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Frustrated magnets and polarized neutrons

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Abstract
A general theoretical approach to polarized neutron chiral scattering is described. It is shown that using polarized neutrons one can study the projection of the spin chirality on the axial-vector interactions and investigate critical chiral fluctuations. Applications of this method to the triangular lattice antiferromagnets, helimagnets and spin glasses are discussed.

1. Introduction

The important role of spin chirality (SC) was recognized during theoretical studies of frustrated magnets. It was shown that the SC is a relevant critical variable for triangular lattice antiferromagnets (TLA), along with the sublattice magnetization [1]. Numerical studies reveal the possibility of a chiral glass state in spin glasses (see [2] and references therein). The SC should be important for other frustrated magnets. Hence, its experimental investigation is an urgent problem.

The SC is determined as a vector product of the lattice spins: $\mathbf{C}_{12} = [\mathbf{S}_1 \times \mathbf{S}_2]$ and the chiral susceptibility is described by a four-spin correlation function. Its direct observation is a difficult experimental problem. Polarized neutrons allow one to partly solve it. If the system as a whole possesses some axial vector (magnetization, Dzyaloshinskii vector, etc), the tensor of the magnetic susceptibility acquires an antisymmetric part, which is a projection of the SC onto the axial-vector interaction. As a result the scattering cross section becomes dependent on the initial neutron polarization $\mathbf{P}_0$ [3–5]. This provides the possibility to study SC using polarized neutrons. By this method new chiral exponents were measured for the first time in the TLA $CsMnBr_3$, $CsNiCl_3$ and helimagnetic Ho [6–8].

In this paper we outline the theoretical basis of this method and its application to the TLA and helimagnets. We discuss also the possibility of using it to investigate spin glasses.

2. General properties

The antisymmetric part of the susceptibility tensor is connected to an axial vector: $\chi^A_{\alpha\beta} = -\mathrm{i}\epsilon_{\alpha\beta\gamma} C_\gamma$, where 'i' is introduced for convenience only and the vector $\mathbf{C}$ is given by [8]

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

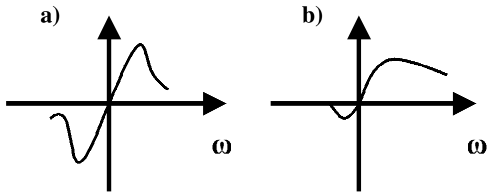

Figure 1. The energy dependence of the DC in classical $\omega_c \ll T$ (a) and quantum (b) cases where ω_c is the characteristic energy of fluctuations.

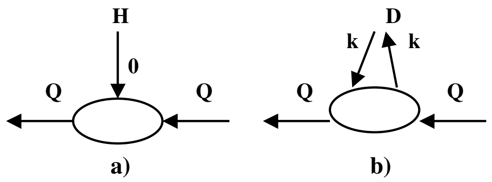

Figure 2. A diagrammatic representation of the polarized neutron chiral scattering in the cases of magnetic field (a) and Dzyaloshinskii–Moriya interaction (b).

where $\mathbf{S}_{-\mathbf{Q}}$ is the Fourier component of the spin density. For the chiral part of the cross section we have [3–5]

$$\sigma_{\rm ch}(\mathbf{Q}, \omega) = 2r^2 F^2(\mathbf{Q})(k_{\rm f}/k_{\rm i}) \frac{(\mathbf{P}_0 \hat{Q})(\hat{Q}\,{\rm Im}\,\mathbf{C}(\mathbf{Q}, \omega))}{1 - {\rm e}^{-\omega/r}}, \tag{2}$$

where $r = 5.4 \times 10^{-13}$ cm, $F(\mathbf{Q})$ is the magnetic form factor and $\hat{Q} = \mathbf{Q}/Q$. The general properties of ${\rm Im}\,\mathbf{C}(\mathbf{Q}, \omega)$ follow from the conditions of detailed balance and symmetry under time reflection. In centrosymmetric systems, ${\rm Im}\,\mathbf{C}$ exists in a magnetic field or in spontaneously magnetized ferromagnets (dynamical chirality (DC)). It is an odd function of the sample magnetization and an even function of ω. Due to the Bose factor in equation (1), $\sigma_{\rm ch}$ changes sign with ω and one has to distinguish two cases: classical ($\omega_c \ll T$) and quantum ($\omega_c \gg T$), where ω_c is the characteristic energy of fluctuations (see figure 1). In the former case, $\sigma_{\rm ch}$ is an odd function of ω and therefore the ω-integrated static SC is zero. This ω oddness was confirmed experimentally [6, 7].

In crystals with the Dzyaloshinskii–Moriya interaction (DMI), $\sigma_{\rm ch}$ is an odd function of $\mathbf{Q}$ but does not change sign with ω. This difference in the ω behaviour of $\sigma_{\rm ch}$ is a result of the difference in t parity of $\mathbf{H}$ and the Dzyaloshinskii vector $\mathbf{D}$.

