Scaling Behavior of Giant Magnetotransport Effects in Co/Cu Superlattices

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We have observed two forms of scaling in magnetotransport properties of (111) Co/Cu superlattices: (1) the extraordinary Hall coefficient R_s correlates with the resistivity $\rho_{xx}(0)$ in the form $R_s \sim \rho_{xx}^2(0)$, and (2) the magnetothermopower $S(H)$ correlates with the magnetoresistivity $\rho_{xx}(H)$ in the form $S(H)/T \sim \rho_{xx}(0)/\rho_{xx}(H)$. The first effect relates to quantum mechanical side jump while the other relates to interfacial spin-dependent density of states (DOS). These results reveal the different roles of bulklike spin-dependent scattering in Co layers and spin-dependent DOS of interfacial layers in causing the observed large positive extraordinary Hall effect and giant longitudinal magnetotransport.

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The discovery of high field giant magnetoresistance (GMR) effects in Co-based superlattices grown by molecular beam epitaxy (MBE) techniques [1-3] suggests that the observed effects do not depend on the dominant spins in these materials since they occur at fields well in excess of the saturation of the Co magnetization. The result questions the generality of the conventional spin-dependent scattering mechanisms based on antiferromagnetic coupling of the neighboring magnetic layers [4,5]. Recent publications further demonstrate the important role of interfacial magnetic states in determining the overall spin-dependent scattering in multilayers [6,7]. Polarized states of noble metal interlayers have also been observed [8,9]. Taken together, these findings call for a careful examination of all of the magnetotransport properties so that the effects of density of states, of spin-orbit coupling, and of spin-dependent scattering potentials can be assessed. In this paper we report scaling behavior in magnetotransport properties of (111) Co/Cu superlattices through a comprehensive study of the Hall effect and the magnetothermopower in addition to the magnetoresistivity. This has made it possible to identify the specific spin-dependent processes in these materials from existing model predictions.

The superlattices were prepared using MBE techniques starting from 500 Å of (110) Ge buffer, then bcc (110) Co and (111) Au, grown on (110) GaAs substrates. Samples studied here each contain typically 30 bilayers of 15 Å Co layer and Cu spacer layer between 6 and 20 Å thick. The growth methods and the high quality of the resulting crystals, particularly the smooth and abrupt nature of the interfaces, are documented elsewhere [2]. In particular, the crystal coherence length of the superlattice, measured by reflection high energy electron diffraction, cross-sectional high-resolution transmission electron microscopy, x-ray diffraction, and spin-echo NMR experiments, is > 200 Å [2]. Samples were further processed for transport measurements using conventional lithography techniques including inert Ar ion etching. Bridge shaped patterns with 0.5 mm wide current channels and 0.2 mm wide voltage terminals were made for standard dc four-terminal measurements. Care was taken to avoid contamination during post-growth procedures. Thermopower measurements were performed using calibrated fine Cu wires as electrodes, and constantan-chromel differential thermocouples. The temperature difference was maintained at about 1 K by a resistive heater attached to one end of the ~10 mm long and ~3 mm wide GaAs substrate, with the other end thermally anchored to the Cu block of the cryostat using Stycast adhesive. Experimental details will be provided in a more complete publication.

We measured the resistivity ρ_{xx}, the Hall resistivity ρ_{xy}, and the thermopower S with field applied along the [111] growth direction [10]. Large magnetotransport effects were observed, and their qualitative behaviors do not vary from sample to sample. Typical field dependences at 161 K are shown in Fig. 1 for a $(\mathrm{Co}_{15\,\text{Å}}/\mathrm{Cu}_{8\,\text{Å}})_{30}$ superlattice and they are compared with the measured magnetization [Fig. 1(b)].

