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## Hall effect in Zr-Ni and Zr-Cu metallic glasses doped with hydrogen

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We have studied the effect of hydrogen doping on the Hall resistivity of paramagnetic  $Zr_{76}Ni_{24}$ ,  $Zr_{40}Ni_{60}$ ,  $Zr_{70}Cu_{30}$ , and  $Zr_{60}Cu_{40}$  metallic glasses. All the alloys have positive Hall coefficients except the  $Zr_{40}Ni_{60}$ . The hydrogen dopant increases the absolute value of the Hall coefficient irrespective of its sign, except in the case of  $(Zr_{70}Cu_{30})_{1-x}H_x$ , where the Hall coefficient decreases for low hydrogen concentrations ( $x < 0.04$ ) and increases at higher ones, while staying positive all the time. The results are explained as being due to the enhanced  $s$ - $d$  hybridization.

### INTRODUCTION

It is well known that in paramagnetic metallic glasses consisting of early and late transition metals the sign of the Hall coefficient,  $R_H$ , reverses from being negative to positive as the concentration of the early transition metal is increased.<sup>1-3</sup>

Weir *et al.*<sup>4</sup> have argued that a positive Hall coefficient in amorphous and liquid transition metals are the result of the anomalous dispersion of the free electrons due to  $s$ - $d$  hybridization, which leads to negative group velocities.

Shulte *et al.*<sup>5</sup> have shown that the sign of  $R_H$  can be related to hybridization between  $s$  and  $d$  electrons and to the finite spread of the electronic spectral function in the presence of disorder. The Hall constant becomes positive if the Fermi energy is located in the  $d$  band and becomes negative again at its edges.

Trudeau *et al.*<sup>6</sup> have invoked a side-jump mechanism, which is a result of the spin-orbit interaction, to explain a positive Hall effect in ferromagnetic as well as in paramagnetic Zr-3d metallic glasses, where this interaction is particularly strong due to the Zr  $d$  band being less than half-filled.

Measurements of the Hall coefficient in zirconium-rich  $(Zr-Ni)_{1-x}Al_x$  ternary compounds<sup>7,8</sup> have shown that  $R_H$  remains positive and becomes larger with increasing Al concentrations, even though the Zr concentration decreases to the value for which  $R_H$  in Zr-Ni alloys becomes negative. In the case of nickel-rich  $(Zr-Ni)_{1-x}Al_x$  alloys,  $R_H$ , being negative for  $x=0$ , diminishes and even becomes positive as  $x$  increases, reaching a sharp maximum for  $x=0.6$  and becoming negative again at  $x=0.7$ .

In this paper we report room-temperature measurements of the Hall coefficient for  $Zr_{76}Ni_{24}$ ,  $Zr_{40}Ni_{60}$ ,  $Zr_{70}Cu_{30}$ , and  $Zr_{60}Cu_{40}$  metallic glasses doped with hydrogen.

Our previous results<sup>9,10</sup> have shown a strong influence of hydrogen on the transport properties and electronic structure in Zr-Ni and Zr-Cu metallic glasses. For instance, the resistivity and the temperature coefficient of

the resistivity increase, the room-temperature spin susceptibility and, hence, the density of states at  $E_F$  decrease, and the spin-orbit interaction—which is strong in Zr-based alloys—decreases with increasing hydrogen concentration. Ultraviolet-photoelectron-spectroscopy<sup>11</sup> (UPS) measurements for  $(Zr-Ni)_{1-x}H_x$  metallic glasses show a change in  $s$ - $d$  hybridization upon hydrogenation. Thus, the use of hydrogen as a light atomic probe in the measurement of the Hall resistivity can give us, at least qualitatively, an insight into the roles the side-jump term and the  $s$ - $d$  hybridization play in the anomalous contribution to the Hall coefficient.

### EXPERIMENTAL TECHNIQUES

Amorphous ribbons were prepared by melt spinning on a single copper wheel in argon, and were doped with hydrogen using an electrolytic method.<sup>10</sup> The content of absorbed hydrogen was determined using a previously established relationship between the gain in resistance and the volumetrically determined hydrogen concentration. The samples cut from the ribbons were 2–2.5 cm long, 2.3–2.6 mm wide, and 20–30  $\mu$ m thick. The thickness of the samples was calculated from measurements of the density, mass, length, and width. The contacts were made either by spot-welding or with silver-conducting paint, and the results were reproducible within the accuracy of the method used.

