Odd time magnetic correlations and chiral spin nematics

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Abstract

The magnetic analog of odd-time superconducting order is introduced. It is shown that the classification of possible odd-time magnetic states admits the existence of a novel state - the chiral spin nematic - which is characterized by an odd-in-time spin-spin correlation function. The known example of the chiral spin liquid appears naturally in this approach. The equal-time correlation function of these odd-in-time states involves non-trivial three spin correlations.
The question of broken symmetry states characterized by some correlation function which is odd in time, was revived recently in the context of superconductivity. An odd-time, or equivalently, odd-frequency, superconducting ordering was introduced in the pioneering work of Berezinskii [1], who considered spin-triplet odd-time pairing as an alternative to the conventional theory of $^3$He pairing. Interest in odd-time ordering was renewed after the work of Balatsky and Abrahams [2], in which the extension of Ref. [1] to odd-time pairing for spin-singlet superconductors was given.

In this paper, in analogy to odd-time superconductivity, we introduce the concept of magnetic states with odd-time correlations. We shall discuss the symmetry constraints for such states and shall classify the different possibilities.

For odd-time superconductivity, the possibility of odd-time states follows immediately from the basic symmetry constraint [2] $F(k, t) = F(-k, -t)$ for the general spin-singlet anomalous correlator $F(k, t) = i(\sigma^y)_{\alpha\beta} \langle T_t c_{\alpha}(t)c_{-\alpha}(0) \rangle$. This symmetry constraint is an immediate consequence of the Fermi statistics of the $c_{\alpha}$ operators and of the singlet nature of the anomalous correlator. The consideration for spin-triplet pairing is analogous and was done by Berezinskii [1]. Then one finds two classes of states: 1) even-time pairing (BCS) states and 2) odd-time pairing states with $F(k, -t) = -F(k, t)$. The orbital parities of these states are opposite [even(odd) and odd(even) for singlet(triplet) respectively] as follows from the symmetry equation.

The generalization of odd-time ordering to spin systems is straightforward and requires the symmetry equation for the dynamic correlation function of the spin density $S_i(r, t)$ where $i = 1, 2, 3$ denotes cartesian components. The spin-spin correlation function is $\Lambda_{ij}(r, r'|t) = \langle T_t S_i(r, t)S_j(r', 0) \rangle$. The symmetry equation which follows directly from the properties of the time-ordering operator in the definition of $\Lambda_{ij}$ is $\Lambda_{ij}(r, r'|t) = \Lambda_{ji}(r', r|-t)$. In matrix form, this is:

$$PT \tilde{\Lambda}(r, r'|t) = \Lambda(r, r'|t), \quad (1)$$

where $P$ is the spatial parity operator $r - r' \to r' - r$, $T$ is the time inversion and $\tilde{\Lambda}$ is the
transposed matrix $\tilde{\Lambda}_{ij} = \Lambda_{ji}$. This equation, valid for any rank spin $S$, allows the existence of odd-time magnetic states. These states are characterized by a nontrivial time structure of the spin-spin correlation function with $\Lambda_{ij}(r, r'|t)$ being an odd function of time.

The main purpose of this paper is to classify magnetic states with odd-time magnetic correlations. We shall give the symmetry analysis for possible odd-time magnets. It follows closely that for even-time magnets which have nontrivial spin correlation functions. All the known chiral states are recovered in this approach: The chiral spin liquid (CSL) and the ferromagnet (a rather dull example of a chiral state) are among them. A new magnetic state is found as well: It is a state which is the odd-time analog of the spin nematic. The spin nematic was first considered by Andreev and Grishchuk [3]. The new odd-time state is also characterized by nematic ordering in spin space [3], but with broken $T$ and $P$. We call this state a “chiral spin nematic” (CSN). We shall discuss the physical properties of this state and we shall show that the low energy lagrangian is identical to that for a conventional spin nematic [3].

We begin with the symmetry of the magnetic correlator. We consider states in which spontaneous breakdown of the $O(3)$ spin rotation group occurs without the appearance of an average microscopic spin density so that $\langle S_i(r, t) \rangle = 0$. This restriction leads us to consider a spin-spin correlation function $\Lambda_{ij}(r, r'|t)$, which is time-dependent and in general is a function of two coordinates $r, r'$. As shown by Andreev and Grishchuk [3], spin-spin correlations without time-reversal violation can still describe a nontrivial magnetic order, i.e. a spin nematic. In that case, $\Lambda_{ij}(r, r'|0)$ transforms as a tensor representation of $O(3)$. This will also be the case for odd-time correlations described by $\Lambda_{ij}$ for $t \neq 0$.

