# Reorientation of Spin Density Waves in Cr(001) Films Induced by Fe(001) Cap Layers

P. Bödeker,[1] A. Hucht,[2] A. Schreyer,[1,3] J. Borchers,[3] F. Güthoff,[4] and H. Zabel[1]
[1]*Institut für Experimentalphysik/Festkörperphysik, Ruhr-Universität Bochum, D-44780 Bochum, Germany*
[2]*Theoretische Physik, Gerhard-Mercator-Universität Duisburg, D-47048 Duisburg, Germany*
[3]*National Institute of Standards and Technology, Gaithersburg, Maryland 20899*
[4]*Forschungszentrum Jülich, D-52425 Jülich, Germany*
(Received 17 November 1997)

Proximity effects of 20 Å Fe layers on the spin density waves (SDWs) in epitaxial Cr(001) films are revealed by neutron scattering. Unlike in bulk Cr we observe a SDW with its wave vector **Q** pointing along only one {100} direction which depends dramatically on the film thickness $t_{\mathrm{Cr}}$. For $t_{\mathrm{Cr}} <$ 250 Å the SDW propagates out of plane with the spins in the film plane. For $t_{\mathrm{Cr}} > 1000$ Å the SDW propagates in the film plane with the spins out of plane perpendicular to the in-plane Fe moments. This reorientation transition is explained by frustration effects in the antiferromagnetic interaction between Fe and Cr across the Fe/Cr interface due to steps at the interface. [S0031-9007(98)06723-4]

PACS numbers: 75.70.–i, 75.25.+z, 75.30.Fv

While the incommensurate spin density wave (SDW) antiferromagnetism is well established for bulk Cr [1], it is presently of high interest to analyze how the magnetic properties of Cr are altered either by reduced dimensionality in thin films or by proximity effects to ferromagnetic (FM) layers. The magnetic state of Cr is particularly interesting since ultrathin Cr films play an important role in exchange coupled Fe/Cr superlattices exhibiting giant magnetoresistance effects [2,3]. Also for theoretical treatments of the exchange coupling it is uncertain whether the Cr spacer layer should be treated as a paramagnet, an antiferromagnet, or as a proximity induced antiferromagnet [4]. In this context, the role of the Fe/Cr interface is a matter of intense study [5,6]. Magnetic domain imaging of an Fe layer deposited on a wedge shaped Cr layer on an Fe whisker shows a domain pattern switching between parallel and antiparallel alignments having a periodicity of two Cr(001) monolayers and a phase shift consistent with a SDW state [7]. More recently, neutron scattering and perturbed angular correlation spectroscopy (PACS) have been used on Fe/Cr(001) superlattices to investigate the magnetic structure of Cr directly for Cr film thicknesses $t_{\mathrm{Cr}}$ of about 30–400 Å [8–10]. Although some inconsistencies still remain, these experiments show that the SDW state collapses for Cr films well below the period $\Lambda$ of the SDW.

The aim of the present work is to gain a basic understanding of the effect of FM proximity layers on the magnetic properties of thin Cr(001) films in the SDW phase. The thickness range of the Cr films (200–3000 Å) is chosen such that the question of the presence of a SDW state is not an issue. Using neutron scattering we find that the propagation direction of the SDW depends dramatically on the Cr film thickness. Our experimental results are rationalized by computer simulations using a Heisenberg model which takes realistic Fe/Cr interfaces with interfacial roughness and interdiffusion into account. Complementary experiments with synchrotron radiation will be discussed elsewhere.

We have grown epitaxial Fe/Cr(001) bilayers by molecular beam epitaxy on $Al_2O_3$ $(1\bar{1}02)$ substrates with a 500 Å thick Nb(001) buffer layer, following well established growth recipes [11,12]. Cr(001) films with thicknesses from 200–3000 Å were grown on the Nb buffer layer at 450 °C with a growth rate of 0.1 Å/s. As evidenced by reflection high energy electron diffraction (RHEED) during growth, the crystalline quality of the Cr near the Cr/Nb interface is not very high due to the 14% lattice mismatch between Nb and Cr. However, with growing Cr thickness the film quality improves dramatically [12]. After annealing for 30 min at 750° the Fe cap layer was grown at 300° at a rate of 0.1 Å/s. For the scattering experiments it was necessary to keep the absolute amount of Cr in the samples roughly constant. Therefore, for samples with $t_{\mathrm{Cr}} < 1000$ Å the Fe/Cr structure was repeated several times up to a minimum total $t_{\mathrm{Cr}}$ of 2000 Å. All additional Fe/Cr layers were grown at 300 °C. For protection against oxidation all samples were covered with a 20 Å Cr layer [13]. X-ray scattering shows that all samples are of high crystalline quality, with an out-of-plane crystalline coherence length of 60%–80% of the total film thickness and a mosaic spread of about 0.2° FWHM measured at the (002) peak.

