Structural and Magnetic Ordering of Chromium in Ag/Cr Multilayers

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We studied the influence of the lattice structure on the chromium magnetism in Ag/Cr multilayers by combining structural characterization techniques and perturbed angular correlation spectroscopy. Chromium in Ag/Cr multilayers orders as an incommensurate longitudinal spin-density wave (SDW) with the spins oriented out of plane. A size effect induces the instability of the SDW below a critical chromium thickness. The absence of the spin-flip transition, the position of the Néel temperature, and the actual character of the SDW are attributed to the specific structural properties. [S0031-9007(98)07171-3]

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Theoretical predictions concerning the magnetism of overlayers can be verified by a variety of modern techniques. Buried spacer layers, however, are more difficult to access experimentally, especially when antiferromagnetically ordered. Neutron scattering but also hyperfine interaction techniques using radioactive probes are very suitable in such cases. This was recently demonstrated [1] by introducing perturbed angular correlation (PAC) spectroscopy [2] for the study of the peculiar magnetism [3] in chromium layers. In bulk Cr the longitudinal spin-density wave (LSDW) is, at 123 K, replaced by a transversal spin-density wave (TSDW) up to a Néel temperature of 311 K. The results obtained by PAC in Fe/Cr multilayers [1] mainly concerned the Cr spin orientation, found perpendicular to the layers and thus perpendicular to the in-plane Fe magnetization, and the collapse of the SDW for Cr thickness below about 5 nm. Concurrently the latter effect was found by resistivity measurements also [4]. This critical thickness almost equals the period of the LSDW observed. Subsequent neutron scattering data agreed with the SDW-instability result, although some important inconsistencies remained [5,6]: (i) on the character of the SDW for Cr layers above the critical thickness and (ii) on the possible replacement of the incommensurate SDW by a commensurate AF_0 state below the critical thickness. In neutron scattering experiments [7] one observes that the propagation vector **Q** as well as the polarization vector **S** of the SDW depends both on the Cr film thickness and on the type of capping layer. The reorientation effect is explained [8] by a stepped Fe/Cr interface which induces frustration in the inter- and intralayer exchange interaction. In addition, below the critical thickness for the stability of the SDW, neutron scattering experiments [6] on Fe/Cr superlattices find Cr in the in-plane AF_0 state, and a spiral spin orientation is suggested as a possible mechanism for the noncollinear interlayer coupling. In contrast, the PAC data for Fe/Cr multilayers show paramagnetic Cr up to 5.1 nm and out-of-plane spin polarization in thicker layers, but independent of the Cr-layer thickness [1]. A new set of PAC data [9], obtained on Cr films characterized by neutron diffraction [8], established that results were independent of the experimental technique, thereby proving that PAC indeed probes the intrinsic Cr magnetism.

In this Letter we offer an explanation for the conflict in results mentioned above. We show that not the degree of frustration at the interface but rather the specific structural properties are the main origin of the controversy. The present PAC data obtained on structurally well characterized Ag/Cr multilayers, combined with our earlier data on Fe/Cr multilayers, undoubtedly prove that chromium layers with thickness above 5.1 nm remain in the LSDW phase from 4.2 K up to 500 K. The condition for this is that the Cr layers are grown such that a small tetragonal distortion can establish and stabilize the LSDW ordering. The instability versus thickness of the SDW is an intrinsic size effect, rather than due to the proximity of the interface or presence of magnetic Fe in the multilayers. Ag/Cr superlattices are hereby promising as a model system to investigate the LSDW in a single-Q state in a broad temperature region and to explore magnetic effects representative for chromium in reduced dimension.