Following the analysis of the Andreev and Grishchuk [3], we consider the invariance group of the system. It is given by the product $O(3) \times T \times G$, where $O(3)$ is the spin rotation group, $G$ is the space group of the lattice and $T$ is time reversal, equivalent to spin inversion. $G$ defines the symmetry of spin scalars; it contains the crystal point group and the translations of the lattice.
\( \Lambda_{ij} \) is a \( 3 \times 3 \) matrix containing 9 elements. It can be decomposed as a sum of terms which transform as particular representations of \( O(3) \). From the decomposition \( 9 = 1 + 3 + 5 \) we find that \( l = 0, 1, 2 \) can be present. Therefore we write:

\[
\Lambda_{ij}(\mathbf{r}, \mathbf{r}'|t) = A(\mathbf{r}, \mathbf{r}'|t)\delta_{ij} + \epsilon_{ijk}B_k(\mathbf{r}, \mathbf{r}'|t) + Q_{ij}(\mathbf{r}, \mathbf{r}'|t). \tag{2}
\]

where \( A(\mathbf{r}, \mathbf{r}'|t) \) is a scalar \( (l = 0) \) and is the only term which contributes to the trace of \( \Lambda_{ij}(\mathbf{r}, \mathbf{r}'|t) \). \( B_k(\mathbf{r}, \mathbf{r}'|t) \) is a vector \( (l = 1) \) and \( Q_{ij}(\mathbf{r}, \mathbf{r}'|t) \) is the tensor part \( (l = 2) \). The latter is symmetric and traceless. The quantities \( A, B_k \) and \( Q_{ij} \) describe a spin liquid with no ordering, magnetic ordering with a pseudovector order parameter and spin nematic ordering, respectively. This is the case whatever the parity and time reversal properties of these objects.

For the case of the even-time states, the order parameter can be found from the equal-time correlator \( \Lambda_{ij}(\mathbf{r}, \mathbf{r}'|t = 0) \). Then we recover the classification of Ref. [3]. In the even-time case, \( \Lambda_{ij}(\mathbf{r}, \mathbf{r}'|t) \) in Eq. (2) is even under time reversal. Then \( B_k(\mathbf{r}, \mathbf{r}'|t) \) and \( Q_{ij}(\mathbf{r}, \mathbf{r}'|t) \) describe two different nematic states, the so-called \( p \) and \( n \) nematics respectively [3]. The \( A \)-term describes a spin liquid with no spin ordering. Complete details for the even-time case may be found in Ref. [3].

We now turn to odd-time correlations. In the case of odd-time superconductivity, it is well-established [5–8] that the equal-time correlators which describe the condensate contain composite operators which involve a conventional Cooper pair bound with, for example, a spin-density operator. In the magnetic case, we shall implement exactly the same approach and the odd-time magnetic correlators will describe the condensate of the higher order spin operators. For any \( \Lambda_{ij}(\mathbf{r}, \mathbf{r}'|t) \) which is odd in time, the time derivative will be nonzero (we assume analyticity of \( \Lambda_{ij} \) at small \( t \)) at \( t = 0 \) and represents a thermodynamic order parameter for a composite condensate:

\[
\partial_t \Lambda_{ij}(\mathbf{r}, \mathbf{r}'|t)_{t=0} = \langle T_t[\partial_t S_i(\mathbf{r}, t)], S_j(\mathbf{r}', 0) \rangle_{t=0} \tag{3}
\]

The equation of motion for \( S_i \) can be used to find the general result
\[ \partial_t S_i(r, t) = i[H, S_i(r, t)] = \epsilon_{ilm} S_l(r, t) M_m(r, t), \]  

(4)

where \( M_m(r, t) \) is the “molecular field” for the hamiltonian \( H \). If the hamiltonian is bilinear in spin operators, the general form of the molecular field will be

\[ M_m(r) = \int dr' K_{mn}(r, r') S_n(r'). \]  

(5)