To determine the magnetic structure, neutron scattering experiments were performed on the triple-axis spectrometers BT-2 of the National Institute of Standards and Technology and UNIDAS at the KFA Jülich, Germany. In both cases we used pyrolytic Graphite PG(002) monochromator and analyzer crystals to select a wavelength of $\lambda = 2.351$ Å. Graphite filters suppressed any $\lambda/2$ contamination.

Bulk Cr exhibits an incommensurate SDW, i.e., the magnitude of the antiferromagnetically aligned Cr magnetic moments $\mu$ varies sinusoidally with a temperature

 0031-9007/98/81(4)/914(4)$15.00 

dependent period $\Lambda$ of about 21 lattice constants at $T = 0$ [1]. The incommensurability is ascribed to a nesting vector along the {100} directions of the Cr Fermi surface. The wave vector **Q** defines the direction of propagation of the SDW. At lowest temperatures a longitudinal SDW (LSDW) forms, i.e., $\mu$ is parallel to **Q**. Above the spin-flip transition temperature, $T_{\mathrm{SF}} = 123$ K, $\mu$ is perpendicular to **Q**, forming a transverse SDW (TSDW). Above $T_N = 311$ K bulk Cr is paramagnetic.

The incommensurate modulation of the antiferromagnetic (AF) spin structure by the SDW causes *two* satellite peaks to occur around the {1,0,0} positions [1], e.g., at $(0,0,1\pm\delta)$, $(1\pm\delta,0,0)$, and $(0,1\pm\delta,0)$ which can be investigated by neutron scattering. Here $\delta = 1 - |\mathbf{Q}|$ with $|\mathbf{Q}|$ in reciprocal lattice units, $\delta = a/\Lambda$, and the Cr lattice constant $a$. In the first case, the position of the satellites indicates **Q** being oriented out of the film plane, the two latter cases occur for either direction of in-plane propagation. In addition, the polarization of the SDW (i.e., TSDW or LSDW) can be obtained by making use of the selection rule for magnetic neutron scattering. It requires a component of the magnetization vector $\mu$ to be perpendicular to the scattering vector $|\mathbf{q}| = (4\pi/\lambda)\sin\theta$ where $\theta$ is the scattering angle. Thus, a longitudinal SDW propagating along $L$, i.e., out of plane, will generate no intensity at $(0,0,1\pm\delta)$. However, satellites will occur at $(1,0,0\pm\delta)$. A commensurate AF ($AF_0$) phase, on the other hand, will yield a *single* peak of purely magnetic origin at the Cr{001} positions. Thus, with neutron scattering we can uniquely determine $AF_0$ and SDW magnetic order as well as SDW propagation and polarization. For a more detailed discussion, see, e.g., Ref. [14].

Figure 1 reproduces neutron scans taken at possible satellite positions around the Cr(001) and Cr(010) positions of a 3000 Å Cr(001) film capped with a 20 Å Fe layer for temperatures of 30–500 K. To make the spectra comparable, all intensities have been normalized with respect to the structural reflections. For all temperatures only satellites in scans along the in-plane $K$ direction occur, whereas in scans along $L$ no satellites are found. Thus, only SDWs propagating in the film plane are present. For $T = 30$ K satellites appear around the (001) position. From the selection rule described above we conclude that at $T = 30$ K a LSDW propagates in the plane with the spins dominantly oriented in plane. For temperatures $T \geq 50$ K, however, we find the reversed situation. The satellites occur only around the (010) position. Thus, we conclude that the SDW still propagates in plane but that the polarization has changed to transverse. Moreover, the absence of any satellites in the K-scan around (001) tells us that for temperatures $T \geq 50$ K we observe a TSDW with the spins pointing out of the film plane, i.e., perpendicular to the Fe/Cr interface. The observed spin flip transition from the LSDW to the TSDW at approximately $T_{\mathrm{SF}}^{\mathrm{film}} = (40 \pm 10)$ K occurs at a much lower temperature than in bulk ($T_{\mathrm{SF}}^{\mathrm{bulk}} = 123$ K). Around

