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Influence of strain on the magnetic properties of thin Cr(001) films

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We report on the influence of strain on the magnetic properties of thin Cr(001) layers in the Fe/Cr/Fe(001) trilayers. The epitaxial strain in the Cr layer is measured via channeling Rutherford backscattering spectroscopy and x-ray diffraction spectroscopy. The magnetic properties of the Cr spacer layer are probed via perturbed angular correlation spectroscopy on the ^{111}In(^{111}Cd) radioactive probe. The results indicate that the strain in the Cr spacer layer influences its Néel temperature, the orientation of the magnetic moments, and the strength of the magnetic hyperfine field on the diamagnetic ^{111}Cd probe.

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I. INTRODUCTION

The antiferromagnetic properties of bulk Cr were very intensely studied in the past. As is comprehensively reviewed in Ref. 1, it is found that the itinerant antiferromagnetism in the bulk chromium is dramatically influenced by external factors such as temperature, pressure, strain, and doping with impurities. More recently, the interest in the magnetic properties of chromium has revived since its use in Fe/Cr multilayers have lead to the discovery of interlayer exchange coupling and the giant magnetoresistance effect.[2,3] The interlayer exchange coupling determines the orientation of the magnetization of the two ferromagnetic layers. In addition to the bilinear interlayer exchange coupling, which ferro- or antiferromagnetically orients the magnetization in the Fe layers with respect to each other, a biquadratic term has also been observed.[4] This biquadratic term tends to orthogonally align the magnetization of the Fe layers to each other and dominates the interlayer exchange coupling for moderately thick ($t_{\mathrm{Cr}} \approx 4$ nm) Cr spacers.

For very thick Cr spacers ($t_{\mathrm{Cr}} > 5$ nm), it was found that the magnetic ordering in the Cr spacer suppresses the biquadratic interlayer exchange coupling.[5–7] The magnetic ordering temperature for thick Cr spacers is, thus, directly related to the onset temperature of the biquadratic coupling between the Fe layers in the Fe/Cr multilayers. As a consequence, it is of fundamental importance to understand the various factors that may influence the magnetic properties of Cr spacer layers.

Apart from the finite size effects that influence the Néel temperature of Cr,[5,6] the residual strain in thin Cr films may also shift the Néel temperature. Early studies on relatively thick Cr films show that the substrate can induce either a positive or a negative tetragonal distortion in the Cr film, which induces an enhancement or a reduction in the Néel temperature, respectively.[8,9] To our knowledge, however, no detailed and systematic study exists that deals with the influence of strain on the magnetic properties of the Cr spacer in Cr-based multilayers.

In this paper, the influence of the strain on the magnetic properties of thin Cr films is investigated. The presence of strain does not only shift the Néel temperature of the Cr spacer but also determines the orientation and the strength of the magnetic hyperfine field in the Cr spacer.

In Sec. II, we will study the structural properties of two series of Fe/Cr/Fe(001) trilayers and, thereby, illustrate the dependence of the tetragonal distortion in the Cr layer on the growth temperature. Subsequently, we will address the magnetic properties of the Cr spacer. At the end, we will discuss the influence of the strain on the magnetic properties of the Cr spacer.

II. STRUCTURAL PROPERTIES

We studied two series of Fe/Cr/Fe(001) trilayers, which were epitaxially grown on MgO(001) substrates with the use of molecular beam epitaxy (base pressure, 2×10^{-11} Torr). The Cr spacer thickness was kept constant at 8 nm and the two series differ only in the thickness of the bottom Fe layer. In the case of series I, the bottom Fe layer had a thickness of 4 nm while, in the case of series II, the thickness of the bottom Fe layer was 10 nm. A thin capping layer was deposited at room temperature to protect the sample against oxidation. The growth temperature was varied between 20 and 300 °C. The epitaxy was monitored by using *in situ* reflection high-energy electron diffraction (RHEED). Irrespective of the growth temperature, the Fe/Cr/Fe(001) trilayers always showed good epitaxial growth, as inferred from RHEED and x-ray diffraction (XRD) experiments. The good structural quality of the Fe/Cr/Fe(001) trilayers was confirmed by Rutherford backscattering spectroscopy (RBS) experiments.

