Nontrivial Magnetic Order: Localized versus Itinerant Systems

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The generalization of the non-Néel spin ordering concept to itinerant systems is discussed. It is shown that some of the *p*-type spin nematic and chiral spin states affect the electronic spectrum near the Fermi surface and can then be related by symmetry with the intrinsic instabilities of the Fermi liquid although the energetic mechanisms for the localized spins and itinerant electrons are different. If the localized and itinerant subsystems coexist, the phase transition is described by the common order parameter. Possible experimental manifestations of the above transitions are discussed.

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Recently, the possibility of the non-Néel ordering for the antiferromagnetically interacting local spins has been widely discussed. The very fact that order parameters other than the average local magnetic moment are possible has been known for a long time [1, 2]. Our interest in this problem is mostly related to peculiarities of the magnetic behavior of some high- T_c oxides and heavy fermion materials. In the high- T_c compounds, a very small amount of free carriers destroys the long-range antiferromagnetic order, particularly in the 214 systems. As is argued in [3], the disappearance of the staggered magnetization at small doping may indicate the instability of the Néel state with respect to the formation of a nontrivial phase rather than the transition to the paramagnetic state. As for the heavy fermions, in URu₂Si₂ and UPt₃ the average magnetic moments appear below 17.5 and 5 K, correspondingly. However, the moments as seen by neutrons are extremely small in both compounds $(\sim 10^{-2} \mu_B)$. On the other hand, the phase transition at 17.5 K in URu₂Si₂ is accompanied by the strong specificheat anomaly which shows that the magnetic entropy involved is quite large. In other words, the spin degrees of freedom are affected more strongly by the transition than can be expected from such a small average magnetization. This contradiction can be resolved if one assumes that unlike for the ordinary antiferromagnet the staggered magnetization here is not a driving order parameter. This is a strong argument in favor of a nontrivial phase, as has been suggested in [4]. Since both high- T_c and heavy fermion compounds have magnetism which coexists with the metallic behavior, in what follows we address the issue of a correspondence between the symmetry classifications of the non-Néel magnetic phases in the localized and itinerant systems and the consequences of the existence of nontrivial phases in a system with both localized and itinerant components. As for the purely itinerant systems, we believe that the above phases could appear in some of the intercalation compounds and organic conductors as a result of electronic instabilities.

For a localized spin system (assuming the exchange approximation), only the spin degrees of freedom are involved. Any order parameter in this case can be written as a multiple-spin average of the form

$$\langle S_1^{\alpha_1} \cdots S_N^{\alpha_N} \rangle = \sum_{\zeta} \mathsf{T}_{\zeta}^{\alpha_1 \cdots \alpha_N} \Phi_{\zeta}(\mathbf{r}_1, \dots, \mathbf{r}_N), \qquad (1)$$

where T_{ζ} are tensors and Φ_{ζ} are the basis functions of an irreducible representation (multidimensional, in general) of the symmetry group of spin scalars G (G does not necessarily coincide with the crystal group of the higher symmetry phase). The basis function Φ_{ζ} transforms as

$$\Phi_{\zeta}(\mathbf{r}_{1} + \mathbf{R}, \dots, \mathbf{r}_{N} + \mathbf{R}) = \exp\left[i\mathbf{Q}_{\zeta} \cdot \mathbf{R}\right] \\ \times \Phi_{\zeta}(\mathbf{r}_{1}, \dots, \mathbf{r}_{N})$$
(2)

under the lattice translation **R**. In what follows, the index ζ enumerating basis functions will be omitted as unimportant for the present discussion.

