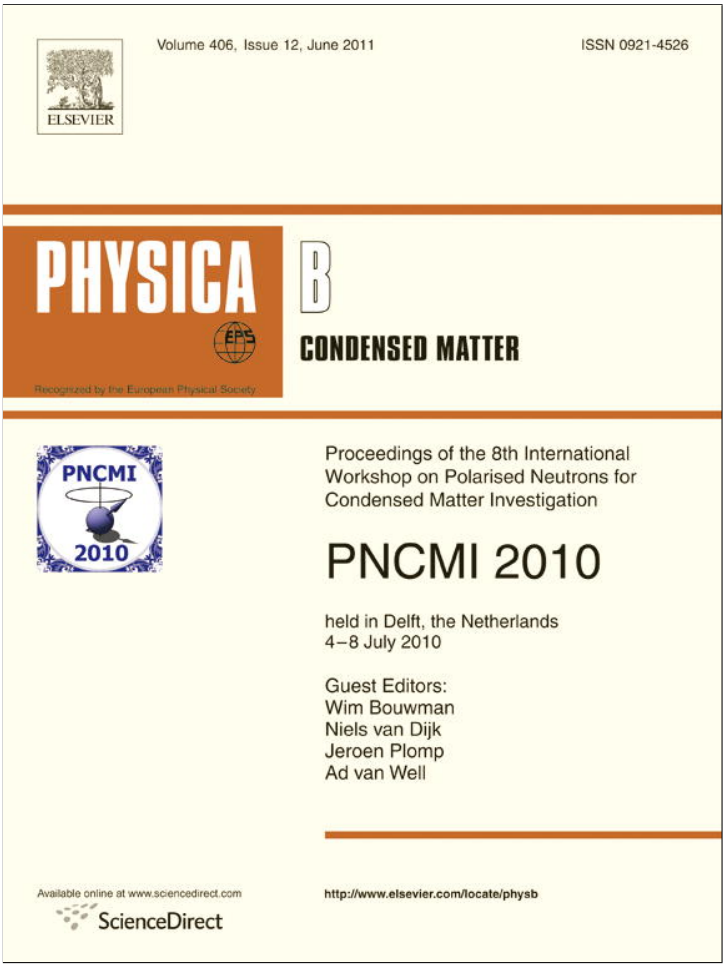

Physica B 406 (2011) 2385–2388

Contents lists available at ScienceDirect

Physica B

journal homepage: www.elsevier.com/locate/physb

Spin chirality of polycrystalline MnSi, or, difficult way from rumours to the solid ground

V.A. Dyadkin [a,*], S.V. Grigoriev [a], D. Menzel [b], E.V. Moskvin [a], S.V. Maleyev [a], H. Eckerlebe [c]

[a] Petersburg Nuclear Physics Institute, 188300 St. Petersburg, Russia
[b] Institut für Physik der Kondensierten Materie, Technische Universität Braunschweig, 38106 Braunschweig, Germany
[c] GKSS Forschungszentrum, 21502 Geesthacht, Germany

ARTICLE INFO

Available online 18 November 2010

Keywords:
Dzyaloshinskii–Moriya interaction
Spin chirality
Small-angle scattering of polarized neutrons

ABSTRACT

Six different polycrystalline samples of MnSi were produced as a rapidly frozen melt after the single crystal growth in the Czochralski process. With the help of small-angle scattering of polarized neutrons we show that the magnetic spin chirality being scanned along the sample length oscillates strongly on a scale of 2–3 mm, revealing the volumes of the samples with high degree of an enantiomeric excess. The average chirality of the samples deviates from zero value.

1. Introduction

Since the middle of the 1980s after Tanaka's work when seven different MnSi crystals as well as three $Fe_{1-x}Co_xSi$ with $x=0.20$, 0.40 and 0.50 were found to be structurally left-handed for all samples [1,2], the "broken chiral symmetry paradigm" has unintentionally puffed up on the level on rumours. This "paradigm" consisted in a statement that MnSi-related compounds can exist as a left-handed structure of $P2_13$ symmetry (cubic noncentrosymmetric B20) only.