The tetragonal distortion ($\epsilon^T = \epsilon^{\parallel} - \epsilon^{\perp}$) for the Fe and Cr layers was determined by measuring the precise channeling direction of 3 MeV He^{++} ions along the [111] crystal axis by using Rutherford backscattering spectroscopy (RBS-C).[10] From the angle between the [001] and [111] crystal axes, the tetragonal distortion in the Fe and Cr layers could be separately determined. In Fig. 1, the tetragonal distortion of the Cr spacer and the Fe layers is plotted as a function of the growth temperature for several Fe/Cr/Fe trilayers with a bottom Fe layer thickness of 4 (series I) and 10 nm (series II). The tetragonal distortion that was deduced via complementary XRD measurements is also shown in Fig. 1 and it is in line with the tetragonal distortion, as determined via RBS-C measurements.

From Fig. 1(a), one readily deduces that the growth temperature dramatically influences the tetragonal distortion in

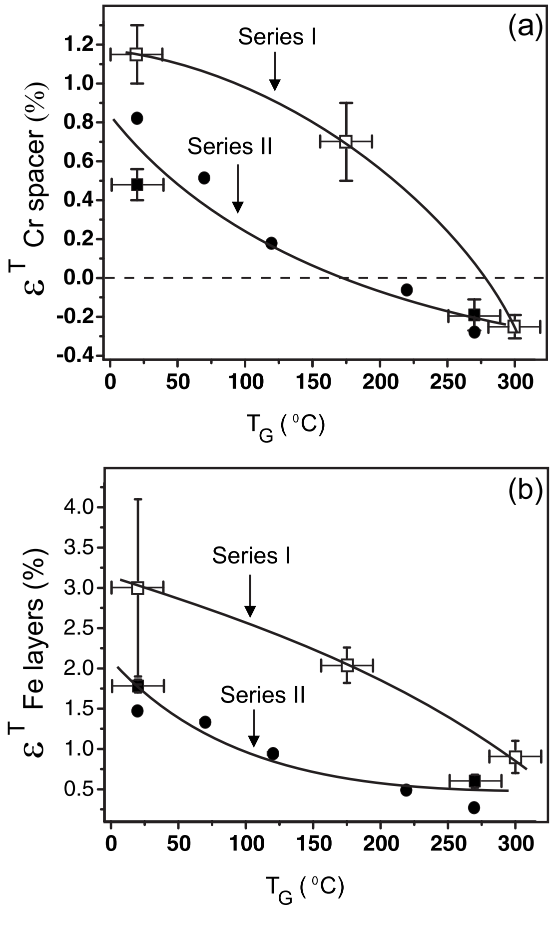

FIG. 1. Tetragonal distortion in the Cr spacer (a) and the Fe layers (b) as a function of the growth temperature. The results for series I are shown as open symbols (RBS-C: □). The results for series II are shown as filled symbols (RBS-C: ■, XRD: ●). The solid lines are a guide to the eye.

the Cr layers. The tetragonal distortion decreases with increasing growth temperature and changes sign around 200 °C. This means that for a high enough growth temperature, the in-plane lattice of the Cr spacer is compressed, whereas it is expanded at low growth temperatures. Furthermore, through the comparison of the results for the two series in Fig. 1(a), it is deduced that the Cr layers have a higher tetragonal distortion if grown on a bottom Fe layer of 4 nm than if they are grown on a 10 nm thick Fe base layer. The difference is especially large for the Fe/Cr/Fe trilayers grown at low and intermediate growth temperatures (20–170 °C).

The tetragonal distortion in the Cr spacer is connected to the tetragonal distortion of the Fe layers since the Cr layer pseudomorphically grows on the bottom Fe layer. In Fig. 1(b), the tetragonal distortion in the Fe layers is shown. The tetragonal distortion of the Fe layers decreases as a function of the growth temperature and the thickness of the bottom Fe layer. It is attributed to the grain size of the Fe layers. RHEED measurements indicated an island type of growth for Fe on MgO at low temperatures (20–70 °C) and a layer-by-layer growth at elevated temperatures, which is confirmed in Ref. 11. We also did observe this trend in the present work. More specifically, the XRD rocking curve measurements around the diffraction at $2\theta = 64.8°$ indicate an increase in the grain size of the Fe layers when the growth temperature of the Fe/Cr/Fe trilayer is increased.

