Field Induced Chirality in the Helix Structure of Dy/Y Multilayer Films and Experimental Evidence for Dzyaloshinskii-Moriya Interaction on the Interfaces

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Polarized neutron scattering experiments have demonstrated that Dy/Y multilayer structures possess a coherent spin helix with a preferable chirality induced by the magnetic field. The average chirality, being proportional to the difference in the left- and right-handed helix population numbers, is measured as a polarization-dependent asymmetric part of the magnetic neutron scattering. The magnetic field applied in the plane of the sample upon cooling below T_N is able to repopulate the otherwise equal population numbers for the left- and right-handed helixes. The experimental results strongly indicate that the chirality is caused by Dzyaloshinskii-Moriya interaction due to the lack of the symmetry inversion on the interfaces.

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The phenomenon of chirality is attracting much attention in a wide variety of disciplines from biology to chemistry and physics. For example, one of the authors of this Letter is left-handed, while the others are right-handed. Considering this fact, one can conclude that there must be an internal "right-handed chiral" force in the bodies of human beings pushing us to the "right" and building obstacles on the way to the "left." A similar phenomenon takes place in the physics of the helical spin structures of some crystals. The lack of a center symmetry creates the chiral force called antisymmetric Dzyaloshinskii-Moriya (DM) interaction and, as a result, one-handed helicity (chirality) [1,2]. However, the right- and left-handed helices are energetically equivalent in materials which have a crystallographic center of inversion. Such chiral degeneracy occurs in the rare-earth elements Ho and Dy, which are planar helical magnets.

The external force must be induced into these helimagnets in order to break the chiral degeneracy. For example, a nonzero average chirality was created in the single crystal of Ho by its torsion deformation [3]. Even more interesting was the experiment on $ZnCr_2Se_4$, where the average chirality of the crystal was controlled through its cooling in presence of both electric and magnetic field [4]. This experiment was explained by the construction of the lowest-order coupling between the spin and electric-field-induced polarization in cases where magnetic ions occupy the sites of the simple hexagonal lattice with planar anisotropy [5]. This magnetoelectric coupling term has the form identical to the usual DM interaction. It was demonstrated that the chiral degeneracy is removed if the electric field is applied perpendicular to the crystallographic basal-plane axis. In all cases with bulk materials the force introduced due to symmetry breaking is supposed to be weak. The situation changes in case of the broken symmetry at surfaces and interfaces. Numerous theoretical works predict the appearance of the complex magnetic configurations due to the broken symmetry in magnetic nanostructures [6,7]. The first clear experimental evidence in which the surface generates a DM interaction due to an obvious lack of inversion symmetry is given recently [8,9]. As is shown in [8], a single atomic layer of manganese on tungsten demonstrated the existence of a spiral spin structure instead of an antiferromagnetic ordering which is characteristic for Mn. This finding changes the whole concept of the magnetism in nanostructures as it introduces a new important DM interaction into the consideration of all nano-objects including the multilayer structures.

In this Letter we demonstrate that degeneracy of the helix structure due to the RKKY interaction is cancelled by the DM interaction caused by the broken symmetry at the interfaces of the Dy/Y multilayer structure. The magnetic field applied upon cooling in the plane of the sample plays the key role in coupling of the spin spiral with the DM vector displaced in the sample plane. Polarized neutron diffraction was used to define the chirality induced by external forces in [3,4] as well as in the present Letter. As it is known the pure magnetic elastic cross section of the polarized neutrons consists of polarization-independent and polarization-dependent parts [10]. The latter part is also asymmetric with respect to the momentum transfer **Q** and associated to the average chirality of the magnetic system. It can be determined by one of the following methods: by measuring the difference between the scattering intensities taken from the incident neutron beam with the polarization along (up) and opposite to (down) the guiding magnetic field at the fixed position in the momentum space **Q**, or by measuring the difference between the scattering intensities at two distinct Bragg points **Q** and −**Q** with a fixed incident polarization. Two factors have to be accounted for in the measurements: the average chirality $\langle C\rangle$ and the difference $(n_L - n_R)$ between the left- and right-handed domains.

The present study was motivated by the observation of the broken chiral degeneracy in the Dy/Y multilayer structure [11]. As was shown in [11] the spiral structure possess nonequal chiral domain population numbers for left- and right-handed domains. This difference could stem from the various sources. The first one is the stress in a multilayer structure which leads to a skew-type deformation of the superlattice. It is known that such a deformation may create nonequal domain population [3]. The second one is the random statistical distribution of right- and left-handed domains, so that $\Delta n = n_L - n_R \sim (n_L^2 + n_R^2)^{1/2}$. The third one is an internal interaction of the DM type similar to that appearing in [4,5]. The present experiment was aimed at establishing the origin of the force stabilizing the system with nonequal spiral domain population.

