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Field induced chirality in helix structure of Ho/Y multilayers

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We study the net chirality in the spin helix structure of Ho/Y multilayers induced by an in-plane applied magnetic field. The chiral symmetry breaking was revealed by means of polarized neutron reflectometry. The three samples of different thicknesses of Ho and Y layers were grown by the molecular-beam epitaxy method. No break of the chiral symmetry is found upon zero field cooling below the critical temperature $T_N = 115 \pm 3$K. The chirality parameter γ rises up upon field cooling procedure in the field range from 0 to 1 T and saturates at a value of 0.12 ± 0.01. The chirality appears stepwise below T_N and depends weakly on temperature. The phenomenon is interpreted in terms of the Dzyaloshinskii-Moriya interaction appeared at the interface between Ho and Y layers.

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The rare-earth magnetism attracted much attention in the light of the discovery of 3D long range order, which can occur in rare-earth/yttrium superlattice (SL) structures [1–6]. Superlattices of Dy/Y and Ho/Y show a helical order in which magnetic moments are aligned in ferromagnetic sheets within each basal plane, but the orientation of these moments changes from one plane to another one, thus forming a spin helix. The long-range coherence of the magnetic structure arises from the conduction electrons propagating coherently throughout the SL. This coherent propagation from the yttrium layers into the magnetic layers maintains the stability of the turn angle and the chirality of the helix.

The new impulse for the investigations of the rare earth superlattice was stimulated by the observation of effects caused by magnetic interfaces, such as, the enhanced interfacial magnetic order [7], the twisted magnetization states [8] and the surface-induced Dzyaloshinskii-Moriya interaction [9–11]. A few years ago Grigoriev *et al.* have demonstrated that Dy/Y magnetic multilayers (MMLs) possess a coherent spin helix with a preferable chirality induced by a magnetic field [10]. It was shown that a 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. It was suggested that the interplay of the Ruderman-Kittel-Kasuya-Yoshida (RKKY) and Zeeman interactions helps to reveal the otherwise hidden antisymmetric Dzyaloshinskii-Moriya interaction (DMI). It was argued that the observed chirality is a fingerprint of the DMI resulting from the lack of the symmetry inversion at the interfaces [12].

One can suggest that the same effect of an applied magnetic field on the chirality of the helix spin structure can occur in the MMLs made of other Rare-Earth elements such as Ho. The magnetic structure of bulk Ho was investigated using neutron scattering by Koehler *et al.* [13]. Below the Neel temperature (T_N=132.2 K) the magnetic system of the hexagonal close packing structure of Ho orders in the spin helix. Similar to the Dy, the moments in Ho are ferromagnetically coupled within the basal (*ab*)-plane, but their orientation rotates at a certain angle while moving along the *c*-axis. It was, however, shown that, contrary to the Dy, the magnetic order in bulk Ho is strongly affected by the crystal-field anisotropy [6, 14, 15]. The magnetic moments is bunching along the six easy axes in the basal plane due to the crystal-field anisotropy, what leads, firstly, to the lock-in of the wave vector into values commensurable with the atomic lattice in certain temperature intervals, and, secondly, to the formation of a series of long period commensurate spin-slip structures [6, 14, 15]. Particularly, the moments are ordered below 18 K in a commensurate cone structure with the wavevector **k** along *c*-axis, forming a twelve-layer magnetic unit cell.

As it was shown in [6] the magnetic structure of Ho/Y multilayers is similar to that of bulk Ho. The coherent spin helix penetrates through the paramagnetic Y layers due to the charge density wave of the conduction electrons [1]. The effective turn angle in Y is found to be constant (about 51^o) at all temperatures, while the turn angle in Ho layers was larger in comparison with bulk Ho. In addition, the ferromagnetic transition at 18 K is suppressed in multilayers. The strains introduced by the lattice mismatch between Ho and Y produce a lattice pressure which reduces the ordering temperature inside the Ho blocks. The corresponding lattice parameter in the bulk is equal to $c = 2.808$ Å for Ho and $c = 2.865$ Å for Y, respectively.