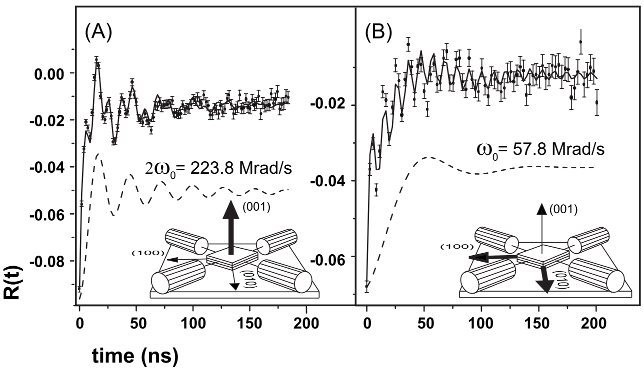

FIG. 2. PAC time spectra for the samples grown at a low (a) and at a high (b) temperature. The contribution in the PAC time spectra from the ^{111}Cd probes in the Cr layer is explicitly shown as a dashed line. ω_0 is the characteristic Larmor precession frequency for the Cr contribution.

A possible explanation for the observed dependence of the tetragonal distortion in the thin films as a function of the growth temperature is the formation of misfit dislocations at the interface between the substrate and the first Fe layer. Misfit dislocations are formed when the strain energy in the layer reaches the energy needed to form a misfit dislocation.[12] The strain energy in the Fe layer is increased when the volume of the grains increases, and consequently, an increase in the grain size will result in the formation of misfit dislocations and a decrease in the strain and the tetragonal distortion in the Fe layer. This correlates well with the observed dependence of the tetragonal distortion and the grain size on the growth temperature. In a similar way, the decrease in the tetragonal distortion for thicker Fe films can be explained. When the thickness of the Fe film increases, the grain size increases, and consequently, more misfit dislocations can be formed in the grain at the Fe/MgO interface, which results in a lower strain and a tetragonal distortion.[13]

Most importantly for the present work, we have shown that the tetragonal distortion of the Cr spacer very strongly depends on the growth temperature and the thickness of the bottom Fe layer.

III. PERTURBED ANGULAR CORRELATION SPECTROSCOPY

The magnetic properties of the Cr spacer were studied by using perturbed angular correlation (PAC) spectroscopy. For that purpose, ^{111}In(^{111}Cd) probes were implanted into the Fe/Cr/Fe trilayer. Implantation energies between 60 and 80 keV were selected such that the radioactive probes end up in the middle of the Cr spacer. The data were acquired by using a high-resolution fast-slow coincidence four-detector setup. For details on the method, we refer to our previous publications.[5,14]

Two representative PAC spectra are shown in Fig. 2. In panel (A), the PAC spectrum for an Fe/Cr/Fe trilayer grown at a low temperature is shown, while in panel (B), the PAC spectrum for an Fe/Cr/Fe trilayer grown at a high temperature is shown. In PAC, one probes the local electric and magnetic environments at the ^{111}Cd nuclei via the Larmor precession (ω) of the nuclear spin induced by the hyperfine field. Since the radioactive atoms end up in substitutional lattice positions in both the Fe and the Cr layers, the PAC time spectra are composed of two contributions. The contribution to the PAC time spectra that corresponds to the well-known hyperfine field for ^{111}Cd probes in Fe is recognized in the spectra as a fast oscillation. The contribution to the PAC time spectra that corresponds to ^{111}Cd probes in the Cr layer is explicitly shown as dashed lines in Fig. 2.

Depending on the orientation of the magnetic hyperfine fields with respect to the detectors, the single or double harmonic of the Larmor precession is observed. In panel (A) of Fig. 2, the second harmonic of the Larmor precession frequency ($2\omega_0$) for the Cr contribution is observed; while in panel (B), the first harmonic (ω_0) is observed. From this observation, we can derive that the magnetic hyperfine field in Cr is oriented out of plane (along the growth direction) for the sample deposited at a low temperature (panel A) and in plane for the sample deposited at a high temperature (panel B).

From the value of the characteristic frequency for the Cr contribution, we can calculate the magnetic hyperfine field for ^{111}Cd in Cr, which gives ($\omega_0 = 111.9$ Mrad/s) 7.62 T for the spectrum in panel (A) and ($\omega_0 = 57.8$ Mrad/s) 3.94 T for the spectrum in panel (B).

The above illustrates the sensitivity of the PAC technique to the value and the orientation of the hyperfine field in the Cr layer of an Fe/Cr multilayer. We will now discuss the temperature dependence of the magnetic hyperfine field and the relation between the direction of the magnetic hyperfine field and the growth temperature.

In Fig. 3, the temperature dependence of the magnetic hyperfine field is shown for the two Fe/Cr/Fe trilayers with a 4 nm thick bottom Fe layer and the results are compared to the hyperfine fields in bulk Cr.[15] The magnetic hyperfine field in the Cr spacer for the Fe/Cr/Fe trilayer grown at 20 °C (■) is enhanced with respect to the magnetic hyperfine field observed for bulk Cr. In contrast, the magnetic hyperfine field observed in the Cr spacer of the Fe/Cr/Fe trilayer grown at 300 °C (△) is reduced compared to the values for bulk Cr. This observation confirms that the strength of the magnetic hyperfine field observed in the Cr spacer depends on the growth temperature of the Fe/Cr/Fe trilayer.

