Nematicity and quantum paramagnetism in FeSe

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In common with other iron-based high-temperature superconductors, FeSe exhibits a transition to a ‘nematic’ phase below 90 K in which the crystal rotation symmetry is spontaneously broken. However, the absence of strong low-frequency magnetic fluctuations near or above the transition has been interpreted as implying the primacy of orbital ordering. In contrast, we establish that quantum fluctuations of spin-1 local moments with strongly frustrated exchange interactions can lead to a nematic quantum paramagnetic phase consistent with the observations in FeSe. We show that this phase is a fundamental expression of the existence of a Berry’s phase associated with the topological defects of a Néel antiferromagnet, in a manner analogous to that which gives rise to valence bond crystal order for spin-1/2 systems. We present an exactly solvable model realizing the nematic quantum paramagnetic phase, discuss its relation with the spin-1 J1–J2 model, and construct a field theory of the Landau-forbidden transition between the Néel state and this nematic quantum paramagnet.

In insulating quantum paramagnets (PM) are magnetic systems with only short-range antiferromagnetic (AF) correlations even at zero temperature. Examples of quantum paramagnetic states include valence bond solids, symmetry-protected topological states, and spin liquids. Interest in quantum paramagnets was greatly intensified following the proposal1 (since shown to be incorrect) that the parent insulator of the cuprate high-temperature superconductors (HTSC) might be a spin liquid. Because of the proposed relevance to the cuprates, special attention has been paid to square lattice $S = 1/2$ systems, and in particular to the $J_1$–$J_2$ Heisenberg model,

$$H = J_1 \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle\langle ij \rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j \tag{1}$$

where $(ij)$ and $\langle\langle jk \rangle\rangle$ denote nearest-neighbour (NN) and second neighbour pairs of sites and $J_1, J_2$ are the associated coupling constants. Numerical studies have shown that the ground state of equation (1) has Néel AF order for $0 \leq J_2/J_1 \leq 0.4$ and stripe AF order (see Fig. 1a,b) for $0.6 \leq J_2/J_1$. Both these phases have gapless $S = 1$ (spin wave) excitations. For $0.4 \leq J_2/J_1 \leq 0.6$ the ground state seems to have no magnetic order. However, there remain important unresolved issues concerning the precise character of this phase or phases; at least for $0.5 \leq J_2/J_1 \leq 0.6$ there is fairly compelling evidence of translation-symmetry-breaking valence band solid order with an energy gap for spin-1 excitations.

The curious fact that on restoring the spin rotation symmetry the breaking of spatial translation symmetry follows, rests on a uniquely quantum mechanical effect associated with the Berry’s phase of the monopole events6,7. A monopole (anti-monopole) is a spacetime event (see Fig. 2) across which the ‘skyrmiom number’ (see the caption of Fig. 2) changes by $+1(-1)$. The proliferation of monopoles randomizes the Néel order parameter, hence causing the system to become a PM. In ref. 5, it was argued that monopole events contribute to the path integral with a phase factor that depends on the monopole’s spatial location (see Fig. 2b,c).

Spin models based on local moments, such as the $J_1$–$J_2$ model, have found renewed applications in the young field of iron-based superconductivity. The iron-based HTSCs have layers of Fe$^{2+}$ ions which form a square lattice at high temperatures. Many experimental and theoretical studies have concluded that, with the possible exception of heavily phosphorus-doped members of the 122 family, the electronic correlations in the iron-based materials, in particular in the iron chalcogenides, are strong8–9. Moreover, relatively large local magnetic moments in many of these materials have been inferred10. For FeSe the itinerant electrons give rise only to tiny Fermi pockets11–14. Thus, it is plausible that the magnetism of FeSe can be addressed using a spin model as a starting point.

For many iron-based materials, depending on the doping level, there is ‘stripe’ AF order at low temperatures (see Fig. 1a,b). Owing to the breaking of crystal rotation symmetry the stripe order is accompanied by a tetragonal to orthorhombic lattice distortion. In some cases—for example, in electron-doped Ba-122 materials—the lattice distortion can exist without stripe order (see Fig. 3a). In ref. 15 it was shown that the distortion is driven by electronic nematicity rather than a lattice (phonon) instability. It has been argued that such nematicity reflects an underlying stripe ordering tendency, and the reason it can exist without the magnetic order is because thermal fluctuations of the continuously varying spin orientation are more severe than those of the discrete nematic director16–18.