The time reversal symmetry is broken only if N in (1) is odd. Thus, for the dimer state, $T^{\alpha\beta} = \delta^{\alpha\beta}$, while for the chiral spin state [5] $T^{\alpha\beta\gamma} = \epsilon^{\alpha\beta\gamma}$. Note that the parity breaking here is described by the spatial part of (1), $\Phi(\mathbf{r}_1, \ldots, \mathbf{r}_N)$. For the *p*-type spin nematic [6, 7], $T^{\alpha\beta} = \epsilon^{\alpha\beta\gamma}P^{\gamma}$. The options for the *n*-type spin nematic [6] are $T^{\alpha\beta} = n^{\alpha}n^{\beta} - \frac{1}{3}\delta^{\alpha\beta}$, $T^{\alpha\beta} = u^{\alpha}v^{\beta} + v^{\alpha}u^{\beta}$ ($\mathbf{v} \perp \mathbf{u}$), or even a more exotic possibility of a symmetric $T^{\alpha\beta} =$ $T^{\beta\alpha}$ with the eigenvalues $\{\sin\frac{\pi}{12}, -\cos\frac{\pi}{12}, \cos\frac{\pi}{4}\}$ [8] (see [8] for a phenomenological classification of the *n*-type spin nematics). For the tensor spin state discussed in [4], T is a third-rank symmetrized spin tensor and $T^{\alpha\alpha\gamma} = 0$.

In principle, (1) allows us to enumerate all order parameters for the localized spins. In the presence of the itinerant electrons, the straightforward generalization is obvious in terms of correlators of the form (1) constructed using the total spin density operators which include both localized and itinerant contributions. An intrasite exchange interaction between the localized and itinerant subsystems

$$H = J \sum_{n} \left(\frac{1}{2} \,\boldsymbol{\sigma}_{\sigma'\sigma} \cdot \mathbf{S}_{n}^{\text{loc}} \right) \,\psi_{\sigma'}^{\dagger}(\mathbf{r}_{n})\psi_{\sigma}(\mathbf{r}_{n}), \qquad (3)$$

in the perturbative approach would induce corrections to the Fermi-liquid features due to the presence of the nontrivial order parameter in the *localized* subsystem. Then, one may attempt to establish a correspondence between the perturbations induced by the nontrivial localized spin order, and possible intrinsic instabilities for the electronic subsystem. In general, such perturbations involve electronic states far away from the Fermi surface and there is no distinct electronic mechanism favoring the formation of a specific phase. However, there are two exceptions. First, for the transitions without translational symmetry breaking ($\mathbf{Q} = 0$) and, second, when the electronic spectrum has a nesting property $\epsilon(\mathbf{p}) \approx -\epsilon(\mathbf{p} + \mathbf{Q})$.

Consider first the case of $\mathbf{Q} = 0$. In the Fermi-liquid approach, deviations from the equilibrium are described with the help of the Landau function

$$\hat{f}(\mathbf{p}, \mathbf{p}') = f^s(\mathbf{p}, \mathbf{p}') + \hat{\boldsymbol{\sigma}} \cdot \hat{\boldsymbol{\sigma}}' f^a(\mathbf{p}, \mathbf{p}')$$
(4)

 $(\hat{\sigma}$ are the Pauli matrices), which can be decomposed into the sum over all irreducible representations of the crystal point group:

$$f^{s(a)}(\mathbf{p}, \mathbf{p}') = \sum_{i} f_{i}^{s(a)} u_{i}(\mathbf{p}) u_{i}(\mathbf{p}'), \qquad (5)$$

where $u_i(\mathbf{p})$ is a complete set of the basis functions. The Fermi liquid is stable if

$$1 + N(0)f_i^s > 0, \quad 1 + N(0)f_i^a > 0 \tag{6}$$

for any i [9] [N(0) is a density of states at the Fermi surface]. As has been recently discussed [4], this approach can be useful in classifying the nontrivial phase transitions for the itinerant system without the unit cell change. If one of the stability conditions (6) is broken, a new symmetry distortion for the equilibrium Fermi surface develops. Such a phase transition is called a Pomeranchuk instability [9]. The energetic mechanism is expressed here in terms of the Fermi surface reconstruction due to the self-consistent interaction between all electrons as it is described by the Landau function. The corresponding variation of the quasiparticle occupation numbers compared to the one for the higher symmetry phase is either spin symmetric,

