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## Weak-localization and Coulomb-interaction effects in hydrogen-doped Zr-Ni and Zr-Cu metallic glasses

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Measurements are described and the results presented for the temperature and magnetic field dependence of the electrical conductivity and for the magnetic susceptibility of Zr-Ni and Zr-Cu metallic glasses doped with hydrogen. Hydrogen is found to reduce both the effective electron diffusion constant and the screening of the Coulomb interaction which leads to a decrease in the conductivity, suppression of the superconducting transition temperature, and to an enhancement of the spin susceptibility at low temperatures. The relative contributions of the spin-orbit and the Maki-Thompson interaction to the magnetoresistivity are also depressed. It is shown that the theoretical models of weak localization and Coulomb interaction provide both a good qualitative and quantitative description of the anomalous temperature and field dependence of the measured properties.

### INTRODUCTION

It is now widely accepted<sup>1-5</sup> that the anomalous transport properties (i.e., the negative temperature coefficient of the electrical conductivity at low temperatures and the magnetoresistivity which is a few orders of magnitude larger than the expected "normal" contribution) of high-resistivity metallic glasses (usually containing a transition metal) can be well described, both qualitatively and quantitatively, with theoretical models of weak localization<sup>6,7</sup> (WL) and Coulomb interaction<sup>8,9</sup> (CI). Bergmann<sup>10,11</sup> has argued that both WL and CI effects can be visualized as the interference of the conduction electrons scattered by impurities leading to an "echo" in the backward direction, due to WL, and to the appearance of a charge hologram, due to CI, and that inelastic scattering should suppress both anomalies.

Quantum-interference theories have been developed for free electrons and isotropical scattering in weakly disordered systems and result from perturbation expansion when  $(k_F l)^{-1} \ll 1$ . That is why their applicability to strong scattering systems ( $k_F l \sim 1$ ) with more than one conduction band (i.e., transition-metal alloys studied in this work with dominant contribution of the  $d$  electrons to the electrical conductivity and  $k_F l \sim 3$ ) may be questioned. However, it has been shown<sup>12</sup> that they could be applied to alloys with more complicated conduction bands and arbitrary impurity scattering because various parameters of the models (such as inelastic, spin-orbit, and magnetic scattering times) are averaged over all electronic states. It has also been argued<sup>13</sup> that the condition  $k_F l \gg 1$  can be relaxed and that the higher-order terms in a perturbation expansion in  $(k_F l)^{-1}$  should be included only when  $k_F l$  is very near to 1. That is why, lacking exact theories for transition metals, we have interpreted our data using the WL and CI models mentioned above.

In this paper we present the results and a detailed

quantitative analysis for the electrical conductivity, magnetoresistivity, and magnetic susceptibility of the hydrogen-doped Zr-Ni and Zr-Cu metallic glasses.

We have already demonstrated that hydrogen<sup>3,14,15</sup> can be used as an atomic probe to study quantum interference at defects and to gain an insight into the way the atomic microenvironment influences the electronic properties of a disordered system, but the early data were mainly described qualitatively.

Zr-Ni and Zr-Cu metallic glasses are characterized by high room-temperature resistivities ( $\sim 200 \mu\Omega \text{ cm}$ ). Both systems are paramagnetic and become superconducting at temperatures below 3 K depending on the alloy composition ( $T_c$  is higher for higher Zr concentration).

The ultraviolet photoelectron spectroscopy (UPS) data<sup>16</sup> and the band-structure calculation<sup>17</sup> have shown that the density of states at the Fermi level of the early-late transition-metal glasses is dominated by the early transition element (Zr in our case).

Soft-x-ray spectroscopy (SXES) measurements of Zr-Ni and Zr-Ni-D metallic glasses have been performed to study the effect of deuterium on the local density of states of Zr and Ni. It has been shown<sup>18</sup> that the Zr  $4d$  band is strongly modified by the deuterium. The addition of deuterium reduces the peak height of the emission spectra and produces a subband at about 6.5 eV below the Fermi level. The peak has been attributed to the formation of Zr-D bonding states. At the same time the emission spectrum of Ni shows no significant changes when deuterium is added and there is no sign of Ni-D bond formation. These results are consistent with the structural analyses<sup>19,20</sup> which reveal that H (or D) atoms preferentially occupy the tetrahedral holes inherent in a glass structure, surrounded by four Zr atoms. The sites defined by three Zr and one Ni atom are characterized by higher internal energy (because Ni atoms are smaller than

Zr ones and therefore their negative contribution to the crystal field at the tetrahedral site will be smaller) and thus a smaller heat of formation with hydrogen. That is why these sites begin to be occupied by hydrogen atoms only at higher H (or D) concentrations. This happens at  $x > 0.4$  as indicated by SXES (Ref. 21) and hydrogen absorption<sup>22</sup> measurements.

