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Spin-density-wave magnetism in layered chromium studied by perturbed-angular-correlation spectroscopy

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Results obtained by perturbed-angular-correlation (PAC) spectroscopy on epitaxial chromium thin films are compared with data obtained from the same samples by neutron-diffraction experiments. We report on the study of a 250-nm-thick Cr single layer and an Fe/Cr multilayer with Cr thickness of 25 nm grown on Nb/Al_2O_3 ($1\underline{1}02$). The chromium single layer film orders below the Néel temperature as a longitudinal (AF_2) spin density wave antiferromagnet with the spins out of plane, while the Cr in the Fe/Cr multilayer orders as a transversal (AF_1) spin density wave antiferromagnet with the spins in plane. In addition we observe a minority volume fraction in a commensurate AF_0 phase, stable up to at least 400 K. The present results prove that PAC probes the *intrinsic* magnetic features of chromium and therefore validate this technique as a complementary approach towards the understanding of the chromium magnetism in thin films and Fe/Cr multilayers. [S0163-1829(98)51210-6]

In spite of the already extensive literature, the typical itinerant magnetism of chromium remains a topic of broad interest. It is well known[1] that bulk chromium orders, at the Néel temperature $T_N = 311$ K, as an incommensurate spin-density-wave (SDW) antiferromagnet with wave vector $Q_{\mathrm{SDW}} = 0.958(2\pi/a)$ along the (100) direction (a = lattice constant, Q is defined as the propagation vector of the SDW). Its polarization changes from $\mathbf{S} \perp \mathbf{Q}$ in the transverse SDW phase (AF_1) above the spin-flip temperature ($T_{\mathrm{SF}} = 123$ K) to $\mathbf{S} \parallel \mathbf{Q}$ in the longitudinal SDW phase (AF_2) below T_{SF}. The crystal structure above T_N is body centered cubic, but transforms for a single Q sample from orthorhombic to tetragonal by cooling through T_{SF}. The commensurate antiferromagnetic structure (AF_0), in which the spins at the corner and at the center of the bcc unit cell are of equal magnitude but point in opposite directions, may be stabilized in alloys. This phase was incorporated also in the magnetic phase diagram of strained chromium.[2]

Recently it became possible to produce high quality epitaxial layers and superlattices. Most novel properties of the interlayer coupled systems were first discovered in the Fe/Cr multilayers. This progress activated the research of the chromium magnetism in reduced dimension. Perturbed-angular-correlation (PAC) spectroscopy on ion implanted Fe/Cr multilayers gave the first evidence[3,4] that the Cr layers lose the bulk Cr spin-density-wave magnetism for thickness below 5.0 nm. Moreover, while the magnetization in the Fe layer is along the *in-plane* [010] or [001] axes, for Cr-thickness above 7.5 nm the observed polarization of the spin-density wave corresponds to chromium spins along the [100] direction *normal* to the layers,[4] thus *perpendicular* to the Fe spins. This type of ordering has been seen in Cr films grown on MgO up to a thickness of at least 40 nm, irrespective of the presence of Fe.[4]

Transport measurements on sputtered epitaxial Fe/Cr superlattices[5] confirmed the suppression of magnetic ordering below a Cr thickness of 4.2 nm. On the other hand, neutron diffraction experiments on Fe/Cr multilayers, grown by magnetron sputtering on MgO (Ref. 6) or by molecular beam epitaxy (MBE) on Nb/Al_2O_3,[7] suggest that very thin chromium layers are not paramagnetic but rather adopt the commensurate antiferromagnetic structure below the critical chromium thickness. A problem arose also on the issue of the SDW polarization. Neutron-diffraction experiments on uncovered or Cu-capped chromium films approximately 300 nm thick find a longitudinal out-of-plane SDW as observed in the PAC study. When the film is covered by a 2.0 nm ferromagnetic (i.e., Fe) capping layer the Q vector reorients into in plane but the magnetic moments still point out of plane. For relatively thick chromium layers, the Fe and Cr moments are thus oriented perpendicular to each other as was also reported for Fe/Cr on MgO.[4] However in contrast to the PAC study, for Fe/Cr multilayers with Cr thickness between 5.1 nm and 19 nm neutron diffraction data[7] indicate a *transverse* SDW with an out-of-plane Q vector and spins in plane.