The Hall resistivity ρ_{xy} shown in Fig. 1(a) strictly follows the phenomenological form [11]

$$\rho_{xy} = R_0 H + R_s 4\pi M_{\mathrm{Co}}, \tag{1}$$

where the first term is the ordinary Hall effect and the second is the extraordinary Hall effect which is proportional to the Co magnetization M_{Co}. In contrast, the longitudinal transport properties shown in Figs. 1(c) and 1(d) exhibit a very different field dependence including a markedly gradual saturation at high fields. Figure 1 establishes unequivocally that, while the Hall effect depends on the Co magnetization, the MR ratio $[\Delta\rho_{xx}/\rho_{xx}(0)]$, and the magnetothermopower $[\Delta S/S(0)]$ do not [12]. This leads to the conclusion that the Co magnetization cannot be the only field dependent state variable in this superlattice system. We note in particular that the magnetization of the superlattices is ferromagnetic in character so that the saturation behavior merely reflects the domain rotation in the bulk of the sample due to magnetic anisotropy. Therefore, the scattering processes that give rise to the observed extraordinary Hall effect [Fig. 1(a)] must occur within each Co layer and the observed giant longitudinal magnetotransport effects [Figs. 1(c) and 1(d)] must arise from the scattering at the interfaces

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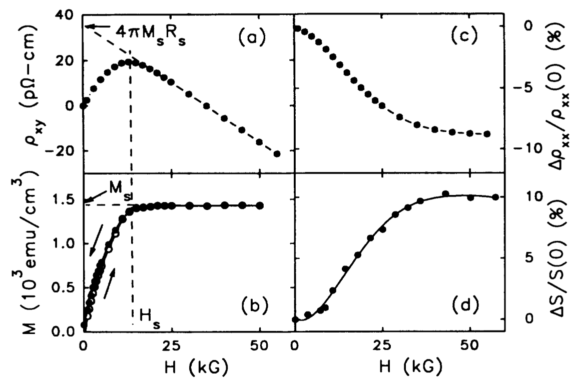

FIG. 1. Field dependence of (a) Hall resistivity, (b) magnetization, (c) magnetoresistivity, and (d) magnetothermopower for $(Co_{15\,Å}/Cu_{8\,Å})_{30}$ at 161 K. Field was along the [111] growth direction. The vertical dashed line indicates the perpendicular saturation field of the Co layers. The linear portion of the Hall resistivity at high fields corresponds to the ordinary Hall effect, and the low field magnetization-dependent behavior is the extraordinary Hall effect (a). The extraordinary Hall coefficient R_s is obtained by extrapolating the high field linear behavior to zero field, as indicated by the dashed line in (a). Magnetization was measured separately using a commercial SQUID magnetometer.

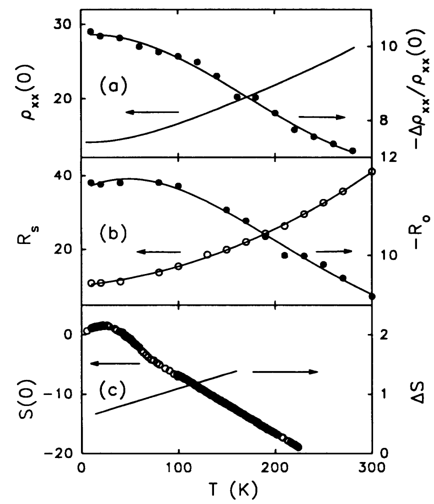

FIG. 2. Temperature dependences of (a) zero field resistivity in units of $\mu\Omega$ cm and magnetoresistivity at 5.7 T transverse field in %, (b) ordinary and extraordinary Hall coefficient in units of 10^{-11} m^3/C, and (c) zero field thermopower and magnetothermopower at 5.7 T transverse field in units of μV/K for $(Co_{15\,Å}/Cu_{8\,Å})_{30}$.

where the spin states are not those of the ferromagnetic Co. Any mechanism that describes the spin-dependent scattering processes in this system must include the spin states of the interfaces.

The presence of strong interfacial scattering is made more evident by the temperature dependence of the resistivity $\rho_{xx}(0)$ [Fig. 2(a)]. $\rho_{xx}(0)$ of the superlattice is more than twice the value of either Co or Cu at high temperatures where phonon scattering usually dominates the electron relaxation. In the presence of relatively long structural coherence length (>200 Å) [2] only interfacial scattering can put such a low limit on the electron mean free path, because diffusive scattering [13] from the interfacial magnetic states reduces the overall mean free path of the superlattice from those of the pure materials. This is completely consistent with the observed low residual resistance ratio (~ 2), as shown in Fig. 2(a).

