Positive Hall effect in paramagnetic amorphous Zr-Fe

M. Trudeau and R. W. Cochrane
Département de Physique, Université de Montréal, Case Postale 6128, Succursale A, Montréal, Québec, Canada H3C 3J7

D. V. Baxter, J. O. Strom-Olsen, and W. B. Muir
McGill University, Rutherford Physics Building, 3600 University Street, Montreal, Quebec, Canada H3A 2T8
(Received 23 March 1987; revised manuscript received 22 October 1987)

We present measurements and analysis of the Hall effect in amorphous paramagnetic Zr-Fe. From the correlation between the Hall coefficient and the valence susceptibility we show that the Hall effect is dominated by an anomalous contribution due to the side-jump effect, the first time such a contribution has been clearly identified in an amorphous paramagnetic system. We propose that the anomalous contribution accounts for the positive value of the Hall coefficient seen in many amorphous alloys.

I. INTRODUCTION

The positive Hall coefficient, R_H, seen in many amorphous metals remains a puzzle. According to the most widely used model of electron transport in noncrystalline metals, the Faber-Ziman theory, R_H should take its free-electron value $-1/n\,|e|$; and though there have been attempts[1] to extend the model to account for the positive sign, none have succeeded in describing the data quantitatively. An alternative approach, adopted by Cochrane and co-workers,[2] has been to invoke the anomalous Hall effect, specifically the so-called side-jump contribution. As explained by Berger[3] the effect results from the interaction between the conduction-electron spin **S** and the ionic orbital moment **L**. The resulting spin-orbit scattering displaces the center of mass of the electron wave packet sideways at each collision and leads to an additional contribution to the Hall conductivity.

In crystalline metals the anomalous Hall effect is usually seen in ordered magnetic systems,[4] where the large magnetization results in a strong contribution. In amorphous metals the contribution is enhanced by the large value of the electrical resistivity ρ. In the present article we provide evidence that in the paramagnetic amorphous alloys $Zr_{1-x}Fe_x$, close to the critical composition for ferromagnetism ($x \sim 0.4$), the Hall effect is in fact dominated by the anomalous contribution. These systems are therefore the first paramagnetic metallic glasses in which the anomalous Hall effect can be unequivocally identified.

II. EXPERIMENTAL TECHNIQUES

Samples of amorphous Zr-Fe were prepared by conventional melt-spinning techniques. Details of sample preparation and characteristics are given elsewhere.[5] Ribbons were typically 25 μm thick 2 mm wide. The Hall resistivity was measured in a field of up to 1 T using a high-resolution ac bridge,[6] modified to permit automatic balance control and data acquisition by microcomputer. The magnetic susceptibility was measured in a field up to 1.7 T by an automated alternating-force magnetometer under microcomputer control. Hall resistivity and magnetization data were taken for temperatures in the range 4.2–300 K, and subsequently analyzed for fields above 0.5 T. This choice was made to limit the influence of sample inhomogeneities which can affect the measurements at lower fields.[7]

III. RESULTS AND DISCUSSION

At Fe concentrations below 25 at.% both R_H and χ are temperature independent, but as the Fe content is increased the temperature variation grows rapidly, becoming very pronounced close to the critical composition for ferromagnetism ($\sim$38 at.% Fe). The relative variation of R_H is greater than that of χ in the sense that its proportionate change between room temperature and 4.2 K is greater but the general behavior of the two parameters is similar, as shown by Fig. 1. In fact both have the same functional dependence, namely of the form $A+B/(T+\Theta)$ with the possible exception of the 37.5 at.% Fe, which is right on the verge of ferromagnetism. The Curie-Weiss form of the susceptibility is expected on the basis of the theory of spin fluctuations in strongly enhanced paramagnets.[8] A detailed analysis of the magnetic behavior will be published elsewhere.[9]

To interpret the above data we assume that the Hall resistivity $\rho_H = R_H B$ is the result of two contributions: one the normal term, $R_0 B$, due to the Lorentz force and the other an extraordinary (or anomalous) term due to the side-jump ρ_H^{SJ}, so that

$$\rho_H = R_0 B + \rho_H^{SJ} \ . \tag{1}$$

As outlined above, ρ_H^{SJ} is usually only important in ferromagnets where it gives rise to a Hall resistivity proportional to the magnetization. However, the effect is not specific to ferromagnetism, but rather results from the

