Spin nematics

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We investigate possible properties of exchange magnets in which the onset of magnetic order leads to spontaneous violation of the isotropy of the spin space, but invariance to time reversal is preserved. These magnets do not differ from antiferromagnets in their macroscopic magnetic properties and can be identified only by neutron scattering or NMR investigations. The possibility of similar ordering in the nuclear system of solid $^3$He is discussed.

The onset of magnetic order in exchange magnets is accompanied by spontaneous symmetry breaking relative to the spin-space rotation group $O(3)$. In ordinary magnets the character of the ordering is such that the order parameter representative of the coordinate dependence of the average microscopic spin density is always transformed in accord with the vector representation of the spin group $O(3)$, i.e., in particular, it reverses sign under spin inversion whose role is played by the time-reversal transformation. It is precisely this last property which is usually regarded as the general criterion of magnetic ordering. Magnets differ in accordance with which representation of the pure space group of the crystal the order parameter is transformed. This is the unitary representation for ferromagnets, the one-dimensional non-unitary representation for collinear antiferromagnets, and so on (see Ref. 1).

We analyze in this paper the possible properties of exchange systems in which magnetic ordering leads to spontaneous breaking of the spin group $O(3)$, but invariance to time reversal is preserved. By virtue of this last property, the average microscopic spin density vanishes in the systems considered. The order parameter should be introduced as a characteristic of the transformation properties of the spin correlation functions and described by the tensor representation of the spin group $O(3)$. The symmetry of the considered systems is similar to the symmetry of ordinary nematics, the only difference being that we are dealing here with spin rather than coordinate space. We shall show below on the basis of general symmetry considerations that despite the invariance to time reversal, the magnetic properties of "spin nematics" are indistinguishable from the properties of antiferromagnets. Namely, all the phenomena typical of antiferromagnets, such as flipping and collapse of sublattices or zero longitudinal susceptibility, a phase transition corresponding to sublattice flipping, etc. Quadrupole ordering is impossible for spins $s = 1/2$. The order parameter should be introduced as a characteristic of the symmetry of the ordering is such that the order parameter representative of the coordinate dependence of the average microscopic spin density is always transformed in accord with the vector representation of the spin group $O(3)$, i.e., in particular, it reverses sign under spin inversion whose role is played by the time-reversal transformation. It is precisely this last property which is usually regarded as the general criterion of magnetic ordering. Magnets differ in accordance with which representation of the pure space group of the crystal the order parameter is transformed. This is the unitary representation for ferromagnets, the one-dimensional non-unitary representation for collinear antiferromagnets, and so on (see Ref. 1).

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1. GENERAL PROPERTIES

Let $\langle \sigma_i(r) \sigma_j(r') \rangle$ be the spin density operator. In states with magnetic order invariant to time reversal, just as in the paramagnetic state, we have $\langle \sigma_i(r) \rangle = 0$. We consider the spin-correlation function $\langle \sigma_i(r) \sigma_j(r') \rangle$. In the paramagnetic state, by virtue of the invariance to the spin group $O(3)$, this function reduces to the spin scalar

$$\langle \sigma_i(r) \sigma_j(r') \rangle = \langle \sigma_i \sigma_j \rangle (r, r').$$

The nonscalar part of the correlation function

$$K_{\alpha}(r, r') = \langle \sigma_i \sigma_\alpha(r) \sigma_j \sigma'_\alpha(r') \rangle = \langle \sigma_i \sigma_\alpha \rangle (r, r')$$

can therefore be regarded as a characteristic of the symmetry of a magnetic order that is invariant to time reversal. Separating in $K_{\alpha}$ the parts symmetric and antisymmetric in the indices $i$ and $k$, we rewrite (1) in the form

$$K_{\alpha}(r, r') = \langle \sigma_i \sigma_\alpha \rangle (r, r') + \langle \sigma_i \sigma_\alpha \rangle (r, r'),$$

where $P_{\alpha}(r, r')$ are antisymmetric functions of the spatial arguments, and the $Q_{\alpha}$ are symmetric in the indices $i$ and $k$ as well as in $r$ and $r'$, with $Q_{\alpha} = 0$.

We do not engage here in classification in all the cases that are possible in principle. As is customarily done in the investigation of spontaneous breaking of the space group $O(3)$ in ordinary nematics, we confine ourselves to structures having an axis of total axial symmetry.

