PHYSICAL REVIEW B VOLUME 50, NUMBER 5 1 AUGUST 1994-I

Increases in giant magnetoresistance by ion irradiation

David M. Kelly and Ivan K. Schuller
Department of Physics, 0319, University of California, San Diego, La Jolla, California 92093-0319

V. Korenivski and K. V. Rao
Department of Condensed Matter Physics, Royal Institute of Technology, 10044 Stockholm, Sweden

Kim K. Larsen and J. Bottiger
Institute of Physics and Astronomy, University of Århus, DK-8000 Århus, Denmark

E. M. Gyorgy and R. B. van Dover
AT&T Bell Laboratories, Murray Hill, New Jersey 07974

(Received 28 February 1994)

We have studied the effect of 500-keV Xe^+ irradiation on the structure, magnetotransport, and magnetic properties of sputtered Fe/Cr multilayers. Initially, with increasing dosage the giant magnetoresistance (GMR) increases with a concomitant increase in the interfacial roughness as indicated by changes in the low-angle x-ray-diffraction spectra. At doses higher than 1×10^{13} ions/cm^2, the value of the GMR progressively decreases, corresponding to progressively increasing regions of the film which are ferromagnetically coupled. The resulting structural disorder increases the overall electronic scattering rate and hence the electrical resistance at these higher doses. These results demonstrate that increased interfacial scattering enhances the GMR and thus plays an important role in the mechanism determining the GMR in Fe/Cr superlattices.

Giant magnetoresistance (GMR) in magnetic/normal multilayers has received considerable attention since its original discovery.[1] Much work has gone into understanding the magnetotransport,[2–11,20] the antiferromagnetic coupling mechanism,[12–15] and the role of different materials systems.[16–20,25] The role of the interfacial structure has received less attention.[10,21–26] For the Fe/Cr system, increases in the magnitude of the GMR have been claimed with increasing interfacial roughness and increasing interdiffusion.[10,22–24] It should be stressed that at very large roughness, when antiferromagnetic coupling is lost, a decrease in the GMR is invariably expected. Moreover, if crystallographic orientation plays an important role studies of interfacial roughness may be complicated by this fact. These types of studies rely on comparisons of different samples in which the thicknesses and structural characteristics are kept constant, but the interfacial structure is varied in a reproducible fashion by varying growth conditions. Independent checks on the structural characteristics are obtained using structural probes, such as x-ray diffraction and/or electron microscopy. Since GMR varies considerably with Cr thickness, it would be desirable to perform a study of the effect of interfacial roughness in a reproducible manner in a *single* sample, when its structure is modified by ion irradiation.

Here we present a series of studies on both single-phase [110] and mixed-phase [100] and [110] Fe/Cr multilayers in which the interfacial structure is varied systematically on a single sample by using 500-keV Xe^+ ion irradiation. In this fashion, any possible systematic errors due to variation in growth conditions from sample to sample are eliminated. Ion-beam irradiation is known to induce increases in the roughness of surfaces in a variety of materials.[27–29] With increasing Xe^+ ion irradiation the GMR initially is unaffected, then increases, and at large dosages it decreases. Simultaneous structural studies show that Xe^+ ion irradiation increases the interfacial roughness for intermediate doses, and thus with increasing interfacial roughness there is an initial increase in GMR until the antiferromagnetic coupling between the layers is progressively lost, thereby decreasing the GMR. At higher irradiation doses, the GMR decreases, the films become smoother and more intermixed.