As these compounds are long-ordered helical magnets with Dzyaloshinkii–Moriya interaction [3–5] the concomitant spin helix chirality measured in those MnSi samples was also found to be left-handed but right-handed for $Fe_{1-x}Co_xSi$ solid solutions [2,6]. In our previous works [7,8] we have shown that the magnetic chirality is strongly connected to the crystallographic handedness: a left-handed $P2_13$ structure generates a left-handed spin helix for MnSi-related compounds but a right-handed helix for $Fe_{1-x}Co_xSi$ alloys. This well established relation between structural and magnetic chirality of MnSi-type compounds allows one to determine the magnetic chirality only.

Surprisingly, the spin helix chirality of the investigated $Fe_{1-x}Co_xSi$ single crystals came out to be left-handed for $x<0.2$ and right-handed for $x\geq 0.2$ [7,8]. This result, however, has to be viewed under the constraint, that the above described experiments were carried out with very few but considerably large single crystals. Therefore, it cannot be undoubtedly concluded that the $Fe_{1-x}Co_xSi$ system exhibits any enantiomeric excess or preference for left- or right-handed crystals. Instead, it can be assumed that the growth technique must affect the crystallization process. Generally, within the Czochralski method a growing single crystal inherits the crystallographic orientation of its seed crystal which is submerged in the melt [9]. Thus, the structural chirality is also transferred from the seed to the new crystal.

The energy gain due to a preference for the left or right crystallographic handedness, if such a tendency exists, must be rather small and, therefore, the usual crystallization temperature dominates the system. In order to detect such a preference one has to inspect a large number of freely solidified single crystals or, for example, polycrystalline samples, in which neither the crystallographic orientation nor the chirality are transferred from one neighbouring crystallite to another. As a result, a large amount of crystalline grains inside a polycrystal sample should tremendously increase the measuring statistics of the chirality "preferred by nature".

The chirality γ of the polycrystalline sample is, by definition, the normalized difference between numbers of grains with left (N^l) and right (N^r) crystallographic or spin-helix handedness, respectively:

$$\gamma=\frac{N^l-N^r}{N^l+N^r}. \tag{1}$$

If there is a tendency of the growing crystal towards a "natural" chirality $\langle\gamma\rangle_n$ present, the number of crystallites in a polycrystal ($N=N^l+N^r$) must be large in order to satisfy the conditions for good statistics:

$$\sqrt{N}\ll\langle\gamma\rangle_n\cdot N. \tag{2}$$

In this work we used the small-angle scattering of polarized neutrons in order to measure the magnetic (crystallographic) chirality of the samples. For the experiments with polarized neutrons the value of γ is connected with the polarizing ability P_s of the sample [10]:

$$P_s=\gamma(\mathbf{P}_0\cdot\hat{\mathbf{q}}), \tag{3}$$

* Corresponding author. Tel.: +7 813 714 61 21.
E-mail address: dyadkin@lns.pnpi.spb.ru (V.A. Dyadkin).

0921-4526/$ - see front matter © 2010 Elsevier B.V. All rights reserved.
doi:10.1016/j.physb.2010.11.023

where $\hat{\mathbf{q}}$ is the unit scattering vector and $\mathbf{P_0}$ is the incident neutron beam polarization vector.

The polarization P_s can be directly determined from the polarized SANS experiment as

$$P_s = \frac{n(+P_0,\mathbf{Q}) - n(-P_0,\mathbf{Q})}{n(+P_0,\mathbf{Q}) + n(-P_0,\mathbf{Q})}, \tag{4}$$

where n is the scattered neutron intensity measured at the same point of the reciprocal space $\mathbf{Q}=\mathbf{k}$ for the polarization along $(+P_0=\mathbf{P}_0\uparrow\uparrow\mathbf{h})$ and opposite $(-P_0=\mathbf{P}_0\downarrow\uparrow\mathbf{h})$ the guiding magnetic field $\mathbf{h}$. Here $\mathbf{k}$ is a spin helix wave vector.

The accuracy of the determination of the polarizing ability P_s and, therefore, the chirality γ is limited by the applied method, i.e. by the statistics of the neutron scattering. To satisfy the conditions of good statistics the number of detected neutrons n must also be large:

$$\sqrt{n} \ll P_s \cdot n. \tag{5}$$

Ideally, the number of detected neutrons must be much larger than the number of the crystallites: $n \gg N$. In this case the accuracy of the determination of the chirality $\langle\gamma\rangle_n$ is limited by the object under investigation (polycrystalline sample) but not by the method of polarized neutron scattering.