Just as any exchange magnetic system, a spin nematic is
The elements of a certain space group $G$ consisting of translations, whose elements are products of certain spin rotations by elements described by specifying the exchange symmetry

$$P(r, \vec{r}) = P(G, \vec{r})$$

at given initial $r$ and $\vec{r}$ as functions on a group $G$ and represent them in the form of a sum of quantities that transform in accordance with irreducible representations of the group $G$. With the aid of the method described by Marchenko and one of us it is easy to verify that for a structure with complete axial spin symmetry only one term in this sum differs from zero and corresponds to a certain one-dimensional (unitary or nonunitary) representation. This means that the structure in question can be characterized by an order parameter $p$ that transforms like a pseudovector under the action of the elements of the spin group $G(3)$ and in accord with one of the one-dimensional representations of the space group $G$. By virtue of the axial symmetry, the symmetric part $Q_{ss}$ of the correlator (2) should take in this case the form

$$Q_{ss}(r, \vec{r}) = q(r, \vec{r}) (\hat{p} \cdot \hat{n}) \delta(\vec{a})$$

where $n$ is a certain constant unit spin vector defined according to the sign, while $Q(r, \vec{r})$, as above, is a scalar with respect to all the transformations.

There exist thus two types of axisymmetric spin nematics, characterized by spin directors $p$ or $n$. In the first case $p$ is a spin pseudovector that transforms in accord with a certain one-dimensional representation of the group $G$, with $p$ and $-p$ different. In the second case $n$ can be regarded as a scalar relative to $G$, and the states $n$ and $-n$ are identical.

The macroscopic low-frequency properties of the spin nematics, particularly their behavior in magnetic fields that are weak compared with the exchange field, can be described on the basis of symmetry considerations alone. Reasoning similar to that expounded in Ref. 1 for the case of collinear antiferromagnets leads to the following Lagrange function:

$$L = \frac{1}{2} \sum_{i} \left[ \omega(s) \psi^{\dagger}(\psi^{\dagger} + \gamma \psi) + \frac{1}{2} \sum_{i} \left( \omega(s) \psi^{\dagger}(\psi^{\dagger} + \gamma \psi) \right) \right]$$

Here $H$ is the external magnetic field, $\gamma$ the gyromagnetic ratio, $\omega(s)$ the inhomogeneous exchange tensor,

$$\omega(s) = \omega(s) \hat{n}(s) \hat{n}(s)$$

the magnetic-susceptibility tensor, $\gamma$ the transverse susceptibility, and $U_{a,b}$ the relativistic anisotropy energy. The magnetization of a spin nematic is equal to

$$M = \langle \hat{M}(s) | \hat{M}(s) \rangle + \chi_{r} H_{r}$$

The foregoing equations, especially formula (6) according to which a spin nematic has no longitudinal susceptibility (at zero temperature), do not differ in substance from the corresponding equations for collinear antiferromagnets. The only difference is that, by virtue of the invariance of the state to time reversal, weak ferromagnetism is impossible in spin nematics; furthermore, Eq. (7) cannot contain additional terms that are linear in the spatial derivatives of the order parameter. Equations (5)-(7) in which $n$ is replaced by $p$, hold also for spin nematics with director $p$. Some difference between the two types of nematic is in the form of the anisotropy energy. In type-$p$ nematics, just as in antiferromagnets, the expansion of $U_{a,b}$ in powers of $n$ has no linear terms. In the case of antiferromagnets this is due to symmetry with respect to time reversal, and in the case of nematics to the identity of the states $n$ and $-n$. As a result, as usual, we have $U_{a,b} = (a_{a,b}/2m, n_{a,b})$, where $a_{a,b}$ is the anisotropy tensor $a_{a,b} = 0$. A similar formula with $n$ replaced by $p$ holds formally for $p$-type nematics only if the one-dimensional representation of the group $G$, according to which $p$ is transformed, is not contained in the pseudovector representation. Otherwise there exists a relativistic invariant linear in the components of $p$ and $U_{a,b} = a_{a,b}$, where $a_{a,b}$ are constants. Let, for example, $p$ transform in a uniaxial magnet like the $z$-component of a pseudovector. Then the anisotropy energy can be written in the form $U_{a,b} = a_{a,b}$. The energy minimum corresponds to $p_{z} = 1$ or $p_{z} = -1$, depending on the sign of $a_{z}$. At small deviations from equilibrium we have

$$U_{a,b} = \frac{1}{2} \sum_{i} (1-\gamma) \left( p_{a,b}^2 + p_{z}^2 \right)$$

which is analogous to antiferromagnets of the easy axis type.

