Dynamical magnetic properties of Cr in an Fe/Cr(110) multilayer

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The magnetism of a 26-nm-thick Cr spacer layer, in an Fe/Cr(110) multilayer previously characterized via neutron diffraction, is studied with perturbed angular correlation spectroscopy. The existence of a commensurate antiferromagnetic state between 200 K and 500 K, and an incommensurate spin-density-wave state below 200 K is confirmed. In addition, the present results reveal dynamical behavior of the Cr magnetic moments at elevated temperature. The observed spin fluctuations may be essential to understand the transition from the incommensurate antiferromagnetic to the paramagnetic state in thin Cr films.

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Bulk Cr is an itinerant antiferromagnetic material that exhibits a spin-density-wave magnetic ordering below the Néel temperature of 311 K. The spin-density-wave is incommensurate with the lattice and characterized by the propagation direction ($\vec{Q}$) and the polarization ($\vec{S}$). Because chromium is an archetypical itinerant antiferromagnet, the magnetic excitations have been studied intensely in Cr single crystals both theoretically[1] and experimentally.[2–5] It was found that field cooling stabilizes a single $\vec{Q}$ state.[6] In such a field cooled state, inelastic neutron diffraction experiments near the Néel temperature revealed critical fluctuations at incommensurate satellite positions at which no elastic scattering occurs.[7] Apart from this observation commensurate diffuse scattering was also observed.[8] It was concluded that, even at temperatures as high as 500 K, magnetic correlations exist between the Cr spins over distances of several bcc unit cells.[8]

The magnetic properties of Cr thin films and surfaces differ drastically from the magnetic properties of bulk Cr.[9–13] For thin films, the magnetic properties depend on the layer thickness, the strain, and the type of boundary layers. Until now, very little is known about the *dynamical* magnetic properties of Cr thin films. Due to the lack of scattered intensity, inelastic neutron diffraction on thin Cr films is extremely difficult. Recently, we demonstrated that one can apply perturbed angular correlation spectroscopy to study the dynamical magnetic properties of Cr in Fe/Cr multilayers and trilayers.[14,15] In the present paper we apply this approach to an Fe/Cr(110) multilayer.

The multilayer is grown at 470 K on a V(110) substrate and consists of 15 repetitions of Cr(26.0 nm)/Fe(1.2 nm) bilayers, capped with 10.0 nm Cr. The same sample was previously characterized via neutron diffraction.[16] From the temperature dependence of the (100) Bragg reflection intensity it was concluded that the Cr spacer below 200 K exhibits an incommensurate spin-density-wave ordering (ISDW). Above 200 K the commensurate antiferromagnetic ordering (AF_0) is the most prominent magnetic phase, while above 450 K, the Cr spacer gradually becomes paramagnetic [see Fig. 2(a)]. In the present work we prove that, in thin Cr films, apart from the static magnetic properties, fluctuations of the magnetic spin structure are also present. We identify the *transition* from the commensurate antiferromagnetic ordering to the paramagnetic state as a dynamical process in which the Cr magnetic moments start to fluctuate.

In order to study the magnetic properties of the Cr spacer using perturbed angular correlation (PAC) spectroscopy, $^{111}In^+$ ions are implanted in the Fe/Cr multilayer with the Leuven Isotope Separator. The $^{111}In^+$ was implanted at energies ranging from 70 to 80 keV. It was verified with SRIM calculations that the probes end up mainly in the top Cr spacer layer.[17] Previous investigations show that after ion implantation, the ^{111}In takes a regular substitutional position in the metal lattice.[9] After each ion implantation, a PAC spectrum was recorded at room temperature and we did not observe a change in the magnetic properties. The ^{111}In has a half life of 2.8 days, and decays through electron capture to the 7/2+ excited state of ^{111}Cd.[18] The $^{111}In/^{111}Cd$ is a suitable probe to perform PAC spectroscopy. PAC spectroscopy is able to probe the magnetic structure through the Larmor precession of the ^{111}Cd nuclear magnetic moment in the local magnetic hyperfine field. Since the latter is proportional to the magnetic moments on the surrounding atoms, PAC is sensitive to the local magnetic ordering around the probe nuclei. For a more detailed description of PAC we refer to Refs. 19 and 20.

