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Depth-dependent investigation of the distribution of the spin density waves in thin chromium films with surface X-ray and neutron scattering

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Abstract

For thin epitaxial Cr(0 0 1) films capped with a ferromagnetic Fe layer a transverse spin density wave (SDW) is expected which propagates in the out-of-plane direction with the Cr spins aligned parallel to the film plane in the direction of the Fe magnetization vector. Synchrotron and neutron scattering experiments show, however, that the SDW wave propagates parallel to the film plane with spins oriented out-of-plane. In addition, a commensurate antiferromagnetic phase is found. The re-orientation of the SDW is caused by a frustrated Fe–Cr exchange coupling introduced by monoatomic steps at the Fe–Cr interface. Complete re-orientation takes place over some distance close to the interface reducing severely the coherence length of the SDW structure. With the surface scattering method we have measured the coherence length of the SDW as a function of depth. Furthermore, we have investigated the role of the commensurate antiferromagnetic phase near the Fe–Cr interface. We find no scattering from a commensurate order, implying a layering of the two phases with the incommensurate phase on top. © 1998 Elsevier Science B.V. All rights reserved.

1. Introduction

Below the Néel temperature of $T_N = 311$ K the itinerant antiferromagnet chromium exhibits an incommensurate spin density wave (SDW) state in which the spin density varies sinusoidally in space [1]. The wave vector $\boldsymbol{Q} = 2\pi/a(1-\delta)$ of the modulation, where δ is a measure of the deviation from commensurability, may point along any of the $\{0\,0\,1\}$ directions in the BCC Cr lattice. In bulk Cr all three possible orientations occur with equal probability. The SDW always is accompanied by a strain wave (SW) and a charge density wave (CDW) which are periodic modulations of the lattice spacing and the charge density, respectively. Those modulations give rise to satellite reflections in the reciprocal lattice of Cr, which can be ob-

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served either with magnetic neutron scattering in case of the SDW or with synchrotron scattering in case of the CDW/SW [2–4]. Only recently the spin density waves in epitaxial Cr films were explored [5,6]. Of particular interest is the effect of ferromagnetic boundary layers on the antiferromagnetic state of Cr and the propagation direction of the SDW. Furthermore, the role of *commensurate* antiferromagnetic order must be understood, since it also occurs in epitaxial Cr films [5,6], From the mutual influence between ferromagnetic Fe layers on one hand and the antiferromagnetism of Cr on the other hand, the exchange coupling in Fe–Cr superlattices [7], the non-collinear orientation of the magnetization vectors in the Fe layers [8,9], and the concomitant giant magneto-resistance may be better understood [10,11].

We have carried out extensive scattering studies to investigate the SDW in epitaxially grown thin Cr(0 0 1) films, including surface and interface effects [6,12,13]. These studies show that unlike in bulk Cr, in a 3000 Å thick Cr(0 0 1) film the SDWs are almost entirely longitudinal, i.e. a single SDW wave vector $\boldsymbol{Q}$ pointing in the out-of-plane with the magnetic moments parallel to $\boldsymbol{Q}$. A thin ferromagnetic Fe cap layer of only 20 Å thickness causes a complete re-orientation of $\boldsymbol{Q}$ from longitudinal out-of-plane to transverse in-plane. The magnetic moments of Cr are oriented in the out-of-plane direction, perpendicular to the in-plane magnetized Fe layer, as schematically shown in Fig. 1. This re-orientational transition can be understood in terms of frustration effects at the Fe–Cr interface, whenever the strong antiferromagnetic interaction

