Investigation of Spin Chirality by Polarized Neutrons

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The projection of the chiral spin fluctuations on the magnetization is discussed. It is shown that it determines the polarization dependent part of the neutron magnetic scattering. A strong enhancement of this scattering appears near the chiral phase transition in triangular lattice antiferromagnets as well as in all cases when the system is soft and its nonlinearity is important. The example of conventional antiferromagnets is considered. A possibility to observe chiral fluctuations in doped CuO_2 planes as a precursor of a proposed chiral state is discussed.

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During the last decade, spin chirality has attracted much attention (see, for example, [1–3] and references therein). Frustrated antiferromagnets on a triangular lattice (TLA's) were the main subject of the discussion. Kawamura suggested [3] that the phase transition in the stacked triangular lattice belongs to a new chiral universality class. This statement was questioned on the basis of a $2+\epsilon$ expansion [4] and three-loop renormalization group calculations [5]. However, recent Monte Carlo simulations [1] and numerous experimental data [6–9] apparently confirm the Kawamura conjecture. Nevertheless, up to now we have no definitive solution of the problem. The doped CuO_2 layers in the high-T_c superconductors were considered as another candidate for chiral spin order with violation of P and T symmetries [10]. In spite of the fact that this suggestion has not been confirmed experimentally [11], the persistence of the chiral fluctuations cannot be ruled out.

For the complete experimental investigation of the spin chirality problem, one should study fluctuations of the chiral variable. However, it is a combination of spin pairs belonging to different lattice sites [1–3,12]. Corresponding fluctuations are related to four-spin correlations, and their direct experimental study is impossible. In this paper we discuss the possibility of studying, using polarized neutrons, a projection of the chiral field on the sample magnetization induced by the applied field.

We will show that this projection determines the part of the cross section which is proportional to the neutron polarization $\vec{P}_0$. It is a pure inelastic part which disappears at $\omega = 0$, and below we will call it the dynamical chirality (DC). In general, the $\vec{P}_0$ dependent part of the cross section consists of two terms: the DC contribution and the interference between nuclear and magnetic scattering. Both terms are proportional to the field induced magnetization. In crystals the latter appears near the nuclear Bragg reflections only, where it is related to weak processes such as magnetovibrational scattering. At the same time, the DC scattering persists near magnetic reflections where, as we will see below, it is strongly enhanced. Therefore in what follows we may neglect the interference scattering completely. As a result, the $\vec{P}_0$ dependent part of the cross section is determined by the DC only, and it may be comparatively easy to separate it from other scattering processes.

The $\vec{P}_0$ dependent part of the critical scattering in ferromagnets has been discussed in [13], where it was ascribed to three-spin dynamical correlations. In ferromagnets the chirality is not a relevant variable. However, very strong nonlinearity of the system near T_c gives rise to significant DC which has been observed in [14]. Recently, this method has been used for investigation of the spin-wave spectra in disordered ferromagnets [15].

In this paper we will study the general properties of DC and discuss the possibilities of its observation in several magnetic systems. First of all, we consider the TLA. In this case, as well as in the Kagomé antiferromagnet [16], the chirality is a relevant variable, and the DC should provide direct information on the nature of the phase transition. We then consider ordinary antiferromagnets, where the DC persists due to the nonlinearity of the system. We discuss also the possibility of studying the DC in doped CuO_2 layers in the high-T_c systems in relation to the supposed chiral order [10].

