Longitudinal spin-density-wave ordering in thin epitaxial chromium layers

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Thin chromium films molecular beam epitaxy grown on partly relaxed MgO/Fe substrates held below $T=150\,°C$ are found either nonmagnetic below a chromium thickness around 5.0 nm or in a single-Q magnetic domain with the longitudinal (spins-out-of-plane) spin-density-wave (SDW) ordering stable up to 500 K. The longitudinal SDW-to-paramagnetic transition, the absence of the spin-flip transition and the actual character of the SDW depends on specific growth conditions. The present results reveal the spin-flip transition in chromium as an anisotropy effect rather than an intrinsic feature of the SDW ordering. © *1999 American Institute of Physics.* [S0021-8979(99)58808-7]

I. INTRODUCTION

The typical itinerant magnetism of chromium remains a topic of broad interest, especially since molecular beam epitaxy (MBE) techniques can produce high quality epitaxial layers and superlattices. Most novel properties of the interlayer coupled systems were first discovered in the Fe/Cr multilayers and therefore activated the research of chromium magnetism in reduced dimension. Buried spacer layers however are difficult to access, especially when antiferromagnetically ordered. In the nanometer thickness range even neutron scattering techniques are not very suitable. Perturbed angular correlation (PAC) spectroscopy was proposed[1] recently as a valuable alternative for the study of the peculiar magnetism in chromium, with some appealing results. It is well known that bulk chromium orders, at the Néel temperature $T_N = 311\,K$, as an incommensurate spin density wave (SDW) antiferromagnet with wave vector $Q_{SDW}=0.958(2\pi/a)$ along the (100) direction. Its polarization changes from $\mathbf{S}\perp\mathbf{Q}$ in the transverse SDW phase (AF_1) above the spin-flip temperature ($T_{SF}=123\,K$) to $\mathbf{S}\|\mathbf{Q}$ in the longitudinal SDW phase (AF_2) below.

The PAC experiments[1] on Fe/Cr multilayers grown on MgO at 150 °C found the Cr spin orientation perpendicular to the layers and thus perpendicular to the in-plane Fe magnetization, independent of the Cr-layer thickness at least up to 400 nm. Furthermore we found[1] the collapse of the SDW for Cr thickness below about 5.0 nm. This critical thickness almost matches with the period of the SDW, in those measurements observed in single Q domain.

Subsequent experiments have indicated that the specific growth conditions are important for the actual type of magnetic order. Indeed, in contrast to the original PAC study, for Fe/Cr multilayers grown on Nb at 300 °C with Cr thickness between 5.1 and 19 nm, both neutron diffraction data[2] and PAC data[3] indicate a *transverse* SDW with an out-of-plane Q vector and spins in-plane, but turning out-of-plane for thicker Cr layers. In addition, below the critical thickness for the stability of the SDW, neutron diffraction experiments[2] on Fe/Cr superlattices find Cr partially in the in-plane AF_0 state.

In order to avoid any influence on the chromium spin structure as due to a magnetic spacer layer the PAC technique was applied on Ag/Cr multilayers.[4] One expects to observe in this system a magnetic state representative for chromium in reduced dimension. These multilayers were grown by molecular beam epitaxy on preannealed MgO substrates. The lattice mismatch for Cr and Ag on MgO is respectively +4.1% and +2.9%. Therefore the roughness of the multilayer was reduced by using a buffer layer of 10 nm Fe, deposited at 150 °C, shown by channeling experiments to be completely relaxed (the lattice mismatch between Ag and Fe is only 0.8%). The face-centered-cubic (fcc) Ag grows on body-centered-cubic (bcc) Cr under the epitaxial relation [110]Ag//[100]Cr. All samples consist of ten bilayers with varying Cr thickness. The deposition rates were 0.5 (Cr) and 0.9 Å/s (Ag) and the growth temperatures 25 °C. Reflection high-energy electron diffraction (RHEED) patterns were recorded to monitor the growth and the quality of the samples was checked by x-ray diffraction (XRD). Rutherford backscattering and channeling experiments, used[4] to measure strain in the epitaxial layers, prove that in the Ag/Cr multilayers the out-of-plane lattice parameter is reduced as compared to the in-plane lattice parameter. For the Cr layers the compression equals 4×10^{-3} while in the Ag layers a much larger reduction of the out-of-plane lattice parameter of 2.4% is observed. The detailed structural characterization analysis implies that the body-centered-tetragonal (bct) structure of the chromium layer should be responsible for the actual magnetic order in the Cr layers.