### EXPERIMENTAL DETAILS

Ribbons of Zr-Ni and Zr-Cu metallic glasses were prepared by rapid solidification of the melt on a single-roll spinning copper wheel in an argon atmosphere. The hydrogenation was carried out electrochemically using the sample as a cathode and a platinum wire as an anode. The electrolyte used was 0.1 N H<sub>2</sub>SO<sub>4</sub>. The dopant concentrations were determined volumetrically.

The structures of the as-quenched and hydrogenated samples were examined by x-ray diffraction using Cu K $\alpha$  radiation to confirm that they were amorphous.

The magnetoresistance was measured by a low-frequency, four-probe ac method, with a relative precision of 10<sup>-6</sup>, using a superconducting magnet. The samples were mounted on an orientable holder, situated in a separate He bath, to facilitate measurements in transverse and longitudinal fields. The temperature range

covered was from 1.7 to 4.2 K in magnetic fields up to 6.5 T.

The electrical resistance was measured with a standard four-probe dc technique in the temperature range from 2 to 300 K. The precision of these measurements extended to a few parts in 10<sup>6</sup>.

Magnetic-susceptibility measurements were carried out by the Faraday method using a Cahn electrobalance combined with a conventional magnet in fields up to 0.94 T. A sensitivity of 10<sup>-7</sup> J T<sup>-2</sup> mol<sup>-1</sup> was maintained in these measurements throughout the temperature range from 2 to 300 K.

### THEORETICAL MODELS

#### Magnetic field dependence

Quantum corrections to the magnetoresistivity are dominated by the localization effects except at very low temperatures and high magnetic fields where the effects arising from electron-electron interaction should be included.

The full expression of the magnetoresistivity due to weak localization, superconducting fluctuations (Maki-Thompson contribution), spin-orbit scattering, spin Zeeman, and orbital effects is given by<sup>23</sup>

$$\begin{aligned} \Delta\rho = & \rho(H, T) - \rho(0, T) \\ = & -A\rho^2 \left\{ h^{1/2} \left[ F_3 \left[ \frac{1+t}{h} \right] - \beta F_3 \left[ \frac{t}{h} \right] \right] + 0.5 \left[ \frac{h}{1-\gamma} \right]^{1/2} \left[ F_3 \left[ \frac{t_+}{h} \right] - F_3 \left[ \frac{t_-}{h} \right] \right] \right. \\ & \left. - \frac{1}{(1-\gamma)^{1/2}} (t_-^{1/2} - t_+^{1/2}) + t^{1/2} - (t+1)^{1/2} \right\}, \end{aligned} \quad (1)$$

with

$$A = \frac{e^2}{2\pi^2\hbar} (D\tau_{so})^{-1/2}, \quad \gamma = \left[ \frac{g^* \mu_B H \tau_{so}}{2\hbar} \right]^2,$$

$$h = \frac{eDH\tau_{so}}{\hbar}, \quad t = \frac{\tau_{so}}{4\tau_i},$$

$$t_{\pm} = t + 0.5[1 \pm (1-\gamma)^{1/2}],$$

and

$$F_3(z) = \sum_{n=0}^{\infty} \left\{ 2[(n+z+1)^{1/2} - (n+z)^{1/2}] - (n+z+\frac{1}{2})^{-1/2} \right\}$$

where  $D = v_F^2\tau/3$  is the diffusion constant;  $\tau$ ,  $\tau_i$ , and  $\tau_{so}$  are the elastic, inelastic, and spin-orbit scattering times, respectively;  $g^*$  is the effective  $g$  factor; and  $\mu_B$  is the Bohr magneton. The term proportional to  $\beta$  arises from the Maki-Thompson interaction. The approximated field dependence of  $\beta$  is given through a field-dependent effective coupling constant  $g$  (on which  $\beta$  depends), which has been recently calculated by McLean and Suzuki:<sup>24</sup>

$$g(T, H)^{-1} = \ln \left[ \frac{T_c}{T} \right] + \Psi\left(\frac{1}{2}\right) - \Psi \left[ \frac{1}{2} + \frac{DeH}{2\pi k_B T} \right], \quad (2)$$

where  $\Psi$  is the digamma function. After calculating  $g(T, H)$  one can use Larkin's tabulation<sup>25</sup> to obtain  $\beta(T, H)$  although this tabulation does not take into account the influence of the magnetic field. (This is the best one could do in the absence of a more comprehensive theory.)