For weak axial-vector interaction the chiral scattering is connected with three- and four-spin fluctuations in the $\mathbf{H}$ and $\mathbf{D}$ cases respectively (see figure 2). Evaluation of ${\rm Im}\,\mathbf{C}$ is a very complex problem except for the simplest case of ferromagnets below T_c.

3. TLA and helimagnets

According to [1] the TLA and helimagnets belong to the same chiral universality classes of the second-order phase transition. Near T_N an expression for ${\rm Im}\,\mathbf{C}$ can be obtained using scaling theory as demonstrated for ferromagnets [3, 5]. A similar approach is appropriate for TLA and helimagnets [5, 8]. According to the scaling theory, any relevant variable A has the special anomalous dimension Δ_A which determines the average value of A below the transition: $\langle A\rangle = (-\tau)^{\beta_A}$, where $\tau = (T - T_N)/T_N$, $\beta_A = \Delta_A \nu$ and ν is the correlation length exponent. The generalized susceptibility of two variables has the form

$$\chi_{AB}(\mathbf{Q}, \omega) = \frac{1}{T_{\mathrm{N}}\tau^{\nu(3-\Delta_A-\Delta_B)}} F\left(\frac{\mathbf{Q}a}{\tau^{\nu}}, \omega\right), \tag{3}$$

where a is of order of the lattice spacing [5, 6] and F depends on A and B. The explicit form of this function is inessential for us (see below). In a weak field, DC describes the correlation between the SC with dimension Δ_c and the magnetization whose dimension is zero and we get

$$\mathbf{C}(\mathbf{Q}, \omega) = \frac{g\mu_{\mathrm{B}}H}{T_{\mathrm{N}}\tau^{\varphi_c}} \phi\left(\frac{Qa}{\tau^{\nu}}, \omega\right), \tag{4}$$

where $\varphi_c = (3 - \Delta_c)\nu$. According to (3), the exponent of the chiral susceptibility $\gamma_c = \nu(3 - 2\Delta_c)$ and $\varphi_c = \beta_c + \gamma_c$. If $\mathbf{Q}$ is at the magnetic Bragg point, the τ dependence of $\mathrm{Im}\,\mathbf{C}$ is determined by the factor $\tau^{-\varphi_c}$ and one can measure this exponent [7, 8]. The exponent β_c could be measured using polarized neutrons too [5, 10, 11].

Some additional improvements of equation (4) allow us to correct experimental data and obtain deeper insight into the structure of $\mathrm{Im}\,\mathbf{C}$. According to figure 2(a), DC describes a process where the spin fluctuation scatters on the uniform magnetization and we can write

$$\mathbf{C}(\mathbf{Q}, \omega) = \left[\frac{\chi(q, \omega)}{\chi(0, 0)}\right]^2 \frac{1}{\tau^{\varphi_c}} f(q, \omega) \frac{g\mu_{\mathrm{B}}H}{T_{\mathrm{N}}}, \tag{5}$$

where $\chi(q, \omega)$ is the conventional susceptibility and q is the distance from the magnetic Bragg point. As the magnetization is not a relevant variable, one should assume nonsingular behaviour of the factor f. In this case, due to the ω evenness of $\mathrm{Im}\,C$, one should assume $f(q, \omega) = (\omega/T_{\mathrm{N}}) f_0$. Assuming also the experimentally established form [12]

$$\chi(q, \omega) = \frac{Z}{q^2 + \kappa^2} \frac{\mathrm{i}\Gamma_q}{\omega + \mathrm{i}\Gamma_q}, \tag{6}$$

where $\kappa \sim \tau^{-\nu}$ is the inverse correlation length and Z is a constant, we get [5]

$$\sigma_{\mathrm{ch}} \sim \frac{g\mu_{\mathrm{B}}H}{T_{\mathrm{N}}^2\tau^{\varphi_c}} \left(\frac{\kappa^2}{\kappa^2 + q^2}\right)^2 \frac{\omega\Gamma_q^3}{(\Gamma_q^2 + \omega^2)^2}. \tag{7}$$

At $q = 0$ the second factor is unity. However, with decreasing τ, the q resolution becomes larger than κ and one has to integrate over q up to $q_{\mathrm{max}} \gg \kappa$. As a result we get $\chi_{\mathrm{ch}} \sim \tau^{\varphi_c - 2\nu}$ [5]. This explains the crossover in the τ dependence of σ_{ch} observed in [6, 7] and corrects the results of those papers [13].

4. Spin glasses

For spin glasses, equations (4) and (5) are applicable above T_g for the small angle scattering. The increase of σ_{ch} at $\tau = (T - T_g)/T_g \to 0$ would be evidence that the SC is a relevant variable.

Below T_g the sample magnetization slowly relaxes after the field is switched off. Then the chiral scattering should be observed both in field cooled (FC) and zero-field cooled (ZFC) experiments. Corresponding studies would provide additional information about the spin glass state.