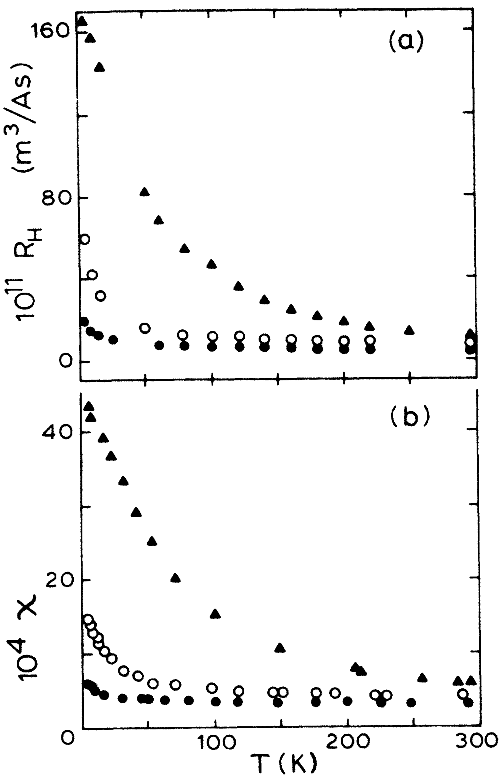

FIG. 1. Temperature dependence of the Hall coefficient (a), and magnetic susceptibility (Ref. 14) (b) of Fe_xZr_{1-x} for $x=33$ (●), $x=35$ (○), and $x=37.5$ (▲).

spin-orbit interaction between the charge carriers and the ions. Several workers[3,10,11] have shown that the interaction contributes a transverse conductivity σ_H^{SJ}, independent of the details of the scattering potential, given by

$$\sigma_H^{SJ}=\frac{\rho_H^{SJ}}{\rho^2}=\sum_i 2\frac{Ne^2}{\hbar k_F}\Delta y_i=\sum_i 2\left[\frac{\sigma_i}{\Lambda_i}\right]\Delta y_i \ , \tag{2}$$

where σ_i is the contribution to the zero-field conductivity of the carriers of type i and Λ_i their mean free path.[12] Δy_i is the magnitude of the side jump at each collision and is given by

$$\Delta y=\lambda_{s.o.}\langle S_z\rangle k_F \ . \tag{3}$$

$\langle S_z\rangle$ is the mean value of the electron spin along the field and $\lambda_{s.o.}$ is the effective spin-orbit interaction[10]

$$\lambda_{s.o.}=A_{s.o.}Id^2\sum_n\frac{|\langle 0|L|n\rangle|^2}{E_n-E_F} \ . \tag{4}$$

$A_{s.o.}$ is the atomic spin-orbit parameter and I the overlap integral (between the conduction-electron wave functions and the Zr-ion wave function). The form of Eq. (4) shows that $\lambda_{s.o.}$ is related to the Van Vleck susceptibility χ_{VV}, which is given by a double summation over all the occupied states:

$$\chi_{VV}=\sum_{m,n}|\langle L\rangle|^2/E_n-E_m, \quad E_m<E_n \ .$$

Although χ_{VV} is positive, the sign of $\lambda_{s.o.}$ and hence the side-jump contribution depends on the position of the Fermi level in the Zr d band. The less-than-half-filled band of Zr should result in a positive contribution to the Hall coefficient,[10] as required in the present case.

The factor σ_i/Λ_i in Eq. (2) implies that the contribution is independent of carrier mobility. Therefore, since E_F is known to be in the Zr d band,[13] Eq. (2) implies that the overwhelming contribution to σ_H^{SJ} comes from Zr d states. Moreover, the Van Vleck susceptibility of Zr is known to be unusually large,[14] suggesting that $\lambda_{s.o.}$ is significant for the Zr-rich alloys under investigation. Neglecting other contributions we combine Eqs. (2), (3), and (4) to give

$$\rho_H^{SJ}=2\frac{N_de^2}{h}\rho^2\lambda_{s.o.}\langle S_z\rangle \ . \tag{5}$$

In the present system $\langle S_z\rangle=\chi_v B/\mu_0N_dg\mu_B$ where χ_v is the relative valence susceptibility.[15] Thus

$$R_H=\frac{\rho_H}{B}=R_0+\frac{2e^2}{\mu_0\hbar\mu_Bg}\rho^2\lambda_{s.o.}\chi_v=R_0+R_s\chi_v \ . \tag{6}$$

The measured susceptibility, χ, may be written as $\chi=\chi_v+\chi_{core}+\chi_{VV}$ where both χ_{core} and χ_{VV} are temperature independent. Thus the temperature dependence of both R_H and χ come about through χ_v and a plot of R_H

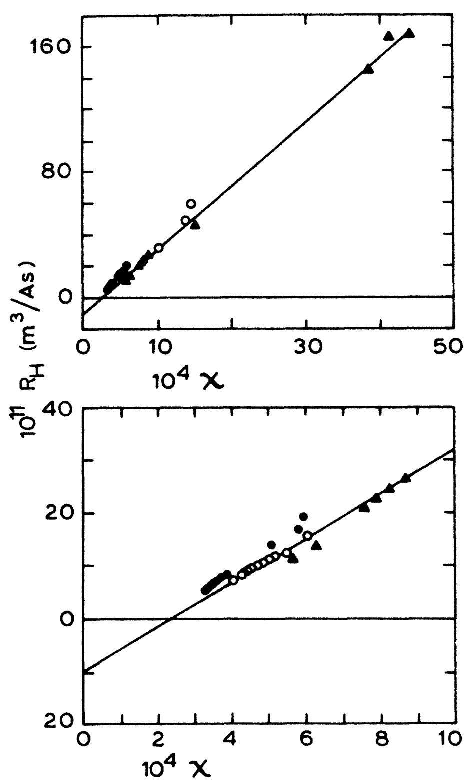

FIG. 2. Linear dependence of the Hall coefficient on the magnetic susceptibility for Fe_xZr_{1-x}. $x=33$ (●), $x=35$ (○), and $x=37.5$ (▲). The lower curve shows the lower left-hand portion of the upper curve in greater detail. The solid line is a least-squares fit to all the points and has a slope $R_s=4.3\pm0.1\times10^{-7}$ m^3/A s.