The measurements of the Hall voltage,  $V_H(B)$ , were performed using a standard low-frequency (29-Hz) ac technique at 295 K and in a dc magnetic field between 0 and 2 T. The samples were fitted with two current and three voltage leads for the measurements of  $V_H(B)$  using the conventional technique of compensation of all voltages occurring at zero magnetic field. The current used was 33 mA.  $R_H$  was obtained from the mean value of the two field-direction slopes of  $V_H(B)$  in order to eliminate any magnetoresistivity signal. An absolute accuracy of 8%, due to the uncertainty in the effective sample thickness and contact position, and a relative resolution of about 5% were achieved.

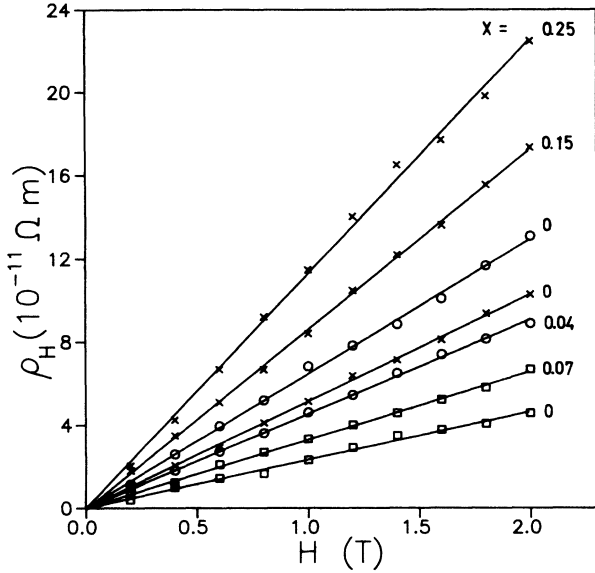


FIG. 1. Hall resistivity  $\rho_H$  vs applied magnetic field for  $(Zr_{76}Ni_{24})_{1-x}H_x$  ( $\square$ ),  $(Zr_{70}Cu_{30})_{1-x}H_x$  ( $\circ$ ), and  $(Zr_{60}Cu_{40})_{1-x}H_x$  ( $\times$ ) at 295 K.

## RESULTS

The Hall-resistivity data for  $(Zr_{76}Ni_{24})_{1-x}H_x$  ( $x=0,0.07$ ),  $(Zr_{70}Cu_{30})_{1-x}H_x$  ( $x=0,0.04$ ), and  $(Zr_{60}Cu_{40})_{1-x}H_x$  ( $x=0,0.15,0.25$ ) metallic glasses as a function of the magnetic field are plotted in Fig. 1. It can be seen that the Hall resistivity is linear in the applied field up to 2 T. In Fig. 2 the Hall-coefficient ( $R_H$ ) data for  $(Zr_{76}Ni_{24})_{1-x}H_x$  ( $x=0,0.07,0.09,0.35,0.44$ ),  $(Zr_{40}Ni_{60})_{1-x}H_x$  ( $x=0,0.07,0.15$ ),  $(Zr_{60}Cu_{40})_{1-x}H_x$  ( $x=0,0.024,0.15,0.25$ ), and  $(Zr_{70}Cu_{30})_{1-x}H_x$  ( $x=0,0.04,0.07,0.15,0.23,0.31$ ) are plotted against the hydrogen