In spin glasses characteristic energies are distributed according to the $1/\omega$ law [14] over the wide range below some cut-off energy Ω_c. This means that below T_g in a very crude approximation we have

$$\sigma_{\mathrm{ch}}(Q, \omega) = \int_{\Gamma_q}^{\infty} \frac{\mathrm{d}\Gamma}{\Gamma} \sigma_{\mathrm{ch}}(Q, \omega, \Gamma). \tag{8}$$

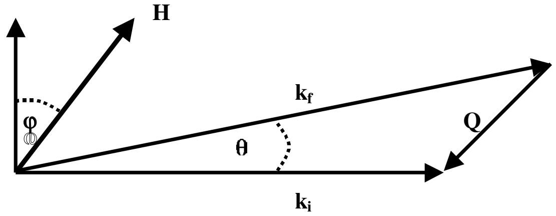

Figure 3. The geometry of the chiral scattering in the inclined magnetic field.

Assuming that below T_g the ω dependence of $\sigma_{ch}(Q, \omega, \Gamma)$ is the same as in equation (7), we obtain

$$\sigma_{ch}(Q, \omega) = \sigma_{ch}(Q) \begin{cases} \dfrac{\omega}{\Gamma_q}, & |\omega| \ll \Gamma_q; \\ \dfrac{\pi}{4} \operatorname{sgn} \omega, & \Gamma_q \ll |\omega| \ll \min(\Omega_0, T). \end{cases} \quad (9)$$

We see that there is a wide ω range where $\sigma_{ch}(Q, \omega)$ depends on $\operatorname{sgn} \omega$ only. Really, one can expect $\sigma_{ch} \sim |\omega|^a \operatorname{sgn} \omega$ with $a \ll 1$.

The prefactor σ_{ch} has to have a different form in the critical region and at low T. In the former case we can write

$$\sigma_{ch} \sim (-\tau)^{\nu_c \Delta_c} \frac{g_B \mu H P_0 \operatorname{sgn} \omega}{T_g^2 [1 + (Qa/\tau^\nu)^2]^2}, \quad (10)$$

where according to [2] we introduce two correlation range exponents ν and ν_c. In the low temperature region where both correlation lengths have to be of the order of a, we have $\sigma_{ch}(Q) \sim g_B \mu H P_0 / T_g^2$ and it is Q independent for $Qa \ll 1$.

In spin glasses the chiral scattering should be weak, and investigation of the ω dependence of σ_{ch} is a difficult experimental problem. At the same time the ω-integrated chiral scattering in a classical case is zero (see above). So one can use the so-called inclined geometry applied for small angle critical scattering in ferromagnets [3, 5, 15]. In this case the field is directed at an angle $90° - \varphi$ to the neutron beam (see figure 3). In spin glasses, $\mathbf{C} = \hat{h}\mathbf{C}$ where $\hat{h} = \mathbf{H}/H$ and $\mathbf{P}_0 = P_0 \hat{h}$ to avoid the Larmor precession. As a result the factor $P_0(\hat{h}\hat{Q})^2$ appears in equation (2). For the small angle scattering it has a term $P_0(2E\vartheta\omega \sin 2\varphi)[\omega^2 + (2E\vartheta)^2]^{-1/2}$; the oddness of $(T \operatorname{Im} C)/\omega$ is compensated by this term and the ω-integrated chiral scattering has the form

$$\sigma_{ch}(\vartheta) = (2r^2 T P_0 \sin 2\varphi) \int \frac{d\omega (2E\vartheta) \operatorname{Im} C(Q, \omega)}{(2E\vartheta)^2 + \omega^2}, \quad (11)$$

where $Q = k(\vartheta^2 + \omega^2/4E^2)^{1/2}$. We see that now instead of ω oddness we get ϑ oddness of $\sigma_{ch}(\vartheta)$. This integrated chirality could be easily extracted from the total scattering intensity, as demonstrated in [15] for the critical scattering in ferromagnets.

At small T in the range where $\sigma_{ch}(\omega) \sim \operatorname{sgn} \omega$, from equation (11) we get

$$\sigma_{ch}(\vartheta) \sim 2E\vartheta \ln \frac{\Omega_0}{2E\vartheta}; \qquad 2E\vartheta > \Gamma_{k\vartheta}, T. \quad (12)$$

Here we have taken into account the Q independence of $\sigma_{ch}(Q)$. Investigation of other limiting cases is beyond the scope of this paper and could be done easily in connection with corresponding experimental studies. However, one has to have in mind that the chiral scattering increases with T.

5. Conclusions

We have described a general theoretical approach to the chiral scattering of polarized neutrons and demonstrated its efficiency for investigation of the spin chirality in the TLA and helimagnets. We discuss also a possible application of the method for investigation of the SC in spin glasses. Obviously there could be many applications to other frustrated magnetic systems.

Acknowledgments

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