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# Transport properties in granular Co–Ag alloys

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**Abstract.** We have investigated the field dependence of the magnetoresistance, Hall effect, thermal conductivity and thermoelectric power on Co–Ag granular films made by electron gun evaporation under ultrahigh-vacuum conditions. A large field dependence correlated with the giant magnetoresistance is observed in all the transport properties. The dependence is anisotropic for 42 at.% Co–Ag between parallel and perpendicular fields. At zero field, a resistivity minimum is found that varies with annealing temperature. The behaviours of the magnetothermal conductivity and the magnetothermoelectric power are generally consistent with previous studies. The extraordinary Hall resistivity exhibits an anomalous field dependence and changes sign with temperature.

## 1. Introduction

Following intense research efforts from both experimental [1] and theoretical [2] points of view, the basic mechanism of the giant magnetoresistance (GMR) in magnetic multilayers is now understood to a considerable extent. However, there still remain many interesting characteristics to be understood [3, 4].

To obtain deeper insight into the GMR effect, measurements of additional transport properties, such as thermal conductivity, thermoelectric power (TEP) and Hall effect have been made on magnetic multilayers [5–11]. From resistance measurements, one can estimate only the change in magnitude of conduction electron scattering. The thermal conductivity allows the inelastic nature of the conduction electron scattering responsible for the GMR to be probed. The TEP gives information about the energy dependence of electron scattering. The Hall effect is advantageous for obtaining information about the left–right asymmetry of the conduction electron scattering, i.e. the asymmetry in the spin-dependent scattering responsible for the GMR effect.

Recently, GMR has also been observed in granular structures such as Co–Cu, Fe–Cu, Fe–Ag and Fe–Au [12–15]. Some measurements of magnetothermal properties and Hall effect have already been reported for Co–Ag granular systems prepared mostly by sputtering, although there still remain several problems in debate [16–20]. In this paper, we present some new results and compare data for sputtered and evaporated samples.

## 2. Experiment

Samples with Co concentrations of 27.2 at.% ( $\approx 20$  vol.%) and 42 at.% ( $\approx 33$  vol.%) were evaporated in ultrahigh vacuum onto glass substrates at room temperature. The samples for the Hall effect and the MR were evaporated with a mask suitable for transport

measurements. The samples for the thermal measurements were evaporated onto glass substrates approximately 80  $\mu\text{m}$  thick without a mask, to reduce thermal shunting by the substrate. The MR was also measured on the latter samples simultaneously with the field dependence of thermal properties. The total sample thickness was always fixed at 500 nm. To investigate the effect of annealing, the samples were annealed at ( $T_A =$ ) 240, 280, 310 and 320  $^\circ\text{C}$  in evacuated glass tubes for 15 min. The TEP and the thermal conductivity measurements were made by the ordinary static heat flow method. The Hall effect and the MR were measured by conventional DC four-probe methods between 1.5 and 300 K up to magnetic fields  $H = 5$  T. The voltages were obtained using Keithley 181 nanovoltmeters. The magnetization measurements were made of a Quantum Design SQUID magnetometer up to 5.5 T.

### 3. Results and discussion

To compare our transport measurements with those previously reported for the sputtered samples, we first show in figure 1(a) typical examples of the field dependences of the resistivity  $\rho(H)$  at 4.2 and 273 K for 27.2 at.% Co samples. The results generally agree with those on the sputtered sample obtained by Xiao *et al* [14]. The MR ratios [ $\rho_{\text{max}} - \rho(5 \text{ T})$ ]/ $\rho_{\text{max}}$  ( $\rho_{\text{max}}$  is the maximum resistivity) at 4.2 K (273 K) are 42% (22%), 45% (24%), 39% (20%) and 37% (20%) for  $T_A =$  room temperature (RT) (non-annealed), 240  $^\circ\text{C}$ , 280  $^\circ\text{C}$  and 310  $^\circ\text{C}$ , respectively. Even for the non-annealed state, the MR ratio of the present sample ( $T_A = \text{RT}$ ) is larger than 35% of the sputtered sample annealed at 200  $^\circ\text{C}$  in [14], which suggests that the separation of Ag- and Co-rich phases has already progressed considerably in the evaporation process. The maximum MR ratio, obtained for  $T_A = 240$   $^\circ\text{C}$ , is almost the same as that reported for the sputtered sample [14].