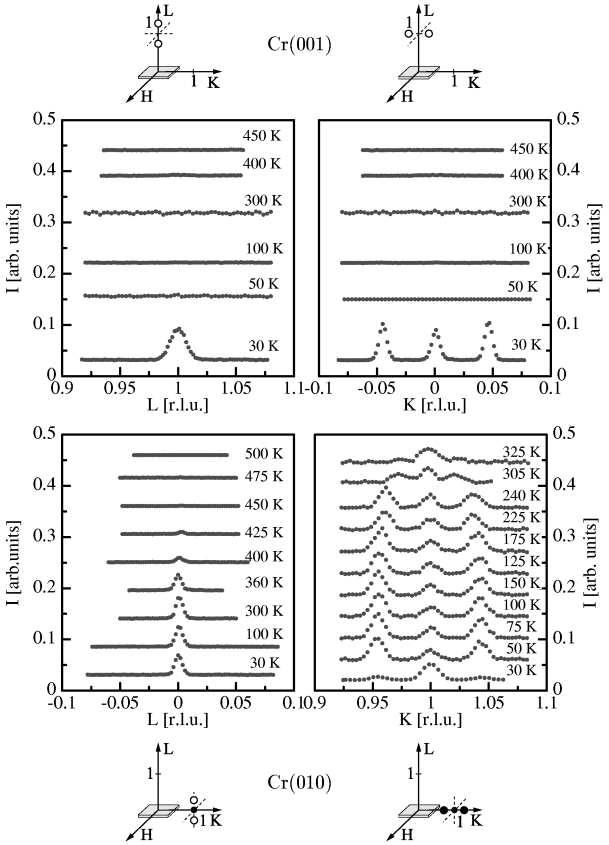


FIG. 1. Neutron scans through the possible satellite positions around the (001) (top) and (010) (bottom) position for a 3000 Å thick Cr(001) film capped with a 20 Å Fe layer for $30 \leq T \leq 500$ K. For each scan direction a schematic picture of the reciprocal lattice with the open and solid circles as a representation of the possible and observed satellites at 100 K is shown.

311 K the SDW satellites disappear consistent with the bulk Néel temperature.

Another feature of the data of Fig. 1 is the presence of intensity commensurate with the Cr(001) and Cr(010) positions indicating an additional $AF_0$ phase. From the temperature dependence the Néel temperature is found to be $T^{\mathrm{com}} \approx 450$ K. Using the selection rule we again observe a flipping of the spins from in plane below $T_{\mathrm{SF}}^{\mathrm{film}} = 40 \pm 10$ K to out of plane above $T_{\mathrm{SF}}^{\mathrm{film}}$. The origin of the $AF_0$ phase will be discussed below.

In summary, we find an in-plane LSDW and an $AF_0$ phase coexisting below 40 K, both with spins in plane. Above 40 K an in-plane TSDW and an $AF_0$ phase coexist, both with spins out of plane. Thus, our measurements imply that the Cr moments are oriented *perpendicular* to the Fe moments since magnetization measurements confirmed that the Fe is magnetized in plane.

In Fig. 2 results of equivalent measurements on a series of samples with $250 \leq t_{\mathrm{Cr}} \leq 3000$ Å are summarized in a qualitative phase diagram for $T = 100$ K. Two points at 42 and 80 Å from earlier experiments by Schreyer *et al.*

[10] have been added. The diagram can be divided into four parts. For $t_{Cr} \leq 45$ Å only an $AF_0$ phase with in-plane Cr spins exists. With increasing $t_{Cr}$ up to 250 Å we observe an out-of-plane TSDW with the Cr spins in plane, consistent with results of Fullerton *et al.* [9]. For 250 Å $\leq t_{Cr} \leq$ 1000 Å in- and out-of-plane TSDWs coexist. Finally, for the thickest films, the reorientation to a TSDW propagating in the film plane with spins out of plane is complete. Interestingly, this reorientation is correlated with the occurrence of a coexisting $AF_0$ phase with the same out-of-plane spin orientation.

