensity approaching the Goldstone modes, along with a very faint gapped mode. As in the f-electron systems, this may be measurable using resonant x-ray scattering. Such experiments would directly probe the order parameter, and could in consequence provide compelling evidence for the existence of spin-nematic order. How these excitations evolve with field, and what their consequences are for NMR $1/T_1$ relaxation rates will be explored in separate publications.

An obvious question for future work is the role of interactions. As in the case of FQ order, interactions between the Goldstone modes of the AFQ state endow these excitations with a finite, $k^2$-dependent lifetime. There is also a corresponding renormalisation of the director stiffness, $\rho_2$, leading to small changes in the velocity of the Goldstone modes. However, the most interesting features come from the interaction between the Goldstone modes and the gapped, long-wavelength “spin-wave” modes. This is true both from an experimental point of view, since the gapped modes support large spin-dipole fluctuations, and a theoretical point of view, where these type of interactions have not been as thoroughly explored as those between Goldstone modes. We will return to these effects in a future paper.

In conclusion, the SU(3) generalisation of the non-linear sigma model developed in this text provides a robust means of characterising spin-nematic states with antiferroquadrupolar order, which is independent of any particular microscopic model. This sigma model approach provides an excellent starting point for understanding the universal behaviour of spin-nematic states, and leads to concrete, testable predictions for experiment. For this reason, it can serve as an important tool for establishing whether spin-nematic order exists in a wide variety of real materials. We hope that the waves predicted by the sigma model will, in the near future, be seen.

Acknowledgments. We are grateful to Tsutomu Momoi for a number helpful comments on this work, and to Karlo Penc for a careful reading of the manuscript. This research was supported under EPSRC grants EP/C539974/1 and EP/G031460/1.

1. P. A. Lee, Science 321, 1306 (2008)
2. M. Blume and Y. Hsieh, J. Appl. Phys. 40, 1249 (1969)
3. H. Chen and P. Levy, Phys. Rev. Lett. 27, 1383 (1971)
4. A. F. Andreev and I. A. Grishchuk, Sov. Phys. JETP 60, 267 (1984)
5. N. Papaniicolou, Nucl. Phys. B305, 367 (1988)
6. P. Chandra and P. Coleman, Phys. Rev. Lett. 66, 100 (1991)
7. V. Barzykin, L. P. Gorkov and A. Sokol, EPL 15, 869 (1991)
8. V. Barzykin and L. P. Gorkov, Phys. Rev. Lett. 70, 2479 (1993)
9. K. Penc and A. Lauchli, Introduction to frustrated magnetism, chapter 13 (Springer-Verlag Berlin Heidelberg 2011)
10. A. V. Chubukov, J. Phys. Cond. Mat. 2, 4455 (1990)
A. Auerbach, *Interacting electrons and quantum magnetism* (Springer-Verlag New York 1994)

S. Chakravarty, B. I. Halperin and D. R. Nelson, Phys. Rev. B, 39, 2344 (1989)

S. Allen and D. Loss, Physica A 239, 47 (1997)

P. Azaria, B. Delamotte, F. Delduc and T. Jolicoeur, Nucl. Phys. B408, 485 (1993)

D. F. McMorrow, K. A. McEwen, U. Steigenberger, H. M. Ronnow and F. Yakhou, Phys. Rev. Lett. 87, 057201 (2001)

H. C. Walker, K. A. McEwen, D. F. McMorrow, S. B. Wilkins, F. Wastin, E. Colineau and D. Fort, Phys. Rev. Lett. 97, 137203 (2006)

H. C. Walker, K. A. McEwen, M. D. Le, L. Paolasini and D. Fort, J. Phys. Condens. Matter 20, 395221 (2008)

A. Smerald and N. Shannon, Phys. Rev. B 84, 184437 (2011)

A. Smerald and N. Shannon, arXiv:1303.4465