Blue Quantum Fog: Chiral Condensation in Quantum Helimagnets

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It is shown that a condensation transition involving a chiral order parameter can occur in itinerant helimagnets, in analogy to the transition between the isotropic phase and the phase known as blue fog or blue phase III in cholesteric liquid crystals. It is proposed that such a transition is the explanation for recent neutron scattering results in MnSi. Predictions are made that will allow for experimental tests of this proposal.

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The unusual behavior of the low-temperature itinerant magnet MnSi has received much attention. At ambient pressure P, the material enters a magnetic phase below a temperature $T_c \approx 30$ K. Over distances of a few lattice spacings, the magnetic order appears ferromagnetic [1]. However, at longer length scales, a helical modulation of the magnetization appears, with a wavelength $2\pi/Q_0 \approx 170$ Å [2]. This helical structure is believed [3,4] to be due to an interaction between magnetic fluctuations $\boldsymbol{M}$ of a form first proposed by Dzyaloshinski [5] and Moriya [6] (DM), $\int d\boldsymbol{x}\boldsymbol{M} \cdot (\boldsymbol{\nabla} \times \boldsymbol{M})$. Such a term, which is produced by the spin-orbit interaction, is allowed by symmetry in MnSi since its lattice structure lacks inversion symmetry. The helix is readily observed via neutron scattering, with scattering intensity appearing only in the $\langle 111 \rangle$ direction since crystal fields lock the direction of the helix [2,7]. With increasing P, the transition temperature to the ordered phase decreases monotonically, and the nature of the transition changes from continuous or very weakly first-order to decidedly first-order at a tricritical point at $P^* \approx 12$ kbar, before the transition temperature drops to zero at $P_c \approx 14.6$ kbar. In the disordered phase, i.e., for $P > P_c$, spectacular non-Fermi-liquid (NFL) behavior of the transport properties has been observed below a crossover temperature T^* [8,9].

Within this extended NFL region, Pfleiderer *et al.* [7] have identified a pressure-dependent temperature scale T_0, with a strong neutron scattering signal at a well-defined wave number $q_0 \approx 0.043$ Å for $T < T_0$. The signal is strong enough to be reminiscent of the one at $Q_0 \approx 0.037$ Å in the ordered phase but is much more isotropic, with broad maxima centered around the $\langle 110 \rangle$ direction. These observations imply the existence of short-ranged helical order even in the nonmagnetic phase, and Ref. [7] has suggested that this short-ranged helical order is at the heart of the NFL transport behavior.

In this Letter, we focus on such local correlations and argue that the existence of the temperature scale T_0 is consistent with a first-order transition from a chiral liquid to a gaseous phase as one crosses the condensation temperature $T_0(P)$. We thus propose that the phase diagram of MnSi is more complicated than previously thought, with a liquid-gas-type transition inside the nonmagnetic phase. This proposed phase diagram is schematically depicted in Fig. 1.

A convenient order parameter for this first-order transition is the chiral composite field $\psi = \boldsymbol{M} \cdot (\boldsymbol{\nabla} \times \boldsymbol{M})$. ψ is a pseudoscalar which has nonvanishing average values both below and above T_0. The two phases separated by T_0 thus have the same symmetry, as do the gaseous and liquid phases, respectively, of ordinary fluids. Crossing the coexistence line $T_0(P)$ is accompanied by a discontinuous change in the expectation value of ψ, which corresponds to stronger short-ranged helical correlations in the liquid than in the gas. This accounts for a stronger neutron

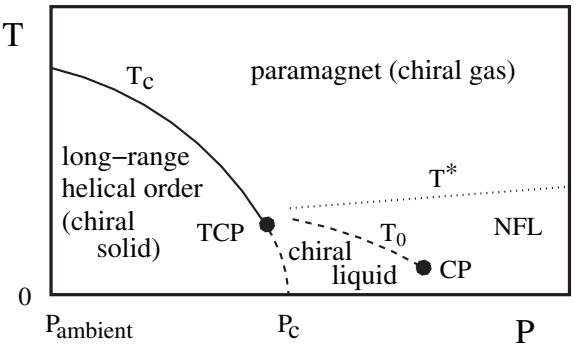

FIG. 1. Phase diagram of MnSi, including the proposed chiral liquid phase with an associated critical point (CP). The CP may be at $T < 0$ and, thus, inaccessible. Second- (or very weakly first-) order transitions, first-order transitions, and crossovers are denoted by solid, dashed, and dotted lines, respectively. The non-Fermi-liquid (NFL) region includes the chiral liquid, and the chiral gas extends below T^*. The precise structure near the tricritical point (TCP) is not known. See the text for further information.

