Unusual magnetic susceptibility and magnetoresistance in [Fe/Cr(001)]₁₀ multilayers at low temperatures

F. G. Aliev,^{a)} R. Guerrero, and V. V. Pryadun

Departamento Fisica de la Materia Condensada C-III, Instituto Nicolás Cabrera de Ciencia de Materiales, Universidad Autonoma de Madrid, 28049, Madrid, Spain

J. L. Martinez

Instituto de Ciencia de Materiales Madrid, Consejo Superior de Investigaciones Científicas (CSIC), 28049 Cantoblanco, Madrid, Spain

R. Villar

Departamento Fisica de la Materia Condensada C-III, Instituto Nicolás Cabrera de Ciencia de Materiales, Universidad Autonoma de Madrid, 28049, Madrid, Spain

(Presented on 10 November 2004; published online 4 May 2005)

Transport and magnetic properties near a field-induced orientation phase transition (OPT) from the easy to the hard axis in the magnetization of antiferromagnetically coupled epitaxial $[Fe/Cr(100)]_{10}$ multilayers have been studied. It was found that in the vicinity of the OPT both the amplitude of the magnetic susceptibility anomaly and the relaxation rate of the magnetoresistance change dramatically below a few Kelvin. These observations, together with the previously observed strong reduction of zero-field magnetic losses at temperatures below 5–7 K, indicate a qualitative transformation of the magnetic dynamics in Fe/Cr multilayers at very low temperatures. © 2005 American Institute of Physics. [DOI: 10.1063/1.1847992]

Recently, a number of unexpected phenomena have been reported to occur in antiferromagnetically coupled magnetic multilayers at very low temperatures. One example is a magnetic-field-tuned quantum phase transition (QPT) at very low temperatures¹ near the region of the field-induced transition between antiferromagnetic and ferromagnetic order in Fe/Cr multilayers. Another example is the unexpected behavior of the low-frequency magnetic dynamics of antiferromagnetically coupled multilayers-a property poorly explored up to now. An investigation of the magnetic susceptibility of epitaxial antiferromagnetically coupled $[Fe/Cr(100)]_{10}$ multilayers reported a dramatic change in the low-frequency magnetic losses at temperatures below 10 K.² The origin of these unusual phenomena remains unclear. One of the possible explanations could be quantum tunneling between two degenerate opposite orientations of the Néel vector (equal to the difference between the magnetizations of the two sublattices), which may occur at zero field in antiferromagnetically coupled nanoclusters formed by neighboring Fe layers within the multilayers.

The magnetic dynamics in epitaxial Fe/Cr multilayers may be also probed at fields close to the orientation phase transition (OPT) between the easy and hard axes. We were able to detect precisely from the relaxation measurements a noticeable change of the magnetoresistance near the OPT, due either to a change in the average angle between coupled layers or to a rotation of the magnetization of biquadratically coupled (BC) domains (or caused by both factors). In the case of ferromagnetic systems, previous direct magnetization relaxation measurements have proved to give rich information on magnetic dynamics.³ However, in antiferromagnetically coupled multilayers the total magnetization is very small due to nearly complete compensation. Therefore, direct magnetization relaxation experiments give a reduced signal-to-noise ratio in comparison with magnetoresistance relaxation. A recent magnetic after-effect study at very large time scales (up to 10^5 s) in Fe/Cr multilayers revealed some features in the ultralow-frequency magnetization dynamics such as BC domain rotation at ultralow temperatures.⁴

Here we compare magnetic (ac susceptibility) and transport (high-resolution low-field magnetoresistance and resistance relaxation) properties of $[Fe/Cr]_{10}$ multilayers when the magnetic field, which is applied along the (110) direction [i.e., along the hard axis (HA)], induces a change in the magnetization orientation of the coupled Fe layers from the easy ($H < H_{OPT}$) to the hard ($H > H_{OPT}$) axis.

