Possibility of Observation of Nontrivial Magnetic Order by Elastic Neutron Scattering in Magnetic Field

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We consider the neutron scattering in the magnetic phases with zero average sublattice magnetization (double- or triple-spin correlator as an order parameter). External magnetic field induces different magnetizations on the different sublattices in such a system. As a result, the new purely magnetic Bragg peaks for the elastic neutron scattering appear. We present the estimates of intensities in the Bragg peaks and their angular dependence on the external magnetic field. This could serve as a test whether the nontrivial magnetic order exists in some heavy fermion compounds, like URu_2Si_2 or UPt_3 and possibly in the high-T_c oxides.

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The vast majority of magnetic systems order antiferromagnetically below some transition temperature, T_N. However, less common ground states may also exist. Theoretically this is expected for the Heisenberg Hamiltonian with frustrated interactions (see, e.g., [1,2]). Experimentally so-called quadrupole order was observed long ago in UPd_3 and CeB_6 [3]. The problem of the non-Néel magnetic states has recently attracted much theoretical attention in connection with the peculiar magnetic properties of both the high-T_c oxides in the normal phase and some of the heavy fermion compounds, like UPt_3 and URu_2Si_2. Both classes display the strong correlated system features together with the itinerancy of the electronic system.

As for the high-T_c oxides, the insulating 3D Néel state seen for the stoichiometric La_2CuO_4 is easily destroyed by a very small amount of dopants (Sr, Ca). On the other hand, the in-plane spin-spin exchange interaction is so strong ($J \sim 0.15$ eV) that it seems to be natural for the system to preserve some long-range magnetic order even if the dopants are present, at least at zero temperature. Unfortunately, the method proposed below probably will not answer this question.

Another challenging example is given by the heavy fermion compounds, UPt_3 and URu_2Si_2, where an antiferromagnetic order is seen below 5 K and 17.5 K, correspondingly. The puzzle is that the staggered magnetization turns out to be extremely small, $\langle m_Z \rangle \sim 10^{-2}\mu_B$ [4], while a large specific heat anomaly is seen at 17.5 K for URu_2Si_2. The transition, as proved by unpolarized neutron scattering, seems to be of a magnetic origin [4], but the smallness of the sublattice magnetization is hardly compatible with the observed entropy loss. It was suggested in [5] that the actual driving order parameter could be the triple-spin correlator while the small moments appear as a perturbation due to a weak antiferromagnetism mechanism. For what follows, it is essential to keep in mind that the magnetic transitions mentioned above double the lattice unit (i.e., there are two sublattices). Some other substances, like organic conductors, may also be suspicious for the unusual magnetic order. To conclude, there are reasons to expect the non-Néel type of ordering in a number of substances.

It has also been known for a long time [6,7] that most of the bulk properties of such non-Néel phases would be indistinguishable from those of an ordinary antiferromagnet: such phenomena as the anomaly in the specific heat at some T_N, the kinks in the temperature dependence of the magnetic susceptibility, and the spin-flop (and spin-flip) transitions in the applied magnetic field would be seen at any magnetic ordering. Quadrupole-type ordering may be seen by neutrons via magnetoelastic coupling, but distortions will be small. All this makes the issue of experimental identification of the new type of ordering a difficult problem.

The neutron scattering remains one of the most powerful methods of identification of the magnetic order. Elastic neutron experiments measure directly the average sublattice magnetizations, $\langle S^i \rangle$, i.e., the average local spins in magnets. This method, however, fails for an unconventional magnetic order, where $\langle S^i \rangle$ identically vanishes (like in the spin nematics) or is small (the triple-spin correlators), so that the leading order parameter is different. To obtain the dependence of the neutron scattering cross section on spin S in a higher power, one needs to go beyond the Born approximation in the scattering amplitude. These corrections, while seen experimentally for the critical scattering in ordinary ferromagnets [8], are very small. Therefore, we discuss below another way to identify the nontrivial magnetic order—the neutron *elastic* scattering measurements in an *applied external magnetic field.*