The contributions to the magnetoresistivity arising from Coulomb interaction are (a) the orbital effects in the Cooper channel<sup>26</sup>

$$\Delta\rho = \rho^2 g(T, H) \frac{e^2}{8\pi^2\hbar} \left[ \frac{eH}{\hbar} \right]^{1/2} \Phi_3 \left[ \frac{2DeH}{\pi k_B T} \right] \quad (3)$$

and (b) the spin splitting of the conduction electron energies in the diffusion channel<sup>27</sup>

$$\Delta\rho = \rho^2 \frac{e^2}{4\pi^2\hbar} \bar{F} \left[ \frac{k_B T}{2\hbar D} \right]^{1/2} g_3 \left[ \frac{g\mu_B H}{k_B T} \right]. \quad (4)$$

Here

$$\bar{F} = \frac{32F}{3} \left[ \left[ 1 + \frac{F}{2} \right]^{3/2} - \left[ 1 + \frac{3F}{4} \right] \right],$$

where  $F$  is the averaged screened Coulomb potential ( $0 \leq F \leq 1$ , depending on the screening length). The func-

tions  $g_3$  and  $\Phi_3$  are similar in form to  $F_3$  which results in the same field dependence of all the contributions to the magnetoresistivity (1), (3), and (4) ( $H^2$  at low fields and  $H^{1/2}$  at high fields).

### Temperature dependence

The temperature-dependent quantum corrections to the conductivity due to localization in the presence of inelastic scattering  $\tau_i$  and spin-orbit scattering  $\tau_{so}$  are given as<sup>23</sup>

$$\Delta\sigma(0, T) = \left[ \frac{e^2}{2\pi^2\hbar} \right] \left[ 3 \left[ \frac{1}{D\tau_{so}} + \frac{1}{4D\tau_i} \right]^{1/2} - \left[ \frac{1}{4D\tau_i} \right]^{1/2} \right] \quad (5)$$

At low temperatures  $T < \Theta_D$ , the dominant contribution to the inelastic scattering comes from the electron-phonon interaction which in metallic glasses is proportional to  $T^2$  (also there exists some experimental evidence that at the lowest temperatures  $\tau_i^{-1} \sim T^p$  with  $2 < p < 4$ ). This gives the localization correction to the classical conductivity which is proportional to  $T$ .

The corrections from the electron-electron interaction, both in the diffusion and the Cooper channel, take the following form:<sup>28</sup>

$$\Delta\sigma(T) = \frac{1.3e^2}{4\pi^2\hbar} \left[ \frac{4}{3} - 3F^* - \frac{2}{\ln\left[\frac{T_c}{T}\right]} \right] \left[ \frac{k_B T}{D\hbar} \right]^{1/2}, \quad (6)$$

where  $T_c$  is the superconducting transition temperature and  $F^* = F - \lambda$  with  $\lambda$  the electron-phonon coupling constant which can be estimated from the measured  $T_c$  using the McMillan formula.<sup>29</sup>  $D$  and  $F$  have been defined in the previous section.

In contrast to the temperature and field dependence of the magnetoresistivity, where the dominant part comes from the localization effects, temperature dependence of the resistivity for temperatures  $T < 20$  K is dominated by the interaction effects. The localization term is small at low temperatures because the number of the inelastic scattering processes which are responsible for the phase breaking of the electron coherence falls off with decreasing temperature.

### Magnetic susceptibility

The corrections to the magnetic susceptibility in a disordered system that are weakly temperature dependent arise from the spin-splitting and the orbital effects. Altshuler and Aronov have shown<sup>8</sup> that the Coulomb interaction between electrons in a disordered metal is enhanced due to their diffusive motion leading to an enhancement in the density of states at the Fermi level and its square-root-law dependence on quasiparticle energy for the three-dimensional systems. This anomalous behavior of the density of states shows up in the anomalous temperature dependence of the magnetic susceptibil-

ity.