The temperature dependences of the Hall coefficients and thermopower are also shown in Fig. 2. Unlike the resistivity, they are more sensitive to the density of states of the superlattice. Since pure Co possesses large density of states at the Fermi level relative to those of the Cu [11] and of the interface [9], one would expect that these bulk-sensitive effects are Co-like. This is confirmed by the measurements which show that both the ordinary Hall coefficient R_0 [Fig. 2(b)] and the thermopower of the superlattice [Fig. 2(c)] are very close to the values of bulk Co.

The connection between the bulklike scattering, which depends on Co magnetization M_{Co}, and the interfacial scattering is demonstrated by the strong correlation exhibited by the extraordinary Hall coefficient R_s [Fig. 2(b)] and resistivity $\rho_{xx}(0)$ [Fig. 2(a)]. Experimental and theoretical work establishes that this type of correlation depends on the specific electron scattering processes through spin-orbit interactions [14]. When the electron relaxation is weak, plane wave states are present, and the classical asymmetrical scattering is dominant, hence a linear correlation. On the other hand, when relaxation is strong, the quantum mechanical side jump takes place for the individual conduction electron wave packets and, in this case a quadratic scaling is predicted, $R_s \sim \Delta y \rho_{xx}^2(0)$ [14]. Δy here is the transverse displacement of the otherwise linearly propagating wave packet. In the case of Co/Cu superlattices studied here, with the short mean free path resulting from strong interfacial scattering, plane wave states are suppressed and side jump must dominate the bulk scattering even at low temperatures. This is observed experimentally (Fig. 3). All samples studied here, with the combined R_s values varying over two decades, exhibit the same power law behavior for temperatures >2 K. This result confirms that the electron scattering in the bulk of the superlattice is dominated by the quantum mechanical side jump. We note in passing that the observation of this universal exponent is quite rare in superlattices, and generally much larger or smaller exponents have been reported [1,15,16]. Such an agreement with existing theory is consistent with the high structural quality and uniformity of the superlattices studied here.

Can the side jump cause the observed GMR since its low field MR is $\sim -\Delta y H M_{Co}$ [14]? The answer is no, simply because the Δy inferred from our measurements is

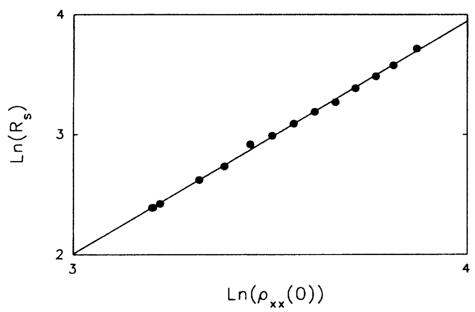

FIG. 3. Scaling behavior between extraordinary Hall coefficient and resistivity for $(Co_{15\,Å}/Cu_{8\,Å})_{30}$. The slope of the log-log plot is 2 indicating the relation $R_s \sim \rho_{xx}^2(0)$.

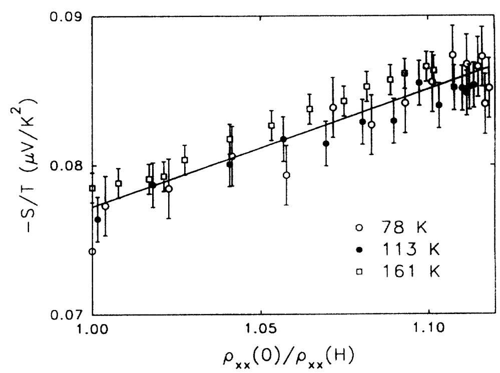

FIG. 4. Scaling behavior of magnetothermopower and magnetoresistance at various temperatures and fields for $(Co_{15\,Å}/Cu_{10\,Å})_{30}$.

at least 10 times too small.

While the MR shows no apparent dependence on the Co magnetization it does exhibit scaling with the thermopower at temperatures where diffusion thermopower is dominant ($T > 50$ K) [see Figs. 1(c) and 1(d), and Figs. 2(a) and 2(c)]. The scaling is of the form [17]

$$S(H)/T \sim \rho_{xx}(0)/\rho_{xx}(H)\,, \tag{2}$$

as illustrated in Fig. 4 for $(Co_{15\,Å}/Cu_{10\,Å})_{30}$ at three different temperatures. Scattering of conduction electrons due to the spin-dependent density of states of the unfilled d bands at the Fermi level leads to precisely this form of scaling [17]. Note that this mechanism does not require the presence of strong spin-orbit interaction or spin-wave excitation [17,18].

