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Spin chirality and polarized neutrons

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Abstract

Investigation of the chiral spin fluctuations by polarized neutrons is discussed. It is shown that it is possible in external magnetic field or in presence of the Dzyaloshinskii–Moriya interaction. Theoretical consideration is illustrated by several examples of the relevant experiments where new information was obtained inaccessible for conventional magnetic scattering. Possible studies of the spin chirality in different systems are discussed briefly.

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1. Introduction

In any correlated spin system except the Ising one there are dynamical noncollinear or chiral spin variable which is defined as a vector product of two lattice spins $\mathbf{S}_1 \times \mathbf{S}_2$. In some compounds at low temperatures it becomes frozen and form helical (spiral) spin structure. Polarized neutrons allow to determine completely the helix structure including the direction of its rotation [1,2]. Investigation of the chiral fluctuations is a more complex problem as the chirality is a two-spin operator and chiral fluctuations are described by the four-spin correlation function. Its direct measurement is impossible. (The so-called scalar chirality $(\mathbf{S}_1[\mathbf{S}_2 \times \mathbf{S}_3])$ is studied by the Raman scattering [3].)

An experimental study of the chiral fluctuations is required for verifying some theoretical results. We mention here predictions of new universality classes of the phase transition in triangular lattice antiferromagnets and helimagnets [4] and the chiral transition in spin glasses [5]. Investigation of the chirality gives important information for other correlated spin systems as it was shown for scattering in ferromagnets (see below).

In this paper we demonstrate how using polarized neutrons one can study chiral fluctuations and give a review of the main experimental achievements in this field. Our principal goal is to attract an attention to this branch of the polarized neutron studies of magnetism.

2. Conventional and chiral magnetic scattering

For clarifying what kind of new information could be obtained in the case of the chiral scattering we outline the theory of the

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conventional magnetic scattering. The inelastic scattering cross-section has the form

$$\sigma(\mathbf{Q},\omega) = N_{\rm in}\,{\rm Im}\,\chi_{\alpha\beta}(\mathbf{Q},\omega)(\delta_{\alpha\beta} - \hat{Q}_\alpha\hat{Q}_\beta), \tag{1}$$

where $\chi_{\alpha\beta}$ is the magnetic susceptibility and

$$N_{\rm in} = \frac{r^2(k_{\rm f}/k_{\rm i})F^2(\mathbf{Q})}{\pi[1-\exp(-\omega/T)]}. \tag{2}$$

If z-axis is along $\mathbf{Q}$ from the cross-section one determines the sum ${\rm Im}\,(\chi_{xx}+\chi_{yy})$. Measuring polarization of the scattered neutrons one can determine ${\rm Im}\,\chi_{xx}$ and ${\rm Im}\,\chi_{yy}$ separately along with the sum ${\rm Im}\,(\chi_{xy}+\chi_{yx})$. Indeed for x component of the scattered neutron polarization we have [6,7]

$$P_x\sigma(\mathbf{Q},\omega) = N_{\rm in}[P_{0x}{\rm Im}\,(\chi_{xx}-\chi_{yy}) + P_{0y}\,{\rm Im}\,(\chi_{yx}+\chi_{xy})], \tag{3}$$

similar equation holds for P_y and $P_z = -P_{0z}$.

In the elastic case ${\rm Im}\,\chi_{\alpha\beta}$ and $N_{\rm in}$ should be replaced by $\langle \mathbf{S}_{-\mathbf{Q}\alpha}\rangle\langle \mathbf{S}_{\mathbf{Q}\beta}\rangle$, where $\langle \ldots \rangle$ denotes the thermal average and $N_{\rm el} = r^2F^2(\mathbf{Q})$, respectively.

So for the symmetric tensor $\chi_{\alpha\beta}$ polarized neutrons allows to determine all its components perpendicular to the momentum transfer. It is well known that antisymmetric part is connected to an axial vector: $\chi^{\rm A}_{\alpha\beta} = -{\rm i}\varepsilon_{\alpha\beta\gamma}C_\gamma$ (we introduce factor "i" to simplify final expressions). It may appear if the system as a whole has some axial vector interaction. We consider below the sample magnetization induced by magnetic field and Dzyaloshinskii–Moriya interaction (DMI). In both cases the $\mathbf{P}_0$ depended part of the cross-section has the form [6,7]

$$\sigma_{\rm ch}(\mathbf{Q},\omega) = 2N_{\rm in}(\mathbf{P}_0\hat{Q})(\hat{Q}\,{\rm Im}\,\mathbf{C}). \tag{4}$$

Corresponding contribution to the scattered neutron polarization is equal to $-(\partial/\partial\mathbf{P}_0)\sigma_{\rm ch}$ and contains the same information as $\sigma_{\rm ch}$.