The correction to the orbital magnetic susceptibility in the Cooper channel in 3D systems, due to the correction in the density of states, can be expressed as<sup>30</sup>

$$\delta\chi = -2\chi_0 \left[ \frac{\pi}{6} \right]^{1/2} \zeta\left(\frac{1}{2}\right) \left[ \frac{T\tau}{\hbar} \right]^{1/2} \ln^{-1} \left[ \frac{T_c}{T} \right], \quad (7)$$

where  $\chi_0 = -\frac{2}{3}\beta^* N_0$  is the diamagnetic susceptibility of electrons,  $\beta^* = \hbar e / 2m^* c$ ,  $N_0$  is the density of states for one spin orientation,  $\zeta(x)$  is the Riemann zeta function,  $\tau$  is the momentum relaxation time, and  $T_c$  is the superconducting transition temperature,

$$T_c \sim \omega_D \exp \left[ \frac{1}{\lambda_c} \right],$$

$$\lambda_c = \lambda_{ph} + \left\{ \lambda_{Coul} / \left[ 1 + \lambda_{Coul} \ln \left[ \frac{E_F}{\omega_D} \right] \right] \right\}.$$

In a magnetic field for which  $1/\tau \gg \Omega_H \gg T/\hbar$  and  $\hbar\Omega_H \gg g\mu_B H = \hbar\omega_s$ , where  $g$  is the  $g$ -factor of conduction electrons, the orbital magnetic susceptibility becomes field dependent and is given as<sup>30</sup>

$$\Delta\chi(H) = - \frac{\chi_0 \left(\frac{1}{2}\right)^{1/2} (1 - 2^{3/2}) 15 \zeta\left(\frac{3}{2}\right) (\tau\Omega_H)^{1/2}}{4\pi^2 \ln \left[ \frac{k_B T}{\hbar\Omega_H} \right]}, \quad (8)$$

where  $\Omega_H = 4DeH/\hbar c$ .

The spin-splitting effect also gives rise to a correction to the magnetic susceptibility. However, as was first shown by Fukuyama,<sup>31</sup> only the Hartree interaction between up and down spin subbands in a disordered Fermi system leads to an enhancement of the susceptibility at low temperatures. At the same time the Hartree term involving equal spins and the exchange term do not affect the spin susceptibility just as electron-phonon enhancement of the density of states leaves the susceptibility unchanged.

The quantum corrections to the spin susceptibility in both the Cooper,  $\delta\chi_s^C$ , and the diffusion channel,  $\delta\chi_s^D$ , in 3D system are expressed as<sup>30</sup>

$$\delta\chi_s = \delta\chi_s^C + \delta\chi_s^D$$

$$= \frac{\zeta\left(\frac{1}{2}\right)(g\mu_B)^2}{16\pi^{3/2} 2^{1/2} (D\hbar)^{3/2}} T^{1/2} \left[ 2 \ln^{-1} \left[ \frac{T_c}{T} \right] - \lambda^{(j=1)} \right], \quad (9)$$

where  $\lambda^{(j=1)}$  is a dimensionless constant for the electron-hole interaction  $\lambda^{(j=1)} > 0$  for an attractive interaction).  $\delta\chi_s^C$  and  $\delta\chi_s^D$  become field dependent for  $\hbar\Omega_H \geq k_B T$  and  $\hbar\omega_s \geq k_B T$ , respectively. This correction is enhanced for transition metals by the Stoner enhancement factor ( $\sim 1.7$  in the system investigated).

## RESULTS

## Magnetoresistivity

The magnetoresistance results pertaining to the  $Zr_{0.67}Ni_{0.33}H_x$  ( $x=0, 0.1, 0.2, 0.25$ ) samples at 4.2 K and  $Zr_{0.70}Cu_{0.30}H_x$  ( $x=0, 0.1, 0.2$ ) at 2.9 K in the magnetic fields up to 6 T are shown in Fig. 1 and Fig. 2, respectively. The magnetoresistance was found to be independent of the sample orientation with respect to the magnetic field as predicted by the theoretical models for 3D systems.

The magnetoresistance slopes are lowered by the hydrogen dopant and the saturation is shifted to lower fields. These effects are more pronounced for  $Zr_{0.67}Ni_{0.33}H_x$  samples.

The solid curves fitted to the experimental data in Fig. 1 and Fig. 2 are derived from the sum of the relations (1), (3), and (4), with the inelastic scattering time  $\tau_i$ , the spin-orbit scattering time  $\tau_{so}$ , the superconducting transition temperature  $T_c$ , the diffusion constant  $D$ , and the screening parameter  $F$ , used as the fitting parameters. The term  $(1-\gamma)^{1/2}$  in (1) can become imaginary for certain values of the magnetic fields and the spin-orbit relaxation times. However, it can be shown that  $\Delta\rho$  remains real. In our calculations we have treated all the variables containing  $\gamma$  as complex. The contribution to the magnetoresistivity from the spin-splitting term [Eq. (4)] is greatly reduced in our systems by the spin-orbit scattering and it amounts to

about 5% of the measured values at the highest magnetic field used. The orbital contribution, Eq. (3), is even smaller (2%).

