PHYSICAL REVIEW B VOLUME 15, NUMBER 2 15 JANUARY 1977

Anomalous Hall effect in *Au*Fe alloys

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(Received 3 May 1976)

*Au*Fe shows a large anomalous Hall effect that is dominated by a skew contribution arising from the asymmetric scattering of the itinerant electrons. We have measured this effect in alloys ranging from the dilute limit up to concentrations for which *d-d* overlap is the dominant intermoment coupling (Au + 211 at. ppm Fe to Au + 8.64-at.% Fe). We show that the qualitative behavior of the skew component does not change over this wide concentration range, and does not correlate with the magnetization. The results suggest a cause more complicated than one involving isolated scattering by ions coupled through the Ruderman-Kittel-Kasuya-Yosida interaction.

I. INTRODUCTION

This paper gives a qualitative description and interpretation of the skew component of the anomalous Hall effect observed in polycrystalline *Au*Fe alloys. An anomalous component of the Hall effect is one which, from its variation with magnetic field strength or temperature, evidently does not arise from the classical Lorentz force. The skew component[1,2] is one of at least two distinctly different anomalous parts of the Hall effect that can be seen in Kondo alloys consisting of a magnetic solute dissolved in a noble-metal host; the second is the spin component.[2]

The *skew component* arises from the spin-orbit coupling between the localized moment's spin and the conduction electron's angular momentum during the electron's temporary residence in the ion's virtual state.[2-5] The result is an elastic scattering event that is asymmetric (or skew) with respect to the plane containing the ion's moment and the electron's incident velocity.[3-7] The skew component is typically evident even at the lowest field strengths and tends to saturate in the Hall resistivity (ρ_H) when the field strength is increased sufficiently; it often increases with decreasing temperature and is approximately proportional to the solute's concentration.[3-5,8,9]

The second anomalous component is the *spin component*.[1,4,5] It is associated with the inelastic spin-flip scattering of the conduction electrons by the localized moments. As the applied field strength is increased and the magnetic ion's Zeeman levels become both increasingly separated in energy and nonuniformly populated, so the total amount of this scattering falls.[10,11] This produces a field-dependent difference between the relaxation times of spin-up and spin-down electrons. The result is a negative magnetoresistance and a field-dependent contribution to the Hall effect.[12,13] Here we are concerned with the behavior of the skew component; the spin component, which appears only incidentally in the following, will be the subject of a later publication.

We wish to extend, to higher concentrations and applied fields, the scope of a previous study of this system[8] which first showed the importance of the skew contribution in *Au*Fe. The aim is to observe the behavior of the skew component as the circumstances pass between the extremes of the dilute limit, where the intermoment coupling is presumably dominated by Ruderman-Kittel-Kasuya-Yosida (RKKY) coupling, to a concentration range where the coupling must be dominated by *d-d* overlap exchange forces. We have measured the Hall resistivity in alloys ranging from Au + 211 at. ppm to Au + 8.64-at.% Fe in the range of fields up to 7 T and temperatures between 1.5 and 80 K.

II. EXPERIMENTAL DETAILS

Polycrystalline ingots of about 1-cm diam. were prepared in a high-vacuum induction furnace as described previously.[8] The starting materials were Au of 99.9999-at.% purity[14] and Fe of approximately[15] 99.99-at.% purity. After chill casting twice to improve homogeneity, the ingots were cold rolled to sheets about 9×10^{-4} m thick. Parts from the top and bottom of each sample were analyzed, using spectrographic and atomic absorption methods, with the mean results quoted in the figures. As a check on each sample's metallurgical state, we plotted its measured electrical resistivity at 4.2 K against the measured solute concentration. In all cases the results were in good agreement with the value of 79.8 nΩ m/at.% determined earlier.[16]

The samples were spark cut from the rolled sheets using a template with the shape described previously.[17] With the exception of the most con-

centrated sample, their chemical cleaning was followed by annealing at 550 °C for 24 h in a Pyrex container which was air cooled to ambient temperature before opening. The Au + 8.64-at.% Fe sample was annealed in vacuum at 900 °C before rapid quenching into iced brine, and was maintained at 77 K until measured.

A standard cryostat-superconducting magnet combination was used together with dc potentiometric instrumentation having a sensitivity of 1×10^{-9} V. The sample could be flipped rapidly through 180° about its transverse axis so that, without reversing the magnet current, the Hall voltage could be determined from the mean of the results obtained from the four permutations of applied electric and magnetic field directions. In the case of the more concentrated samples, where the experiment could encompass the spin-glass regime, care was taken always to cool only in zero applied field. The accuracy of any Hall resistivity datum cited in the following figures is dominated by the error in the determination of the sample's thickness. This quantity was measured to within about 1.5×10^{-5} m, leading to an overall uncertainty of about ±1.6% in the Hall resistivity ρ_H.