II. THE Cr MAGNETISM IN Ag/Cr(001) MULTILAYERS

In order to characterize the magnetism in the samples, we use a nuclear technique based on the hyperfine interaction: perturbed angular correlation spectroscopy (PACs).[1,3] A trace amount of typically 10^{11} radioactive ^{111}In probes is introduced into the sample by ion implantation at an energy of 80 keV, thereby distributing the probes throughout the multilayer. The experimental result of a PAC measurement is the so-called $R(t)$ spectrum, a more or less periodic pattern representing the nuclear Larmor precession. The method is not only sensitive to the magnitude of the extranuclear elec-

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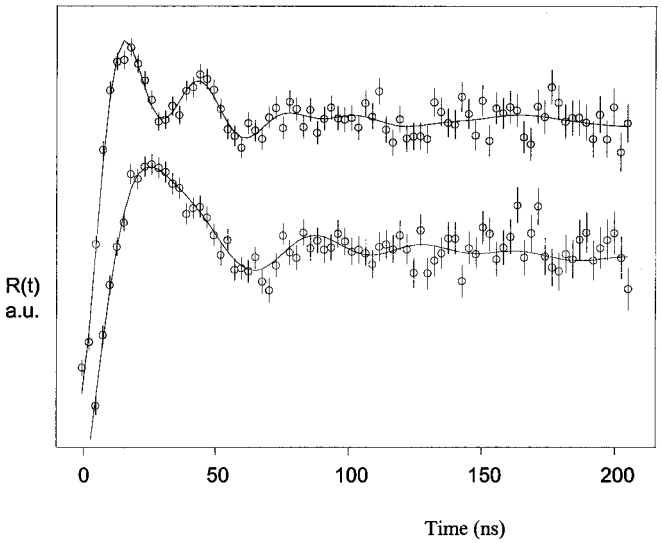

FIG. 1. PAC time spectra for $(2.8\,\text{nm Ag}/7.9\,\text{nm Cr})_{10}$ measured at 77 K. The upper spectrum was recorded with the sample normal perpendicular to the detector plane, the lower spectrum with the sample normal in the detector plane.

tromagnetic fields through the frequency, but important also to the orientation of these hyperfine fields relative to the detectors. Recent PAC experiments[3] on thin films and multilayers have proven that the diamagnetic ^{111}Cd nuclear probe indeed senses the intrinsic magnetic behavior of the Cr layer giving results compatible with neutron diffraction experiments when measured on the same samples.

We have studied samples with varying Cr thickness at temperatures from 4.2 up to 550 K and in different detector geometries. A typical $R(t)$ spectrum, measured simultaneously in two detector geometries in order to derive the hyperfine field orientation in a straightforward way, is shown in Fig. 1. In the analysis[5] we accounted for three In probe locations: apart from probes at locations near to the interface, we expect probes at substitutional positions in the Cr as well as the Ag layer. The probes ending up within the Cr layer contribute to the oscillation seen in the spectra (Fig. 1),

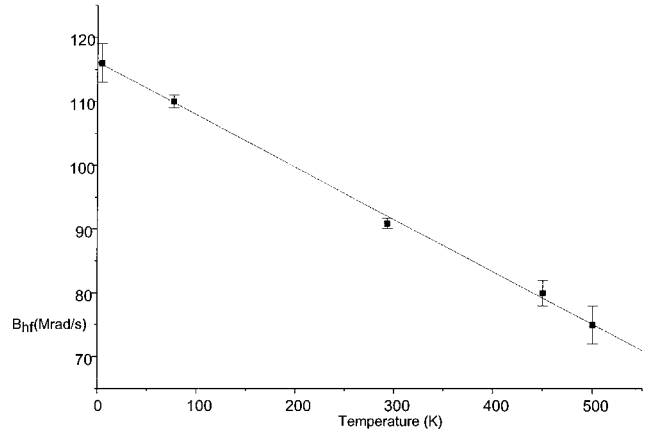

FIG. 2. Hyperfine field (B_{hf}), or equivalently the chromium magnetic moment amplitude, as a function of temperature. The data points represent averages over the different layer thicknesses.

