Appl. Phys. A 74 [Suppl.], S1566–S1569 (2002) / Digital Object Identifier (DOI) 10.1007/s003390201810

Applied Physics A
Materials
Science & Processing

Deuterium-induced magnetic decoupling in a Ho(00.1)/Y superlattice

V. Leiner[1,2,*], D. Labergerie[1], M. Ay[1], H. Zabel[1]

[1] Institut für Experimentalphysik/Festkörperphysik, Ruhr-Universität Bochum, 44780 Bochum, Germany
[2] Institut Laue-Langevin, B.P. 156, 38042 Grenoble Cedex 9, France

Received: 17 July 2001/Accepted: 11 December 2001 –

Abstract. Ho/Y superlattices have been studied extensively in the past concerning their phase transition and the propagation of the magnetic spiral through the Y spacer layer. Hydrogen in rare-earth metals is known to alter the electronic and magnetic properties. By means of neutron scattering, we have investigated the change of the magnetic spiral and phase transition of a $[77\,\text{Å}\,\text{Ho}(00.1)/52\,\text{Å}\,\text{Y}]_{30}$ superlattice upon deuteration. Neutron reflectivity confirms that deuterium preferentially occupies the Y spacer layers, maintaining the structural coherence of the superlattice. Low-temperature reciprocal space maps of the pristine Ho/Y superlattice exhibit a magnetic satellite peak τ, convoluted with the satellite peaks of the chemical period, indicative for exchange coupling between the Ho layers mediated by the Y spacers. After deuterium uptake the splitting of the τ peak vanishes. The spiral becomes confined to the individual Ho blocks, similar to single thin Ho films. The ordering temperature, T_N, is found to be ≈ 118 K and constant under deuterium loading. Thus coupling–decoupling the Ho blocks does not affect the ordering temperature, whereas it depends on the individual Ho layer thickness via scaling.

PACS: 75.70.Cn; 61.12.-q; 68.55. Ln

In ferromagnetic/paramagnetic heterostructures the amplitude and sign of the interlayer exchange coupling (IEC) between ferromagnetic layers can be tuned if the mediating spacer layer allows hydrogen up-take. This has been demonstrated for Fe/Nb(H) [1] as well as for Fe/V(H) [2, 3], where a sign reversal of the interlayer coupling constant, J', can be achieved for specific hydrogen concentrations. The more complex interlayer exchange coupling in RE films and superlattices may also be tuned with hydrogen. However, since hydrogen may enter both the magnetic and the non-magnetic spacer layers, more caution is appropriate for interpreting the results.

*Corresponding author. (Fax: +49-2343/214-173, E-mail: leiner@ill.fr)

It is well-known that e.g. Y, La, and the rare earth (RE) metals readily take up hydrogen. In these systems the introduction of hydrogen leads to three different phases with different crystal structures: a) the hexagonal α-phase for low H-concentrations. In this phase the hydrogen behaves like a lattice gas, expanding the host lattice without changing its symmetry. b) The cubic β-phase near the stoichiometric dihydride REH_2 with a tetrahedral occupancy of the interstitial sites; and c) the hexagonal γ-phase near to the trihydride REH_3. Thus hydrogen in Y has been used for optical switching between the metallic YH_2 and a transparent YH_3 phase signaling a metal-insulator transition [4].

Ho exhibits an incommensurate spin helix below the Néel temperature, $T_N = 131.5$ K. The spin helix consists of ferromagnetically ordered moments in the hexagonal basal plane and a turn angle α between the magnetisation vector from one plane to the next. The reciprocal lattice vector of the spiral $\tau(T)$ and the turn angle are related via $\alpha = 180° \times \tau^\star$, where $\tau^\star$ is given in reciprocal lattice units of the c-axis. The helical magnetic structure is only preserved in the α-phase, but with increasing H- concentration the Néel temperature drops with a rate of ≈ 1 K per atomic percent of hydrogen and the turn-angle slightly decreases [5]. The dihydride phase exhibits a complex antiferromagnetic structure below about 5 K with a reduced magnetic moment [6], whereas the trihydride phase shows no magnetic order.

Ho/Y superlattices offer a model system for investigating potential switchable interlayer coupling in a rare earth system. Ho/Y superlattices have been studied extensively in the past as concerns their phase transition and the propagation of the magnetic spiral through the Y spacer layer [7]. For a Y spacer thickness of not more than 80 Å it has been found that the interlayer coupling between adjacent Ho layers is mediated by the Rudermann-Kittel-Kasuya-Yosida (RKKY) interaction, supported by the nesting feature of the Y Fermi surface in the c-direction. Ho/Y superlattices are also very favorable for neutron scattering studies because of the high atomic moment of Ho of $\approx 10\mu_B$ and the incommensurate helical magnetic structure, giving rise to intense τ-peaks at low q-values. Furthermore, the very good growth properties via molecular beam epitaxy (MBE) are well-established.