The observed reorientation effect can be explained by considering a realistic Fe/Cr interface structure. In the Fe/Cr system three different interactions are present: a FM Fe-Fe and an AF Cr-Cr intralayer interaction within each Fe or Cr layer, and in addition an AF [15] interlayer interaction between the Fe and the Cr. At an ideally flat interface all three interactions can coexist without any frustration as long as all moments are oriented in the film plane. However, at real interfaces steps and interdiffusion may occur. Any step height of an uneven number of Cr layers along the Fe/Cr interface introduces frustrations between the Fe and Cr intralayer interaction on one hand, and the interlayer interaction on the other hand [16]. It is not possible to minimize all three coupling energies independently. Thus, the resulting spin structure depends on the values of the respective coupling constants. The following four limiting cases can be distinguished assuming a single monoatomic step at the interface. If the interface coupling is large compared to the Fe or Cr coupling constant, a domain wall forms in the Fe (case 1) or in the Cr (case 2). For a very small interface coupling the ideal FM and AF order in the Fe and Cr layers can be preserved by a domain wall forming along the interface (case 3). If, however, the AF interface coupling is of intermediate magnitude, the system can react by reorienting the Cr moments perpendicular to the Fe (case 4).

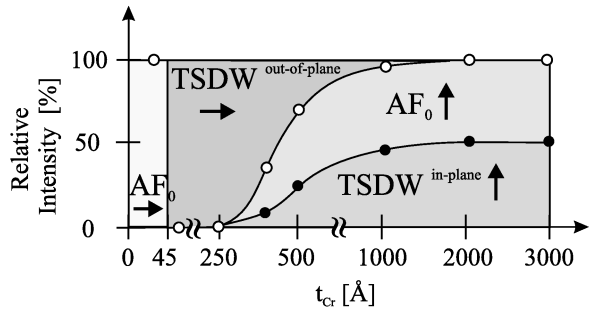


FIG. 2. Qualitative magnetic phase diagram of Cr films capped with 20 Å thin Fe layers as a function of the Cr thickness. The superscripts describe the orientation of the $Q$ vector, the arrows the orientation of the Cr spins. The solid and open circles represent the relative intensities of the TSDW and the $AF_0$ peaks, respectively.

This phenomenological description is confirmed by computer simulations of a classical Heisenberg model with the following Hamiltonian:

$$\mathcal{H} = -\frac{1}{2}\sum_{\langle ij\rangle} J_{e_i e_j}\mathbf{s}_i \cdot \mathbf{s}_j - \sum_i D_{e_i}(s_i^z)^2, \qquad (1)$$

where $\mathbf{s}_i = (s_i^x, s_i^y, s_i^z)$ are spin vectors of unit length at site $i$ on a bcc(001) lattice, and $e_i = \mathrm{Fe}, \mathrm{Cr}$ is the element at this site. $J_{e_i e_j}$ is the nearest-neighbor exchange coupling constant between elements $e_i$ and $e_j$, and $D_{e_i}$ is the uniaxial anisotropy of element $e_i$ which, if positive, favors the $z$ direction perpendicular to the film. We assume that this anisotropy does not depend on the position of the atoms. We choose $J_{\mathrm{FeFe}}/k_B = 375$ K and $J_{\mathrm{CrCr}}/k_B = -170$ K, consistent with the critical temperatures $T^{\mathrm{com}} \approx 450$ K for Cr and $T_c \approx 1000$ K for Fe in mean field solution of this model. The interaction between Fe and Cr is chosen as $J_{\mathrm{FeCr}}/k_B = -40$ K [17]. The shape anisotropy of Fe induced by the dipole interaction is modeled by a negative uniaxial anisotropy of $D_{\mathrm{Fe}}/k_B = -1.5$ K. Finally, we introduce a very small uniaxial anisotropy $D_{\mathrm{Cr}}/k_B = 50$ mK at the Cr sites induced by epitaxial strain, consistent with our results on uncovered Cr films [14]. Using a combination of over-relaxation dynamic and conjugate gradient method, we determine the ground state of this system for various configurations of the Fe/Cr interface. This method is much faster than the tight binding method used by Freyss *et al.* [18], while the magnetic structure obtained is qualitatively the same. To make our model even more realistic we have included interdiffusion at the Fe/Cr interface [5] in addition to well defined steps [19]. In Fig. 3 the resulting ground state spin configuration is shown. Clearly, the frustration induces an effective 90° coupling between the Fe and the Cr order parameter (case 4). Together with the small uniaxial anisotropy of the Cr atoms this leads to an orientation of the Cr

