Experimental report

03/09/2018

Proposal: **5-53-283** **Council:** 4/2018

Title: Impact of Dzyaloshinski-Moriya interaction on the spin-polarized SANScross section of defect-rich materials

Research area: Physics

This proposal is a new proposal

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Samples: Tb

Instrument	Requested days	Allocated days	From	To
D33	6	5	07/06/2018	12/06/2018

Abstract:

We will study the impact of the Dzyaloshinski-Moriya interaction (DMI) on the spin-polarized SANS cross section of defect-rich polycrystalline ferromagnets (nanocrystalline terbium). A recent theoretical study has prediced a magnetic-field-dependent asymmetry in the SANS cross section. The outcome of this experiment will contribute to our fundamental understanding of polarized magnetic SANS and one will learn on the defect-related DMI mechanism.

Impact of Dzyaloshinskii-Moriya interaction on the spin-polarized SANS cross section of defect-rich materials

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Based on a recent theoretical prediction using the continuum theory of micromagnetics we show experimentally that the microstructural-defect-induced Dzyaloshinskii-Moriya interaction (DMI) gives rise to a polarization-dependent asymmetric term in the small-angle neutron scattering (SANS) cross section. This effect—conjectured already by Arrott in 1963—is demonstrated for *nanocrystalline* terbium (large grain-boundary density). Analysis of the scattering asymmetry allows one to determine the defect-induced DMI constant, $D = 0.40 \pm 0.07\,\mathrm{mJ/m^2}$ for Tb at 100 K. To the best of our knowledge, our study proves for the first time the *generic* relevance of the DMI for the magnetic microstructure of polycrystalline bulk ferromagnets, with the technique of polarized SANS playing the pivotal role for disclosing its signature. Moreover, the results open up the way to study defect-induced skyrmionic magnetization textures in disordered materials.

The Dzyaloshinskii-Moriya interaction (DMI) [1, 2] has recently moved again into the focus of condensed-matter research in the context of the discovery of skyrmion lattices in a variety of magnetic materials (see, e.g., Ref. 3 and references therein). The origin of the DMI is related to relativistic spin-orbit coupling and in inversion asymmetric crystal-field environments it gives rise to antisymmetric magnetic interactions. In the above mentioned references, the DMI is the essential ingredient for the stabilization of various types of skyrmion textures [4–7], and its origin is indeed related to the noncentrosymmetric crystal structures of the materials under study.

However, it has been conjectured for a long time that lattice imperfections in the microstructure of polycrystalline bulk magnetic materials are accompanied by the presence of local chiral DM couplings due to the breaking of inversion symmetry at defect sites [8]. Arrott suggested that the DMI is present in the vicinity of any lattice imperfection and that it gives rise to inhomogeneous magnetization states. In a sense, microstructural defects are supposed to act as a source of additional local chiral interactions, similar to the above mentioned *intrinsic* DMI in noncentrosymmetric crystal structures. It is important to realize that defect-induced DMI is generally operative in polycrystalline materials, even in high-symmetry lattices, where the "usual" intrinsic DMI term vanishes. This point of view has already been adopted by Fedorov *et al.* [9], who studied the impact of torsional-strain-induced DM couplings near dislocations on the helix domain populations in Ho single crystals. Similarly, Grigoriev *et al.* [10] investigated the field-induced chirality in the helix structure of Dy/Y multilayer films and provided evidence for interface-induced DMI. Butenko and Rößler [11] have developed a micromagnetic model for dislocation-induced DMI couplings. These authors considered a disk-like film element with a screw dislocation at its center and showed that the defect-induced DMI leads to a chirality selection of the vortex state.

Supported by theory [12] we provide in this investigation experimental evidence for the *generic* impact of the defect-induced DMI on the spin structure of polycrystalline defect-rich ferromagnets. Examples for such systems are nanomagnets, which are characterized by a large volume fraction of internal interfaces (e.g., grain boundaries), or mechanically-deformed metals containing a large density of dislocations. In the vicinity of both types of lattice imperfections—interfaces and dislocations—inversion symmetry is likely to be broken, so that the DMI may be operative. Defect-related DMI is therefore expected to manifest in other measurements as well (e.g., magnetization data), however, the technique of polarized small-angle neutron scattering (SANS) is probably the only one which is able to disclose its signature: this is brought about by the unique polarization dependence of the SANS cross section.