The magnetic properties have been studied by using the ac option of a physical properties measurement system (PPMS) (Quantum Design), and measurements have been done at different temperatures above 2 K, with ac drive frequencies below 10 kHz and drive amplitudes below 4 Oe. The dc electron-transport measurements were carried out in a Janis magnetic system. dc electrical resistance was measured on lithographically patterned multilayers with a Keithley 2182 nanovolt meter at a rate of one data point each 4 sec, on times up to 2000 sec, by using the four-point method, and changing the electric current polarity for each data point in order to suppress thermoelectric effects. In this way we could determine the relaxation of the magnetoresistance, and therefore of the magnetization, with a precision better than 10^{-5} . All the studied [Fe/Cr]10 multilayers were grown on MgO(100) substrates and had Fe thicknesses from 12 to 30 Å and Cr thickness of 12 Å, corresponding to the first maximum in antiferromagnetic coupling, providing a giant mag-

0021-8979/2005/97(10)/10C505/3/\$22.50

97, 10C505-1

a)Electronic mail: farkhad.aliev@uam.es



FIG. 1. Magnetoresistance (and its derivative at 50 mK, inset) near the orientation phase transition (OPT) at two different temperatures. The lower inset sketches the relative orientation of the magnetic axes and the applied magnetic field H.

netoresistance (GMR) of about 100%, and saturation fields corresponding to the field-induced parallel state of about 10 kG at 10 K. Details of sample preparation and characterization can be found elsewhere.⁵

First of all we show in Fig. 1 typical data of magnetoresistance in the applied magnetic-field range close to the orientation phase transition measured at two different low temperatures. The inset presents the derivative dR/dH at 50 mK, clearly indicating the strong variation of the resistance induced by the change of orientation of coupled Fe layers. Interestingly, the OPT has been observed both for increasing and decreasing magnetic fields, but its position was somewhat hysteretic. This behavior was also confirmed by the magnetic susceptibility measurements presented in Fig. 2. As in the electron-transport data of Fig. 1, the OPT is clearly observed in the magnetic susceptibility both for increasing (low maxima) and decreasing (large maxima) magnitudes of magnetic field. We have found, however, that only for the field-induced OPT (increasing field) a strong change in the susceptibility anomaly occurs when the temperature decreases below 10 K (see inset which shows the corresponding amplitude of the maximum in magnetic susceptibility), while for the OPT occurring when reducing the magnetic field, this anomaly is nearly independent of temperature.

Next we shall compare the changes in the ac magnetic dynamics with magnetic relaxation observed via magnetoresistance. We shall concentrate on the relaxation rate measured near the field-induced OPT. For the magnetic field along the easy axis (100) the OPT was absent.

Figure 3 presents the variation of the relaxation rate of the magnetoresistance when the magnetic field is tuned through the OPT after cycling it each time between ± 2000 Oe, and fixing the field with the permanent superconducting switch. We have observed that at temperatures above



FIG. 2. Magnetic ac susceptibility at three different temperatures with the applied magnetic field along the hard axis. The right inset expands the data in the region of the OPT. The left inset shows the amplitude of the maxima in the real part of the susceptibility near the OPT as a function of temperature for two different ac drive frequencies.

10 K the normalized relaxation is nearly temperature independent, being logarithmic in time and smaller than 10^{-4} . By using a Hall probe we have proved that this relaxation is mainly due to the relaxation of the magnetic field of the superconducting magnet in the persistent mode, which is logarithmic with time. Therefore, at temperatures above 10 K the main factor contributing to the relaxation is not the magnetization proper but a small variation of magnetic field $dH \sim (-\ln t)$ resulting in the anomaly in the relaxation rate $[dR/d(\ln t)]$ similar to the anomaly in the derivative -dR/dH of the magnetoresistance near the OPT (see inset to Fig. 1). Having in mind that the form of the derivative dR/dH re-



FIG. 3. Normalized magnetoresistance relaxation rate $[dR/d(\ln t)]R^{-1}(0)$, with the time *t* in seconds, as a function of the magnetic field near the OPT for different temperatures (FC stands for field cooled). The inset shows the dependence of the relaxation rate on temperature for a fixed magnetic field near the OPT.

mains nearly the same for the whole studied temperature range, a qualitative transformation of the relaxation rate versus magnetic field with a maximum above 10 K to a minimum below 3.5 K indicates an increase in the relaxation rate of the magnetoresistance proper, due to the appearance at low enough temperatures of new relaxation channels in the magnetization near the OPT.