In the TLA the chirality of the elementary triangle is determined as [3]

$$\vec{K}_{123} = \vec{S}_1 \times \vec{S}_2 + \vec{S}_2 \times \vec{S}_3 + \vec{S}_3 \times \vec{S}_1\,, \tag{1}$$

where $\vec{S}_i$ is a spin in a vertex i. The corresponding staggered chirality introduced in [12] has the form

$$K_{\vec{Q}} = i\vec{S}_{\vec{Q}} \times \vec{S}_{-\vec{Q}} = N^{-1} \sum_{\vec{R}_1,\vec{R}_2} \vec{S}_{\vec{R}_1} \times \vec{S}_{\vec{R}_2} \sin\vec{Q}\vec{R}_{21}\,. \tag{2}$$

In the case of 120° structure of the TLA, the chiral order is determined by [17]

$$\vec{S}_R = \vec{\ell}\cos\vec{Q}_0\vec{R} + \vec{m}\sin\vec{Q}_0\vec{R}\,, \tag{3}$$

where $\vec{\ell}^{\,2} = \vec{m}^2 = S^2$, $\vec{\ell}\vec{m} = 0$, and $\vec{Q}_0 = 2\pi \times (2/3, 0, 1/2)$. As a result, in the elastic scattering cross

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section a term proportional to $\vec{K}^{\perp}_{\hat{Q}}\vec{P}_0$ appears [18], where

$$\vec{K}^{\perp}_{\hat{Q}} = \frac{1}{2}\langle[\vec{\ell}_{\perp} \times \vec{m}_{\perp}]\rangle \times \sum_{\vec{\tau}}[\Delta(\vec{Q} - \vec{Q}_0 + \vec{\tau}) - \Delta(\vec{Q} + \vec{Q}_0 + \vec{\tau})]. \quad (4)$$

Here $\vec{\tau}$ is the reciprocal lattice vector $\Delta(0) = N$; $\Delta(\vec{Q} \neq 0) = 0$ and $\vec{x}_{\perp} = \vec{x} - (\vec{x}\hat{Q})\hat{Q}$. This elastic scattering may be observed in the case of a one-domain sample, where the chiral degeneracy is removed. It may be done by cooling the sample in an electric field [19].

We are interested in the DC contribution to the neutron scattering. It is determined by the antisymmetric part of the spin Green's function

$$G_{\alpha\beta}(\vec{Q},\omega) = i\int_0^{\infty} dt e^{i\omega t}\langle[S^{\alpha}_{\vec{Q}}(t), S^{\beta}_{-\vec{Q}}(0)]\rangle. \quad (5)$$

According to the general rule [20], in the magnetic field we have $G_{\alpha\beta}(\vec{H}) = G_{\beta\alpha}(-\vec{H})$ and the symmetric $G^{(S)}$, and antisymmetric $G^{(A)}$ parts of G are even and odd functions of $\vec{H}$, respectively. It is convenient below to introduce the function $S_{\alpha\beta} = iG^{(A)}_{\alpha\beta}$. Using the conventional intermediate state decomposition of the Green's function [20], one can show that $\mathrm{Im}G^{(S)}$ and $\mathrm{Re}S$ are odd and $\mathrm{Re}G^{(S)}$ and $\mathrm{Im}S$ even functions of ω, respectively. At the same time, the neutron cross section is determined by $\mathrm{Im}G^{(S)}$ and $\mathrm{Im}S$.

Multiplying (5) by the unit pseudotensor $\epsilon_{\alpha\beta\gamma}$, we extract the DC contribution

$$\vec{K}_{\vec{Q}}(\omega) = i\int_0^{\infty} dt e^{i\omega t}\langle\vec{S}_{\vec{Q}}(t) \times \vec{S}_{-\vec{Q}}(0) - \vec{S}_{-\vec{Q}}(0) \times \vec{S}_{\vec{Q}}(t)\rangle. \quad (6)$$

The dependence of $\vec{K}_{\vec{Q}}$ on ω is a direct consequence of the nonlocality of the chiral operator. Indeed, in the definition (2) we may consider spins $\vec{S}_{R_1}$ and $\vec{S}_{R_2}$ at different times on the same footing as at the different lattice points.

