Quantum Spin Nematics: Moment-Free Magnetism

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Treating antiferromagnetism as a spin superfluid, we discuss the possibility of moment-free magnetism. Specifically, we show that large quantum fluctuations induce an anisotropy in Heisenberg spiral structures, driving a biaxial-uniaxial transition to a spin nematic; this state is characterized by *tensor* spin order and a gapless Goldstone mode of spin-pair excitations. Experimental signatures and realizations of this phenomena are suggested.

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The essence of broken rotational symmetry in Heisenberg systems is a finite spin stiffness; specifically, the development of a local moment is not a necessary condition for spin superfluidity. The 1D half-integer Heisenberg spin chain, a superfluid spin system at criticality, provides a well-known example.[1] In this paper we discuss another "moment-free magnet," the spin nematic, characterized by long-range order in the tensor order parameter

$$Q^{\alpha\beta}(x,x') = \langle S^\alpha(x) S^\beta(x')\rangle - \tfrac{1}{3}\delta^{\alpha\beta}\langle \mathbf{S}(x)\cdot\mathbf{S}(x')\rangle . \qquad (1)$$

Unlike conventional magnets, the low-lying excitations of a spin nematic are tensor in character and correspond to bound states of spin waves. Motivated by recent studies of charge-induced magnetic incommensuration in doped Mott insulators, we examine the formation of a spin nematic in strongly fluctuating spiral spin structures.

Spin-nematic behavior was first introduced to explain anomalous transitions in magnetic pnictides[2] where quadrupolar order is driven by biquadratic[3] interactions $H' = J(\mathbf{S}_i\cdot\mathbf{S}_j)^2$. Andreev and Grischuk have postulated a second "p-type" spin nematic where the director $\hat{\mathcal{T}}$ is a pseudovector associated with a twist of the underlying spin configurations:[4]

$$\mathcal{T}(x,x') = \langle \delta\mathbf{S}(x)\times\delta\mathbf{S}(x')\rangle = F(x-x')\hat{\mathcal{T}} . \qquad (2)$$

Such anisotropic fluctuations are present in a quantum helimagnet; here, additional symmetry breaking leads to the development of an incommensurate magnetization in the plane perpendicular to $\hat{\mathcal{T}}$, and the system becomes biaxial. Technically, the twist in a helimagnet contains a "condensate" contribution from the ordered moments,

$$\langle \mathbf{S}(x)\times\mathbf{S}(x')\rangle = \mathcal{T}(x,x') + \langle \mathbf{S}(x)\rangle\times\langle \mathbf{S}(x')\rangle , \qquad (3)$$

but only the first term is an independent order parameter; specifically, $\hat{\mathcal{T}}$ is responsible for the *longitudinal* susceptibilities and stiffnesses associated with biaxial behavior in a helimagnet.[5,6] In the presence of magnetic order, fluctuations in the twist director are constrained to lie in a plane perpendicular to the magnetization; in two dimensions, the resulting x-y-like behavior in $\hat{\mathcal{T}}$ leads to suppression of the fluctuation renormalizations and to a finite-temperature SO(3) vortex transition.[7] In this paper we examine the effect of quantum fluctuations at zero temperature in frustrated, two-dimensional quantum helimagnets. We argue that the fluctuation contributions to the twist make it robust against the loss of an ordered moment; melting of a biaxial quantum helimagnet to an isotropic spin fluid thereby occurs in two stages, via the intermediate formation of a uniaxial p-type spin nematic (see Fig. 1).

The essence of our approach is a quantum-fluid treatment of magnetism in two-dimensional frustrated Heisenberg models.[8-10] From this perspective, antiferromagnetism corresponds to a spin condensate surrounded by a normal fluid of spin fluctuations. In a spin nematic, *spin-pair* condensation occurs in the absence of a local moment. To examine the development of such phases we imagine arbitrarily twisting the spin reference frame, introducing a "fictitious" twist vector potential $\mathbf{A}_l$ ($l=1,2$) into the Heisenberg model,

$$H = \tfrac{1}{2}\sum J(\mathbf{R}_{ij})\mathbf{S}_i\cdot\exp\left[-\int_j^i \mathbf{A}_l\, dR_l\right]\times\mathbf{S}_j - \sum_j \mathbf{B}_j\cdot\mathbf{S}_j . \qquad (4)$$

