TEMPERATURE-DEPENDENT STUDY OF ION-CHANNELING IN Fe/Cr SUPERLATTICES*

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Giant-Magneto-Resistance (GMR), as large as 150% at 4K, occurs in Fe/Cr superlattices as a result of antiferromagnetic interlayer coupling. We have successfully grown epitaxial single-crystal Fe/Cr multilayers using magnetron sputtering. Ion channeling was employed to study the structural and vibrational properties of the sputter-deposited Fe/Cr superlattices, and of a Cr thin film, between temperatures of 100 and 330K. Channeling in the latter specimen was used to investigate the importance of depositing a Cr buffer-layer in order to obtain superlattices with large GMR values. Once the buffer layer exceeded a critical thickness, a high quality Cr film was observed. The epitaxial quality of the superlattices grown on such buffer layers by sputtering was found to be excellent. Minimum yields nearly equal to theoretical predictions were found for channeling along the $\langle 001 \rangle$ growth direction; slightly higher values were found along the $\langle 111 \rangle$ axis. Because of the high structural quality of the sputter-deposited films, it was possible to investigate changes in thermal vibration amplitudes, even though their magnitude is only of the order of a few pm (10^{-12} m). No unusual structural changes of this magnitude were observed in angular channeling scans obtained while cooling the Fe/Cr superlattice from 330 down to 100K.

1. Introduction

Because they exhibit Giant-Magneto-Resistance (GMR) [1], i.e. a strong dependence of the resistance on the applied magnetic field, as well as exotic magnetic coupling phenomena [2], Fe/Cr superlattices have attracted considerable attention. Magnetoresistance loops measured [3] at three different temperatures for a 50-period (100)-oriented $[\text{Fe}(14\text{\AA})/\text{Cr}(8\text{\AA})]$ superlattice prepared by magnetron sputtering are displayed in Fig. 1. The most striking feature is the magnitude of the GMR: 150% at 4.2K and 30% at room temperature.

Experiment and theory have shown that the nature of the coupling of the Fe layers in these superlattices is strongly dependent upon the Cr layer thickness [4]. With increasing thickness of the Cr layer the coupling of the Fe layers oscillates between ferromagnetic and antiferromagnetic, exhibiting a period of $18\text{\AA}$. The amplitude of the oscillation decreases with increasing Cr thickness. The oscillatory coupling is attributed to the formation of a

static spin density wave in the Cr layer, or equivalently to spin polarized quantum well states confined to the Cr layers. Bulk Cr is antiferromagnetic below its Néel-temperature and paramagnetic above. The Néel-temperature for a given superlattice depends on the Cr-layer thickness, dropping to zero if the thickness is less than 42Å. In the thickness regime that exhibits pronounced GMR behavior, the Cr does not exhibit a Néel temperature. When the Cr-thickness is greater, the Fe-layers decouple below the Néel-temperature.

High crystalline quality is correlated with a high GMR. Through the use of a Cr buffer layer, it has become possible to produce [3] high-quality single-crystalline films using sputter deposition. The quality of these sputtered films is comparable with those produced by molecular beam epitaxy.

In the present work, we first used ion channeling [5] to understand the role of the initially deposited buffer layer in producing high-quality superlattices by magnetron sputtering. For sufficient Cr-layer thicknesses, channeling measurements along the (100) growth direction revealed excellent crystalline quality, allowing changes in thermal vibrational amplitudes to be investigated as a function of temperature. Slightly poorer crystalline quality was found along the (111) directions, although it was still possible to observe changes in vibration amplitudes. Next we investigated channeling behavior in an Fe/Cr superlattice where again, quite high crystalline quality was observed, particularly along the (100) growth axis. We therefore employed axial channeling scans in the superlattice to search for evidence of either temperature-dependent lattice strain, or a subtle periodic lattice distortion accompanying the spin-density wave that communicates the coupling information.

2. Experimental Procedures

A few (100)-oriented Cr films of varying thicknesses, and an [Fe(14Å)/Cr(62Å)] superlattice containing fourteen periods, were grown by d.c. magnetron sputtering onto epitaxially polished MgO(100) substrates. A 100 Å Cr buffer layer was initially deposited at a substrate temperature of 600 °C to establish the epitaxial orientation with the substrate and provide a template for the growth of the superlattice. The MgO (100)-surface ($a_0 = 4.213$ Å) has a 2.98 Å square surface net which is only 3.3% lattice mismatched with the Cr (100)-

surface upon a 45° rotation about the surface normal (i.e., Cr[010]/MgO[011]). The substrate was subsequently cooled to 190°C , where the superlattice was grown by sequential sputter deposition of Fe and Cr layers [3].