Fe/Cr multilayers were prepared using dc magnetron sputtering (base pressure of 1×10^{-7} Torr) on ambient temperature Si [111] substrates. Structural studies were performed using high- and low-angle x-ray diffraction using a Rigaku rotating anode diffractometer with Cu $K\alpha$ radiation. The in-plane magnetization was measured using a superconducting-quantum-interference-device magnetometer. Four lead magnetotransport measurements were performed at 77 K and room temperature in fields (parallel to the plane of the films and perpendicular to the current) up to 5 T. Normal incidence, 500-keV Xe^+ ion irradiation mastered over a 1.2×1.2 cm^2 area was performed in a vacuum of $\sim3\times10^{-7}$ Torr and a current level below 0.6 μA/cm^2. Sample heating was avoided during irradiation by the low, rastered current and by properly heat sinking the sample. For the film thickness used, it is expected that 500-keV Xe^+ ions will completely traverse the film. Rutherford backscattering analysis gave no evidence for Xe^+ in the films. The results presented here were observed in two independent irradiation experiments on a series of [Fe(30 Å)/Cr (12 Å)]$_{10}$ samples, where the subindex indicates the total number of bilayers. We chose this particular Cr thickness because earlier measurements showed that the maximum value of the GMR is observed at a Cr thickness of ~12 Å.[22,24]

0163-1829/94/50(5)/3481(4)/$06.00 © 1994 The American Physical Society

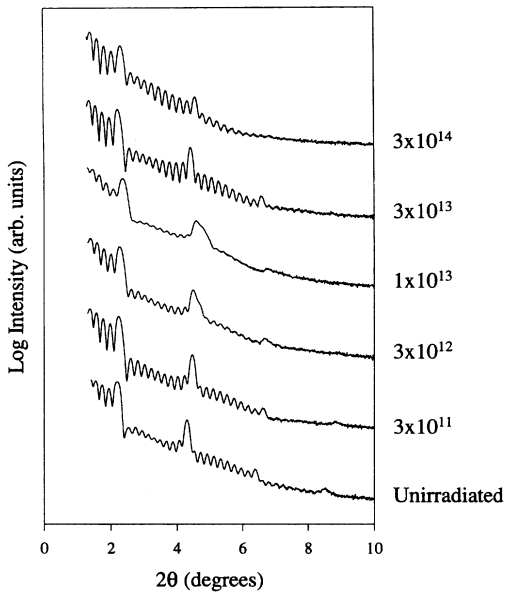

FIG. 1. Low-angle x-ray diffraction spectra for a [Fe(30 Å)/Cr(12 Å)]$_{10}$ superlattice irradiated by 500-keV Xe^+ ions. The fluence, in units of Xe^+/cm^2, is indicated on the right-hand side of the figure. The spectra have been offset for clarity.

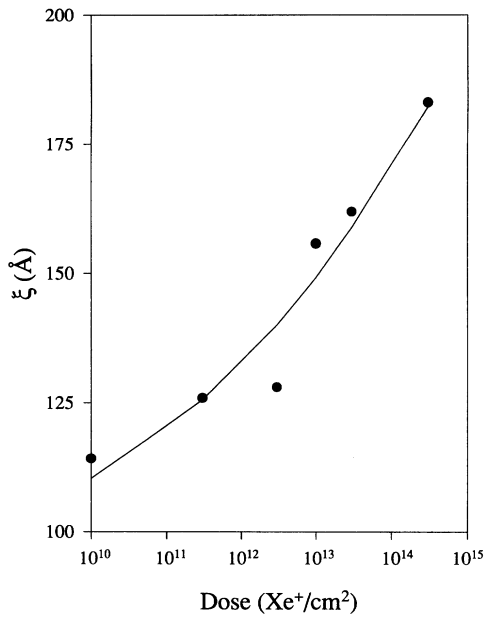

FIG. 2. Structural coherence length ξ calculated from the linewidth of the [110] x-ray peak (in the direction perpendicular to the film) using Sherrer's equation.

Since the interlayer coupling may be affected by crystallographic orientation[31,32] we have paid particular attention to thoroughly characterizing the crystallographic texture. High-sensitivity, high-angle x-ray diffraction shows that to the accuracy of our measurements (better than 1:100) our first sample consisted of a single [110] bcc texture, i.e., the growth is with the [110] bcc direction perpendicular to the substrate and random orientation in the plane of the film. Rocking curves full width at half maximum of 10° are typical of sputtered samples.

