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Extraordinary Hall effect in giant magnetoresistive Fe/Cr multilayers: The role of interface scattering

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We have investigated the Hall effect in giant-magnetoresistive Fe/Cr multilayers as a function of systematic changes in interfacial roughness. Two approaches have been used to modify the interfacial roughness: sputter deposition under different argon pressures, and isothermal annealing at fixed temperatures. We find that interfacial roughness enhances the magnetic (extraordinary) contribution to the Hall effect. Furthermore, the presence of roughness at the interfaces modifies the relationship between the Hall resistivity and the longitudinal resistivity. These results indicate that interface scattering cannot be neglected and should be included in theories of the extraordinary Hall effect in magnetic multilayers. [S0163-1829(96)51218-X]

Ability to tailor antiferromagnetic exchange coupling[1] among the layers in ferromagnetic/nonmagnetic multilayer structures and the discovery of giant magnetoresistance (GMR) in Fe-Cr multilayers[2] have stimulated considerable interest and active studies investigating the magnetism and magnetotransport phenomenon in such materials. GMR has been extensively investigated experimentally and is currently ascribed to arise from spin-dependent scattering of conduction electrons at the magnetic/nonmagnetic interfaces and/or inside the magnetic layers.[3] The relative importance of these two contributions to the GMR for different systems is yet to be established. However, there is growing evidence that, at least for some GMR systems, interface scattering is predominant[4] and it may even enhance the GMR.[5,6] In order to critically investigate the role of interface scattering and also the consequence of the physical nature of the interface boundary, such as roughness, simultaneous determination of the Hall and electrical resistivities is useful. Especially in a magnetic material, the anomalous Hall effect is an additional physical property which reflects both the magnetic nature as well as the transport characteristics of the material. Theoretical guidance in this respect is not clear as to what extent the Hall effect is different in a multilayer with individual layers of only a few tens of angströms thick when the interface constitutes an appreciable fraction of the sample. In this paper we show that in Fe/Cr multilayers increasing interfacial roughness enhances the magnetic contribution to the Hall effect (the extraordinary Hall effect, EHE). This result indicates that interfacial scattering should be an ingredient in theories of the EHE in magnetic multilayers.

In bulk ferromagnetic materials the Hall effect is commonly described by the phenomenological equation[7]

$$\rho_H = R_o B + R_s 4\pi M, \tag{1}$$

where ρ_H is the Hall resistivity, B is the external magnetic field, R_o is the ordinary Hall coefficient, and M is the magnetization. R_o has the usual meaning and is related to the number of conduction carriers per atom. R_s, the extraordinary Hall coefficient is characteristic of magnetic materials and is typically much larger and has a stronger temperature dependence than R_o. It has been well established, both experimentally and theoretically, that there is a direct correlation between the extraordinary Hall coefficient and longitudinal resistivity in the form

$$R_s \propto \rho^n, \tag{2}$$

where n depends on the predominant scattering mechanisms involved: $n=1$ for skew scattering, and $n=2$ for side jump.[7,8] The skew scattering term, believed to arise from the spin-orbit coupling between the magnetic moment and the conduction electron, is expected to dominate in pure materials at low temperatures whereas the side jump mechanism is predominant at higher temperatures and in materials with high resistivities. In the resistivity range studied here, the side jump mechanism with $n=2$ is known to dominate the EHE in homogeneous Fe and dilute Fe-Cr alloys (see Refs. 8 and 9 and references therein).

