Interlayer exchange coupling in Er|Tb superlattices mediated by short range incommensurate Er order

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Abstract. We study the magnetic correlations in Er|Tb superlattices by means of off-specular scattering of polarized neutrons. We show here the co-existence of inhomogeneous magnetic states: i) ferromagnetic order of moments within the Tb layers below 230 K (FM), correlation length of about 10 bilayer, ii) an incommensurate modulated magnetic order, restricted to single Er layers and iii) antiferromagnetic coupling of ferromagnetic layers below 70K (AFC). Polarised off-specular neutron scattering under grazing incidence reveals that i) magnetic fluctuations appear when the sample is cooled below 70 K, ii) these fluctuations lead to AFC, when the sample is cooled to 10 K, which iii) persists, when the sample is subsequently heated up to 45 K, while the order is not present during the cooling cycle. Also the short range incommensurate order changes accordingly, implying that the magnetic order in the Er layers mediates the interlayer coupling between ferromagnetic Tb layers.

1. Introduction

The discovery of magnetic interlayer exchange coupling initiated intense research activities concerning rare-earth superlattices (overview in [1,2]). While earlier studies focused on systems featuring non-magnetic spacers (e.g. consisting Y layers [3,4]) more recent work was devoted to systems containing two magnetic layers [5-11]. The latter cases reveal a variety of novel magnetic phases due to the competing anisotropies, proximity effects and epitaxial strain. In Er|Tb superlattices the competing anisotropies prevent the formation of long range incommensurate structures as found in the bulk materials, except for the thinnest Tb layer thickness ([Er₂₁|Tb₅] indices denoting the number of atomic layers) [7]. The presence of epitaxial strain stabilizes the ferromagnetic order in the Tb layers for Tb layer thicknesses larger than 10 atomic layers below $T_C = 230$ K. Wide angle neutron diffraction and x-ray resonance exchange scattering revealed an antiferromagnetic coupling (AFC) between adjacent layers in contrast to the macroscopic magnetization that has been observed by SQUID measurements [10]. Furthermore short range incommensurate magnetic correlations appear below 80 K, which seem to be restricted to single Er layers.

Here we present off-specular scattering of polarized neutrons to resolve the nature of the AFC in an [Er₂₁|Tb₁₀] superlattice containing 61 bilayers and to probe the range of the incommensurate correlations perpendicular and parallel to the surface plane. The sample was grown with the hexagonal [0001] direction parallel to the surface normal, which is also parallel to the propagation vector of the incommensurate bulk magnetic structures. The high epitaxial
quality was confirmed by low energy electron diffraction and x-ray diffraction. Details of the preparation and characterization can be found elsewhere [7].

2. Macroscopic magnetization

For the macroscopic characterization we used a SQUID magnetometer (Quantum Design MPMS) in a temperature range from 10 K to 250 K. For these measurements a small piece was cut from the sample later used for neutron scattering. Subsequent x-ray diffraction assured that the sample quality was not deteriorated. The piece was aligned with the surface normal perpendicular to the applied magnetic field. The temperature dependence of the in-plane magnetization at two different applied fields is shown in Fig. 1. The curves have been measured by cooling the sample in the respective magnetic field and subsequent heating to avoid a reduction of the measured magnetization by the formation of randomly oriented domains, when cooled in zero field. We tentatively normalize the magnetization to the number of Tb atoms within the sample, since the basal plane ferromagnetism is an original phase of bulk Tb. Hence we may compare the magnetization to the saturation value of $9.72 \mu_B$ for metallic Tb. The number of Tb atoms was determined from the area of the substrate, the number of Tb layers as known from the preparation, and the unit cell area in the epitactically strained sample. As the latter can not be quantified exactly we estimate the error on the Tb atom number to 4%.

Below $T_C \approx 230$ K the magnetization steeply increases indicating a ferromagnetic order of the Tb moments throughout all the applied field. Below 90 K the qualitative behavior is different for the 180 mT data and the 1 mT data. Upon cooling we observe a kink, that has been attributed to an additional contribution from the Er moments to the magnetization [10]. Below the maximum at 60 K the magnetization decreases significantly when the sample is cooled in an applied field $\mu_0 H = 1$ mT. For this magnetic field, we observe a clear thermal hysteresis upon heating, as the

Figure 1. (Color online) Temperature dependence of the in-plane magnetization of an Er$_{21}$Tb$_{10}$ superlattice in two different magnetic fields. Blue curve measured during cooling, red curve measured during heating. The diamagnetic contribution from sample container and substrate was corrected for. The applied magnetic fields are representative for the field conditions during the neutron scattering experiment.