The commensurate antiferromagnetic ordering results in a single hyperfine field, corresponding to a simple cosine function in the PAC spectrum. An incommensurate spin-density-wave ordering is characterized by an Overhauser distribution of hyperfine fields, corresponding to a zeroth order Bessel function in the PAC spectrum.[21] The influence of fluctuations on the PAC spectra is observed as a gradual damping of the oscillations. In the numerical analysis, the fluctuations are described by a stochastic model for fluctuating hyperfine fields.[22,23] The fluctuations are parametrized by a hyperfine field fluctuation time τ, which is the average time a magnetic moment is observed in a certain state. For very slow spin fluctuations, the model leads to the well known cosine or Bessel function in the PAC spectrum, i.e., the static case. When the spin fluctuation frequency is comparable to the hyperfine field frequency, a damped cosine or Bessel function is observed in the PAC spectrum. Finally, for very fast

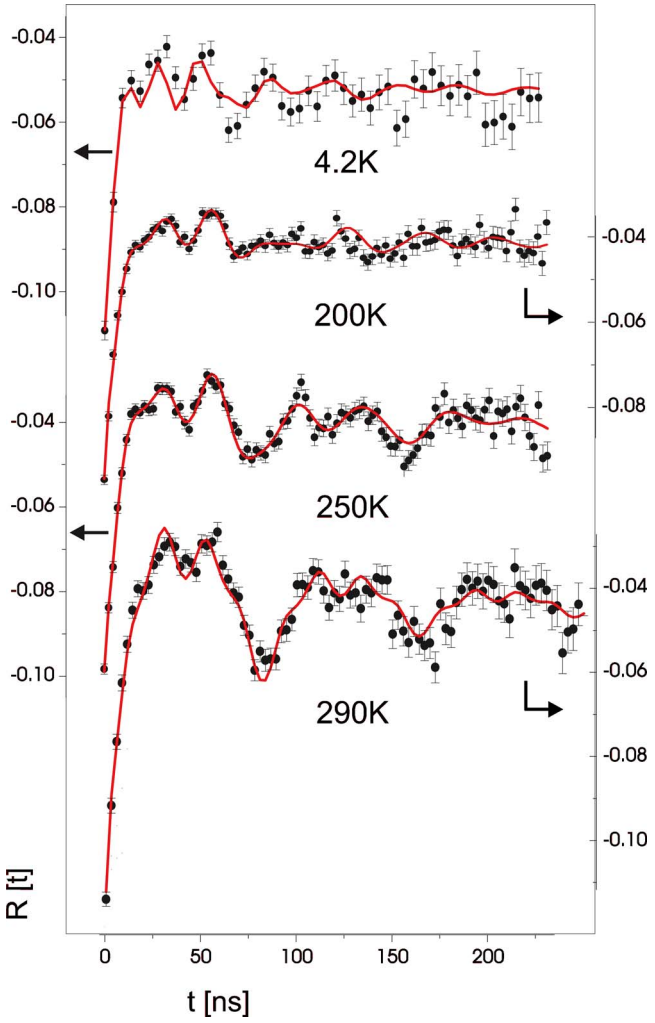

FIG. 1. (Color online) Selection of perturbed angular correlation measurements recorded between 4.2 and 290 K (see Ref. 24). The dots are the data and the solid line corresponds to a fit through the data. The data illustrate the transition from the incommensurate spin-density-wave phase, which is present up to 200 K, to the commensurate antiferromagnetic phase at 290 K.

spin fluctuations, the nuclear probes can no longer follow the fast switching hyperfine field and a nonmagnetic signal without oscillations is observed in the PAC spectrum; this is the paramagnetic case.

Figure 1 shows a selection of PAC spectra taken between 4.2 and 290 K. Up to 200 K, the general shape of the spectra, and thus also the magnetic state in the Cr spacer, remains unchanged. Between 200 and 290 K the spectra change, reflecting a change in the magnetic state of the Cr spacer.

