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The giant magnetoresistance and the anomalous Hall effect in molecular-beam-epitaxy grown Co/Cu superlattices

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Abstract. We have investigated the dependence of the magnetoresistance (MR) and the Hall effect on both the Cu layer thickness d_{Cu} and the Co layer thickness d_{Co} in (111)-oriented epitaxial Co/Cu superlattices. The MR ratio has a single peak near 9 Å as a function of d_{Cu}, while it shows a broader faint peak near 10 Å as a function of d_{Co}. The maximum MR ratio up to 81% was observed at 4.2 K for $d_{Co} = 9.0$ Å and $d_{Cu} = 9.3$ Å. For both d_{Cu} and d_{Co} dependence, the saturation field increases monotonically with decreasing layer thickness. As a function of d_{Co}, the anisotropy field changes its sign from in plane for large d_{Co} to perpendicular near $d_{Co} = 9$ Å.

The extraordinary part of Hall resistivity shows a single peak against magnetic field for samples with a large MR ratio. The field dependence is explained in terms of the field dependence of both the skew and the side-jump scattering components associated with the giant magnetoresistance. The ratio of the two components is almost unchanged between zero field and saturation field, which suggests that the main scattering mechanisms at both fields are microscopically the same.

1. Introduction

The giant magnetoresistance (GMR) effect was first discovered by Baibich *et al* [1] for Fe/Cr multilayers. Parkin *et al* [2] have subsequently found oscillations both in the MR ratio and in an exchange coupling energy between magnetic layers as a function of non-magnetic spacer layer thickness, which is directly related to the origin of the GMR. Since then, considerable combinations of ferromagnetic/non-magnetic multilayers [3–7] have been reported to show the GMR effect and oscillatory behaviour of the exchange coupling strength. Among them, the Co/Cu system has been most intensively investigated, since it shows the largest MR ratio at relatively low fields. In the sputter-deposited samples, clear oscillations both in the MR ratio and in the saturation field have been observed [6, 7], while the MBE-grown epitaxial superlattices were reported to show considerable differences from the sputtered samples, which are still controversial [8–13]. The key issues are summarized as follows.

(a) For the sputtered samples, a MR ratio over 100% is reported at 4.2 K while, for the MBE-grown epitaxial samples, the MR ratio is usually three or four times smaller. Recently Harp *et al* [11] reported a systematic study of the dependence of the MR on the growth temperature in MBE-grown wedged superlattices and obtained a maximum MR ratio of about 45%.

(b) For sputtered Co/Cu multilayers, oscillations both in the interlayer magnetic coupling and in the MR magnitude have been clearly observed while, for MBE-grown samples, no oscillations were observed and the large saturation field for small d_{Cu} has been invoked as evidence of strong antiferromagnetic coupling. However, there is some inconsistency between the MR value and the large saturation field. Recently, Hall *et al* [12] reported the first observation of oscillations in the saturation field, but they have not succeeded in observing oscillations in the MR ratio for the same samples.

In this paper, we describe the systematic measurement of the dependence of the MR on both d_{Cu} and d_{Co} in order to investigate these issues. In addition, we have measured the Hall effect which gives information on the left–right asymmetric part of conduction electron scattering. For a better understanding of the GMR effect, it is important to characterize the conduction electron scattering by the simultaneous measurement of other electrical transport properties. In fact, until now, thermoelectric power [14–16], thermal conductivity [17] and Hall effect [18, 19] measurements on multilayer systems have been undertaken. We have found an anomalous field dependence of the Hall resistivity for an Fe/Cr multilayer with a large MR and explained the dependence as a reduction of left–right asymmetric conduction electron scattering correlated with the GMR [19]. It is of interest to know whether it is a common character of multilayers exhibiting GMR.

2. Experimental procedure

All samples were grown on sapphire (0001) substrates on which a 62 Å Cu(111) buffer layer was deposited under a working pressure lower than 3×10^{-9} Torr. The layer periodicity is always 50. Both Co and Cu were evaporated using electron beam sources at a rate of 0.4 Å s^{-1}. The Cu buffer layer was first deposited at room temperature (RT) and was annealed at 100 °C for 30 min in order to improve the surface smoothness. The Co/Cu superlattices were then grown at RT. *In situ* reflection high-energy electron diffraction (RHEED) showed a sixfold symmetry for both the Cu buffer layer and the superlattices, which suggests that they were (111) oriented but twinned texture. Twinning seems to be common for MBE growth of the (111)-oriented Co/Cu superlattices [11, 20, 21]. The artificial periodicity was also confirmed by x-ray diffraction analyses from superlattice peaks in both small- and large-angle regions for all the samples. For the smaller-layer-periodicity samples, only the first-order satellite has been observed. The samples were shaped to an appropriate form for transport measurements using the photolithographic method. The details of preparation and characterization of samples have been described elsewhere [9, 22].

Two series of samples were prepared by an identical method except that the second series were grown several months later. Judging from x-ray analyses, no clear structural difference was observed between two series. The d_{Cu} and d_{Co} dependence of MR characteristics are roughly similar; however, the absolute values of MR ratio are somewhat larger for the second series of samples on the whole owing to some uncontrolled variation in sample preparation conditions.