It is well known that the magnetic part of the Born scattering amplitude has the form

$$f_{\mathbf{q}} = -r_0\gamma \sum_j F(\mathbf{q}) e^{i\mathbf{q}\cdot\mathbf{R}_j} \left(\delta_{\alpha\beta} - \frac{q_\alpha q_\beta}{q^2}\right) S_j^\alpha \sigma^\beta , \qquad (1)$$

where $\mathbf{q} = \mathbf{p} - \mathbf{p}'$, p and p' are the initial and the final

momenta of the neutron, r_0 is the classical electron radius, γ is the gyromagnetic ratio of the neutron, $\sigma/2$ is its spin, $\mathbf{S}_j$ is the spin of the atom at the point $\mathbf{R}_j$, and $F(\mathbf{q})$ is the form factor of the atom.

Thus, the Born cross section for the elastic Bragg scattering is determined by $\langle S^j \rangle$, the average local magnetization. The latter, however, depends explicitly on the double- and triple-spin correlators (we assume below $\langle S^j \rangle_0 = 0$):

$$\langle S_l^k \rangle = (g\mu_B) H^i T_l^{ik} + \frac{(g\mu_B)^2}{2} H^i H^p T_l^{ipk}, \tag{2}$$

where in the Matsubara representation

$$T_l^{ik} = \sum_a \int_0^\beta d\tau \langle T\{S_a^i(\tau) S_l^k(0)\}\rangle ,$$
$$T_l^{ipk} = \sum_{ab} \int_0^\beta \int_0^\beta d\tau_1 d\tau_2 \langle T\{S_a^i(\tau_1) S_b^p(\tau_2) S_l^k(0)\}\rangle, \tag{3}$$

(l, a, b) are the lattice indices and (i, p, k) are the spin indices. $\hat{T}_l$ can be different for the different sublattices. We shall see that Eq. (2) provides a sizable effect already in moderate magnetic fields. Unfortunately, much stronger magnetic fields are needed to check the magnetic symmetry in the high-T_c oxides.

Nontrivial spin arrangements are known to exist in the microscopic localized spin models with $S \geq 1$ [9] (quadrupole spin arrangements). There were also numerous attempts to describe the exotic magnetic ground states microscopically in terms of the Hubbard or t-J model using the mean field approach or $1/N$ expansion [10, 11]. However, a phenomenological approach may be taken instead, which would be applicable for any system where the Néel state is unfavorable. We shall now summarize this approach in brief.

Independently of specific approximations any of these new ground states should manifest a breaking of the spin symmetry. Any breaking of the rotational spin group (in the exchange approximation), in its turn, can be classified near T_N by introducing average multiple-spin correlators in the form [7, 12]

$$\langle S^i(\mathbf{r}_1) S^k(\mathbf{r}_2) \cdots S^l(\mathbf{r}_n)\rangle = \sum_\beta T_{\alpha\beta}^{ik\ldots l} \Phi^{\alpha,\beta}(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_n), \tag{4}$$

where α is a representation of the spatial lattice group in the paramagnetic phase, so that $\Phi^{\alpha,\beta}(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_n)$ transforms according to this representation, while the n-rank spin tensors, $T_{\alpha\beta}^{ik\ldots l}$, describe the spontaneous breaking of the spin rotational symmetry. For $n=2$ this is an obvious generalization of the local (on site) quadrupole averages and is applicable for any spin value. If n is even, the time-reversal symmetry remains preserved, while at any odd n the time-reversal symmetry is broken. The symmetry in the spin space is lowered and varies depending on the specific form for the tensor $\hat{T}$ in Eq. (4) (examples are given below). The spin tensors $\hat{T}$ in Eq. (4) show up in the time-dependent correlators in Eqs. (2) and (3). To be more specific, we discuss below only the pair and triple-spin correlators

$$K_{ab}^{ik}(\tau) = \langle T[S_a^i(\tau) S_b^k(0) - \tfrac{1}{3}\delta^{ik} S_a^j(\tau) S_b^j(0)]\rangle ,$$
$$K_{abc}^{ipk}(\tau_1, \tau_2) = \langle T\left[S_a^i(\tau_1) S_b^p(\tau_2) S_c^k(0)\right]\rangle. \tag{5}$$