A potential problem with this perspective is the thermal evolution observed in FeSe crystals, in which nematicity onsets at $T_{\text{nem}} \sim 90$ K, but there is no magnetic ordering down to the lowest measured temperatures, suggesting the possibility of a zero-temperature nematic quantum PM phase (Fig. 3b). This fact, coupled with the absence of any observed enhancement of the low-frequency magnetic fluctuations at $T_{\text{nem}}$ (refs 19,20), has led many to conclude that the nematicity in FeSe is driven by orbital ordering.

Certainly, it is clear that within a local moment picture the lack of magnetic order implies that the spin–spin interactions must be highly frustrated; this is true regardless of what causes the nematic ordering. Thus in the following we ask, ‘Can frustrated spin interactions alone drive a nematic quantum PM state?’

We consider models in which each Fe$^{2+}$ possesses a localized spin 1 (for example, the $S = 1$ version of the model in equation (1)).

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Given the strength of Hund’s coupling and the crystal-field splittings, $S = 1$ is a reasonable possibility for the spin of Fe$^{3+}$ ions. However, it is important to keep in mind that FeSe is not an insulator, and such localized models neglect the effects of itinerant carriers. We shall return to this point later.

First we demonstrate the existence of a nematic quantum PM phase in an exactly solvable Hamiltonian. Consider a square lattice of $S = 1$ spins interacting via the short-range, spin rotationally invariant Hamiltonian

$$H_K = K \sum_{\langle ij \rangle} P_i \{S_i + S_j + S_k\}$$

(2)

where $K > 0$ and $\sum_{\langle ij \rangle}$ sums over all elementary triangles of sites (see Fig. 1c), and where $\hat{T}$ and $\hat{R}$ are NN bonds and $\hat{K}$ is a next-NN bond. Here $P_i(S_i) = (1/720)S_i(S_i - 2)(S_i - 1)$ is the projection operator onto $S = 3$. We note that because it involves spin-1 operators, equation (2) possesses a global spin SO(3) (rather than SU(2)) symmetry. In addition it possesses all crystalline symmetries of the square lattice (that is, translation and point group symmetries). Moreover, there are two degenerate ground states which can be constructed exactly as follows: any closed loop $\eta_{i_1 \ldots i_n}$ on the lattice can be thought of as a spin-1 chain. The well-known AKLT (Affleck–Keeley-Lieb–Tsai) state$^{21}$ of such chains can be written as the matrix product state $|C_{\eta_{i_1 \ldots i_n}}\rangle = \sum_{m_{i_1 \ldots i_n}} \text{Tr}[A(m_{i_1}) \ldots A(m_{i_n})] |m_{i_1}\rangle \ldots |m_{i_n}\rangle$, where $m_i = -1, 0, 1$ is the $S\_q$ quantum number of the spin on site $i$, and the matrices $A(m)$ are $A(\pm 1) = (\mp \sigma_x + i \sigma_y) / \sqrt{2}$, $A(0) = \sigma_z / 2$, with $\sigma_{x,y,z}$ being the Pauli matrices. We identify the two adjacent sites $i_1$ and $i_{n+1}$ in an AKLT loop as an ‘AKLT-entangled pair’, and graphically represent it by a blue bond connecting $i_1$ and $i_{n+1}$ in Fig. 4. The two ground states of $H_K$ are constructed as the direct product of NN AKLT-loops on all the loops made by connecting NN sites in the $x$ direction, $|X\rangle = \prod_{\langle ij \rangle} |C_{\eta_{i_1 \ldots i_n}}\rangle$, or in the $y$ direction, $|Y\rangle = \prod_{\langle ij \rangle} |C_{\eta_{i_1 \ldots i_n}}\rangle$, where $(x, y)$ with $x, y = 1, \ldots, N$ labels the sites of an $N \times N$ square lattice. The graphical representations of $|X\rangle$ and $|Y\rangle$ are shown in Fig. 4. Because the maximum total spin of an AKLT-entangled pair is $S = 1$ (ref. 21), it follows that the maximum spin component of a triplet of spins $\langle ijk \rangle$ containing at least one AKLT-entangled pair is $S = 2$. From this it follows trivially that $H_K$ annihilates $|X\rangle$ and $|Y\rangle$; moreover, as $H_K$ is positive semidefinite, this proves that they are ground states.

It is only slightly more difficult to see that any other state constructed as a direct product of AKLT-loop states has positive energy: as each site can be AKLT-entangled with at most two other sites, and as there are four elementary triangles per site, in any zero-energy state exactly two of the two sites must be AKLT-entangled, and each entangled pair must form an edge of four distinct elementary triangles. The two states $|X\rangle$ and $|Y\rangle$ are the only ones that satisfy this constraint. For instance, if the blue bonds in Fig. 4 make a turn, an elementary triangle with its vertex at the corner of the turn will not contain any AKLT-entangled pair. Although we do not have a general proof that no more complex ground states exist, explicit finite cluster diagonalization (up to system size $4 \times 4$) suggests that there are none.