Based on micromagnetic theory using the DMI energy of *cubic* symmetry (e.g., [7]),

$$E_{\mathrm{DMI}} = \frac{D}{M_s^2} \int_V \mathbf{M} \cdot (\nabla \times \mathbf{M}) dV, \qquad (1)$$

we have theoretically investigated in Ref. 12 the impact of the DMI on the magnetic SANS cross section. In addition to the above DMI energy, we have taken into account the energies due to the isotropic exchange interaction, magnetic anisotropy, and the magnetostatic interaction. The central prediction is that the difference $\Delta\Sigma$ between the polarized "spin-up" and "spin-down" SANS cross sections depends on the so-called chiral function $2i\chi(\mathbf{q})$, which contains the lattice-defect-induced effect related to the DMI:

$$\Delta\Sigma \propto 2i\chi(\mathbf{q}) = -\frac{2\widetilde{H}_p^2 p^3 \left(2 + p\sin^2\theta\right) l_D q \cos^3\theta + 4\widetilde{M}_z^2 p(1+p)^2 l_D q \sin^2\theta\cos\theta}{\left(1 + p\sin^2\theta - p^2 l_D^2 q^2\cos^2\theta\right)^2}, \quad (2)$$

where $\widetilde{H}_p^2(q\xi_H)$ denotes the anisotropy-field Fourier coefficient, and $\widetilde{M}_z^2(q\xi_M)$ is the Fourier coefficient of the longitudinal magnetization. These functions characterize the strength and spatial structure of, respectively, the magnetic anisotropy field (with correlation length ξ_H) and of the *local* saturation magnetization $M_s(\mathbf{r})$ (with correlation length ξ_M); $p = p(q, H_i) = M_s/[H_i(1 + l_H^2 q^2)]$ is a known function of q and of the internal magnetic field $H_i = H_0 - NM_s$ (N: demagnetizing factor); $l_H(H_i) = \sqrt{2A/(\mu_0 M_s H_i)}$ and $l_D = 2D/(\mu_0 M_s^2)$ represent micromagnetic length scales which characterize, respectively, the size of inhomogeneously magnetized regions around defects and the range of the DMI (A: exchange-stiffness constant; D: DMI constant). The function $2i\chi(\mathbf{q})$ is asymmetric in $\mathbf{q}$, which is due to the defect-induced DMI: at small fields, when the anisotropy term $\propto \widetilde{H}_p^2$ in the enumerator of Eq. (2) dominates, two extrema parallel and antiparallel to the field axis are observed, whereas at larger fields, when the magnetostatic term $\propto \widetilde{M}_z^2$ comes into play, additional maxima and minima appear approximately along the detector diagonals.

Polarized SANS experiments on nanocrystalline Tb were carried out at the instrument D33 at the Institut Laue-Langevin, Grenoble, France [13]. We used polarized incident neutrons with a mean wavelength of $\lambda = 6$ Å and a wavelength broadening of $\Delta\lambda/\lambda = 10\,\%$ (FWHM). The external magnetic field $\mathbf{H}_0$ was provided by a cryomagnet and applied perpendicular to the wave vector $\mathbf{k}_0$ of the incident neutron beam. The beam was polarized by means of a remanent FeSi supermirror transmission polarizer ($m = 3.6$), and a radio-frequency (rf) spin flipper allowed us to reverse the initial neutron polarization. The flipping efficiency of the rf flipper was $\epsilon = 99.8\,\%$ and the polarization of the beam was $P = 95.2\,\%$.

Evaluating Eq. (2) along the horizontal direction ($\theta = 0°$) and assuming that $\widetilde{H}_p^2$ depends only on the magnitude of $\mathbf{q}$, we have (independent of $\widetilde{M}_z^2$)

$$2i\chi(q, H_i) = -\frac{4\widetilde{H}_p^2 p^3 l_D q}{\left(1 - p^2 l_D^2 q^2\right)^2}, \quad (3)$$

which can be used to analyze experimental data. Note that $\chi(q) = 0$ for $\theta = 90°$, which allows its clear separation from the nuclear-magnetic interference term $\propto \sin^2\theta$ (provided that both $\widetilde{N}$ and $\widetilde{M}_z$ are isotropic). In the analysis below, we have assumed that the anisotropy-field Fourier coefficient is described by a squared Lorentzian, $\widetilde{H}_p^2(q\xi_H) = \langle H_p^2\rangle / \left(1 + \xi_H^2 q^2\right)^2$, where $\langle H_p^2\rangle$ is the mean-square anisotropy field, and ξ_H denotes the correlation length of the anisotropy field. For an idealized nanocrystalline ferromagnet, where each grain is a single crystal and the anisotropy field jumps randomly in direction at grain boundaries due to the changing set of crystallographic easy axes, the correlation length ξ_H is expected to be related to the average grain size.