According to Eq. (4), below the transition point, T_N, one can write

$$K_{ab}^{ik}(\tau) = \sum_\beta T_\beta^{ik} \Phi_{\mathbf{Q}}^\beta(\tau; \mathbf{r}_a, \mathbf{r}_a - \mathbf{r}_b) ,$$
$$K_{abc}^{ipk}(\tau_1, \tau_2) = \sum_\beta T_\beta^{ipk} \Phi_{\mathbf{Q}}^\beta(\tau_1, \tau_2; \mathbf{r}_a, \mathbf{r}_a - \mathbf{r}_b, \mathbf{r}_a - \mathbf{r}_c). \tag{6}$$

As mentioned above, $\Phi_{\mathbf{Q}}^\beta$ are responsible for the space symmetry of the order parameter. In most cases of interest the magnetic order involves two sublattices, so that $\Phi^\beta(\tau_{1\ldots n-1}; \mathbf{r}_{1\ldots n})$ in Eq. (6) have to be of the form

$$\exp[i\mathbf{Q}\cdot(\mathbf{r}_1 + \mathbf{r}_2 + \cdots + \mathbf{r}_n)/n] \Phi_{\mathbf{Q}}^\beta(\tau_{1\ldots n-1}; \mathbf{r}_{1\ldots n}), \tag{7}$$

where $\mathbf{Q}$ is the sublattice wave vector [for instance, $\mathbf{Q} = (\pi, \pi)$ for the CuO_2 planes in the high-T_c oxides] and $\Phi_{\mathbf{Q}}^\beta(\tau_{1\ldots n-1}; \mathbf{r}_{1\ldots n})$ realize an irreducible representation of the small group of vector $\mathbf{Q}$. Of course, the explicit form for functions $\Phi_{\mathbf{Q}}^\beta$ (i.e., their space and time dependence) in (4) or (7) is not known except in the model calculations. Nevertheless, the basic conclusion from Eqs. (2) and (3) is that the two sublattices acquire different magnetic moments in the applied magnetic field seen as the Bragg peaks by neutrons. According to Eqs. (2) and (3) one has

$$T_l^{ik} = \sum_\beta T_\beta^{ik} \Phi_{\mathbf{Q}}^\beta(\omega = 0; \mathbf{r}_l, \mathbf{q} = 0) ,$$
$$T_l^{ipk} = \sum_\beta T_\beta^{ipk} \Phi_{\mathbf{Q}}^\beta(\omega_1, \omega_2 = 0; \mathbf{r}_l, \mathbf{q}_1, \mathbf{q}_2 = 0), \tag{8}$$

where we have introduced the Fourier components for the basis functions of Eq. (6). Here β enumerates the basis functions of the irreducible representation with the vector $\mathbf{Q} = (\pi, \pi)$. As usual, the orientation of the spin tensor is fixed by some magnetic anisotropy.

(*i*) *The nonpolarized elastic neutron cross section. The new peaks.*—The purely magnetic contribution into elastic neutron cross section can be expressed in the following way [13]:

$$\frac{d\sigma}{d\Omega} = r_0^2 N \frac{(2\pi)^3}{v_0} \sum_{\mathbf{q}} \delta(\mathbf{k} - \mathbf{q}) \exp\{-2W(\mathbf{q})\} |F_M(\mathbf{q})|^2 \times \{1 - (\hat{\mathbf{q}}\cdot\hat{\boldsymbol{\eta}})^2\}_{\text{av}} . \tag{9}$$

Here $F_M(\mathbf{q})$ is the magnetic unit-cell structure factor defined by

$$F_M(\mathbf{q}) = \sum_d \frac{1}{2} g_d \langle S_d \rangle_H F_d(\mathbf{q}) \lambda_d e^{i\mathbf{q}\cdot\mathbf{r}_d}. \quad (10)$$

λ_d is either $+1$ or -1 according to whether the spin at cite d is on the "plus" or "minus" sublattice, $\hat{\mathbf{q}}$ and $\hat{\boldsymbol{\eta}}$ are the unit vectors following the direction of $\mathbf{q}$ and $\mathbf{L}$, where $L = \langle S_1 \rangle_H - \langle S_2 \rangle_H$ and $\langle S_1 \rangle_H$, $\langle S_2 \rangle_H$ are the field-induced spin averages on the two sublattices. The average is taken over the orientations of $\hat{\boldsymbol{\eta}}$, $\exp\{-2W(\mathbf{q})\}$ stands for the Debye-Waller factor.