The magnetoresistance and the Hall voltages were measured by the standard DC four-probe method using a computer-controlled current source and Keithley-181 nanovoltmeters. A magnetic field strength of up to 5 T was applied both in the layer plane and perpendicular to the plane using a superconducting magnet. The magnetization measurement was performed using a Quantum Designs SQUID magnetometer.

3. Results and discussion

3.1. Magnetoresistance

Figure 1 shows the transverse MR of Co(9.0 Å)/Cu(9.3 Å) for a magnetic field both in

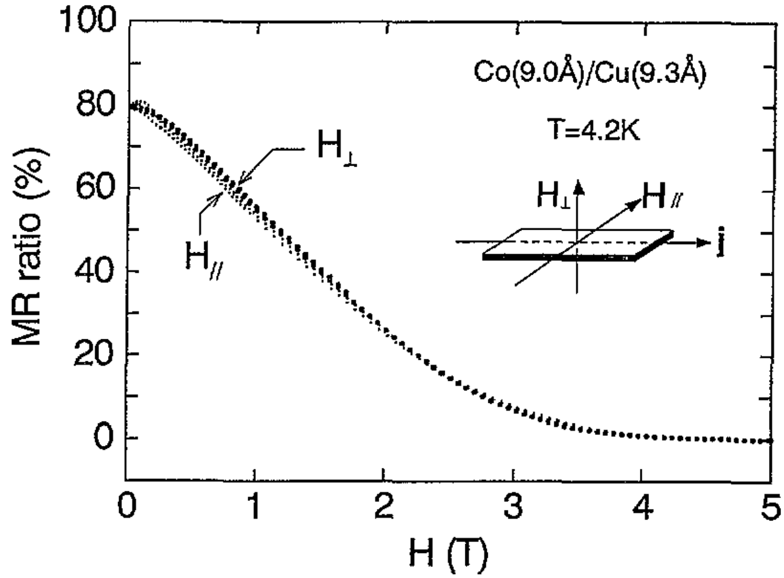

Figure 1. The field dependence of the MR ratio for Co(9.0 Å)/Cu(9.3 Å) with the field in the sample plane ($H_{\parallel}$) and perpendicular to the plane ($H_{\perp}$). The experimental geometries are also shown.

plane ($H_{\parallel}$) and perpendicular to the plane ($H_{\perp}$) at 4.2 K. From this figure, the following characteristic features can be realized.

(a) The MR ratio has a record value of 81% at 4.2 K ever reported for MBE-grown Co/Cu superlattices. In this paper, the MR ratio is defined as $(\rho_m - \rho_s)/\rho_s$ using the maximum resistivity (ρ_m) and the resistivity (ρ_s) at saturation field. ρ_m is replaced by the field-dependent resistivity $\rho(H)$ when we discuss the field dependence.

(b) The saturation field H_s is larger than that reported for the sputtered samples with a comparable layer thickness, in agreement with the results reported before [8–13]. H_s is defined here as the field where the field dependence of the MR ratio becomes less than 1%.

(c) Both the anisotropy against the field direction and the hysteresis against the field strength are very small in this sample.

Figures 2(a) and 2(b) show the field dependences of the MR ratio and of H_s on d_{Cu} for a constant d_{Co} of 16.8 Å. When the scatter in the data is taken into account, the MR ratio shows only a single peak near 9 Å, and H_s keeps increasing with decreasing d_{Cu} down to $d_{Cu} = 5$ Å. The dependence is roughly similar to the results obtained by Harp *et al* [11] and Hall *et al* [12]; however, the peak value of the MR ratio is about twice their result. The large MR ratio may be mainly due to the optimized buffer layer thickness and the layer periodicity. We have first made samples with a 1000 Å Ag buffer layer in order to obtain high-quality epitaxial samples. However, they showed a positive MR at low temperatures, resulting from the normal MR of the Ag buffer layer. In general, the thicker buffer layer improves the sample quality. However, the thicker buffer layer results in a larger shunting effect. In order to reduce the shunting effect, a large layer periodicity is preferable. However, we know that the topological roughness of samples tends to increase with increasing layer periodicity. We have tried to optimize both the buffer layer thickness and the layer periodicity in order to obtain a larger MR ratio. In the present experiment, a

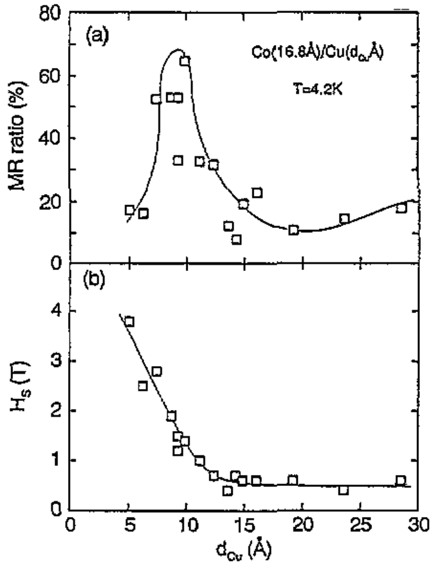

Figure 2. The Cu layer thickness d_{Cu} dependence of (a) the MR ratio and (b) the saturation field H_s for the field in plane measured at 4.2 K: ——, guides to the eye.