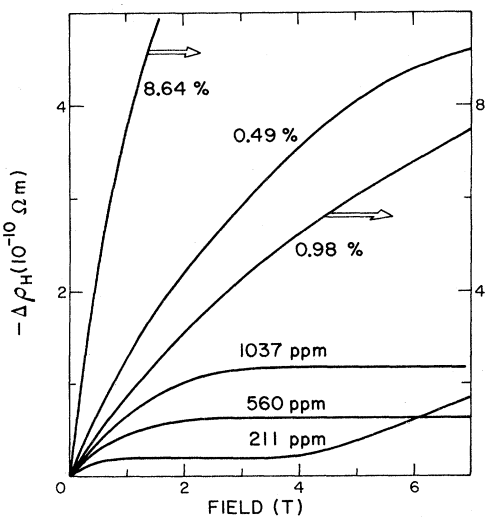

FIG. 1. Comparison of the anomalous part of the Hall resistivity ($\Delta\rho_H$) seen at 4.2 K in the alloys indicated. (Note that the curves for the four most dilute cases refer to the left-hand ordinate.) Similar results for still more dilute alloys appear in Fig. 6 of Ref. 8. Here and in Fig. 2 the data points are omitted for clarity; they are spaced by 0.25 T at the lower field strengths and by 0.5 T at the higher.

III. DISCUSSION OF RESULTS

A. Isothermal field dependence

The anomalous component of the Hall effect originates from the localized moments in the alloy, thus its extraction from the total effect requires a knowledge of the behavior of a fictitious alloy for which the solute's moment has been eliminated. Our pragmatic solution[8] is to use the classical Lorentz part of $\rho_H(B)$ expected for the pure solvent metal, since this is the behavior of an alloy whose concentration is reduced to the point where no anomalous component is evident. We have thus subtracted the classical Lorentz component for polycrystalline Au from our results to produce the field dependences of the skew component $\Delta\rho_H(B)$ shown in Fig. 1 for six alloys at 4.2 K.[18] The most dilute sample (Au + 211 at. ppm Fe) has previously been studied up to 1.5 T and provides a link to the comparable behavior seen in the more dilute samples of Ref. 8.

For the three more dilute samples the skew component saturates in the fields available. However, for the concentrated ones the anomalous component is not saturated even at 7 T. A qualitative comparison of the saturation behavior of the skew component (Fig. 1) with that of the magnetization in equivalent circumstances shows that the former saturates in lower fields than the latter. For example, in the Au + 1037-at. ppm Fe alloy, the skew component at 4.2 K is saturated above 3 T, whereas the magnetization of a 0.1-at.% alloy[19] showed no sign of saturation even at 3.3 T. Thus there seems to be no direct correlation between the skew component of the Hall resistivity and the magnetization for this system. The estimated saturation values of $\Delta\rho_H(B)$ from Fig. 1 and Ref. 8 are found to vary roughly linearly with solute concentration, although the $\Delta\rho_H(B)$ curves show qualitatively different shapes for different concentrations. We find the curves of Fig. 1 do not scale to produce a universal curve[20] of $\Delta\rho_H/x$ vs B/x (x is the solute concentration), and it is unlikely that the $\Delta\rho_H(B)$ of Fig. 1 can be fitted to a universal Brillouin function as was done for the more dilute alloys.[8]

Even though the effective Bohr magneton value of Fe in Au (~3.7) is smaller than that of Mn in Au (~5.4) the skew component per at.% is largest for Fe. This can be seen, for example, from a comparison of $\Delta\rho_H(B)$ for an Au + 1022 at. ppm Mn sample (Fig. 2) with the results for a sample of approximately the same concentration of Fe (Fig. 1). The larger skew component arises from the stronger resonant coupling between the Fe virtual levels and the conduction electrons. This is also reflected in the resistivity per at.%, which is about 3 times larger for Fe in Au than for Mn in Au. In systems such as *Ag*Mn or *Cu*Mn the

