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1990 J. Phys.: Condens. Matter 2 9227

(http://iopscience.iop.org/0953-8984/2/46/023)

J. Phys.: Condens. Matter 2 (1990) 9227–9234. Printed in the UK

LETTER TO THE EDITOR

Triangular antiferromagnet as a biaxial quantum fluid

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Received 21 August 1990

Abstract. We apply a Schwinger boson scheme for quantum helimagnets to the two-dimensional triangular lattice Heisenberg antiferromagnet. Fluctuations are shown to lead to biaxial behaviour and the generation of an additional longitudinal Goldstone mode that appears as a pole in the longitudinal magnetic susceptibility.

The Heisenberg quantum antiferromagnet on a two-dimensional triangular lattice is the prototypical example of a frustrated antiferromagnet. Interest in this problem has been traditionally divided between the extreme quantum spin-$\frac{1}{2}$ and the large-S classical limits. At both extremes, spin fluctuations are endemic and profoundly modify the properties of the magnet. Classically, short wavelength thermal fluctuations drive a finite temperature topological phase transition associated with the unbinding of vortices in the (SO(3)) order parameter field [1]. In the extreme quantum limit of the spin-$\frac{1}{2}$ model, there has been a long-standing interest in the Anderson and Fazekas [2, 3] hypothesis, that short wavelength quantum fluctuations might destroy the ordered moment forming a resonating valence bond state with no ordered moment [2, 4]. Interest in the quantum limit has been recently revived by the possibility of a link with high temperature superconductivity [2–9].

Despite interest in these extreme limits of the triangular lattice Heisenberg model, there has been comparatively little effort to understand the properties of this model at intermediate S. EPR studies on quasi-two-dimensional triangular spin systems do indicate the presence of spin vortices at finite temperatures, suggesting that the order parameter is intrinsically biaxial in the quantum spin system [10]. In this letter, we discuss this development of biaxial order in the quantum triangular lattice model, and examine its consequences for the low-energy properties.

The classical triangular lattice antiferromagnet is a commensurate helimagnet, with three magnetic sublattices lying at 120° to each other in a plane. In the ground state, classical Heisenberg antiferromagnets are locally uniaxial, the order parameter being defined by a vector on the unit sphere (S^2), so that the longitudinal magnetic susceptibility is zero and in general there are two transverse Goldstone modes with different velocities, corresponding to slow rotations of the spins about axes perpendicular to the local magnetization.

In non-collinear helimagnets, zero-point fluctuations modify the properties qualitatively at finite spin S. Spin fluctuations in a non-collinear magnet are anisotropic

0953-8984/90/469227+08$03.50

about the local magnetization axis: the axial symmetry of the order parameter is thereby broken, driving it 'biaxial'. For helimagnets, we may consider the fluctuation component of the 'twist' between neighbouring spins in the triangular lattice spin system, which is written

$$\mathcal{T}_{ij} = \langle S_j \times S_i \rangle - \langle S_j \rangle \times \langle S_j \rangle \tag{1}$$

Quantum zero-point spin fluctuations generate a ground-state expectation value for this vector which lies normal to the plane of magnetization. Low-energy excitations about the ground state will distort the overall spin configuration, *maintaining the orthogonality* of the spin plane and the axis of the zero-point fluctuations. In this way, the character of the order parameter changes from S^2 to P^3 (SO(3)), where it is characterized by two orthogonal unit vectors.

There are several immediate qualitative consequences of the change in character of the order parameter. First, the long-wavelength longitudinal magnetic susceptibility becomes finite. Second, an additional 'longitudinal' Goldstone mode must appear [11], corresponding to slow rotations of the order parameter about the local magnetization axis. This mode will appear as a pole in the longitudinal magnetic susceptibility around $q = 0$. Finally, the topological properties of the order parameter are changed: for an SO(3) order parameter, free vortices with spin become topologically stable, whilst 'hedgehog' point-defect configurations that occur for S^2 order are no longer stable.