The multilayers were grown by molecular beam epitaxy (MBE) on MgO(001) substrates which were cleaned in isopropanol and annealed at 600 °C for 30 min prior to growth. The buffer layer of 10 nm of Fe, grown at 175 °C, is partly relaxed. Thereby the subsequent layers of Ag and Cr have a much smaller lattice mismatch with the substrate, compared to direct growth on MgO itself. The fcc Ag grows on bcc Cr following the azimuthal epitaxial relationship MgO[100]//Fe[110]//Cr[110]//Ag[100]. The growth temperature is 20 °C, whereas the previously investigated Fe/Cr multilayers were grown at 150 °C. In all samples the Ag layer thickness is approximately 3 nm, while the Cr thickness ranges from 3 to 8 nm.

The large angle x-ray diffraction spectra, fitted to model calculations using the SUPREX program [10], include information on the perpendicular lattice parameter, whereas the low-angle spectra are used for the layer thickness

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determination. The perpendicular lattice spacings of Ag lie between 1.97(2) Å and 1.98(2) Å, a significant reduction compared to the bulk value of 2.043 Å. A much smaller reduction of the perpendicular lattice spacing of Cr is seen; values lie between 1.437(2) Å and 1.439(2) Å. For Cr the bulk value equals 1.442 Å.

Depth sensitive information on strain and azimuthal orientation in the layers can be inferred from channeling experiments along nonperpendicular lattice directions [11]. During the angular scan different energy windows correspond to different layers in the sample. Since no strain is expected in MgO, the minimum in the yield indeed shows up to 45° (Fig. 1). Because of the epitaxial relationship, unstrained Ag would again give rise to a minimum at 45°. The unit cell of Cr, however, is azimuthally turned. For undistorted Cr, the minimum would thereby be expected at 54.74°. From the position of the actual minima, one determines the tetragonal distortion e_T in the layers ($e_T = e_{//} - e_{\perp}$; see Table I and [11]). The magnitude of this distortion is definitely much larger in Ag than in Cr. We calculate the perpendicular lattice spacing assuming a simple Poisson behavior and using values for the Poisson ratio of 0.43 for Ag and 0.16 for Cr [12,13]. In Table I we compare the results of the channeling

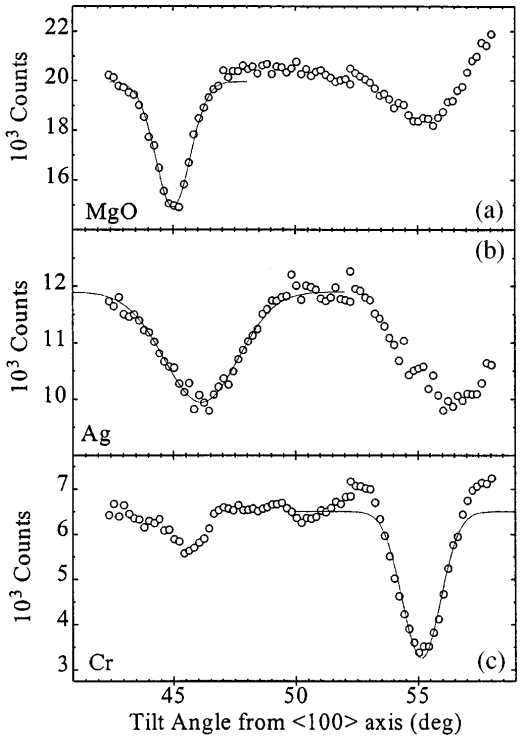

FIG. 1. Result of a channeling experiment on MgO/Fe/(2.9 nm Ag/6.3 nm Cr)$_{10}$, using 2 MeV He particles. Unstrained MgO, Ag, and Cr would give rise to a minimum at tilt angles of 45°, 45°, and 54.74°, respectively. The actual minima appear at 45° for MgO (a), 46.13° for Ag (b), and 55.14° for Cr (c), respectively, indicating tetragonal distortion of the layers in the superlattice.

experiments for the MgO/Fe/(2.9 nm Ag/3.6 nm Cr)$_{10}$ and MgO/Fe/(2.9 nm Ag/6.3 nm Cr)$_{10}$ samples with the results of the x-ray diffraction fits. The values agree well. The obtained perpendicular lattice spacings suggest growth on a partly relaxed Fe buffer layer.