From the known properties of AKLT states$^{21,22}$, it follows immediately that $|X\rangle$ and $|Y\rangle$ are gapped PM states with exponentially falling spin–spin correlations which break crystal rotation symmetry: $G_\pm(R) = (|X\rangle\langle X| + |Y\rangle\langle Y|)/2$ with $Q_N = \pi (\hat{x} + \hat{y})$ and $\xi = 1/\ln(3)$ in units in which the lattice constant is 1. The gap implies that the nematic PM phase is perturbatively stable. However, the asymptotic form of $G$ is non-generic—this reflects the fact that $H_K$ lies on a ‘disorder line’$^{23}$ where, although there is no associated thermodynamic non-analyticity, the oscillatory character of the short-range order changes.

Turning to less ‘engineered’ models, we discuss the very interesting numerical (density matrix renormalization group) study by Jiang et al.$^{24}$ of the spin-1 version of the $J_z$–$J_y$ model in equation (1). They found that as a function of $J_z/J_y$ there is an intermediate paramagnetic phase (characterized by gapped $S = 1$ excitations) between the Néel and stripe-ordered phases (see Fig. 3c). Moreover the Néel and stripe order parameters vanish continuously as $J_z/J_y$ approaches the respective critical values from the magnetically ordered side (see Fig. 4 of ref. 24). In the Supplementary Information we perform exact diagonalization on small lattices and demonstrate the adiabatic connectivity of the ground-state phases of $H_2$ (equation (2)) and the $J_z$–$J_y$ model when $J_z/J_y$ falls within the PM regime. Furthermore, we compare the excitations in different spin and momentum sectors.

To obtain an analytic understanding of the nematic quantum PM phase, we consider a field theory description valid in the neighbourhood of a continuous or weakly first-order quantum phase transition from the Néel phase to a nematic PM phase. Because the unbroken symmetries of the Néel and nematic PM phases do not have a subgroup relationship, classical Landau theory would imply that such a continuous phase transition is forbidden. However, as pointed out in ref. 25, when the topological defects of one order carry the quantum number of the other order a continuous transition becomes possible.

According to ref. 5, in the path integral describing the quantum fluctuations of the $S = 1$ Néel antiferromagnet in 2D, the weight associated with each field configuration is determined by the usual nonlinear sigma model (analogous to the first term in equation (3)), but there is also an additional Berry’s phase factor. For a charge $q_m$ monopole (which causes the skyrmion number to jump by $q_m$) centred on plaquette $\mathcal{P}$ (which designates a point on the dual lattice) this phase factor (up to a global ‘gauge’ ambiguity) is $\eta_\mathcal{P} = e^{iq_m/\sqrt{2}}$, where $\mathcal{P}$ is a plaquette on the dual lattice that designates a point on the dual lattice) this phase factor (up to a global ‘gauge’ ambiguity) is $\eta_\mathcal{P} = e^{iq_m/\sqrt{2}}$ (see Fig. 2c). Rotation by $90^\circ$ about a lattice site transforms $\eta_\mathcal{P} \rightarrow -\eta_\mathcal{P}$ but keeps $q_m$ invariant. So long as this symmetry is preserved, the Feynman amplitude of any field configuration with an odd $q_m$ cancels that associated with the configuration rotated by $90^\circ$. As a result, odd $q_m$ monopoles cannot proliferate, although even $q_m$ monopoles can. Consequently, states can be classified by a conserved skyrmion number parity ($\sim 1$). It turns out that the two-fold degeneracy associated with opposite skyrmion parity
accounts for the two nematic ground states depicted in Fig. 4. If we introduce a perturbation that breaks the crystal 90° rotation symmetry, the absolute value of the Feynman amplitudes associated with monopoles sitting at the +1 and −1 locations in Fig. 2c no longer need to be the same, hence their Feynman amplitudes no longer cancel, which means the odd \( q_m \) monopoles can proliferate, rendering the PM state non-degenerate. This observation justifies identifying the two-fold degeneracy in the symmetric system with spontaneous breaking of 90° rotation symmetry.

