Spin chirality and polarized neutrons

S.V. Maleyev

Petersburg Nuclear Physics Institute named by B.P. Konstantinov of National Research Centre "Kurchatov Institute" E-mail: maleyevsv@mail.ru

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There are the static and dynamical spin chiralities. The former is a result non-collinear spin structure. The second takes place in the magnetized samples. Both are considered and illustrated by results of the neutron scattering experiments.

Keywords: non-collinear spin structure, spin chiralities, neutron scattering experiments.

1. Historical notes

Ural Winter School Kourovka 1962. After the session devoted to magnetic spirals three young theoretics, Vitia Bariahtar, Rob Suris, and the author discussed this problem with Ruben Alihanian, one of the first Soviet neutronists. Ruben asked, "What can do the polarized neutrons (PN) in this field?" Our answer was published in the end of that year [\[1\]](#page-2-0). Similar results were published by M. Blune in the next year [\[2\]](#page-2-1)). In these papers, general expressions were derived for the PN scattering and new phenomena were predicted: i) Rotation of the polarization at the scattering. ii) Dependence of the scattering on the polarization in the case of the non-collinear (chiral) magnets. To investigate these phenomena one must study the behavior of the polarization (**P**) in zero magnetic field. There were several steps in the solution of this nontrivial experimental problem. First of all, it was solved in the case of the nonscattered neutrons by Rekveldt (1971, Delft) [\[3\]](#page-2-2) and Drabkin and Gordeev (1972, Gatchina) [\[4\]](#page-2-3). Then it was solved for the small angle scattering by Okorokov (1975, Gatchina) [\[5\]](#page-2-4). General solution was presented by Tasset (1987, ILL, Grenoble) [\[6\]](#page-2-5). The full-scale PN studies were possible after these achievements. I remember several conferences devoted to the PN scattering: Grenoble, Gatchina, Aachen, Jülich, Berlin, and Delft. European Neutron Conference (Petersburg 2019) should be mentioned too.

In this paper, I outline the main features of the PN scattering theory and illustrate them by some experimental results by my Gatchina friends.

2. Theoretical background

Based on $[1,2]$ $[1,2]$ (see $[7]$ also) we present general results of the neutron magnetic scattering theory. The scattering amplitude is given by [\[8\]](#page-2-7)

$$
f(\mathbf{q}) = -1.9r_0 F(\mathbf{q})(\mathbf{S}_{\mathbf{q}}^{\perp} \cdot \boldsymbol{\sigma}), \ \mathbf{S}_{\mathbf{q}}^{\perp} = \mathbf{S}_{\mathbf{q}} - (\mathbf{S}_{\mathbf{q}} \cdot \hat{q})\hat{q}, \ \hat{q} = \mathbf{q}/q, \tag{1}
$$

where r_0 is the classical electron radius, F is the magnetic ion form-factor, **S** is the spin density, and σ is the neutron Pauli. For the scattering intensity *I* and polarization of the scattered neutrons $P = \langle \sigma \rangle$ we get [\[1](#page-2-0)[,2,](#page-2-1)[7\]](#page-2-6)

$$
I \sim \left\langle \left\langle \mathbf{S}_{\mathbf{q}}^{\perp} \cdot \mathbf{S}_{-\mathbf{q}}^{\perp} \right\rangle + i \left(\mathbf{P}_{0} \cdot \left\langle \left[\mathbf{S}_{\mathbf{q}}^{\perp} \times \mathbf{S}_{-\mathbf{q}}^{\perp} \right] \right\rangle \right) \right\rangle, \tag{2}
$$

$$
I \sim \left\{ \left\langle (\mathbf{P}_{0} \cdot \mathbf{S}_{\mathbf{q}}^{\perp}) \mathbf{S}_{-\mathbf{q}}^{\perp} + \mathbf{S}_{\mathbf{q}}^{\perp} (\mathbf{S}_{-\mathbf{q}}^{\perp} \cdot \mathbf{P}_{0}) - \mathbf{P}_{0} (\mathbf{S}_{\mathbf{q}}^{\perp} \cdot \mathbf{S}_{-\mathbf{q}}^{\perp}) \right\rangle - \right. \\ \left. - i \left\langle \left[\mathbf{S}_{\mathbf{q}}^{\perp} \times \mathbf{S}_{-\mathbf{q}}^{\perp} \right] \right\rangle \right\rangle, \tag{3}
$$

where P_0 is the initial polarization and $\langle ... \rangle$ is the thermal average [\[7\]](#page-2-6). The scattering intensity is a scalar, the neutron polarization is an axial vector which changes the sign at the time reflection. So the second term in Eq. (2) appears if the scatterer has a corresponding property which is known as the spin chirality. There are two types of the chirality:

i) The static chirality (SC). It is in non-collinear spin structures and may be observed at the elastic PN scattering.

ii) The dynamical chirality (DC). It takes place in the magnetized samples, where the magnetization has the same symmetry as **P**. It is seen in the inelastic PN scattering.