The coupling \( K_{mn}(r, r') \) is an explicit function of the two co-ordinates \( r, r' \). Finally, we obtain

\[ \partial_t \Lambda_{ij}(r, r'|t=0) = \epsilon_{ilm} \int dr'' K_{mn}(r, r'') \langle S_l(r) S_n(r'') S_j(r') \rangle. \]  

(6)

If the hamiltonian has the form

\[ \mathcal{H} = -\sum_{mn} \int drdr' S_m(r) L_{mn}(r, r') S_n(r'), \]  

(7)

then the molecular field kernel is given by

\[ K_{mn}(r, r') = L_{mn}(r, r') + L_{nm}(r', r) = 2L_{mn}(r, r'). \]  

(8)

We see that any pure pairwise exchange terms of the hamiltonian will not contribute to the time derivative of odd-time correlators, since the corresponding kernel \( K_{mn}(r, r'') \) for pairwise exchange is symmetric in \( r, r'' \), while the correlator on the r.h.s of Eq. (6) is antisymmetric. Therefore for a non-zero result, \( K(r, r'') \) must contain a spatially-odd component. We shall give an explicit example of a hamiltonian which produces these equations of motion for the specific case of a CSL. In general, complicated interactions are required for chiral magnetic order.

We now discuss the specific phases which can be found in odd-time magnets. The possible terms in the decomposition Eq. (2) will be considered in turn.

1). Chiral Spin Liquid. Consider a spin-spin correlator \( \Lambda_{ij} \) containing only the \( A \)-term in the r.h.s. of Eq. (2). The spin scalar term \( A(r, r'|t) \) is odd in time and, therefore is odd under spatial parity \( P \), as can be easily seen from the symmetry equation (1) for any cartesian-symmetric odd-time \( \Lambda_{ij} \). Taking the time derivative as in Eqs. (3-6), we find
\[ \partial_t A(r, r')|0 = \int dr'' K(r, r'') \langle S(r) \times S(r'') \cdot S(r') \rangle, \]  

where, for the assumed CSL state with no anisotropy in spin space, we have taken 
\[ K_{mn}(r, r'') = \delta_{mn} K(r, r''). \]  
Note that \[ \partial_t A(r, r'|0 \) obtained in this way is explicitly odd under \( P \).

The necessary condition for the odd-time CSL phase is the existence of a non-zero correlator 
\[ X_P(r_1 r_2 r_3) = \langle S(r_1) \times S(r_2) \cdot S(r_3) \rangle, \]  
which when defined on a plaquette is exactly the usual CSL order parameter \[ 9 \]. We argue, therefore, that an alternative way to describe a CSL phase is in terms of an odd-time spin singlet correlator 
\[ A(r, r'|t) = \langle S_i(r_t) S_i(r'') |0 \rangle \]  
The well-known picture of the CSL state as the state where all spins precess so that local handedness is preserved is naturally supported in this description. In order for the time derivative \( \partial_t A(t = 0) \) to exist, the coupling \( K(r, r') \) should contain a part which is odd in \( r - r' \). This puts a severe constraint on the possible exchange models which can support the CSL ground state. One of them is given by the hamiltonian \[ 10 \]
\[ H = -\frac{\lambda}{2} \sum_{\langle ij \rangle} [S_1 \times S_2 \cdot S_3]_{P_i} [S_4 \times S_5 \cdot S_6]_{P_j}, \]  
with \( \lambda \)-positive. The sum runs over nearest neighbor plaquettes \( P_i, P_j \) with spins 1, 2, 3 and 4, 5, 6 belonging to the plaquettes \( P_i \) and \( P_j \) respectively \[ 9 \] and we consider here the triangular lattice. This hamiltonian has a CSL ground state with ferromagnetic ordering of chiralities with nonzero \( X_P(r_1, r_2, r_3) \). It can be shown that the equations of motion obtained from this hamiltonian do indeed lead to Eq. \( 9 \) for \( \partial_t A \), where the spin scalar kernel \( K(r, r') \) is a certain combination of the pair spin \( \langle S(r_i) S(r_j) \rangle \) correlation functions.