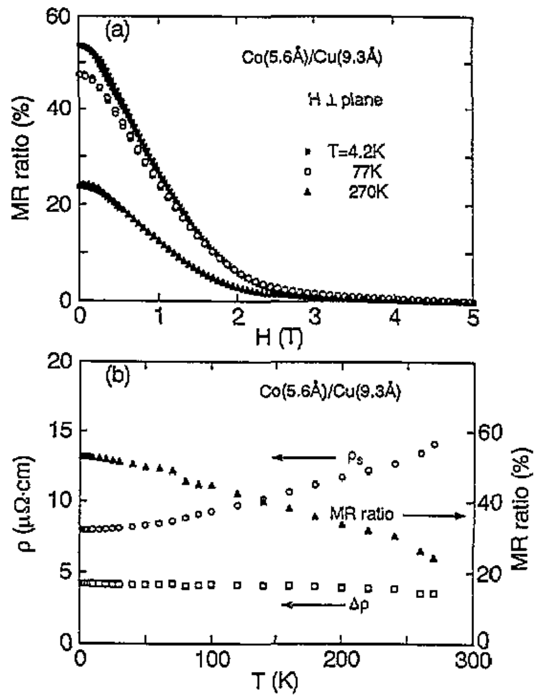

Figure 3. (a) The field dependence of the MR ratio for the field perpendicular to the plane at $T = 4.2$, 77 and 270 K. The H_s-values are 2.4 T, 2.4 T and 2.3 T for $T = 4.2$ K, 77 K and 270 K, respectively. (b) The temperature dependence of ρ at 5 T (ρ_s), $\Delta\rho$ and the MR ratio for Co(5.6 Å)/Cu(9.3 Å).

62 Å Cu buffer layer and a layer periodicity of 50 were selected. The sample quality may not be much different from those for the samples used by Harp *et al* and Hall *et al*, except for some minor differences in the microscopic interfacial structure invisible by RHEED and x-ray analyses. In order to discuss the reason for the large MR ratio quantitatively, however, broader knowledge such as the absolute values of ρ or magnetization in their experiments is needed.

In the work by Harp *et al* (figure 5 in [11]), the MR ratio has a single peak near $d_{Cu} = 10$ Å, and the H_s increases monotonically with decreasing d_{Cu} without oscillation. The results obtained by Hall *et al* are different. They observed oscillations in H_s as a function of d_{Cu} with two peaks at 7 and 18 Å, although no oscillations were observed in the MR ratio. We cannot discard the effect of topological roughness as a possible origin of the absence of oscillations. With increasing layer number, the topological quality of samples usually becomes worse. In fact, for the trilayer samples, for which better sample quality is expected than for the superlattices, oscillatory behaviour in the interlayer coupling between Co layers through a Cu (111) spacer has been reported [20, 23]. Even in the trilayer structures, the interlayer coupling is reported to depend crucially on the samples [20, 23–25]. Dupas *et al* [25] have succeeded in observing oscillations in MR on UHV-evaporated Co/Cu(111) trilayers, although the MR ratio obtained was only a few per cent. Very recently, Schreyer *et al* [21] have succeeded in observing oscillations in the interlayer coupling strength on carefully deposited Co/Cu(111) superlattices.

We have also measured the temperature dependence of MR on several samples. The resistivities both at zero field and at 5 T vary approximately as T^2 at low temperatures, suggesting s–d scattering as in pure ferromagnetic materials [26]. $\Delta\rho$ depends only weakly on temperature (figure 3(b)). The temperature-dependent reduction in the MR ratio is thus mainly due to the increase in ρ_s, which is consistent with the results reported for the sputtered samples [6]. Figure 3(a) shows that H_s also depends only weakly on temperature.