For a Co concentration of 42 at.%, an anisotropy is found in the field dependence of

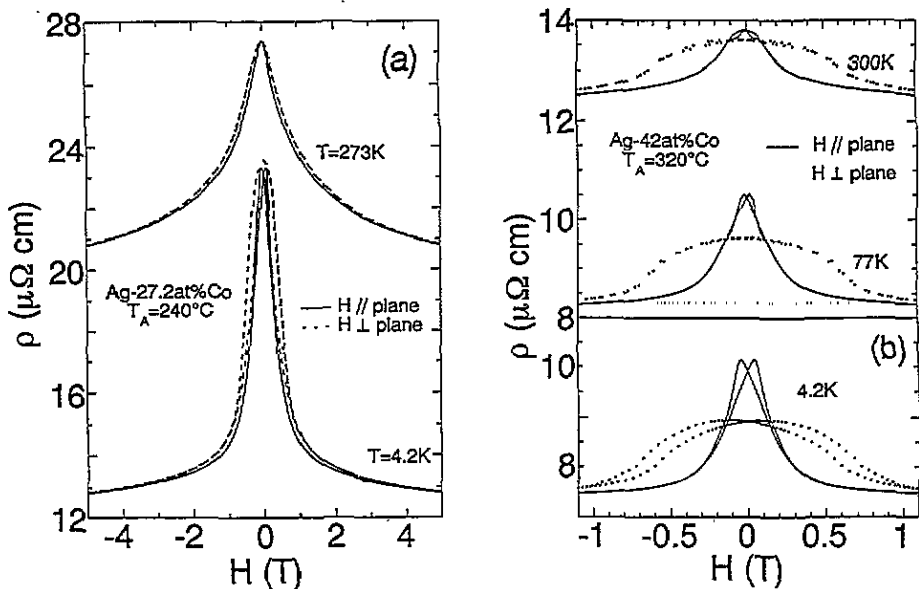


Figure 1. (a) Field dependence of the resistivity for 27.2 at.% Co-Ag annealed at ( $T_A =$ ) 240  $^\circ\text{C}$ . (b) Anisotropy of the field dependence of the resistivity for 42 at.% Co-Ag annealed at 320  $^\circ\text{C}$ .

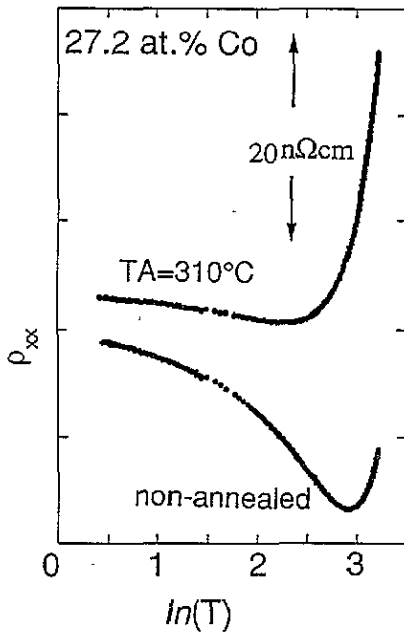


Figure 2.  $\ln T$  dependence of the electrical resistivity for 27.2 at.% Co (non-annealed and  $T_A = 310^\circ\text{C}$ ). The vertical axis is shifted arbitrarily for clarity. The resistivity at the minimum is  $26.0 \mu\Omega \text{ cm}$  for the non-annealed specimen and  $10.7 \mu\Omega \text{ cm}$  for the  $T_A = 310^\circ\text{C}$  specimen.