This procedure results in the growth of epitaxial Fe/Cr(100) superlattices which exhibit the expected $18\text{ \AA}$ long-period oscillatory interlayer coupling and magnetoresistance values as large as 150% at a temperature of 4K (as shown in Fig. 1). Néel transition temperatures are observed for relatively thick Cr spacer layers. For Cr-spacer thicknesses greater than $42\text{ \AA}$, the Néel temperature first rises rapidly with increasing Cr thickness, and then asymptotically approaches its bulk value.

Ion channeling in combination with Rutherford Backscattering Spectrometry (RBS) [5,6] was conducted using a well-collimated (divergence $<0.05^\circ$) beam of 1.5 MeV He^+ ions. The target holder, which was mounted on a precision double-axis goniometer having an angular resolution of 0.01° , could be cooled to 100K using a closed-cycle refrigeration system. Backscattered α particles were detected by a Si-surface-barrier detector [full width at half maximum (FWHM) of 16 keV] placed at a scattering angle of 138° with respect to the incident beam.

3. Results and Discussion

Three kinds of channeling measurements have been performed on these samples: axial channeling- to determine the epitaxial quality; angular dependence of axial channeling at several temperatures- to determine the thermal vibration amplitudes and search for any structural or phase changes; and Resonance Planar Dechanneling (RPD)- to determine the strain. In the present paper we focus on the first two types of measurements. For RPD it is necessary to produce a special sample where the thickness of the layer is matched to the wavelength of the beam [7].

Angular scans were performed by changing the angle of incidence, ψ , between the beam and the sample normal. The minimum yield and the half-width at half-maximum (FWHM) of an angular scan, also called the critical angle, $\psi_{1/2}$, for channeling provide information on the crystalline quality and, when done as a function of temperature, the thermal vibration amplitudes of the atoms. These two quantities can be calculated [5,6] as outlined below.

The minimum yield for axial channeling can be estimated from the following relationship:

$$\chi_{\min} = 18.8 N d u_1^2 (1 + \zeta^{-2})^{1/2} \quad (1)$$

Here N is the number of crystal atoms per unit volume (in cubic-angstroms), d the atomic spacing along the rows (in angstroms), u_1 the one-dimensional rms of the thermal vibration amplitude (in angstroms) and:

$$\zeta = 126 u_1 / (\psi_{1/2} d) \quad (2)$$

where $\psi_{1/2}$ is given in degrees:

$$\psi_{1/2} = 0.80 F_{RS} (1.2 u_1 / a) \psi_1 \quad (3)$$

$$\psi_1 = 0.307 \sqrt{\frac{Z_1 Z_2}{d E}} \quad (4)$$

E is the energy of the incident ion in MeV, and a is the Thomas-Fermi screening radius in angstroms:

$$a = 0.4685 \left(\sqrt{Z_1} + \sqrt{Z_2} \right)^{-2/3} \quad (5)$$

if the incident ion is not fully ionized:

$$a = 0.4685 Z_2^{-1/3} \quad (6)$$

if the incident ion is fully ionized. The function F_{RS} is defined as:

$$F_{RS}(\xi) = \sqrt{f_{RS}} = \sqrt{\int_0^\infty (\xi^2 + \eta^2)^{-1/2} \varphi[(\xi^2 + \eta^2)^{1/2}] d\eta} \quad (7)$$

where φ is the Molière potential (x in angstroms):

$$\varphi(x) = 0.1e^{-6x} + 0.55e^{-1.2x} + 0.35e^{-0.3x} \quad (8)$$

Applying the Debye-Theory for the lattice vibration:

$$u_1 = 12.1 \sqrt{\{\Phi(x)/x + 0.5\}/\Theta M_2} \text{ \AA} \quad (9)$$

x is given by $x=\Theta/T$, with the Debye-temperature, Θ , and the temperature, T , of the target. M_2 is the mass of the target and $\Phi(x)$ is the Debye-function defined by:

$$\Phi(x) = \frac{1}{x} \int_0^x \frac{\eta}{e^\eta - 1} d\eta \quad (10)$$

From this, a nearly linear dependence for the FWHM and minimum yield is expected on temperature within the range from 100 to 350K. Calculated values for the various quantities defined in Eqs. 1-10 for pure Cr are listed in Table I.