The sample is a superlattice of the layer sequence $\mathrm{Y}_{50}[\mathrm{Dy}_{4.3\,\mathrm{nm}}/\mathrm{Y}_{2.8\,\mathrm{nm}}]_{350}/\mathrm{Y}_{234\,\mathrm{nm}}/\mathrm{Nb}_{200\,\mathrm{nm}}\mathrm{Al}_2\mathrm{O}_3$ (substrate) produced by molecular-beam-epitaxy techniques, the same as used in [11]. The superlattice was grown along the c axis [001] of the Dy and Y hcp structure. The average lattice parameter along the c axis is 2.845 Å, the coherent length is about 500 Å, and the mosaic spread is 0.41°. The sample has a slight misorientation of 5° between the normal to the layer surface and its crystallographic c axis. A coherent helical phase extended over many bilayers is observed in the range from $T_N = 166$ K to $T = 0$ K [12]. As shown in [13], the interlayer interaction has the RKKY origin, the same as the interaction between Dy atoms inside the layer. Thus, similar to the helical structure of the bulk Dy, the Dy/Y superstructure orders in the coherent helical phase propagating through the paramagnetic Y layers due to the scalar oscillating RKKY interaction.

The polarized neutron experiment was carried out at the NeRo reflectometer at GKSS. A neutron beam with the polarization $P_0 = 0.96$, the wavelength $\lambda = 0.435$ nm, and $\Delta\lambda/\lambda = 0.02$ was used. The c axis of the multilayer sample was set almost perpendicular to the incident beam (Fig. 1). The sense of the polarization followed the guide magnetic field (of 1 mT) applied perpendicularly to the multilayer surface (along the c axis). This geometry was used to study the polarization-dependent part of the scattering. An additional field (up to 900 mT) could be applied vertically and parallel to multilayer surface.

The methodology of the experiment for studying the chirality of the magnetic system is described in [14], where it was shown that the neutron elastic cross section on the magnetic helix has the following form:

$$\frac{d\sigma}{d\Omega} \sim S^2(1 + (\hat{\mathbf{q}}\,\hat{\mathbf{c}})^2) + 2\langle C\rangle(\hat{\mathbf{q}}\mathbf{P}_0)(\hat{\mathbf{q}}\,\hat{\mathbf{c}})(n_L - n_R), \quad (1)$$

with $\hat{c}$ the unit vector of the chirality $\mathbf{C} = [\mathbf{S}_1 \times \mathbf{S}_2]$ and $\hat{\mathbf{q}} = \mathbf{q}/q$. n_L, n_R are the population numbers of the left- and right-handed helices, respectively. From Eq. (1), one can extract *polarization-independent* part, namely, $\Sigma\sigma(\mathbf{q}) = \sigma(\mathbf{q}, \mathbf{P}_0) + \sigma(\mathbf{q}, -\mathbf{P}_0) \sim S^2$, and *polarization-dependent* part, namely, $\Delta\sigma(\mathbf{q}) = \sigma(\mathbf{q}, \mathbf{P}_0) - \sigma(\mathbf{q}, -\mathbf{P}_0) \sim \langle C\rangle(\hat{\mathbf{q}}\mathbf{P}_0)(n_L - n_R)$. For convenient comparison, we normalize $\Delta\sigma(\mathbf{q})$ by $\Sigma\sigma(\mathbf{q})$ and introduce the chiral parameter γ of the scattering at $\mathbf{q} = \mathbf{k} \parallel \mathbf{c}$ as

$$\gamma = \frac{\Delta\sigma(\mathbf{q})}{\Sigma\sigma(\mathbf{q})} \sim \frac{\langle C\rangle(\hat{\mathbf{q}}\mathbf{P}_0)(n_L - n_R)}{S^2}. \quad (2)$$