Figure 2. (Color online) Schematic experimental setup for the neutron scattering. Magnetic field and neutron polarization aligned parallel to the basal plane. Magnetization parallel to this contributes to NSF channel and perpendicular to SF channel.
Figure 3. (Color online) NSF and SF maps of scattering intensity as a function of \( q_z \) in units of the superlattice periodicity \( 2\pi/D \) and \( \alpha_i - \alpha_f \), yielding the information about correlations parallel to the growth direction vertically and the lateral correlations horizontally. Color coded intensities are normalized to monitor counts. Both rows were measured at 45 K. The upper row shows intensity maps measured during the cooling cycle, the lower row was recorded upon heating after the sample had been cooled to 10 K in the respective applied magnetic field.

magnetization starts to increase only above 50 K. The heating curve crosses the cooling curve at \( \approx 80 \) K and remains slightly above the cooling curve up to \( T_C \). For larger applied fields, the thermal loop is also not completely closed. We observe an additional kink at a slightly different temperature for the cooling and the heating cycle, respectively. For comparison the macroscopic magnetization was measured at the same values of applied field and temperature as the neutron measurement. For this being only a few points this data is not displayed in Fig. 1. Nevertheless there is still a small thermal hysteresis to be observed under these conditions and the magnetization curve at 180 mT is representative.

3. Neutron Measurements
To get an insight into the magnetization of a single layer and the range of the magnetic coupling we measured the polarized off-specular neutron scattering along the specular rod at the TREFF instrument (\( \lambda = 4.73 \) Å) at FRM II (formerly HADAS at FRJ-2 [13]). For this we again applied the magnetic field along the in-plane direction and the scattering vector \( \mathbf{Q} \) was parallel to the out-of-plane direction. As the scattering vector has a large component parallel to the surface normal and only a small component within the surface plane, we probe mainly the magnetization within the surface plane, which coincides with the \( ab \) plane of the hexagonal lattice. The external magnetic field and the neutron polarization are aligned perpendicular to the scattering plane, i.e. within the superlattice surface. Magnetic moments parallel to the guide field cause non-spin-flip
scattering (NSF), while magnetic moments perpendicular to the field flip the spin (spin-flip, SF).

We have measured all 4 channels of incoming and outgoing polarization. Since the differences between the up-up and down-down states of the neutron polarization are negligible, we present only the sum of the respective intensities. The same applies for the up-down and down-up states. Due to the fact that the nuclear scattering lengths of Er and Tb are similar ($b_{E_r} = 7.8$ fm and $b_{Tb} = 7.4$ fm) the nuclear contrast between the layers almost vanishes, leaving only a small nuclear contribution to the NSF. Fig. 3 shows the off-specularly scattered intensity maps as a function of $\alpha_i - \alpha_f \approx \frac{2\pi}{9\theta}$ and $q_z$ in units of $\frac{2\pi}{D}$, with the bilayer thickness $D = 8.5$ nm. The former gives a measure of the lateral correlations on the length scale from several 1-100 $\mu$m. Parallel to $q_z$ one can probe correlations on a length scale of hundreds of nm. We show representative scans at 2 mT and at 338 mT, which is the maximum field that can be applied.