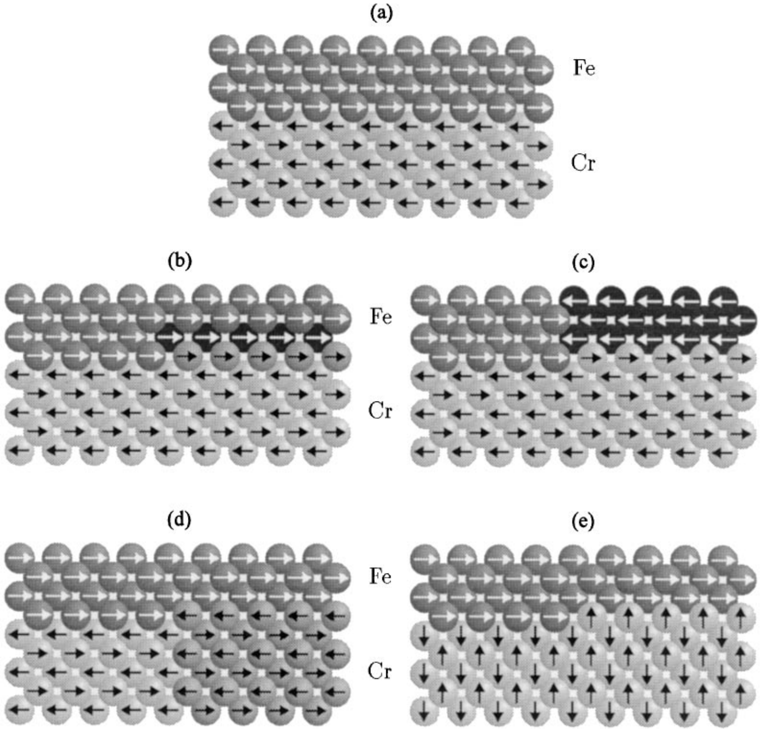

Fig. 1. Schematics of different spin density wave structures in Cr films covered with a ferromagnetic Fe cap layer. In (a) the ideal structure is shown with a sharp interface between the ferromagnetic top layer and the antiferromagnetic Cr film. In this case a transverse spin density wave with propagation direction perpendicular to the film plane and magnetic moments parallel to the interface are expected. Monoatomic steps at the interface cause frustrations (b), which can be overcome via domain formation in the Fe layer (c), or Cr film (d). For thick Cr films the system reacts, however, by rotation of the spin density wave vector with propagation parallel to the film plane and Cr magnetic moments at right angles to the magnetization vector of the Fe cap layer (e).

between Fe and Cr across the interface is frustrated in the presence of monatomic steps and kinks. In addition to the incommensurate SDW, commensurate antiferromagnetic order is also observed [6,12,13]. This phase may be attributed to strains [1] induced by the epitaxial growth process. Whereas the commensurate phase cannot be observed with synchrotron radiation, it yields strong (1 0 0) peaks in neutron scattering. In the following we show with depth controlled surface synchrotron scattering experiments that the transverse in-plane SDW originates at the Fe–Cr interface and continues to deeper regions. Furthermore, we demonstrate with neutron surface diffraction, that no commensurate antiferromagnetic order occurs at the Fe–Cr interface, implying a layering of the commensurate and incommensurate phases.

2. Experimental

The epitaxial Cr(0 0 1) films were prepared by molecular beam epitaxy on an $Al_2O_3(1\,\bar{1}\,0\,2)$ substrate with a 500 Å thick Nb(0 0 1) buffer layer. Nb is well known for its excellent growth quality on sapphire substrates and its use as a buffer for the growth of Cr on sapphire [14]. The Cr film was grown on top of Nb at a substrate temperature of 450°C and a growth rate of 0.1 Å/s. To improve the structural coherence and to smoothen the surface of the Cr film the sample is annealed for 30 min at 750°C immediately after the growth. Then, a 20 Å thick magnetic Fe cap layer is grown at 300°C with a growth rate of 0.1 Å/s. These specific growth parameters are chosen to avoid interdiffusion between the Fe and the Cr layers. To protect the Fe layer from oxidation an additional 20 Å thin Cr layer is deposited on top of the Fe layer. It is well known that Cr forms a thin and stable protective oxide scale when exposed to atmospheric conditions with a thickness of not more than 20 Å at room temperature [15]. For the neutron experiments large surface samples were grown on $5 \times 5\,\mathrm{cm}^2$ substrates.

The synchrotron experiments were carried out at the TROIKA beamline at the ESRF. The wavelength was set by a diamond monochromator to 1 Å. Measurements were taken between 10 and 300 K by using a He displex cryostat with Be windows. For maximum intensity the glancing exit angle α_f was set to be equal to the critical angle for total reflection α_c. The penetration depth of the evanescent wave was controlled by the incident glancing angle α_i.

The neutron experiments were carried out on the EVA grazing incidence diffractometer at the ILL at a wavelength of 2.75 Å [16]. Measurements were taken between 20 and 300 K using a special displex cryostat for large samples with Al windows. Scans were taken as a function of α_i with the in-plane scattering vector aligned either on commensurate or incommensurate peaks. A position sensitive detector provided resolution in α_f.