In this Rapid Communication we show that the chiral symmetry of the helix structure can be broken by an in-plane magnetic field applied upon cooling of

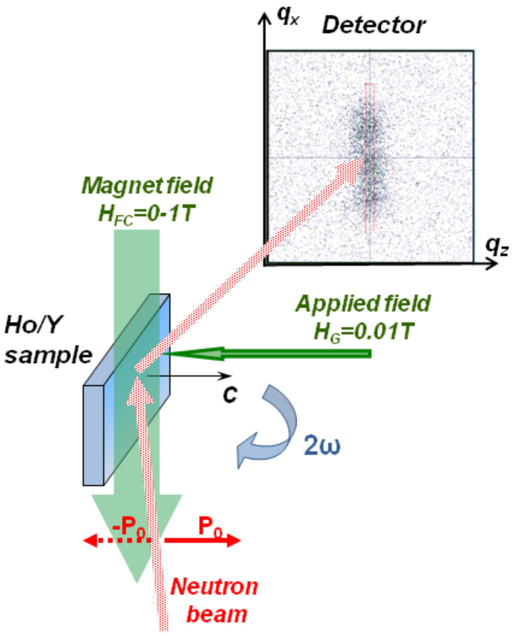

FIG. 1: The schematic drawing of the experiment.

holmium/yttrium multilayers. The effect of an applied magnetic field was studied using the following three samples: $[\mathrm{Ho}45\text{Å}/\mathrm{Y}30\text{Å}]_n$, $[\mathrm{Ho}25\text{Å}/\mathrm{Y}20\text{Å}]_n$ and $[\mathrm{Ho}20\text{Å}/\mathrm{Y}30\text{Å}]_n$ denoted as (Ho45Y30), (Ho25Y20) and (Ho20Y30), respectively. The number of bilayers n is 20 in (Ho45Y30) and (Ho25Y20), and 30 for (Ho20Y30). The samples were grown along the c axis [001] of the Ho and Y hcp structure by molecular-beam-epitaxy techniques at the Uppsala University [16] on a sapphire substrate with a 150 Å Nb buffer layer and a 200 Å Y seed layer below the superlattice. The samples were capped by a 50 Å Nb layer to prevent the oxidation of the magnetic material. The good chemical and crystallographic quality of the SLs was verified by x-ray diffractometry and reflectometry using a standard x-ray diffracometer at the Helmholtz-Zentrum Geesthacht. The structural coherence lengths along the c axis are estimated from the FWHM of the central Bragg peak and are between from 450 to 650 Å with an average mosaicity of about 0.34^o. The lattice parameters along the c axis is larger for the holmium blocks and smaller for yttrium blocks compared to the individual bulk materials.

To answer the question whether any preferable chirality arises for these structures, polarized neutrons are especially useful since they allow one to determine the chirality of magnetic structures [17]. The total magnetic elastic neutron cross section for polarized neutrons can be separated into a polarization-dependent contribution and a polarization-independent part. The latter part is also asymmetric with respect to the momentum transfer Q and can be associated with the average chirality of the magnetic system.

The polarized neutron experiments were carried out at the MARIA reflectometer at the FRM II (JCNS). An incident neutron beam with the polarization $P = 0.98$, the wavelength $\lambda = 6$ Å and $\Delta\lambda/\lambda$=0.1 were used. In order to provide a perpendicular guide field in respect to the sample plane at the sample position, additional magnetic guide fields were mounted. Due to the non-trivial setup the polarization at the sample position was reduced to about $P_0 = 0.90$. The c axis of the multilayer sample was set perpendicular to the incident beam (FIG. 1). A magnetic field of up to 1 T could be applied parallel to the multilayer surface during the field cooling (FC) procedure from T>T_N to T<T_N. The reflectivity profile at the Bragg peak position of the helices were taken at different temperatures after zero field cooling (ZFC) and the FC procedures from T>T_N to T<T_N. The scattering intensity is measured in a small guide field H_G, with $(\mathbf{H}_G\|\mathbf{P}_0\|\mathbf{k})$, thereupon the in-plane field H_{FC} was switched off. The sense of the polarization followed a guide magnetic field of 1 mT applied perpendicularly to the multilayer surface (along the c axis). Such geometry was used to study the polarization-dependent part of the scattering cross section. At this configuration with the polarization of the incident beam aligned along the direction of the applied field $(\mathbf{P}_0\|\mathbf{Q})$, the corresponding scattered intensities, $[I^+ = I(\mathbf{Q}, +\mathbf{P}_0)$ and $I^- = I(\mathbf{Q}, -\mathbf{P}_0)]$, are due to scattering on either the right- or left-handed domains, respectively. The average chirality, which is proportional to the difference in the population of the left- and right-handed helices, was measured as the polarization-dependent asymmetric part of the magnetic neutron-scattering cross section [10].