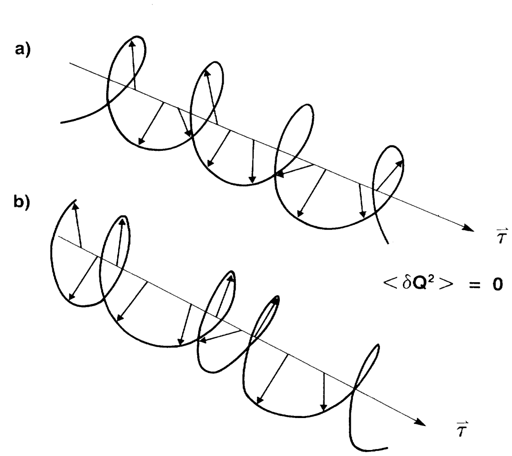

FIG. 1. Pictorial representation of (a) helimagnet and (b) twisted spin nematic.

A uniform twist vector potential $\mathbf{A}_l = Q_l \hat{\mathbf{k}}$ is equivalent to a uniformly twisted spin coordinate axis or, alternatively, twisted boundary conditions with a twist of angle $(Q_x L_x, Q_y L_y)$ about the $\hat{\mathbf{k}}$ axis in the x and y directions. The stiffnesses of the broken-symmetry ground state are found by computing the second derivative of the free energy with respect to the twist, in a manner reminiscent of the Abrikosov-Gorkov computation of superfluid density in a superconductor. We parametrize the frustrating interactions in terms of the Fourier transform of the bond strengths

$$J(\mathbf{q}) = 2J_1(c_x + c_y) + 4J_2(c_x c_y) + 2J_3(c_{2x} + c_{2y}) + \cdots , \quad (5)$$

where $c_l = \cos(q_l a)$ $(l = x, y)$ and J_1, J_2, J_3 are first-, second-, and third-nearest-neighbor couplings, respectively.

Following Arovas and Auerbach[8] we adopt a Schwinger boson representation, whereby each spin $\mathbf{S}_i = \frac{1}{2} b^\dagger_{i\sigma} \boldsymbol{\sigma}_{\sigma\sigma'} b_{i\sigma'}$ is represented as an "incompressible" ensemble of $2S$ spin quanta $b^\dagger_{i\sigma} b_{i\sigma} = 2S$. We assume that the ground state can be characterized by a simple uniform twist and transform into a twisted reference frame where spin correlations are of even parity and the twist vanishes. Setting $\mathbf{A}_l = Q_l \hat{\mathbf{k}}$, in the twisted reference frame the Hamiltonian must then be decoupled in terms of even-parity pairing fields; at the mean-field level this is equivalent to the BCS pairing Hamiltonian

$$H_{\rm BCS} = \frac{1}{4} \sum_{\mathbf{q}\mathbf{q}'} [\mathcal{J}^+_{\mathbf{q}\mathbf{q}'} D^\dagger_{\mathbf{q}} D_{\mathbf{q}'} - \mathcal{J}^-_{\mathbf{q}\mathbf{q}'} B^{(t)\dagger}_{\mathbf{q}} B^{(t)}_{\mathbf{q}'}] + \sum_{\mathbf{q}\sigma} \lambda (b^\dagger_{\mathbf{q}\sigma} b_{\mathbf{q}\sigma} - 2S) , \quad (6)$$

where $\mathbf{B}^\dagger_{\mathbf{q}} = \sum_\sigma b^\dagger_{\mathbf{q}\sigma} b^\dagger_{\mathbf{q}-\sigma}$ and $D^\dagger_{\mathbf{q}} = \sum b^\dagger_{\mathbf{q}\sigma} b_{\mathbf{q}\sigma}$ are the Cooper and particle-hole pairing fields, respectively, N is the number of sites, and