2. EXAMPLES

Let exchange-interacting spins $s = 1$ be located at the sites of a crystal lattice. The most general pair interaction of the spins is described by the Hamiltonian

$$H = \sum_{i} \left[ J_{a} s_{a,i}^{\dagger} + J_{b} s_{b,i}^{\dagger} G_{a,b}^{(a,b)} s_{a,b} \right]$$

where $s_{a,b}^{\dagger}$ are matrices of unity spin,

$$G_{a,b}^{(a,b)} = \sum_{i} s_{a,b} \sum_{j} s_{a,b}^{\dagger} - \sum_{i} s_{a,b}^{\dagger} s_{a,b}$$

and the subscripts $a$ and $b$ number the lattice sites. Assuming that the exchange parameters $J_{a}$ and $J_{b}$ are positive and long-range, we shall use the self-consistent-field method, i.e., seek the wave function of the ground state in the form of the product

$$\Psi = \prod_{i} \psi_{i}(s_{a,i})$$

where $s_{a,b} = +1, 0, -1$ are the values of the spin projections on the $z$ axis, and $\psi_{i}(s_{a,i})$ is a single-node wave function defined by the condition that the energy be a minimum.

By averaging the Hamiltonian (8) over the wave function (9) we obtain the energy per site:

$$E = \frac{1}{2} \sum_{i} \left( J_{a} s_{a,i}^{\dagger} + J_{b} s_{b,i}^{\dagger} G_{a,b}^{(a,b)} s_{a,b} \right)$$
the angle brackets denote averaging over the function \( \psi_J \), and the subscript \( z \) will hereafter be omitted. The wave function \( \psi^{nm}(m, n = 1, 2) \) is expressed in terms of the components of \( \mathbf{q} \) as follows:

\[
(q_n) = \langle \mathbf{q}_n \rangle, \quad (q_m) = \langle \mathbf{q}_m \rangle, \quad \langle \mathbf{q} \rangle = 0.
\]

(11)

The general phase factor of the wave function \( \psi \) can always be chosen such that the vectors \( \mathbf{R}_n \) and \( \mathbf{Im} \phi \) become mutually perpendicular. We choose next a coordinate frame in which the \( x \)-axis is directed along \( \mathbf{Re} \phi \) and the \( z \)-axis is perpendicular to the plane of \( \mathbf{Re} \phi \) and \( \mathbf{Im} \phi \). Taking the normalization condition into account we obtain

\[
\psi = \psi_0 \psi_0, \quad \psi_0 = (1 - \psi)^{1/2}, \quad \psi_0 = 0.
\]

(12)

where \( 0 \leq \psi_0 \leq 1 \) and \( \lambda = \pm 1 \). Substitution of (12) in (11) and (10) yields

\[
(q_n) = \langle \mathbf{q}_n \rangle, \quad (q_m) = 2 \Delta q_n, \quad (q_0) = 2 \Delta q_0.
\]

(13)

If \( G < J \) the energy is a minimum at \( \psi = 2/\sqrt{2} \). Then \( (s_j) = 1 \), i.e. the ground state corresponds to an ordinary ferromagnet. If, however, \( G > J \), the minimum is reached at two points: \( \psi = 0 \) and \( \psi = 1 \). In both cases we have \( (s_j) = 0 \). This is the so-called quadrupole ordering.\textsuperscript{23} In the considered simplified model the function (11) differs from zero only at \( \psi = 0 \). Therefore \( \psi = \mathbf{R}_n \cdot \mathbf{Q}_n \) or the spin director \( \mathbf{n} \) determined by the formula \( \mathbf{Q}_n = -2n_1 \mathbf{e}_n + (1/3) \mathbf{S}_n \). At \( \psi = 0 \) and \( \psi = 1 \) we have respectively \( n_1 = 1 \) and \( n_1 = 1 \). In the general case at \( G > J \) the ground state of the Hamiltonian (18) is characterized by the equality \( (s_j) = 0 \) and is infinitely degenerate in the direction of the vector \( \mathbf{n} \), with the states \( n_1 = -n_1 = 1 \) identical.