Thus we introduce here a chiral parameter directly related to the measured intensities and to the imbalance between the left- and right-handed domains:

$$\gamma = \left| \frac{1}{P_0} \frac{I(+P_0) - I(-P_0)}{I(+P_0) + I(-P_0)} \right| \quad (1)$$

The measured value of γ was normalized to the polarization P_0 at the sample position.

Figure 2 shows reflectivity profiles, $I(+P_0)$ and $I(-P_0)$, for the sample Ho25Y20 after the ZFC procedure (a) and the FC procedure at $H = 1$ T cooled down to $T = 30$ K. The observed peaks are obviously originating from the incommensurate helical spin structure since they appear only below T_N and at a Q value not corresponding to the structural superlattice period or a magnetically commensurate Q-state. No difference in the scattering profiles is observed upon the ZFC procedure within the error bars [Fig.2(a)]. The FC procedure, on the other hand, show a nonzero difference between the two scattering intensities of opposite polarizations, $I(+P_0)$ and $I(-P_0)$, demonstrating the appearance of a nonzero average chirality in the sample.

The temperature dependence of the integrated magnetic peak intensity after the FC procedure is shown in Fig.3(a). We extrapolated the intensity of magnetic peak to the zero value and found that the such determined ordering temperature T_N in Ho layers of these samples is

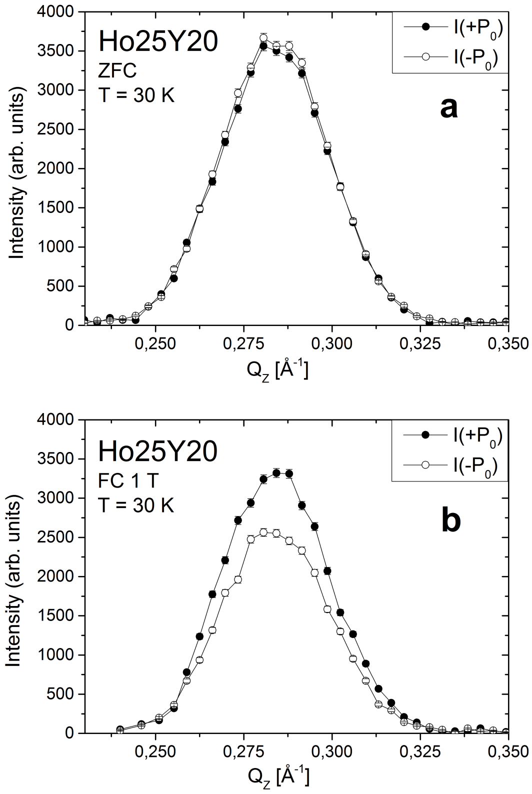

FIG. 2: The Q dependence of the neutron-scattering intensity (reflectivity profile) for the samples Ho25/Y20, taken for two polarizations of the incident beam at T = 30 K after ZFC procedure (a) and FC procedure at the applied field H=1 T (b).