RBS axial channeling scans obtained at room temperature with 1.5-MeV He incident along the $\langle 001 \rangle$ and $\langle 111 \rangle$ directions of a pure chromium film grown on an $\langle 001 \rangle$ MgO substrate are displayed along with scans obtained along a random direction in Figs. 2 and 3, respectively. The minimum yield for the $\langle 001 \rangle$ direction, 2.2% just behind the Cr leading edge, is the same as that expected, 2.2% (cf. Table I), on the basis of the equations presented above using a Debye temperature of 485 K [6] for Cr.

Along the $\langle 111 \rangle$ direction a slightly poorer match between experiment, 2.5%, and theory, 2.1%, is observed. The dechanneling rate, i.e., the increase in RBS yield with depth is also significantly greater in the $\langle 111 \rangle$ compared to the $\langle 100 \rangle$ directions. These results both demonstrate that the epitaxial quality of the sputtered-deposited films is higher along the growth direction than perpendicular to it, probably due to dislocations which are generated in response to the 3.3% lattice mismatch described above.

The channeling behavior observed as a function of depth helps clarify the importance of the buffer layer in producing high quality Fe/Cr superlattices. A significant amount of disorder is clearly evident near the back edge of the Cr film, i.e., that closest to the MgO substrate, in both the $\langle 001 \rangle$ and $\langle 111 \rangle$ channeling spectra. Because of the significantly lower dechanneling rate, the thickness of this disordered layer appears better defined in the $\langle 001 \rangle$ channeling spectrum. The total thickness of this Cr film as determined from the RBS spectrum is 310 nm. From Fig. 2, then, the thickness of this initial disordered layer can be estimated to be on the order of a few tens of nm's. Once

the buffer layer exceeds a critical thickness, however, the disorder disappears and a high quality Cr film is produced.

Because of the high crystalline quality of the Cr films, as evidenced by the channeling spectra in Figs. 1 and 2, angular scans were taken at several temperatures between 150 and 330K. Three of those measured along the $\langle 001 \rangle$ growth axis are reproduced in Fig. 4; three obtained along the $\langle 111 \rangle$ direction in Fig. 5. The (100) and (111) scans were accumulated using the counts in channel numbers 330-370 and 328-368, respectively. Critical angles and minimum yields obtained from these and the other scans are compiled in Table II. In general, the minimum yield measured experimentally decreases with decreasing temperature, and the angular width (critical angle) increases, as expected. We note that because of the fewer number of counts involved, the minimum yield values listed in Table II have larger error bars associated with them than do the critical angles. This general decrease in minimum yield and increase in critical angle reflect the fact that the atomic rows are becoming smoother [5,6], because of a reduction in thermal vibration amplitude with decreasing temperature.

We now turn to the channeling measurements made on the [Fe(14Å)/Cr(62Å)] superlattice containing fourteen periods. RBS axial channeling scans obtained at room temperature with 1.5-MeV He incident along the $\langle 001 \rangle$ and $\langle 111 \rangle$ directions of this specimen are displayed along with scans obtained along a random direction in Figs 6 and 7, respectively. Because the layers are thin, individual layers cannot be identified. Again, high crystalline quality is observed along the (100) growth axis, with some degradation along the (111) direction. The (100) minimum yield is $\leq 5\%$, while the (111) value is below 10%.

The temperature dependent angular scans for the Cr film demonstrated that small ($\sim$ pm) structural changes can be observed using ion channeling. We therefore decided to perform similar measurements on the superlattice, in order to search for any such changes that might be responsible for the strong temperature dependence (Fig. 1) observed in the magnetoresistance.

Angular scans from the superlattice were therefore taken at several temperatures between 150 and 300K. Three of those measured (using channels 345-375) along the $\langle 001 \rangle$ growth axis are reproduced in Fig. 8; three obtained (using channels 328-368) along the $\langle 111 \rangle$ direction in Fig. 9. Critical angles and minimum yields obtained from these and the other scans are displayed in Figs. 10 and 11, respectively. Only the expected slight increase in critical angle

(Fig. 10), and decrease in the minimum yield (Fig. 11), is observed with decreasing temperature for both the (100) and (111) directions. Hence, no unusual structural changes were observed while cooling the Fe/Cr superlattice from 330 down to 100K.

4. Conclusion

Epitaxial single-crystal Fe/Cr multilayers were grown using magnetron sputtering. Ion channeling was employed to study the structural and vibrational properties of the sputter-deposited Fe/Cr superlattices, and a Cr-only thin film, between temperatures of 100 and 330K. The channeling behavior observed as a function of depth in the latter specimen clarified the importance of the buffer layer in producing high quality Fe/Cr superlattices. A significant amount of disorder was clearly evident near the back edge of the Cr film, i.e., that closest to the MgO substrate. The thickness of this initial disordered layer was estimated to be on the order of a few tens of nm's. However, once the buffer layer exceeds a critical thickness, the disorder disappears and a very high quality Cr film is produced.