We have also studied the influence of the degree of antiferromagnetic coupling on the magnetoresistance relaxation (not shown). For the $[Fe/Cr]_{10}$ multilayer with the largest magnetoresistance (about 120% at 10 K) the OPT shifts to higher magnetic fields (H_{OPT} of about 700 Oe). The relaxation rate near the OPT is small and also weakly dependent on the temperature between 250 and 10 K, increasing in 1-2 orders of magnitude below 7 K. The sharper OPT allowed us to follow the corresponding coercive field (H_c) as a function of temperature. We found that H_c , being practically temperature independent above 10 K, shows a notable, nearly steplike increase of about 15% below 10 K. In [Fe/Cr]₁₀ multilayers with reduced antiferromagnetic coupling (GMR less than 15% at 10 K) we fail to observe any indication of an OPT. This could be caused by the reduced epitaxy and enhanced interface disorder, suppressing GMR. At the same time a noticeable (1%) resistance change at a small field near 20 Oe, most probably caused by anisotropic magnetoresistance (AMR), allowed us to follow the relaxation rate near the AMR transition. For the uncoupled Fe/Cr multilayer the magnetization relaxation rate was found to be only weakly temperature dependent between 1.8 and 25 K with a tendency to gradually reduce logarithmic relaxation with decreasing temperature. Details on the dependence of the relaxation on the degree of antiferromagnetic coupling will be published elsewhere.6

In order to understand the strong change in the relaxation rate near the OPT below 10 K one could suppose that it is due to the transition from superparamagnetism to magnetic blockage of nanoparticles representing a deviation from perfect epitaxy in the multilayers. This could in part explain the slight enhancement of the coercive field which has been observed below the blocking temperature for nanopatricles.⁷ Other experimental observations, however, contradict this hypothesis. One would expect that the blocked nanoparticles would provide a strong maximum in the ac susceptibility near the blockage temperature. Experimentally we observe only a reduction of the ac susceptibility near the OPT. Also the change in the coercive field is nearly steplike instead of the expected square root on temperature dependence for nanoparticle arrays $H_c = H_c(0)(1 - \text{const}) \sqrt{T}$.⁷ Finally, we note that large antiferromagnetic coupling with better-defined OPT (better epitaxy and reduced structural/magnetic disorder) increases the change in the relaxation rate at low temperature.

Alternatively, the essential change in the magnetic dynamics of antiferromagnetically coupled Fe/Cr multilayers at $T \le 10$ K could be due to a classical-quantum transition in the escape rate of antiferromagnetically coupled Fe/Cr/Fe nanoclusters forming multilayers.8 Such field-induced transition is predicted to be even of the first order for large enough barriers.⁸ This could provoke a well-defined onset of enhanced magnetic relaxation and smearing of the OPT in the ac susceptibility near QPT. Such explanation is strengthened by the previous observation of a sudden decrease of zerofield magnetic losses below 7 K in antiferromagnetically coupled multilayers² and the absence of strongly temperature-dependent relaxation in uncoupled multilayers below 25 K.⁶ Many important questions, however, still remain to be answered. One example is the direction of the change of coercive field with the onset of quantum tunneling of the Néel vector.

The authors thank R. Schad for the preparation of epitaxial Fe/Cr multilayers and A. P. Levanyuk, A. Fert, and W. Wernsdorfer for useful discussions. The work was supported by Spanish MCYT (MAT2003-2600) and CAM (07N/0050/ 2002).

- ¹F. G. Aliev, V. V. Moshchalkov, and Y. Bryunseraede, Phys. Rev. Lett. **81**, 5884 (1998).
- ²F. G. Aliev, J. L. Martinez, V. V. Moshchalkov, Y. Bryunseraede, A. P. Levanyuk, and R. Villar, Phys. Rev. Lett. 88, 187201 (2002).
- ³E. Del Barco, E. del Barco, M. Duran, J. M. Hernandez, J. Tejada, R. D. Zysler, M. Vasquez Mansilla, and D. Fiorani, Phys. Rev. B **65**, 052404 (2002).
- ⁴F. G. Aliev, Ch. Van Haesendonck, and R. Villar, J. Magn. Magn. Mater. **272–276**, 299 (2004).
- ⁵R. Schad, C. D. Potter, P. Belein, G. Vrback, V. V. Moshchalkov, and Y. Bryunseraede, Appl. Phys. Lett. **64**, 3500 (1994).
- ⁶F. G. Aliev *et al.* (unpublished).
- ⁷W. C. Nunes, W. S. D. Folly, J. P. Sinnecker, and M. A. Novak, Phys. Rev. B **70**, 014419 (2004).
- ⁸B. Zhou, J. Q. Liang, and F. C. Pu, Phys. Rev. B 64, 132407 (2001).