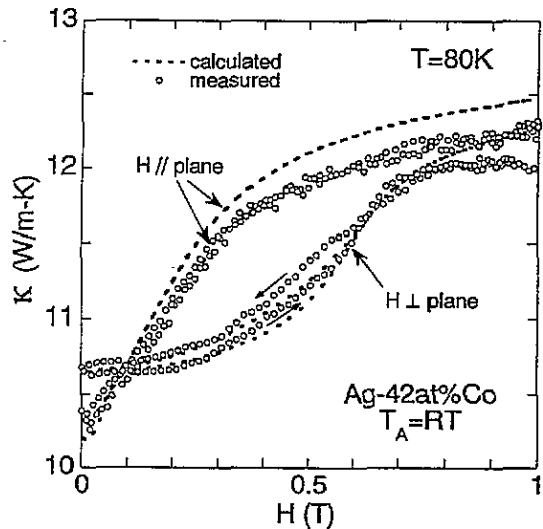


Figure 3. Field dependence of thermal conductivity  $\kappa(H)$  for the 42 at.% Co-Ag sample at 80 K for the field both parallel and perpendicular to the plane. The calculated thermal conductivity  $\kappa(H)$  from  $\rho_{xx}(H)$  based on the WFL is also shown. The experimental curve is shifted vertically to coincide with the calculated curve at zero field.

the MR (figure 1(b)). The MR ratio for the field parallel to the film plane ( $H \parallel$  plane) falls off more rapidly than that for the field perpendicular to the plane ( $H \perp$  plane) and is also larger, at 6 K (272 K) reaching values of 33% (14%) for  $T_A = \text{RT}$  and 26% (9.5%) for  $T_A = 320^\circ\text{C}$ . Similar behaviours have recently been reported for vacuum-evaporated Co-Ag granular films and epitaxial Co-Ag alloy films [21, 22] although those anisotropies are not as great as in the present experiment. Such anisotropy is advantageous in the other transport properties for sorting out GMR-related effects from parasitic effects. We presume that the origin of this anisotropy lies in the formation of cluster networks as increasing the Co concentration causes magnetic particles to connect to adjacent particles. The concentration of 42 at.% is in the critical region where cluster networks with finite size coexist with smaller granules [15, 23, 24].

Figure 2 shows that the electrical resistivity  $\rho_{xx}(0 \text{ T})$  in zero field, exhibits an approximate low-temperature  $\ln T$  dependence on  $T$  for both the non-annealed and the  $T_A = 310^\circ\text{C}$  samples, with a resistance minimum in the vicinity of 20 K for the non-annealed sample and near 10 K for  $T_A = 310^\circ\text{C}$ . The increase in  $\rho_{xx}(0)$  below the minimum is eight times that for the non-annealed sample, while the minimum resistivity ( $26.0 \mu\Omega \text{ cm}$ ) for the non-annealed sample is only 2.4 times that ( $10.7 \mu\Omega \text{ cm}$ ) for  $T_A = 310^\circ\text{C}$ . Similar resistance minima have been reported for Au/Fe, Co/Cu and Fe/Cr multilayers [25–27].

From the low absolute values of the residual resistivity, the minima are not due to

weak-localization effects. We previously explained [25] the temperature-dependent electrical resistivity of Au/Fe superlattices in terms of a model related to the Kondo effect associated with loose spins proposed by Slonczewski [28]. However, while the Kondo temperature  $T_K$  for Co–Ag is not known, the  $T_K$  plot for noble metals given by Daybell and Steyert [29] suggests that  $T_K$  for Co–Ag will be above 100 K, making it unlikely that the present result is due to an ordinary dilute Kondo effect. Hasegawa [30] recently reported a theoretical investigation of GMR in magnetic multilayers at finite temperatures that gave  $\rho_{xx}(0\text{ T})$  decreasing with increasing temperature in the limit of low sample purity, and Saito *et al* [26] explained their observation of a low-temperature decrease in resistivity with increasing temperature in Co/Cu multilayers as due to the decreasing spin-dependent scattering responsible for the GMR overwhelming an increasing spin fluctuation scattering. Whether this explanation can describe our granular data will require further theoretical analysis.