Loading Ho/Y superlattices with deuterium (D) instead of hydrogen offers the advantage of adding a positive scattering length. From neutron reflectivity data it is then possible to decide the preferential occupation of deuterium in one of the sublattices.

1 Experimental

The investigated superlattice was grown by MBE on an $Al_2O_3\,(11\bar{2}0)$ substrate using a Nb/Y buffer system and capped with Y/Nb/Pd to ensure symmetric interfaces for the Ho blocks; the Pd cap layer serves as a protection against oxidation and as a window for the D uptake.

The neutron experiments were carried out using the ADAM reflectometer [8–10] at the Institut Laue-Langevin, Grenoble. Scans were taken with an unpolarised beam of a fixed wavelength, $\lambda = 4.41$ Å. In order to permit an in-situ loading of the sample, a close-cycle refrigerator was equipped with a sample container, which, in turn, was connected to an outside loading station by a capillary. A temperature range of 10 K to 400 K was accessible during the experiment. Specular and off-specular data were collected simultaneously by using a multi-wire ^{3}He position-sensitive detector (PSD).

2 Results

Owing to the considerable cross-section of deuterium for neutrons, the scattering length density profile in the sample strongly depends on the D concentration within the different layers. In Fig. 1 neutron reflectivity curves for the virgin superlattice and the superlattice loaded at three different pressures, p_D, are plotted. Several qualitative but important features are immediately noticeable from the data: In the virgin state a superlattice peak corresponding to the Ho/Y bilayer period is clearly observable at $q = 0.05\,\text{Å}^{-1}$. With increasing deuterium pressure the superlattice peak first vanishes (15 mbar) and reappears at higher D pressures. The loss of contrast at 15 mbar arises from a matching of the scattering length densities in the Ho and Y blocks of the bilayers. As the scattering length density of Ho is higher than that of Y ($\varrho_{Ho} = 2.57 \times 10^{-6}\,\text{Å}^{-2}$, $\varrho_Y = 2.35 \times 10^{-6}\,\text{Å}^{-2}$), the D uptake has to occur primarily in the Y layer. At 30 mbar and above contrast is recovered as the scattering length density in the YD_x becomes larger than that of HoD_x.

The formation of YD_3 or HoD_3 can be excluded as the formation of either trihydrate would shift the critical edge to higher q-values [11].

We also performed temperature-dependent measurements of the magnetic satellite peaks on the virgin sample and on the sample loaded at $p_D = 60$ mbar. In Fig. 2 the observed intensities at 20 K (top row) and 115 K (bottom row) as a function of momentum transfer perpendicular and parallel to the sample surface are depicted.

The virgin sample displays sharp satellite peaks sampling an envelope corresponding to the magnetic structure factor of one Ho layer. This is clear evidence for the long range coherence of the spin helix within the Ho blocks mediated across the Y spacers. Upon deuteration the splitting of the τ-peak is lost. The broad peak corresponds to the magnetic signal one

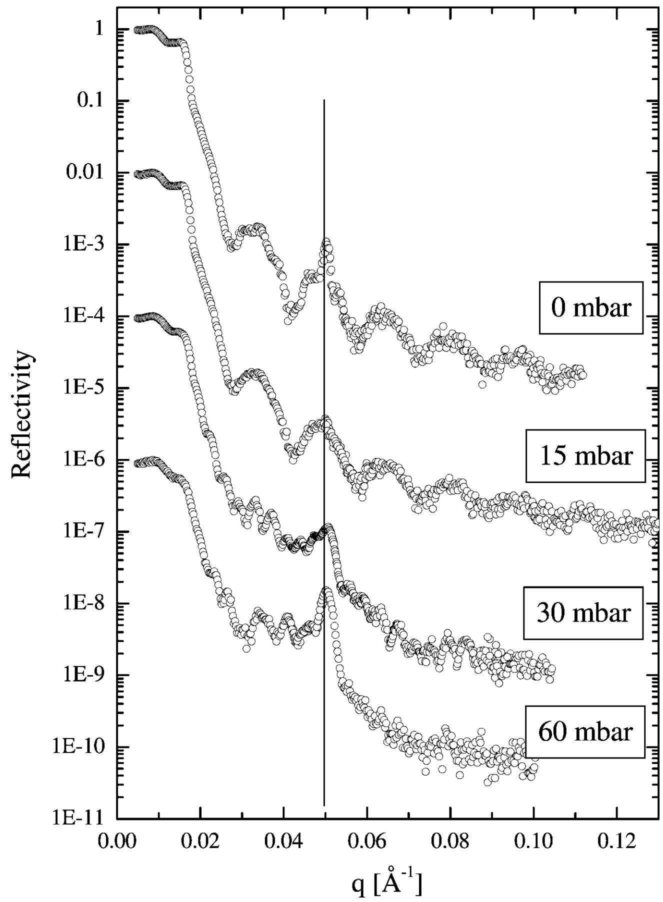

Fig. 1. Reflectivity curves taken with an unpolarised beam for different D concentrations