$$\mathcal{J}^\pm_{\mathbf{q}\mathbf{q}'} = \tfrac{1}{2} \{ J(\mathbf{q}+\mathbf{q}') \pm \tfrac{1}{2} [J(\mathbf{q}+\mathbf{q}'+\mathbf{Q}) + J(\mathbf{q}+\mathbf{q}'-\mathbf{Q})] \}_S \quad (7)$$

are twist-dependent pairing potentials. (The subscript S denotes symmetrization with respect to q and q'.) In particular, the twist may be energetically selected so as to maximize the attractive pairing interactions between the spin quanta. The last term in (6) is the constraint; physically, it is the local Onsager correction to the Weiss field caused by strong spin fluctuations.[10,11] A mean-field decoupling of this Hamiltonian then yields

$$H_{\rm MF} = \sum_{\mathbf{q}} \{ (h_{\mathbf{q}} - \lambda) [b^\dagger_{\mathbf{q}\uparrow} b_{\mathbf{q}\uparrow} + b_{-\mathbf{q}\downarrow} b^\dagger_{-\mathbf{q}\downarrow}] - [\Delta_{\mathbf{q}} b^\dagger_{\mathbf{q}\uparrow} b^\dagger_{-\mathbf{q}\downarrow} + \mathrm{H.c.}] \} , \quad (8)$$

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

$$h_{\mathbf{q}} = \int_{\mathbf{q}'} \mathcal{J}^+_{\mathbf{q}\mathbf{q}'} \alpha_{\mathbf{q}'}, \quad \Delta_{\mathbf{q}} = \int_{\mathbf{q}'} \mathcal{J}^-_{\mathbf{q}\mathbf{q}'} \eta_{\mathbf{q}'}, \quad S + \tfrac{1}{2} = \int_{\mathbf{q}} \alpha_{\mathbf{q}} , \quad (9)$$

with $\int_{\mathbf{q}} \equiv \int d^2 q/(2\pi)^2$ and

$$(2\alpha_{\mathbf{q}}, 2\eta_{\mathbf{q}}) = (\langle D^\dagger_{\mathbf{q}} \rangle, \langle B^{(t)\dagger}_{\mathbf{q}} \rangle) = [\coth(\tfrac{1}{2}\beta\omega_{\mathbf{q}})/\omega_{\mathbf{q}}] (\tilde{h}_{\mathbf{q}}, \Delta_{\mathbf{q}}) . \quad (10)$$

The twist vector is determined by minimizing the total energy with respect to $\mathbf{Q}$. The spectrum of the corresponding Bogoliubov quasiparticles $a^\dagger_{\mathbf{q}\sigma} = u_{\mathbf{q}} b^\dagger_{\mathbf{q}\sigma} - v_{\mathbf{q}} b_{-\mathbf{q}-\sigma}$ is then $\omega^2_{\mathbf{q}} = (h_{\mathbf{q}} - \lambda)^2 - \Delta^2_{\mathbf{q}}$; in the untwisted reference frame, they annihilate a twisted resonating-valence-bond (RVB) ground-state wave function

$$|\Psi\rangle = P_{2S} \hat{g} \exp\left[\sum_{\mathbf{q}} (v_{\mathbf{q}}/u_{\mathbf{q}}) b^\dagger_{\mathbf{q}\uparrow} b^\dagger_{-\mathbf{q}\downarrow} \right] |0\rangle , \quad (11)$$

where P_{2S} is the Gutzwiller projector and $\hat{g} = \exp[i \sum_j \mathbf{Q} \cdot \mathbf{R}_j S^z_j]$ is a twist operator. For the special case of collinear spin correlations where $2Q \equiv 0$ and $(u_{\mathbf{q}}, v_{\mathbf{q}}) = (u_{\mathbf{q}+\mathbf{Q}}, -v_{\mathbf{q}+\mathbf{Q}})$, it is straightforward to show that (11) becomes a singlet RVB wave function.[12]

In the large-S limit the spin quanta condense at $\mathbf{q} = 0$ in the twisted reference frame and the mean-field theory recovers the transverse-spin-wave dispersion of the classical helimagnet,[13]

$$\omega^2_{\mathbf{q}} = S^2 \{ [\mathcal{J}^+_{\mathbf{q}0} - J(\mathbf{Q})]^2 - [\mathcal{J}^-_{\mathbf{q}0}]^2 \} . \quad (12)$$