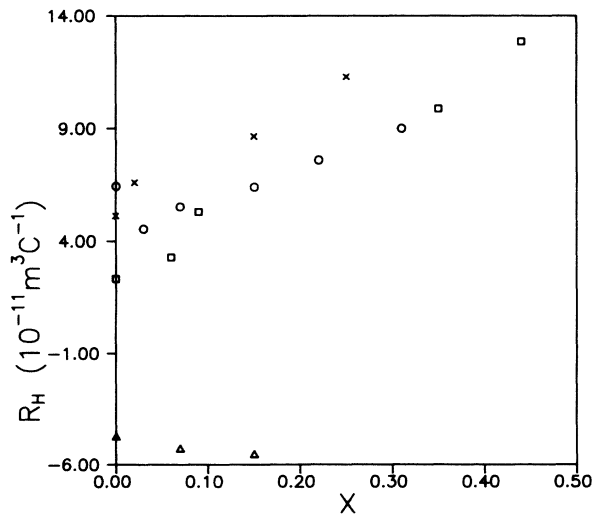


FIG. 2. Hall coefficient  $R_H$  vs hydrogen concentration  $x$  for  $(Zr_{76}Ni_{24})_{1-x}H_x$  ( $\square$ ),  $(Zr_{40}Ni_{60})_{1-x}H_x$  ( $\triangle$ ),  $(Zr_{60}Cu_{40})_{1-x}H_x$  ( $\times$ ), and  $(Zr_{70}Cu_{30})_{1-x}H_x$  ( $\circ$ ) at 295 K.

concentration. The Hall coefficient is positive for  $(Zr_{76}Ni_{24})_{1-x}H_x$ ,  $(Zr_{60}Cu_{40})_{1-x}H_x$ , and  $(Zr_{70}Cu_{30})_{1-x}H_x$ . It increases with the hydrogen concentration for  $(Zr_{76}Ni_{24})_{1-x}H_x$  and  $(Zr_{60}Cu_{40})_{1-x}H_x$ . In the case of  $(Zr_{70}Cu_{30})_{1-x}H_x$ , the Hall coefficient decreases for low hydrogen concentrations and a minimum occurs at  $x=0.04$ , after which it increases again.  $(Zr_{40}Ni_{60})_{1-x}H_x$  has a negative Hall coefficient, and its magnitude increases in the negative direction with increasing hydrogen concentration, but the enhancement is less than in the case of  $(Zr_{76}Ni_{24})_{1-x}H_x$ .

## DISCUSSION

The positive sign of the Hall coefficient cannot be explained by perturbative corrections to the Boltzmann conductivity assuming a free-electron density of states.

Mott<sup>12</sup> modified the nearly-free-electron (NFE) expression for the case of strong scattering:

$$R_H = R_{H,FE} \frac{N_{FE}(E_F)}{N(E_F)}, \quad (1)$$

where  $N(E_F)$  is the density of states at the Fermi level,  $R_{H,FE}$  is the NFE value of the Hall coefficient, and  $N_{FE}(E_F)$  is the NFE density of states.

Fukuyama *et al.*<sup>13</sup> have evaluated the Kubo-Greenwood formula for the NFE system and have obtained

$$R_H = R_{H,FE} \left[ \frac{N_{FE}(E_F)}{N(E_F)} \right]^2. \quad (2)$$

Thus,  $R_H$  calculated from Eqs. (1) or (2) is always negative.

Howson and Morgan<sup>14</sup> have calculated the Hall conductivity from a linear-response theory using an  $s$ - $d$  hybridization model similar to that described by Weir *et al.*<sup>4</sup> They showed that the positive Hall coefficient observed in many transition-metal-based amorphous alloys results from  $s$ - $d$  hybridization through the S-shaped dispersion curve. Hybridization can lead to a drastic modification of the  $s$ -electron dynamics and a reduction of the  $s$ -electron density of states at the Fermi energy in the vicinity of the  $d$  band. In their model the dispersed free-electron band plays a major role in the Hall conductivity, and the contribution of  $d$  electrons can be ignored, as opposed to their significant contribution in the ordinary conductivity.

Nguyen-Manh *et al.*<sup>15</sup> have used the linear-response formula derived by Howson and Morgan<sup>14</sup> to show that the Hall coefficient is proportional to the derivative of the density of states at the Fermi energy; it is given as

$$R_H = \frac{-\alpha}{2eN_s^2(E_F)} \left. \frac{dN_s(E)}{dE} \right|_{E=E_F}, \quad (3)$$

where  $\alpha$  is a constant in the range 1–3, and  $N_s(E)$  is the density of hybridized  $s$  states. The  $s$ - $d$  hybridization leads to negative values of  $dN_s(E)/dE$ , and thus a positive  $R_H$  when  $E_F$  lies in the lower half of the  $d$  band. They calculated  $R_H$  as a function of concentration  $x$  for  $Zr_{1-x}Cu_x$  amorphous alloys using electronic-structure calculations,

and obtained results that are in good agreement with experiment. The change of sign of the Hall coefficient from positive to negative occurs when the Fermi level located at the Zr 4d states moves to the edge of the Zr 4d resonance, thus changing the electronic structure at  $E_F$  from  $d$ -electron- to  $sp$ -electron-dominated states.