2). Ferromagnet. Consider the \( B_k \) term in Eq. \( 3 \). \( B_k \) transforms as a vector under the spin rotation group \( O(3) \); it is even under spatial inversion \( P \) and odd under time reversal \( T \), as follows from Eq. \( 4 \). A pairwise exchange interaction hamiltonian, therefore, is sufficient for the present discussion. One can take \( H = \frac{1}{2} \int dr dr' K(r, r') S(r) \cdot S(r') \) following the same approach as for the CSL case, we find for the time derivative \( \partial_t B \)
\[ \partial_t B(r, r'|0) = \int dr'' K(r, r'') \{ \langle (S(r'') \cdot S(r')) S(r) \rangle - S^2 \langle S(r''') \rangle \}. \]  

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Therefore, we conclude that a nonzero $B$-term in the decomposition of Eq. (2) describes a vector ordering $\langle \mathbf{S}(\mathbf{r}) \rangle \neq 0$. This is trivial ferromagnetic order. In particular, for $\mathbf{r} = \mathbf{r}'$ it follows from Eq. (11) that $\partial_t B(\mathbf{r}, \mathbf{r}|0) \propto \langle \mathbf{S}(\mathbf{r}) \rangle$. The ferromagnet corresponds to the reducible three-spin correlator in Eq. (3) and can be described in terms of a single spin operator expectation value. This contradicts our initial restriction to states having no average spin density and we do not consider this ferromagnetic case any further.

3). Chiral Spin Nematic. As remarked earlier, the last term in the decomposition of Eq. (2) represents a spin nematic order belonging to the tensor $l = 2$ representation of $O(3)$. The crucial difference from previously considered spin nematics [3] is that in our case $Q_{ij}(\mathbf{r}, \mathbf{r}'|t)$ is an odd function of $t$ and therefore it is an odd function under $P$. In the presence of an external magnetic field, which expressly violates time reversal and spin rotation symmetries, a $P$-odd nematic state was considered by Chubukov [12]. In our considerations, however, the external magnetic field is zero.

The parity operation is equivalent to interchange of $\mathbf{r}, \mathbf{r}'$. The conventional spin nematic with $Q_{ij}(\mathbf{r}, \mathbf{r}')$ considered at equal times is even under $P$, as follows from Eq. (1) for $\Lambda_{ij}(\mathbf{r}, \mathbf{r}'|t) = Q_{ij}(\mathbf{r}, \mathbf{r}'|t)$. We emphasize this distinction because it shows that the CSN state cannot be obtained within the old classification scheme [3] and is a new type of state.

As before, we take the time derivative to write the equal-time correlator. We find

$$L_{ij}(\mathbf{r}, \mathbf{r}') = \partial_t Q_{ij}(\mathbf{r}, \mathbf{r}'|0) = \int d\mathbf{r}''K(\mathbf{r}, \mathbf{r}'')\epsilon_{iln}V_{lnj}(\mathbf{r}, \mathbf{r}'', \mathbf{r}').$$

Here $V_{lnj}(\mathbf{r}, \mathbf{r}'', \mathbf{r}') = \langle S_i(\mathbf{r})S_n(\mathbf{r}'')S_j(\mathbf{r}') \rangle$ is the three-spin correlator and an isotropic tensor $K_{ij} = \delta_{ij}K$ was taken. As mentioned above, only the $P$-odd part of $K(\mathbf{r}, \mathbf{r}'')$ contributes to the integral and this constraint requires special models for exchange interactions.

The transformation properties of $V_{lnj}$ under permutation of the spin indices are given by a particular representation of the permutation group of three objects $S_3$ and in this particular case it is the $l = 2$ representation. The symmetry of this tensor can be seen from Eq. (12): $V_{lnj} = V_{[ln]j}$, where the square brackets stand for antisymmetrization with respect to the indices contained within. The even-time totally symmetric three-spin correlator has been
considered by Gor’kov [11]. This symmetry is obvious from the fact that the product \( \epsilon_{ln} V_{lnj} \) enters Eq. (12). On the other hand, the time derivative \( \partial_t Q_{ij} = L_{ij} \) yields the symmetric traceless tensor \( L_{ij} \). Therefore, the product \( \epsilon_{ln} V_{lnj} \) should be symmetrized with respect to \( i, j \). In what follows, we limit ourselves to the case of uniaxial solutions for the CSN. An allowed solution is

\[
L_{ij}(r, r') = L(r, r')(n_i n_j - \frac{1}{3} \delta_{ij}),
\]

where \( n \) is the nematic director (\( n \) and \(-n\) are equivalent) in spin space. The director specifies the plane of the spin modulation, as in the conventional spin nematic. Although the form of \( L_{ij} \) in the above equation is identical to that for the conventional even-time spin nematic with \( Q_{ij}(r, r') = Q(r, r')(n_i n_j - \frac{1}{3} \delta_{ij}) \) [3], here the physics is fundamentally different. The odd-time \( L_{ij} \) is inherently connected to the three spin correlator \( V_{lnj}(r, r'', r') \) in Eq. (12), in contrast to a two spin correlator as in the conventional spin nematic. In addition, the spatial parity of \( L_{ij} \) is opposite to the parity of the standard spin nematic \( Q_{ij} \). 