A second sample was grown to verify that the effects observed with the first sample were reproducible. This sample exhibited a ~70% majority [110] and ~30% minority [100] oriented grains. Despite that, *all our observations regarding the changes in the structure and magnetotransport of the film with* Xe^+ *irradiation were identical.* Since more extensive data was obtained on the second sample in the region of changing magnetotransport, we present here only the data from this sample which shows mixed [100] and [110] orientations. We should emphasize again that the same trends are observed in both sets of samples, and therefore the conclusions seem to be independent of crystallographic orientation. Nevertheless, crystallographic orientation dependencies cannot be ruled out and will be the subject of future investigations.

Figure 1 shows the low-angle diffraction for a [Fe(30 Å)/Cr(12 Å)]$_{10}$ sample which has been progressively irradiated with 500-keV Xe^+ ions. The unirradiated sample shows well-developed superlattice Bragg peaks around 2°, 4°, 6°, and 8° due to the superlattice periodicity, with finite-size peaks in between these Bragg peaks due to the 10 bilayers present in the multilayer. A fitting of this x-ray-diffraction scan[33] sets an upper limit on the layer thickness fluctuation of ~1 Å and interdiffusion of ~1.8 Å. A detailed comparison of the x-ray-diffraction scans with increasing irradiation shows that initially the Bragg and finite-size peaks progressively broaden and decrease in intensity until a fluence of 1×10^{13} ions/cm^3. At the larger doses, the finite-size peaks are enhanced, indicating the film becomes smoother. However, the higher-order Bragg peaks are greatly suppressed compared to the unirradiated spectra (the 4th-order Bragg peak in the 3×10^{13}, and both 3rd and 4th in the 3×10^{14}), indicating an increase in the interdiffusion at the interfaces. These trends in the roughness of the films is confirmed by the corresponding trends in the Yoneda scattering,[30] and have been observed previously.[24] It should be pointed out at this stage that the Xe^+ ion irradiation is expected to produce structural changes both at the interfaces and the bulk of the multilayers.[29] The details of the mechanism by which the interfacial disorder first increases and then decreases is not understood at the present time. A possible origin for this may be an increase in the roughness due to an increase in the interfacial strain and a progressive release of this strain with increasing radiation dosage, which gives rise to the possible breakup of the local regions which are antiferromagnetically coupled.

Figure 2 shows the structural coherence length (ξ) of the multilayers calculated from the linewidth of the [110] x-ray-diffraction peak using Sherrer's equation. In contrast to the nonmonotonic changes in the low-angle spectra above, there is a systematic, monotonic increase in ξ with increasing Xe^+ fluence. This shows that there is a monotonic enhancement of the crystallinity of the films with radiation dosage.

The magnetization versus field in the as-grown samples shows the typical behavior found in antiferromagnetically coupled Fe/Cr layers, i.e., a gradual increase in magnetization up to saturation magnetization M_s in a saturation field H_s, with very little remanent magnetization, M_r in zero

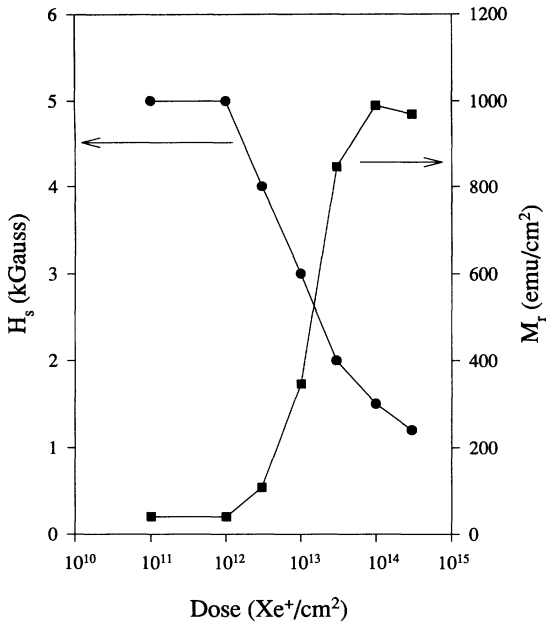

FIG. 3. Saturation field H_s and remanent magnetization M_r vs Xe^+ ion fluence. Measurements performed at 77 K, with the field parallel to the plane of the film. Lines are guides to the eye.