In the absence of long-range chiral order we have $\vec{K}_{\vec{Q}}(0) = 0$, and at the same time $\vec{K}_{\vec{Q}}(\omega)$ is an odd function of $\vec{H}$. We may consider $\vec{K}_{\vec{Q}}(\omega)$ as a dynamical projection of the chiral operator $\vec{K}_{\vec{Q}}$ onto the magnetization induced by the magnetic field.

Another approach to the problem is the following. In a low magnetic field the Matsubara Green's functions acquire an additional term

$$\delta G_{\alpha\beta}(i\omega_n) = -g\mu H_{\gamma}\sqrt{N}\int_0^{1/T} d\tau\, d\tau_1 e^{i\omega_n\tau} \times \langle TS^{\alpha}_{\vec{Q}}(\tau)S^{\beta}_{-\vec{Q}}(0)S^{\gamma}_0(\vec{\tau}_1)\rangle, \quad (7)$$

where $g\mu > 0$. The function $G^{(A)}_{\alpha\beta}(\omega)$ is an analytical continuation of this expression to the real axis. We see that in low field the DC is determined by the three-spin correlation function introduced in [13,21] for the case of critical fluctuations in ferromagnets.

The antisymmetric tensor $S_{\alpha\beta}(\hat{Q},\omega)$ is determined by an axial vector $\vec{A}$: $S_{\alpha\beta} = \epsilon_{\alpha\beta\rho}A_{\rho}$. We are interested in uniaxial systems with the $\hat{c}$ axis perpendicular to the layers. In this case the general expression for $\vec{A}$ has the form

$$\vec{A}(\vec{Q},\omega) = \hat{h}S_1(\vec{Q},\omega) + (\hat{c}\hat{h})\hat{c}S_2(\vec{Q},\omega), \quad (8)$$

where $S_{1,2}$ are scalar functions. As a result we obtain from Eqs. (6) and (8) $K = -2i[\hat{h}S_1 + (\hat{c}\hat{h})\hat{c}S_2]$. For the isotropic Heisenberg and XY models we have $S_2 = 0$ and $S_1 = 0$, respectively.

Using (8) for the chiral part of the cross section we have

$$\left(\frac{d\sigma}{d\Omega\, d\omega}\right)_{P_0} = \frac{2}{\pi}P_0(r_0\gamma)^2|F(\vec{Q})|^2\frac{k_f}{k_i}\frac{1}{1-\exp(-\omega/T)} \times \{(\hat{Q}\hat{h})^2\mathrm{Im}S_1(\vec{Q},\omega) + (\hat{h}\hat{Q})(\hat{Q}\hat{c})(\hat{c}\hat{h})\,\mathrm{Im}S_2(\vec{Q},\omega)\}. \quad (9)$$

Here we have taken into account that in real experiments, $\vec{P}_0$ is directed along or opposite to the magnetic field.

In (9) $\mathrm{Im}S_{1,2}$ are even functions of ω, and if the characteristic energy is less than T the chiral cross section is an odd function of ω and the chiral contribution to the quasielastic scattering disappears. It is a consequence of the orthogonality of the chiral field and magnetization. At the same time, it was shown [13–15] that due to the ω dependence of $\hat{Q}$ the DC contribution to the small-angle quasielastic scattering persists if the angle between the beam and the field is not equal to 0° or 90°.

In a low magnetic field we have the following very crude estimate:

$$\mathrm{Im}S_{1,2}(\vec{Q},\omega) = \left(\frac{g\mu H}{T^2_{\mathrm{int}}}\right)g_{1,2}(\vec{Q},\omega), \quad (10)$$

where $g_{1,2}$ is a dimensionless function and T_{int} is the characteristic energy of the spin-spin interaction. We will see below that this estimation does not take into account any enhancement which appears if the system becomes soft in some region of the $\vec{Q}$ space.