In the following discussions we will use Euclidean spacetime and denote the imaginary time as \( \tau \). Consider a 4-component real vector field of unit norm, \( \Omega(x, y, \tau) = (\Omega_1, \Omega_2) \) with \( |\Omega|^2 = 1 \), whose first three components, \( \Omega_1(x, y, \tau) \) are the Néel order parameter and \( \Omega_2 \) is the Ising-like nematic order parameter. For example, for a spin model on a square lattice, we can take \( \Omega_1(x, y, \tau) \propto (S \cdot S_{x+i} - S \cdot S_{y+i}) \). The nonlinear sigma model action we shall consider is

\[
S = \int d^2 x d\tau \left[ \frac{1}{2g} |\dot{\Omega}|^2 + V(\Omega^2) \right] + i \frac{\Theta}{2\pi} \int d^2 x d\tau \epsilon^{\alpha\beta\gamma\delta} \partial_\alpha \Omega_\beta \bar{\partial}_\gamma \Omega_\delta \bar{\partial}_\epsilon \Omega_\epsilon
\]

where \( \Theta = \pi \) and \( \epsilon \) is the Levi-Civita symbol. This field theory has been introduced in refs 26,27, which discuss the effects of topological terms on the spectrum and phases of nonlinear sigma models. For our purposes a derivation of equation (3) is given in the Supplementary Information. Note that this particular topological term is possible only in spatial dimension \( d = 2 \) with a \( N = 4 \) component order parameter. Here \( V \) is an anisotropy term that favours the first three components of \( \Omega_1 \) (for example \( V = \Delta |\Omega_1|^2 \)), hence equation (3) has \( O(3) \times Z_2 \) symmetry. Consider a monopole configuration centred on the origin, \( (x, y, \tau) = (0, 0, 0) \). Because of the anisotropy term, \( \Omega \) on a two-sphere (\( S^2 \)) in spacetime far away from the centre of the monopole is largely constrained to lie in the space spanned by the first three components of \( \Omega_1 \)—that is, \( \Omega_2 \approx (\hat{n}, 0) \). Topologically the configurations of \( \hat{n} \) on the two-sphere are classified by the skyrmion number which, in this case, is equal to the monopole charge \( q_m \). However, such far-field configuration is compatible with two different \( \Omega_1 \) orientations in the monopole core. Consider the following configurations which reduce to the same \( q_m = 1 \) configuration in \( \hat{n} \) far from the monopole centre

\[
\hat{\Omega}^{(2)}(x, y, \rho) = \left( \sin \phi_1(\rho) \hat{n}(x, y, \rho), \cos \phi_1(\rho) \right)
\]

In equation (4) \( \rho = \sqrt{x^2 + y^2 + \tau^2}/R \), \( \phi_1(\rho) = \pi/2 \tan h(\rho) \) and \( \phi_2(\rho) = \pi(1 - 1/2 \tan h(\rho)) \). Here \( R \) is the size of the ‘monopole core’. \( \hat{\Omega}^{(2)} \) both describe \( q_m = 1 \) monopoles but with \( \Omega = (0, \pm 1) \) in the monopole core. It is straightforward to show that \( \exp(i\Theta/2\pi i) \int d^2 x d\tau \epsilon^{\alpha\beta\gamma\delta} \partial_\alpha \Omega_\beta \bar{\partial}_\gamma \Omega_\delta \bar{\partial}_\epsilon \Omega_\epsilon \equiv e^{i\pi/2} = \pm i \) for these two types of monopole. A similar discussion holds for the \( q_m = -1 \) monopole. It is easy to generalize the above argument to \( q_m \) monopoles and obtain the corresponding phase factors \( e^{i\pi/2} = -1 \). A consequence of the \( Z_2 \) (nematic) symmetry is that the Feynman amplitudes of the two monopoles described above have the same absolute values.

These monopoles represent events (as a function of imaginary time) at which the skyrmion number changes. We can thus think of the quantum disordered phase as an interacting fluid of skyrmions and anti-skyrmions. Because of the destructive interference between the two types of \( q_m \) monopoles discussed above, events in which the skyrmion number changes by one are forbidden—the net skyrmion number is thus conserved modulo 2 and the Hilbert space breaks into an even and an odd sector. As the stiffness constant \( 1/g \) renormalizes to zero in the quantum disordered state, and as there is a non-zero (possibly small) density of skyrmions and anti-skyrmions, the ground-state energy in these two sectors should be equal in the thermodynamic limit. This is the ground-state degeneracy associated with the breaking of \( C_4 \) symmetry to \( C_2 \). In the Supplementary Information we discuss the effects of an explicit \( C_4 \to C_2 \) symmetry-breaking field.