The other interpretation<sup>6,16</sup> of the positive Hall coefficient invokes the side-jump contribution that results<sup>17</sup> from the interaction between the conduction-electron spin and the ionic orbital moment. The resulting spin-orbit scattering displaces the center of mass of the electron wave packet sideways at each collision, leading to an additional contribution to the Hall conductivity of the form

$$R_H^{SJ} = \frac{2e^2}{\mu_0 h \mu_B g} \rho^2 \lambda_{s.o.} \chi_V, \quad (4)$$

where  $\chi_V$  is the valence susceptibility,  $\lambda_{s.o.}$  the effective spin-orbit interaction, and  $\rho$  the electrical resistivity. Since  $\rho$  is large in disordered systems, this effect is expected to be enhanced. ( $R_H^{SJ}$  is the side-jump term.)

The effective spin-orbit interaction is given by

$$\lambda_{s.o.} = \frac{A_{s.o.} I d^2 \chi_{VV}}{2\mu_0 \mu_B^2 N}, \quad (5)$$

where  $A_{s.o.}$  is the atomic spin-orbit parameter,  $I$  the overlap integral,  $d$  the interatomic distance,  $N$  the density of states, and  $\chi_{VV}$  the orbital (Van Vleck) susceptibility, which is large for partly filled degenerate bands (as is the case in Zr-based metallic glasses).

In order to interpret the measured data, we shall assume that the Hall resistivity,  $\rho_H = R_H B$ , is the result of two contributions: the first is the Lorentz term  $R_0 B$ , due to the Lorentz force and  $s$ - $d$  hybridization [Eq. (3)], and the second is the side-jump term [Eq. (4)].

The side-jump contribution can be estimated from Eq. (4) using Eq. (5) for calculating the effective spin-orbit interaction,  $\lambda_{s.o.}$ . Taking as typical values  $A_{s.o.} = 0.1$  eV,  $I = 0.2$ ,  $d = 3.45$  Å,  $\chi_{VV} = 1.03 \times 10^{-3}$  J T<sup>-2</sup> mol<sup>-1</sup>,  $\rho = 166 \times 10^{-8}$  Ω m,  $N_{Zr} = 3.5 \times 10^{28}$  m<sup>-3</sup>, and  $\chi_V = 0.73 \times 10^{-3}$  J T<sup>-2</sup> mol<sup>-1</sup>, we find  $R_H^{SJ} = 2.18 \times 10^{-11}$  m<sup>3</sup> C<sup>-1</sup> for the undoped sample. To compare this result with the measured value, one would have to know the effective number of conducting electrons in order to calculate the free-electron part of  $R_0$ . Nevertheless, the relative change of  $R_H$  with hydrogen concentration can be estimated.

We can use our previous results (Ref. 10), which show that the electrical resistivities of (Zr-Ni)<sub>1-x</sub>H<sub>x</sub> and (Zr-Cu)<sub>1-x</sub>H<sub>x</sub> metallic glasses increase with hydrogen concentration (see Table I). At the same time the valence magnetic susceptibility decreases (Ref. 10), as does the spin-orbit-scattering rate determined from our magneto-resistivity data (Ref. 10). Thus, for instance, for Zr<sub>67</sub>Ni<sub>33</sub>,  $\rho = 170 \times 10^{-8}$  Ω m,  $\tau_{s.o.} = 2.44 \times 10^{-12}$  s, and  $\chi_V = 73 \times 10^{-5}$  J T<sup>-2</sup> mol<sup>-1</sup>; for Zr<sub>67</sub>Ni<sub>33</sub>H<sub>33</sub>,  $\rho = 224 \times 10^{-8}$  Ω m,  $\tau_{s.o.} = 6.71 \times 10^{-12}$  s, and  $\chi_V = 50 \times 10^{-5}$  J T<sup>-2</sup> mol<sup>-1</sup>; and for Zr<sub>60</sub>Cu<sub>40</sub>,  $\rho = 169 \times 10^{-8}$  Ω m,  $\tau_{s.o.} = 1.04 \times 10^{-12}$  s, and

TABLE I. Sample characteristics for the metallic glasses studied.