2. Samples and the experimental setup

In this work we inspected six large polycrystalline MnSi samples, which were left over after the Czochralski single crystal growth process. The samples were prepared as a stoichiometric mixture of Mn and Si which was molten by the tri-arc method under argon atmosphere. A seed crystal with a defined chirality (see Table 1) was submerged into the melt, and a new single crystal was pulled out by the Czochralski technique. After the new single crystal was completely removed from the liquid phase, the temperature of the melt was abruptly decreased and the liquid has crystallized. Such obtained MnSi polycrystals, which were a rest round lump with a diameter of about 10 mm, had been used for the measurements.

The SANS experiments were carried out at the PNR scattering facility of the FRG-1 research reactor in Geesthacht (Germany). A polarized neutron beam with a neutron wavelength $\lambda=0.6$ nm, a bandwidth $\Delta\lambda/\lambda=0.1$, was used. The scattered neutrons were detected by a position sensitive ^{3}He detector with 128×128 pixels. A magnetic field guiding the polarization was applied horizontally within the sample's plane. The temperature was measured with an accuracy better than 0.05 K.

Six polycrystalline MnSi samples were placed inside an aluminic closed-cycle refrigerator which was mounted on the step motor. The exit slits were about 1 mm. The sample was cooled down to $T=15$ K in a magnetic field about 80 mT converting the sample into a conical monodomain phase with the helix wave vector parallel to the field axis ($\mathbf{k}\|\mathbf{H}$) [11]. As a result, the scalar product in Eq. (3) is equal to $|\mathbf{P}_0|$. The conditions described above are well suited to the measurements of the sample's chirality, since the magnetic field does not change the chirality of MnSi single crystal below $T_c\approx29$ K as shown in Ref. [11]. The measurements of the polarizing scattering ability P_s were performed within a scan across the whole sample with a step width of 1 mm. Combining Eqs. (3) and (4) the chirality γ has been calculated for every step along the sample length x.

Table 1
The average chirality of polycrystalline MnSi samples.

MnSi	$\langle\gamma\rangle_I$	Seed's chirality	Coincide
a	-0.180 ± 0.009	Left	+
b	-0.111 ± 0.007	Right	−
c	-0.012 ± 0.006	Left	+
d	$+0.102\pm0.005$	Right	+
e	-0.048 ± 0.005	Left	+
f	$+0.160\pm0.006$	Right	+
Sum	-0.09 ± 0.02	–	–

3. Results and discussion

The results of the scans for the six samples are shown in Fig. 1. Negative values of γ represents the regions with prevailing left-handed spin helices whereas positive γ values display a predominating right handedness. It should be noted that in accordance to Refs. [7,8] the measured value of the magnetic chirality for MnSi is exactly equal to the crystallographic handedness $\gamma\equiv\gamma_{cr}$.

As is well seen in Fig. 1, the chirality of every sample oscillates strongly, reaching at some points rather high values. For example, for sample $MnSi_a$ the chirality $\gamma\approx-0.8$ at $x=3.5$, and for sample $MnSi_d$ the chirality $\gamma\approx0.6$ at $x=10$. For sample $MnSi_f$ the chirality demonstrates a quasi-periodic behaviour reaching absolute values of $\gamma\approx0.7$ for both the left- and right-handed counterparts.

These large oscillations of the chirality can be explained under the assumption that these polycrystals consist of rather large monocrystalline domains. The surface photograph of the $MnSi_f$ shown in Fig. 2 has confirmed that the polycrystals contain grains with the size of about $2\times2\times2$ mm^3. If the non-zero value of the average chirality is caused by the statistics of the crystallite's number, one can estimate from Fig. 1 the number of the crystallites under inspection using the following equation: $\gamma=\sqrt{N}/N\to N\approx\gamma^{-2}$.

Taking the mean over many crystallites of the polycrystal, we calculated the average chirality of the whole sample $\langle\gamma\rangle_I$ as

$$\langle\gamma\rangle_I = \frac{\sum_x I_x(+P_0,\mathbf{q}) - \sum_x I_x(-P_0,\mathbf{q})}{\sum_x I_x(+P_0,\mathbf{q}) + \sum_x I_x(-P_0,\mathbf{q})}\cdot\frac{1}{P_0}. \tag{6}$$

The calculated values for all six samples are summarized in Table 1. The chirality of the seed crystals used during the crystal growth in the Czochralski method is also given in Table 1.