Application of spin-wave theory to the triangular magnet encounters divergences associated with this development of biaxial order. For a general helimagnet, a uniform field applied in the plane of the spins produces changes in the *magnitudes* of the local ordered moments, resulting in a longitudinal component to the uniform susceptibility. In spin-wave theory, to leading order in $1/S$, the uniform longitudinal susceptibility is determined by the density fluctuations of the Holstein–Primakoff bosons, at the magnetic wavevector

$$\chi_l = \langle |n(Q,\omega)|^2 \rangle_{\omega=0} = \frac{1}{4} \sum_q \frac{(u_{q+Q} v_q + u_q v_{q+Q})^2}{[\omega_q + \omega_{q+Q}]} \tag{2}$$

where ω_q are magnon frequencies and u_q and v_q are the Bogoliubov coefficients in the expansion of the magnon creation operators [12, 13]. For the triangular antiferromagnet, Goldstone modes in the spin-wave spectrum around $q = Q$ and $q = 2Q$ lead to a power-law infrared divergence $\chi_l \sim 1/\Lambda$, where Λ is the momentum cutoff around $q = Q$. A related divergence occurs in the the wavefunction renormalization constant of the magnons. These divergences are a symptom of the biaxial behaviour: to regularize them, a new energy scale associated with the deformation of the fluctuation twist axis relative to the magnetization axis must be introduced into the theory.

One of the more expedient methods that has recently been applied to the problem of low dimensional antiferromagnets, is the Schwinger boson technique, developed by Arovas and Auerbach [14], which describes strongly fluctuating magnets in terms of an incompressible superfluid of spins, where a classical spin condensate is surrounded by a normal fluid of spin fluctuations. A related method has been developed by Takahashi [15, 16]. Chandra, Coleman and Larkin [17] (hereafter referred to as CCL) have extended this approach to encompass biaxial behaviour through the introduction of a triplet spin pairing in the fluid of spin fluctuations. In this letter, we study the finite, but large-S triangular lattice Heisenberg model by using the quantum fluids approach.

Following CCL we imagine arbitrarily twisting the spin reference frame introducing a 'fictitious' twist vector potential $\boldsymbol{A}_l$, $(l = 1, 2)$ into the Heisenberg model

$$H = \frac{1}{2}\sum_{ij} J(\boldsymbol{R}_{ij})\boldsymbol{S}_i \exp\left(-\int_j^i \boldsymbol{A}_l \, \mathrm{d}R_l \times\right)\boldsymbol{S}_j - \sum_j \boldsymbol{B}_j \cdot \boldsymbol{S}_j \tag{3}$$

A uniform twist vector potential $\boldsymbol{A}_l = Q_l\hat{\boldsymbol{k}}$ is equivalent to a uniformly twisted spin coordinate axis or alternatively, twisted boundary conditions, with a twist of angle (Q_xL_x, Q_yL_y) about the $\hat{\boldsymbol{k}}$ axis in the x and y directions. The stiffnesses of the broken symmetry ground state are found by computing the second derivatives of the free energy with respect to the twist, in a manner reminiscent of the Abrikosov–Gorkov computation of superfluid density in a superconductor.

Let $\hat{\boldsymbol{x}} = \hat{\boldsymbol{\imath}}$ and $\hat{y} = \frac{1}{2}\hat{\boldsymbol{\imath}} + \frac{1}{2}\sqrt{3}\hat{\boldsymbol{\jmath}}$ be the Bravais lattice vectors of the triangular lattice, then in a reciprocal lattice basis the Fourier transform of $J(\boldsymbol{R})$ is

$$J_{\boldsymbol{q}} = 2J\left[c_x + c_y + c_{x-y}\right] \tag{4}$$

where $c_l = \cos q_l$ $(l = 1, 2)$ and $c_{x-y} = \cos(q_x - q_y)$. Following CCL, we transform to a twisted reference frame where the configuration of spins S^g is locally ferromagnetic, so $\langle \boldsymbol{S}_i^g \times \boldsymbol{S}_j^g \rangle = 0$. The transformed Hamiltonian is formally unchanged, but the spin vector potential is now $\boldsymbol{A}_l = Q_l\hat{\boldsymbol{z}}$. Next we employ the Schwinger boson spin representation ($\boldsymbol{S}_i = \frac{1}{2}b^\dagger{}_\sigma \boldsymbol{\sigma}_{\sigma\sigma'} b_{i\sigma'}$, $n_b(i) = 2S$), and decouple the twisted Heisenberg model entirely in terms of even parity Cooper and particle-hole pairs. At a mean field level, this is equivalent to discussing a BCS pairing Hamiltonian

$$H_{\mathrm{BCS}} = \frac{1}{4}\sum_{\boldsymbol{q}\boldsymbol{q}'}[\mathcal{J}^+_{\boldsymbol{q}\boldsymbol{q}'} D_{\boldsymbol{q}}{}^\dagger D_{\boldsymbol{q}'} - \mathcal{J}^-_{\boldsymbol{q}\boldsymbol{q}'} B^\dagger_{\boldsymbol{q}} B_{\boldsymbol{q}'}] - \frac{1}{2}NJ(\boldsymbol{Q})S^2 \tag{5}$$