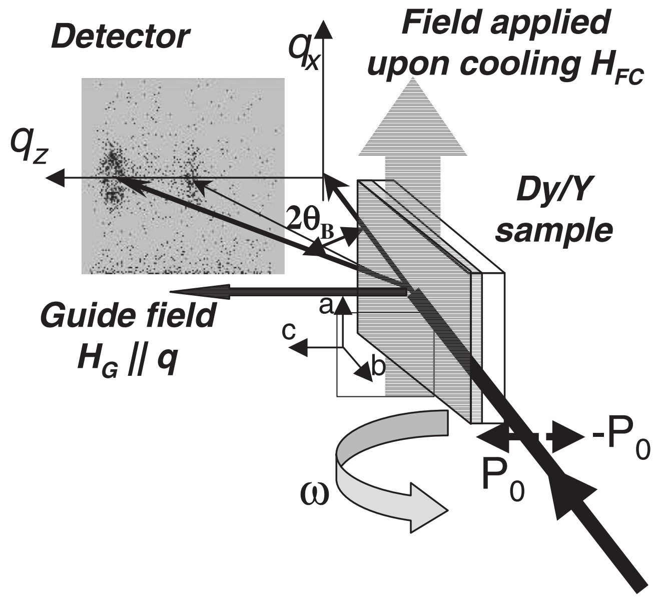

FIG. 1. Schematic drawing of the experiment.

The circularly polarized x rays could also be used to probe the chirality of the sample by exploiting the magnetic circular dichroism effect. Polarized neutrons used in the above-described experimental setup, however, are better suited here due to their direct polarization dependency on the chirality. Moreover, they allow one to probe all 350 bilayers of the sample due to their very low absorption cross section with the sample material.

The typical reflectivity curve from the Dy/Y superlattice taken below T_N is shown in Fig. 2. Closest to $\theta = 0$ is the nuclear peak with $q_N = 0.89\ \mathrm{nm}^{-1}$ (NP) originated from the multilayer structure with the period equaled to the thickness of the bilayer $d = d_{\mathrm{Dy}} + d_{\mathrm{Y}} = 7.1$ nm. Neither the position of this peak nor its intensity change with temperature, giving evidence of its nuclear (nonmagnetic) nature. The peaks at higher q values, denoted as M0 and M1, respectively, and positioned at $q_{\mathrm{M1}} = 1.64\ \mathrm{nm}^{-1}$ and $q_{\mathrm{M0}} = 2.54\ \mathrm{nm}^{-1}$, originate from the scattering on the spin helix and appear below 166 K. Coexistence of two magnetic peaks is a consequence of the modulation imposed by the nuclear multilayer structure on the spiral structure. In a first approximation the M1 peak can be identified as a nuclear satellite of the magnetic one M0 since its position is $q_{\mathrm{M1}} = 1.64\ \mathrm{nm}^{-1} \simeq (q_{\mathrm{M0}} - q_N) = 1.65\ \mathrm{nm}^{-1}$. The presence of well-separated magnetic peaks can also be interpreted in terms of the long-range correlations of the helix structure covering several bilayers as it was considered in [13].

The intensity of the magnetic peak M0 (see inset of Fig. 2) increases with decreasing temperature below T_N.

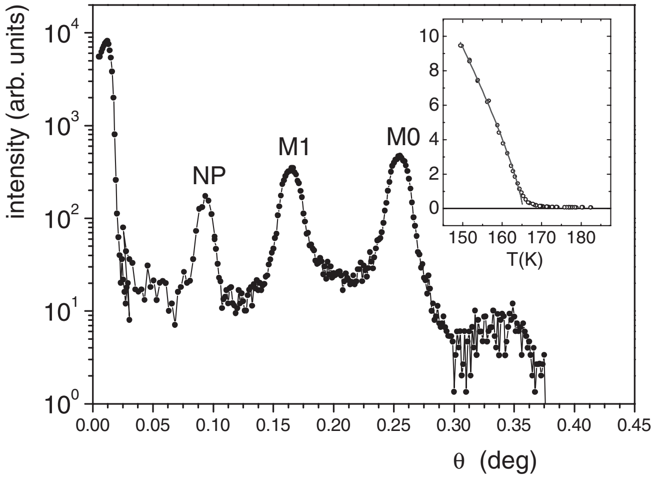

FIG. 2. The q dependence of the neutron scattering intensity at $T = 150$ K. The peaks NP, M1, and M0 are the nuclear and two magnetic reflexes, respectively. The inset shows the temperature dependence of integral intensity for peak M0.

The data obey the scaling law $\Sigma I = I_0 \tau^{2\beta}$ in the range $\tau < 0.1$, where $\tau = (T_N - T)/T_N$ and β is the critical exponent for the magnetization. The values of $T_N = 165.3 \pm 0.2$ and $\beta = 0.40 \pm 0.01$ is in good agreement with that of bulk Dy [15] and previous experiment [11].