The error bars shown in Fig. 1 and in Table 1 are taken from the statistics of the scattering intensity. The numbers of detected neutrons during one measurement was $n\approx10^4$. Thus, the relative error was $\sqrt{n}/n\approx10^{-2}$. This statistics as well as the obtained error bars were small enough to determine the chirality of these samples. The number of the grains contributing to the statistics for the chirality is rather low in these samples. As soon as the typical size of a crystallite is about $2\times2\times2$ mm^3 and the sample volume is 1 cm^3, then the sample contains $N\approx100$ crystallites. Therefore, the statistical error in the determination of the chirality $\langle\gamma\rangle_n$ is $\sqrt{N}/N\sim10^{-1}$ and, therefore, in the order of the error for the average chirality shown in Table 1. This implies that these values are solely determined by poor statistics. Thus, one can conclude that condition given by Eq. (2) is not well fulfilled.

The average chirality $\langle\gamma\rangle_I$ shown in Table 1 deviates from zero and is of the order of 0.1–0.2. This deviation coincides with the seed's chiralities for five samples. Although noticeable, this is most probably an accident, and one would not expect a causal relation between the average chirality of the polycrystal and that of the seed.

We have summed up the average values of the chirality for all samples and have obtained the value of $\gamma_{overall}=-0.09\pm0.02$. We remind to the reader that this error bars are based on the neutron statistics. This indeed allows one to measure the chirality of all

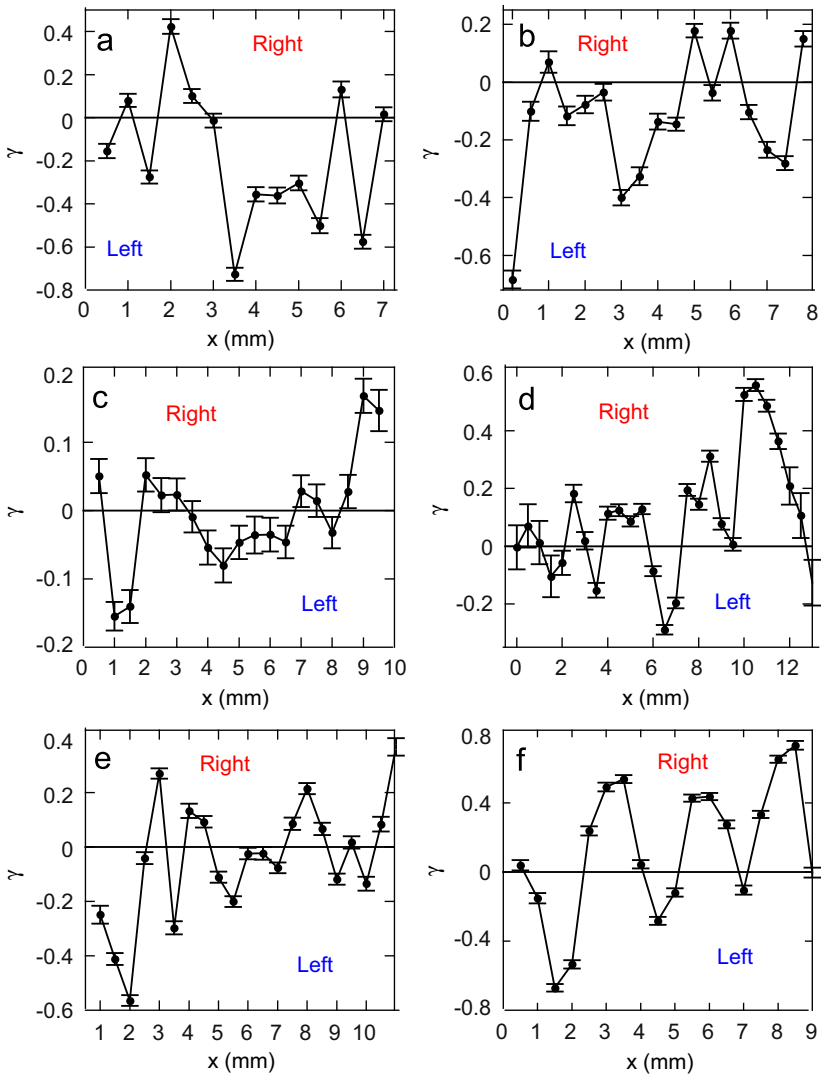

Fig. 1. Space-resolved spin chirality γ along the sample length x for six MnSi polycrystals. Positive and negative values of γ represent right- and left-handed spin helices, respectively.