where $B^\dagger_{\boldsymbol{q}} = b^\dagger{}_{q\sigma} b^\dagger{}_{-q-\sigma}$ and $D^\dagger_{\boldsymbol{q}} = b^\dagger{}_{q\sigma} b_{q\sigma}$ are triplet Cooper and particle–hole pairing fields for the Schwinger bosons. In triangular antiferromagnets, the high degree of symmetry in the lattice preferentially selects a uniform twist with $\boldsymbol{Q} = \pm\frac{2}{3}\pi(1, 2)$, producing a twist of 120° between neighbouring spins. In this case the pairing potentials simplify and become

$$(\mathcal{J}^+_{\boldsymbol{q}\boldsymbol{q}'}, \mathcal{J}^-_{\boldsymbol{q}\boldsymbol{q}'}) = \tfrac{1}{4}(1, 3)J_{\boldsymbol{q}+\boldsymbol{q}'} \tag{6}$$

A mean-field decoupling of this Hamiltonian then yields

$$H_{\mathrm{mf}} = \sum_{\boldsymbol{q}}\{h_{\boldsymbol{q}} b^\dagger{}_{q\sigma} b_{q\sigma} - [\Delta_{\boldsymbol{q}} b^\dagger{}_{q\uparrow} b^\dagger{}_{-q\downarrow} + \mathrm{HC}]\} \tag{7}$$

where the mean field parameters are self-consistently determined through the equations

$$\begin{aligned} h_{\boldsymbol{q}} + \lambda &= \int_{\boldsymbol{q}'} \mathcal{J}^+_{\boldsymbol{q}\boldsymbol{q}'} \alpha_{\boldsymbol{q}'} \\ \Delta_{\boldsymbol{q}} &= \int_{\boldsymbol{q}'} \mathcal{J}^-_{\boldsymbol{q}\boldsymbol{q}'} \eta_{\boldsymbol{q}'} \\ S + \frac{1}{2} &= \int_{\boldsymbol{q}} \alpha_{\boldsymbol{q}}. \end{aligned} \tag{8}$$

Here $\int_q \equiv \int d^2q/(2\pi)^2$, and

$$(2\alpha_q, 2\eta_q) = [\coth(\beta\omega_q/2)/\omega_q](h_q, \Delta_q). \tag{9}$$

The mean field spin quanta have energy $\omega_q = \left(h_q^2 - \Delta_q^2\right)^{1/2}$. The corresponding ground-state wavefunction is an RVB wavefunction of triplet pairs, which when written in the untwisted reference frame contains an admixture of both singlet and triplet valence bonds

$$|\Psi\rangle = P_{2S} \exp\left[\sum_q f_q b^\dagger{}_{q+Q/2\uparrow} b^\dagger{}_{-q-Q/2\downarrow}\right] |0\rangle. \tag{10}$$

Here $f_q = v_q/u_q$ is a symmetric function of q, with $2u_q^2 = [h_q/\omega_q + 1]$ and $2u_q v_q = \Delta_q/\omega_q$. P_{2S} projects the component of the wavefunction with $2S$ spin quanta per site. Equation (10) generalizes the Liang–Doucot–Anderson wavefunction to the triangular lattice [18].

In the large-S limit at zero temperature the bosons condense at $q = 0$, so $\alpha_q \sim \eta_q \sim S^*\delta_q$ giving the classical ordered ground state with magnetization S^*. In this limit, the equations for h_q and Δ_q are saturated by the contribution from the spin condensate, so

$$(h_q + \lambda, \Delta_q) = S^*(\mathcal{J}_{q0}^+, \mathcal{J}_{q0}^-) = (1, 3)\frac{S^* J_q}{4}. \tag{11}$$

The constraint field is set by the condition that $q = 0$ is a Goldstone mode, which then yields $\lambda = S^* J_Q = -3S^* J$, so that

$$\omega_q^2 = \frac{(S^*)^2}{2}[6J - J_q][J_q + 3J] \tag{12}$$

which is the large-S spin-wave spectrum. A first approximation to the critical spin $S_c \sim S - S^*$ is obtained by substituting the large-S values of h_q and Δ_q into the constraint equation (8), giving $S_c \sim 0.258$ in agreement with spin-wave theory results [19], consistent with an ordered ground state for any spin. (Iteration of the mean field equations to full self-consistency only includes some of the higher order corrections in $1/S$, but also gives $S_c < \frac{1}{2}$). At low temperatures solution of the constraint equation gives an accumulation of bosons around $q = 0$, a small gap Δ_0 and a coherence length ξ_0 given by

$$\xi_0 = c_0/\Delta_0 = \frac{2T}{3JS} \exp 2\pi JS^2/T \tag{13}$$

where c_0 is the spin-wave velocity.