Recently, there have been a few reports on the Hall effect measurements in magnetic multilayers. $n=2$ was reported for molecular beam epitaxy grown Co/Cu superlattices,[10] $n=2.6$ was found for electron beam evaporated Fe/Cr multilayers,[11] and n as high as 3.7 was reported for heterogeneous giant magnetoresistive films of Co-Ag.[12] Clearly, the EHE in spatially inhomogeneous magnetic systems is affected by parameters, other than those present in the bulk. The only theoretical treatment of the EHE in magnetic multilayers, recently put forth by Zhang,[13] uses the Kubo formalism and shows that, in general, the commonly used scaling relation [Eq. (2)] between the EHE and longitudinal resistivity is not valid. In this work, Zhang considered only the side jump mechanism, since the overall resistivities are much higher than those of the constituents. He also neglected intersurface scattering. In the local limit, where the mean free path is less than the layer thicknesses, the scaling law mentioned above [Eq. (2)] with $n=2$ is recovered. In contrast, when the mean free path is comparable or greater than the superlattice modulation length, the simple relationship expressed by Eq. (2) no longer holds, the Hall resistivity depends on the ratio of relaxation times (mean free paths) in magnetic layers and nonmagnetic layers, and as a result the power n in the scaling law may be smaller or greater than 2. In multilayers the mean free path is usually larger than layer thicknesses, hence nonlocal effects must be important. But even in the absence of such effects, the fact that the layers and *interfaces* conduct in parallel would by itself result in n not necessarily equal to 2: as the temperature increases the resistance of the layers increases (because of the phonon term), while the resistance of the rough interface remains constant. As a result more current goes through the interface, the interface contribution to the Hall voltage increases which would probably cause a change of n. In this paper we demonstrate that interface scattering *modifies* the relationship between the extraordinary Hall coefficient and longitudinal resistivity in multilayers.

Fe/Cr multilayers were prepared using dc magnetron sputtering (base pressure of 1×10^{-7} Torr) on ambient temperature Si [111] substrates. The interface roughness was varied by changing the sputtering gas (Ar) pressure. The structure of the samples was characterized by x-ray diffraction using a Rigaku rotating anode diffractometer with Cu-Ka radiation. The magnetization loops were measured using a superconducting quantum interference device (SQUID) magnetometer. For the transport measurements the films were patterned using chemical etching into bridge shaped structures with current channels and voltage terminals of about 0.5 mm in width. To eliminate possible spurious signals ac current was used and the Hall voltage was averaged for two field directions. The resistive and Hall voltages were phase sensitively detected using lock-in amplifiers. Some as-prepared samples were annealed in a N_2 atmosphere at temperatures up to 335 °C for 30 minutes with a heating and/or cooling rate of about 40 deg/min, and the changes in the magnetotransport caused by the structural modification were studied. The results presented here are for [Fe(30 Å)/Cr(12 Å)]$_{10}$ samples, where the subindex indicates the number of bilayers. The thickness of Cr was chosen to correspond to the first antiferromagnetic peak in the interlayer exchange coupling, and thus give the maximum GMR value for this system.[5]

Figure 1(a) shows low-angle θ-2θ x-ray diffraction spectra of [Fe(30 Å)/Cr(12 Å)]$_{10}$ superlattices. The same conditions were used for the deposition of the samples except for the pressure of the Ar sputtering gas, which was fixed at 4, 7, and 10 mTorr for the five samples in this study (three samples were from batch deposited at 10 mTorr). The 4-mTorr sample exhibits clear superlattice Bragg peaks up to the third order and clean-cut finite-size peaks between the Bragg peaks. The finite-size peaks are due to interference of x-ray reflections from the top and bottom surfaces of the film. Their periodicity is determined by the total thickness of the film with respect to the modulation length (in this case 10 bilayers). Both the Bragg and the finite-size peaks are considerably reduced in intensity and visibly broadened for the 7-mTorr and 10-mTorr samples. The broadening of the superlattice Bragg peaks and the loss of the finite-size peaks is characteristic of increased interface roughness, i.e., cumulative random variations in layer thicknesses (see Ref. 5 and references therein). This means that the 4-mTorr sample has layers that are significantly flatter than the layers in the 7-mTorr and 10-mTorr samples. It is also obvious from Fig. 1(a) that the 7-mTorr sample has much flatter layers than the 10-mTorr sample. Thus, the interfacial roughness increases with increasing Ar pressure.