For our experiment ($\mathbf{P}_0 \parallel \mathbf{k}$) and in accord with Eq. (1) the scattering intensity with a certain polarization is related to one of two types of the helix domains only: left- or right-handed ones. The rocking scans of the peak M0, shown in Fig. 3, gives an example of the nonzero difference between two intensities of the opposite polarizations $I(+P_0)$ and $I(-P_0)$. The scans demonstrate the existence of the nonzero average chirality in the sample, i.e., the nonequal population of the left- and right-handed domains. Similar scans taken for the nuclear peak NP show no difference in $I(+P_0)$ and $I(-P_0)$.

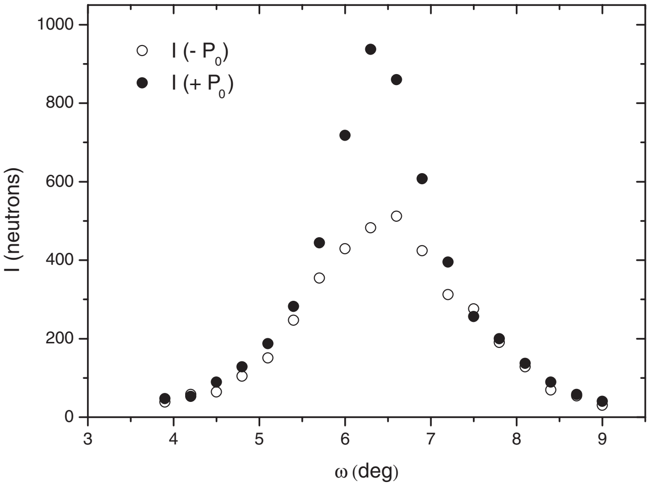

FIG. 3. The rocking scans $I(\omega)$ of the peak M0 (after FC to 150 K at $H = 900$ mT) taken for two opposite polarizations at $T = 150$ K and $H = 1$ mT ($\mathbf{H} \parallel \mathbf{P}_0 \parallel \mathbf{q}$).

The chiral parameter [Eq. (2)] shown in Fig. 4 was calculated as $\gamma = [I(+P_0) - I(-P_0)]/[I(+P_0) - I(-P_0)]$ and normalized to the polarization P_0 of the experimental setup. For zero field cooling (ZFC, solid symbols), γ is negative and decreases from zero above $T = T_N$ to $\gamma = -0.1$ at low temperatures. Fitting of γ to the scaling function of τ gives the value $\beta_C - 2\beta = 0.22$, in good agreement with the previous experiment [11]. It should also be noticed that the thermocycling (cooling up and down through the phase transition in zero field) has no effect on the value of γ, giving the same result within the error bars. This excludes the hypothesis of a random distribution of the chiral domains and strongly suggests the existence of an internal force resulting in the appearance of the average chirality in this system.

A major finding of the present study is the fact that γ depends on the temperature or magnetic-field prehistory. The field up to 1 T applied parallel to the c axis does not significantly change the helix structure or the value of γ. On the contrary, the field of the order of 1 T applied in the plane (ab) tends to destroy the helix transforming it to the ferromagnetic state. The intensity of the peak M0 decreases to 75% of the initial value upon the increase of the field up to 900 mT. The consequent decrease of the field restores the initial intensity as well as the helix structure. The important feature of this experiment is that no chirality could be measured when the field is applied in (ab) plane and $\mathbf{P} \parallel \mathbf{H} \perp \mathbf{k}$ [see Eq. (1)]. Therefore, in order to study the effect of the in-plane field on the chirality, the sample was cooled first from $T > T_N$ to $T < T_N$ in the in-plane field, and then the in-plane field was switched off and the γ

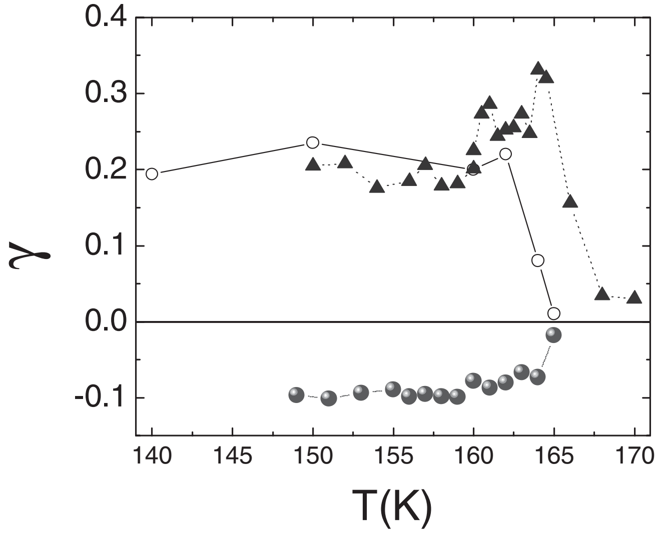

FIG. 4. The temperature dependence of the chiral parameter γ for different temperature or magnetic-field prehistory of the sample (closed circles, ZFC; open circles, FC in $H = 900$ mT to the temperature of interest T; triangles, ZF warming after FC in $H = 900$ mT to $T = 150$ K).