All PAC spectra could be fitted consistently by applying a two component model: a Bessel function of zeroth order and a cosine function. The spectra taken up to 200 K are essentially described by a Bessel function of zeroth order. Hence, from 4.2 up to 200 K an ISDW is present in the Cr spacer. At 290 K, the spectrum is solely described by a cosine function. This reflects the presence of the AF_0 state at high temperature. Between 200 and 290 K the PAC spectra can be described by a combination of a cosine and a zeroth order Bessel function. The relative fraction of the cosine function increases (while the relative fraction of the Bessel function decreases) with increasing temperature, indicating a gradual transition from the ISDW to the AF_0 phase. From the fits of the PAC spectra, the volume fraction of the various contributions could be determined.

In Fig. 2, the temperature dependence of the magnetic phases in the Cr spacer as obtained from PAC spectroscopy [panel (b)] is compared to the results obtained from neutron diffraction measurements [panel (a)]. To allow a more straightforward comparison between both techniques, the PAC results are presented as a weighted intensity, obtained by multiplying the volume fraction of the magnetic state with the strength of the hyperfine field (also obtained from the PAC fits). This weighted intensity compares to the relative amount measured by neutron diffraction. Both techniques locate the phase transition from the ISDW to the AF_0 phase between 200 and 290 K. Also, the transition from the AF_0 phase to the paramagnetic phase is observed at the same temperature by both techniques. The only noticeable difference between both results is that at low temperatures, up to 200 K, a small fraction of the AF_0 phase is detected by PAC [panel (b)] while this is not observed by neutron diffraction [panel (a)]. However, PAC spectroscopy probes only the top Cr spacer, whereas neutron diffraction probes the whole multilayer stack. It could be that the magnetic properties of the Cr spacer layers are not homogeneous over the whole multilayer stack, explaining the small difference between both results. Apart from that, PAC and neutron diffraction agree very well on the phase diagram for the Cr spacer.

In Fig. 2, we also show the temperature dependence of the hyperfine field [panel (c)]. The experimental results are well reproduced by the scaling law $B_{\mathrm{hf}}(T)=B_{\mathrm{hf}}(0)\left(1-\frac{T}{T_N}\right)^{\beta}$. The fit yields a Néel temperature $T_N=320(20)$ K for the ISDW phase, and a much higher Néel temperature of 451(7) K for the AF_0 phase. The critical parameter β is found to be 0.29(2) for both phases.

In Fig. 3, a selection of PAC spectra illustrates the transition from the commensurate antiferromagnetic phase to the paramagnetic phase. Above 290 K the oscillations in the spectra become more and more damped as the temperature increases. At 450 K, when the Néel temperature of the Cr spacer is reached, the oscillations have completely disappeared.

A static antiferromagnetic state results in an un-damped oscillatory pattern of the PAC spectrum. Therefore, a consistent analysis of the spectra in Fig. 3 excludes a static antiferromagnetic state. Instead, the fluctuation model mentioned above was used for the AF_0 phase, yielding excellent agreement with the data. We can thus conclude that the PAC measurements identify strong spin fluctuations in the transition from the commensurate antiferromagnetic phase to the paramagnetic phase. The spin fluctuation times τ determined from the analysis are indicated in Fig. 3.

Figure 4 shows the temperature dependence of the spin fluctuation time in the AF_0 phase. Perturbed angular correlation spectroscopy is most sensitive to spin fluctuation frequencies that are comparable to the hyperfine interaction frequency, i.e., the Larmor precession frequency. As a result, only between 290 K ($\tau=200(-50/+170)$ ns) and 450 K (τ