Let the system be in an external magnetic field \( H \) directed along the \( z \)-axis. For states described by Eqs. (12) this is a case of field transverse to \( \mathbf{n} \). An extra term \(-\mu_0 H(\mathbf{n} \cdot \mathbf{q})\) is added to expression (10) for the energy and leads, when (11) and (12) are taken into account to an additional term \(-2\mu_0 H \mathbf{q}(1 - \psi)^{1/2} \) in (13). If \( H > H_c \), where \( H_c = 2 \mathbf{q}(1 - J/\gamma) \), the minimum of the energy corresponds to \( \psi = 0 \) and \( q(1 - \psi)^{1/2} = H/2H_c \). In the presence of a field the energy \( E \) is decreased by \( \gamma H^2/2H_c \). From which it follows that the nematic state is characterized by a field-independent transverse susceptibility \( \chi = \gamma H \), per atom.\textsuperscript{4} At \( H > H_c \) the energy minimum corresponds to \( \psi = 1/\sqrt{2} \) and \( (s_j) = 1 \). The field \( H_c \) is thus the critical magnetic field of the phase transition from the nematic to the usual completely polarized state. This transition is analogous to collapse of antiferromagnetic lattices.

It is also easy to see that in the nematic state, just as in collinear antiferromagnets, the longitudinal susceptibility is zero. Indeed, let \( \psi_0 = 1 \) and \( \psi_0 = 0 \) in the absence of a field; this is equivalent to \( n_1 = 1 \) or \( n_1 = 1 \). We consider closely lying states, putting \( \psi_0 = 1 + \psi_0 \) and \( \psi_0 = \psi_0 \), where \( |\psi_0| < 1 \). In the approximation linear in \( \psi \) the first of formulas (11) yields

\[
E = -|\mathbf{G}_J + (G - J)| \psi^2 (1 - \psi^2).
\]

Thus, the contribution \( -\mu_0 H_c (s_j) \) to the energy from the longitudinal magnetic field does not contain terms linear in the deviations of the state of the system from the initial minimum. In a sufficiently weak field the system remains therefore in exactly the same state as in the absence of a field.

Weak relativistic interactions can be accounted for by introducing in the Hamiltonian the one-ion-anisotropy energy. This leads to the appearance in the energy (10) of a relativistic term \( \mu_0 G \mathbf{S}_n \cdot \mathbf{S} \), and \( \mathbf{S} \) the spin director is determined by the formula \( \mathbf{S} = -2n_1 \mathbf{e}_n + (1/3) \mathbf{S}_n \). In the general case at \( G > J \) the ground state of the Hamiltonian is characterized by the equality \( (s_j) = 0 \) and is infinitely degenerate in the direction of the vector \( \mathbf{n} \), with the states \( n_1 = -n_1 = 1 \) identical.

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nents of the average total spin of the cell, but of each individual spin.

Averaging the Hamiltonian (14) over the function \( |\psi\rangle \), we obtain the energy per unit cell

\[
E = -J_1 \langle \sigma^x \rangle - J_2 \langle \sigma^y \rangle - J_3 \langle \sigma^z \rangle \langle \chi \rangle - G \langle \sigma^z \rangle^2 - G' \langle \sigma^x \sigma^y \rangle^2,
\]

where the averaging is now over the two-spin wave function,

\[
I_{1,2,3} = \sum_{\alpha} \langle \alpha \uparrow \alpha \uparrow | J_{1,2,3} | \alpha \uparrow \alpha \uparrow \rangle,
\]

\[
\langle \sigma^x \rangle = \frac{1}{2} \sum_{\alpha} \langle \alpha \uparrow | \sigma^x | \alpha \uparrow \rangle, \quad \langle \sigma^z \rangle = \frac{1}{2} \sum_{\alpha} \langle \alpha \uparrow | \sigma^z | \alpha \uparrow \rangle
\]

A two-spin-1/2 wave function is equivalent to an asymmetric spinor $^{0}$ of rank 2. Putting

\[
\psi_0 = |\sigma \rangle \chi
\]

we arrive at a representation in which the wave function of an aggregate of a vector $\bar{\mathbf{a}}$ and a scalar $\chi$; the two satisfy the normalization condition

\[
|\psi_0|^2 = 1.
\]