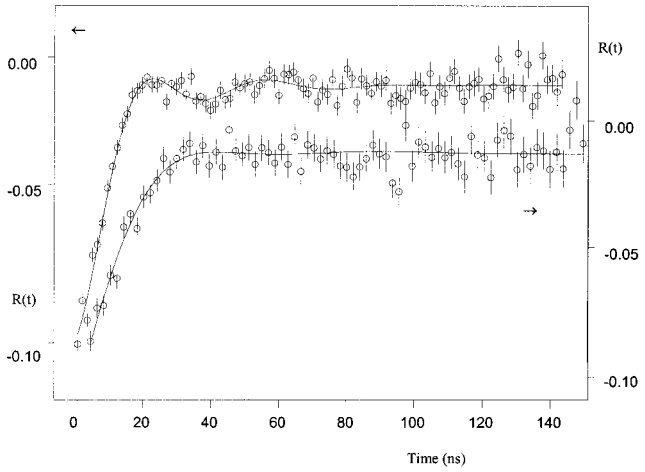

FIG. 3. PAC time spectra for $(2.8\,\text{nm Ag}/5.9\,\text{nm Cr})_{10}$ (upper) and $(3.2\,\text{nm Ag}/5.1\,\text{nm Cr})_{10}$ (lower) measured at 293 K.

which indeed appears as soon as magnetic order establishes. The corresponding hyperfine field however reflects a distribution typical for an incommensurate SDW. The value of the hyperfine field indicates[3] longitudinal polarization thus the LSDW phase (PAC is only indirectly sensitive to the Q vector). The orientation of the magnetic field is derived from the relative amplitude of the first and second harmonic in the $R(t)$ spectrum (see caption to Fig. 1). The hyperfine field in our samples and thus the magnetic moments in the Cr layers are oriented out-of-plane.

The ^{111}CdCr hyperfine fields, magnitude, and orientation, measured at room temperature in the Ag/Cr system are nicely consistent with those determined[1] in Fe/Cr multilayers and Cr single 400-nm-thick layers grown also on MgO. As a function of temperature and for all samples measured the magnitude of the hyperfine field decreases almost linearly with an average slope of $-7.0(2)\times10^{-4}$ K. Interestingly, this linear behavior (Fig. 2) which extends from 4.2 up to 500 K, is a perfect extrapolation of the temperature dependence of the integrated intensity at the $(0,1,\delta)$ satellite measured by neutron scattering[6] in bulk Cr in the LSDW low temperature phase. This observation clearly proves that in the present PAC experiments the *longitudinal* SDW state remains stable up to 500 K, far above the spin-flip temperature at 123 K in bulk Cr. At 500 K Cr is still magnetic, but this magnetic phase has completely disappeared at 550 K. Finally, while in the sample with $t_{Cr}=5.9$ nm the precession pattern due to magnetic ordering is still clearly present, this contribution disappears completely for the thinner layers (Fig. 3). Moreover the absence of any precession signal further proves that the collapsed SDW is not replaced by a commensurate antiferromagnetic ordering.

III. CONCLUSIONS

The observation of the SDW collapse at a chromium thickness comparable with the SDW period, in Fe/Cr but also in Ag/Cr suggests that this effect is an intrinsic size effect of Cr in reduced dimension. The elastic information derived from our channeling experiments reveals that the

perpendicular spin orientation observed in chromium layers can be attributed to residual tetragonal lattice distortion favoring a longitudinal SDW in a single-*Q* magnetic domain. This magnetic order persists up to 500 K and has been observed in Cr layers with thickness at least up to 400 nm. The ''enhanced'' Néel temperature in those samples is understood as the ''true'' LSDW-to-paramagnetic transition temperature, seen here without the intervening TSDW as usual in bulk Cr. The transition to the paramagnetic state should be rather sharp since we observe at 550 K no magnetic order. Therefore the message of this article is: very thin Cr is paramagnetic because the SDW collapses due to *finite size effect* and the magnetic ordering for thicker layers is *LSDW without a spin-flip transition*, due to residual tetragonal lattice distortion. The latter (bct) structure is realized by MBE growth of chromium layers on a Fe/MgO buffer/substrate combination held below 150 °C. Furthermore, the linear temperature dependence of the chromium magnetic moment (the amplitude of the SDW) as reflected by the magnetic hyperfine field is an unusual behavior for a three-dimensional (3D) system and a challenge to theory.

In conclusion, those appealing observations may possibly initiate further studies on the itinerant magnetism in reduced dimension and contribute to understand the nature of the spin-flip transition. Based on the present results, this phase transition appears as an anisotropy effect rather than an intrinsic feature of the SDW ordering. Forthcoming refined electronic structure calculations may give further support on the conclusion that indeed a tetragonal distortion of chromium results in a lowering of the energy for the longitudinal relative to the transversal SDW polarization.

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