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Investigation of magnetic coupling phenomena in $Fe_{1-x}Cr_x$/Cr-superlattices with spin-polarized neutrons

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Abstract

We present the results of temperature dependent measurements of magnetically coupled $Fe_{1-x}Cr_x$/Cr-superlattices. These results are supplementary to the ones known for non-collinearly coupled Fe/Cr-superlattices. By systematically varying the Cr concentration x we cover a wide range of the $Fe_{1-x}Cr_x$-phase diagram. As an experimental technique spin-polarized neutron reflectivity with spin analysis and high-angle neutron scattering proves to be ideal for this work.

Keywords: Magnetic exchange coupling; Curie temperature; Néel temperature; Neutron reflectivity

1. Introduction

Twelve years ago an antiferromagnetic (AFM) interlayer coupling of ferromagnetic (FM) thin films separated by Cr spacer layers was reported [1]. Since then the Fe/Cr system has been of considerable interest in the field of magnetic exchange coupling in metallic superlattices. After the collinear (C) AFM coupling, the non-collinear (NC) coupling was discovered [2,3]. It was pointed out that the magnetic structure of the Cr-spacer layer, known to be an incommensurate AFM spin density wave in the bulk [4], should play an important role in explaining the magnetic coupling in the Fe/Cr-system [5,6]. It was, however, only recently that by use of high angle neutron scattering and polarized neutron reflectometry (PNR) experiments the NC coupling in Fe/Cr-superlattices could be related with the spin structure of the Cr spacer layers [7,8] In agreement with the proximity magnetism model for NC coupled multilayers [9] it was demonstrated that the NC coupling in Fe/Cr-superlattices originates from a frustrated spiral modulation of the Cr moments [7]. Temperature dependent measurements showed that for the Cr Néel temperature $T_{N,\,Cr} = 500$ K, the NC interlayer coupling vanished. To get a deeper insight into this phenomenon we changed those system parameters which are of importance for the magnetic coupling. One way to do this is to exchange the Fe layers by an

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$Fe_{1-x}Cr_x$ alloy. This alloy is FM for a wide range of x and its bulk phase diagram is well established [10]. In this way, the influence of the $Fe_{1-x}Cr_x$ FM intralayer order on the Cr AFM intralayer order and vice versa can be studied systematically. With increasing x, both the average magnetic moment per atom and, of even more interest, the Curie temperature $T_{C,\,FeCr}$, will be shifted to lower values. By varying $T_{C,\,FeCr}$, the effect on $T_{N,\,Cr}$ can be studied. $T_{C,\,FeCr}$ can be selected to be lower or even higher than $T_{N,\,Cr}$. By varying the sample temperature for a given x one may "switch off" or "on" the coupling by either crossing $T_{C,\,FeCr}$ or $T_{N,\,Cr}$.

2. Samples

A set of $[Cr/Fe_{1-x}Cr_x]_N/Cr/Nb/Al_2O_3$ $(1\,\bar{1}\,0\,2)$-superlattices with different x were prepared using molecular beam epitaxy. All samples were grown under the same conditions [7] since structural properties may influence the coupling mechanism [9,11–13]. Table 1 summarizes the samples and their compositions. The $Fe_{1-x}Cr_x$ film thicknesses t_{FeCr} lie between 24 and 31 Å. The Cr spacer layer thickness t_{Cr} was chosen to be smaller than 45 Å, since in this case the Cr AF structure is known to be commensurate below $T_{N,\,Cr}$ in Fe/Cr samples, favouring magnetic coupling [7]. The large number of superlattice periods $N \geqslant 60$ increases the amount of Cr, which yields a better signal to background ratio in neutron scattering experiments. X and the thicknesses t_{Cr}, t_{FeCr} were determined by combining the results of energy dispersive X-ray analysis and superlattice periods Λ, measured with neutron reflectivity.