In view of the above controversy, new data to illustrate the compatibility of the various methods in determining the character of the Cr magnetic ordering are imperative. The perturbed-angular-correlation spectroscopy technique on bulk polycrystalline chromium was found[8] to reproduce the important transitions revealed by neutron diffraction studies. However, up to now no direct comparison between the two techniques has been done on single crystalline, single-**Q** samples. Therefore the PAC method has been applied to study the same chromium films as used for the neutron diffraction experiments. The samples were grown[9] by MBE to a thickness of 250 nm (film) and 25 nm (multilayer: $[2\ \mathrm{nm\ Fe}/25\ \mathrm{nm\ Cr}]_{10}$) on the (50 nm) Nb/Al_2O_3 ($1\underline{1}02$) buffer/substrate system. The epitaxial relationship was checked[9,10] during the growth by *in-situ* reflective high-energy electron diffraction (RHEED) spectroscopy and confirmed by x-ray diffraction measurements.

Transport measurements are frequently used to determine the antiferromagnetic ordering temperature in chromium and chromium alloys.[1] A clear magnetic contribution to the re-

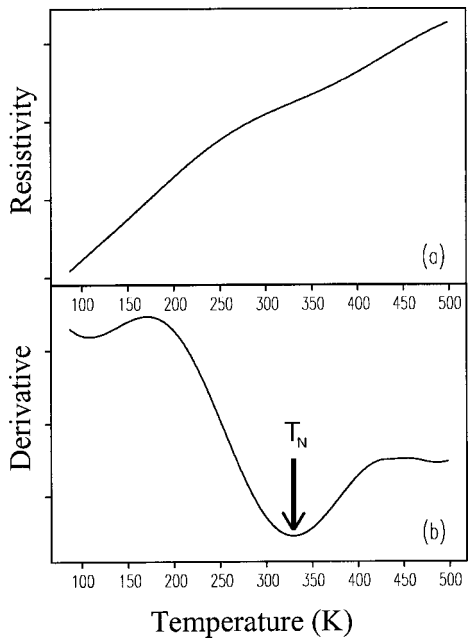

FIG. 1. The temperature dependence of the resistivity and the derivative of the resistivity for the 250 nm Cr/(50 nm)Nb/Al_2O_3 ($11\bar{2}$) film. The Néel temperature is determined by the inflection point in the $d\rho$ vs dT curve at (330 ± 10) K.

sistivity, ρ, is observed below the paramagnetic-antiferromagnetic phase transition temperature, T_N. Also in thin films this method can be applied to determine the phase transition temperature.[5] Figure 1(a) shows the resistivity vs temperature for the present 250 nm Cr film, measured using the standard four-contact dc method. The Néel temperature is found by the inflection point in the $d\rho/dT$ vs T curve at (330 ± 10) K, as shown in Fig. 1(b), where the resistivity amounts to 10.08 $\mu\Omega$ cm. Similar results are obtained for the $[2\text{ nm Fe}/25\text{ nm Cr}]_{10}$ multilayer. As observed earlier,[4] the resistivity anomaly in the Cr layers is considerably broadened compared with the singularity in ρ observed at T_N in a chromium single crystal. Comparison with the results of Geerkens[11] suggests that the pronounced anomaly at 330 K indicates a bulklike antiferromagnetic-to-paramagnetic phase transition, rather than a transition from the incommensurate to commensurate magnetic phase.