At a paramagnetic-ferromagnetic interface, MR due to differences in the spin-dependent density of states, by symmetry, should depend on the superposition of the quadratic terms of the two magnetizations, namely, the interfacial magnetization $M_i(H)$ and $M_{Co}(H)$. To the lowest order, the three possible terms are $[M_i(H)]^2$, $[M_{Co}(H)]^2$, and $[M_i(H)M_{Co}(H)]$. The lack of dependence on $[M_{Co}(H)]^2$ is apparent from the above discussions, and a dependence on $[M_i(H)]^2$ must also be weak owing to the following observations: the anisotropy between the longitudinal and transverse MR effects [Figs. 5(a) and 5(b)] at low fields requires that, if MR depends on $[M_i(H)]^2$, $M_i(H)$ is comparable to that of a Co film owing to shape demagnetizing anisotropy and this certainly is not consistent with the observed small Cu polarization [9,19]. It is not difficult to show that in the limit of strong relaxation at the interfaces, i.e., when the mean free path along the [111] direction is comparable to the layer thickness, the MR ratio $[\Delta\rho_{xx}/\rho_{xx}(0)]$ only depends on $[M_i(H)M_{Co}(H)]$. Since direct evidence of polarized interfacial Cu states and the associated interfacial spin-dependent density of states is now available [8,9], we can probe the behavior of the interfacial magnetization $M_i(H)$, particularly its field dependence, through the observed MR. By assuming a paramagnetic interface, we fit the measured MR to the product of measured Co magnetization and a Langevin function in place of M_i [2], as illustrated in Fig. 5. The usual small positive MR contribution, which is quadratic in H [13], is also included in the fit. The quality of the fit is excellent for both field directions.

The size of the MR in this picture depends on the differences in the interfacial spin-dependent density of states at the Fermi level, rather than those of the conventional anti-aligned Co layers. The observed small differences of interfacial spin-dependent density of states [9] would result in a somewhat smaller MR relative to those observed in the antiferromagnetically coupled Co-based multilayers.

In summary, we have investigated magnetotransport properties of MBE grown (111) Co/Cu superlattices. The results reveal that the quantum mechanical side jump in the bulk of the superlattice is responsible for the observed large extraordinary Hall effect with the scaling $R_s \sim \rho_{xx}^2(0)$ for temperatures > 2 K, and that the effect of spin-dependent density of states of the interfacial layers causes the observed high field GMR and leads to the associated scaling between MR and thermopower. The latter effect requires the polarization of the interfacial Cu, and this has now been confirmed by recent spin

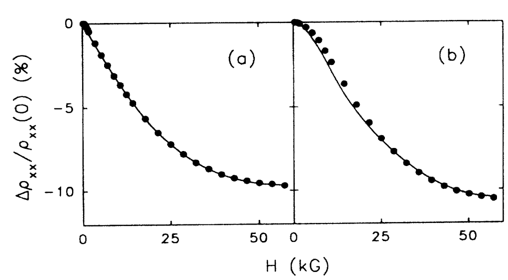

FIG. 5. MR at 50 K for sample $(Co_{15\,Å}/Cu_{6\,Å})_{30}$ (a) for field along the current direction and (b) for field along the [111] growth direction perpendicular to the current. Closed circles are experimental points and lines are fits discussed in the text.

resolved photoemission experiments [9]. From a broader perspective, GMR can arise from a variety of spin-dependent scattering mechanisms not only limited to antiferromagnetically coupled systems and granular films. All of the magnetotransport properties must be studied on an equal footing both experimentally and theoretically so that a better understanding of the microscopic origins of GMR effects can be obtained.

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[1] W. Vavra, C. H. Lee, F. J. Lamelas, H. He, R. Clarke, and C. Uher, Phys. Rev. B **42**, 4889 (1990).