The chirality vector $\mathbf{C}(\mathbf{Q},\omega)$ is described by the vector product of the spin operators [8]

$$\mathbf{C}(\mathbf{Q},\omega) = \frac{1}{2}\int_0^\infty {\rm d}t{\rm e}^{{\rm i}\omega t}\langle \mathbf{S}_{-\mathbf{Q}}(t)\times\mathbf{S}_{\mathbf{Q}}(0) - \mathbf{S}_{\mathbf{Q}}(0)\times\mathbf{S}_{-\mathbf{Q}}(t)\rangle. \tag{5}$$

General properties of ${\rm Im}\,\mathbf{C}(\mathbf{Q},\omega)$ follow from the conditions of detailed balance and symmetry under time reflection [7,8]. We formulate here the final results only. For the centrosymmetric systems ${\rm Im}\,\mathbf{C}(\mathbf{Q},\omega,\mathbf{H})$ is the odd function of the magnetic field and the chiral scattering disappears at $\mathbf{H}=0$. It is an even function of ω and due to the Bose factor in $N_{\rm in}$ the $\sigma_{\rm ch}(\omega)$ changes sign at $\omega=0$. We name it dynamical chirality (DC). Its ω dependence differs qualitatively in classical $\omega_{\rm c}\ll T$ and quantum $T\ll\omega_{\rm c}$ cases where $\omega_{\rm c}$ is the characteristic energy of the spin (chiral?) fluctuations. In the classical case $\sigma_{\rm ch}$ is an odd function of ω. It was verified experimentally in the case of critical scattering in iron [9] and in the triangular lattice antiferromagnet $CsMnBr_3$ [10].

In the case of the DMI due to lack of inversion the vector ${\rm Im}\,\mathbf{C}$ is $\mathbf{Q}$ odd. It is ω odd too and $\sigma_{\rm ch}$ does not change sign at $\omega=0$. This difference in the ω dependence in both cases is a consequence of different t-parity of the magnetic field $\mathbf{H}$ and Dzyaloshinskii vector $\mathbf{D}_{\mathbf{Q}}$. The former is t odd and the latter is t even.

For small $\mathbf{H}$ the vector $\mathbf{C}(\mathbf{Q},\omega)$ is proportional to the field and the chiral scattering is determined by the three-spin dynamical fluctuations. It is a new physical object which contains additional information in comparison with the conventional magnetic scattering. The same holds for the DMI where the chiral scattering is determined by the four spin correlations. It is evident from the following expression for the DMI:

$$V_{\rm DM} = \frac{1}{2}\sum_{l,m}\mathbf{D}_{lm}[\mathbf{S}_l\times\mathbf{S}_m], \tag{6}$$

where the sum is over the pairs of spins connected by the vector $\mathbf{D}$.

3. The chiral scattering from magnetic helices

In the simple static helix the direction of the lattice spin $\mathbf{S}_m$ is given by

$$\mathbf{S}_m = \tfrac{1}{2}[\mathbf{S}{\rm e}^{-{\rm i}\mathbf{k}\mathbf{R}_m} + \mathbf{S}^*{\rm e}^{{\rm i}\mathbf{k}\mathbf{R}_m}], \tag{7}$$

where $\mathbf{S} = \mathbf{S}_1 + {\rm i}\mathbf{S}_2$. If $\mathbf{S}_{1,2}$ and $\mathbf{k}$ are mutually perpendicular and $S_1 = S_2 = S$ the scattering cross-section has the form [1,2]

$$\sigma_{\rm el}(\mathbf{Q}) = \tfrac{1}{4}[rSF(\mathbf{Q})]^2\{[1 + (\hat{Q}\hat{m})^2 + 2(\mathbf{P}_0\hat{Q})(\hat{Q}\hat{m})]\Delta_{\mathbf{Q}+\mathbf{k}} + [1 + (\hat{Q}\hat{m})^2 - (\mathbf{P}_0\hat{Q})(\hat{Q}\hat{m})]\Delta_{\mathbf{Q}-\mathbf{k}}\}, \quad (8)$$