scattering signal in the liquid, with the signal being isotropic to zeroth approximation. Our scenario is analogous to the transition from the isotropic phase to the phase known as "blue phase III" or "blue fog" in cholesteric liquid crystals, for which a theory based on liquid-gas-type first-order transition is the currently most successful interpretation [10–14]. This condensation interpretation makes blue fog a very appropriate name for the phase below the coexistence temperature. We will first show, starting from a microscopic action for a quantum helimagnet, that there is an attractive interaction between chiral fluctuations described by ψ, which makes a gas-liquid-like condensation of these degrees of freedom possible for appropriate values of T and P. Assuming that this condensation can be identified with the observed temperature scale $T_0(P)$, we will focus on experimental predictions that follow from this proposal.

Let us start with a Landau-Ginzburg-Wilson (LGW) functional appropriate for a quantum helimagnet that adds a DM interaction to a ferromagnetic action:

$$S_{\mathrm{DM}}[\boldsymbol{M}] = S_{\mathrm{fm}}[\boldsymbol{M}] + c\int dx \boldsymbol{M}(x)\cdot[\boldsymbol{\nabla}\times\boldsymbol{M}(x)], \tag{1a}$$

$$S_{\mathrm{fm}}[\boldsymbol{M}] = \frac{1}{2}\int dx dy \boldsymbol{M}(x)\Gamma(x-y)\boldsymbol{M}(y) + \frac{u}{4}\int dx(\boldsymbol{M}^2(x))^2. \tag{1b}$$

The two-point vertex Γ reads, in Fourier space,

$$\Gamma(\boldsymbol{p}, i\omega_n) = t + a\boldsymbol{p}^2 + b|\omega_n|/|\boldsymbol{p}|. \tag{1c}$$

Here $x = (\boldsymbol{x}, \tau)$ is a four-vector that comprises position $\boldsymbol{x}$ and imaginary time τ, $\int dx = \int_V d\boldsymbol{x}\int_0^{1/T} d\tau$. $\boldsymbol{p}$ is a wave vector, and $\omega_n = 2\pi T n$ denotes a bosonic Matsubara frequency. $\boldsymbol{M}$ is the order parameter field with components $M_i (i = 1, 2, 3)$ whose expectation value is proportional to the magnetization. t, a, b, c, and u are the parameters of the LGW theory; they are functions of T and P. V is the system volume. S_{fm} is Hertz's action for a quantum ferromagnet [15,16]. The additional term in S_{DM} is the chiral DM term. The sign of c determines the handedness of the helix; we will take $c > 0$ without loss of generality.

For the Gaussian propagator $G_{ij}(\boldsymbol{p}, i\omega_n) \equiv \langle M_i(\boldsymbol{p}, i\omega_n)M_j(-\boldsymbol{p}, -i\omega_n)\rangle$, we obtain from Eq. (1)

$$G_{ij}(\boldsymbol{p}, i\omega_n) = \frac{1}{\Gamma^2(\boldsymbol{p}, i\omega_n) - c^2\boldsymbol{p}^2}[\Gamma(\boldsymbol{p}, i\omega_n)\delta_{ij} - ic p_l \epsilon_{ijl} - c^2 p_i p_j/\Gamma(\boldsymbol{p}, i\omega_n)]. \tag{2}$$

An analysis of the eigenvalue problem given by the quadratic form G_{ij} shows that the paramagnetic phase is unstable against the formation of helical order for $t < c^2/4a$. The instability first occurs at $|\boldsymbol{p}| = q_0 = c/2a$, the pitch of the helix. $t(T_c, P) = c^2/4a$ thus determines T_c in Fig. 1 in a mean-field approximation.