against χ should be linear. Furthermore, as these alloys span a narrow concentration range over which only the magnetic enhancement factor is changing, we should expect to find the *same straight line* for all three alloys. This striking result is shown in Fig. 2, where we note that the linear behavior is in fact followed over about two orders of magnitude in χ. From the slope in Fig. 2 we deduce $R_S = 4.3 \pm 0.1 \times 10^{-7}$ m^3/A s (Ω m/T).

As a point of comparison we estimate R_S directly from Eq. (6) using $\chi_{VV}/2\mu_0 N_{\rm Zr}\mu_B^2$ to approximate the matrix element in the expression (4) for $\lambda_{\rm s.o.}$. Taking as typical values, $A_{\rm s.o.} \sim 0.1$ eV, $I \sim 0.1$, $d \sim 0.32$ nm, $\chi_{VV} = 1.03 \times 10^{-4}$,[5] $\rho = 1.7 \mu\Omega$ m, and $N_{\rm Zr} = 3.5 \times 10^{28}$ m^{-3} we find $R_S = 1.2 \times 10^{-7}$ m^3/A s. Given the uncertainty of the various parameters we consider the agreement between the measured and estimated values to be a strong support for the proposed model.

We conclude with the proposal that the same anomalous contribution is responsible for the widespread observation of positive Hall coefficients in metallic glasses with open *d* bands. The important ingredients are a significant spin-orbit coupling, a large density of states, and a large resistivity. These criteria are met for the many Zr-, Ti-, Hf-, La-, and Y-based alloys where positive Hall coefficients are found. Of equal significance is the free-electron-like Hall coefficient of the Ca-Al (Ref. 16) alloys in spite of their high resistivities. Neither Ca nor Al possess the crucial spin-orbit coupling required to activate the side-jump mechanism.

[1]G. J. Morgan and G. F. Weir, Philos. Mag. B **47**, 177 (1983); R. Harris, J. Phys. F **16**, 53 (1986).

[2]R. W. Cochrane, J. Destry, R. Legault, and M. Trudeau, J. Appl. Phys. **55**, 1939 (1984); R. W. Cochrane, M. Trudeau, and J. O. Strom-Olsen, *ibid.* **57**, 3207 (1985).

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

[4]C. M. Hurd, *The Hall Effect in Metals and Alloys* (Plenum, New York, 1972), pp. 172–175.

[5]Z. Altounian, C. Volkert, and J. O. Strom-Olsen, J. Appl. Phys. **57**, 1777 (1982).

[6]R. W. Cochrane, B. J. Kastner, and W. B. Muir, J. Phys. E **15**, 425 (1982).

[7]R. W. Cochrane and M. Trudeau (unpublished).

[8]L. Hedman and Ö. Rapp, Phys. Lett. **100A**, 251 (1984).

[9]G. G. Lonzarich and L. Taillefer, J. Phys. C **18**, 4339 (1985); D. Baxter, J. O. Strom-Olsen, and R. W. Cochrane (unpublished).

[10]L. Berger and G. Bergmann, in *The Hall Effect and its Applications,* edited by C. L. Chien and C. R. Westgate (Plenum, New York, 1980), p. 55.

[11]P. Nozières and C. Lewiner, J. Phys. (Paris) **34**, 901 (1973).

[12]In addition to the side-jump effect, spin-orbit coupling contributes a further term to the anomalous Hall resistivity from skew scattering, which varies as ρ rather than ρ^2. We neglect this form here as the experimental evidence from crystalline and amorphous ferromagnets suggests that it is much smaller than the side-jump term. [See R. C. O'Handley, Phys. Rev. B **18**, 2577 (1978)].

[13]V. L. Morruzi, P. Oelhafen, A. R. Williams, R. Lapka, H.-J. Güntherodt, and J. Kubler, Phys. Rev. B **27**, 2049 (1983).

[14]H. J. Eifert, B. Elschiver, and K. H. J. Buschow, Phys. Rev. B **25**, 7441 (1982).

[15]All quantities are in SI units. In particular χ_v and χ_{VV} are the dimensionless susceptibilities defined by $B = \mu_0(H + M)$, $\chi = \partial M/\partial H$.

[16]U. Mizutani and T. Matsuda, J. Phys. F **13**, 2115 (1983); C. L. Tsai and F. Chu, J. Non-Cryst. Solids **61&62**, 1403 (1984).