3. Experimental

Polarized neutron scattering techniques are ideally suited for the nondestructive study of the magnetic profile in thin metallic films [14–16] . By means of PNR with spin analysis (SA) we determined the in-plane vector magnetization profile [17,18] of the $Fe_{1-x}Cr_x$ layers along the axis perpendicular to the sample surface. Λ was also determined by neutron reflectometry since the X-ray scattering contrast between the FeCr-alloy and the Cr was not sufficiently large to result in reasonable X-ray reflectivity spectra. With temperature dependent PNR with SA, we obtained the transition temperatures $T_{C,\,FeCr}$. All neutron reflectivity experiments were carried out on the high flux neutron reflectometer ADAM at the ILL [19]. A first sample was studied with high angle neutron scattering to access the spin structure of the Cr spacer layers on the triple axis spectrometer IN-12 at ILL. A single purely magnetic peak of the commensurate AF structure of the BCC-Cr is studied around $\{0\,0\,1\}$ positions [4,7].

4. Results

The results obtained by means of PNR with SA are summarized in Table 1. No NC coupling was

Table 1
List of $[Cr/Fe_{1-x}Cr_x]_N/Cr/Nb/(1\,\bar{1}\,0\,2)Al_2O_3$ samples with systematic variation of the Cr concentration x. The samples were grown under equivalent conditions by molecular beam epitaxy. The spacer layer thicknesses, $t_{Cr} < 45$ Å, was chosen to assure a commensurate AF spin structure. The large number of superlattice periods increases the amount of Cr which improves the signal to background ratio in neutron scattering experiments. The measured Curie temperatures for samples with higher concentrations are considerably lower than the bulk values

x (at% Cr)	t_{Cr} (Å)	t_{FeCr} (Å)	Superlattice period N	Coupling type at $T = 20$ K	Bulk $T_{C,\,FeCr}$ (K) literature [10]	$T_{C,\,FeCr}$ (K) measured
38	28	31	100	AFM	820	–
45	40	33	100	FM	760	770
57	28	24	60	AFM	490	450
66	36	27	190	FM	325	210
70	22	26	60	FM	250	185
80	21	25	60	(Spin glass)	55	–

observed at low temperatures, $T \leqslant 20$ K. This was an unexpected result for the $Fe_{1-x}Cr_x/Cr$ samples suggesting that their interface topology is different to that of the Fe/Cr samples [7,12]. For $x = 0.8$ no remanent magnetization of the Fe_xCr_{1-x} layer was found, consistent with spin glass behaviour. Bulk Fe–Cr alloys exhibit such spin glass character between 81 at% Cr and 84 at% Cr [10] below about 25 K. Above this temperature the bulk alloy becomes paramagnetic. A comparison of the measured Curie temperatures with those of the bulk shows that there is a considerable reduction of $T_{C,\,FeCr}$ for higher Cr concentrations. $T_{C,\,FeCr}$ with $x = 0.38$ was experimentally not accessible with our cryo-furnace. Inferring from the continuous change in the transition temperatures, $T_{C,\,FeCr}$ for this sample should be comparable to the bulk. The AFM coupled samples show a change in the coupling type with increasing temperature. Fig. 1 shows three of the four reflectivity cross R^{++}, R^{--} and $R^{+-} = R^{-+}$. A clear chemical first-order Bragg peak (R^{++}, R^{--}) and a purely magnetic half order Bragg peak (R^{-+}) at $T = 35$ K are observed. This result and the fact that there was no intensity splitting of R^{++} and R^{--} at the first order peak, indicates an AFM alignment perpendicular to the neutron polarisation axis. While the temperature was increased, PNR radial scans with SA of both peaks were taken in order to record the temperature dependence of the magnetization vector profile. The half order peak intensity in R^{-+} diminished continuously, vanishing at $T = 380$ K, whereas a splitting in the first order intensities appeared as shown by the PNR data in the inset of Fig. 1. Thus a rotational transition from AFM towards FM takes place. Beyond this point there is only a decrease in the magnetic net moment up to $T_{C,\,FeCr} = 450$ K. For the FM sample with