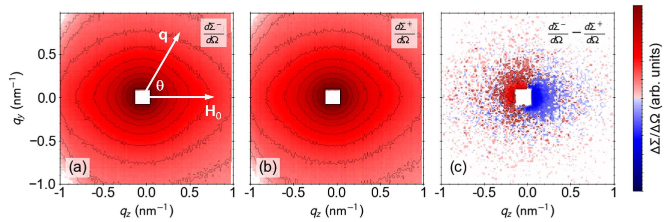

FIG. 1. Polarized SANS results on nanocrystalline Tb at 100 K and 5 T. (a) Two-dimensional flipper-on SANS cross section $d\Sigma^-/d\Omega$; (b) flipper-off SANS cross section $d\Sigma^+/d\Omega$; (c) $\Delta\Sigma = d\Sigma^-/d\Omega - d\Sigma^+/d\Omega \propto 2i\chi(\mathbf{q})$. $\mathbf{H}_0$ is horizontal in the plane. The difference signal amounts to $\sim 8.5\,\%$ of the $d\Sigma^\pm/d\Omega$.

Figure 1 depicts the results of half-polarized SANS experiments on nanocrystalline Tb at $T = 100$ K and at an applied magnetic field of $\mu_0 H_0 = 5$ T. Both spin-resolved data sets $d\Sigma^-/d\Omega$ [Fig. 1(a)] and $d\Sigma^+/d\Omega$ [Fig. 1(b)] are characterized by a maximum of the scattering intensity along the horizontal applied-field direction. This is the signature of spin-misalignment scattering due to the presence of transversal magnetization components. The difference between the two SANSPOL cross sections, $\Delta\Sigma = d\Sigma^-/d\Omega - d\Sigma^+/d\Omega$ [Fig. 1(c)], clearly exhibits an *asymmetric* contribution related to the chiral function. The asymmetry is most pronounced along the horizontal direction ($\theta = 0°$), which by comparison to the theoretical prediction [Eq. (2)] can be attributed to the anisotropy-field term $\propto \widetilde{H}_p^2\cos^3\theta$. Data taken at smaller momentum transfers additionally show the "usual" *symmetric* $\sin^2\theta$-type anisotropy (with maxima at $\theta = 90°$ and $\theta = 270°$), which is due to the polarization-dependent nuclear-magnetic interference term in the SANS cross section.

By taking an angular average of $\Delta\Sigma$ along the horizontal direction and by carrying out a weighted non-linear least-squares fit of the resulting data to Eq. (3) (see Fig. 2), the following parameters are obtained: $D = 0.40 \pm 0.07\,\mathrm{mJ/m^2}$; $A = 25 \pm 17$ pJ/m; $\xi_H = 13.6 \pm 1.3$ nm. The value for D is comparable to bulk DMI values (e.g., [14, 15]), the effective A value is also within the range of experimental data [16, 17], while the correlation length ξ_H of the anisotropy-field is smaller than the average crystallite size of the Tb sample ($L = 40$ nm). The latter finding indicates that there is significant spin disorder on a intraparticle length scale, in agreement with the results

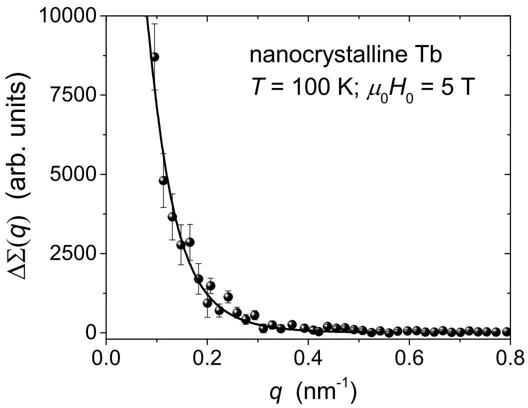

FIG. 2. (•) Azimuthally-averaged $\Delta\Sigma(q)$ of the data shown in Fig. 1(c) ($\pm 10°$ horizontal sector average). Solid line: fit to the negative of Eq. (3).

of an earlier unpolarized SANS study [17].

To summarize, using polarized small-angle neutron scattering (SANS) and micromagnetic theory we have provided evidence that the Dzyaloshinskii-Moriya interaction (DMI) which is due to the lack of inversion symmetry at microstructural-defect sites gives rise to an asymmetry in the polarized SANS cross section. The reported effect is believed to be generic to polycrystalline ferromagnets and, consequently, it should also show up in other magnetic measurements as well, for instance, in the approach-to-saturation regime of a magnetization curve. However, due to the unique dependence of the polarized neutron scattering cross section on the chiral function, the SANS method is particularly powerful for revealing the fingerprint of the defect-induced DMI. Since it is the ratio of the DMI strength D and the isotropic Heisenberg exchange coupling J which determines the wavevector magnitude of modulated structures, it is of interest to extend the present study into the antiferromagnetic helical states of the heavy rare-earth magnets (e.g., $220\,\mathrm{K} < T < 230\,\mathrm{K}$ for Tb) and to search for defect-induced skyrmion textures.

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