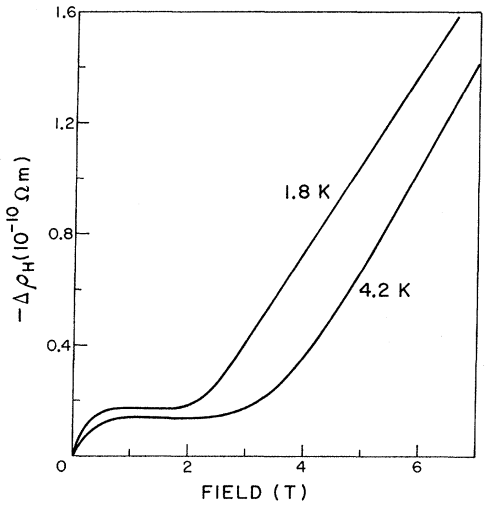

FIG. 2. Field dependence of the anomalous part of the Hall resistivity ($\Delta\rho_H$) obtained at the temperatures shown for a polycrystalline sample of Au + 1022 at. ppm Mn. It illustrates the behavior seen in a system having both a skew and a spin component.

resonant coupling can be so weak as to show very little or no skew component in ρ_H.[21]

The curves in Fig. 2 help to clarify the source of the upturn in $|\Delta\rho_H(B)|$ at fields greater than that needed to saturate the skew component. This upswing in $|\Delta\rho_H(B)|$ occurs at a field that varies significantly with temperature (Fig. 2). This indicates that kT is the determining parameter associated with the upturn rather than $\omega\tau$, which would be the characteristic parameter of a high-field–low-field transition, previously thought to be the explanation of this feature.[8] Evidence from the transverse magnetoresistance of[16] *Au*Fe suggests that the high-field–low-field transition would occur at ~1.7 T for the Au + 211-at. ppm Fe, which is somewhat less than the ~4 T observed in Fig. 1. Furthermore the field at which the upturn becomes evident correlates with the approach to saturation of the negative magnetoresistance. The upturn in $|\Delta\rho_H(B)|$ at the higher fields is therefore identified as the appearance of the spin component, for which kT is the determining parameter.

B. Isomagnetic temperature dependence

Figure 3 shows the temperature dependence of ρ_H in fields chosen so that in each case the skew component (Fig. 1) is saturated at 4.2 K. For the two more concentrated cases the skew component is the dominant feature in ρ_H and the classical Lorentz component, which has not been subtracted from the data, can safely be neglected. Except for the Au + 0.98-at.% Fe sample, the chosen field strength in Fig. 3 is large enough to disrupt any vestiges of the spin-glass state.[21] The exception in Fig. 3 shows the spin-glass transition (T_{SG}) at about 8 K, and we have already described[21] the qualitative behavior that is seen in such a case. Briefly, in the temperature range above T_{SG}, where the thermal energy of the localized spins is sufficient to overcome the ordering influences such as the RKKY and the direct *d-d* overlap interactions, the spins are free to align in the applied field in the usual Curie fashion. The measured skew component depends on the balance between the applied field's ordering influence—which, by aligning the moments throughout the alloy, gives the additive combination of their microscopic skew scattering events[6,7]—and the disordering influence of the ion's thermal energy. Hence, above T_{SG}, the skew component *reduces* as the temperature is increased at a fixed field. Below T_{SG}, the majority of the localized spins are locked in the spin-glass arrangement. Only those having sufficient thermal energy to be unlocked from this state by the applied field can alter the skew component of ρ_H. Hence, below T_{SG}, the skew component *increases* as the temperature is increased. For the Au + 0.98-at.% Fe alloy, when the applied field strength is increased sufficiently to destroy the spin-glass state, $\rho_H(T)$ switches to the same qualitative behavior[21] as

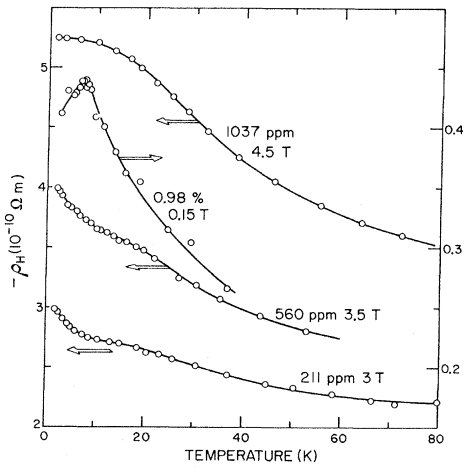

FIG. 3. Temperature dependence of the total Hall resistivity ρ_H obtained at the fields indicated. Although the classical Lorentz component has not been subtracted from the data shown, it is small compared with the anomalous component and can be safely ignored in this qualitative interpretation. In the case of the most concentrated sample, the applied field strength is reduced sufficiently so that the spin-glass state, which forms below $T_{SG} = 8$ K in this case, is not completely disrupted.

that seen for Au + 1037-at. ppm Fe in Fig. 3.