As mentioned above the effective coupling constant  $g(H, T)$  has been calculated from Eq. (2) and then  $\beta(H, T)$  determined using the tabulation by Larkin. In this computation the diffusion constant was used as the fitting parameter so that the experimental and theoretical points derived from Eqs. (1), (3), and (4) coincide. The values of the diffusion constants derived in this way are only about 10% lower than the values of  $D$  calculated from the Einstein relation  $D = [e^2 N(E_F) \rho]^{-1}$  using measured resistivities and  $N_0(E_F)$  from the specific heat results<sup>32</sup> (e.g.,  $\rho = 180 \mu\Omega \text{ cm}$ ,  $D = 3.3 \times 10^{-5} \text{ m}^2 \text{ s}^{-1}$ ,  $N(E_F) = 2.04 \text{ states eV}^{-1} \text{ at.}^{-1}$  for  $Zr_{0.67}Ni_{0.33}$ ).

This procedure is very sensitive to the choice of  $D$ . With higher values of  $D$ , calculated on the basis of the free-electron model, we were unable to fit our experimental data successfully and the magnetic field dependence of  $\beta$  was much stronger than determined experimentally.

It can be seen from Table I that the diffusion constant is lowered and the spin-orbit scattering time is enhanced with increasing hydrogen concentration (e.g.,  $\tau_{so} = 2.44 \times 10^{-12} \text{ s}$  in  $Zr_{0.67}Ni_{0.33}$  and  $6.71 \times 10^{-12} \text{ s}$  in  $Zr_{0.67}Ni_{0.33}H_{0.25}$ ). Since most of the spin-orbit scattering takes place on Zr atoms and in the  $d$  band ( $\tau_{so}^{-1} \sim Z^8$  in a free-electron model, and Hickey *et al.*,<sup>33</sup> have found that  $\tau_{so}^{-1} \sim Z^{12}$  in CuTi alloys) the reduction of the effective spin-orbit contribution to the magnetoresistivity by the dopant can be taken as the evidence that hydrogen atoms migrate mainly to the Zr-rich sites. Comparison between the behavior of (Zr-Ni)-H and (Zr-Cu)-H systems in this

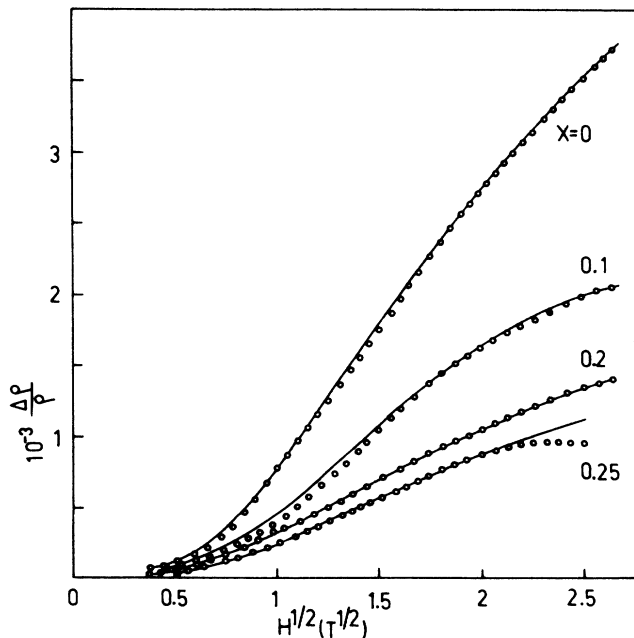


FIG. 1. Magnetoresistance of  $Zr_{0.67}Ni_{0.33}H_x$  metallic glasses, for different values of hydrogen concentration  $x$ , obtained at 4.2 K. Solid lines are theoretical fits to Eqs. (1), (3), and (4).

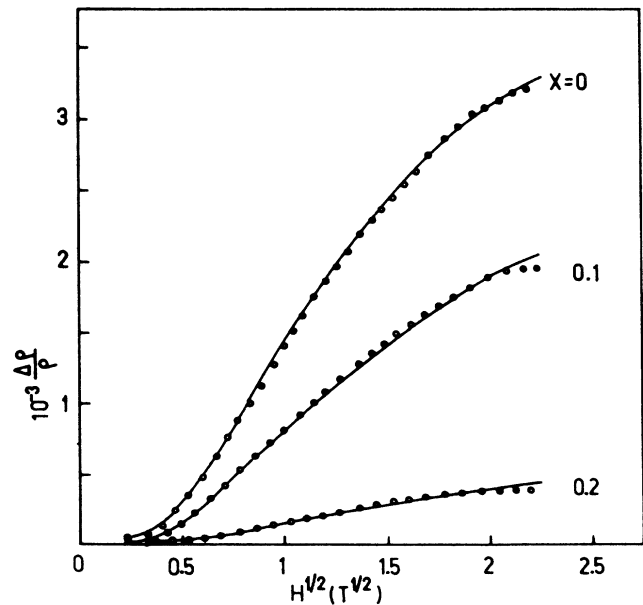


FIG. 2. Magnetoresistance of  $Zr_{0.7}Cu_{0.3}H_x$  metallic glasses obtained at 2.9 K.