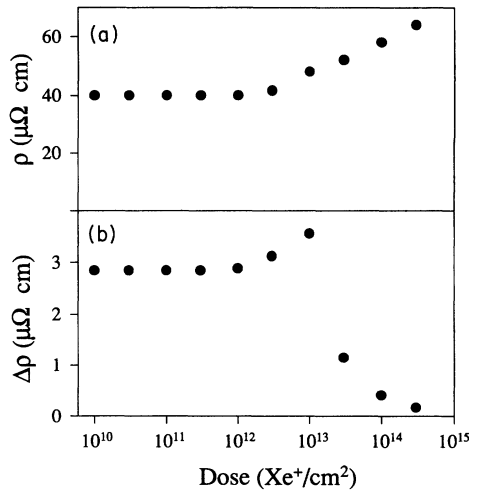

FIG. 4. (a) Saturation resistivity ρ and (b) magnetoresistance $\Delta\rho$ vs ion-beam fluence at 77 K. The field was applied in the plane of the film, perpendicular to the current.

field. M_s is approximately 1200 emu/cm^3, about 70% of the bulk Fe value. This indicates that about 30%, or 9 Å, of each Fe layer is nonmagnetic. This corresponds to ~4.5 Å of interdiffused or disordered Fe at each interface which is not magnetic, in qualitative agreement with the x-ray results and the fact that FeCr alloys are nonmagnetic for Cr concentrations larger than 25%. With increasing Xe^+ ion dosage the remanent magnetization, M_r (at zero field), increases monotonically as indicated in Fig. 3. M_r approaches the saturation magnetization at high fluences, yielding an almost 90% squareness of the magnetization loop for 3×10^{14} Xe^+ ions/cm^2. This shows that with increasing dosage the ferromagnetically coupled portion of the sample increases. H_s, which characterizes the field necessary to ferromagnetically align the coupled Fe layers, decreases monotonically with increasing Xe^+ ion dosage (see Fig. 2). These monotonic changes in H_s are also consistent and expected for a progressive increase of ferromagnetic shorts with increasing radiation.

The resistivity above saturation (ρ), in a field larger than 1 T, behaves as expected with increasing radiation dosage as shown in Fig. 4(a), an initial lack of sensitivity to irradiation followed by a monotonic increase for higher dosages. We should stress that the resistivity is sensitive both to increased interfacial as well as bulk disorder. The giant magnetoresistance ($\Delta\rho$), i.e., the maximum change in resistivity with field shown in Fig. 3(b), shows an interesting *nonmonotonic* change with irradiation. Initially, $\Delta\rho$ is insensitive to the Xe^+ irradiation, then increases and is followed by a decrease. The changes in M_r, H_s, and ρ are all consistent with increased ferromagnetic shorts which would explain the decrease observed in $\Delta\rho$ above a fluence of 1×10^{13} ions/cm^2. The increase observed up to 1×10^{13} ions/cm^2 clearly shows that increasing interfacial roughness increases $\Delta\rho$. This is in spite of the fact that the fraction of the ferromagnetically coupled sample increases with increased radiation dosage, as shown in Fig. 3 by the dependence of the M_r. It is quite remarkable that the largest $\Delta\rho$ is observed for the dosage for which the interfacial roughness is the largest as shown by a comparison between Figs. 1 and 4(b).

It is also interesting to note that although the dependence of $\Delta\rho$ is nonmonotonic that of H_s is monotonic with Xe^+ ion dosage. This suggests that the values of $\Delta\rho$ and H_s are not necessarily connected and may be controlled by different mechanisms. A similar argument can be used for ξ and $\Delta\rho$. Also, it seems clear that the changes in ρ are not due to the changes in ξ, as the opposite trend would be expected.