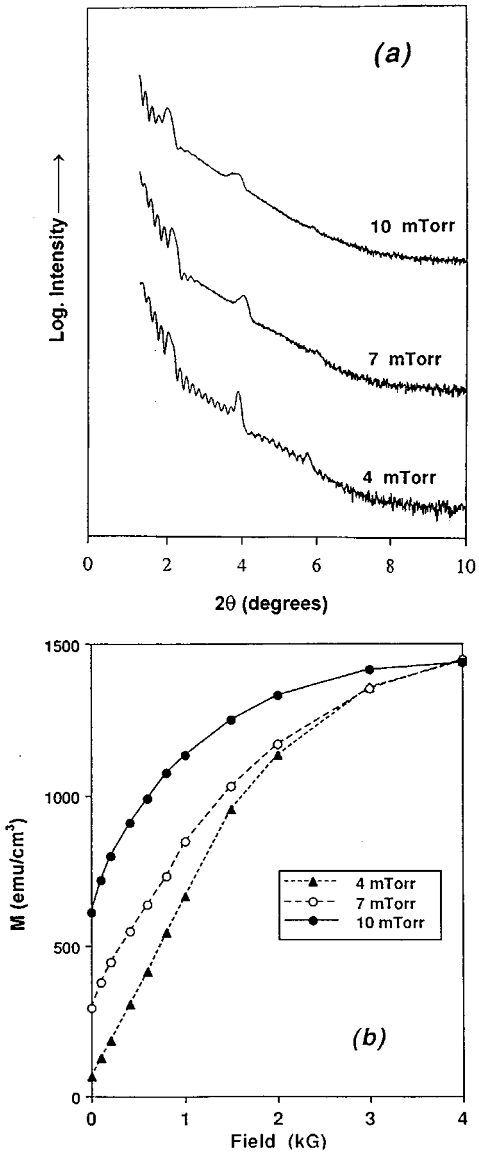

FIG. 1. (a) Low-angle x-ray diffraction spectra and (b) in-plane magnetization curves for [Fe(30 Å)/Cr(12 Å)]$_{10}$ superlattices sputtered at 4, 7, and 10 mTorr Ar pressure. The x-ray spectra are offset for clarity.

Three magnetization curves for the 4-mTorr, 7-mTorr, and 10-mTorr samples are shown in Fig. 1(b). The M versus H curve for the 4-mTorr sample looks typical of an antiferromagnetically coupled multilayer. A field close to 4 kG is required to overcome the antiferromagnetic coupling and saturate the magnetization of the sample. As the magnetic field is decreased the magnetization returns to zero as is typical for antiferromagnetically coupled magnetic layers. The remanence sharply increases for the 7-mTorr and 10-mTorr samples due to variations in the Cr layer thickness resulting in regions of the sample that are coupled ferromagnetically. On the other hand, the saturation magnetization is almost constant for the three samples (1500 emu/cc). This implies that the interdiffusion into the bulk is not changing appreciably with increased roughness.

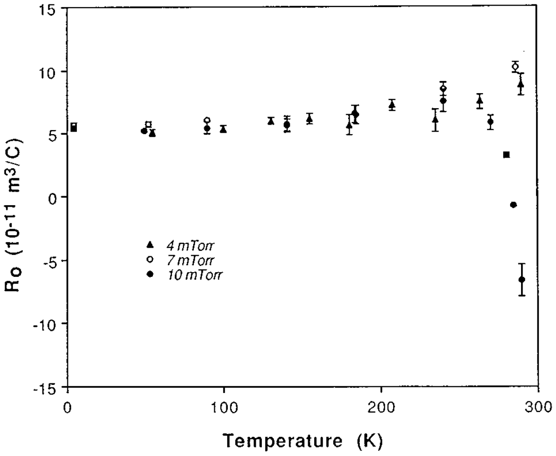

FIG. 2. Ordinary Hall coefficient R_o as a function of temperature for the same samples shown in Fig. 1. The error in determination of R_o for each data point is indicated.

The temperature dependence of the ordinary Hall coefficient is shown in Fig. 2. Remarkably, R_o is virtually the same for the three samples and exhibits a similar temperature dependence in almost the whole temperature range, from 5 to about 250 K. Below about 250 K, R_o is practically independent of temperature exhibiting only a slight increase at 200–250 K. In contrast, above 250 K, R_o *continues to increase* somewhat for the 4-mTorr and 7-mTorr samples, and *changes sign* for the 10-mTorr sample. The similar magnitude and temperature dependence of the ordinary Hall coefficient (below 250 K implies similar electronic structure for the three samples. The value 6×10^{-11} m^3/C is characteristic of thin Fe and Cr films, where R_o may vary between about 2 and 12×10^{-11} m^3/C depending on the film thickness and preparation conditions (see Ref. 7 and references therein). For pure Fe, Cr, and Fe-Cr alloys R_o is positive.[7,9] We note, that the low sensitivity of R_o to increasing roughness also implies negligible changes in the interdiffusion (alloying) at the interfaces, since alloying is expected to significantly modify the electronic structure in Fe-Cr.[9] The change in the sign of R_o in our ''roughest'' sample around room temperature is not understood at present. We only note, that a different sign of R_o at 5 and 300 K has been indicated in Fe/Cr multilayers.[11] We limit our discussion of the temperature variation of the EHE to low temperatures (250 K) where the ordinary Hall coefficient is the same for the three samples.