For the Fe/Cr/Fe trilayer grown at 20 °C, the hyperfine field value at 400 K is still large. This proves that the Néel temperature of this Cr spacer is considerably higher than the Néel temperature for bulk Cr ($T_N = 311$ K). For the Fe/Cr/Fe trilayer grown at 300 °C, the Cr spacer is already in the paramagnetic phase at 293 K. The Néel temperature of the Cr spacer grown at an elevated temperature is reduced compared to the Néel temperature of bulk Cr. Thus, we observed that the Néel temperature of the Cr spacer strongly depends on the growth temperature.

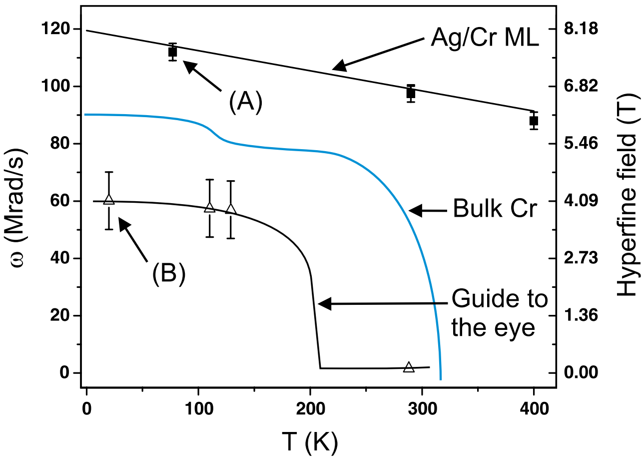

FIG. 3. (Color online) Temperature dependence of ^{111}Cd in the Cr hyperfine field for the Fe/Cr/Fe trilayer grown at 20 °C (■) and at 300 °C (△) (series I). The upper solid line corresponds to the data for the Ag/Cr multilayers (Ref. 10) whereas the lower line is a guide to the eye. The middle blue line corresponds to the values obtained for bulk Cr (Ref. 15). The points indicated by (a) and (b) correspond to the PAC spectra shown in Fig. 2.

In Table I, we give an overview of the orientation of the magnetic hyperfine field in various Fe/Cr/Fe(001) trilayers studied in this work. A growth temperature of 20 °C results in the magnetic hyperfine field being oriented along the growth direction. For an Fe/Cr/Fe trilayer grown at 300 °C, the magnetic hyperfine field is oriented perpendicular to the growth direction. These results are in good agreement with the results in literature.[5,10,16–18] Therefore, the third observation from the PAC results is also that the orientation of the magnetic hyperfine field depends on the growth temperature.

IV. DISCUSSION

We have shown in Secs. II and III that the tetragonal distortion, as well as the magnetic properties of the Cr spacer, strongly depends on the growth conditions of the Fe/

TABLE I. Fraction of the magnetic hyperfine fields (HFFs) in the Cr spacer ($t_{Cr} = 8$ nm) oriented along the [001] direction (growth direction).

Sample	T_G (°C)	HFF//[001] direction (%)
C0206 (series I)	20	100
C1222 (series II)	20	100
Ag/Cr ML[a]	20	100
Fe/Cr ML[b]	50	100
C1218 (series II)	70	50
C0205 (series I)	300	0
Fe/Cr ML[c]	300	0
Fe/Cr ML[d]	300	0

[a] Reference 10.
[b] Reference 5.
[c] Reference 16.
[d] Reference 17.

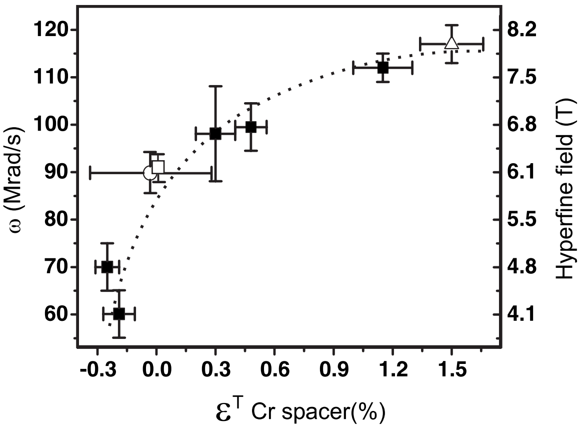

FIG. 4. Hyperfine field of ^{111}CdCr at a low temperature (77 K or lower) as a function of the tetragonal distortion of the Cr spacer for the Fe/Cr(001) trilayers (■). The triangle (△) corresponds to the Ag/Cr multilayers (Ref. 10), the circle (○) to the Fe/Cr multilayers (Ref. 5), and the boxes (□) to the bulk Cr (Ref. 15). The dotted line is a guide to the eye.