Since Piraux *et al* [19] have reported an extensive investigation of both the absolute thermal conductivity and the magnetothermal conductivity on Co–Ag (containing 20 vol.% Co  $\approx$  27.2 at.% Co) granular alloys, we only briefly describe our experimental results for the magnetothermal conductivity of our 42 at.% Co alloy. Figure 3 shows an example of the field-dependent part  $\Delta\kappa(H)$  of the thermal conductivity compared with the curve calculated from the Wiedemann–Franz law (WFL):

$$\kappa(H) = L_0 T / \rho_{xx}(H) \quad (1)$$

using the  $\rho_{xx}(H)$  data measured on the same sample and  $L_0 = 2.44 \times 10^{-8} \text{ W } \Omega \text{ K}^{-2}$ . Since we measured only the field-dependent part of  $\kappa(H)$  with sufficient accuracy, the experimental curves have been shifted vertically to coincide with the theoretical curves at  $H = 0$ . In agreement with the results of Piraux *et al* for 27.2 at.% Co–Ag, the field dependences of our  $\Delta\kappa(H)$  and  $\Delta\kappa(H)_{\text{WFL}}$ -values for 42 at.% Co–Ag lie within our experimental accuracy of 10% at both 10 and 80 K including the anisotropy. The conduction electron scattering responsible for GMR in 42 at.% Co–Ag granular alloys is thus primarily elastic in nature, as previously found for more dilute Co–Ag and for magnetic multilayers [10, 19].

The TEP for Co–Ag granular alloys has been measured by Shi *et al* [18] (20 at.% Co) [18] and Piraux *et al* [19] (27.2 at.% Co). While their data roughly agreed at high temperatures, they reported different temperature dependences at low temperatures. Shi *et al* found that the TEP changed sign from negative at high temperatures to positive below about 25 K. Piraux *et al*, in contrast, observed no sign change, but found a small negative peak near 70 K. Figure 4 shows our results for 27.2 at.% Co–Ag samples non-annealed and  $T_A = 310^\circ\text{C}$ , at both 0 and 1 T, and for the non-annealed 42 at.% Co–Ag sample at 0 T. Overall, our results are similar to those of Piraux *et al*—especially in that we do not see a low-temperature sign change—except that we do not see negative peaks in the vicinity of 70 K. We conclude that neither the negative-to-positive sign change at low temperatures nor the negative peaks in the vicinity of 70 K are intrinsic to Co–Ag alloys. What do seem to be intrinsic at zero-field are

(a) a linear component of the TEP, with a magnitude that decreases slightly with increasing annealing temperatures (from about  $0.029 \rightarrow 0.020 \mu\text{V K}^{-2}$  from non-annealed to  $T_A = 605^\circ\text{C}$  specimens for the data of Piraux *et al* and from about  $0.020 \rightarrow 0.017 \mu\text{V K}^{-2}$  from non-annealed to  $T_A = 310^\circ\text{C}$  specimens for our data) and

(b) a non-linear component that rises rapidly with increasing  $T$  at low temperatures and then roughly saturates at high temperatures, and that decreases in magnitude with increasing annealing temperature (*the component almost disappears for  $T_A = 310^\circ\text{C}$* ).