where $\Delta_{\mathbf{Q}} = (2\pi)^3/v_0\sum_\tau \delta(\mathbf{Q}-\tau)$, v_0 is the unit cell volume, τ is the reciprocal lattice vector and $\hat{m} = [\mathbf{S}_1 \times \mathbf{S}_2]S^{-2}$. Hence, at proper choice of $\mathbf{P}_0$ one can suppress completely one of the Bragg reflections. It was demonstrated for itinerant magnet MnSi [12] where the direction of the spin rotation (sense of the helix) is fixed by the DMI. In the centrosymmetric systems the helix energy is independent on its sense and the sample should be split on domains. In the case of equal population of both kinds of domains the chiral part in Eq. (8) disappears. It can survive if the domain populations differ accidentally as it was observed in $CsMnBr_3$ [13]. The large difference in the domain populations could be obtained by cooling the sample subjected by some external forces which imitate the DMI. Proposed methods were reviewed in Ref. [6]. In particular elastic chirality is absent in helimagnet Ho but it was created by cooling the twisted sample below $T_N = 133$ K. [14] and the temperature dependence of the static chirality C was measured in critical region below T_N. It was found that $C \sim (-\tau)^{\beta_c}$ with $\beta_c = 0.90(3)$ and $\tau = (T - T_N)/T_N$. At the same time the intensity of the nonchiral part of the Bragg reflection (independent on $\mathbf{P}_0$) is proportional to $(-\tau)^{2\beta}$ where $2\beta = 0.76(2)$. Hence, the chirality $[\mathbf{S}_1 \times \mathbf{S}_2]$ decreases with τ faster than the square of the site magnetization. It contradicts to the naive model of [1,2] [Eqs. (7) and (8)] and confirm the idea that chirality is the independent relevant variable [4]. However, in $CsMnBr_3$ within the error bars we have $\beta_c = 2\beta \simeq 0.44$ in agreement with calculations [4]. The reason for this difference is not clear yet.

4. Chiral scattering in the triangular-lattice antiferromagnets and helimagnets

In the stacked triangular-lattice antiferromagnets (TLA) crystal structure consists of planes where spins are arranged in triangular lattice ($CsMnBr_3$, $CsNiCl_3$, etc.). In each basic spin triangle the 120° structure is double degenerated. This degeneracy is described by the Ising discrete variable which coincides with the sign of the triangle chirality $\mathbf{S}_1 \times \mathbf{S}_2 + \mathbf{S}_2 \times \mathbf{S}_3 + \mathbf{S}_3 \times \mathbf{S}_1$. In ferro and antiferromagnets the spin structure is determined by the spin direction at one of the lattice points. For the TLA along with the spin direction one has to take into account the discrete chirality and we have new classes of universality [4]. The chirality is now the relevant (critical) variable and there are two new critical exponents β_c and γ_c. The former describes the τ dependence of the average chirality below T_N and the latter the chiral susceptibility.

Evaluation of the vector $\mathbf{C}(\mathbf{Q},\omega)$ is a complex problem except the simplest case of the Heisenberg ferromagnets at low T. At the same time near the phase transition it could be analyzed using scaling theory [15], where any relevant variable $A(\mathbf{r})$ has special anomalous dimension Δ_A. The generalized susceptibility of two variables has the form

$$\chi_{AB}(\mathbf{Q},\omega) = \frac{1}{T_N\tau^{\nu(3-\Delta_A-\Delta_B)}}F\left(\frac{Qa}{\tau^\nu},\omega\right), \quad (9)$$

where a is of order of the lattice spacing and ν is the exponent determining the correlation length $\xi = a\tau^{-\nu}$.

For small magnetic field the DC describes correlation between the spin chirality and uniform magnetization for which dimensions are Δ_c and zero, respectively, and we obtain [6,8]

$$\mathbf{C}(\mathbf{Q},\omega) = \frac{g\mu_b H}{T_N^2\tau^{\phi_c}}\mathbf{\Phi}\left(\frac{\mathbf{Q}a}{\tau^\nu},\omega\right), \quad (10)$$

where $\phi_c = (3-\Delta_c)\nu = \gamma_c + \beta_c$. If $\mathbf{Q}$ is at the antiferromagnetic Bragg point the amplitude of the chiral scattering is proportional to $\tau^{-\phi_0}$. Results of corresponding measurements for $CsMnCl_3$ are shown in Fig. 1.