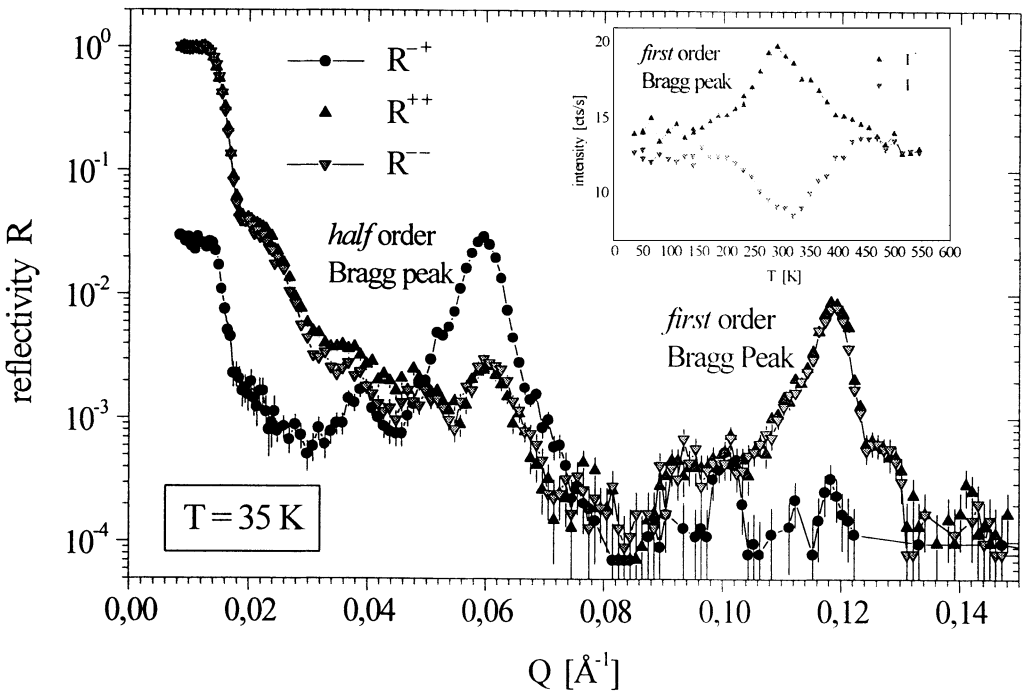

Fig. 1. Reflectivity measurement with spin-polarized neutrons and spin analysis at $T = 35$ K for a $Fe_{0.43}Cr_{0.57}/Cr$ sample. The half order Bragg peak in $R^{+-}(= R^{-+})$ and no intensity splitting at the chemical first order Bragg peak position in R^{++} and R^{--} indicate an AFM coupling perpendicular to the neutron polarisation axis. The high intensity in R^{++} and R^{--} at the half order position is related to a high diffuse magnetic scattering which was confirmed with off specular scans. The inset documents the magnetic reorientation from AFM to FM by its temperature-dependent splitting. For more details see text.

$x = 0.66$, $T_{C,\,FeCr}$ was found to be 180 K. A first high angle neutron scattering experiment yields a Cr Néel temperature $T_{N,\,Cr} = 360$ K for this sample which is about 30% smaller than in equivalent Fe/Cr-samples [7].

5. Conclusions

In a set of $Fe_{1-x}Cr_x$/Cr-superlattices with a systematic variation of x we found reduced Curie temperatures $T_{C,\,FeCr}$ compared to bulk FeCr-alloys. Instead of an expected NC coupling all the samples couple either FM or AFM. For the AFM samples a continuous magnetic reorientation into the FM state takes place with increasing temperature. $T_{N,\,Cr}$, which we have determined for one sample so far, is also considerably lower than in the case of Fe/Cr-superlattices. Thus, $T_{N,\,Cr}$ can be varied via $T_{C,\,FeCr}$, indicating a proximity effect between the FM Fe_xCr_{1-x} and the AF Cr layers. We are currently investigating this proximity effect systematically as a function of alloy concentration x.

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