In the cases of the two most dilute alloys in Fig. 3, the Lorentz component of the measured ρ_H is no longer negligible (because the skew component is relatively small). Since this classical component has not been subtracted, its particular temperature dependence will be manifested in the $\rho_H(T)$, and two features seen in Fig. 3 are attributed to this. The first is the upturn in $|\rho_H(T)|$ seen below about 10 K, and the second is the slight local maximum in $|\rho_H(T)|$ centered at about 20 K.

The first feature is believed to be a manifestation of the shape of the Fermi surface of the matrix metal as the experiment passes into the dynamical high-field condition.[16] As this condition is approached, each crystallite makes a contribution to the Hall effect which depends[22] upon its crystallographic orientation with respect to $\vec{B}$. The net effect for a polycrystal is a complicated average behavior which leads to the upswing in $|\rho_H(T)|$ below about 10 K. The local maximum in $|\rho_H(T)|$ centered at about 20 K is observed more prominently as the concentration is reduced (Fig. 2 of Ref. 23), and is characteristic of $\rho_H(T)$ for pure Au. Much controversy has surrounded the appearance of this feature for the group $1B$ metals,[24] but Barnard's work[25] supports the view that the transition from phonon to impurity dominated regimes is implicated. Figure 3 shows that these two features, which are intrinsic to the host metal, can persist and manifest themselves in $\rho_H(T)$ of *Au*Fe alloys up to a concentration of a few hundred at. ppm.

IV. SUMMARY AND CONCLUSIONS

Our results confirm the indication[8] that the field dependence of the skew component of ρ_H in *Au*Fe alloys has features that are difficult to reconcile with the view that the effect arises simply from scattering by isolated ions. The skew effect in *Au*Fe is between 3 and 23 times bigger than the effect seen in analogous alloys in similar circumstances,[3] and $|\Delta\rho_H(B)|$ shows an approach to saturation in lower fields than the magnetization. If we try to match $\Delta\rho_H(B)$ for a given alloy to a Langevin or Brillouin behavior, the resulting equivalent moment deduced for the scattering center[8] is several times that possessed by an isolated Fe ion in Au.

There is a lot of evidence from previous magnetic and electron transport measurements in *Au*Fe to show that only below a concentration of about 25 at. ppm can the Fe ions be considered as isolated scattering centers (see Ref. 26 for a recent summary). Above that concentration, the presence of d-d coupled groups consisting of pairs, triplets, or perhaps larger numbers, is manifested in the transport properties. We suggest[8] that in this alloy system the skew component of ρ_H arises to a large extent from electron motion within such groups, which we shall call magnetic clusters.[27] The concept of a magnetic cluster, in which the direct d-d interaction is the intracluster coupling force that aligns the constituent's moments, has already been applied[28] to concentrated *Au*Fe alloys, and the term "cluster glass" has been suggested to cover the region (~10–16-at.% Fe) between the spin-glass regime and the percolation limit. Our suggestion is that in the Hall effect such clusters are probably important down to low concentrations. In such a model most of the skew component would arise from within the clusters, where the dominant intermoment force is always the d-d exchange interaction, whatever the Fe concentration. This explains why no qualitative change is seen in the behavior of $\Delta\rho_H(B)$ as the experiment passes from the dilute limit, where the RKKY interaction is dominant, to the higher concentrations where d-d exchange coupling is very significant. In Au–rare-earth alloys[5] the behavior of $\Delta\rho_H(B)$ is closer to the picture of isolated ionic spins coupled through RKKY forces. Direct f-f coupling of the ion's moments to form a cluster is unlikely because the magnetic f shells are buried deep within an ion's core.

A feature of our results, for which we have presently no sure explanation, is the lack of correlation in *Au*Fe alloys between the saturation of $\rho_H(B)$ and that of the magnetization (described in Sec. III). Even if the magnetic clusters of Fe ions are important as suggested, it is difficult to see why both the magnetization and the macroscopic skew components do not saturate in equivalent circumstances. This may be an artifact of the extraction of the skew component; our method has ignored the spin component of $\rho_H(B)$ which must exist in these alloys because they show a negative magnetoresistance. It is possible that the combination of the field dependences of the skew and spin components gives rise to the apparent saturation of the skew component before the magnetization.

ACKNOWLEDGMENTS

It is a pleasure to thank G. F. Turner for his careful sample preparation and general technical assistance, and the members of the Analytical Section for the work done on our behalf.