3. Experimental results and discussion

In Fig. 2 we show synchrotron scans of the strain wave under surface scattering conditions in a 3300 Å thick Cr(0 0 1) oriented film capped with a 20 Å thick Fe layer. The in-plane (0 2 0) fundamental Bragg peak is excited together with the satellite peaks at $\pm 0.09\,\text{Å}^{-1}$ on either side of the fundamental peak. The (0 2 0) peak is removed for clarity because its intensity is many orders of magnitude higher than the intensity of the satellite reflections. The data were taken at 10 K and the penetration depth was varied between 30–120 Å. Each scan reproduced in Fig. 2 is labeled with the normalized glancing incident angle α_i/α_c and the corresponding penetration depth Λ. For a better comparison, all scans are normalized to the intensity of their respective fundamental (0 2 0) peak and they are shifted against each other by a constant amount for clarity.

From 30 to 42 Å, no satellite intensities are visible which is expected due to the protective oxide surface cover and the Fe layer. At a penetration depth of about 60 Å, the satellite peaks become first noticeable and they grow in intensity as the penetration depth is further increased. The existence of these peaks shows that the incommensurate SWs in Cr indeed propagate parallel to the interface. From the synchrotron experiments we can only determine the propagation of the SWs or SDWs, which are parallel, but not the orientation of the Cr spins.

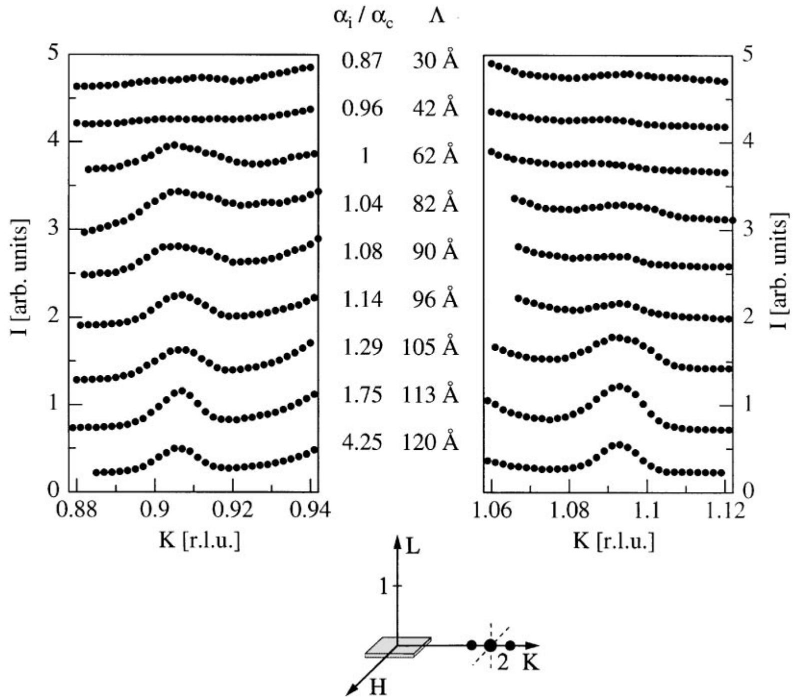

Fig. 2. Depth-dependent measurement of the in-plane CDW/SW satellite reflections around the Cr(0 2 0) Bragg-reflection from 3300 Å thick Cr film covered with a 20 Å thick Fe layer at $T = 10\,\mathrm{K}$. For each scan the incident angle α_i and the resulting penetration depth Λ is given.

On the other hand, from neutron scattering experiments we know that the Cr spins point out-of-the plane, as schematically depicted in Fig. 1e [12]. Thus the surface scattering experiments reveal that the transverse in-plane SDWs in Cr(0 0 1) films exist already close to the Fe–Cr interface.