The DC in the TLA.—For definiteness we will base our analysis on the Kawamura suggestion about the new class of chiral universality [3,17] and use the conventional analysis of the scaling dimensionality [22]. According to the definition (2) in $\vec{r}$ space, the chirality $\vec{K}$ is given by the product $\vec{S}_{\vec{R}_1} \times \vec{S}_{\vec{R}_2}$ and depends on two variables $\vec{R} = (\vec{R}_1 + \vec{R}_2)/2$ and $\vec{\rho} = \vec{R}_1 - \vec{R}_2$. The chiral critical fluctuations are related to the $\vec{R}$ dependence of $\vec{K}$, and $\vec{\rho}$ may be considered as an intrinsic variable of the chiral field. The same holds for the t dependence of $\vec{K}$. The scaling dimensionality is determined as $\vec{K}(\vec{R}s) = \vec{K}(\vec{R})s^{-\Delta_K}$, where Δ_K is related to the corresponding susceptibility and correlation length exponents in the usual way [22], $\Delta_K = (3 - \gamma_K/\nu)/2$.

Below T_N we have $\langle\vec{K}\rangle \sim \xi^{-\Delta_K} \sim (-\tau)^{\beta_K}$, where ξ is the correlation length $\tau = (T - T_N)/T_N$ and $\beta_K = \nu\Delta_K$. However, we are interested in the dynamical chirality

$K(\tilde{p}, t)$ above T_N. According to Eqs. (2) and (7), for small $\vec{H}$ the DC is a correlation function $\langle K_Q \vec{S}_0 \rangle \vec{H}$, where $\vec{S}_0$ is the uniform spin density. We may now use the general expression for the correlation function of two fields A and B: $G_{AB} = \xi^{d-\Delta_A-\Delta_B}$, where d is the spatial dimensionality and $\Delta_{A,B}$ are corresponding scaling dimensionalities [22]. In our case $\vec{S}_0$ is a noncritical variable and $\Delta_{\vec{S}_0} = 0$.

As a result, we have

$$S_{1,2} \sim \xi^{3-\Delta_K} = \tau^{-\varphi_K}, \qquad (11)$$

where $\varphi_K = \nu(3 - \Delta_K) = \beta_K + \gamma_K$ is the chiral crossover exponent [3].

From (10) and (11) we get

$$\mathrm{Im} S_{1,2}(\vec{Q}, \omega) = \left(\frac{g\mu H}{T_N^2 \tau^{\varphi_K}}\right) \Phi_{1,2}(\vec{Q}, \omega), \qquad (12)$$

where τ^{φ_K} gives the above-mentioned enhancement of the chiral scattering. For the Heisenberg and XY models we have $\Phi_2 = 0$ and $\Phi_1 = 0$, respectively. The exponents γ_{ch} and ν were calculated in [3] for both models, and we get $\varphi_H = 1.26(9)$ and $\varphi_{XY} = 1.20(6)$.

Obviously, the functions $\Phi_{1,2}$ have maxima at reciprocal lattice points corresponding to a 120° spin structure, and below we will consider $\Phi_{1,2}$ as functions of $\vec{q} = \vec{Q}_0 - \vec{\tau}$. Chiral scaling cannot be applied to a parametrization of $\Phi_{1,2}$, as $\vec{q}$ and ω are intrinsic variables of the chiral field. They correspond to the correlation of $\vec{S}_1$ and $\vec{S}_2$ and should be governed by the conventional TLA scaling for the two-spin correlation function:

$$\Phi_{1,2}(\vec{q}, \omega) = \tau^{-\gamma} F\left(\vec{q}\xi, \frac{\omega}{T_N \xi^{-z}}\right), \qquad (13)$$

where γ is the staggered susceptibility exponent, which was calculated in [3] for both cases also. As a result, we have an additional enhancement and $S_{1,2} \sim \tau^{-(\varphi_K+\gamma)}$, where $\varphi_K + \gamma = 2.43(16)$ and $2.33(11)$ for the H and YX models, respectively. Comparing (13) and (12) and taking into account that (13) has to be less than or of the same order as the cross section at $H = 0$, we conclude that the low-field approximation used above is applicable if the condition $g\mu H < T_N \tau^{\varphi_K}$ is fulfilled. In other cases the positiveness of the cross section would be violated.

