High-temperature biquadratic coupling in Fe/Cr trilayers

D. Aernout,[a] S. M. Van Eek, B. Croonenborghs, C. L'abbé, M. Rots, and J. Meersschaut
Instituut voor Kern- en Stralingsfysica, K.U. Leuven, Celestijnenlaan 200D, B-3001 Leuven, Belgium

(Received 28 July 2003; accepted 16 September 2003)

We report on the study of epitaxial Fe/Cr trilayers with a Cr thickness of 8 nm grown at different temperatures. We show that it is possible to produce systems that exhibit interlayer exchange coupling above room temperature, while being uncoupled at lower temperatures. Through perturbed angular correlation measurements, this macroscopic effect is identified as a direct result of the growth induced characteristics of the antiferromagnetic spin density wave ordering in the Cr spacer. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1625796]

A good knowledge of the characteristics of the coupling between magnetic thin films in multilayers is of both fundamental and technical importance. In the present work, we demonstrate that it is possible to fabricate multilayer systems that are interlayer coupled *above* room temperature, while being uncoupled below. It is achieved on Fe/Cr/Fe trilayers, in which the magnetic properties of the Cr spacer can be altered by varying the growth conditions. When the samples are grown at room temperature, the Néel ordering temperature of the Cr spacer layer is enhanced with respect to the bulk value and, correspondingly, the onset of biquadratic coupling is shifted to high temperatures.

When the magnetic layers are coupled through nonmagnetic spacer layers, then the interlayer coupling strength shows a monotonous decrease with temperature.[1] A substantially different behavior occurs when the spacer layer itself exhibits long-range magnetic ordering. This can be exploited in the Fe/Cr multilayer system, as bulk Cr metal orders antiferromagnetically below the Néel temperature of 311 K.[2] Indeed, both theoretical studies and experimental results already indicated that the spin density wave antiferromagnetic ordering within the Cr spacer layer drastically changes the interlayer coupling in Fe/Cr trilayers and multilayers.[3,4] In Fe/Cr multilayers grown on MgO, the biquadratic interlayer coupling was found to be suppressed for Cr thicknesses larger than 4.2 nm when Cr orders antiferromagnetically, i.e., below its (reduced) Néel temperature.[5] Above the Néel temperature, fluctuating spins are present, acting as loose spins[6] and mediating the biquadratic interlayer coupling.[7]

Generally, the Néel temperature is reduced for thin Cr layers, and approaches the bulk value asymptotically with increasing thickness. We will demonstrate that the Cr spacer layer turns out to be considerably strained when the Fe/Cr/Fe trilayers are produced at room temperature, and this strain leads to an enhanced Néel temperature of the Cr spacer. Associated with the enhanced ordering temperature, an onset temperature for the biquadratic interlayer coupling of 380 K is found.

We studied Fe (4 nm)/Cr (8 nm)/Fe (4 nm) trilayers, epitaxially grown with molecular beam epitaxy on MgO(001) substrates (base pressure 2×10^{-11} Torr). A thin capping layer against oxidation was deposited at room temperature. The epitaxy was monitored with *in situ* reflection high-energy electron diffraction. The structural quality was confirmed with X-ray diffraction and Rutherford backscattering spectroscopy. For the magnetization measurements an Oxford vibrating sample magnetometer with continuous flow cryostat and oven was used. Several trilayers, grown at different temperatures, were studied. For intermediate growth temperatures (450 K) we found antiferromagnetic ordering temperatures and onset temperatures for biquadratic coupling in good agreement with previous results.[5] The samples grown at room temperature and at elevated temperature, however, show a different behavior. In this letter, we will focus on these results and specifically compare two samples, one grown at room temperature (290 K), and another grown at high temperature (575 K).

Magnetization measurements on the two samples are presented in Fig. 1. For the Fe/Cr/Fe trilayer grown at room temperature, square hysteresis loops are observed up to 380 K. Noncoupled or ferromagnetic coupled systems exhibit a remanent magnetization equal to the full saturation magnetization. Above 380 K, a step is visible in the hysteresis curve and the remanent magnetization drops to one half of the saturation magnetization. This indicates that the trilayer couples biquadratically above 380 K. The Fe/Cr/Fe trilayer grown at high temperature behaves differently: it exhibits square hysteresis curves at very low temperatures, it is biquadratically coupled at 130 K, and it has again square hysteresis loops at room temperature. The bottom panel in Fig. 1 shows the temperature dependence of the remanent magnetization for the two samples. The results are substantially different. In particular, they show a different onset temperature for the biquadratic coupling. The observed onset temperatures are 380(10) and 100(10) K for the sample grown at room temperature and at high temperature, respectively. In the following, we will show that the onset temperature for the biquadratic interlayer coupling is related to the magnetic state of the Cr spacer.