There are two types of transverse Goldstone modes at $\mathbf{q} = 0$ and $\mathbf{q} = \pm\mathbf{Q}$, respectively, corresponding to rotations of the local magnetization about the twist axis and about an axis perpendicular to the twist and local magnetization. At finite S, the twist of the spins develops a fluctuation component $\langle \delta\mathbf{S}(x) \times \delta\mathbf{S}(x') \rangle = F(x - x') \hat{\mathbf{k}}$, where

$$F(x) = \tfrac{1}{4} \sin[\mathbf{Q}\cdot\mathbf{x}] \int_{\mathbf{q}} [u_+ v_- + v_+ u_-]^2 e^{i\mathbf{q}\cdot\mathbf{x}} \quad (\pm \equiv \mathbf{q} \pm \mathbf{Q}/2) . \quad (13)$$

Similar results are obtained in spin-wave theory. Fluctuations of the twist director and magnetization away from orthogonality are suppressed and are found to give rise to a pairing gap $\Delta_{\mathbf{Q}} \sim \sqrt{S}$ at $\pm\mathbf{Q}$ in the mean-field spectrum, indicating the formation of spin-wave triplet bound pairs of size $\sim c/\Delta_{\mathbf{Q}}$. Slow rotations of the twist director about the local magnetization axes now give rise to an additional "longitudinal" Goldstone mode, which appears in this approach as a collective mode of the twisted spin condensate.

For small $S < S_c$, where $S_c + \frac{1}{2} = \int_{\mathbf{q}} h_{\mathbf{q}}/2\omega_{\mathbf{q}}$, spin fluctuations become too large to sustain an ordered moment; the spin quanta no longer individually condense. A gap $\Delta_0 \sim c/\xi_0$ develops in the spin-wave spectrum, where ξ_0 is the finite spin-correlation length and c is a typical spin-wave velocity. In this phase, the long-range twist is sustained by the correlated spin fluctuations of the normal fluid. Equation (11) then describes a p-type spin nematic; in the absence of a local moment the twist operator $\mathcal{T}_Y(X) = \mathbf{S}(X + Y/2) \times \mathbf{S}(X - Y/2)$ displays long-range order,

$$\langle \mathcal{T}_Y(X)\cdot\mathcal{T}_Y(0)\rangle \rightarrow |F(Y)|^2 . \quad (14)$$

This state may be visualized as a helimagnet where quantum fluctuations in the pitch dephase the spins, leading to a distribution of magnetic wave vectors with variance $\langle dQ^2\rangle = \xi^{-2}$. At length scales longer than the spin-correlation length, the residual transverse fluctuations of the uniaxial twist director are described by an O(3) σ model,

$$I = \tfrac{1}{2}\chi\int d^2x\, dt\{(c_l\nabla_l\hat{\mathcal{T}})^2 - (\partial_t\hat{\mathcal{T}})^2\}, \quad (15)$$

where $\hat{\mathcal{T}}(x)$ is the twist director, and the twist wave velocity c_l is determined by the ratio of the spin-wave stiffness to the susceptibility $(c_l)^2 = \gamma^l/\chi$.

Using our microscopic calculation, we can compute the zero-temperature susceptibility and the "London kernel" relating spin currents $\mathcal{J}^l(x) = -\partial H/\partial \mathbf{A}^l(x)$ to the long-wavelength twist: $\mathcal{J}^l(x) = \gamma^l(x-x')\mathbf{A}_l(x)$. The current response kernel $\gamma^l_{\alpha\beta}(x) \equiv [\gamma^l(x)]_{\alpha\beta}$ can be divided into a "diamagnetic" spin-wave stiffness $\mathcal{N}^l$ and a "paramagnetic," or fluctuation, correction $\tilde{\gamma}^l(x-x') = \langle \mathrm{T}\mathcal{J}^l(x)\mathcal{J}^l(x')\rangle$ (where T denotes time ordering),