Sample	$x$	$\rho$ ( $10^{-8}$ Ω m)	$R_H$ ( $10^{-11}$ m <sup>3</sup> /c)
(Zr <sub>60</sub> Cu <sub>40</sub> ) <sub>1-x</sub> H <sub>x</sub>	0	172	5.15
	0.024	176	6.64
	0.15	198	8.66
	0.25	214	11.29
(Zr <sub>70</sub> Cu <sub>30</sub> ) <sub>1-x</sub> H <sub>x</sub>	0	161	6.47
	0.04	167	4.55
	0.07	172	5.56
	0.15	185	6.43
	0.23	197	7.60
	0.31	211	9.00
(Zr <sub>76</sub> Ni <sub>24</sub> ) <sub>1-x</sub> H <sub>x</sub>	0	162	2.33
	0.07	167	3.29
	0.09	172	5.32
	0.35	218	9.88
	0.44	233	12.84
(Zr <sub>40</sub> Ni <sub>60</sub> ) <sub>1-x</sub> H <sub>x</sub>	0	174	-4.74
	0.07	186	-5.27
	0.15	200	-5.53

$\chi_V = 50.4 \times 10^{-5}$  J T<sup>-2</sup> mol<sup>-1</sup>; and for Zr<sub>60</sub>Cu<sub>40</sub>H<sub>11</sub>,  $\rho = 187 \times 10^{-8}$  Ω m,  $\tau_{s.o.} = 1.37 \times 10^{-12}$  s, and  $\chi_V = 33.4 \times 10^{-5}$  J T<sup>-2</sup> mol<sup>-1</sup>. If we use the above values to calculate the change of  $R_H$  upon hydrogenation, we may conclude that the side-jump term cannot explain the increase of the Hall coefficient for (Zr<sub>76</sub>Ni<sub>24</sub>)<sub>1-x</sub>H<sub>x</sub>, (Zr<sub>70</sub>Cu<sub>30</sub>)<sub>1-x</sub>H<sub>x</sub>, and (Zr<sub>60</sub>Cu<sub>40</sub>)<sub>1-x</sub>H<sub>x</sub> with increasing hydrogen concentration, because the increase of  $\rho^2$  in Eq. (4) is canceled by an equal decrease of  $\lambda_{s.o.}$  and  $\chi_V$ . In the case of low hydrogen concentration ( $x < 0.04$ ) in (Zr<sub>70</sub>Cu<sub>30</sub>)<sub>1-x</sub>H<sub>x</sub>, the decrease of the effective spin-orbit interaction and valence susceptibility is stronger than the increase of the resistivity; hence, in the case of weak hybridization, the value of the Hall coefficient can be expected to decrease, as is, in fact, observed.

This semiquantitative analysis shows that  $R_H^{SJ}$  stays constant with increasing hydrogen concentration, and the increase in the Hall coefficient has to be explained within the model of  $s$ - $d$  hybridization.

This is even more evident for (Zr<sub>40</sub>Ni<sub>60</sub>)<sub>1-x</sub>H<sub>x</sub> samples, which exhibit a negative Hall coefficient that increases in magnitude but remains negative with increasing hydrogen concentration. The increase of the side-jump term  $R_H^{SJ}$  that could explain the increase of the positive Hall coefficient would lead to a decrease in the magnitude of the negative Hall coefficient, which is contrary to what we observe.

It can also be argued that the part of the term coming from free electrons,  $R_0 = -1/ne$ , stays constant upon hydrogenation, because the increase in the number of conduction electrons [0.2 per H atom (Ref. 18)] is compensated by almost the same decrease in mass density, so that the concentration of conduction electrons stays constant.