TABLE I. Values of the diffusion coefficient  $D$ , the inelastic scattering time  $\tau_i$ , the spin-orbit scattering time  $\tau_{so}$ , the screening parameter  $F$ , the Maki-Thompson coefficient  $\beta$  as determined from magnetoresistivity data, and superconducting transition temperature  $T_c$ .

Sample	$D$ ( $10^{-5} \text{ m}^2 \text{ s}^{-1}$ )	$\tau_i$ ( $10^{-11} \text{ s}$ )	$\tau_{so}$ ( $10^{-12} \text{ s}$ )	$F$	$\beta$ (4.2 K)	$T_c$ (K)
$\text{Zr}_{0.67}\text{Ni}_{0.33}$	2.8	3.3	2.44	0.45	3.97	2.78
$\text{Zr}_{0.67}\text{Ni}_{0.33}\text{H}_{0.1}$	2.7	3.3	4.37	0.2	1.32	1.76
$\text{Zr}_{0.67}\text{Ni}_{0.33}\text{H}_{0.2}$	2.65	6.1	4.37	0.2	1.035	1.54
$\text{Zr}_{0.67}\text{Ni}_{0.33}\text{H}_{0.25}$	2.55	5.7	6.71	0.19	0.59	0.9
$\text{Zr}_{0.7}\text{Cu}_{0.3}$	3.3	5.4	1.03	0.3	1.22	1.7
$\text{Zr}_{0.7}\text{Cu}_{0.3}\text{H}_{0.1}$	3.15	10.4	1.16	0.2	0.64	1.0
$\text{Zr}_{0.7}\text{Cu}_{0.3}\text{H}_{0.2}$	3.0	12.2	1.4	0.2	0.2	0.3

regard shows a much stronger influence of the dopant on the latter one. This indicates that some of the hydrogen atoms in Zr-Ni tend to migrate to sites coordinated with three Zr atoms and one Ni atom while this process seems to be absent in the Zr-Cu system.

The field dependence of  $\beta(H, T)$ , determined as described above, at two different temperatures for  $\text{Zr}_{0.7}\text{Cu}_{0.3}$  and  $\text{Zr}_{0.7}\text{Cu}_{0.3}\text{H}_{0.2}$  samples are plotted in Fig. 3. In the case of  $\text{Zr}_{0.7}\text{Cu}_{0.3}$  (with  $T_c = 1.7$  K) the field dependence is already seen at 4.2 K and it is enhanced as the temperature is lowered and approaches  $T_c$ , because the Maki-Thompson contributions increases. In the case of  $\text{Zr}_{0.7}\text{Cu}_{0.3}\text{H}_{0.2}$  (with  $T_c = 0.3$  K)  $\beta$  is small and thus practically field independent.

The values of  $T_c$  derived from  $\beta$  (at  $T = 4.2$ ,  $H = 0$  T) together with the samples resistivity, for  $\text{Zr}_{0.67}\text{Ni}_{0.33}\text{H}_x$ , are plotted as a function of dopant concentration in Fig. 4. On the same figure are the data for  $\text{Zr}_y\text{-Pd}_{1-y}\text{H}_x$  taken from Mizutani *et al.*<sup>32</sup>

The suppression of  $T_c$  by the hydrogen is observed to be very fast at smaller dopant concentrations and then

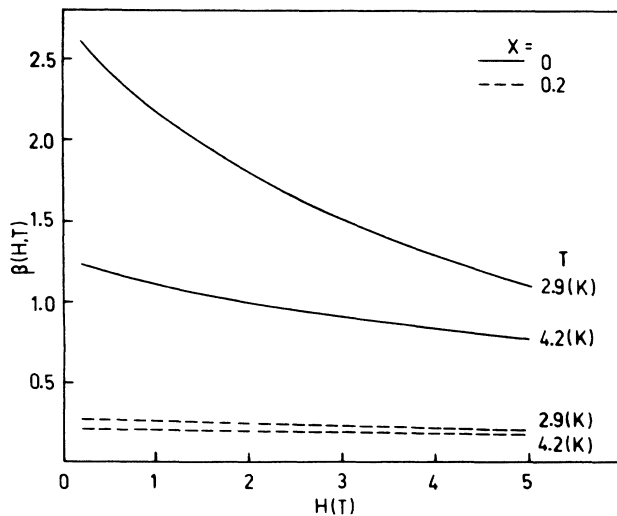


FIG. 3. The field dependence of the Maki-Thompson parameter  $\beta$  for  $\text{Zr}_{0.7}\text{Cu}_{0.3}\text{H}_x$  at two different temperatures. The process by which  $\beta$  is determined is described in the text.