The dynamical scaling in the TLA has been confirmed experimentally for $CeMnBr_3$, which is an XY magnet [23]. It was found that $z \simeq 1.5$. However, there is no theoretical estimation of z for XY systems.

The above consideration has been based on the Kawamura suggestion [3]. In this case one can determine the chiral exponent Δ_K by combining the conventional and chiral scattering data. Another way to determine Δ_K is the elastic scattering below T_N when the cross section has a term proportional to $\vec{K}_\perp P_0$, where $\vec{K}_\perp$ is given by (4). Near T_N we have $\vec{K}_\perp \sim (-\tau)^{\beta_K}$. However, for this study one has to prepare a one-domain sample using the method of Ref. [19].

If the Kawamura conjecture is wrong it may be checked by the chiral scattering. For example, if according to [1] there are two successive transitions to the chiral and Néel states with $T_K > T_N$, we get, instead of (12) and (13), $S_{1,2} \sim \tau_K^{-\varphi_K} \tau_N^{-\gamma} F(q\xi_N, \omega/T_N\xi_N^z)$, where $\tau_{K(N)} = (T - T_{K(N)})/T_{K(N)}$ and ξ_N is the AF correlation length. In the same way one can analyze other possibilities [4,5].

It is well known that the TLA's are quasi-2D or quasi-1D systems (see, for example, [7–9] and references therein). In both cases with increasing τ above T_N, crossover occurs to low-dimensional behavior. In the quasi-2D region the principal excitations are Z and Z_2 vortexes in the XY and Heisenberg TLA's, respectively [24]. The corresponding correlation length has the form [25] $\xi_{\mathrm{KT}} = a \exp(b\tau_{\mathrm{KT}}^{-1/2})$, where a is of the order of the lattice spacing, $b \sim 1$, and $\tau_{\mathrm{KT}} = (T - T_{\mathrm{KT}})/T_{\mathrm{KT}}$, where T_{KT} is the Kosterlitz-Thouless transition temperature. According to [26] we have $T_{\mathrm{KT}} < T_N$, and in the 2D region ξ and the factor $\tau^{-(\lambda+\gamma)}$ in the expression for $S_{1,2}$ should be replaced by ξ_{KT} and $(\xi_{\mathrm{KT}}/a)^{7/8}$, respectively. At the same time, determination of the ω dependence remains a complex problem which we do not consider here (cf. [27]). We do not discuss here the quasi-1D problem either.

Below T_N the DC may be calculated using spin-wave theory with proposed nonlinear complications [28]. The results of the corresponding calculations will be published elsewhere.

Conventional antiferromagnets.—As state above, DC persists in any interacting spin system. We illustrate it by the example of an isotropic two-sublattice antiferromagnet, where the chirality is not a relevant variable. In the critical region above T_N the DC should be governed by conventional scaling, and instead of (12) and (13) we get $S_2 = 0$ and

$$S_1 = \left(\frac{g\mu H}{T_N^2 \tau^{\gamma}}\right) g\left[q\xi, \frac{\omega}{T_N \tau^{3\nu/2}}\right], \qquad (14)$$

where γ is the staggered susceptibility exponent, and we used the well-known value for the dynamical scaling exponent, $z = 3/2$. In this case the low-field approximation holds if $g\mu H \ll T_N$.