Because of the scatter in the resistivity data from sample to sample (typically 10%) due to geometrical uncertainties and other uncontrollable experimental parameters in determination of the absolute value of ρ, a direct comparison of the Hall signals for samples with different roughnesses is not straightforward. In contrast, R_s and ρ can be measured quite accurately ($<0.1\%$) on the same sample as a function of temperature. Figure 3 shows the extraordinary Hall coefficient as a function of the longitudinal resistivity for the samples having different degrees of roughness as extracted from the experimental R_s and ρ at a given temperature. The lines through the data points are linear fits. The experimental data can be approximated well by a linear function with the slope corresponding to the power n in Eq. (2). For our ''smoothest'' sample (4 mTorr) we obtain $n\approx2.0$, while for the ''roughest'' sample (10 mTorr) $n\approx2.3$. Different triangles in Fig. 3 denote different samples from the same 10-mTorr batch. The scatter in the n value between different samples within the same batch is less than 0.04, which appears to be the main source of uncertainty in n (the error in the linear regression of Fig. 3 corresponds to 0.01 uncertainty in n). To better visualize the changes in the R_s versus ρ dependence, we plot the data for the 4-mTorr and 10-mTorr samples offset by a constant in the inset to Fig. 3. Clearly, $R_s\propto\rho^2$ relationship is not unique. We attribute this result to the presence of additional scattering by the interface roughness, and note that it is not caused by the increased overall resistivity of the superlattice. In Fig. 3 we show R_s versus ρ data for the 7-mTorr sample having a higher overall resistivity than the 10-mTorr sample; the slope is $n\approx2.1$. The systematic increase in n for the three samples correlates with increasing roughness in the superlattice. We also note, that interdiffusion is expected to produce the opposite effect: a decrease in n from about 2.15 to 1.85 was observed on Cr alloying into Fe.[9]

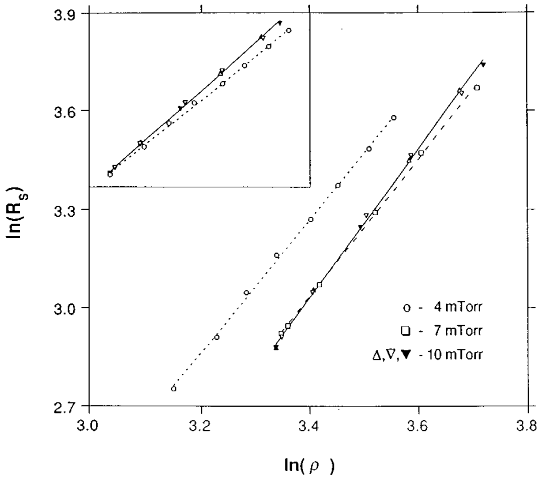

FIG. 3. Log-log plot of the extraordinary Hall coefficient R_s as a function of longitudinal resistivity ρ for the same samples shown in Fig. 1. The inset shows the data for the 4-mTorr and 10-mTorr samples offset by a constant.

The above discussion relies on the validity of the scaling law [Eq. (2)] between the EHE and ordinary resistivity, which was originally derived for bulk ferromagnetic materials. As an independent check, it would be desirable to directly study the changes in the Hall effect in the same sample as the interfacial roughness is modified. This can be done in several ways. We have previously observed high sensitivity of the interface microstructure and the GMR in Fe/Cr, to ion irradiation[6] and heat treatment.[14] Both ion irradiation and annealing cause an increase in the interfacial roughness accompanied by nonmonotonic changes in the GMR.[6,14] The GMR as a function of either ion dose or annealing temperature first increases due to increased roughness and then decreases due to eventual loss of the antiferromagnetic coupling in the multilayer. However, in contrast to low-temperature heat treatment resulting mainly in modification of the interface between the layers, ion irradiation also results in structural disorder which increases the overall electronic scattering rate and hence the resistivity. This is why