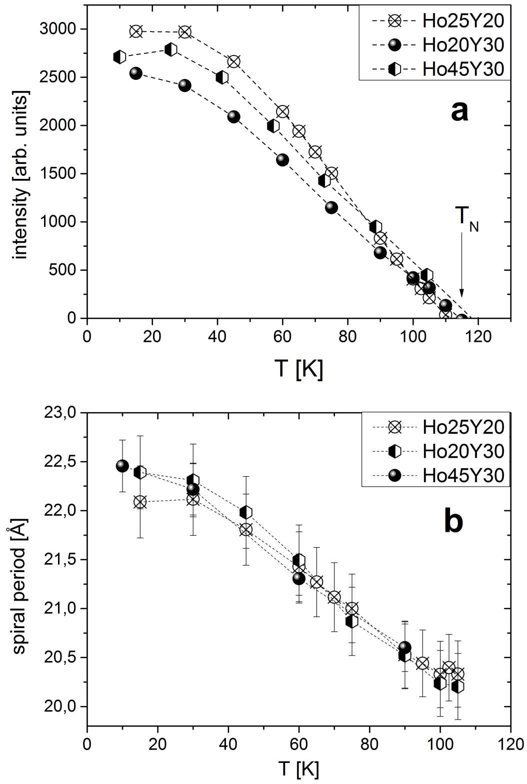

FIG. 3: Temperature dependence of magnetic intensity (a) and the spiral period in Ho layer (b).

significantly reduced in respect to the bulk material to 115 ± 3 K. It should be noted that the beforehand applied magnetic field H_{FC} does not affect the position of the magnetic peak and the spiral period does not depend on the applied field procedure. The spiral period d_s can be calculated from the peak center of magnetic reflection (Q_{cen})

$$d_s = \frac{2\pi}{Q_{cen}} \tag{2}$$

Figure 3(b) shows that the values of d_s are practically the same for the investigated samples and independent of the holmium and yttrium thicknesses of the individual sample. The spiral period d_s is equal to 22.3 ± 0.5 Å at low temperatures and decreases with increasing temperature to about 20 Å in the vicinity of T_N. As it was shown in ref.[6], the scattering data can be reasonably modeled if one assumes that the phase shift across the Y layer, associated with a wave vector $\mathbf{k}_Y$, is different from the wave vector of Ho ($\mathbf{k}_Y \neq \mathbf{k}_{Ho}$). According to [6] the value of $\mathbf{k}_Y$ of 0.31 Å^{-1} is temperature independent, corresponding to a turn angle between Y atomic planes along the c axis of 51^o and the period of helix of 20 Å. Thus we associate the changes of the spiral period d_s to changes in the phase shift across the Ho layer and to the changes of its wave vector $\mathbf{k}_{Ho}$ with the temperature.

Figure 4 shows the value of γ for the samples Ho25Y20, Ho20Y30 and Ho45Y30 in dependence of the temperature after the ZFC and FC procedure, respectively. After the ZFC procedure shown in Fig. 4(a), the chirality γ is quasi zero within the error bars over the complete temperature range for all three samples. This observation can be easily understood considering the RKKY interaction as the dominant interaction for forming the helical structures. In this case, the right- and left-handed helices are energetically equivalent to each other and both states will be occupied in equal measures. The value of chirality γ measured after the FC procedure in a magnetic field of 1 Tesla, on the other hand, show clearly non-zero values of up to 12% suggesting strongly that the chiral symmetry is now broken. The value of γ drops sharply to zero when the temperature approaches T_N indicating that the introduction of the chirality in the systems dominately occur in a very limited temperature range close to the transition temperature for all three samples. In Fig. 5 the chirality γ is plotted as a function of the applied field H_{FC} during the FC procedure with subsequent cooling down to a temperature of 30 K. The value of γ increases with the increase of the applied field in the range $H_{FC} < 0.5$ T, indicating that the strength of the field has a considerable influence on magnetic structure