levels off at  $x \geq 0.5$ . The reduction of  $T_c$  is proportional to the sample resistivity which is the dopant increased and the leveling out of  $T_c$  is accompanied by saturation in the resistivity.

The resistivity saturation can be explained by the tendency of the electron mean free path ( $l_0$ ) to converge to a constant value, i.e., to the interatomic distance (Ioffe-Regel limit).<sup>34</sup> If one calculates  $l_0$  using the relation  $\rho^{-1} = e^2 N_0(E_F) v_F l_0 / 3$  one obtains, for example, for undoped  $\text{Zr}_{0.67}\text{Ni}_{0.33}$   $l_0 = 3.5$  Å which corresponds to Zr nearest neighbors while for  $\text{Zr}_{0.67}\text{Ni}_{0.33}\text{H}_{0.33}$   $l_0 = 1.8$  Å which corresponds to the Zr-H distances.<sup>35</sup>

Anderson *et al.*<sup>36</sup> have estimated the resistivity dependence of the superconducting transition temperature  $T_c$  and have found that in the case of strong disorder ( $k_F l_0 \sim 1$ ), as measured by resistivity, the slowing down of electron diffusion enhances the Coulomb repulsive interaction and thus reduces  $T_c$ . Their results describe well

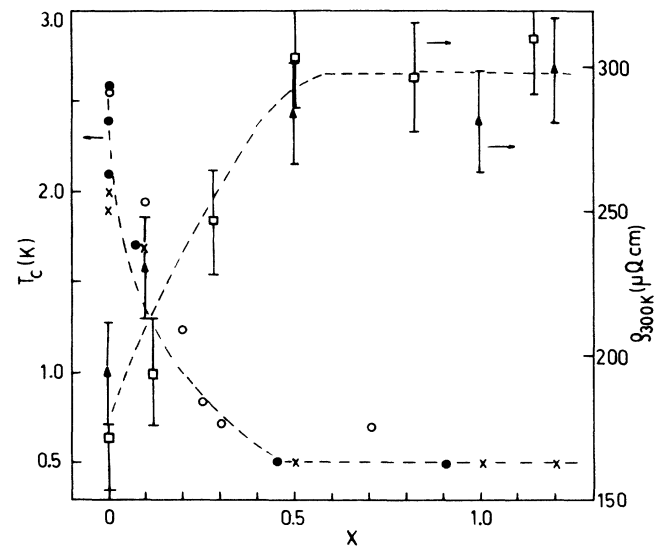


FIG. 4. Superconducting transition temperature  $T_c$  of  $\circ$ ,  $\text{Zr}_{0.67}\text{Cu}_{0.33}\text{H}_x$  (our results);  $\bullet$ ,  $\text{Zr}_{0.67}\text{Ni}_{0.33}\text{H}_x$  (Ref. 32);  $\times$ ,  $\text{Zr}_{0.65}\text{Pd}_{0.35}\text{H}_x$  (Ref. 32); and electrical resistivity  $\rho$  of  $\square$ ,  $\text{Zr}_{0.4}\text{Ni}_{0.6}\text{H}_x$  (our results);  $\blacktriangle$ ,  $\text{Zr}_{0.65}\text{Pd}_{0.35}\text{H}_x$  (Ref. 32) as a function of hydrogen concentration. Lines are only to guide the eye.

the experimentally observed degradation of  $T_c$  with the sample resistivity in some disordered A15 compounds.