For penetration depths close to the interface the width of the satellite peaks is considerably broader than for larger penetration depths, indicating a reduced coherence length of only about 50 Å for the SW in proximity to the ferromagnetic Fe layer. This behavior can be understood as follows. For an ideal sharp interface, the Fe and Cr magnetic moments favor an antiparallel alignment, as indicated in Fig. 1a. Monoatomically high steps at the interface cause frustrations of the interlayer exchange coupling. This frustration can be overcome in several different ways as shown in Fig. 1b–e. Either domains are formed in the Fe layer as shown in Fig. 1c or in the Cr layer as indicated in 1d, or the Fe and Cr magnetic moments are parallel over the length of a terrace (b). In either case, energy is required to form domains at the expense of exchange energy. Re-orientation of the Cr magnetic moments from parallel to perpendicular is energetically the most favorable compromise. Then no interface exchange energy is gained, on the other hand, no domain energy has to be paid. Furthermore, re-orientation of the Cr moments requires less energy than re-orientation of the Fe moments, for which the shape anisotropy has to be overcome. Recent Monte Carlo simulations using a Heisenberg Hamiltonian for describing the spin structure and exchange coupling confirm that in the presence of steps the Cr magnetic moments re-orient in the direction perpendicular to the Fe moments [17,12,13]. Starting from parallel alignments at the interface, this re-orientation takes place over some distance from the interface. Finally the SWs on the left and right side of the step combine to set up a joint SW now propagating parallel to the surface. The broadening of the SW satellite reflections as observed by surface synchrotron experiments is most likely due to finite coherence lengths in the region where the re-orientation takes place.

Whereas with synchrotron scattering the incommensurate SDW can be easily observed via the concomitant strain wave, any commensurate antiferromagnetic order would be overlooked. The observed incommensurate and commensurate phases could coexist laterally in domains or they could be stacked on top of each other, one phase in contact with the Fe–Cr interface and the other with Nb the buffer. From the synchrotron results it can already be concluded that the incommensurate SDW is in contact with the Fe. To establish the role of the commensurate phase, neutron diffraction under grazing incidence was performed. Measurements were carried out with the in-plane scattering vector aligned either with the commensurate (1 0 0) peak or with an incommensurate (0.953 0 0) SDW reflection. In Fig. 3 an α_i scan for the latter case is shown at $T = 50$ K for a 3000 Å thick Cr film with 20 Å Fe on top. A peak is clearly observed at the critical angle α_c, identifying it as a surface peak. Thus, consistent with the synchrotron data, we find an incommensurate SDW near the Fe–Cr interface. In an equivalent scan at the (1 0 0) position no such peak was found. Therefore, we can conclude that near the Fe–Cr interface the incommensurate SDW prevails. The commensurate reflections observed with conventional high angle neutron scattering must come from deep within the film. Consequently the occurence of the commensurate phase in this system appears not to be induced by the Fe–Cr interface.

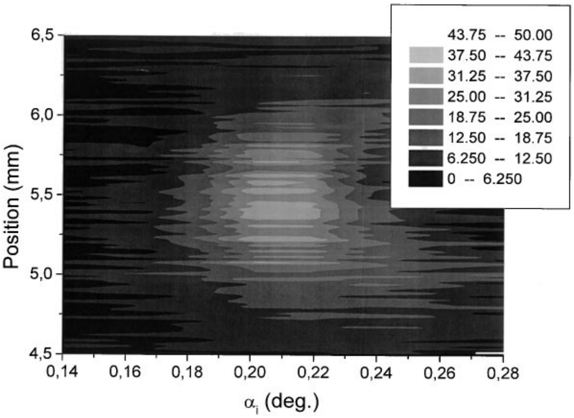

Fig. 3. Neutron grazing incidence diffraction at the incommensurate (0.953 0 0) position of the Cr SDW at $T = 50$ K. Shown is a contour plot of the spectra taken with the position sensitive detector as a function of grazing angle α_i. To correct for background, a spectrum measured away from the surface peak has been subtracted.

4. Summary

We have studied with surface synchrotron scattering experiments the propagation of incommensurate strain waves in thin epitaxial Cr(0 0 1) films. The Cr films are covered with a 20 Å thick Fe layer causing the SWs to re-orient from a propagation in the out-of-plane direction to the in-plane direction. The re-orientation is the result of a frustrated Fe–Cr interfacial exchange coupling introduced by monoatomically high steps at the interface and takes place over a distance of about 30 Å from the interface. The transition region is recognized by broadened strain wave satellite reflections indicating a reduced coherence length close to the Fe–Cr interface. Neutron grazing angle diffraction experiments confirm the synchrotron results and furthermore establish that the additionally observed commensurate antiferromagnetic phase does not persist up to the Fe–Cr interface. Thus, the incommensurate and the commensurate phase coexist in a layered fashion rather than in lateral domains.

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