To study the magnetic state of the Cr spacer layer, we used perturbed angular correlation spectroscopy. In this technique, one observes the time evolution that corresponds to the Larmor precession of the nuclear spin in the local hyperfine field. The hyperfine fields are detected at the $(^{111}\mathrm{In})^{111}\mathrm{Cd}$ probe nuclei that are implanted into the Cr spacer layer with an ion mass separator at 50 keV. The spectra were recorded in a high-resolution fast-slow coincidence setup with two mutual orthogonal four-detector combina-

[a]Electronic mail: dominique.aernout@fys.kuleuven.ac.be

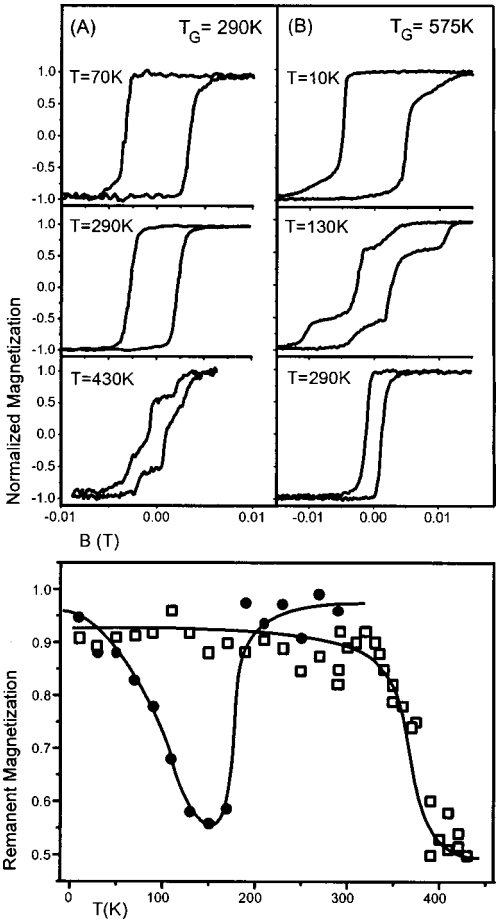

FIG. 1. Hysteresis curves measured at different temperatures for the trilayers grown at room temperature (left) and high temperature (right). Below: the temperature dependence of the remanent magnetization for the samples grown at room temperature (open squares) and high temperature (solid circles). The line is a guide to the eye.

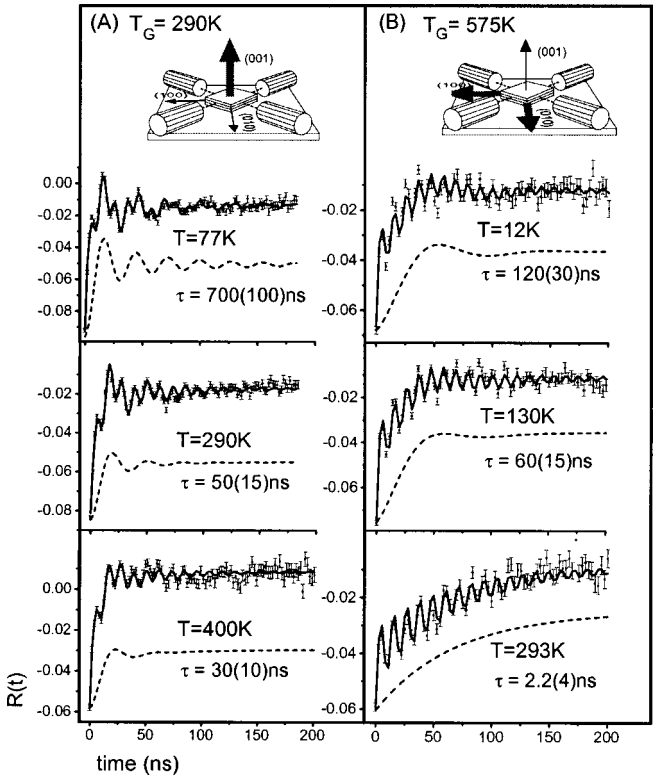

FIG. 2. Perturbed angular correlation spectra recorded at different temperatures for the samples grown at room temperature (left) and high temperature (right). On top, a schematic view of the experimental setup is shown. The direction of the Cr magnetic moments are indicated by the bold arrow. The dots are the data while the solid line through the data is a fit. The dashed line shows the contribution of the Cr. The temperature at which the spectra were measured are indicated, as well as the corresponding spin fluctuation times τ obtained from the analysis.