Cr/Fe trilayer. Therefore, it is worthwhile to inspect the direct relation between the structural and the magnetic properties of the Cr spacer layers.

First, we will show that the strength of the magnetic hyperfine field in the Cr spacer depends on the tetragonal distortion in the Cr spacer. Then, we will also argue that the orientation of the magnetic hyperfine field is determined by this tetragonal distortion.

In Fig. 4, we plot the low temperature (77 K or lower) magnetic hyperfine field in the Cr spacer as a function of the tetragonal distortion. For reference, we also show the hyperfine fields for bulk unstrained Cr (□) and for Ag/Cr (△) and Fe/Cr (○) multilayers available from literature.[5,10] For a large positive tetragonal distortion, an enhancement of the magnetic hyperfine field is found. In contrast, the hyperfine field is reduced in case of a slightly negative tetragonal distortion. From Fig. 4, we can conclude that there exists a direct relation between the tetragonal distortion of the Cr spacer and the strength of the magnetic hyperfine field in the Cr spacer.

The observed relation between the strength of the magnetic hyperfine field and the tetragonal distortion may be attributed to the change in the volume of the Cr unit cell. For a positive tetragonal distortion, the in-plane lattice constant of the Cr unit cell is stretched, and since the Poisson ratio of the bulk Cr is 0.21, an increase in the volume of the Cr unit cell has occurred. As a consequence, the magnetic hyperfine field in the Cr spacer increases. The opposite is true for a negative tetragonal distortion. Similar results have also been obtained with hydrostatic pressure measurements on bulk Cr.[19] It is also corroborated by *ab initio* calculations, which indicate that the magnitude of the magnetic hyperfine field primarily depends on the volume of the Cr unit cell.[20]

The orientation of the magnetic hyperfine field in thin Cr films has been intensively studied for Ag/Cr(001),[10] V/Cr(001),[21] and Fe/Cr(001) multilayers.[5,14,18] The frequently observed perpendicular orientation of the Cr magnetic moments (i.e., along the growth direction) was attributed to magnetic frustration at the Fe/Cr interface[18] or to the strain that is present in the Cr spacer.[22]

The hyperfine field parameters are the same for both the Fe/Cr/Fe trilayers grown at 20 °C with a bottom Fe layer of 4 nm and the Ag/Cr multilayers (see Fig. 3 and Table I). Remarkably, the tetragonal distortion of the Cr spacer is also similar in both systems: $\epsilon^T = +1.50\%$ in Ag/Cr multilayers[10] and $\epsilon^T = +1.15(15)\%$ for the Fe/Cr/Fe trilayer with a bottom Fe layer of 4 nm grown at 20 °C. Since the magnetic properties of the Cr spacer are the same for epitaxial systems, which have either magnetic or nonmagnetic boundary layers, we conclude that the tetragonal distortion of the Cr spacer, which is similar for both cases presented above, determines the magnetic properties of the Cr spacer. In other words, the orientation of the Cr magnetic moments along the growth direction is determined by an anisotropy effect that is stabilized by the tetragonal structure in the Cr.

Finally, for a high growth temperature, e.g., 300 °C, a considerable amount of diffusion of Fe into the Cr spacer at the Fe/Cr interface is expected.[23] The consequences of this interfacial interdiffusion for the magnetic properties of the Cr spacer are not fully known. *Ab initio* calculations show that the presence of an FeCr alloy at the Fe/Cr interface can cause a reduction in the magnetic moment in thin Cr films.[24] As a consequence, the interdiffusion at the interfaces can also contribute to the reduction in the magnetic hyperfine field in the Cr spacer of the Fe/Cr/Fe trilayers grown at elevated temperatures.

V. CONCLUSION

We have shown that there exists an intimate relation between the tetragonal distortion and the magnetic properties of the Cr spacer in the epitaxial Fe/Cr/Fe(001) trilayers. The tetragonal distortion in the Cr spacer shifts the Néel temperature of the Cr spacer, and it determines the orientation and the strength of the magnetic hyperfine field in the Cr spacer.

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