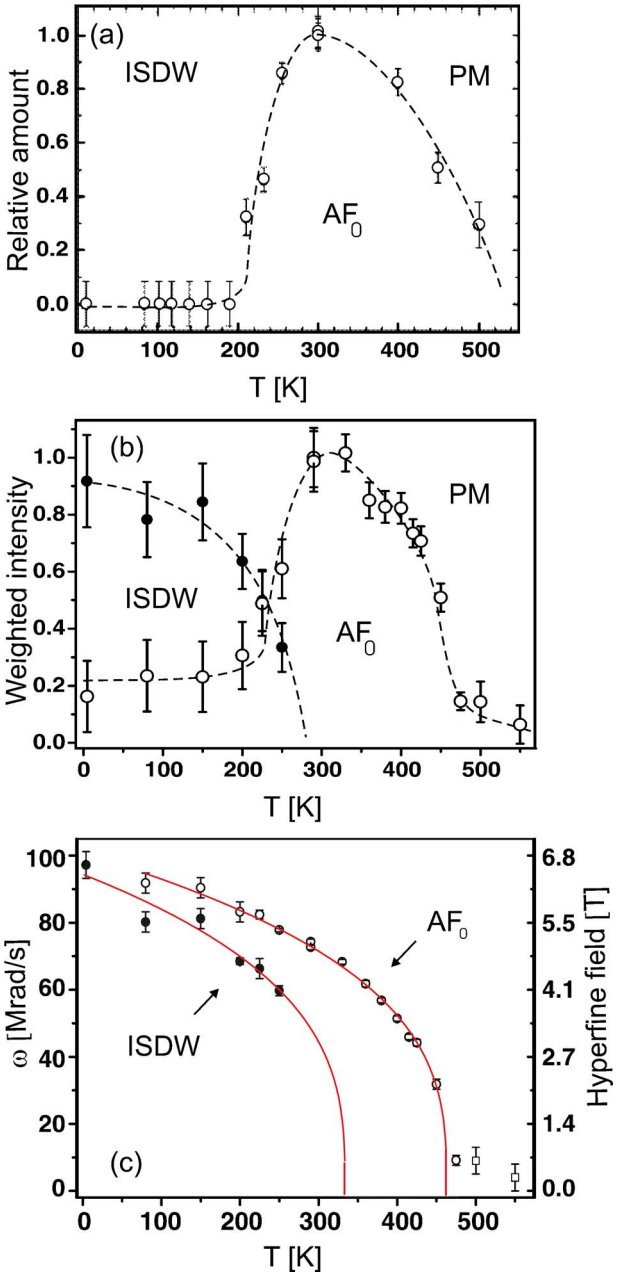

FIG. 2. (Color online) (a) Normalized intensity of the commensurate antiferromagnetic (AF_0) phase in the Cr spacer as a function of temperature, determined by neutron diffraction. (This figure is a reproduction from Ref. 16.) (b) Weighted intensity for the incommensurate spin-density-wave (ISDW) (●) and commensurate antiferromagnetic (AF_0) phase (○), determined by perturbed angular correlation spectroscopy. The dashed lines are a guide to the eye. (c) Hyperfine interaction frequency ω (and the corresponding hyperfine field) as a function of temperature, extracted from the perturbed angular correlation spectra [ISDW (●), AF_0 (○)]. The line through the data corresponds to a fit.