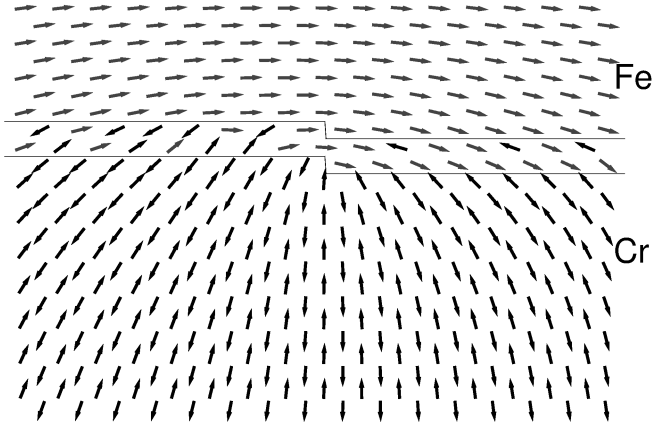


FIG. 3. Ground state spin structure near an Fe/Cr interface with monoatomic steps from computer simulations assuming interdiffusion over two layers. Note that the Fe moments are also affected.

spins perpendicular to the surface, consistent with the experiment.

In our model we find this 90° orientation independent of the presence of interdiffusion as long as there are steps. Two length scales are important, the thickness of the Cr layer and the separation of the steps at the interfaces. When $t_{Cr}$ is reduced below the distance between steps, more energy can be gained by the exchange interaction at the interface than is lost by roughness induced domain wall formation within the Cr. Consequently, for thin Cr films the Cr moments are predicted to be oriented *in* the film plane with domain walls in the Cr layer connecting the interfacial steps. Thus, our model also explains the observed reorientation transition with $t_{Cr}$ (Fig. 2) [20]. For the thinnest Cr films the simulation yields a frustrated spiral structure in the Cr, which induces strong noncollinear coupling between the Fe layers in superlattices as predicted theoretically by Slonczewski [21] and confirmed experimentally by Schreyer *et al.* [10]. Interestingly, $AF_0$ Cr induces such coupling between the Fe layers whereas SDW Cr does not [10,22].

Finally, we discuss the origin of the $AF_0$ phase. For the smallest $t_{Cr}$ well below the SDW period no SDW can form (see Fig. 2). Instead, the system becomes $AF_0$ [9,10] consistent with theory [23]. Thus, the $AF_0$ order is induced by a finite size effect. However, for thick films and in-plane propagation of the SDW, $AF_0$ order also occurs (see Figs. 1 and 2). Grazing incidence x-ray and neutron experiments with depth resolved information have revealed that the in-plane SDW phase is located close to the top Fe/Cr interface [24]. The $AF_0$ phase seems to be limited to the lower Nb/Cr interface of the Fe/Cr/Nb/sapphire structure. The lower quality RHEED data of the Cr near the Nb interface mentioned above indicates small *in-plane* crystalline grain dimensions near the interface due to strain relaxation effects. This can induce $AF_0$ order due to a finite size effect for the in-plane SDW near the Cr/Nb interface. With increasing $t_{Cr}$ the *in-plane* crystalline quality improves according to RHEED, allowing the formation of an in-plane SDW far away from the Cr/Nb interface. On the other hand, for out-of-plane SDW propagation (Fig. 2) no $AF_0$ phase occurs near the Nb/Cr interface, since in this case the limiting factor is the *out-of-plane* crystalline grain size. Using x-ray scattering we measure a much larger out-of-plane coherence length than in the plane.