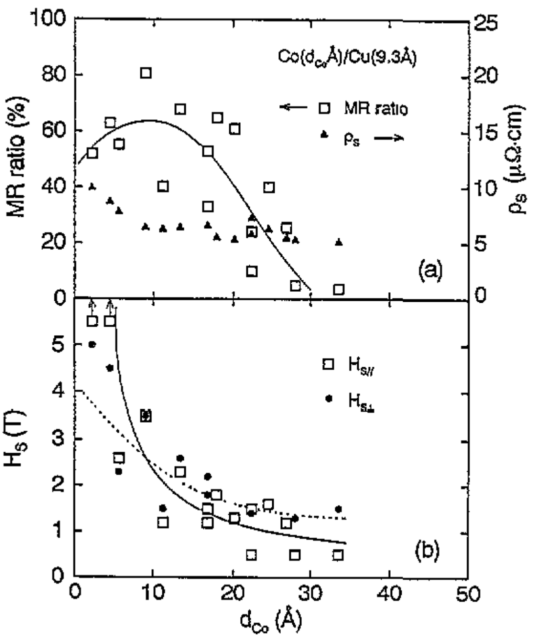

Figure 4. (a) The Co layer thickness d_{Co} dependence of the MR ratio and the saturation resistivity ρ_s at 4.2 K for Co(d_{Co} Å)/Cu(9.3 Å). (b) The d_{Co} dependence of H_s for the field in plane ($H_{s\parallel}$) and perpendicular to the plane ($H_{s\perp}$) at 4.2 K on Co(d_{Co} Å)/Cu(9.3 Å). For $d_{Co} < 4.5$, $H_{s\parallel}$ becomes larger than 5 T: ——, – – –, guides to the eye.

The dependences of the MR ratio and H_s on d_{Co} have not been reported for Co/Cu multilayers (or superlattices) previously, except for our preliminary report [13]. Figure 4(a) shows the dependence of the MR ratio on d_{Co} for a constant d_{Cu} of 9.3 Å. As ρ_s, we employed ρ-values at 5 T under $H_\parallel$ geometry for $d_{Co} > 9$ Å and under $H_\perp$ geometry for $d_{Co} < 9$ Å. The d_{Co} dependence of the MR ratio is similar to that for the sputtered samples reported in [27], except that the MR ratio for MBE-grown samples decreases more sharply for larger d_{Co}. These behaviours may also be related to the larger topological roughness.

Figure 4(b) shows H_s determined from the MR as a function of d_{Co}. H_s increases monotonically with decreasing d_{Co} below about $d_{Co} = 25$ Å. Very recently, it was reported that the interlayer coupling also depends in an oscillatory manner on the Co layer thickness in Co/Cu(100) trilayers [28]. In the present experiment, however, we could not confirm oscillations. A crossover in the anisotropy in H_s can be seen from $H_{s\parallel} > H_{s\perp}$ to $H_{s\parallel} < H_{s\perp}$ near $d_{Co} = 9$ Å, taking into account the large scatter in the data points. For the samples with $d_{Co} < 4.5$ Å, 5 T is not a sufficiently high magnetic field strength to saturate the MR for the $H_\parallel$ geometry. This suggests perpendicular magnetic anisotropy in the samples with $d_{Co} < 9$ Å, which is consistent with the result reported by Harp *et al* [11]. They measured a d_{Cu} dependence of the MR ratio for a constant d_{Co} of 8 Å and obtained perpendicular magnetic anisotropy for all d_{Cu}-values. In order to see the anisotropy more clearly, typical examples of the field dependence of the MR and the magnetization M are shown in figure 5. For $d_{Co} = 11.2$ Å, both the MR and M saturate at lower fields for in-plane geometry. The reverse is the case for $d_{Co} = 5.6$ Å, although the field dependence of M is not simple below 0.5 T.

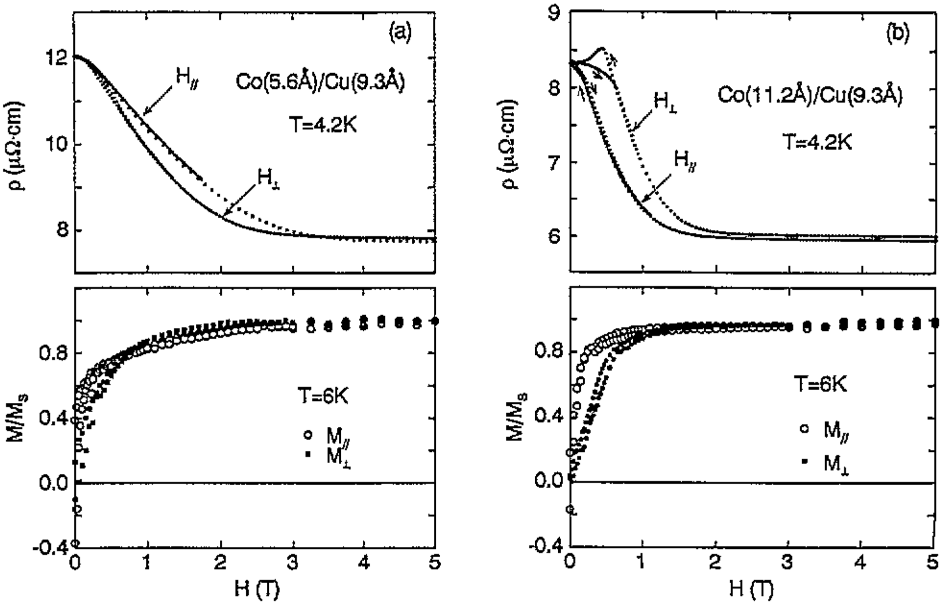

Figure 5. The field dependence of MR and the magnetization at 4.2 K for the field in plane ($H_{\parallel}$ and $M_{\parallel}$) and perpendicular to the plane ($H_{\perp}$ and $M_{\perp}$) for (a) Co(5.6 Å)/Cu(9.3 Å) and (b) Co(11.2 Å)/Cu(9.3 Å).