The first three terms in Eq. (3) describe the change of the polarization after scattering. For paramagnets, we have ${\bf P} = -\hat{q}(\hat{q} \cdot {\bf P}_0)$ [8]. This effect was observed the first time at the critical scattering in iron by Drabkin *et al.* [\[9\]](#page-2-8).

In the ordered magnets the P_0 rotation predicted in [\[10\]](#page-2-9). It depends on the Bragg reflection. So it is used to study very complex magnetic structures (see Brown *et al.* [\[11\]](#page-2-10) as an example). The last term in Eq. (3) is the polarization which appears at the scattering in the chiral crystals.

3. Static chirality

The SC is a result of an non-collinear spin structure. It is characterized by the axial vector $C = [S_k \times S_{-k}]$, where **k** is the wave vector of the structure.

The simplest non-collinear helical structure is determined as follows

$$
\mathbf{S}_{\mathbf{R}} = S \left[\hat{a} \cos \mathbf{k} \cdot \mathbf{R} + \hat{b} \sin \mathbf{k} \cdot \mathbf{R} \right],\tag{4}
$$

where the unit vectors $\hat{a} \perp \hat{b}$, $[\hat{a} \times \hat{b}] = \hat{c}$ and $\mathbf{C} = S^2 \hat{c}$. The relative \hat{c} , **k** orientation determines the type of the helix. For $\mathbf{k} \perp \hat{c}$ and $\mathbf{k} \parallel \hat{c}$ we have the cycloid (multiferroics) and planar helix (*B*20 magnets) respectively. In both these cases, the helical structure is a result of a competition between the exchange and Dzyaloshinskii–Moriya (DM) interactions.

From Eqs. (2) and (4) we get $[1,2]$ $[1,2]$

$$
I \sim S^2 \left\{ [\delta(\mathbf{q} - \mathbf{k}) + \delta(\mathbf{q} + \mathbf{k})] [1 + (\hat{c} \cdot \hat{q})^2] \right\} +
$$

+ 2S² [\delta(\mathbf{q} - \mathbf{k}) - \delta(\mathbf{q} + \mathbf{k})] (\hat{c} \cdot \hat{q}) (\hat{q} \cdot \mathbf{P}_0). (5)

This expression does not change at the time reflection. Indeed, at $t \to -t$ we have $\mathbf{q} \to -\mathbf{q}$, $\mathbf{P}_0 \to -\mathbf{P}_0$ and the intensity remains unchanged.

In B20 magnets (MnSi etc.) we have $\mathbf{k} \parallel \hat{c}$, if $\mathbf{P}_0 \parallel \pm \mathbf{k}$ one of the Bragg reflections may be suppressed. It was observed in MnSi [\[12\]](#page-2-11).

The 1D spin chain with the DMI was studied in [\[13](#page-2-12)[,14\]](#page-2-13). The existence of SC fluctuations was demonstrated. This theoretical result has been confirmed by the critical PN scattering in MnSi above T_c .

The dependence of the scattering on the sign of P_0 in paramagnetic state contradicts to the standard Bak–Jensen theory [\[15\]](#page-2-14), where the DM vector was defined as $\mathbf{D}_{q} = d\mathbf{q}$. The difference was ignored between the left and right crystal chirality which connected with the magnetic one. We propose $D_q = d [q \times \hat{N}]$, where the unit vector \hat{N} is along one of the cubic axes and the sign depends on the crystal chirality. In this case the second term in Eq. (2) is a scalar proportional to $(P_0[q \times N])$ instead of the pseudo-scalar

Fig. 1. Chiral scattering in MnSi above T_c for opposite directions of P_0 along $(1,1,0)$.

 $(q \cdot P_0)$ in the Bak–Jensen model. The similar expression for D_q was used in [\[16\]](#page-2-15).