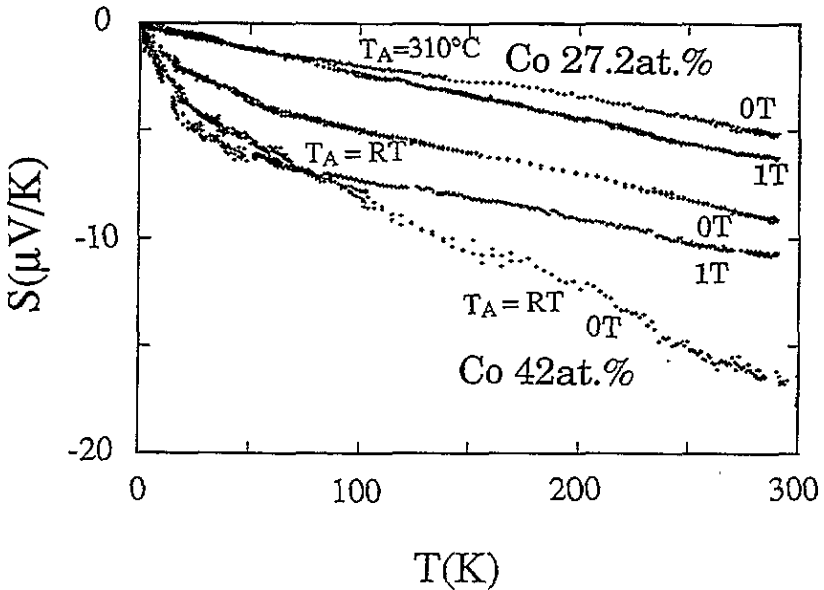


Figure 4. The temperature dependences of TEP for 27.2 at.% Co-Ag for non-annealed ( $T_A = RT$ ) and  $T_A = 310^\circ\text{C}$  both at zero field and at 1 T and for non-annealed 42 at.% Co-Ag at zero field.

The behaviour of the data for our non-annealed 42 at.% Co-Ag sample is similar in form, but with a larger linear component (about  $0.046 \mu\text{V K}^{-2}$ ). Whether a decomposition into two such independent components appropriate is not yet clear.

When a field is applied, all three studies on Co-Ag agree that  $S$  becomes more negative and retains roughly the same shape, and we agree with Piraux *et al* that the magnitude of the change with field decreases with increasing annealing temperature. We want to conclude with one quantitative point not made by Piraux *et al* that will have to be taken into account in interpreting the change with annealing of both the TEP data in figure 4 and the  $\rho_{xx}$  data in figure 2, namely that the form of the field-dependent magnetization changes with annealing. As illustrated in figure 5, we find  $M(H)$  is Langevin like for the non-annealed sample but gradually changes to a more ferromagnetic form by  $T_A = 310^\circ\text{C}$ , where it fully saturates above 1 T. Clearly the magnetic microstructure of the sample is changing upon annealing. Since Piraux *et al* have made a complete analysis of the field dependence  $S(H)$  for 27 at.% Co-Ag, we report here only that the anisotropy found in the MR for 42 at.% Co-Ag was clearly observed also in the field dependence of the TEP.

The Hall resistivity  $\rho_{xy}$  is usually described as the sum of a normal part  $\rho_{xy}^N$  proportional to the magnetic field  $H$  and an extraordinary part  $\rho_{xy}^M$  proportional to the magnetization  $M$ :

$$\rho_{xy} = R_O H + R_S M \quad (2)$$

where  $R_O$  and  $R_S$  are the normal and extraordinary Hall coefficients, respectively.  $R_S$  consists of two asymmetric scattering mechanisms, skew scattering and side jump, and has the form [31]

$$R_S = a\rho_{xx} + b\rho_{xx}^2 \quad (3)$$

where the parameters  $a$  and  $b$  determine the sizes of the skew scattering and the side-jump components, respectively.

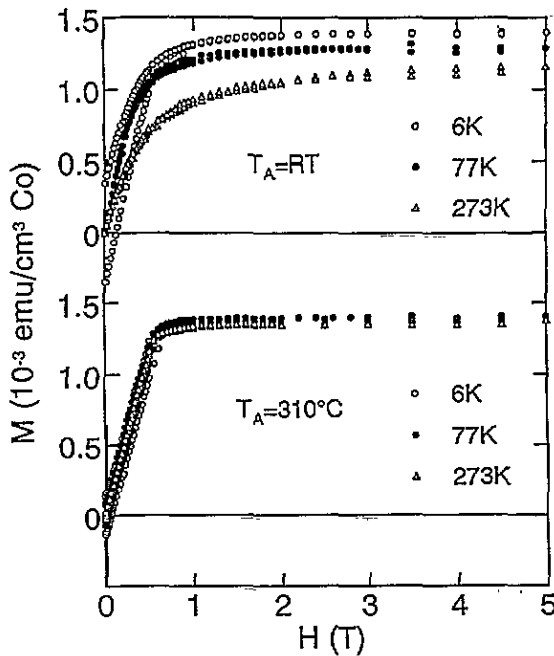


Figure 5. The field dependence of the magnetization at 6, 77 and 273 K for 27.2 at.% Co-Ag (non-annealed and  $T_A = 310^\circ\text{C}$ ).