The fluctuation corrections to the spin-wave spectrum are calculated to leading order in $1/S$ by iteration of the pairing equations (8), adjusting λ to satisfy the constraint equation. The most striking change in the spin-wave spectrum is the development of a gap

$$(\Delta_Q)^2 = \left(\frac{3}{4}\right)^3 S \int_q (-JJ_q)\left[\frac{2(6J - J_q)}{(J_q + 3J)}\right]^{1/2} \tag{14}$$

in the excitation spectrum at $\boldsymbol{q} = \pm\boldsymbol{Q}$. This gap is a signal of biaxial character, and can be identified as the characteristic frequency of the breathing mode in which the fluctuation component of the twist $\boldsymbol{\mathcal{T}}$ and $\langle \boldsymbol{S}(x)\rangle \times \langle \boldsymbol{S}(y)\rangle$ rotate in opposite directions. At low temperatures $\Delta_{\boldsymbol{Q}} \sim 1.50J\sqrt{S}$, whilst at high temperatures $\Delta_{\boldsymbol{Q}} \sim \sqrt{JT}$. The crossover from $\sqrt{S}$- to $\sqrt{T}$-dependence is typical of this kind of quantum exchange mode, where the restoring force is derived from zero-point quantum fluctuations at low temperatures, and from the entropy of thermal fluctuations at high temperatures [20]. The healing length $\xi_{\boldsymbol{Q}} = \Delta_{\boldsymbol{Q}}/c_{\boldsymbol{Q}}$ for these out-of-phase fluctuations is only a few lattice spacings (figure 1). In the large-S limit the Goldstone mode associated with slow rotations of the plane of magnetization appears as two gapless modes in the magnon spectrum at $\boldsymbol{q} = \pm\boldsymbol{Q}$. Once fluctuations are included, the ordered moment magnetization axis and the anisotropic fluid of spin excitations become locked together, so this Goldstone mode at finite S becomes a collective mode associated with slow rotations of the triplet pairing field. In spin-wave theory, this phenomenon is manifested by the development of spin-wave bound states [21, 22]. A calculation of the residue $Z_{\boldsymbol{q}}$ of this Goldstone mode in the one magnon channel reveals that it vanishes at finite S. In the spin wave language, the long-wavelength excitations about $\boldsymbol{q} = \boldsymbol{Q}$ are entirely composed of bound-magnon pairs [13].

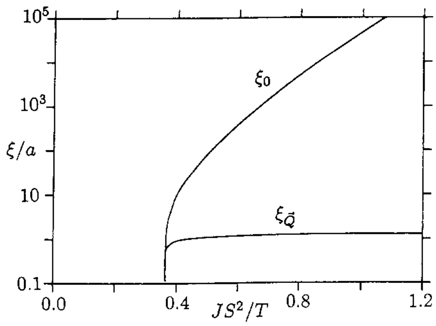

Figure 1. Coherence lengths ξ_0 and $\xi_{\boldsymbol{Q}}$ against JS^2/T for $S = \frac{1}{2}$.

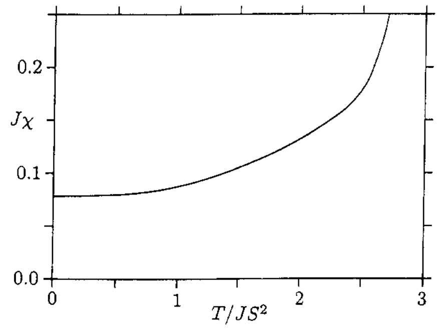

Figure 2. Uniform susceptibility against T/JS^2 for $S = \frac{1}{2}$.