$$\gamma^l(x) = \mathcal{N}^l\delta^3(\mathbf{x}) - \tilde{\gamma}^l(x), \quad (16)$$

where

$$\mathcal{N}^l_{\alpha\beta} = \tfrac{1}{2}\sum_{\mathbf{R}} J(\mathbf{R})(R_l)^2[\langle S^\alpha(0)S^\beta(\mathbf{R})\rangle - \delta^{\alpha\beta}\langle \mathbf{S}(0)\cdot\mathbf{S}(\mathbf{R})\rangle] \quad (17)$$

is the normal component of the stiffness. In a paramagnet these two components cancel. In the spin nematic, spin pairing generates anisotropy, and although the stiffness is still zero about the twist axis, the cancellation is incomplete for components of the stiffness about axes perpendicular to the twist. To compute the spin current response to an external twist we express the paramagnetic spin currents in terms of the spin quasiparticles. Calculating the diamagnetic stiffness and the one-loop fluctuations in the spin currents, the stiffness about an axis perpendicular to the twist is

$$\gamma^l = \tfrac{1}{2}\sum_{\mathbf{k}}(\alpha_{\mathbf{k}}\nabla^2_{\mathbf{k}}h^*_{\mathbf{k}} - \eta_{\mathbf{k}}\nabla^2_{\mathbf{k}}\Delta^*_{\mathbf{k}}) - \tilde{\gamma}^l_\perp , \quad (18)$$

where $h^*_{\mathbf{k}} = \frac{1}{4}(2h_{\mathbf{k}} + h_{\mathbf{k+Q}} + h_{\mathbf{k-Q}})$ and

$$\Delta^*_{\mathbf{k}} = \tfrac{1}{4}(2\Delta_{\mathbf{k}} - \Delta_{\mathbf{k+Q}} - \Delta_{\mathbf{k-Q}}),$$

while the fluctuation term is

$$\tilde{\gamma}^l_\perp = \tfrac{1}{4}\sum_{\mathbf{k}}\frac{1}{\omega_{\mathbf{k}^+}+\omega_{\mathbf{k}^-}}[\nabla_{\mathbf{k}}h^+_{\mathbf{k}}(u_+v_- - u_-v_+) - \nabla_{\mathbf{k}}\Delta^-_{\mathbf{k}}(u_+u_- - v_+v_-)]^2, \quad (19)$$

where $u_\pm = u_{\mathbf{k}^\pm}$, $v_\pm = v_{\mathbf{k}^\pm}$, and $\mathbf{k}^\pm = \mathbf{k} \pm \mathbf{Q}/2$.

The frustrated J_1-J_2-J_3 and Kagomé Heisenberg models (see Fig. 2) provide simple examples of biaxial-uniaxial melting in two-dimensional spin structures; in both cases we find that the twist stiffness has a fluctuation-induced component which allows it to survive the absence of an ordered moment at small S. Unlike the magnetization, the residual twist stiffness is dependent on the presence of local SO(3) order. On length scales below the spin-correlation length the spin nematic behaves like a helimagnet, and vortex configurations of the twist director will be metastable due to the SO(3) core. Bethe-ansatz solutions of 2D classical SO(3) σ models suggest that topological considerations are very important when considering spin-nematic formation from the point of view of a long-wavelength action: Indeed, the very formation of a twist can be regarded as a soliton condensation process reminiscent of soliton condensation in the 2D sine-Gordon model. This process dramatically upsets the nature of the long-wavelength renormalizations;[14] in the absence of a twist the scaling behavior of a continuum 2D SO(3) σ model is essentially identical to its O(3) counterpart.[15] A complete investigation of the topological effects of a twist on the renormalization flows at long wavelengths has yet to be done.