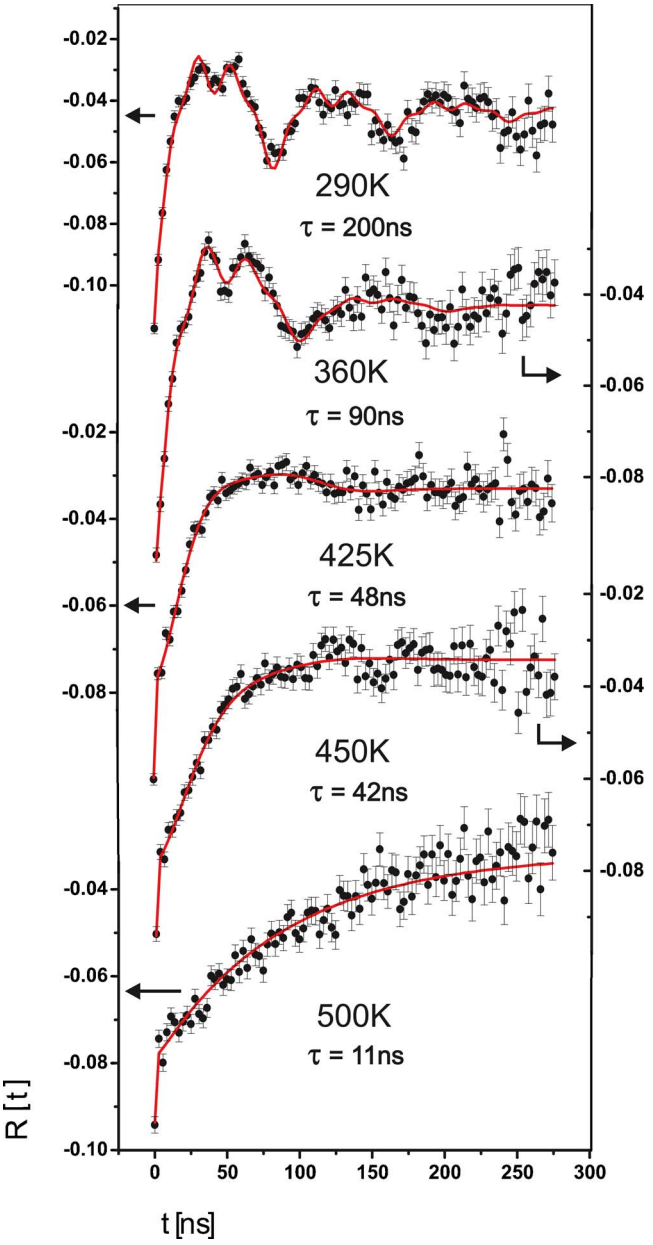

FIG. 3. (Color online) Selection of perturbed angular correlation spectra measured between 290 and 500 K. The dots are the data and the solid line corresponds to a fit through the data. The spectra clearly illustrate damping of the oscillations due to dynamical spin fluctuations. The characteristic fluctuation time is shown for each spectrum.

$=42(-10/+15)$ ns) can accurate information on the fluctuation times be extracted. Below 290 K, a damping of the spectra is already present, but the fluctuations are too slow to be quantized by PAC spectroscopy.

As shown in Fig. 4, a thermal activation process with an activation energy $E_a=109(15)$ meV describes the temperature dependence of the experimentally determined spin fluctuation times well. The interpretation of the obtained activation energy and the origin of the observed spin fluctuations in thin Cr films require further investigation.

When the thickness of the Cr film is reduced below a critical value, the incommensurate spin-density-wave collapses, and either the paramagnetic state[9] or the commensurate antiferromagnetic state remains.[25] Until now, the collapse of the ISDW was attributed to finite size effects. We would like to point out that similar dynamics of the Cr mo-

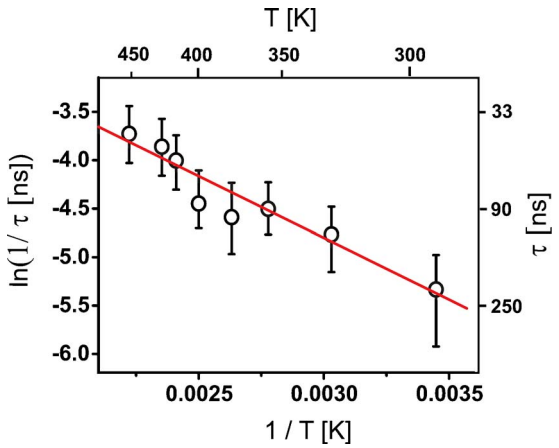

FIG. 4. (Color online) The $\ln(\tau^{-1})$ plotted as a function of T^{-1} for the commensurate antiferromagnetic phase extracted from the perturbed angular correlation spectra.

ments as observed here are also observed in Fe/Cr(100) multilayers.[14,15] This may be a hint that spin fluctuations are a general phenomenon in thin Cr films. However, further theoretical work is needed to support this proposition.

Perturbed angular correlation spectroscopy and neutron diffraction spectroscopy probe dynamical processes on a different time scale. While PAC is sensitive to dynamics in the MHz-GHz regime, neutron diffraction is sensitive to dynamics in the GHz-THz regime. Hence, when PAC observes spin fluctuations, neutron diffraction yields a signal compatible with a static magnetic ordering. This explains the different behavior in the phase diagram of Fig. 2 around 450 K.

In conclusion, where inelastic neutron diffraction becomes very challenging, due to the small scattering volume, perturbed angular correlation spectroscopy can be used as an alternative method to study the dynamics present in low dimensional systems. The present results reveal fluctuating magnetic moments in the Cr spacer of an Fe/Cr(110) multilayer. The Cr magnetic moments fluctuate in the MHz-GHz regime at the magnetic phase transition from the commensurate antiferromagnetic to the paramagnetic state.

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