In the spin-wave region one can calculate the DC using linear spin-wave theory. There are two distinct cases: (1) the field along $\hat{c}$, where $\hat{c}$ is the direction of the sublattice magnetization, and (2) $\vec{H} \perp \hat{c}$. In the second case, if one takes into account the spin canting provoked by the field, Eq. (7) for the DC becomes exact, and after standard calculations for $\vec{Q}$ in the vicinity of the AF reciprocal lattice point, one gets

$$S_1(\vec{q}, \omega) = -2g\mu H S^2 J_0 \omega \{(\omega^2 - \epsilon_{\vec{q}}^2) \times [\epsilon_{\vec{q}}^2 + (g\mu H)^2]\}^{-1}, \qquad (15)$$

where $J_0 = \Sigma_{\vec{R}} J_{\vec{R}}$ and $\epsilon_{\vec{q}}$ is the spin-wave energy at $\vec{H} = 0$. This expression has a maximum at $g\mu H = \epsilon_{\vec{q}}$. It is valid if $H \ll H_{\text{spin flip}}$. The case $\vec{H} \| \hat{c}$ is realized for rather small $H < H_{\text{spin flop}}$ and we do not write here the corresponding expression. It should be noted that Eq. (15) holds if $T < \epsilon_{\vec{q}}$ only, otherwise the spin-wave interaction becomes important. This problem will be considered elsewhere.

Doped CuO_2 layers.—Eqs. (14) and (15) hold for quasi-2D antiferromagnets such as $YBa_2Cu_2O_{6+x}$ and La_2CuO_4. With doping, the AF order in the CuO_2 planes is destroyed, and the system becomes superconducting. In [10,29] it was suggested that in this case the chirality may be a relevant spin variable. However, due to very strong spin-spin interaction ($T_{\rm int} \sim J \sim 100$ meV) the DC is small, and direct measurement of the energy distribution related to the chiral scattering is hardly possible. Meanwhile, the energy transfer at the scattering is large as compared with T, and one may examine the integrated DC. In the conventional geometry [30] when the outgoing neutrons are perpendicular to the CuO_2 planes, from (9) and (10) one gets

$$\left(\frac{d\sigma}{d\Omega}\right)_{P_0} = 2P_0(r_0\gamma)^2|F(\vec{Q})|^2\frac{g\mu H}{T_{\rm int}^2}\int_{-\infty}^{E_i}\frac{d\omega(1-\omega/E_i)^{1/2}}{1-e^{-\omega/r}}\times\begin{cases}[g_1(\vec{Q}_\|,\omega)+g_2(\vec{Q}_\|,\omega)]Q_\perp^2/(Q_\|^2+Q_\perp^2), & \hat{h}\|\hat{c}\,,\\ g_1(\vec{Q}_\|,\omega)(\vec{Q}_\|\hat{h})^2/(Q_\|^2+Q_\perp^2), & \hat{h}\perp\hat{c}\,,\end{cases} \tag{16}$$

where $\vec{Q}_\|$ is a fixed two-dimensional momentum transfer, $Q_\perp^2 = k_i^2[\cos\alpha - (1-\omega/E_i)^{1/2}]^2$, and α is the angle between $\vec{k}_i$ and the $\hat{c}$ axis. If $H \sim 10T$, we have $g\mu H/T_{\rm int} \sim 10^{-2}$. In this case, $(d\sigma/d\Omega)_{P_0}$ may be of the order of a few percent as compared with the ordinary magnetic scattering and may be observed. It would be very instructive to study a possible enhancement of $(d\sigma/d\Omega)_{P_0}$ as a function of T and the doping. In such a way one can establish if any precursor to the chiral order really exists.

In summary, it has been demonstrated that the polarization dependent part of the neutron cross section is determined by the projection of the spin chirality on the sample magnetization induced by an applied field. This part of the scattering is purely inelastic, and due to its $\vec{P}_0$ dependence may be extracted from other scattering processes. It makes the chiral scattering very suitable for investigation of the spin dynamics. It was demonstrated that the chiral scattering should be strongly enhanced in the case of a phase transition in TLA's as well as in ordinary antiferromagnets. In particular, experimental study of the DC may clarify the problem of phase transitions in the TLA. The problem of the chiral scattering on CuO_2 layers in the high-T_c superconductors is discussed also.

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