The spin susceptibility may be calculated from the propagators of the mean field spin quanta, as is described by CCL. We find

$$\chi_3 = \frac{1}{2T}\sum_{\boldsymbol{q}} \operatorname{cosech}^2 \frac{\omega_{\boldsymbol{q}}}{2T}$$

for static the susceptibility parallel to the twist axis and

$$\chi_\perp = \frac{1}{4}\sum_{\boldsymbol{q}}\left[\frac{(u_+v_- + u_-v_+)^2 n[\omega_+]n[\omega_-]}{n[\omega_+ + \omega_-](\omega_+ + \omega_-)} - \frac{(u_+u_- + v_-v_+)^2 n[\omega_+]n[-\omega_-]}{n[\omega_+ - \omega_-](\omega_+ - \omega_-)}\right] \tag{15}$$

for the average susceptibility $\chi_\perp = \frac{1}{2}(\chi_1 + \chi_2)$ perpendicular to the twist axis.Here $\pm \equiv \boldsymbol{q} \pm \boldsymbol{Q}/2$, and $n(\omega)$ is the Bose function. At $T = 0$ and large S, this reduces to the isotropic classical susceptibility. Unlike a uniaxial magnet, the fluid of spin fluctuations is itself anisotropic, and there is a fluctuation corrections $\delta\chi$ to this susceptibility at finite S associated with the moment of of inertia of this fluid. These quantities are calculated from appropriate RPA polarizabilities of the spin quanta, and we find that $\delta\chi_3 = 0$, whilst $\delta\chi_\perp \neq 0$, confirming that the spin fluctuations are axially symmetric about the normal to the spin plane. At finite temperatures only the average of the susceptibilities is observed due to the lack of long range order. A numerical calculation of $\chi(T)$ for $S = \frac{1}{2}$ is shown in figure 2.

Next we consider the long-wavelength behaviour of the triangular antiferromagnet. At finite temperatures, the co-ordinate system of the twisted reference frame experiences slow distortions which we may study by deriving the long wavelength action at low temperatures. The magnetization $\hat{\boldsymbol{S}}$ and twist $\hat{\boldsymbol{k}}$ define an SO(3) order parameter with orthogonal axes $(\hat{\boldsymbol{e}}_1, \hat{\boldsymbol{e}}_2, \hat{\boldsymbol{e}}_3) = (\hat{\boldsymbol{S}}, \hat{\boldsymbol{k}} \times \hat{\boldsymbol{S}}, \hat{\boldsymbol{k}})$. In general, these vectors precess in space according to $\nabla_l \hat{\boldsymbol{e}}_\lambda = (\boldsymbol{\omega}_l + Q_l \hat{\boldsymbol{e}}_3) \times \hat{\boldsymbol{e}}_\lambda$ where the $\boldsymbol{\omega}_l = 0$ in the ground state. Quite generally, we may write the long-wavelength action

$$I = \frac{1}{2} \int \mathrm{d}^2x\, \mathrm{d}t \{\gamma_\lambda((\omega_1^\lambda)^2 + (\omega_2^\lambda)^2) - \chi_\lambda(\omega_0^\lambda)^2\} \tag{16}$$

where the spacetime precession vectors $\boldsymbol{\omega}_\mu = (\boldsymbol{\omega}_0, \boldsymbol{\omega}_l)$, $(l = 1,2)$ are resolved along the local principal axes $\boldsymbol{\omega}_\mu = \omega_\mu^\lambda \hat{e}^\lambda$. The velocities of the associated Goldstone modes are $(c^\lambda)^2 = \gamma^\lambda / \chi_l^\lambda$. From the rotational invariance of the problem, the change in the long wavelength action due to a slow variation of the order parameter is precisely the change induced by the application of a small external magnetic or twist vector potential field $(\delta B, \delta \boldsymbol{A}_l) = \boldsymbol{\omega}_\mu$. We may determine these coefficients by studying the linear spin current response of the spin fluid to an external spin vector potential $\boldsymbol{\mathcal{R}}_l = \gamma_l\, \delta \boldsymbol{A}_l$. For technical details we refer to CCL. Table 1 summarizes our results.

Table 1. Stiffnesses and susceptibilities for the quantum triangular magnet, computed in the quantum fluids approach.