The present samples were characterized[7] by synchrotron x-ray scattering and high-angle neutron-diffraction experiments. The combination of both techniques accurately determines[7] the spin orientation and the Q vector of the SDW, however not yet simultaneously the magnetic moment and the absolute volume fraction of any coexisting phases. In the 250-nm-thick chromium single film, a dominant longitudinal out-of-plane spin-density wave has been observed[12] at low temperatures, together with a minority out-of-plane transverse incommensurate phase. At temperatures above 330 K the incommensurate spin-density wave flips, at least partially, to the commensurate AF_0 phase with the spins out of plane.[13] For the Fe/Cr multilayer, a transverse SDW with in-plane spins is observed, associated with a minority AF_0 (spins in plane) phase. At temperatures above 330 K the Cr layers in the $[2\text{ nm Fe}/25\text{ nm Cr}]_{10}$ superlattice are in a paramagnetic state coexisting with a commensurate phase with the spins in plane.

The PAC technique[4] identifies, through the hyperfine interaction parameters, the microscopic surrounding of a nuclear probe (in this case ^{111}Cd). Each probe environment is characterized by a (Larmor) frequency, ν, proportional to the local hyperfine field. Because a diamagnetic probe such as Cd senses a *transferred* hyperfine field only, the experiment is sensitive to the magnetic moments in the near environment of the probe. In the measurements, the Larmor precession frequency may occur in the spectra together with its second harmonic. The relative amplitude of both frequencies is determined by the direction of the hyperfine field relative to the detector geometry. One expects the magnetic hyperfine field to be collinear with the local magnetization of the matrix.[4] The polarization of the SDW then follows immediately and unequivocally from a PAC experiment. The propagation vector **Q** cannot be determined directly. However, in Ref. 4 we deduced the type of antiferromagnetic ordering (AF_1 versus AF_2) from the hyperfine field value, the latter being enhanced in the longitudinal phase.[8] By the present experiments we will prove that indeed the **S** as well as the **Q** orientation can be derived from the observed hyperfine field.

The experimental PAC time spectra are fitted to the expression

$$R(t)=f_{\mathrm{CSDW}}\sum_{n=0,2}a_n e^{-n\delta\nu_C t}\cos(n\nu_C t)$$
$$+f_{\mathrm{ISDW}}\sum_{n=0,2}b_n e^{-n\delta\nu_I t}J_0(n\nu_I t) \qquad (1)$$

to yield the *magnitudes* of the hyperfine fields and their *relative* fractions. The first term accounts for a commensurate antiferromagnetic (AF_0) ordering described by a frequency component with mean value ν_C and Lorentzian distribution width $\delta\nu_C$. The second term accounts for an incommensurate SDW antiferromagnetic ordering (AF_1 or AF_2). The zero-order Bessel function, $J_0(n\nu_I t)$, results from the Overhauser distribution[1,4] in the hyperfine field.

For the PAC experiments the nuclear probe ^{111}In(^{111}Cd) is incorporated in the sample in trace quantities by ion implantation at an energy of 80 keV. For these implantation conditions, the implantation profile of the ^{111}In probes as estimated by the TRIM code (Ref. 14) has a projected range of 18 nm and a longitudinal straggling of 7 nm. In the multilayer sample, therefore, we do not expect to observe a significant signal due to probes in the Fe environment. The PAC time spectrum was measured simultaneously in two detector geometries, different by the actual orientation of the sample normal (the [100] direction) relative to the plane of the detectors. In the perpendicular geometry, the sample normal is perpendicular to the detector plane, whereas in the in-plane geometry the sample normal points at 45° in between the detectors.[4]

The PAC time spectra measured at 77 K on the 250-nm-thick Cr layer together with their simulation according to Eq. (1) are shown in Fig. 2. The main contribution to the experimental spectrum (explicitly shown) is well reproduced by a distribution of the Overhauser type [second term in Eq. (1)] and thus corresponds to an incommensurate spin-density wave. In the 250-nm-thick chromium film we observe a single frequency for this fraction in both geometries. In the