In order to characterize the magnetism in the Cr layers, we use a nuclear technique based on the hyperfine interaction: perturbed angular correlation spectroscopy [1,2]. The method is sensitive to the magnitude of the extranuclear electromagnetic fields through the frequency in the so-called PAC time spectrum [or R(t) spectrum], as well as to the orientation of these hyperfine fields relative to the detectors [1]. Furthermore, the PAC technique has the advantage that the absolute contributions from different phases can be determined.

Samples with different Cr thickness were implanted [9] with the ^{111}In/^{111}Cd probe at an energy of 80 keV keeping the dose below 2×10^{13} atoms/cm^2. The doped samples were measured at temperatures from 4.2 K up to 550 K and in different detector geometries. The PAC time spectra, measured on MgO/Fe/(2.8 nm Ag/7.9 nm Cr)$_{10}$, simultaneously in two detector geometries, are shown in Fig. 2. The spectra on all samples reflect three ^{111}Cd probe locations: apart from probes at locations near the interface, we expect probes at substantial positions in the Cr as well as the Ag layer. In view of the much larger strain in the Ag layer compared to the Cr layer, these probes experience finite electric field gradients (EFG's) in the Ag, while in the Cr fraction the EFG is nearly zero. For the "interface fraction" we expect hyperfine fields and/or EFG's with a large distribution in magnitude and orientation. This third fraction, together with the Ag fraction, accounts for the damping of the amplitude within 50 ns. The probes ending up in the Cr layer contribute, due to the magnetic hyperfine interaction provided by magnetic ordering, to the oscillation in the time spectra (Fig. 2). This oscillation is fit to a zeroth order Bessel function; hence [1] the antiferromagnetic ordering in the Cr layers reflects an incommensurate SDW-type. The orientation of the magnetic hyperfine field is derived from the relative amplitude of the first and second frequency harmonic in the $R(t)$ spectrum: with the magnetic field normal to the detector plane one observes solely the second harmonic [Fig. 2(a)], while only the first harmonic is observed if the field lies in the plane of the detectors [Fig. 2(b)]. Evidently the hyperfine field in our samples points along the sample normal. This observation directly proves that the magnetic moments in the Cr layers are oriented out of plane. From the value of the hyperfine field of 6.14(5) T at room temperature, one may deduce [9] indirectly the longitudinal character of the SDW. The same perpendicular orientation of the Cr moments and the LSDW phase have been observed with PAC in Fe/Cr multilayers [1] as well as in Cr thin films [9]. The present results suggest that, at least under our growth conditions, frustration at

TABLE I. Comparison of the results for MgO/Fe/(2.9 nm Ag/6.3 nm Cr)$_{10}$ and MgO/Fe/(2.9 nm Ag/3.6 nm Cr)$_{10}$. Included are the measured tetragonal distortionn e_T, the in-plane and perpendicular elastic deformations ($e_{//}, e_\perp$), and the lattice spacings derived from the channeling experiments ($d_{\perp,\mathrm{chan}}$), compared with the values obtained from the x-ray diffraction spectra using the SUPREX program ($d_{\perp,\mathrm{xrd}}$) [10].

	e_T (%)	$e_{//}$ (%)	$e_\perp$ (%)	$d_{\perp,\mathrm{chan}}$ (Å)	$d_{\perp,\mathrm{xrd}}$ (Å)
(2.9 nm Ag/	3.9(4)	1.6(2)	−2.3(4)	1.995(7)	1.972(2)
6.3 nm Cr)$_{10}$	1.50(16)	1.10(15)	−0.40(6)	1.436(1)	1.437(2)
(2.9 nm Ag/	4.4(5)	1.7(2)	−2.6(5)	1.990(7)	1.98(2)
3.6 nm Cr)$_{10}$	1.50(16)	0.94(12)	−0.53(8)	1.436(1)	1.439(2)

the Fe/Cr interface is not at the origin of this anisotropy and this type of Cr magnetism.