Both exponents ϕ_c and β_c were measured for $CsMnBr_3$ [13] and Ho [14]. For $CsNiCl_3$ the ϕ_c was determined only [17]. Hence, for the former two compounds two new chiral critical exponents were measured. For $CsMnBr_3$ the conventional exponents γ, ν, β and the specific heat exponent α were measured before. Their list with corresponding references may be found in Ref. [6]. All set of

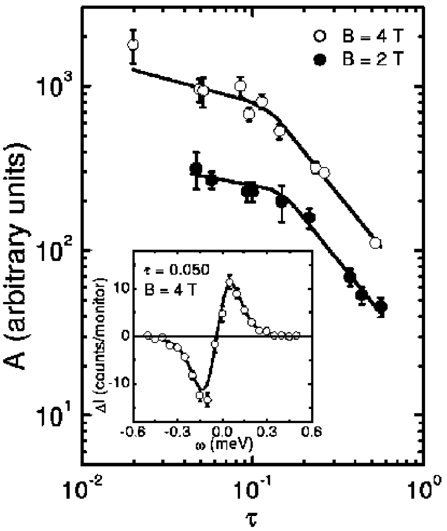

Fig. 1. The τ dependence of the DC in $CsMnBr_3$ for different fields and $\mathbf{Q} = (1/3, 1/3, 1)$. The ω oddness of the DC is shown in the inset [13]. The crossover at $\tau \sim 0.1$ disappears if one properly takes into account the resolution [16].

exponents is in a satisfactory agreement with the theory [4].

5. Critical chiral scattering in ferromagnets

The first time the DC was studied using small angle neutron scattering in iron near T_c [9,19,20]. To measure ω integrated chiral scattering the special inclined geometry was used with the field directed at angle $90° - \varphi$ to the neutron beam [6,11]. In this case ω oddness of σ_{ch} is compensated by the ω terms in expression for $\hat{Q}$ in Eq. (4) and the dependence on the scattering angle ϑ is studied. As $\mathbf{C} = \hat{h}C$ where $\hat{h} = \mathbf{H}/H$, and $\mathbf{P}_0$ is directed along or opposite to $\mathbf{H}$ to avoid the Larmor precession we have [6,11]

$$\sigma_{ch}(\vartheta) = \left(2r^2 T \frac{P_0}{\pi} \sin 2\varphi\right) \times \int \frac{d\omega (2E\vartheta) \mathrm{Im}\, C(Q,\omega)}{(2E\vartheta)^2 + \omega^2}. \qquad (11)$$

In weak field the DC is the three spin correlation function. In Ref. [18] general properties of multi-particle correlation functions were studied. It was shown that if one momentum Q is much larger than others and the inverse correlation length the dependence on Q singled out as a factor. It is a consequence of the conformal invariance of the theory at T_c. Scaling analysis gives $\sigma(\vartheta) \sim \tau^{-0.67}$ [6,11]. Corresponding experimental results for iron are in agreement with this prediction [19].

The crossover from the exchange to the dipolar critical dynamics near T_c in the chiral channel was observed in iron [20] in complete agreement with theory [11] (see also Ref. [6]).

Chiral scattering may be useful for solution of the conventional problems. As an example we mention determination of the spin-wave stiffness for two amorphous ferromagnets FeNiCrP near T_c [21]. The results are in agreement with the dynamical scaling.

6. DMI and chiral fluctuations

The DC study of the chiral scattering in the DMI magnets is in very beginning now. In this case $\sigma_{ch}(\mathbf{Q}, \omega)$ as function of ω has a definite sign and ω integrated chirality is not zero. It was observed in MnSi well above the transition temperature [22].

The chiral fluctuations were studied theoretically in the case of one-dimensional (1D) anti-ferromagnetic chain with the DMI [23,24]. It was shown from the exact solution of the problem that if the DMI has the same sign for each spin pairs there are incommensurate spin fluctuations with the chiral part. However, for the alternating DMI the fluctuations are commensurate without any chiral contribution. The mean-field calculations confirmed this result in 3D case [6,24]. Recently, these incommensurate fluctuations above T_c were studied in MnSi [25].

7. Conclusions

We have shown how using polarized neutrons one can study the chiral spin fluctuations and demonstrated that their investigation allows to get information which is inaccessible in conventional magnetic scattering. Really these studies of the spin chirality are in very beginning and one can mention a lot of problems which could be solved by this method.

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