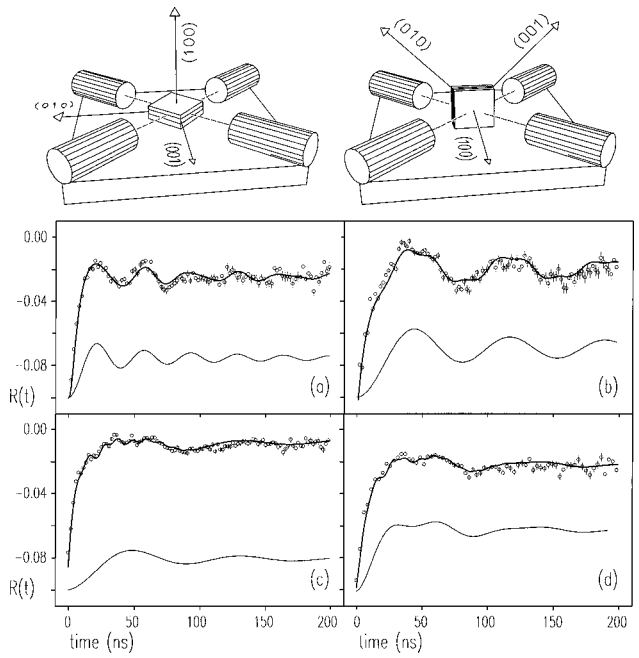

FIG. 2. Comparison of the PAC spectra measured in two detector geometries for (a) and (b) the 250 nm Cr/(50 nm)Nb/Al_2O_3 ($11\bar{0}2$) film, (c) and (d) the $[2\text{ nm Fe}/25\text{ nm Cr}]_{10}$/(50 nm)Nb/$Al_2O_3$ ($11\bar{0}2$) multilayer. The spectra on the left correspond to the out-of-plane geometry; the spectra on the right to the in-plane geometry. The main contribution arising from the chromium incommensurate SDW has been explicitly shown. A minor contribution is due to a volume fraction in a commensurate SDW ordering.

perpendicular [Fig. 2(a)] and in-plane geometry [Fig. 2(b)] we observe the second and the first harmonic, respectively. This immediately proves that the Cr hyperfine field is oriented along the [100] direction normal to the Cr layer. In contrast, the PAC time spectra for the $[2\text{ nm Fe}/25\text{ nm Cr}]_{10}$ multilayer do *not* correspond to an out-of-plane spin orientation. The precession pattern obtained in the perpendicular geometry [Fig. 2(c)] is approached using the first harmonic only, while the in-plane geometry [Fig. 2(d)] leads to a superposition of the single and double frequency. Therefore, the hyperfine field in the $[2\text{ nm Fe}/25\text{ nm Cr}]_{10}$ multilayer is oriented in plane. This result, in conjunction with the evidence from neutron-diffraction experiments for perpendicular spin orientation in the Cr thin film and in-plane spin orientation in the Fe/Cr multilayer, explicitly proves the collinearity of the hyperfine field with the local magnetization of the matrix. We thus prove that perturbed-angular-correlation spectroscopy unequivocally determines the spin polarization of the SDW.

The interaction frequency in the chromium thin film for the majority contribution at 77 K is 14.3(2) MHz, which agrees closely with the value observed at 77 K in bulk chromium[8] and in Fe/Cr multilayers.[4] This hyperfine field value thus corresponds to the longitudinal spin-density-wave structure. Comparison with the neutron diffraction data further establishes the fact that indeed an enhanced hyperfine field represents a longitudinal spin-density-wave phase. The frequency measured at 77 K in the present $[2\text{ nm Fe}/25\text{ nm Cr}]_{10}$ multilayer is, however, markedly reduced and has a value of 12.1(2) MHz. This hyperfine field value is comparable with the value obtained at 250 K in bulk