would obtain from a single isolated Ho film, strongly indicating that the magnetic correlation is lifted by the deuterium uptake in the mediating Y layer. However, the off-specular intensity of the loaded sample still exhibits weak features reminiscent of the coupled superlattice. While a long long range propagation of the magnetic helix across the superlattice is suppressed through D uptake, some correlation appears to persist, which, however, does not extend over more that a few Ho layers at most. For both, the virgin and the fully loaded sample an ordering temperature of ≈ 118 K is observed.

In Fig. 3 line scans are shown that are extracted from the intensity maps in Fig. 2 by interpolating the original data to a rectangular grid and then summing the intensities along the q_x direction. In Fig. 3a results are shown for the virgin sample (solid dots). $2\pi/\Lambda$ corresponds to the reciprocal period of the Ho/Y superlattice. The dashed line represents the structure factor for a single isolated Ho layer; the position and width of this (00τ)-peak is governed by the turn-angle of the magnetic helix and the number of coherently scattering Ho atomic planes, respectively. The lower panel (b) shows line scans for the deuterium loaded sample. The solid points were obtained by summing over the specularly reflected intensity at $q_x \pm 1.22 \times 10^{-3}\,\text{Å}^{-1}$. The resulting curve has the same width as the one calculated for a single Ho layer (dashed line in panel (a)), supporting the notion of decoupled Ho layers with an incoherent superposition of the scattered intensity.

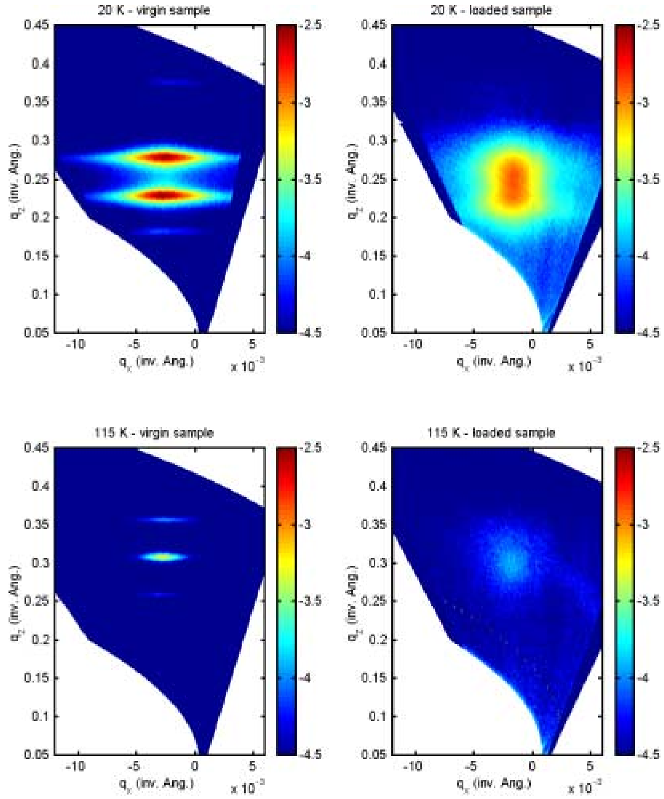

Fig. 2. Specular and off-specular intensities from the virgin and fully loaded sample at 20 K and 115 K