We now consider the completeness of the action given by Eq. (1). The only chiral term so far is the quadratic-in-$\boldsymbol{M}$ DM term with coupling constant c, and the action S_{DM} is the magnetic analog of the action for cholesteric liquid crystals that was the starting point for the theory developed by Lubensky and Stark [10]. However, symmetry allows for a quartic chiral term of the form $\int dx(\boldsymbol{M}\cdot(\boldsymbol{\nabla}\times\boldsymbol{M}))^2$. It is easy to see that such a term is indeed generated by a perturbative renormalization-group procedure, with a *negative definite* coupling constant $0 > -d_1 \propto -u^2c^2$. Another quartic term that is allowed by symmetry and generated by renormalizing the action S is $-d_2\int dx \boldsymbol{M}^2(\boldsymbol{M}\cdot(\boldsymbol{\nabla}\times\boldsymbol{M}))$, with $0 > -d_2 \propto -u^2c$. Adding these two terms to Eq. (1a), we obtain our final LGW action, with $d_1, d_2 > 0$,

$$S[\boldsymbol{M}] = S_{\mathrm{DM}}[\boldsymbol{M}] - d_1\int dx[\boldsymbol{M}(x)\cdot(\boldsymbol{\nabla}\times\boldsymbol{M}(x))]^2 - d_2\int dx\boldsymbol{M}^2(x)[\boldsymbol{M}(x)\cdot(\boldsymbol{\nabla}\times\boldsymbol{M}(x))]. \tag{3}$$

The d_1 term is conceptually crucial for the physical picture we are proposing. The DM term ensures a nonzero expectation value $\langle\boldsymbol{M}\cdot(\boldsymbol{\nabla}\times\boldsymbol{M})\rangle \neq 0$ everywhere in the phase diagram. The presence of $d_1 > 0$ implies an *attractive* interaction between the chiral fluctuations described by $\boldsymbol{M}\cdot(\boldsymbol{\nabla}\times\boldsymbol{M})$ [20]. This, in turn, means that the chiral fluctuations may condense into a chiral liquid as the temperature is lowered, with a discontinuous behavior of $\langle\boldsymbol{M}\cdot(\boldsymbol{\nabla}\times\boldsymbol{M})\rangle$ across a first-order phase transition. This is the central idea of the present Letter.

The above considerations suggest considering the composite field $\psi = \boldsymbol{M}\cdot(\boldsymbol{\nabla}\times\boldsymbol{M})$ an order parameter for a possible chiral first-order phase transition. It is thus desirable to construct an effective action in terms of ψ. The simplest way to do this is to perform a Hubbard-Stratonovich decoupling of the combined d_1 and d_2 terms in the action, Eq. (3), with $\psi(x)$ the auxiliary field. Alternatively, one can constrain $\boldsymbol{M}\cdot(\boldsymbol{\nabla}\times\boldsymbol{M})$ to ψ by means of a Lagrange multiplier field that is later integrated out [10]. Either method yields an effective action which contains all terms allowed by symmetry and which leads to the same partition function as the original action. If one integrates out $\boldsymbol{M}$, one obtains an action in terms of ψ alone, which is of the form of a LGW action that describes a liquid-gas transition [21]. This is not very illuminating, since the ψ-correlation functions are not directly measurable. It is, therefore, advantageous to integrate out only the "fast" (i.e., large-momentum and high-frequency) components of the field $\boldsymbol{M}$ and write the theory in terms of ψ and the slow components of $\boldsymbol{M}$, whose correlation functions *are* directly measurable. If we denote the slow components of $\boldsymbol{M}$ by the same symbol for simplicity, we thus obtain the following final effective action for chiral fluctuations ψ and slow magnetization fluctuations $\boldsymbol{M}$:

$$S_{\mathrm{eff}}[\boldsymbol{M}, \psi] = S_{\mathrm{DM}}[\boldsymbol{M}] + S_{\psi}[\psi] + S_c[\boldsymbol{M}, \psi]. \tag{4}$$

The first part of the effective action has the same functional form as the action S_{DM} given by Eq. (1), only the parame-

ters have different values. The chiral part $S_\psi[\psi]$ is a LGW functional for a scalar order parameter with no invariance under the transformation $\psi \to -\psi$,

$$S_\psi[\psi] = \int dx[r\psi^2(x) - h\psi(x) + s|\nabla\psi(x)|^2 - v\psi^3(x) + w\psi^4(x)]. \quad (5)$$