The ^{111}Cd*Cr* hyperfine fields measured at room temperature in the Ag/Cr system are consistent with those determined earlier [1] in Fe/Cr multilayers and in a single Cr layer of 400 nm. As a function of temperature [Fig. 3(a)] the magnitude of this hyperfine field decreases almost linearly with a slope of $4.8(1) \times 10^{-3}$ T/K within accuracy independent of the layer thickness. Interestingly, this almost linear behavior, which extends from 4.2 K up to 500 K, is a perfect extrapolation of the low temperature neutron data of Werner *et al.* [14] on bulk Cr. In Fig. [3(b)], we compare the temperature dependence of the integrated intensity at the $(0, 1, \delta)$ satellite, as measured in neutron scattering [14], with the (sample average) hyperfine fields determined by PAC measurements. The integrated satellite intensity was normalized to the hyperfine field at 4.2 K. We find an excellent consistency between both data sets thereby proving that indeed the LSDW, the low temperature phase in bulk Cr, remains stable up to 500 K in the (thin) Cr layers of the Ag/Cr multilayer. At 500 K the oscillation in our spectra, due to the long-range magnetic ordering in the Cr layers, is still present, but it has completely disappeared at 550 K. Likewise, a similar enhanced Néel temperature was observed in MBE-grown Fe/Cr multilayers and Cr thin films on MgO. The major reason for the higher Néel temperature in all these samples cannot be the proximity of the interface, but rather the structural properties resulting from the specific growth conditions on MgO. The small tetragonal distortion in the Cr layers, revealed by the channeling experiments, determines the character of the SDW. We observe from 4.2 K up to the magnetic transition around 550 K a *longitudinal* SDW, in contrast to bulk chromium.

Finally, by decreasing the Cr thickness from 5.9 nm to 5.1 nm a drastic change in the spectra is illustrated in Fig. 4. While in the sample with $t_{\mathrm{Cr}} = 5.9$ nm the precession pattern due to the magnetic ordering is still clearly present [Fig. 4(a)], this contribution disappears completely for the thinner layers [Fig. 4(b)]. Moreover, the absence of any oscillation again proves that the collapsed SDW is not replaced by a commensurate antiferromagnetic ordering.

The observation of the SDW collapse at a chromium thickness comparable with the SDW period, in Fe/Cr but now also in Ag/Cr, suggests that this effect is an intrinsic size effect of Cr in reduced dimension. We do not observe (at least for the present growth conditions on

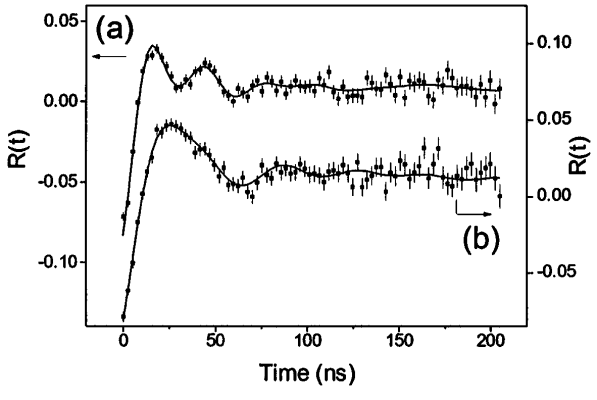

FIG. 2. PAC time spectra for MgO/Fe/(2.8 nm Ag/7.9 nm Cr)$_{10}$ measured at 77 K, recorded with the sample normal oriented perpendicular to the detector plane (a), with the sample normal oriented in the detector plane, in-between two detectors (b).