The mean values in (16) are equal to

\[
\langle \sigma^x \rangle_{\psi_0} = \langle \sigma^z \rangle_{\psi_0} = 0, \quad \langle \sigma^y \rangle_{\psi_0} = 0.
\]

We choose the gauge of the wave function and of the coordinate system such as to satisfy the equalities

\[
\psi_0 = u, \quad \psi_0 = iv, \quad \chi = 0
\]

with positive $v$ and real $\chi$. We set also $\phi_0 = \phi + i\eta$.

We assume first that the constants $G, G'$ and $G$ in (16) greatly exceed in absolute value the constants $J_{1,2,3}$. The system energy is then

\[
E = \frac{1}{2} \left[ (u^2 + v^2) - 3u^2 \right] - 8G' (\chi^2 + u^2) - 4G (1 - 4(\chi^2 + u^2)),
\]

If the variable $\chi$ is excluded with the aid of the normalization condition $\chi^2 + 1 = u^2 - v^2 - \eta^2$, the resultant function $E(u', v', \eta)$ should be minimized in the region $0 < u', v', \eta^2 < 1$. The constants $G, G'$ and $G$ should be regarded as positive to justify the choice of the wave function in the form (15) with a two-spin function that is the same for all unit cells. Since the second derivative

\[
\frac{\partial^2 E}{\partial (u')^2} = -16 \left( \frac{G}{3} + 2G' \right)
\]

is negative everywhere, the minimum energy is reached at the boundary of the region considered. Moreover, for that part of the boundary where $u' = 1 - u^2 - v^2$ and $\eta^2 < 1$ we have

\[
\frac{\partial E}{\partial (u')^2} = -16 \left( \frac{G}{3} + 2G' \right) < 0,
\]

so that the minimum can be reached only at those points of the region where at least one of the quantities $u, v, \eta$ is zero. Simple analysis of (19) shows that three types of state can correspond to the energy minimum.

1. If $3G > \max (G, G')$, the energy is a minimum at $u = v = 0$. In this case $\phi_0 = 0$ and $|\psi_0| = 1$, so that we are dealing with a nonmagnetic state in which the spin symmetry $O(3)$ is not broken.

2. At $3G < 2G' < G$ the energy reaches its minimum in two states: $u = 1, v = \eta = 0$ and $u = 1, v = 1$. The meanvalues in (16) are equal to

\[
\langle \sigma^x \rangle = \langle \sigma^z \rangle = \langle \chi \rangle = 0.
\]

The system is a spin nematic of type $n$. The order parameter $q_{\alpha \beta} = 2(\alpha_1 \alpha_1 - \delta_{\alpha_1} / 3)$ corresponds to a director $n$ directed along the $z$ axis in the first state and along the $x$ axis in the second.

We consider now the total energy (16) under conditions (18) near the state $u = 1$ as a function of small $v, \chi$, and $\eta$. Expanding up to quantities of second order of smallness, we get

\[
E = \frac{1}{2} \left[ (u^2 + v^2 - 3u^2) - 8G' (\chi^2 + u^2) - 4G (1 - 4(\chi^2 + u^2)) 
\right] + 8G' (\chi^2 + u^2) + G (1 - 4(\chi^2 + u^2))
\]

The mean value of the projection on the total spin $I/2 = \langle \sigma^z \rangle$ is equal to

\[
\langle \sigma^z \rangle = \langle \chi \rangle = 0.
\]

The total energy can be easily calculated by noting that by virtue of (20) the conditions (18) remain in force also in the presence of a weak field directed along the $z$ axis. With the aid of (20) and (21) we get

\[
\chi = -1/2 (3G - J_1 - J_1).
\]

3. At $3G < \min (G', 3G' - 2G)$ the energy (19) is a minimum for states with $v = 0, u = \cos \theta, \eta = \sin \theta$, where

\[
\cos \theta = -2(3G - 2G') / (3G' - 2G).
\]

The same energy is possessed by states with $u = \eta = 0$. 