In most experiments on ordinary ferromagnetic films,  $\rho_{xy}(H)$  has been described fairly well by equation (2) assuming a field-independent  $R_S$ . Then  $\rho_{xy}^M(H)$  mimics the field dependence of  $M$ . However, in recent experiments on Fe/Cr [8,9] and Co/Cu [11] multilayers, an anomalous peak, due to the field dependence of  $R_S$ , was found in  $\rho_{xy}^M(H)$ . It was thus somewhat surprising when Xiong *et al* [16] reported that  $\rho_{xy}^M(H)$  for a 27.2 at.% Co-Ag granular alloy that exhibited GMR could be described in terms of a field-independent  $R_S$ . Taking  $\rho_{xy}^M(H) \propto M$ , they derived an unusual scaling relation  $\rho_{xy}^M(8\text{ T}) \propto \rho_{xx}(0\text{ T})^{3.7}$  as illustrated by the full circles in figure 6. In attempting to repeat their results, in figure 7, we show  $\rho_{xy}^M(H)$  calculated from equation (2) using  $\rho_{xy}(H)$  and  $R_0$  determined from the linear slope at high fields, and only after the sample had first been taken to saturation. Except for  $T_A = \text{RT}$ , it is immediately clear that  $\rho_{xy}^M(H)$  differs qualitatively from  $M(H)$  (see figure 5). A similar peaked structure to those in figure 7 for  $T_A > 200^\circ\text{C}$  has been seen previously for magnetic multilayers [8,9,11]. If the MR is small, so that  $\rho_{xx}(H) \approx \rho_{xx}(0)$ , then  $R_S$  is independent of  $H$ . If, however, there is a GMR, as in the present Co-Ag granular samples, then equation (3) produces the field dependence in  $R_S$  even for constant  $a$  and  $b$ . If  $a$  and  $b$  have the same sign, then equation (2) naturally gives a peak in  $\rho_{xy}^M(H)$  for a GMR system, since  $\rho_{xx}$  decreases with increasing field, whereas  $M$  increases. It can, of course, also give a peak if they have opposite signs, but with more complex behaviour. Re-examining their data, we also find evidence for such a peak, and thus a field-dependent  $R_S$ , in all the data in figure 1 of [16] for  $T_A > 200^\circ\text{C}$ . On the other hand, if we plot the high-field saturation limits of our values of  $\rho_{xy}^M(H)$  as the open and full squares in figure 6, we find that they differ only modestly from the data of Xiong *et al* (open circles) and also evidence a rapid variation with  $\rho_{xx}$ .

Before discussing the variation in  $R_S$  with annealing temperature (figure 6), we try to