One sees that the cross section in the new purely magnetic Bragg peaks is determined by the square of the difference of the sublattice magnetizations, L^2. Therefore, by comparing Eqs. (4) and (8), one would expect that the intensities of the Bragg peaks are $(2\mu_B H/T_N)^2$ times weaker for the spin nematics than those in the antiferromagnets and $(2\mu_B H/T_N)^4$ times weaker for the triple-spin correlator systems. So, the effect should be seen on the scale of the magnetic fields of the order of

$$H_0 = \frac{T_N}{2\mu_B}. \quad (11)$$

While for $(LaSr)_2CuO_4$, this estimate would give us $\simeq$ 200 T, which is unrealistic as we mentioned above, this is only $\simeq$ 10 T for URu_2Si_2. The intensities of the new Bragg peaks are proportional to $(H/H_0)^2$ for the spin nematics and $(H/H_0)^4$ for the triple-spin correlator systems.

(*ii*) *The symmetry of the spin correlators.*—According to Eqs. (3) and (8), only the totally symmetric tensors T^{ik} and T^{ipk} for the double- and triple-spin correlators could be seen by the elastic neutron scattering. Therefore, the so-called "p-nematic" $T^{ik} = \epsilon^{ikm} P_m$ [7] and the chiral spin state $T^{ipk} = \epsilon^{ipk}$ [11,12], if present in the spin system, must be detected by some other means [2].

For the symmetric two-spin correlators (spin nematics or quadrupole order) the classification of all possible symmetry groups has been done in [14] and it was shown that quite a rich variety of different possibilities exist even for a simple tetragonal structure. Some triple-spin correlator symmetries were considered in [5].

To demonstrate the versatility of the neutron approach, as described above, we consider only a few examples. (a) For the n-type spin nematic [7], where $T^{ik} = (n_i n_k - \frac{1}{3}\delta^{ik})$ (the uniaxial symmetry, $\mathbf{n}$—the unit vector) the spin polarization, $\mathbf{L}$, is proportional to

$$L^i \propto n^i(\mathbf{n}\cdot\mathbf{H}) - \tfrac{1}{3} H^i. \quad (12)$$

(b) For the so-called (u,v) nematic [14],

$$L^i = u^i(\mathbf{v}\cdot\mathbf{H}) + v^i(\mathbf{u}\cdot\mathbf{H}), \quad (13)$$

where $\mathbf{u} \perp \mathbf{v}$ are the two mutually perpendicular unit vectors. (c) The triple-spin correlator order parameter can be generally expressed through three independent vectors in the following way [5]:

$$T^{ipk} = \sum_{\mathbf{P}} [a^i b^p c^k - \tfrac{2}{5}(\mathbf{b}\cdot\mathbf{c})(\delta^{ip}a^k + \delta^{ik}a^p + \delta^{pk}a^i)]. \quad (14)$$

Here the summation is taken over all possible permutations of $(\mathbf{a},\mathbf{b},\mathbf{c})$ at fixed indices (ipk). Again, the applied external magnetic field induces magnetizations on different sublattices, so that their difference has now the following form:

$$L^i = T^{ipk} H_p H_k = \sum_{\mathbf{P}} [a^i \{(\mathbf{b}\cdot\mathbf{H})(\mathbf{c}\cdot\mathbf{H}) - \tfrac{2}{5}(\mathbf{b}\cdot\mathbf{c})H^2\} - \tfrac{4}{5}(\mathbf{b}\cdot\mathbf{c})(\mathbf{a}\cdot\mathbf{H})H^i]. \quad (15)$$