$$\delta \hat{n}(\mathbf{p}) = \hat{1} \Delta^s(\mathbf{p}) \,, \tag{7}$$

or spin antisymmetric,

$$\delta \hat{n}(\mathbf{p}) = (\hat{\boldsymbol{\sigma}} \cdot \mathbf{d}) \,\Delta^a(\mathbf{p}),\tag{8}$$

where $\Delta^{s}(\mathbf{p})$ or $\Delta^{a}(\mathbf{p})$ transforms as one of the representations, $u_{i}(\mathbf{p})$. For a spin-symmetric instability, the spin rotational symmetry is preserved. As for the spinantisymmetric phase, it is ferromagnetic for the identical representation and, as one can show explicitly, antiferromagnetic for any nonidentical and even $\Delta^a(\mathbf{p}) = \Delta^a(-\mathbf{p})$ representation (the magnetic cell coincides with the lattice unit cell).

The phase (8) with odd $\Delta^a(\mathbf{p}) = -\Delta^a(-\mathbf{p})$ is neither ferromagnetic nor antiferromagnetic [10]. Essentially, it is a "spin-split" state [11] with different Fermi surfaces for different spin projections. (This state seems to possess a macroscopic spin current [11]. An arbitrarily small dissipation eliminates this current, while the symmetry remains intact.) The time reversal symmetry is preserved while the spin rotation symmetry is broken with only the axial symmetry in the spin space; i.e., the symmetry is actually the same as in the *p*-type spin nematic state for the localized spins characterized by the order parameter

$$\mathbf{P}_{12} = \langle [\mathbf{S}_1 \times \mathbf{S}_2] \rangle, \quad \mathbf{P}_{12} = -\mathbf{P}_{21} \tag{9}$$

 $[\mathbf{P}_{12} = \mathbf{P}(\mathbf{r}_1 - \mathbf{r}_2)$ for $\mathbf{Q} = 0]$. For the itinerant system, the symmetry breaking (8) explicitly involves spatial degrees of freedom while for the localized spins the analogous role is played by the change in the spin Young scheme. As one can calculate, the itinerant instability introduces a twist for the localized spins

$$\langle [\mathbf{S}_1 \times \mathbf{S}_2] \rangle = \left(\frac{JS(S+1)}{3T} \right)^2 \\ \times n_{12} \frac{1}{N} \sum_{\mathbf{p}} \operatorname{Tr}[\hat{\boldsymbol{\sigma}} \delta \hat{n}(\mathbf{p})] \sin [\mathbf{p} \cdot \mathbf{r}_{21}] \quad (10)$$

(for the sake of simplicity, we neglect the interactions in the localized subsystem in this formula), where $\mathbf{r}_{ab} = \mathbf{r}_a - \mathbf{r}_b$, and

$$n_{ab} = \frac{1}{N} \sum_{\mathbf{p}} n(\mathbf{p}) \cos\left[\mathbf{p} \cdot \mathbf{r}_{ab}\right]$$
(11)

is a Fourier transform of the unperturbed Fermi occupation number $n(\mathbf{p})$, although the genuine order parameter here is the distortion of the Fermi surfaces for different spin projections. When both the itinerant and localized subsystems coexist, the order parameters induce each other through a term in a free energy linear in both \mathbf{P}_{12} and $\delta \hat{n}(\mathbf{p})$. All of this allows us to call the odd spin-antisymmetric Fermi-liquid instability (8) an *itinerant p-type spin nematic.*