Three types of features can be distinguished in Fig. 3: i) Reflections at integer values of $q_z$ indicating FM order of Tb layers as anticipated from the SQUID measurements. They are represented by narrow Bragg sheets indicating long range vertical correlations spanning across some 10-15 bilayer. The sheets exist already at 150 K, the highest measured temperature. ii) Reflections at half integer values denoting a doubled magnetic unit cell and therefore indicating antiferromagnetic coupling of ferromagnetic layers. They appear only after the sample had been cooled to 10 K. Under small field conditions the NSF channel exhibits true specular reflectivity at these $q_z$ values, while only the Bragg sheets are visible in the SF channel. Hence the AFC is long range only for the component parallel to the external field with a correlation length $> 1.5 \mu$m. For the component perpendicular to the field we estimate the correlation length from the width of the Bragg sheet to be smaller than 500 nm. When the sample is cooled in a large field, the situation is different: The reflections at half integer values exist only in the SF channel, indicating that only the component perpendicular to the external field couples antiferromagnetically. Upon heating, these reflections still exist at $T = 45$ K. During the cooling cycle diffuse intensity is observed in the $q_z$ range $0.3 < q_z(\frac{2\pi}{D}) < 1$ at this temperature. iii) Broad diffuse intensity at $3.5 < q_z(\frac{2\pi}{D}) < 4$. For this feature the thermal history has two effects. Comparing the data measured at low field for the cooling and heating cycle, the center of the intensity is shifted towards smaller $q_z$ in the heating run. But the center and the magnitude of the intensity in the SF and NSF channel remain similar. In combination with earlier magnetic resonant x-ray scattering results [7], this intensity could be attributed to short range helix-like magnetic correlation reminiscent of the magnetic structure found in bulk Er. It has to be noted that the intensity at $q_z = 3.5\frac{2\pi}{D}$ is enhanced in the heating run in the NSF and the SF channel. The situation is different, when the sample is cooled in a strong field: upon cooling, the intensity is centered around $q_z = 4\frac{2\pi}{D}$ and it is broader in the SF than in the NSF channel. Upon heating the NSF intensity remains centered around $q_z = 4\frac{2\pi}{D}$. In the SF channel the diffuse intensity is found for $3.5 < q_z(\frac{2\pi}{D}) < 4$ with a maximum around $q_z = 3.5$. So far it is not clear, how the intensity at strong magnetic fields has to be interpreted. The experimental result indicates different Fourier components for the parallel and perpendicular components of the magnetic moments. It seems plausible that the coupling of ferromagnetic layers is connected with the short range correlations, particularly the antiferromagnetic coupling of the perpendicular component.

4. Discussion and Outlook

The macroscopic data and the neutron reflectivity show that the [Er$_{21}$|Tb$_{10}$] superlattice develops ferromagnetic order below $T_C = 230$ K. The intriguing features of the superlattice are revealed by the off-specular scattering. The surprising suppression of the macroscopic magnetization at low temperature can be linked to the appearance of magnetic fluctuations causing diffuse scattering. The temperatures at which we observe anomalies in the magnetization coincide roughly with the phase transition temperatures in bulk Er. The magnetization starts decreasing at temperatures, at which the basal plane order changes in bulk Er [12]. The off-
specular scattering reveals the fluctuations associated with the suppression of the magnetization. While upon cooling the diffuse scattering at low $q_z$ is rather unstructured, clear AFC develops between neighboring layers once the sample is cooled to base temperature. Due to the limited neutron flux we were not able to determine exactly the temperature, where the AFC becomes long range. It is intriguing to realize that the turn angle accumulated across 21 atomic layers of Er approximates $\Phi = 5\pi$ for the ground state wave vector in bulk Er being $5/21\pi$ [15], which favours the AFC between neighbouring Tb layers. The application of a magnetic field leads to a ferromagnetic alignment of the magnetization component parallel to the field, while the AFC survives for the component perpendicular. So far we cannot explain the origin of the thermal hysteresis, but we speculate that magneto-elastic interactions are involved, as they drive the lock-in of the propagation vector in bulk Er [14]. To conclude we have shown, that ferromagnetic order appears below a transition temperature $T_C = 230$ K. The ferromagnetic alignment is disturbed at low temperatures, leading first to magnetic fluctuations and finally to AFC. This might be attributed to the propagation vector in the Er layers becoming commensurate with the superlattice periodicity. Further refinement of the presented data is ongoing and will give quantitative information about the layer magnetization in Er and Tb layers, respectively. To fully understand the complex magnetic structure of the [Er$_{21}$|Tb$_{10}$] superlattice it is necessary to develop a model that takes the different energy contributions and disorder at the interfaces into account. A systematic study of the composition dependence is ongoing and will reveal new hints to solve this problem.

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