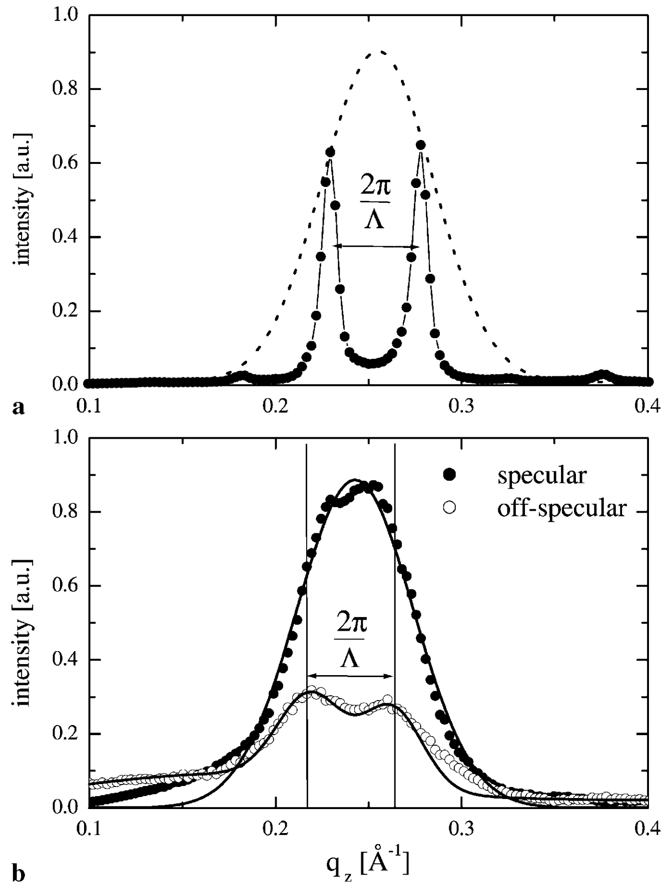

Fig. 3. Linescans extracted from the intensity maps for the virgin and fully loaded sample. Data were taken at 20 K

A slight shift of the (00τ) peak is observed from $\tau^* = 0.225c^*$ to $\tau^* = 0.217c^*$. This may be due to the formation of an alpha-phase HoD_x with $x \leq 2\%$.

The off-specular diffuse scattering apparently shows a different intensity pattern than the specular ridge. Instead of one peak, a double peak structure is observed after deuterium up take, with a distance between the peaks corresponding to the original superlattice period, $2\pi/\Lambda$. However, the peak widths have considerable increased with respect to the superlattice peaks in the virgin sample. This may be explained by the coexistence of α- and β-phase Yttrium hydride. Whereas the Ho films are predominantly decoupled by Yttrium in the β-phase, some α-YH_x domains remain which create "shortcuts" for a correlation of the magnetic helix extending across a few Ho-layers only. In the measurements the specular line is found to be primarily given by those domains where a complete decoupling of the Ho blacks has occurred, whereas the off-specular regime shows an enhanced sensitivity to those regions in the sample where some correlation is retained. The decoupling of the Yttrium layers in the β-phase can be understood as arising from a depletion of the Y density of states at the Fermi surface by opening a new hydrogen derived band below the $4d5s$ Y-band, thereby reducing the Y susceptibility for RKKY coupling of the Ho layers.

3 Conclusion

We have demonstrated that the exchange coupling in a Ho/Y superlattice can be altered through deuterium uptake in the Y spacer layer. Reflectivity measurements confirm that the deuterium resides primarily in the Yttrium spacer layer. In the virgin sample, a long-range coherence of the magnetic helix across the Y spacer layer is found. The measured ordering temperature of 118 K and the turn angle of of the magnetic spiral corresponds to the value expected for an undeuterated single Ho film of 77 Å. At the highest deuterium pressure of 60 mbar investigated here, a almost complete suppression of the Ho exchange coupling via the Y spacer layers could be observed. Thus the Y layers in the β phase effectively block the magnetic correlation of the Ho spiral. Some weak and short range correlation remains as recognized in the diffuse part of the scattering pattern, which is most likely due to residual Y domains in the α phase.

Acknowledgements. This work was supported by the Bundesministerium für Bildung und Forschung under Contract No. 03-ZA5BC20 and the DFG-SFB 491: Magnetic heterostructures.

References

1. F. Klose, Ch. Rehm, D. Nagengast, H. Maletta, A. Weidinger: Phys. Rev. Lett. **78**, 1150 (1997)
2. B. Hjörvarsson, J.A. Dura, P. Isberg, T. Watanabe, T.J. Udovic, G. Andersson, C.F. Majkrzak: Phys. Rev. Lett. **79**, 901 (1997)
3. D. Labergerie, C. Sutter, H. Zabel, B. Hjörvarsson: J. Magn. Magn. Mater. **192**, 238 (1999)

4. J.N. Huiberts, R. Griessen, J.H. Rector, R.J. Wijngarten, J.P. Dekker, D.G. de Groot, N.J. Koeman: Nature (London) **380**, 321 (1996)
5. C. Sutter, D. Labergerie, A. Remhof, H. Zabel, C. Detlefs, G. Grübel: Europhys. Lett. **53 (2)**, 257 (2001)
6. L. Grier: Ph.D. thesis, (University of Oxford 2000)
7. D.A. Jehan, D.F. McMorrow, R.A. Cowley, R.C.C. Ward, M.R. Wells, N. Hagmann, K.N. Claussen: Phys. Rev. B **48**, 5594 (1993)
8. A. Schreyer, R. Siebrecht, U. Englisch, U. Pietsch, H. Zabel: Physica B **248**, 349 (1998)
9. R. Siebrecht, A. Schreyer, U. Englisch, U. Pietsch, H. Zabel: Physica B **241**–**243**, 169 (1998)
10. www.ill.fr/yellowbook/adam
11. A. Remhof, G. Song, Ch. Sutter, A. Schreyer, R. Siebrecht, H. Zabel: Phys. Rev. B **59**, 6689 (1999)