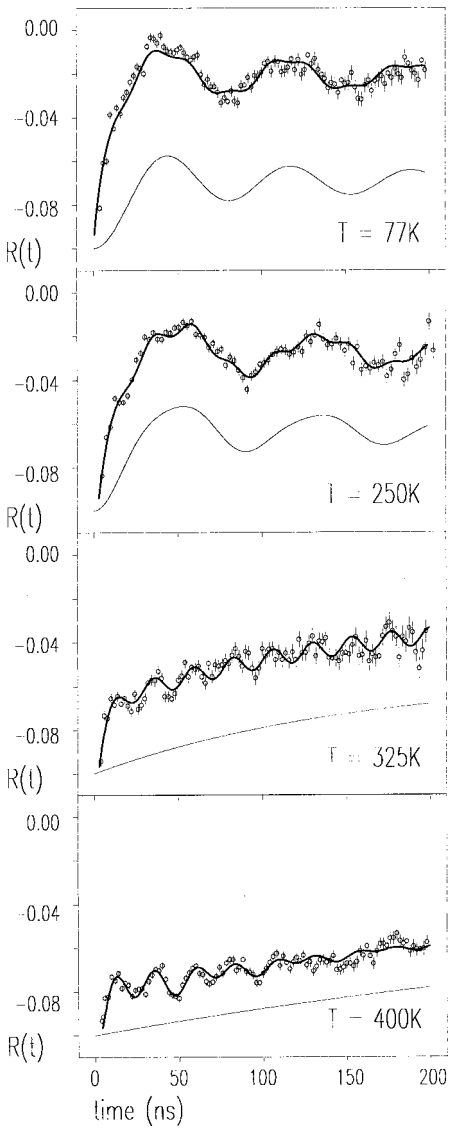

FIG. 3. Temperature dependence of the PAC time spectra for the 250 nm Cr/(50 nm)Nb/Al_2O_3 ($11\bar{0}2$) film in the in-plane geometry measured at (a) 77 K, (b) 250 K, (c) 325 K, and (d) 400 K. The thin lines represent the incommensurate SDW chromium contribution.

chromium[8] and thus indicates the presence of the transverse spin-density-wave phase, in agreement with the neutron-diffraction experiments. Therefore, the type of magnetic ordering of the spin-density wave (AF_1 or AF_2) can thus be deduced from the hyperfine field value.

The spectra measured at different temperatures on the 250-nm-thick Cr film are shown in Fig. 3. The main contribution to the spectra is explicitly shown. Clearly, the pronounced slow precession pattern due to the SDW observed at low temperatures disappears at 325 K. At high temperatures, we observe a rather slow decay corresponding to probes in paramagnetic bcc chromium with a weak nuclear electric quadrupole interaction due to defects or strain in the crystal. Therefore we do not observe an incommensurate-commensurate transition at 330 K. On the contrary, above 325 K the majority of the probes is in an environment with vanishing moments indicating that the majority volume fraction becomes paramagnetic, consistent with the resistivity measurement (Fig. 1).

In the PAC time spectrum obtained at 400 K, a minority fraction is clearly visible as the rapid precession superimposed on the slow signal. This high-frequency modulation corresponds to a commensurate SDW fraction which is definitely also present at lower temperatures (Fig. 3). We obtain from the f_{ISDW} and f_{CSDW} fractions in Eq. (1), independent of the magnetic moments and temperature, the relative volume fractions of 85(5)% incommensurate and 15(5)% commensurate chromium. The minority fraction reflects a substantially enhanced Cr magnetic hyperfine field of 21.1(2) MHz corresponding to a chromium magnetic moment $\mu = 0.90(2)\ \mu_B$. In the temperature interval studied, the hyperfine field due to the minority fraction remains nearly unchanged.

The commensurate antiferromagnetic structure found here has earlier been identified by neutron diffraction[7] and shown to be present in Cr films to a volume fraction depending on thickness and temperature. In single crystalline bulk chromium this phase is not present, whereas in strained polycrystalline chromium, the neutron diffraction experiments of Ref. 2 could not distinguish between the AF_0 or the AF_2 phases. We have shown that perturbed angular correlation spectroscopy is definitely sensitive to the commensurate antiferromagnetic phase, but this phase was not disclosed in our Fe/Cr multilayers grown on MgO studied previously.[4] Further investigations are necessary to clarify the origin of this commensurate phase apparently present in the samples grown for neutron diffraction experiments.