It should be pointed out that increasing interfacial roughness not only increases the electronic scattering. In a well-textured [110] Fe/Cr sample as the interfacial roughness increases, the fraction of non-[110] planes at the interface also increases. If the GMR is affected by the crystallographic orientation at the interface, this may also explain the increase of GMR with increasing interfacial roughness. It should be noted that increases in the GMR have also been observed in samples where the Fe and Cr are alloyed at the interfaces.[10] It is possible that the interdiffusion is increasing while the roughness is increasing, and the increase in the GMR is due to both effects.

Several theoretical models have shown that an increase in $\Delta\rho$ is expected with an increase of interfacial roughness.[5–8,21] Naively, this increase in $\Delta\rho$ originates in the increase of sampling of the antiferromagnetically coupled layers with increasing roughness.

There are still disagreements over the location of the spin-dependent scattering in Co/Cu and NiFe/Cu multilayers.[25,26,34,35] For the parallel resistor model,[11] it has been demonstrated that increases or decreases in the GMR with increasing roughness are expected, increases if the spin-dependent scattering is occurring at the interfaces, decreases if the spin-dependent scattering is occurring in the bulk of the magnetic material.[34] However, other experiments seem contradictory.[25] The present data are consistent with the spin-

dependent scattering occurring at the Fe-Cr interface.

In summary, we have studied the effect of 500-keV Xe^+ ion irradiation on the structure, magnetization, and magnetotransport of Fe/Cr multilayers. The structure, magnetization, and saturation resistivities are in agreement with the idea that Xe^+ ion damage changes the interfacial roughness. This is manifested by a monotonic increase on the resistivity and remanent magnetization and a monotonic decrease in the saturation field. This is consistent with a progressive increase in the ferromagnetically coupled portion of the sample with increased radiation dose. The giant magnetoresistance, on the other hand, has a nonmonotonic dependence with ion damage. This nonmonotonic dependence is correlated with a nonmonotonic dependence of the small-angle x-ray diffraction. This indicates that interfacial scattering increases the giant magnetoresistance in spite of the decreasing portion of the sample, which is antiferromagnetically coupled. These conclusions are in agreement with earlier studies in which the interfacial roughness was varied by the growth conditions.[21–23] However, the possibility that GMR is predominantly affected by the crystallographic orientation at the interface cannot be ruled out by the present experiments.

We thank P. Levy, K. Hathaway, G. Prinz, and A. Fert for useful conversations. This work was supported by the US-DOE and NSF at UCSD and the Swedish-Natural Research Council.

[1] M. N. Baibich, J. M. Broto, A. Fert, F. Nguyen Van Dau, F. Petroff, P. Etienne, B. Creuzet, A. Frederich, and J. Chazelas, Phys. Rev. Lett. **61**, 2472 (1988).

[2] A. Barthelemy, A. Fert, M. N. Baibich, S. Hadjoudj, F. Petroff, P. Etienne, R. Cabanel, S. Lequien, F. Nguyen Van Dau, and B. Creuzet, J. Appl. Phys. **67**, 5908 (1990).

[3] G. Binasch, P. Grünberg, F. Saurenbach, and W. Zinn, Phys. Rev. B **39**, 4828 (1989).

[4] P. M. Levy, S. Zhang, and A. Fert, Phys. Rev. Lett. **65**, 1643 (1990).

[5] R. E. Camley and J. Barnas, Phys. Rev. Lett. **63**, 664 (1989).

[6] N. Garcia and A. Hernando, J. Magn. Magn. Mater. **99**, L12 (1991).

[7] J. Inoue, A. Oguri, and S. Maekawa, J. Phys. Soc. Jpn. **60**, 376 (1991).

[8] P. Baumgart, B. A. Gurney, D. R. Wilhoit, T. Nguyen, B. Dieny, and V. S. Speriosu, J. Appl. Phys. **69**, 4792 (1991).

[9] H. Hasegawa, Phys. Rev. B **47**, 15 073 (1993).

[10] L. H. Chen, T. H. Tiefel, S. Jin, R. B. Van Dover, E. M. Gyorgy, and R. M. Fleming, Appl. Phys. Lett. **63**, 1279 (1993).