Next we discuss the origins of the large saturation field H_s. On comparison of figures 2(b) and 4(b), it is inferred that the large H_s in MBE-grown Co/Cu superlattices compared with that in the sputtered multilayers is not simply related to the antiferromagnetic coupling between adjacent continuous Co layers. Down to the thinnest layer thickness, the x-ray analysis confirms the existence of an artificial periodicity and gives no clear sign of structural change. It is reported that MBE-grown Co/Cu(111) superlattices show a larger surface roughness than do the sputtered samples [11]. A STM analysis [22] shows that our samples also have topologically rough surfaces, and we naturally expect the interfaces between layers also to be irregular. Taking into account the fact that granular Co–Cu alloy systems also show a GMR with large H_s [29, 30], we infer that a granular-like component exists in MBE-grown Co/Cu superlattices resulting from the topological irregularity, as first proposed by Harp *et al.* They have discussed the existence of the granular component in MBE-grown Co/Cu(111) superlattices based on detailed structural analysis and suggested that the absence of oscillations in the MR ratio and the large H_s are due to the granularity [11]. This explains why Dupas *et al* [25] have observed the oscillations in the MR ratio, since the smoother interface is easily expected for samples with only trilayers. As an origin of granularity, de la Figuera *et al* [31] recently suggested the FCC (111) twin-related structure, which is inevitable for the growth of (111)-oriented MBE-grown Co/Cu superlattices.

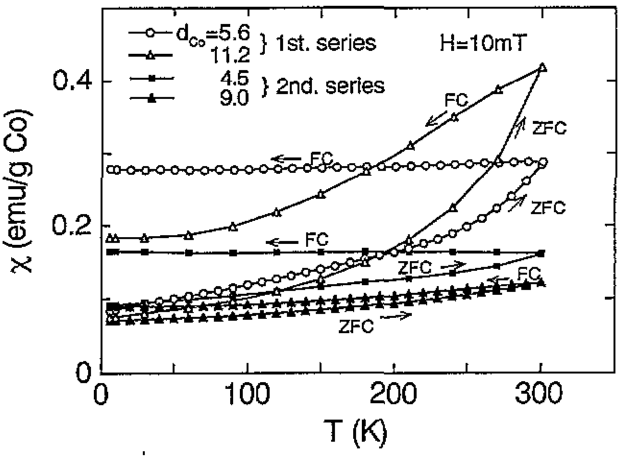

Figure 6. The temperature dependence of the susceptibility for Co(d_{Co} Å)/Cu(9.3 Å) (d_{Co} = 4.5, 5.6, 9.0 and 11.2) measured under an applied field strength of 10 mT for zero-field-cooled (ZFC) (from 6 to 300 K) and field-cooled (FC) (from 300 to 6 K) conditions, where the ZFC susceptibilities were measured with increasing temperature after being cooled to 6 K under zero field. ——, guides to the eye.

If such a granular-like component plays a role, a characteristic behaviour can be expected in the temperature dependence of the susceptibility; that is zero-field-cooled and field-cooled susceptibilities follow different paths [29]. In confirmation of this view, we measured the temperature dependence of the susceptibility for several samples. As shown in figure 6, one can clearly see the remanence effect described above, which suggests the existence of a granular-like component in the MBE-grown Co/Cu superlattices. A similar behaviour has been observed for Ag/Co multilayers in thin Co regions [32]. For Ag/Co, the contribution from the granular-like component possibly dominates in the thin-layer-thickness samples, since the maximum MR ratio of about 80% reported for granular samples [33] is larger than for the multilayer samples (about 15% for current parallel to the layer planes) reported before [34]. For the Co/Cu system, however, the maximum MR ratio reported for granular samples is 50% [35], while that in the present experiment is 81%. We believe that the granular-like component contributes only partly to the large MR ratio in the present experiment and the multilayer structure is still essentially important. Firstly, the artificial periodicity was confirmed down to the smallest-layer-periodicity sample, which was also supported by the anisotropy observed in the MR curves in figure 5. Secondly, the difference between the low-temperature remanence, of the two sample series cannot be understood consistently if we assume only a granular-like contribution. We naturally expect that the larger remanence results from the more granular components. In fact, the remanence effect in the magnetization measurement increases monotonically with decreasing d_{Co} within each series, in agreement with the increasing H_s with decreasing d_{Co}. On comparison of the two series, however, the granular-like component in the first series (open symbols) is larger than that in the second series (full symbols) judging from figure 6, while the MR ratio and H_s are generally smaller for the first series. These facts also show that the granular-like component does not contribute exclusively but only partly to the GMR effect in the MBE-grown Co/Cu superlattices [36].

3.2. Hall effect

In ordinary ferromagnetic thin films, the field dependnece of the Hall resistivity is described empirically as

$$\rho_H = R_0 H + R_S 4\pi M \qquad (1)$$

where the first term is the ordinary Hall component (R_0 is the ordinary Hall coefficient), and the second term, resulting from magnetic conduction electron scattering, is called an extraordinary Hall component ρ_H^M (R_S is the extraordinary Hall coefficient). If R_S is field independent, the field dependence of ρ_H is a simple superposition of a linear ordinary component and an extraordinary component proportional to M.