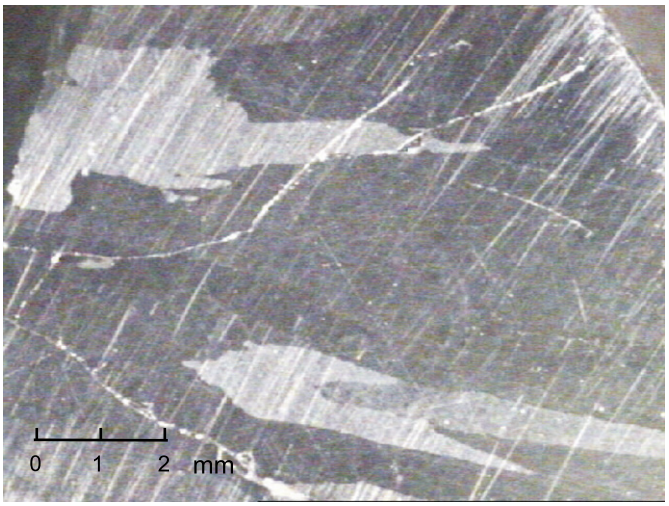

Fig. 2. Photograph of the surface of the $MnSi_f$ polycrystal.

six samples with acceptable accuracy. The estimated number of crystallites inside all six samples is roughly $6 \times 100 = 600$ and the statistical error is of the order of 0.04.

4. Concluding remarks

In this work we measured the spin helix chirality of six different MnSi polycrystals obtained after the Czochralski single crystal growth. The average chirality of these polycrystals deviates unexpectedly high from zero. This net chirality maybe related to the yet poor statistics in the number of the left or right crystallites inside the polycrystals.

Acknowledgments

The PNPI-team acknowledges GKSS for their hospitality. The work is partly supported by the RFBR project No. 10-02-00282-a and Goskontrakt No. 02.740.11.0874. V.A.D. acknowledges the Organizing Committee of PNCMI2010 and the RFBR (Project no. 10-02-09464-mob_z) for the given opportunity to participate in the Conference.

References

[1] M. Tanaka, H. Takayoshi, M. Ishida, Y. Endoh, Journal of the Physical Society of Japan 54 (1985) 2970.
[2] M. Ishida, Y. Endoh, S. Mitsuda, Y. Ishikawa, M. Tanaka, Journal of the Physical Society of Japan 54 (1985) 2975.
[3] Y. Ishikawa, K. Tajima, D. Bloch, M. Roth, Solid State Communications 19 (1976) 525.

[4] P. Bák, M.H. Jensen, Journal of Physics C Solid State Physics 13 (1980) L881.
[5] J. Beille, J. Voiron, F. Towfiq, M. Roth, Z.Y. Zhang, Journal of Physics F: Metal Physics 11 (1981) 2153.
[6] G. Shirane, R. Cowley, C. Majkrzak, J.B. Sokoloff, B. Pagonis, C.H. Perry, Y. Ishikawa, Physical Review B 28 (1983) 6251.
[7] S.V. Grigoriev, D. Chernyshov, V.A. Dyadkin, V. Dmitriev, S.V. Maleyev, E.V. Moskvin, D. Menzel, J. Schoenes, H. Eckerlebe, Physical Review Letters 102 (2009) 037204.
[8] S.V. Grigoriev, D. Chernyshov, V.A. Dyadkin, V. Dmitriev, E.V. Moskvin, D. Lamago, T. Wolf, D. Menzel, J. Schoenes, S.V. Maleyev, H. Eckerlebe, Physical Review B 81 (2010) 012408.
[9] S. Kasap, P. Capper, Handbook of Electronic and Photonic Materials, Springer, 2006.
[10] S.V. Maleyev, Physical Review Letters 75 (1995) 4682.
[11] S.V. Grigoriev, S.V. Maleyev, A.I. Okorokov, Y.O. Chetverikov, P. Böni, R. Georgii, D. Lamago, H. Eckerlebe, K. Pranzas, Physical Review B 74 (2006) 214414.