Consider now the spin-symmetric instability (7) with odd $\Delta^s(\mathbf{p}) = -\Delta^s(-\mathbf{p})$. Once again, some of the order parameters of this kind seem to allow an energetically unfavorable persistent macroscopic current. However, exactly as above, a dissipation would eliminate the current without changing the symmetry itself. Depending on the representation, currents may not appear at all if a pseudovector (i.e., the orbital moment) is not allowed by the representation (for instance, the representation invariant with respect to all rotations and odd under the space inversion). Unlike the previous case, this state does not involve electronic spin degrees of freedom at all. However, as one can show, the symmetry here corresponds to that of the chiral spin state for localized spins which has the following order parameter:

$$\mathsf{R}_{123} = \epsilon^{\alpha\beta\gamma} \langle S_1^{\alpha} S_2^{\beta} S_3^{\gamma} \rangle. \tag{12}$$

As in the case of a p-type spin nematic, if the localized and itinerant subsystems coexist, the order parameters induce each other so that the anomalous local spin correlator for the itinerant-driven transition is

$$R_{123} = \frac{3}{2} \left(\frac{JS(S+1)}{3T} \right)^{3}$$
$$\times n_{12}n_{23} \frac{1}{N} \sum_{\mathbf{p}} \operatorname{Tr}[\delta \hat{n}(\mathbf{p})] \sin (\mathbf{p} \cdot \mathbf{r}_{31})$$
$$+ \text{cyclic permutations of (123)}$$
(13)

 $[n_{ab}$ is determined by (11)]. Correspondingly, one can call this state an *itinerant chiral spin state*.

Turn now to the case of broken translational symmetry ($\mathbf{Q} \neq 0$). The coupling interaction (3) introduces matrix elements between electronic states with wave vectors \mathbf{p} and $\mathbf{p} + \mathbf{Q}$. The problem involves states far away from the Fermi surface unless it has a nesting property $\epsilon(\mathbf{p}) \approx -\epsilon(\mathbf{p} + \mathbf{Q})$. In this case, the reconstruction of the electronic spectrum near the nested parts of the Fermi surface is energetically most important. As is well known, such a system can be unstable with respect to the electron-hole condensation where the order parameter is

$$F_{\sigma\sigma'}(\mathbf{p}) = -\langle T_{\tau} a_{\sigma}(\mathbf{p}) a_{\sigma'}^{\dagger}(\mathbf{p} + \mathbf{Q}) \rangle.$$
(14)

The symmetry with respect to spin rotations allows the singlet or triplet form of (14):

$$\hat{F}(\mathbf{p}) = \hat{1}F^{s}(\mathbf{p}) \quad \text{or} \quad \hat{F}(\mathbf{p}) = (\hat{\boldsymbol{\sigma}} \cdot \mathbf{d})F^{t}(\mathbf{p}), \qquad (15)$$

where $F(\mathbf{p})$ transforms as an irreducible representation of the small group of \mathbf{Q} . In what follows, we assume the commensurate vector \mathbf{Q} (2 \mathbf{Q} is a reciprocal lattice vector). If the representation was identical, $F^s(\mathbf{p})$ in (15) would describe the charge density wave (CDW) while $F^t(\mathbf{p})$ means the spin density wave (SDW) formation.

The transition manifests itself in the divergence of the electron vertex part $\Gamma(12; 34)$. In the weak coupling regime, there is a large parameter $\ln(\epsilon_F/T_0) \gg 1$ (T_0 is a transition temperature) which allows one to solve the problem rigorously in a justified approximation. The intersite Coulomb and exchange interactions favor the development of the phases with nonidentical representations [12]. In some cases, such phases are accompanied by the persistent staggered microscopic spin or charge currents [13]. Time reversal transforms electronic annihilation and creation operators as follows: $a_{\sigma'}(\mathbf{p}) \rightarrow g_{\sigma'\sigma}a^{\dagger}_{\sigma}(-\mathbf{p}), a^{\dagger}_{\sigma'}(\mathbf{p}) \rightarrow g_{\sigma'\sigma}a_{\sigma}(-\mathbf{p})$, where $g_{\sigma'\sigma} = i\sigma^{y}_{\sigma'\sigma}$, so that $F^{s}(\mathbf{p}) \rightarrow F^{s}(\mathbf{Q} - \mathbf{p})$ and $F^{t}(\mathbf{p}) \rightarrow -F^{t}(\mathbf{Q} - \mathbf{p})$.