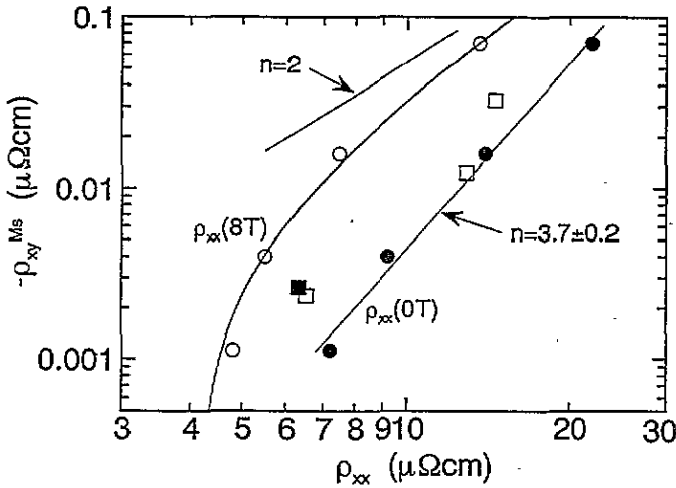


Figure 6.  $\ln(\rho_{xy}^M)$  at 8 T versus  $\ln \rho_{xx}$  at 0 T plot by Xiong *et al* [16] (●) along with a new plot (O) versus  $\ln \rho_{xx}$  at 8 T. The present data (□, ■) both  $\rho_{xy}^M$  and  $\rho_{xx}$  measured at 5 T for 27.2 at.% Co-Ag are also included. For the full square the sign is reversed. The full curve is the best fit using equation (3).

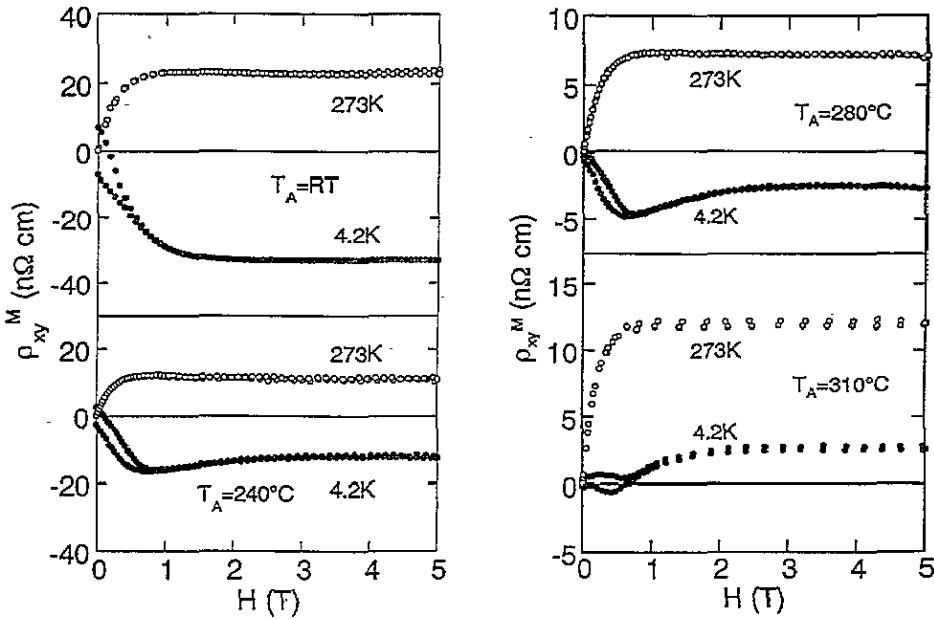


Figure 7. Typical field dependences of the extraordinary Hall resistivity for 27.2 at.% Co-Ag samples at 273 and 4.2 K.

explain the dependences of  $R_S$  in equation (2) on field strength and measuring temperature. To see whether the field dependence of our present results (figure 6) can be explained by equation (3), we plotted  $R_S(H)/\rho_{xx}(H)$  versus  $\rho_{xx}(H)$  [20]. For Fe/Cr and Co/Cu multilayers, such a plot was found to be roughly linear, indicating that the parameters  $a$



and  $b$  were independent of magnetic field. This independence was taken as evidence that the mechanism with the same ratio of the skew to the side-jump components (probably the same scattering centres) dominates conduction electron scattering in both the low-field high-resistance state and the high-field low-resistance state of these multilayers. In contrast, we see that the plots for Co–Ag samples are not linear. In addition, both the form and the magnitude of the curves depend delicately on the annealing temperature. If we stick with equation (3), we must conclude from the lack of linearity of most of our data that the conduction electron scattering in our granular Co–Ag samples is not dominated by only a single mechanism, but that at least two different mechanisms must be present. As a simple example of such a possibility, we might have scattering near the interfaces between relatively large granules and the Ag matrix dominant at low fields, but scattering from localized Co spins or smaller Co clusters dominating at high fields.