tions. From the coincidence spectra the anisotropy ratio $R(t)$ is constructed.[8] The method can quantify the magnitude of the local hyperfine field, the fraction of different hyperfine interaction contributions, and the orientation of the hyperfine field.[9,10] Since the magnetic hyperfine fields are related to the magnetic moments, perturbed angular correlation spectroscopy can determine the magnetic state of the probed layer on a microscopic scale. The chromium contribution to the perturbed angular correlation spectra is analyzed using a spin-density-wave fluctuation model. This model is based on the stochastic theory for fluctuating hyperfine fields and accounts for the Overhauser distribution that is characteristic for the spin density wave ordering.[11] Here the spin fluctuation time τ describes the average time a spin is observed in a certain state. For the static incommensurate spin density wave state, i.e., with very large spin fluctuation times τ, the model leads to the Bessel function of zeroth order to describe the experimental perturbed angular correlation spectrum. In the paramagnetic case, when the spin fluctuations are faster than the hyperfine interaction frequency, a very slow decay of the anisotropy ratio $R(t)$ is obtained. In addition, the model allows to analyze the *transition* from the static antiferromagnetic ordering to the paramagnetic state through a dynamical process in which the Cr magnetic moments start to fluctuate.

The perturbed angular correlation spectra recorded at different temperatures are shown in Fig. 2. The fast and the slow frequency in the spectra correspond to the Fe and Cr contribution, respectively. The orientation of the Cr magnetic moment was deduced from the ratio of the single to double harmonic of the Larmor precession. For the sample grown at room temperature, the double harmonic is observed in the geometry depicted in Fig. 2(a), while in the orthogonal detector geometry (not shown) the single harmonic is observed. These results prove that the hyperfine field is oriented out of the plane of the film for the trilayer grown at room temperature. In an analogous way, we found that the Cr spins of the sample grown at high temperature are in the plane. The temperature dependence of the Cr contribution is also markedly different for the two samples. At low temperatures the Cr contribution in both samples shows a clear oscillation, described by a Bessel function. This confirms the presence of an incommensurate spin density wave. For the sample grown at high temperature, the oscillation in the Cr contribution has disappeared at 293 K, which indicates that a transition from the incommensurate spin density wave (12 K) to the paramagnetic phase (293 K) has occurred. The dynamical behavior of this transition is clearly expressed by the strong damping in the Cr contribution for increasing temperature resulting in a dynamical paramagnetic contribution at high temperature.[7] For the trilayer grown at room temperature, this transition occurs at a much higher temperature. Note that in perturbed angular correlation spectroscopy the commensurate antiferromagnetic ordering is not described by a Bessel function and is characterized by a high magnetic hy-

perfine field and remarkably high transition temperature.[12] Whereas a static commensurate antiferromagnetic ordering was believed to mediate the biquadratic coupling,[13,14] this phase was clearly not observed in our samples. Our results indicate the presence of an incommensurate spin density wave ordering at low temperatures and a paramagnetic phase at high temperatures.

From the perturbed angular correlation experiments, we conclude that the growth at room temperature produces a magnetic state in Cr characterized by spins out of plane and the transition to the paramagnetic state above room temperature. The growth at high temperatures, on the other hand,[9] produces a magnetic state in Cr with spins in plane and a transition to the paramagnetic state far below room temperature. Moreover, for both samples, spin fluctuation rates in the order of 20 MHz are observed at the onset for the biquadratic coupling (Fig. 2). This clearly demonstrates that the interlayer coupling in Fe/Cr/Fe trilayers is closely related to the microscopic magnetic properties of the Cr spacer. An explanation for the different magnetic states in the Cr spacer is to be found in the structural properties of the layers.

The tetragonal distortion ($e^T = e^{\parallel} - e^{\perp}$) for the Fe and Cr layers was determined by measuring the channeling along the [111] crystal axis using Rutherford backscattering spectroscopy. The [111] axis of the bcc Cr lattice was observed at an angle of 55.10(4)° with respect to the normal [001] direction for the sample grown at room temperature. This corresponds to a tetragonal distortion of 1.15(15)% in the Cr spacer. Analogously, for the sample grown at high temperature, a tetragonal distortion of $-0.25(6)\%$ was obtained. The same behavior of decreasing strain with increasing growth temperature was also observed for the Fe layers. This indicates that the tetragonal distortion due to the lattice misfit of the Fe/Cr structure with the substrate is reduced with increasing growth temperature.