The epitaxial quality of the superlattices grown on such buffer layers by sputtering was also found to be excellent. Minimum yields nearly equal to theoretical predictions were measured for channeling along the $\langle 001 \rangle$ growth direction, slightly higher values for the $\langle 111 \rangle$ axis. Because of the high structural quality of the sputter-deposited films, it was possible to investigate structural changes on the order of a few pm (10^{-12} m). No unusual structural changes were observed while cooling the Fe/Cr superlattice from 330 down to 100K.

5. Acknowledgement

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7. Figure Captions

- Fig. 1. Magnetoresistance loops measured at three different temperatures for a (100)-oriented [Fe(14Å)/Cr(8Å)] superlattice prepared by magnetron sputtering.
- Fig. 2. Random and (100)-channeling RBS spectra from a 3100Å thick Cr film. The growth axis is also (100).
- Fig. 3 Random and (111)-channeling RBS spectra from the same film used to obtain the data in Fig. 1.
- Fig. 4. Angular scans taken about the (100) axis of the 3100Å thick Cr film at temperatures of 150, 200 and 330 K using the counts in channels 330-370.
- Fig. 5. Angular scans taken about the (111) axis of the 3100Å thick Cr film at temperatures of 150, 200 and 330 K using channels 328 - 368.
- Fig. 6 Random and (100)-channeling RBS spectra from a Fe(14Å)/Cr(62Å) superlattice containing fourteen periods.

- Fig. 7 Random and (100)-channeling RBS spectra from a Fe(14Å)/Cr(62Å) superlattice containing fourteen periods.
- Fig. 8. Angular scans taken about the (100) axis of the [Fe(14Å)/Cr(62Å) superlattice at temperatures of 150, 200 and 330 K using the counts in channels 345 - 375.
- Fig. 9. Angular scans taken about the (111) axis of the Fe/Cr superlattice at temperatures of 150, 200 and 330 K using the counts in channels 348 - 374.
- Fig. 10. Angular widths for the (100) and (111) scans from the Fe(14Å)/Cr(62Å) superlattice as a function of temperature.
- Fig. 11. Minimum yields for the (100) and (111) scans from the Fe(14Å)/Cr(62Å) superlattice as a function of temperature.

Table I Calculated Channeling Quantities for Cr

T(K)	330	300	250	200	150	100
x	1.470	1.617	1.940	2.425	3.233	4.850
$\Phi(x)$	.6913	.6666	.6159	.5484	.4562	.3297
u_1 (Å)	.0646	.0620	.0574	.0526	.0476	.0430
ξ	.5652	.5425	.5022	.4602	.4165	.3762
FRS(ξ)	1.012	1.020	1.035	1.051	1.068	1.083
$\Psi_{1/2}^{100}(0)$	.828	.834	.847	.860	.874	.886
$\Psi_{1/2}^{111}(0)$	.890	.897	.910	.924	.939	.952
$\chi_{\min}^{100}$	.024	.023	.020	.017	.015	.012
$\chi_{\min}^{111}$	.021	.020	.017	.015	.012	.010

Table II Measured Channeling Quantities for Cr

T(K)	330	300	250	200	150	100
$\Psi_{1/2}^{100}(0)$	0.98	0.99	1.03	1.05	1.10	-
$\Psi_{1/2}^{111}(0)$	1.02	1.06	1.07	1.14	1.16	-
$\chi_{\min}^{100}$	.023	.023	.022	.021	.022	-
$\chi_{\min}^{111}$	.050	.048	.046	.042	.037	-

Fig. 2
950726 001 Cr Ran & Chn

Fig. 3
950725 111 Cr Ran & Chn

Fig. 4
950726 Cr 001 Axial Scans
Channel 330-370

Fig. 5
950725 Cr 111 Axial Scans
Channel 328-368

Fig. 6
950727 001 FeCr Ran & Chn

Fig. 7
950728 111 FeCr: Ran & Chn

Fig. 8
950727 FeCr 001 Axial Scans
Channel 345-375

⊕

Fig. 9
FR950728 FeCr 111 Axial Scans
Channels 348-374

Fig. 10 (Frank's Fig. 12)
Fe/Cr [14A/62A] (Figure 12)

Fig. 11
Fe/Cr [14A/62A] $\langle 111 \rangle$