[2] D. Barlett, F. Tsui, D. Glick, L. Lauhon, T. Mandrekar, C. Uher, and R. Clarke, Phys. Rev. B **49**, 1521 (1994); D. Barlett, F. Tsui, L. Lauhon, T. Mandrekar, C. Uher, and R. Clarke, in *Magnetic Ultrathin Films*, edited by B. T. Jonker *et al.*, MRS Symposia Proceedings No. 313 (Materials Research Society, Pittsburgh, PA, 1993), p. 35.

[3] D. Greig, M. J. Hall, C. Hammond, B. J. Hickey, H. P. Ho, M. A. Howson, J. J. Walker, N. Wiser, and D. G. Wright, J. Magn. Magn. Mater. **110**, L239 (1992); M. J. Hall, B. J. Hickey, M. A. Howson, M. J. Walker, J. Xu, D. Greig, and N. Wiser, J. Phys. Condens. Matter **4**, L495 (1992).

[4] P. M. Levy, S. Zhang, and A. Fert, Phys. Rev. Lett. **65**, 1643 (1990); Phys. Rev. B **45**, 8689 (1992); S. Zhang and P. M. Levy, Phys. Rev. B **43**, 11 048 (1991).

[5] Y. Wang, P. M. Levy, and J. L. Fry, Phys. Rev. Lett. **65**, 2732 (1990); D. M. Edwards, J. Mathon, R. B. Muniz, and M. S. Phan, *ibid.* **67**, 493 (1991); P. Bruno and C. Chappert, *ibid.* **67**, 1602 (1991).

[6] S. S. P. Parkin, Phys. Rev. Lett. **71**, 1641 (1993).

[7] F. Tsui, C. Uher, and C. P. Flynn (to be published).

[8] S. Pizzini, C. Giogetti, A. Fontaine, E. Dartyge, G. Krill, J. F. Bobo, and M. Piecuch, in *Magnetic Ultrathin Films* (Ref. [2]), p. 625.

[9] K. Garrison, Y. Chang, and P. D. Johnson, Phys. Rev. Lett. **71**, 2801 (1993); C. Carbone, E. Vescovo, O. Rader, W. Gudat, and W. Eberhardt, *ibid.* **71**, 2805 (1993).

[10] We have observed weak anisotropy in the high field magnetotransport properties for fields applied along various orientations with respect to the current. In MR the form of the anisotropy in high fields is $\sim d\cos 2\theta$ about the current direction with d about 1%. The twofold symmetry evidently arises from the effect of spin-orbit coupling [see B. E. Miller, E. Y. Chen, and E. D. Dahlberg, J. Appl. Phys. **73**, 6384 (1993)].

[11] C. M. Hurd, *The Hall Effect in Metals and Alloys* (Plenum, New York, 1972).

[12] The absence of correlation between the MR and the Co magnetization is even more evident for applied fields in the growth plane, where the saturation field of the ferromagnetic Co layers is at least 100 times smaller than that of the MR [2].

[13] J. M. Ziman, *Electrons and Phonons* (Oxford Univ. Press, Oxford, 1960), Chap. 11.

[14] L. Berger, Phys. Rev. B **2**, 4559 (1970).

[15] P. Xiong, G. Xiao, J. Q. Wang, J. Q. Xiao, J. S. Jiang, and C. L. Chien, Phys. Rev. Lett. **69**, 3220 (1992).

[16] S. N. Song, C. Sellers, and J. B. Ketterson, Appl. Phys. Lett. **59**, 479 (1991).

[17] L. Xing, Y. C. Chang, M. B. Salamon, D. M. Frenkel, J. Shi, and J. P. Lu, Phys. Rev. B **48**, 6728 (1993); J. Shi, S. S. P. Parkin, L. Xing, and M. B. Salamon, J. Magn. Magn. Mater. **125**, L251 (1993).

[18] I. A. Cambell, A. Fert, and A. R. Pomeroy, Philos. Mag. **15**, 977 (1967); A. Fert, J. Phys. C **2**, 1784 (1969); A. Fert and I. A. Cambell, J. Phys. F **6**, 849 (1976).

[19] The observed total moment for a 15 Å thick Co layer is within 5% of the pure Co value. This sets an upper limit for the interfacial magnetization of $< 1/5$ of the Co moment.