In conclusion, we have shown that PAC experiments determine the magnetic state of chromium and particularly the Cr spin orientation. Although PAC is insensitive to the direction of the Q vector itself, the polarization of the spin-density wave can be determined from the hyperfine field value. The present PAC experiments agree with the neutron-scattering data and indeed confirm the longitudinal incommensurate spin-density-wave ordering with the spins out of plane for the single-layer 250 nm Cr film, while for the Fe/Cr multilayer a transverse SDW with the spins in plane is observed. This consistency proves that the radioactive probes used in perturbed-angular-correlation spectroscopy indeed sense the *intrinsic* chromium magnetism. By virtue of this result we further support the existence of a *longitudinal* SDW with single Q *normal* to the layers for Cr (thickness above 5.0 nm) in Fe/Cr superlattices, MBE grown on MgO, as reported earlier.[4] Interestingly, in the $[2\ \mathrm{nm\ Fe}/25\ \mathrm{nm\ Cr}]_{10}$ multilayer grown for neutron diffraction experiments, a *transversal* SDW with *in-plane* Cr spins is present. In addition, we have recognized the presence of the commensurate antiferromagnetic phase as an enhanced hyperfine field. Further complementary study therefore should aim to explain the difference in both types of Fe/Cr multilayers.

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[1] E. Fawcett, Rev. Mod. Phys. **60**, 209 (1988); E. Fawcett, H. L. Alberts, V. Yu. Galkin, D. R. Noakes, and J. V. Yakhmi, *ibid.* **66**, 25 (1994).

[2] G. E. Bacon and N. Cowlam, J. Phys. C **2**, 238 (1969).

[3] B. Swinnen, J. Meersschaut, J. Dekoster, and M. Rots, J. Magn. Magn. Mater. **140-144**, 543 (1995).

[4] J. Meersschaut, J. Dekoster, R. Schad, P. Beliën, and M. Rots, Phys. Rev. Lett. **75**, 1638 (1995).

[5] E. Fullerton, K. T. Riggs, C. H. Sowers, S. D. Bader, and A. Berger, Phys. Rev. Lett. **75**, 330 (1995).

[6] E. Fullerton, S. D. Bader, and J. L. Robertson, Phys. Rev. Lett. **77**, 1382 (1996).

[7] H. Zabel, A. Schreyer, P. Bödeker, and P. Sonntag, in NATO ASI Series C: Mathematical and Physical Sciences, edited by A. Skjeltorp *et al.* (Kluwer Academic, Dordrecht, in press).

[8] R. Venegas, P. Peretto, G. N. Rao, and L. Trabut, Phys. Rev. B **21**, 3851 (1980).

[9] P. Sonntag, P. Bödeker, T. Thurston, and H. Zabel, Phys. Rev. B **52**, 7363 (1995).

[10] P. Bödeker, P. Sonntag, A. Schreyer, J. Borchers, K. Hamacher, H. Kaiser, and H. Zabel, Physica B **234-236**, 464 (1997); P. Bödeker, P. Sonntag, A. Schreyer, H. Zabel, J. Borchers, K. Hamacher, and H. Kaiser, J. Appl. Phys. **81**, 5247 (1997).

[11] B. M. Geerkens, R. Griessen, G. Benediktsson, H. U. Åström, and C. van Dijk, J. Phys. F **12**, 1603 (1982).

[12] P. Sonntag, P. Bödeker, A. Schreyer, H. Zabel, K. Hamacher, and H. Kaiser, J. Magn. Magn. Mater. (to be published).

[13] P. Bödeker, A. Hucht, A. Schreyer, J. Borchers, F. Güthoff, and H. Zabel (unpublished).

[14] J. F. Ziegler, J. P. Biersack, and U. Littmark, *The Stopping and Ranges of Ions in Solids* (Pergamon, New York, 1985).