Two mechanisms were proposed to explain the out-of-plane orientation of the Cr moments: the strain present in the Cr layer on the one hand, and magnetic frustration at the Fe/Cr interface on the other hand.[4,15,16] Strain in thin Cr layers was found to enhance the Néel temperature of the layer.[17] Since a considerable strain is present in the Fe/Cr trilayer grown at room temperature, we can conclude that the strain is the most convincing explanation for the out-of-plane orientation of the Cr moments and the enhanced magnetic ordering temperature. However, for the sample grown at high temperature, the onset temperature for the biquadratic coupling is 100(10) K. This is much lower than the expected value of 200 K for similar Cr thicknesses.[5] Such a reduction of the onset temperature for the biquadratic coupling was observed previously and explained by diffusion of Fe into the Cr spacer.[18] Interdiffusion of Fe in the Cr spacer is highly probable in the case of epitaxial growth at high temperature.[19] We also observe that the biquadratic coupling suddenly disappears in a narrow temperature region around 190 K. This behavior is reminiscent of the one observed for Fe/Cr superlattices in which the Cr was alloyed with 6% of Fe.[20]

Our results emphasize the intimate relation between the magnetic state of the Cr spacer layer and the biquadratic interlayer coupling between the Fe layers. Biquadratic interlayer coupling appears when the Cr spacer goes from the incommensurate spin density wave ordering to the paramagnetic phase. The ordering temperature of the Cr, depends on the growth temperature. By reducing the growth temperature, the magnetic properties of the Cr spacer can be altered in such a way that the onset of biquadratic coupling appears above room temperature. Based on structural investigations, we attributed the modified magnetic properties of the Cr spacer to the epitaxial strain.

This work was supported by the F.W.O.-Vlaanderen Project Nos. G.0194.00 and G.0137.95. The authors are grateful to T. Ślęzak, D. Wilgocka, A. Vantomme, and H. Guérault for help in samples preparation and characterization. S.M.V.E. was supported by the Research Council of K.U.L. (F-00-040). J.M. and C.L. thank the Fund for Scientific Research Flanders, Belgium (F.W.O.-Vlaanderen) for financial support.

[1] P. Bruno, Phys. Rev. B **52**, 411 (1995).
[2] E. Fawcett, Rev. Mod. Phys. **60**, 209 (1988).
[3] D. T. Pierce, J. Unguris, R. J. Celotta, and M. D. Stiles, J. Magn. Magn. Mater. **200**, 290 (1999).
[4] R. S. Fishman, J. Phys.: Condens. Matter **13**, R235 (2001).
[5] E. E. Fullerton, K. T. Riggs, C. H. Sowers, S. D. Bader, and A. Berger, Phys. Rev. Lett. **75**, 330 (1995).
[6] J. C. Slonczewski, J. Appl. Phys. **73**, 5957 (1993).
[7] J. Meersschaut, C. L'abbé, M. Rots, and S. D. Bader, Phys. Rev. Lett. **87**, 107201 (2001).
[8] G. Schatz and A. Weidinger, *Nuclear Condensed Matter Physics*, 2nd ed. (Wiley, New York, 1996), p. 63.
[9] J. Meersschaut, J. Dekoster, R. Schad, P. Beliën, and M. Rots, Phys. Rev. Lett. **75**, 1638 (1995).
[10] J. Meersschaut, J. Dekoster, S. Demuynck, S. Cottenier, B. Swinnen, and M. Rots, Phys. Rev. B **57**, R5575 (1998).
[11] C. L'abbé, J. Meersschaut, M. Rots, and S. D. Bader, J. Appl. Phys. **93**, 8250 (2003).
[12] P. Peretto, R. Venegas, and G. N. Rao, Phys. Rev. B **23**, 6544 (1981).
[13] A. Schreyer, C. F. Majkrzak, T. H. Zeidler, T. Schmitte, P. Bödeker, K. Theis-Bröhl, A. Abromeit, J. A. Dura, and T. Watanabe, Phys. Rev. Lett. **79**, 4914 (1997).
[14] T. Schmitte, A. Schreyer, V. Leiner, R. Siebrecht, K. Theis-Bröhl, and H. Zabel, Europhys. Lett. **48**, 692 (1999).
[15] S. Demuynck, J. Meersschaut, J. Dekoster, B. Swinnen, R. Moons, A. Vantomme, S. Cottenier, and M. Rots, Phys. Rev. Lett. **81**, 2562 (1998).
[16] M. Almokhtar, K. Mibu, and T. Shinjo, Phys. Rev. B **66**, 134401 (2002).
[17] P. Sonntag, P. Bödeker, A. Schreyer, H. Zabel, K. Hamacher, and H. Kaiser, J. Magn. Magn. Mater. **183**, 5 (1998).
[18] J. Dekoster, J. Meersschaut, S. Hogg, S. Mangin, E. Nordström, A. Vantomme, and G. Langouche, J. Magn. Magn. Mater. **198–199**, 303 (1999).
[19] D. Venus and B. Heinrich, Phys. Rev. B **53**, R1733 (1996).
[20] E. E. Fullerton, C. H. Sowers, and S. D. Bader, Phys. Rev. B **56**, 5468 (1997).