value was measured in the small guide field ($\mathbf{H}_g \parallel \mathbf{P}_0 \parallel \mathbf{k}$). As shown in Fig. 4, such a field cooling (FC) procedure at $H = 900$ mT gives the positive chirality ($\gamma = +0.2$), opposite to the ZFC experiment. Even more intriguing behavior of γ is observed for the measurements after FC ($H = 900$ mT) to 150 K with the subsequent warming up in zero field (Fig. 4, triangles). In this case the chiral parameter γ stays constant in the range 150–160 K and then increases as temperature approaches T_N. It has a maximum at T_N and then decreases to zero in the paramagnetic range. One can conclude that the appearance of the chirality is strongly related to the critical temperature range where the softening of the magnetic structure allows the weak chiral force to induce the nonequal population of the left- and right-handed domains.

Furthermore, γ depends on the strength of the field applied in the sample plane (ab) in the FC procedure but does not depend on the direction ($\pm\mathbf{H}_{\mathrm{FC}}$) of the field. As shown in Fig. 5, γ increases in its absolute value as the field increases. The similar measurements, conducted for two other Dy/Y multilayer samples, qualitatively revealed the same dependence of the increasing chirality on the magnetic field. This implies that the field-induced chirality is genetic for the Dy/Y multilayer structures.

The here-described phenomena can be interpreted in terms of the DM type of interaction, which is expected to appear on the magnetic-nonmagnetic interfaces of the multilayer structures similar to that observed in [8,9]. It is important that the DM vector is oriented in the interface's plane, i.e., perpendicular to the helix wave vector **k**, and therefore does not induce the chirality into the system. However, first, the crystallographic c axis along with **k** is inclined to the normal to the interfaces. Second, the dispersion of **k** direction, related to the out-of-plane deviation of spins, rises upon application of the magnetic field. Thus, the chirality can be switched on in the sample upon FC procedure. A similar phenomenon of switching the DM interaction on and off can be found in the classical α-Fe_2O_3 at the Morina point $T_M = 250$ K. The anisotropy axis changes at $T = T_M$ orienting antiferromagnetically ordered spins parallel to the DM vector at $T < T_M$ and perpendicular to the DM vector at $T > T_M$. The spin structure with ($S \parallel$ DM) remains collinear and hides the presence of DM interaction while the one with ($S \perp$ DM) is distorted by DM interaction resulting in a weak ferromagnetic component of the spins.

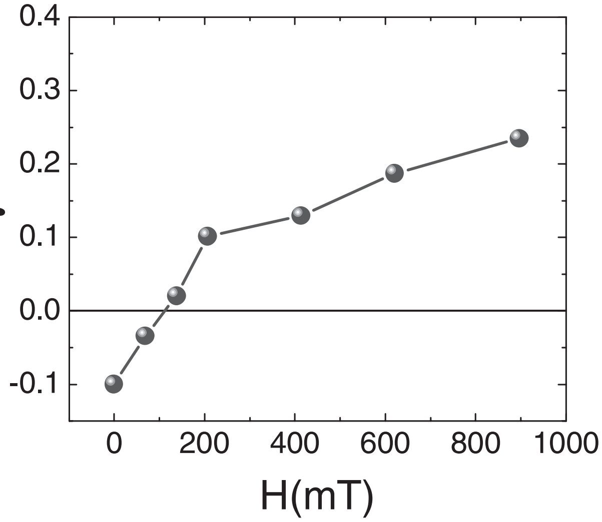

FIG. 5. The chiral parameter γ as a function of the magnetic field H applied in the FC procedure to $T = 150$ K.

In conclusion, we give the experimental evidence for Dzialoshinskii-Moriya interaction on the interfaces of the Dy/Y multilayer structure. The DM interaction reveals itself via nonzero average chirality measured by the method of polarized neutron diffraction. The application of magnetic fields plays a key role in coupling of the existing spiral spin structure and the DM interaction lying in the sample's plane. This experiment demonstrates that the spin and chiral components of the order parameter can be decoupled with the help of the magnetic field as a tool for manipulating these parameters.

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