For suitable parameter values, ψ can thus undergo a first-order phase transition. Finally, the coupling term reads

$$S_c[\mathbf{M}, \psi] = \int dx[g_1\mathbf{M}(x)\cdot[\nabla\times\mathbf{M}(x)]\psi(x) + g_2\boldsymbol{M}^2(x)\psi(x)]. \quad (6)$$

$g_1 > 0$, $g_2 > 0$ have the same sign as d_1, d_2. The coupling constants r, h, s, etc., are functions of temperature and pressure, as are the coupling constants of the starting LGW theory in Eq. (1). The structures of all terms in the action S_{eff} are governed by symmetry requirements. Once one has introduced the pseudoscalar order parameter ψ in addition to $\boldsymbol{M}$, one therefore can, in principle, just write down S_{eff} based on symmetry considerations.

In what follows, we will assume that the first-order transition inherent in the theory occurs in the experimentally accessible range of P and T and can be identified with the observed temperature scale T_0. We will now discuss some simple observable consequences of this proposal. For simplicity, we will treat ψ in mean-field approximation, $\psi(x) \approx \langle\psi(x)\rangle \equiv \psi = \text{const.}$ ψ increases discontinuously as one crosses the coexistence curve $T_0(P)$ from above, and the discontinuity goes to zero as one approaches the critical point that marks the end of the coexistence curve $T_0(P)$; see Fig. 1.

Observable consequences of the first-order transition arise from the coupling of the chiral order parameter ψ to the magnetization via Eq. (6). In our mean-field approximation, which treats ψ as a constant, this coupling simply renormalizes the terms quadratic in $\boldsymbol{M}$ in the action S_{DM}, leading to renormalized coupling constants

$$c_{\text{R}} = c + g_1\psi, \qquad t_{\text{R}} = t + g_2\psi, \quad (7a)$$

and a renormalization of the vertex Γ, Eq. (1c), given by

$$\Gamma_{\text{R}}(\boldsymbol{p}, i\omega_n) = t_{\text{R}} + a\boldsymbol{p}^2 + b|\omega_n|/|\boldsymbol{p}|. \quad (7b)$$

Since the proposed first-order transition occurs within the magnetically disordered phase, the terms of higher order in $\boldsymbol{M}$ are not qualitatively important; they can be treated perturbatively and lead to further renormalizations of the Gaussian action. The physical magnetic susceptibility tensor $\chi_{ij}(\boldsymbol{p}, i\omega_n) = \langle M_i(\boldsymbol{p}, \omega_n)M_j(-\boldsymbol{p}, -\omega_n)\rangle$, therefore, has the same form as the Gaussian propagator given by Eq. (2), but it now depends on the chiral order parameter ψ,

$$\chi_{ij}(\boldsymbol{p}, i\omega_n) = \frac{1}{\Gamma_{\text{R}}^2(\boldsymbol{p}, i\omega_n) - c_{\text{R}}^2\boldsymbol{p}^2}[\Gamma_{\text{R}}(\boldsymbol{p}, i\omega_n)\delta_{ij} - ic_{\text{R}}p_l\epsilon_{ijl} - c_{\text{R}}^2 p_i p_j/\Gamma_{\text{R}}(\boldsymbol{p}, i\omega_n)]. \quad (7c)$$

In particular, the thermodynamic magnetic susceptibility $\chi_{ij} = \delta_{ij}\chi_m$, defined as $\chi_{ij} = \lim_{\boldsymbol{p}\to 0}\int d\omega \text{Im}\chi_{ij}(\boldsymbol{p}, \omega + i0)/\omega$ is ψ dependent,

$$\chi_m = 1/t_{\text{R}}. \quad (8)$$

We now discuss these results. The energy-resolved neutron scattering cross section $d^2\sigma/d\Omega d\omega$, with Ω the solid angle and ω the frequency or energy, is related to the quantity $\chi(\boldsymbol{q}) \equiv (\delta_{ij} - \hat{q}_i\hat{q}_j)\chi_{ij}(\boldsymbol{q}, i\omega_n = 0)$ by [21]

$$\chi(\boldsymbol{q}) = \text{const}\times\int_{-\infty}^{\infty} d\omega\,\frac{1}{\omega}\left(1 - e^{-\omega/T}\right)\frac{d^2\sigma}{d\Omega d\omega}. \quad (9)$$