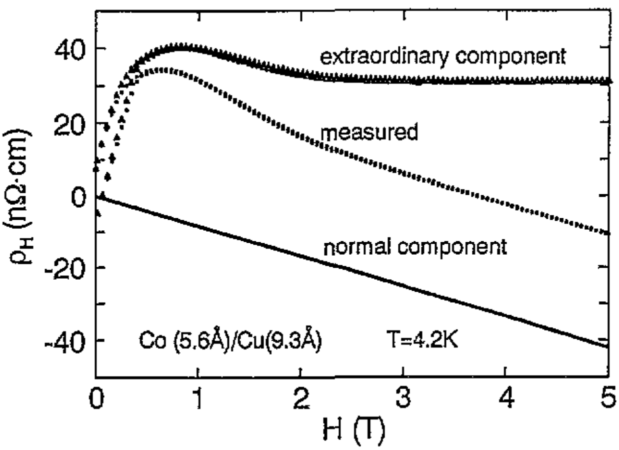

Figure 7. An example of the procedure to decompose measured Hall resistivity into the normal and the extraordinary components for Co(5.6 Å)/Cu(9.3 Å) at 4.2 K.

Figure 7 shows an example of the procedure to decomose the measured Hall resistivity ρ_H into the normal component and the extraordinary component in Co/Cu superlattice with a large MR ratio according to equation (1). We have assumed that the extraordinary component saturates at higher fields and the slope of ρ_H versus H gives the ordinary Hall coefficient R_0. Thus the R_0 determined is always negative and close to the reported bulk Cu value for large d_{Cu} samples and is close to that of bulk Co for large d_{Co} [37]. This indirectly supports the assumption that we applied in the analysis. Typical examples of the thus obtained ρ_H^M are compared with the corresponding MR curve in figure 8. ρ_H^M for the samples with a large MR ratio shows an anomalous field dependence compared with ordinary ferromagnetic thin films, where we never expect to find such a peak against magnetic field. A similar field dependence has been reported for the Fe/Cr system [18, 19]. In [19], we proposed that the anomalous field dependence is directly related to the field dependence of electrical resistivity correlated with GMR. Since R_S represents the magnitude of the asymmetric part of conduction electron scattering, it is naturally expected that the reduction in ρ is also reflected in R_S. In fact, experimentally, the anomalous field dependence of ρ_H^M was observed only for samples with a large MR ratio.

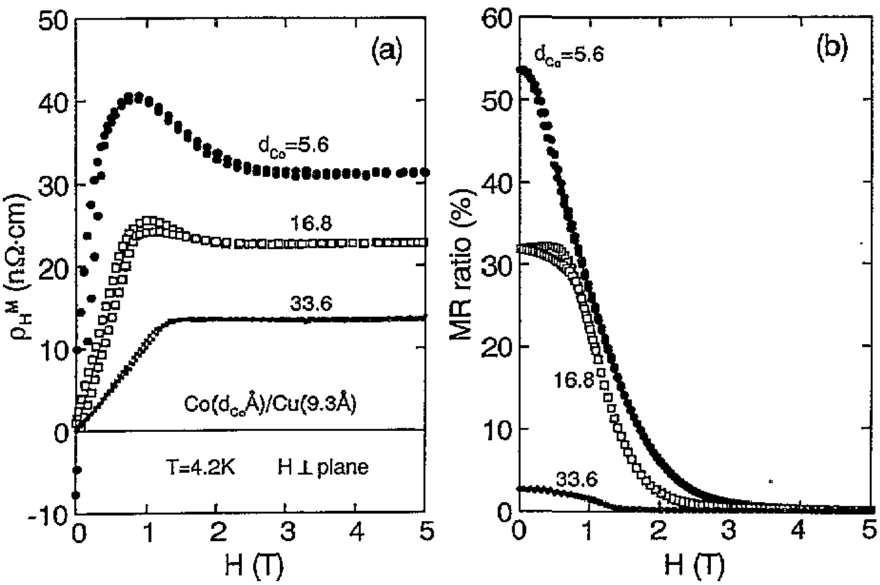

Figure 8. Typical examples of the field dependence of (a) the extraordinary part of Hall resistivity together with (b) the corresponding MR ratio for the field perpendicular to the plane for Co(d_{Co} Å)/Cu(9.3 Å) at 4.2 K.