In order to calculate the contribution of the hybridization between H 1s electrons and Zr 4d states to the Hall coefficient, one would need detailed electronic-state calculations of the  $(\text{Zr-Ni})_{1-x}\text{H}_x$  or  $(\text{Zr-Cu})_{1-x}\text{H}_x$  system, which are difficult to perform. Nevertheless, we can make some semiquantitative predictions of the variation of  $R_H$  with hydrogen concentration using the results for the variation of  $N_{s,d}(E_F)$  (Refs. 10 and 19) and  $dN_{s,d}/dE$  (Refs. 11 and 19) with  $x$ .

Ultraviolet-photoelectron-spectroscopy data<sup>20</sup> and the band-structure calculation<sup>21</sup> for Zr-Ni and Zr-Cu amorphous alloys show that the density of states at the Fermi level is dominated by the Zr  $d$  band, its peak lying somewhat above  $E_F$ , while the  $d$  band of the late transition metal is at higher binding energies. As the concentration of the late transition metal is increased, the peak at the higher binding energies is shifted closer to  $E_F$  and becomes more prominent, while  $N(E_F)$  decreases according to the reduced zirconium content in the system.

X-ray-photoelectron-spectroscopy (XPS) and UPS measurements<sup>10</sup> on hydrogenated  $\text{Zr}_{67}\text{Ni}_{33}$  and  $\text{Zr}_{36}\text{Ni}_{64}$  metallic glasses show a marked decrease in intensity close to  $E_F$  and the appearance of a subband at about 6.5 eV below  $E_F$ , both effects being proportional to the hydrogen concentration. This structure close to the bottom of the valence band is attributed to the formation of Zr 4d–H 1s bonding states. On the basis of these measurements, the estimated decrease in  $N_d(E_F)$  due to interaction with hydrogen is 12% for  $(\text{Zr}_{67}\text{Ni}_{33})\text{H}_{1.15}$  and  $(\text{Zr}_{36}\text{Ni}_{64})\text{H}_{0.34}$ , which is in good agreement with a decrease of 10% in the  $(\text{Zr}_{67}\text{Ni}_{33})\text{H}_{1.15}$  obtained from the low-temperature specific-heat<sup>19</sup> and a magnetic-susceptibility<sup>10</sup> measurements. At the same time the  $s$ - $d$  hybridization is stronger

in  $(\text{Zr}_{67}\text{Ni}_{33})\text{H}_{1.15}$  than in  $(\text{Zr}_{36}\text{Ni}_{64})\text{H}_{0.34}$ .<sup>10</sup> The emission spectra of Ni show no significant changes when hydrogen is added, and there is no sign of Ni–H bond formation. The effect of hydrogen on  $N_s(E_F)$  is—at present—not entirely clear. A small increase in  $s$ -electron population due to the hydrogen  $s$  electron is expected to be more than offset by increased  $s$ - $s$  as well as  $s$ - $d$  interaction. A decrease in  $N_s(E_F)$  is, however, consistent with our data, since it leads to an increase in magnitude of  $R_H$  [Eq. (3)], whether it is positive or negative.

The derivative of the  $s$  density of states  $dN_s/dE$  that enters Eq. (4) depends on the  $s$ - $d$  hybridization. Thus, a stronger hybridization would lead to a stronger increase of  $R_H$ , as is observed in Zr-rich alloys as compared to the Zr-poor ones (Fig. 2).

This explanation of the variation of  $R_H$  with hydrogen concentration as being due to the  $s$ - $d$  hybridization is also consistent with the concept of Shulte *et al.*<sup>5</sup>

In conclusion, we have found that the addition of hydrogen to the Zr-Ni and Zr-Cu metallic glasses greatly increases the magnitude of  $R_H$  regardless of its sign.

Using measured values for  $\rho$ ,  $\chi_V$ , and  $\lambda_{s.o.}$  or  $\chi_{VV}$  (Ref. 10), we have calculated the side-jump contribution to  $R_H$  for various hydrogen concentrations and have found that it does not change much upon hydrogenation. Thus, interpretation of the present data along these lines is not possible. Moreover, the increase of the side-jump term, which could explain an increase of the positive  $R_H$ , would lead to a decrease of the negative  $R_H$ , contrary to what is observed.

We conclude, therefore, that the observed behavior of  $R_H$  can be explained in terms of an increased hybridization of hydrogen  $s$  electrons with the Zr  $d$  band.<sup>10,11</sup>

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