Next, to estimate reasonable values for  $a$  and  $b$  of our granular Co–Ag samples at high fields, we have plotted  $R_S(T)/\rho_{xx}(T)$  versus  $\rho_{xx}(T)$  at higher temperatures on each sample at 5 T, which is the simplest case to be compared with the theory since the dominant scattering mechanism may not change with  $\rho_{xx}$  [32]. We found opposite signs for  $a$  and  $b$ ;  $b \approx 7 \times 10^4 \Omega^{-1} \text{ m}^{-1} \text{ T}^{-1}$  nearly independent of both Co concentration and  $T_A$ , while  $a$  varies from  $-12$  to  $-3 \times 10^{-3} \text{ T}^{-1}$  depending on the condition.

Finally, we discuss the dependence of  $R_S$  on  $T_A$ , reported by Xiong *et al* using the plot in figure 6. To fit the  $\rho_{xy}^M(8 \text{ T})$  data (open circles) plotted against  $\rho_{xx}(8 \text{ T})$  with equation (3), we now need a positive  $a$  and a negative  $b$ . The best fit, shown as the broken line, is for  $a \approx 7.3 \times 10^{-3} \text{ T}^{-1}$ , and  $b \approx -1.7 \times 10^5 \Omega^{-1} \text{ m}^{-1} \text{ T}^{-1}$ . Our data (open and full squares) for  $\rho_{xy}^M(5 \text{ T})$  versus  $\rho_{xx}(5 \text{ T})$ , which are shifted horizontally compared with the data of Xiong *et al*, can also be fitted similarly with  $a \approx 4.7 \times 10^{-3} \text{ T}^{-1}$ , and  $b \approx -7.6 \times 10^4 \Omega^{-1} \text{ m}^{-1} \text{ T}^{-1}$ , if we take into account the scatter in the data points. We note that opposite signs for  $a$  and  $b$  provide a mechanism for the higher law than 2 reported by Xiong *et al* and illustrated in figure 6; a rapid change in  $\ln R_S$  versus  $\ln \rho_{xx}$  appears in the limited range of  $\rho_{xx}$  near  $a + b\rho_{xx} = 0$ . Whereas  $a$  and  $b$  of the same sign cannot give a variation in  $\rho_{xy}^M(8 \text{ T})$  versus  $\rho_{xx}(8 \text{ T})$  faster than  $\rho_{xx}^2$ . Figure 6 shows that  $a$  and  $b$  of opposite signs can give an apparently higher power, including the change from negative values of  $R_S$  to a positive value for a sufficiently high  $T_A$ . However, there is an inconsistency in the signs of these values of  $a$  and  $b$  with those estimated from measuring the temperature dependence. Taking into account the result for the field dependence experiment, we infer that more than two scattering mechanisms are competing in granular alloys, and  $a$  and/or  $b$  vary with the annealing temperature. Of course, we cannot rule out the possibility that the scaling relation (equation (3)) has already broken down for granular alloys. Very recently, Zhang [32] tried to calculate the extraordinary Hall effect in magnetic multilayers and found that the ordinary scaling relation for the side-jump mechanism should not be valid. This lack of validity might be one of the sources of our troubles in trying to fit our granular Co–Ag data.

To summarize, with a few minor exceptions, our MR,  $\kappa(H)$ , TEP and Hall data on electron-gun-prepared 27.2 at.% Co–Ag granular alloys agree with data on similar samples prepared by sputtering. We confirm large isotropic MRs in 27.2 at.% Co–Ag granular alloys and report large anisotropic MRs in 42 at.% Co–Ag granular alloys. We find a resistivity minimum, which cannot be explained as either an ordinary Kondo effect or due to weak localization, and which is found to depend upon annealing temperature. We confirm, with anisotropy for 42 at.% Co–Ag, previous evidence from thermal conductivity measurements that the scattering responsible for the GMR effect in Co–Ag is elastic below 80 K. We find an anomalous peaked structure in the extraordinary Hall resistivity, which is not well described

by existing theories and probably indicates that more than two scattering mechanisms are competing in granular alloys.

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