[11] D. M. Edwards, J. Mathon, and R. B. Muniz, IEEE Trans. Magn. **MAG-27**, 3548 (1991).

[12] P. Grünberg, R. Schreiber, Y. Pang, M. B. Brodsky, and H. Sowers, Phys. Rev. Lett. **57**, 2422 (1986).

[13] J. C. Slonczewski, Phys. Rev. Lett. **67**, 3172 (1991).

[14] Y. Wang, P. M. Levy, and J. L. Fry, Phys. Rev. Lett. **65**, 2732 (1990).

[15] S. Mirbt, H. L. Skriver, M. Alden, and B. Johansson, Solid State Commun. **88**, 331 (1993).

[16] J. Q. Xiao, J. S. Jiang, and C. L. Chien, Phys. Rev. Lett. **68**, 3749 (1992).

[17] A. E. Berkowitz, J. R. Mitchell, M. J. Carey, A. P. Young, S. Zhang, F. E. Spada, F. T. Parker, A. Hutten, and G. Thomas, Phys. Rev. Lett. **68**, 3745 (1992).

[18] S. S. Parkin, Phys. Rev. Lett. **67**, 3598 (1991).

[19] E. E. Fullerton, J. E. Mattson, S. R. Lee, C. H. Sowers, Y. Y. Huang, G. Felcher, S. D. Bader, and F. T. Parker, J. Magn. Magn. Mater. **117**, L301 (1992).

[20] V. S. Speriosu, B. Dieny, P. Humbert, B. A. Gurney, and H. Lefakis, Phys. Rev. B **44**, 5358 (1991).

[21] H. Nakanishi, A. Okiji, and H. Kasai, J. Magn. Magn. Mater. **126**, 451 (1993).

[22] E. E. Fullerton, D. M. Kelly, J. Guimpel, I. K. Schuller, and Y. Bruynseraede, Phys. Rev. Lett. **68**, 869 (1992).

[23] F. Petroff, A. Barthelemy, A. Hamzic, A. Fert, P. Etienne, S. Sequien, and G. Creuzet, J. Magn. Magn. Mater. **93**, 95 (1991).

[24] D. M. Kelly, E. E. Fullerton, F. T. Parker, J. Guimpel, Y. Bruynseraede, and I. K. Schuller, Int. J. Mod. Phys. B **7**, 419 (1993).

[25] S. S. P. Parkin, Appl. Phys. Lett. **61**, 1358 (1992).

[26] B. A. Gurney, D. R. Wilhoit, V. S. Speriosu, and I. L. Sanders, IEEE Trans. Magn. **26**, 2747 (1990).

[27] E. A. Eklund, R. Bruinsma, J. Rudnick, and R. S. Williams, Phys. Rev. Lett. **67**, 1759 (1991).

[28] J. Krim, I. Heyvaert, C. Van Haesendonck, and Y. Bruynseraede, Phys. Rev. Lett. **70**, 57 (1993).

[29] G. Gladyszewski, Ph. Goudeau, A. Naudon, C. Jaouen, and J. Pacaud, Appl. Surf. Sci. **66**, 28 (1992).

[30] S. K. Sinha, Physica B **173**, 25 (1991).

[31] W. F. Egelhoff and M. T. Kief, IEEE Trans. Magn. **28**, 2742 (1992).

[32] S. S. P. Parkin, R. F. Marks, F. F. C. Farrow, G. R. Harp, Q. H. Lam, and R. J. Savoy, Phys. Rev. B **46**, 9262 (1992).

[33] E. E. Fullerton, I. K. Schuller, H. Vanderstraeten, and Y. Bruynseraede, Phys. Rev. B **45**, 9292 (1992).

[34] M. J. Hall, B. J. Hickey, M. A. Howson, C. Hammond, M. J. Walker, D. G. Wright, D. Grieg, and N. Wiser, J. Phys. Condens. Matter **4**, L495 (1992).

[35] V. S. Speriosu, J. P. Nozieres, B. A. Gurney, B. Dieny, T. C. Huang, and H. Lefakis, Phys. Rev. B **47**, 11 579 (1993).