There are two types of magnetic helices.

i) The DM helices. Their structure is determined by the minimum of the sum of the exchange and DM energies: $\nabla (J_{\mathbf{q}} + \hat{c} \cdot \mathbf{D}_{\mathbf{q}}) = 0$. In this case, the vectors \hat{c} and **k** are connected.

ii) Frustrated helices. In this case, the **k** structure is determined by $\nabla J_{\mathbf{q}} = 0$. The \hat{c} directions are determined by the crystal anisotropy. So for each, **k** there is even number of \hat{c} with the same energy.

According to Kawamura [\[17\]](#page-2-16) the second-order phase transition in the frustrated magnets belongs to the new class of the universality where the chirality is an additional critical variable. In particular it was predicted that $\langle \hat{c} \rangle \sim (T_N - T)^{p_c}$. The PN scattering in triangular antiferromagnets CsMnBr₃ confirmed it. Plakhty found that $\beta_c = 0.44(2)$ in agreement with Monte Carlo calculations [\[18\]](#page-2-17).

4. Dynamical chirality

The dynamical chirality appears in the magnetized spin systems. It is studied by the inelastic PN scattering.

The general expression for dynamical chirality (DC) part in Eq. (2) is following

$$
D \sim Z^{-1} \sum e^{-E_a/T} \left[(S_q^x)_{ab} (S_{-\mathbf{q}}^y)_{ba} - (S_q^y)_{ab} (S_{-\mathbf{q}}^x)_{ba} \right] \times
$$

$$
\times \delta(\omega + E_{ab}) \hat{q}_z (\hat{q} \cdot \mathbf{P}_0), \tag{6}
$$

where z is the direction of magnetization, $E_{a,b}$ are the energies of the sample, $Z = \sum \exp(-E_a / T)$, $E_{ab} = E_a - E_b$, $ω = E_i - E_f$ is the energy transfer, E_i and E_f are the neutron energies before and after scattering respectively.

To elucidate this general expression we consider **D** for ferromagnetics. Using the spin-wave theory we get

$$
\mathbf{D}_{S_W} \sim 2S \left[-(N_{\mathbf{q}} + 1) \delta(\omega + \varepsilon_{\mathbf{q}}) + N_{\mathbf{q}} \delta(\omega - \varepsilon_{\mathbf{q}}) \right] \hat{q}_z (\hat{q} \cdot \mathbf{P}_0),
$$
\n(7)

where $\varepsilon_q = Aq^2$ is the spin wave energy and N_q is Plank function. Replacing in the first term $-\rightarrow$ + and $\hat{q}_z(\hat{q} \cdot \mathbf{P}_0) \rightarrow 1 + q_z^2$ we obtain corresponding expression for the conventional spin-wave scattering. So using polarization one can suppress the spin-wave emission or absorption.

The P_0 dependence of the scattering allows to study the spin waves in amorphous ferromagnets $Fe_{50}Ni_{22}P_{18}Cr_{10}$ and $Fe_{48}Ni_{24}P_{18}Cr_{10}$ near T_c (Okorokov *et al.* [\[19,](#page-2-18)[20\]](#page-2-19)). In these compounds, due to strong disorder, conventional methods do not work. The measurements od the chirality allows determining the temperature dependence or the spin-wave stiffness. It was found that $A \sim (T_c - T)^x$ where $x = 0.36 \pm 0.02$ and $x = 0.31 \pm 0.02$ for the first and second compounds respectively in agreement with the dynamical scaling.

In $[21]$, it was shown that in ferromagnets above T_c in weak field *D* may be represented as the three spin fluctuations. It depends on **q** and the inverse correlation radius κ. The many spin fluctuations above T_c were studied by [\[22\]](#page-2-21). In our case at $q \gg \kappa$ we have $D \sim 1/(q^{7/2} \kappa^{3/2})$. In ferromagnets $\kappa \sim \tau^{\nu}$, where $\tau = (T - T_c)/T_c$ and $\nu \approx 2/3$. So $D \sim 1/\tau$. This prediction was confirmed by the chiral scattering in Fe [\[23\]](#page-2-22) and EuS [\[14\]](#page-2-13).

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Спінова кіральність та поляризовані нейтрони

С.В. Малєєв

Існують статичні та динамічні спінові кіральності. Перша є результатом неколінеарної спінової структури. Друга спостерігається у намагнічених зразках. Обидві розглядаються та ілюструються результатами експериментів з розсіяння нейтронів.

Ключові слова: неколінеарна спінова структура, спінові кіральності, розсіяння нейтронів.