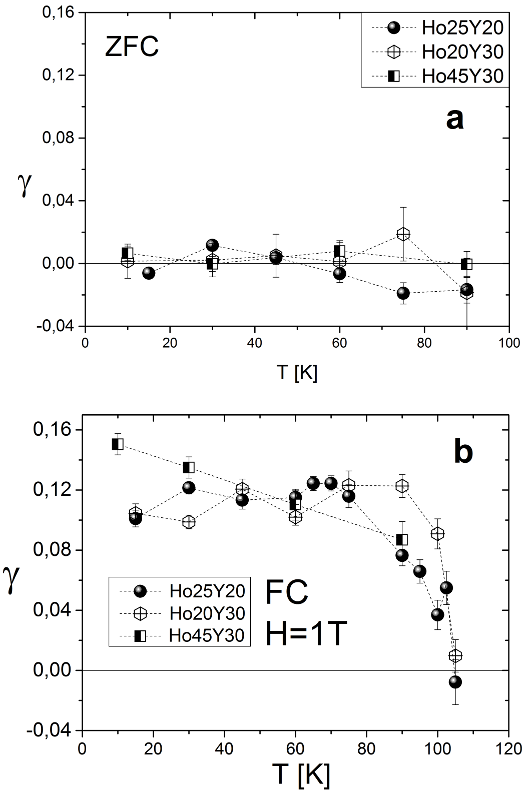

FIG. 4: The chirality for the samples Ho25Y20, Ho20Y30 and Ho45Y30 in dependence on temperature prehistory: (a) T-dependence after ZFC, (b) T- dependence after FC under applied field $H = 1$ T

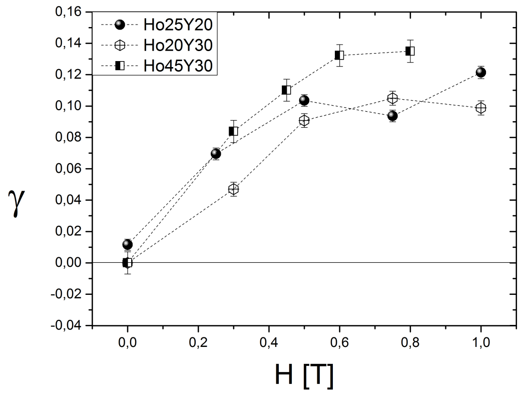

FIG. 5: The chirality for the samples Ho25Y20, Ho20Y30 and Ho45Y30 in dependence on field prehistory: H-dependence after FC to 30K.

of the samples in this regime. The chirality γ, however, saturates at a value of $0.10 - 0.12$ with no further significant increase for $0.5 < H_{FC} < 1$ T. The FC procedure clearly demonstrates the predominance for one type of the helix domains over other one induced by the in-plane magnetic field applied during the FC procedure.

As it was already mentioned above, a very similar behavior in respect of the net chirality was detected before in Dy/Y multilayers. The assumption of a breaking of the chiral symmetry in these systems due to appearance of DMI on interfaces was theoretically confirmed by Haraldsen and Fishman in 2010 [18]. They had shown that interfacial defects, emerging due to the overlap between magnetic Dy and nonmagnetic Y atoms, can produce a non-zero DM contribution normal to the interface in the magnetic heterostructures. By application of an in-plane magnetic field, particularly in the vicinity of the transition temperature when the RKKY interaction is weak and comparable to the applied Zeemann energy, the DMI can be coupled into the system and further transferred by the increasing strength of the RKKY interaction with decreasing temperature throughout the whole SL. Once the RKKY is strong enough, the imprinted chirality during the transition temperature remains unchanged.

We assume that the net chirality in Ho/Y systems has the same nature and appears due to symmetry breaking at the magnetic-non-magnetic interfaces. The x-ray characterizations clearly indicate that the interfaces of all of the examined Ho/Y SL are of good quality, but are also partly intermixed and thus fulfill the necessary conditions for the occurence of a non-zero DM energy contribution at the interfaces. On the other hand, the chirality of the investigated samples does not exceed a maximum value of about 13% what is twice as less that in Dy/Y multilayers [10, 11]. We explain this decrease by a strong influence of the crystal-field anisotropy in Ho layers. In general, we gave a new experimental evidence for Dzialoshinskii-Moriya interaction on the interfaces of the magnetic multilayer structure taking Ho/Y multilayers, for examples. Further experimental and theoretical studies can reveal the role of the crystal-field anisotropy in Ho layers as well as the role of the possible interplay the RKKY and DM interactions in these types of the magnetic multilayer systems.

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