Consider the time reversal invariant triplet order parameter $[F^t(\mathbf{p}) = -F^t(\mathbf{Q} - \mathbf{p})]$. Like the case of $\mathbf{Q} = 0$, this phase retains axial symmetry in a spin space and time reversal invariance, and therefore is analogous by symmetry to the *p*-type spin nematic [14]. If the spin

$$\mathbf{P}(\mathbf{r}_1 + \mathbf{R}, \mathbf{r}_2 + \mathbf{R}) = \exp\left[i\mathbf{Q}\cdot\mathbf{R}\right]\mathbf{P}_{12}$$
(16)

 $(\exp [2i\mathbf{Q} \cdot \mathbf{R}] \equiv 1)$, the existence of the electron-hole condensate results in the anomalous average for the local spins

$$\langle [\mathbf{S}_1 \times \mathbf{S}_2] \rangle = 2 \left(\frac{JS(S+1)}{3T} \right)^2 \\ \times n_{12} \frac{1}{N} \sum_{\mathbf{p}} [i\mathbf{F}(\mathbf{p})] e^{-i(\mathbf{p}+\mathbf{Q})\cdot\mathbf{r}_2 + i\mathbf{p}\cdot\mathbf{r}_1} \quad (17)$$

[the average (17) is always real]. Therefore, exactly as the spin-antisymmetric and odd Pomeranchuk instability (8) is a generalization of the *p*-type spin nematic phase for a transition without a change of the unit cell, the above electron-hole pairing is an analog of the *p*-type spin nematic phase with a doubling of the unit cell.

Unlike for $\mathbf{Q} = 0$, the action of the lattice inversion $\mathbf{r} \to -\mathbf{r}$ is generally different from that of time reversal. The former operation transforms $F^t(\mathbf{p})$ to $F^t(-\mathbf{p})$ while the latter connects $F^t(\mathbf{p})$ with $-F^t(\mathbf{Q} - \mathbf{p})$. Hence, for the lattice with center of symmetry there are two different kinds of itinerant spin nematic phases: $F^t(\mathbf{p}) = F^t(-\mathbf{p})$ and $F^t(\mathbf{p}) = -F^t(-\mathbf{p})$. In the former case, $F^t(\mathbf{p})$ is imaginary while in the latter case it is real. In both cases the parity is broken: $\mathbf{P}_{12} = -\mathbf{P}_{21}$. However, for the even instability $\mathbf{P}_{\mathbf{r}_1,\mathbf{r}_2} = -\mathbf{P}_{-\mathbf{r}_1,-\mathbf{r}_2}$ while for the odd instability $\mathbf{P}_{\mathbf{r}_1,\mathbf{r}_2} = -\mathbf{P}_{-\mathbf{r}_1,-\mathbf{r}_2}$.

As was mentioned, for the quasi-one-dimensional model in the weak coupling regime, there is a well-defined large parameter, $\ln(\epsilon_F/T_0) \gg 1$, so that the mean field approach becomes a well justified approximation. Therefore the low-temperature properties of the itinerant spin nematic phase can be obtained in the same manner as for the spin density wave transitions. Unlike the case of Pomeranchuk instability for $\mathbf{Q} = 0$, the electronic spectrum possesses a gap near the nested regions of the Fermi surface. Below the transition point, spin wave excitations with linear dispersion appear due to the continuous symmetry breaking. The macroscopic properties of the spin nematic phase such as the specific heat or the bulk spin susceptibility χ_0 are indistinguishable from those for a SDW [2, 6]. It is known that the χ_0 stops being isotropic once the magnetic anisotropy energy fixes the direction of **d**. One can calculate that in the nesting model, $\chi_0(\mathbf{H} \| \mathbf{d})$ decreases as temperature decreases and vanishes at T = 0 while $\chi_0(\mathbf{H} \perp \mathbf{d})$ does not tend to zero at small temperatures. At the same time, the properties near the nesting wave vector of the spin nematic phase are qualitatively different from those of the spin density wave. For instance, the staggered susceptibility $\chi(\mathbf{Q})$ which for the spectrum with nesting increases as temperature decreases would saturate at the value of the order of $\chi_0 \ln(\epsilon_F/T_0) \gg \chi_0$ for the spin nematic transition, while for the SDW it diverges, resulting in the onset of long-range antiferromagnetic order. As one can