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we get

\[ \langle \hat{p}_x \rangle = 0, \]

If the pseudovector \( \mathbf{p} \) is defined by the inequality

\[ \rho_g \sin \theta > 0, \]

we get \( |\mathbf{p}| = 1 \).

\[ \langle \hat{\mathbf{p}} \rangle = -2 \cos \theta \langle \hat{\mathbf{p}} \rangle \]

and at \( \rho_g = 0 \) and \( \rho_u = \rho_v = 0 \) we have respectively

\[ p_x = \text{sign} \sin \theta \rho_u \quad \text{and} \quad p_y = -\text{sign} \sin \theta \rho_v. \]

The system symmetry corresponds thus, to a spin nematic with director \( \mathbf{p} \).

Let us consider states close to the state with \( |\mathbf{p}| = 1 \).

At small \( \rho_u \) and \( \rho_v \) the first three terms of (16) are of second

order of smallness, as follows from (17), therefore the charac-
ter of the equilibrium state does not change at exchange con-
stants \( J_{\rho_u}, J_{\rho_v} \) that are not too large in absolute value.

The magnetic susceptibility that is longitudinal in \( \rho \) is

likewise zero in this case, since \( (\rho_u^2 + \rho_u^2) \) does not contain

terms linear in the deviations of the wave-function compo-
nents from values corresponding to a state without a field.

The z-component of the total spin is now

\[ \rho_z = \rho_u \cos \theta, \]

so that a nonzero \( \rho_u \) appears in a magnetic field directed along the

z axis. Expanding the energy (16) near the equilibrium

value in powers of \( \rho_u \) or \( \rho_v \), we find

\[ E(\rho_u) = E(0) = -h \approx \cos \theta \left( \frac{G}{3} + G' -4G - J_3 \right) \frac{1}{2}(\rho_u + \rho_v)^2 + 3G \].

From (24) and (25) we obtain the following expression for the

transverse susceptibility of a \( \rho \)-nematic:

\[ \chi_{\perp} = \frac{1}{4} \rho_u \cos \theta \left( \frac{G}{3} + G' - 4G - J_3 \right) \frac{1}{2}(\rho_u + \rho_v)^2 + 3G. \]

If \( 3G' - 2G - 3G - J_3 \), it can be seen from (23) that \( \cos \theta \rightarrow 1 \)

and the \( \rho \)-nematic is transformed into an \( \rho \)-nematic. Equa-
tion (26) is then transformed into (22).

The foregoing examples show that magnetic ordering with the symmetry of a spin-nematic should be sought for in

experiments on substances in which, for one reason or an-
other, the many-particle exchange is not small compared with the usual exchange. Since the magnetic properties of

spin nematics are the same as those of antiferromagnets, they can be distinguished only by using neutron scattering or

NMR, methods that can determine the average microscopic spin density.

It is of particular interest to discuss here the possibility of

nematic ordering in an exchange nuclear magnet, solid

\( ^{3}\text{He} \).

At present it is universally accepted (see Refs. 7 and 8)

that the principal mechanism of exchange interaction of nu-

clear spins in solid \( ^{3}\text{He} \) is four-particle cyclic exchange, which exactly contributes to nematic ordering. On the other hand, the conclusion that antiferromagnetic ordering was

observed in solid \( ^{3}\text{He} \) at temperatures below 1 mK was based

either on a study of macroscopic magnetic properties, or on an investigation of the resonant properties of the systematic frequencies much lower than the exchange frequencies. 14 It is clear from our present results these properties are typical of antiferromagnets to the same degree as for spin nematics.

It can therefore not be excluded that what is realized is not a structure of the \( uuud \) type but a nematic structure in which the director \( \mathbf{p} \) behaves under spatial transformation like the antiferromagnetic vector \( \mathbf{l} \) in the \( uuud \) structure. Since we are dealing here with ordering of nuclear spins, the only

method capable of unambiguously answering this question is

investigation of neutron scattering.

We note in conclusion that a specific example of nema-

tic spin ordering is the spin system of superfluid \( ^{3}\text{He}-f \). The

pair spin correlator \( (\rho_u \rho_v) \) in \( ^{3}\text{He}-f \) is anisotropic and has the

structure discussed above and determined by the spin direc-
tor. The distinguishing feature is that \( \rho_u \rho_v \) is not invariant to time reversal. The equality \( (\rho_u \rho_v) = 0 \) holds in it by virtue of the homogeneity and of the absence of ferromagnetism.

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