One can also arrive at the PM phase in Fig. 3c from the stripe-ordered side by proliferating the monopole of the stripe order parameter. Generalizing the calculation of ref. 5 we find the Berry’s phase factor associated with the stripe monopole is trivial. Consequently, the charge \( \pm 1 \) monopole can proliferate, rendering the resulting quantum disordered state non-degenerate. Here, because the symmetries of the two phases have subgroup relationship, a continuous transition is allowed in the Landau theory. We expect such a transition to be in the O(3) universality class.

Our key theoretical conclusion is that for a spin-1 model on a square lattice, any ‘intermediate’ quantum disordered phase.
between the Néel and stripe-ordered magnetic phases is likely to be a nematic PM. This is consistent with the numerical evidence for the \(J_1\text{−}J_2\) model (equation (1)). Because the nematic quantum PM phase straddles closely between the Néel and nematic phases, we expect it to have low-lying spin excitations near both momenta \((\pi, 0)\) and \((\pi, \pi)\). The nematic order considered here might be thought of as 'vestigial' order\(^\text{9}\) left behind when quantum fluctuations have restored spin-rotational symmetry.

Before speculating on the relevance of these results to the interesting case of FeSe, it is necessary to comment on the effects of itinerant carriers. Angle-resolved photoemission spectroscopy (ARPES) and scanning tunnelling microscope experiments show very small electron and hole pockets for FeSe (refs 13,14,29) and at the same time quantum oscillations associated with these tiny pockets are seen\(^\text{30}\). Because the latter experiment has a very stringent requirement on the long lifetime of the quasiparticles we take it as an indication that the coupling between the itinerant carriers and the local moments is weak. This phase with decoupled magnetic and itinerant carrier degrees of freedom is analogous to the antiferromagnetic metal phase in heavy fermion systems. However, owing to the itinerant carriers the nematic phase can no longer be characterized as having a spin-gap and, close enough to criticality, the universality class of the quantum critical point is likely to be altered. In addition, FeSe is at best 'quasi-2D', which is to say that it is ultimately three-dimensional, and this, too, will alter the nature of the phase transitions, but not necessarily affect the sequence of ordered phases.

As the iron-based superconductors involve multiple 3d orbitals it has long been suspected that orbital ordering is the cause of nematicity. Although we focus on the nematicity caused by frustrated magnetism, we do not intend to imply that orbital degrees of freedom play no role at all. As is well known, regardless of the driving mechanism the nematic order parameter induces orbital splitting and lattice distortion. Therefore, a cooperative effect involving many different degrees of freedom could well be necessary to describe the system behaviour quantitatively. In addition, the so-far ignored spin orbit coupling is ultimately necessary for the nematicity in the magnetic degrees of freedom to induce the band splitting observed by ARPES, the anisotropic magnetic susceptibility observed by NMR and so on.

With these caveats, the present results suggest that it may be possible to view the nematic phase in the FeSe as being driven primarily by frustrated magnetism. The fact that the underlying nematic quantum PM is gapped implies that there need not be any enhancement of the low-energy magnetic fluctuations associated with the proximity of stripe long-range order of the sort that has been detected by NMR in other Fe-based materials. Recent neutron scattering experiments\(^\text{31}\) show relatively short-range magnetic correlations, but with reasonably large intensity (that is, with substantial magnitudes of the fluctuating moments) at the stripe ordering wavevector at low energies. Consequently, the lack of any strong evidence of slow magnetic fluctuations in NMR (ref. 20) and the short range of the magnetic correlations seen in neutron scattering seem to be entirely consistent with a magnetic origin of nematicity. As discussed previously, we expect the nematic PM phase to also exhibit spin excitations with relatively low energy at the Néel ordering wavevector. The nematic quantum PM state consists of a stacking of spontaneously formed spin-1 AF chains. It can be viewed as a 'weak' symmetry-protected topological state. A potentially testable consequence of this observation is that gapless spin-1/2 excitations can arise on certain surfaces and domain boundaries. In addition we expect nonmagnetic impurities to cut the chain and induce low-lying spin excitations in the PM gap. This can be detected by electron spin resonance in a manner analogous to that done for spin-1 chains\(^\text{32}\). Further experimental studies of nematic order and fluctuations in these materials—including elasto-resistance, nuclear quadrupole resonance, and Raman scattering studies—as well more complete neutron scattering studies of the magnetic fluctuations would clearly be helpful.

Finally after the submission of our paper, related theoretical studies appeared\(^\text{33,34}\), including one based on an itinerant electron picture of the nematicity in FeSe (ref. 35).

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Additional information
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Competing financial interests
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