show, the dynamic response function $\chi''(\mathbf{q}, \omega)$ which determines the inelastic neutron scattering intensity and the NMR relaxation rate develops a peak at $\mathbf{q} = \mathbf{Q}$ as temperature decreases. The temperature dependence of the relaxation rate has a maximum below T_0 . This maximum is due to the quasiparticle scattering with negligible energy transfer near the gap edge where the density of states is divergent, and in this sense is similar to the Hebel-Slichter peak for the superconducting transition. Below the transition $\chi''(\mathbf{q}, \omega)$ is anisotropic. However, the NMR line is not shifted, unlike for the SDW.

Consider now the case of the singlet electron-hole order parameter which breaks time reversal symmetry $[F^s(\mathbf{p}) = -F^s(\mathbf{Q} - \mathbf{p})]$. Generally, there could be a microscopic staggered current which creates staggered orbital magnetic moments although the macroscopic current is absent (this phenomenon is known as the orbital antiferromagnetism [13]). Whether the average local orbital magnetic moments are zero or not depends on the choice of the representation (see above). The same as for $\mathbf{Q} = 0$, the itinerant and chiral spin order parameters induce each other through the term in the free energy linear in both R_{123} and $F^s(\mathbf{p})$; the anomalous local spins correlator is

$$R_{123} = 3 \left(\frac{JS(S+1)}{3T} \right)^{3}$$

$$\times n_{12}n_{23} \frac{1}{N} \sum_{\mathbf{p}} \left[iF^{s}(\mathbf{p}) \right] e^{-i(\mathbf{p}+\mathbf{Q})\cdot\mathbf{r}_{3}+i\mathbf{p}\cdot\mathbf{r}_{1}}$$

$$+ \text{cyclic permutations of (123)}$$
(18)

 $(\text{Im }R_{123}=0)$, and this itinerant phase is an analog of the chiral spin state with a doubled unit cell.

In the case of the singlet electron-hole wave function for the lattice with the center of symmetry, odd $F^{s}(\mathbf{p}) = -F^{s}(-\mathbf{p})$ is real while even $F^{s}(\mathbf{p}) = F^{s}(-\mathbf{p})$ is imaginary, and the anomalous spin average R_{123} , correspondingly, changes sign with $\mathbf{r} \to -\mathbf{r}$ or remains invariant. All properties of this phase can again be calculated rigorously. χ_{0} remains isotropic and decreases rapidly below the transition while the staggered susceptibility $\chi(\mathbf{Q})$ saturates. Like above, there is a peak at \mathbf{Q} in the isotropic dynamic response function as well as a Hebel-Slichter-type maximum in the NMR relaxation rate below T_{0} .

To summarize, we have shown that in the exchange approximation only the chiral spin state and the *p*-type spin nematic state influence the electronic spectrum near the Fermi surface. In the system with both localized spins and itinerant carriers, the above two phases may be the result of either the Pomeranchuk instability if $\mathbf{Q} = 0$, or the electron-hole condensation near the nested Fermi surfaces if \mathbf{Q} is commensurate. Note that in the heavy fermions the magnetic anisotropy is large, the exchange approximation is not applicable, and, generally, the Fermi surface can also be sensitive to other order parameters. Experimentally, the transitions discussed would be of the second order and similar to the ordinary lattice or AFM instabilities, as concerns, say, the specific-heat and bulk susceptibility anomalies. The above phases may exist not only in the heavy fermion and high- T_c materials, but also in intercalation compounds and organic conductors.

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