The left–right asymmetric scattering of conduction electrons responsible for the extraordinary Hall effect are classified into two mechanisms, namely the skew scattering and the side-jump mechanisms [37, 38]. The most general expression for R_S is

$$R_S = a\rho + b\rho^2 \qquad (2)$$

where the first and the second terms represent the skew and the side-jump components, respectively. The coefficients a and b are characteristics of samples. If a and b are field independent and the field dependence of the conduction electron scattering is only through ρ, $R_S(H)/\rho(H)$ of a single sample will be plotted on a linear line against $\rho(H)$. Typical examples of such plots are shown in figure 9. When the accuracy of the experiment is taken into account, the plot for each curve is approximately linear. We do not go into more details of the result in the present paper, although a slight upturn for the larger ρ region (the lower fields) seems to be a general tendency. The independence of the parameters a and b of the magnetic field gives an important hint that the microscopic character of conduction electron scattering (at low fields) responsible for the GMR is basically the same as that of residual scattering at high fields ($H > H_s$), judging from their left–right asymmetry.

From the intercept and the slope of a least-squares fitting, we determined the mean values of a and b for each sample. In order to see how a single pair of field-independent a and b can reproduce the experimental $\rho_H^M(H)$, typical examples of the comparison between the computed and the experimental $\rho_H^M(H)$ are shown in figure 10. In the computation, we combined equations (1) and (2) using the experimental values of $\rho(H)$ and $M(H)$ measured in the same experimental geometry and at the same temperature. For $d_{Co} = 5.6$ Å, we also show the skew and the side-jump components separately. The field dependence is reasonably well reproduced for all the samples from the low-field hysteresis to the high-field saturation.

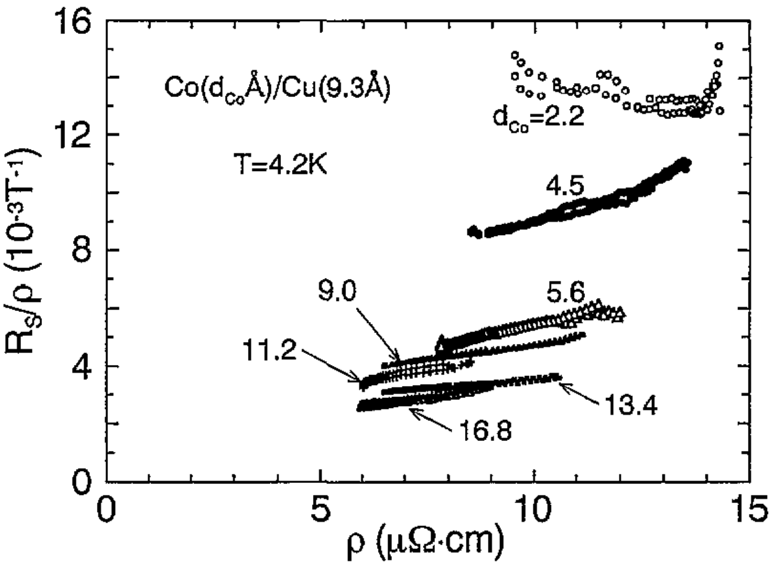

Figure 9. $R_S(H)/\rho(H)$ versus $\rho(H)$ for Co(d_{Co} Å)/Cu(9.3 Å) at 4.2 K.

The existence of the peak largely correlates with the fact that the saturation field of the MR is larger than that of the magnetization.

The dependences of a and b (magnitude of the skew and the side-jump components) on d_{Co} are shown in figure 11. The signs of both components are always positive except for d_{Co} = 2.2 Å. It should be noted that the error in the computed $\rho_H^M(H)$ is large for d_{Co} = 2.2 and 4.5 Å because of the difficulty in measuring the small magnetization. Taking into account the error in the determination, we can say that the skew and the side-jump components increase slightly with decreasing d_{Co} except for d_{Co} = 2.2 Å. In order to test the effect of temperature on the parameters a and b, we also repeated the same procedure at 270 K for several samples. An example for d_{Co} = 5.6 Å is shown in figure 12, where the $R_S(T)/\rho(T)$ versus $\rho(T)$ at 5 T is also plotted, with temperature as a parameter. We can judge that the skew component a is larger at higher temperatures, although the scatter in the data points at 270 K is large owing to the temperature drift. It is usually believed that the skew component decreases at high temperatures in paramagnetic metals and alloys, since the magnetization decreases with increasing temperature [38]. For Co/Cu superlattices, however, the reverse seems to be true as seen from the $R_s(T)/\rho(T)$ versus $\rho(T)$ at 5 T. At low temperatures, ρ_H^M (5 T) and ρ (5 T) are only weakly dependent on temperature experimentally. The rather sharp increase in $R_S(T)/\rho(T)$ against $\rho(T)$ between 8 and 9 $\mu\Omega$ cm at 5 T in figure 12 is accordingly dominated by the reduction in M, which varies almost linearly up to room temperature as expected for a two-dimensional ferromagnet. Under such conditions, the parameters a and b in equation (2) do not depend on the magnetic field at a fixed temperature but depend on the temperature.

Finally we must mention our assumption that the multilayer samples obey the same model as homogeneous ferromagnetic alloy films in the analysis; we used M-values normalized not to the Co volume but to the total sample volume, since we have no practical formula to analyse the Hall effect of multilayer samples. Whichever M is used in the analysis, however, our discussion undergoes no essential change exept for the thickness dependence of R_S.