Consider for example the two triple correlator phases studied in [5]. The first phase is uniaxial with $\mathbf{a} = \mathbf{b} = \mathbf{c} = \mathbf{n}$ in Eq. (14). In the second phase $\mathbf{a} \perp \mathbf{b} \perp \mathbf{c}$. For these two phases one has, correspondingly,

$$L^{(1)} = n^i[(\mathbf{H}\cdot\mathbf{n})^2 - \tfrac{1}{5}H^2] - \tfrac{2}{5}(\mathbf{H}\cdot\mathbf{n})H^i,$$
$$L^{(2)} = a^i(\mathbf{b}\cdot\mathbf{H})(\mathbf{c}\cdot\mathbf{H}) + b^i(\mathbf{a}\cdot\mathbf{H})(\mathbf{c}\cdot\mathbf{H}) + c^i(\mathbf{a}\cdot\mathbf{H})(\mathbf{b}\cdot\mathbf{H}). \quad (16)$$

All the above phases are, therefore, experimentally distinguishable by their different angle dependence on the orientation of the applied magnetic field. It was, of course, assumed that the directions of $\mathbf{n}$ in Eq. (12), $(\mathbf{u},\mathbf{v})$ in Eq. (13), or $(\mathbf{a},\mathbf{b},\mathbf{c})$ in Eq. (15) are somehow fixed with respect to the lattice axes by the magnetic anisotropy, so that the magnetic field is below its "spin-flop" value.

(*iii*) *The scattering of the polarized neutron beam.*—Similar information about the symmetry of the spin tensor may be obtained from the polarization measurements on the scattered beam. The polarization of the scattered beam is given by the formula

$$P_\mu^{\text{Bragg}} = \frac{2L_\perp^\mu(\mathbf{P}_0\cdot\mathbf{L}_\perp) - P_0^\mu L_\perp^2}{L_\perp^2}, \quad (17)$$

where $\mathbf{P}_0$ is the polarization of the oncoming neutron beam. Now $\mathbf{L}_\perp$ is the projection of the difference of the sublattice magnetizations into the plane perpendicular to the scattering vector. The dependence of $\mathbf{L}$ on the direction of the magnetic field is different for the n-type uniaxial spin nematic and the $u\perp v$ symmetric spin nematic, as we have just seen, and this allows us to distinguish between these two types of symmetric spin nematics. The same is true for both symmetric triple-spin correlator phases.

(*iv*) *The nuclear-magnetic Bragg peaks intensities.*—To determine $\mathbf{L}$, one might find it possible to measure the nuclear-magnetic neutron peaks by polarized neutrons. The intensities of those will be given by the same formulas as for conventional magnets, but are now only

(H/H_0) times weaker for the spin nematics and $(H/H_0)^2$ times weaker for the triple spin correlator order parameter.

(*v*) *The inelastic scattering.*—The inelastic neutron measurements on spin nematics should see the low lying spin wave spectrum that is not necessarily symmetric near the Γ and X points [7]. Of course, the spin wave spectrum should also be seen for the triple-spin correlator phases.

The anomalous ordering can exist in many substances. On the other hand, if the phenomena like an onset of the anisotropy in *the temperature dependence* of the bulk susceptibility or a spin-flip transition are observed in a material, but *no spontaneous magnetization* is seen by neutrons, this would again indicate that below this onset temperature a novel non-Néel state develops. This could be the case, for instance, in URu_2Si_2. In principle, in addition to the case of URu_2Si_2 or UPt_3, this comment could be important in the high-T_c oxides, where a magnetic transition may appear at $T_N \sim 200$ K, but the method proposed would be not applicable for the realistic values of the magnetic field.

In conclusion, if T_N is low enough, the method provides a simple way to determine whether the given structure is an unconventional magnet or not: in a large enough applied magnetic field the new Bragg peaks appear due to the sublattice magnetization, which is proportional to H/H_0 for spin nematics and to $(H/H_0)^2$ for the triple-spin correlators. Only symmetric (see above) tensor phases can be seen in this approach. However, at least in principle, since the overall symmetry of the spin scalars may change in higher orders (see [14]), this can be noticed by the study of the magnetic scattering anisotropy with respect to the applied magnetic field for the Bragg peaks with $\mathbf{Q} = 0$. It is also worth mentioning that there could exist magnetic systems where, in general, similar magnetic states could develop for the wave vector $\mathbf{Q} = 0$. Most of the above considerations would be applicable as concerns the anisotropy of the magnetic peak intensity on the direction of applied magnetic field for this case as well. The estimates obtained are quite general, because they are based upon a phenomenological analysis of the order parameters.

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