In the exchange approximation, the coordinate and spin indices are independent and one can factorize \( V_{ijm} : \langle S_i(r_1) S_n(r_2) S_j(r_3) \rangle = T_{lnj} \Phi(r_1, r_2, r_3) \). The spin part is \( T_{lnj} = \epsilon_{ln}(n_i n_j - \frac{1}{3} \delta_{ij}) \), and the orbital part \( \Phi \) is to be integrated with \( K(r, r') \) in Eq. (12) to yield \( L(r, r') \). One finds

\[
Q_{ij}(r, r'|t) = Q(r, r'|t)(n_i n_j - \frac{1}{3} \delta_{ij}),
\]

\[
\partial_t Q(r, r'|t) = L(r, r').
\]

The lagrangian for the CSN state is identical to the lagrangian of the conventional spin nematics [3] and is

\[
\mathcal{L} = \frac{1}{2\gamma^2} \chi_{ij}(n \times \partial_t n + \gamma H)_i(n \times \partial_t n + \gamma H)_j - J_{kl} \partial_k n \partial_l n - U_{anis},
\]

where \( \gamma \) is the gyromagnetic ratio, \( H \) is the external magnetic field, \( J_{ij} \) is the inhomogeneous exchange tensor, \( \chi_{ij} = \chi_{\perp}(\delta_{ij} + n_i n_j) \) is the transverse susceptibility tensor, and \( U_{anis} \) is the relativistic anisotropy energy. Linear gradient terms are not allowed in the lagrangian.
We mention briefly possible ways to generate a CSN phase. To lower the symmetry of the system down to the CSN one can consider the quadrupolar interaction in the CSL phase \( l = 0 \), which will generate the nematic \( l = 2 \) component. In the \( P, T \) violating ground state this nematic state will have a chiral component. Another possibility is spontaneous breakdown of \( P \) and \( T \) in an already existing nematic state.

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REFERENCES

* Also at the Landau Institute for Theoretical Physics, Moscow, Russia.

[1] V.L. Berezinskii, JETP Lett. 20, 287 (1974) [ZhETP Pisma 20, 628 (1974)].

[2] A.V. Balatsky and E. Abrahams, Phys. Rev. B 45, 13125 (1992).

[3] A.F. Andreev and I.A. Grishchuk, Sov. Phys. JETP 60, 267 (1984) [Zh. Eksp. Teor. Fiz. 87 467 (1984)].

[4] E. Abrahams, A.V. Balatsky, J.R. Schrieffer and P. B. Allen, Phys. Rev. B47, 513, (1993).

[5] E. Abrahams, A.V. Balatsky, D.J. Scalapino and J.R. Schrieffer, unpublished.

[6] P. Coleman, E. Miranda and A. Tsvelik, Phys. Rev. Lett. 70, 2960, (1993).

[7] V.J. Emery and S. Kivelson, Phys. Rev. B46, 10812, (1992).

[8] A.V. Balatsky and J. Bonca, Phys. Rev. B47, 7445, (1993).

[9] X.G. Wen, F. Wilczek and A. Zee, Phys. Rev. B39, 11413, (1989). V.I. Marchenko, Pisma ZhETP, 48, 387, (1988) (JETP Lett., 48, 427, (1989)).

[10] Here the simplest possible model has been assumed to illustrate the symmetry statements to be made about different types of odd-time magnetic states. The microscopic origin of the model Hamiltonian will not be considered here. More complicated Hamiltonians are possible, such as to include other fields, i.e. electrons, in the problem. Also, for the higher spins another higher order invariants are possible as well. We will not address these questions here either.

[11] L.P. Gor’kov, Europhys. Lett., 16, 301, (1991)

[12] A. V. Chubukov, Phys. Rev. B44, 4693, (1991).