	1: Longitudinal	2: Transverse	3: Transverse
χ	$\dfrac{0.017}{J\sqrt{S}}$	$\dfrac{2}{9J} + O(\frac{1}{\sqrt{S}})$	$\dfrac{1}{9J} + O(\frac{1}{S})$
γ	$0.080JS$	$\dfrac{3JS^2}{4} + O(S)$	$\dfrac{3JS^2}{4} + O(S)$
c	$2.17JS^{3/4}$	$1.84JS$	$2.60JS$

As expected, in the large-S limit the the longitudinal susceptibility and stiffness are zero. We find $\chi_3 = \frac{1}{2}\chi_2$, corresponding to an isotropic magnetic susceptibility in the untwisted reference frame, as obtained in previous studies [7]. For finite S the longitudinal stiffness is finite, and the system develops a longitudinal Goldstone mode with velocity $c^{(1)} = 2.17\, JS^{3/4}$. The non-analytic dependence of the velocity on S reflects its fluctuation origins. In the vicinity of the magnetic Bragg peaks, neutron scattering will now observe three distinct poles in the scattering intensity, given approximately by

$$\chi''(\boldsymbol{q}, \omega) \sim \sum_{\lambda=1,3} \left(\frac{f_q^\lambda}{\chi_\lambda \omega}\right) \delta(\omega - c^\lambda q). \tag{17}$$

Here the form factor $f_q^{\lambda} = 1$ for the transverse modes ($\lambda = 2, 3$), but is $f_q^1 = (\gamma_3 q)^2$ for the longitudinal mode since the longitudinal magnetization depends on derivatives of the twist ($M_1 \sim \hat{\chi}_1 T \times \partial_t \hat{T}$). The presence of the additional pole in the longitudinal spin susceptibility provides an unambiguous experimental test for the development of biaxial order.

Let us briefly consider the finite-temperature behaviour of the quantum triangular magnet, and in particular its relationship to the topological phase transition. In addition to smooth variations in the spin reference frame and the associated vector potential $\boldsymbol{A}_l$, the biaxial nature of the magnet permits the introduction of stable 'Z_2 vortices' into the order parameter field [1, 23, 24]. In the locally ferromagnetic spin reference frame, the phase integral of the spin vector potential around a Z_2 vortex precesses through 2π

$$\oint \boldsymbol{A}_l \, \mathrm{d}R_l = 2\pi\hat{v}. \tag{18}$$

These vortices carry a net moment S, and can be loosely regarded as unbound spins. Rotational invariance enables two Z_2 vortices to be adiabatically deformed into a singlet, thereby annihilating one-another. The free energy of an isolated vortex is $F \sim (JS^2 - 2T)\ln[\xi(T)/a]$ which may be neglected until $T \sim JS^2$. Fortunately, this is also the temperature where $\Delta_Q \sim T$ and spin waves unbind, destroying the biaxial order, so our assumption of a smoothly varying twist vector potential is consistent with the topological picture of SO(3) order.

Finally, we should like to discuss the possibilities of 'disordered' magnetic ground states on a triangular lattice. In the case of a square lattice Heisenberg model, increasing the fluctuations is thought to cause a topological transition into a dimer state, via constructive interference between 'hedgehog' instanton configurations of the order parameter [25, 26]. For spiral magnets, there are no stable point defects in $2+1$ dimensions, ($\pi_2(SO(3)) = 0$) and it would seem that the analogous phase transition into a dimer is ruled out. An interesting alternative is the formation of a 'spin nematic' [27], where the magnetisation drops to zero, but the twist axis, the associated stiffness and topological character are sustained by the continued condensation of magnon pairs. One way to suppress the magnetization is to add next-nearest neighbour interactions. On the 2-dimensional square lattice Heisenberg antiferromagnet, once $J_2/J_1 \sim \frac{1}{2}$, the Goldstone mode spectrum becomes quadratic and the magnetisation drops to zero [28, 29]. In the triangular lattice this approach fails, for the softening of the Goldstone modes and a divergence in the magnetic fluctuations is pre-empted by a first order phase transition into a collinear magnetic state [30, 31]. A more successful approach is to remove one quarter of the sites from the lattice to form a tessellated array of hexagons and triangles called the Kagome lattice. This model has no classically ordered moment, but may nevertheless display a topological SO(3) phase transition. The study of this system is currently of great experimental and theoretical interest [13, 32, 33].

In conclusion, we have shown how quantum fluids treatment of the triangular lattice antiferromagnet helps to elucidate the fluctuation driven biaxial character of the quantum triangular magnet. In particular, this leads to the development of quantum exchange gaps, which we have interpreted in terms of the development of a biaxial order parameter, and a longitudinal Goldstone mode.

We are grateful to P Chandra for a number of stimulating discussions. Part of this work was supported by NSF grants DMR-89-13692 and PHY-82-17853, supplemented by funds from the Sloan Foundation. IR would like to thank Rutgers University where much of this work was carried out whilst visiting, and to acknowledge the support of the SERC and Trinity College, Cambridge. PC would like to thank Cambridge University where the final stages of this work were carried out.

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