From Eq. (7c), one finds for this weighted frequency average of the cross section

$$\chi(\boldsymbol{q}) = 2\frac{t_{\text{R}} + a\boldsymbol{q}^2}{(t_{\text{R}} + a\boldsymbol{q}^2)^2 - c_{\text{R}}^2\boldsymbol{q}^2}. \quad (10)$$

For $c_{\text{R}}^2/4a < t_{\text{R}} < c_{\text{R}}^2/a$, the system is in the disordered phase, and $\chi(\boldsymbol{q})$ has a maximum at $q = q_0 > 0$ with

$$aq_0^2 = c_{\text{R}}\sqrt{t_{\text{R}}/a} - t_{\text{R}}. \quad (11)$$

A measure for the height of the peak at q_0 is

$$\chi(q_0)/\chi(0) = 1/(2\sqrt{y} - y), \quad (12)$$

which depends only on $y = c_{\text{R}}^2/t_{\text{R}}a$. The peak is higher and sharper for smaller t_{R} (at fixed c_{R}).

The theory thus yields, in the disordered phase not too far from the boundary to long-range magnetic order, a sharp peak in $\chi(\boldsymbol{q})$ at a wave number on the same order as the pitch of the helix in the ordered phase, in qualitative agreement with experiment [7,22]. Upon crossing the first-order transition from chiral liquid to the gas, ψ decreases and the peak in $\chi(\boldsymbol{q})$ becomes less pronounced [23], again in qualitative agreement with the interpretation of T_0 as a chiral first-order transition.

Two predictions of other observable effects are (i) a latent heat Q across the coexistence line $T_0(P)$ (the absolute value of Q will be small, since the transition takes place at low temperatures) and (ii) a discontinuity of the magnetic susceptibility χ_m across $T_0(P)$ [see Eq. (8)], even though no magnetic transition occurs at that temperature. Again, this effect will be small since g_2 must be small in order for the first-order transition to occur in the first place [23]. If such signs of a first-order transition are observed, it would also be worthwhile to look for the critical point (CP in Fig. 1), which will be characterized by critical fluctuations in the Ising universality class.

The present theory cannot easily explain the magnitude of the increase in the neutron scattering observed below T_0. We note that the original form of the Lubensky-Stark theory for liquid crystals [10] also did not explain the huge increase observed by light scattering in the blue fog phase. Later theories that took into account more sophisticated correlation effects did find anomalously large fluctuations [12–14], and we expect the same to be true for the

present theory. A quantitative understanding of the anomalously large scattering below T_0 may also be needed to understand the NFL transport behavior mentioned in the introduction.

Finally, we need to discuss the relation between the present theory and related treatments of chiral liquid crystals, as the very possibility of such a relation has been questioned. Early work on blue phases in liquid crystals focused on double-twist cylinder configurations of the director. In this context, Wright and Mermin [24] have argued that there cannot be analogs of blue phases in helical magnets, and they bolstered this argument by free-energy considerations at the mean-field level. These arguments do not apply to our proposal, for several reasons. (i) It has since become clear that blue phase III needs to be considered separately from the other blue phases. Double-twist cylinder configurations are not central to the current understanding of blue phase III; they are just one of many speculations concerning local order in this phase [25]. (ii) The free-energy argument of Ref. [24] is most applicable to the crystalline blue phases I and II because entropic contributions that arise in disordered phases are not taken into account. (iii) The free-energy argument breaks down when the terms with coupling constants d_1, d_2 are included in the free-energy functional.

Given that a magnetic analog of the blue fog phase or blue phase III cannot be ruled out, an obvious first step is to construct a theory analogous to the one put forward in Ref. [10], which is what we have done above. There are two main differences between the two theories. One is the purely technical point that we deal with a quantum vector order parameter (the magnetization) instead of a tensor classical one (the director). The other is the existence in our theory of the couplings d_1 and d_2 in Eq. (3). These terms are allowed by symmetry and also arise in the explicit derivation sketched above. The attractive sign of d_1 is crucial, since it forms the physical basis for a condensation scenario. In the final effective action, these terms give rise to the coupling constants g_1 and g_2 in Eq. (6). The analog of the former was also present in Ref. [10], although its sign was not obvious.

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