Consequently, a pure SDW propagating out of plane can form without any $AF_0$ contribution in the $t_{Cr}$ range between 45 and 250 Å. Thus, we can consistently explain the occurrence of the $AF_0$ phase by finite size effects.

In conclusion, we have studied proximity effects between Fe and Cr in the Fe/Cr system with neutron scattering. We have focused on the regime of large $t_{Cr}$ about which no studies exist so far. As opposed to bulk Cr we find a single $Q$ state SDW whose direction of propagation is reoriented from in plane to out of plane upon reducing $t_{Cr}$. Compared to bulk, the spin flip temperature is reduced to about 40 K. The occurrence of commensurate $AF_0$ structures can be attributed to finite size effects. Using ground state calculations of a classical Heisenberg Hamiltonian the observed reorientation transition is explained by a realistic Fe/Cr interface with steps causing frustration of the system.

We thank Professor Usadel for valuable discussions and J. Podschwadek and W. Oswald for technical assistance. The work in Bochum and Duisburg was supported by the Deutsche Forschungsgemeinschaft through SFB 166.

[1] E. Fawcett, Rev. Mod. Phys. **60**, 209 (1988), and references therein.
[2] G. Binasch *et al.*, Phys. Rev. B **39**, 4828 (1989).
[3] M. N. Baibich *et al.*, Phys. Rev. Lett. **61**, 2472 (1988).
[4] M. D. Stiles, Phys. Rev. B **54**, 14 679 (1996).
[5] D. Venus and B. Heinrich, Phys. Rev. B **53**, R1733 (1996); A. Davies *et al.*, Phys. Rev. Lett. **76**, 4175 (1996).
[6] D. Stoeffler and F. Gautier, J. Magn. Magn. Mater. **147**, 260 (1995); A. Vega *et al.*, Europhys. Lett. **31**, 561 (1995); M. Freyss, D. Stoeffler, and H. Dreyssee, Phys. Rev. B **56**, 6047 (1997).
[7] J. Unguris *et al.*, Phys. Rev. Lett. **69**, 1125 (1992).
[8] J. Meersschaut *et al.*, Phys. Rev. Lett. **75**, 1638 (1995).
[9] E. E. Fullerton, S. D. Bader, and J. L. Robertson, Phys. Rev. Lett. **77**, 1382 (1996).
[10] A. Schreyer *et al.*, Phys. Rev. Lett. **79**, 4914 (1997).
[11] S. M. Durbin *et al.*, J. Phys. F **11**, L223 (1981); **12**, L75 (1982).
[12] K. Theis-Bröhl *et al.*, Phys. Rev. B **57**, 4747 (1998).
[13] A. Stierle and H. Zabel, Europhys. Lett. **37**, 365 (1997).
[14] P. Sonntag *et al.*, J. Magn. Magn. Mater. **183**, 5 (1998).
[15] See, e.g., R. Jungblut *et al.*, J. Appl. Phys. **70**, 5923 (1991).
[16] A. Berger and Eric E. Fullerton, J. Magn. Magn. Mater. **165**, 471 (1997).
[17] This value was determined from a preliminary high field magnetization measurement on an Fe/Cr superlattice.
[18] M. Freyss, D. Stoeffler, and H. Dreysse, in MRS Symposia Proceedings, Symposium M, Spring Meeting, San Francisco, 1997 (Materials Research Society, Pittsburgh, 1997).
[19] We use the same $J_{e_i e_j}$ and $D_{e_i}$ for the interdiffused region since deviations make no qualitative difference.
[20] Certain interdiffusion profiles can also cause a 90° orientation between Cr and Fe spins for thick Cr films. However, only steps explain the observed reorientation as a function of Cr thickness. Thus the steps explain *all* observations, interdiffusion without steps does not.
[21] J. C. Slonczewski, J. Magn. Magn. Mater. **150**, 13 (1995).
[22] E. E. Fullerton, C. H. Sowers, and S. D. Bader, Phys. Rev. B **56**, 5468 (1997).
[23] Z. P. Shi and R. S. Fishman, Phys. Rev. Lett. **78**, 1351 (1997).
[24] P. Bödeker *et al.*, Physica (Amsterdam) B (to be published).