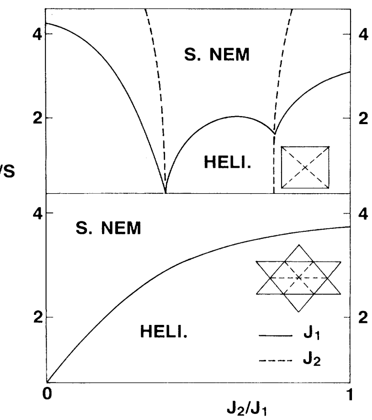

FIG. 2. Phase diagram of the critical spin as a function of frustration in the Heisenberg model for (top) a square lattice with $1/4 > J_3/J_1 > 0$ and (bottom) a Kagomé lattice (where J_2 is the bond strength within the hexagons), with the spin-nematic and helimagnetic regions shown.

At this point our methods do not include instanton effects, and thus we cannot distinguish between specific values of the spin S which may lead to competing nonuniform dimer ground states.[16-18] We can, however, rule out the possibility of a direct transition from helimagnetic to dimer phase because "hedgehog" instanton configurations are specific to uniaxial order parameters. A numerical study of the extended Liang-Doucot-Anderson wave function may serve to resolve this controversial question.[12]

Several experimental features of a helimagnet will be retained by a spin nematic *even* in the absence of elastic Bragg peaks. As in cholesterics and helimagnets, p-type spin nematics will be optically active. The magnetic order in a spin nematic decays on a slow time scale $t \sim \xi/c_s$, where c_s is the spin-wave velocity. Therefore, given single-spin-nematic domains of size L, where $L \ll \xi(c/c_s) \sim 10^4 \xi$, electromagnetic radiation will perceive a spin nematic as a disordered helimagnet with a distribution of pitch lengths. The magnetic susceptibility in a helimagnet contains a nonuniform rotating component of the form $\delta\chi^{\alpha\beta}(\mathbf{R}) \sim \chi_a S^\alpha(\mathbf{R}) S^\beta(\mathbf{R})$, and, as in cholesterics, this generates an optically active permittivity tensor. Following de Vries,[19] we conclude that a p-type spin nematic will exhibit a frequency-dependent circular dichroism with optical rotation ϕ of approximate size

$$\phi(k) \sim \int dQ\,\rho(Q) \frac{QL}{32} \left(\frac{\chi_a}{\mu_0 \mu} \right)^2 \frac{k^4}{Q^2(Q^2-k^2)}, \quad (20)$$

where $\rho(Q) \sim \xi/\pi[(Q-Q_0)^2\xi^2+1]$. As in a cholesteric, a plane-polarized beam reflected off a spin nematic will acquire a circularly polarized component with the same handedness as the nematic.[19] For optical frequencies, the dichroism scales as $\sim\omega^4$, whereas at x-ray frequencies, as in helimagnets, a marked circular polarization will develop.[20] Finally, we note that, as in a ferromagnet, low-angle neutron scattering can detect poles in the susceptibility associated with the Goldstone mode of the twist. Gradients in the twist correspond to a spin current; from spin conservation the autocorrelation function of the divergence of the spin current and the spin precession $\dot{S}(\mathbf{q})$ are equal, so that at long wavelengths $\chi_s(q) \sim (q^4/\omega^2)\chi_{\hat{T}}$. Since the twist is not a conserved quantity, the imaginary part of the twist susceptibility will contain poles of the form $\chi''(\mathbf{q},\omega) \sim (1/\omega_\mathbf{q})\delta(\omega - cq)$ which will appear in the low-angle neutron scattering, multiplied by the form factor q^2.

We would like to mention some possible realizations of this phenomenon. In two dimensions Ramirez and coworkers[21,22] have recently studied a chromium Kagomé lattice in the magnetoplumbite compound $SrCr_{8-x}Ga_{4+x}O_{19}$;[23] they find no ordered moment but a broad neutron peak at the hexagonal wave vector. At low temperatures, the specific heat has a purely T^2 dependence that is robust against disorder;[21] this reflects the linear density of states expected in a 2D spin nematic. A related 2D realization is the nuclear magnetism of ^{3}He films adsorbed on graphite which may also form a Kagomé lattice.[24] Though our discussion has focused on 2D compounds, it would be interesting to reexamine the phase diagrams of classic rare-earth spin helimagnets. Finally, there is the question of whether spin-nematic behavior can occur in conductors as an intermediate state between paramagnetic metallic behavior and insulting antiferromagnetism.

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