[1]In an earlier paper (Ref. 8) we did not differentiate between the two anomalous components; we referred to them collectively as the "spin contribution," on the grounds that their presence was evidently associated with the localized spins. The term "spin component" has since acquired the more specialized use described in the text.

[2]Communications by A. Fert, P. Monod, B. Giovannini, and C. M. Hurd, in *Proceedings of the Saint-Cergue Meeting on the Extraordinary Hall Effect*, edited by G. Cohen, B. Giovannini and D. Sorg (Université de Genève, Genève, 1973).

[3]A. Fert and O. Jaoul, Phys. Rev. Lett. 28, 303 (1972).

[4]A. Fert, J. Phys. F 3, 2126 (1973).

[5]A. Fert and A. Friederich, Phys. Rev. B 13, 397 (1976).

[6]C. M. Hurd, Contemp. Phys. 16, 517 (1975).

[7]C. M. Hurd, *Electrons in Metals: An Introduction to Modern Topics* (Wiley-Interscience, New York, 1975), p. 128.

[8]J. E. A. Alderson and C. M. Hurd, J. Phys. Chem. Solids 32, 2075 (1971).

[9]P. Monod and A. Friederich, *Proceedings of the Twelfth International Conference on Low Temperature Physics*, edited by E. Kanda (Keigachu, Tokyo, 1971), p. 755.

[10]M-T. Béal-Monod and R. A. Weiner, Phys. Rev. 170, 552 (1968).

[11]Reference 7, p. 127.

[12]R. M. More, Solid State Commun. 7, 237 (1969).

[13]M-T. Béal-Monod and R. A. Weiner, Phys. Rev. B 3, 3056 (1971).

[14]Supplied by Consolidated Mining and Smelting Co.

[15]Obtained privately from the Batelle Memorial Institute.

[16]C. M. Hurd and J. E. A. Alderson, Phys. Rev. B 6, 1894 (1972).

[17]J. E. A. Alderson, T. Farrell, and C. M. Hurd, Phys. Rev. 174, 729 (1968).

[18]This is open to criticism because it treats the elements of the magnetoresistivity tensor as additive. The approach that is correct in principle is to determine experimentally at any given field strength the separate magnetoresistivity tensors for the alloy and for the solvent in identical circumstances. These should be inverted, subtracted, and the result inverted again to give the magnetoresistivity elements appropriate to the anomalous effect. In the face of these complications, it seems that the simpler approach used is adequate, and has the advantage of showing at a glance the spin and skew content of the Hall resistivity.

[19]R. J. Borg and T. A. Kitchens, J. Phys. Chem. Solids 34, 1323 (1973).

[20]A similar lack of scaling, which has been attributed to the restraint produced by the strong concentration-dependent RKKY coupling, is also found for the magnetic susceptibility by J. C. Liu and F. W. Smith, Solid State Commun. 17, 595 (1975).

[21]S. P. McAlister and C. M. Hurd (unpublished); Solid State Commun. 19, 881 (1976).

[22]J. E. A. Alderson and C. M. Hurd, Phys. Rev. B 7, 1226 (1973).

[23]C. M. Hurd and J. E. A. Alderson, Phys. Rev. B 7, 1233 (1973).

[24]C. M. Hurd, *The Hall Effect in Metals and Alloys* (Plenum, New York, 1972), Chap. 4.

[25]R. D. Barnard, *Proceedings of the Fourteenth International Conference on Low Temperature Physics, Helsinki, 1975*, edited by M. Krusius and M. Vuorio (North-Holland–American Elsevier, Amsterdam, 1975), p. 71.

[26]J. A. Mydosh, P. J. Ford, M. P. Kawatra, and T. E. Whall, Phys. Rev. B 10, 2845 (1974).

[27]In common metallurgical use the term "cluster" has connotations of a large entity consisting of hundreds or perhaps thousands of members. By "magnetic cluster" we mean here any group of magnetic ions whose moments are coupled together by d-d exchange. As the concentration ranges from the dilute to the percolation limit, we envision an enormous variation in the mean cluster size. We are thus simply extending an existing picture of the *Au*Fe system (Ref. 28) to the case of much more dilute alloys.

[28]A. P. Murani, S. Roth, P. Radhakrishna, B. D. Rainford, B. R. Coles, K. Ibel, G. Goeltz, and F. Mezei, J. Phys. F 6, 425 (1976); and D. A. Smith, *ibid.* 5, 2148 (1975).