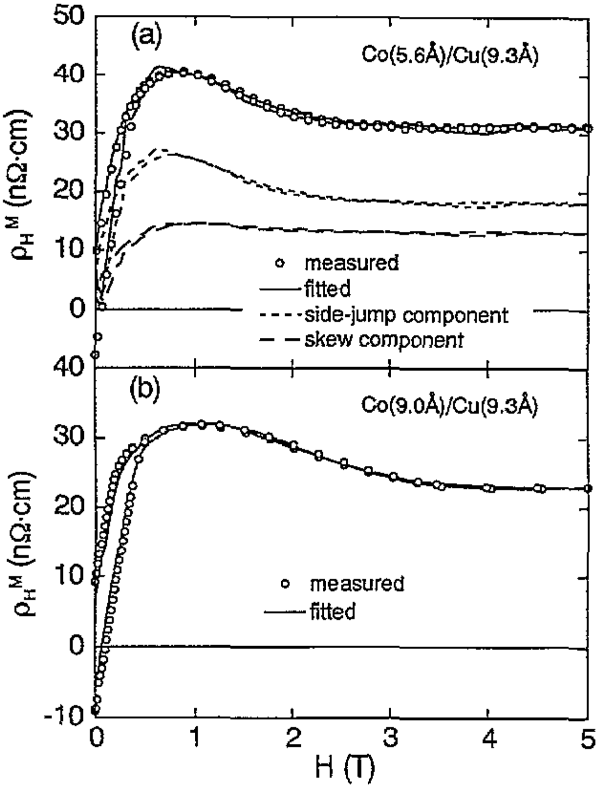

Figure 10. The results of the fitting to $\rho_{\mathrm{H}}^{M} = (a\rho + b\rho^2)M$ using the measured $\rho(H)$ and $M(H)$ for (a) Co(5.6 Å)/Cu(9.3 Å) and (b) Co(9.0 Å)/Cu(9.2 Å). For Co(5.6 Å)/Cu(9.3 Å), the contribution from each component $a\rho M$ (skew component) and $b\rho^2 M$ (side-jump component) are also shown.

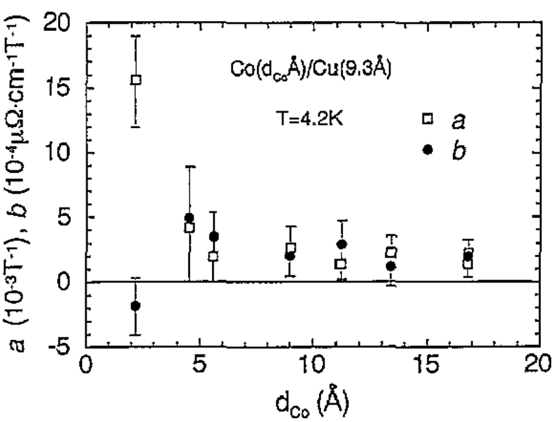

Figure 11. The dependences of a and b on d_{Co}.

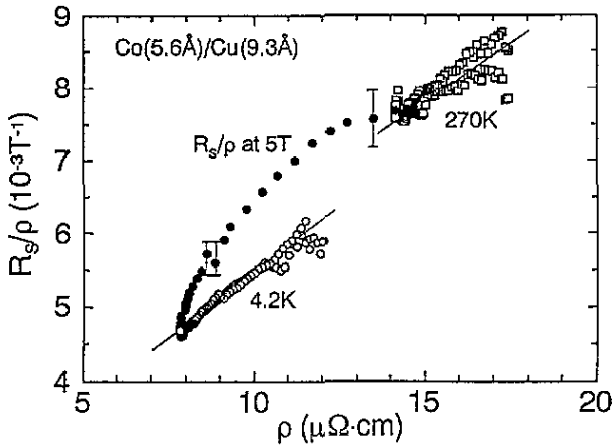

Figure 12. $R_s(H)/\rho(H)$ versus $\rho(H)$ at 4.2 and 270 K for Co(5.6 Å)/Cu(9.3 Å). $R_s(T)/\rho(T)$ versus $\rho(T)$ at 5 T is also shown, with the temperature T as a parameter.

4. Conclusion

A maximum MR ratio of 81% has been observed, suggesting that the small MR ratio reported previously is not an essential feature of MBE-grown Co/Cu superlattices. From the comparison of the d_{Cu} and d_{Co} dependences of H_s, we infer that the large H_s observed for MBE-grown samples is not simply due to the large antiferromagnetic coupling between Co layers. The temperature dependence of the low-field magnetic susceptibility verified the existence of some granular-like component. In order to understand the experimental results, the effects from both the artificial periodicity and the granular-like component must be taken into account.

An anomalous field dependence of the extraordinary Hall effect was observed. The approximate linear variation in the R_S/ρ versus ρ plot was obtained by the analysis using equation (2). This suggests that the microscopic mechanisms of conduction electron scattering responsible for the GMR is essentially the same as that or residual scattering at saturation fields in MBE-grown Co/Cu superlattices, from the viewpoint of the left–right asymmetry.

Acknowledgments

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