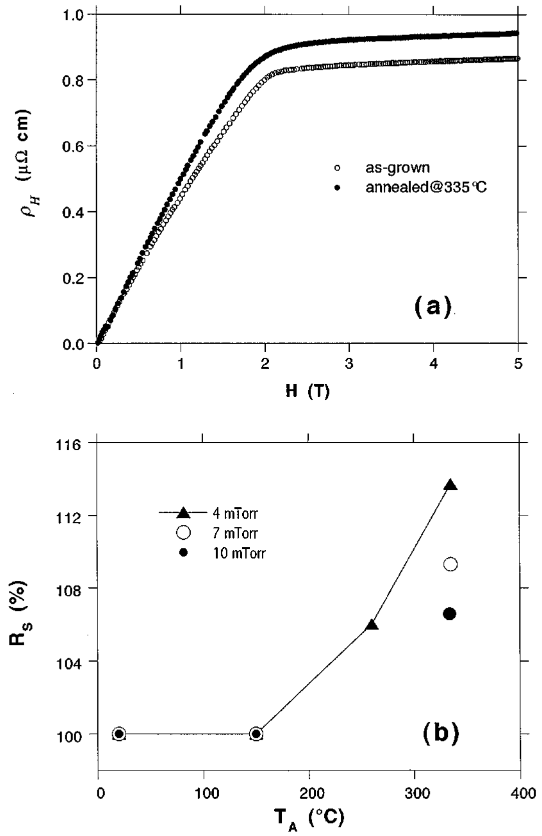

FIG. 4. Room temperature (a) Hall resistivity ρ_H as a function of field H for the 7-mTorr sample as-grown and annealed at 335 °C and (b) extraordinary Hall coefficient R_s as a function of annealing temperature T_A for the same samples shown in Fig. 1.

we have chosen here low-temperature annealing to study the influence of the modified structure on the Hall effect. In Fig. 4(a) we plot the Hall resistivity as a function of field for the 7-mTorr sample as-grown and annealed at 335 °C. The increase in the EHE is obvious. Figure 4(b) shows the extraordinary Hall coefficient as a function of annealing temperature for the 4-mTorr, 7-mTorr, and 10-mTorr samples. The enhancement in R_s is less pronounced for samples having initially rougher layers, which is consistent with the idea that this enhancement is due to scattering by interfacial roughness. We observe practically no change in the ordinary Hall coefficient with annealing. It is important to note, that the overall resistivity does not increase, i.e., no additional disorder or mixing, with annealing at low temperatures, and hence cannot be the origin of the enhancement in EHE. In fact, the resistivity of the 4-mTorr sample is about 5% lower after annealing at 335 °C. This behavior of ρ with annealing is consistent with our previous results,[14] and is most probably caused by general improvement of the structure (bulk defect annihilation, release of atomic strain, etc.). We used annealing at low temperatures, where the multilayer character of the samples is preserved, the total resistivity is practically unaffected, while the interfacial microstructure is significantly modified. The observed low sensitivity of the ordinary Hall coefficient and saturation magnetization to low temperature annealing indicates small changes in the interdiffusion. Annealing at higher temperatures ($>$350 °C) causes significant interdiffusion resulting in the loss of the multilayer character and a considerable increase in the overall resistivity, which complicates the study of the role the interface plays in the EHE.

In view of the recent theory for the EHE in multilayers[13] the experimentally observed $R_s \propto \rho^2$ scaling in samples with good interface quality (see also Ref. 10) is somewhat surprising, since it is expected to be valid only when the ratio of the mean free paths in magnetic and nonmagnetic layers is constant as a function of temperature.[13] More work, both experimental and theoretical, is required to determine the important parameters affecting the EHE in ultrathin magnetic multilayers. What is clear, however, is that the scaling law $R_s \propto \rho^2$ is not unique and is structure dependent. In particular, we find that the presence of roughness enhances the EHE in magnetic multilayers and modifies the relationship between the Hall resistivity and ordinary resistivity. We suggest that this effect may be one of the reasons for the large values of the exponent n recently reported in the literature.[10,11] Theoretical work to clarify the role of the interface scattering in the EHE in spatially inhomogeneous magnetic systems is encouraged.

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