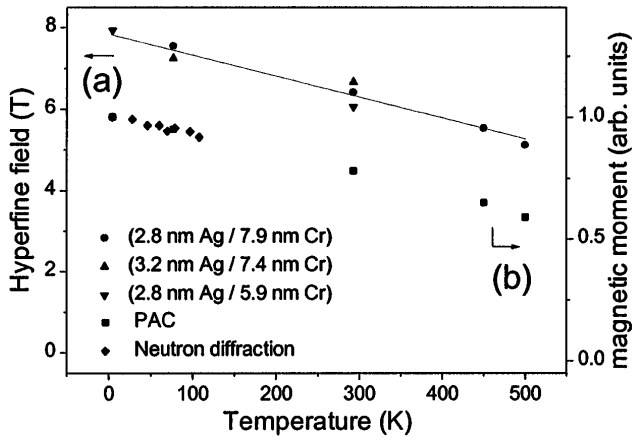

FIG. 3. Hyperfine field versus temperature (a). The solid line is a linear fit through the mean values. Magnetic moments (normalized at 4.2 K), obtained by neutron diffraction and PAC, versus temperature (b).

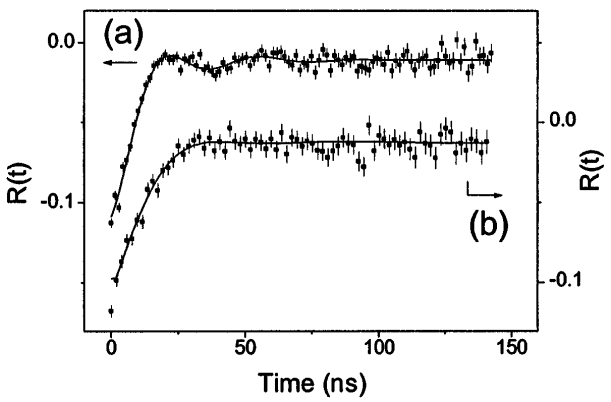

FIG. 4. PAC time spectra for MgO/Fe/(2.8 nm Ag/5.9 nm Cr)$_{10}$ (a) and MgO/Fe/(3.2 nm Ag/5.1 nm Cr)$_{10}$ (b), both measured at 293 K, with the sample normal oriented perpendicular to the detector plane.

MgO) any TSDW (with spins in-plane) in either type of multilayers or in Cr films. The growing of the Cr layers at an increased temperature and/or on another substrate, results in a different growth mode and/or increased interdiffusion at the Fe/Cr interface [15]. This can explain the occurrence of a different type of Cr-SDW magnetism and/or Néel temperature as seen in neutron data and confirmed also by PAC data [9]. The perpendicular spin orientation observed in our chromium layers is attributed to residual tetragonal lattice distortion favoring a longitudinal SDW in a single-Q magnetic domain. The "enhanced" Néel temperature is understood as a "true" LSDW-to-paramagnetic transition temperature, seen here without the intervening TSDW as usual in bulk Cr. The transition to the paramagnetic state is rather sharp because this hyperfine field versus temperature behavior definitely holds until 500 K, whereas we observe no magnetic ordering at 550 K. Interestingly, the position of this Néel temperature also depends on growth conditions, because T_N equals 330 K for a 250 nm Cr film on Nb growth at 300 °C [9].

We conclude that MBE growth of very thin chromium films on partly relaxed MgO/Fe substrates at $T = 150$ °C (Fe/Cr) or at room temperature (Ag/Cr) produces a single-Q magnetic domain with the LSDW (spins out of plane) ordering stable up to 500 K far above the spin-flip temperature of bulk chromium. This appealing observation may possibly initiate further studies of the itinerant magnetism in reduced dimension and contribute to understanding the nature of the spin-flip transition. Based on inelastic neutron scattering with polarization analysis [16], this phase transition was understood as an anisotropy effect rather than an intrinsic feature of the SDW ordering. This conjecture, supported by the present results, needs further founding by forthcoming refined electronic structure calculations. It has been shown already that relativistic effects indeed result in a lowering of the energy for the longitudinal relative to the transversal SDW polarization [17].

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