Using  $T_c$  values from Table I and  $\Theta_D$  from the specific heat measurements<sup>32</sup> ( $\Theta_D = 234$  K for  $Zr_{0.67}Ni_{0.33}$  and 273 K for  $Zr_{0.67}Ni_{0.33}H_{0.7}$ ), the McMillan's relation for the electron-phonon coupling constant  $\lambda_{ph}$ ,<sup>29</sup>

$$\lambda_{ph} = \frac{1.04 + \mu^* \ln \left[ \frac{\Theta_D}{1.45T_c} \right]}{(1 - 0.67\mu^*) \ln \left[ \frac{\Theta_D}{1.45T_c} \right] - 1.04}, \quad (10)$$

with  $\mu^*$  the screened Coulomb potential ( $\mu^*$  is usually taken to be 0.13 for transition metals because it is very weakly dependent on the density of states), gives  $\lambda_{ph}$  that decreases from 0.57 to 0.41 with increasing hydrogen concentration (up to  $x \approx 0.5$ ) and then saturates. At the same time the density of states at the Fermi level continues to fall.<sup>32</sup> The same trends have been observed in Zr-Pd-D, Zr-Ni-D through the specific heat and electrical resistivity measurements (Ref. 15 of Ref. 32).

The electron-phonon coupling constant is usually expressed in the form

$$\lambda_{ph} = \frac{N_0(E_F) \langle I^2 \rangle}{M \langle \omega^2 \rangle}, \quad (11)$$

where  $N_0(E_F)$  is the bare density of states at  $E_F$ ,  $\langle I^2 \rangle$  is an average of the squared electron-phonon matrix element,  $M$  is the ionic mass, and  $\langle \omega^2 \rangle$  an average square phonon frequency which is taken as varying as  $\Theta_D^2$ .

The tight-binding approximation applied to transition metals predicts a near constance of the product  $N_0(E_F) \langle I^2 \rangle$ , because  $\langle I^2 \rangle$  is proportional to  $\langle v_F^2 \rangle$ .  $N_0(E_F)$  and  $\langle v_F^2 \rangle$ , in general, behave inversely and thus tend to neutralize each other in the superconductivity parameter  $\lambda_{ph}$ . Thus the density of states is eliminated from the coupling constant. The experimental results show<sup>37</sup> that this is really a case for amorphous Zr-Ni and Zr-Cu alloys, that is, the value of the Hopfield parameter [ $\eta = N_0(E_F) \langle I^2 \rangle$ ] is constant and independent of both Zr and the specific  $3d$  element and their concentration. That is why we believe that the depression of  $T_c$  with hydrogen is caused partly by the increase of  $\Theta_D$  (due to the additional coupling of electrons to the hydrogen modes) and partly by the increase of the effective Coulomb pseudopotential which is a localization effect. A more quantitative analysis will be given elsewhere.

#### Temperature dependence of the electrical resistivity

The temperature-dependent part of the electrical resistivity for hydrogen-doped  $Zr_{0.67}Ni_{0.33}H_x$  ( $x = 0, 0.004, 0.01, 0.104, 0.108, 0.123$ ) metallic glass is shown in Fig. 5.

The temperature coefficient of the resistivity is negative in the temperature range investigated (2–300 K) and increases (faster below 100 K) with hydrogen concentration. As mentioned earlier the dopant causes the resistivity to increase and the superconducting transition temperature to decrease.

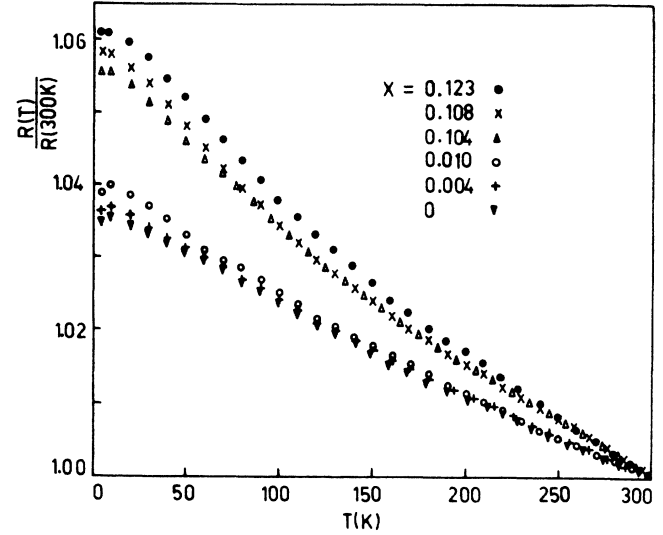


FIG. 5. Temperature dependence of the resistance normalized to the value at 300 K, for  $Zr_{0.67}Ni_{0.33}H_x$  metallic glasses.

The resistivity increase is partly caused by the observed decrease of the density of  $d$  states at the Fermi level (Table III) with the dopant concentration. This is consistent with the theoretical and experimental results which show that the electrical conductivity of a transition metal alloy is dominated by the  $d$  electrons.

The experimental data are fitted to the relation

$$\frac{\Delta\rho(T)}{\rho} = -A[3(B + C^2T^2)^{1/2} - CT + ST^{1/2}E], \quad (12)$$

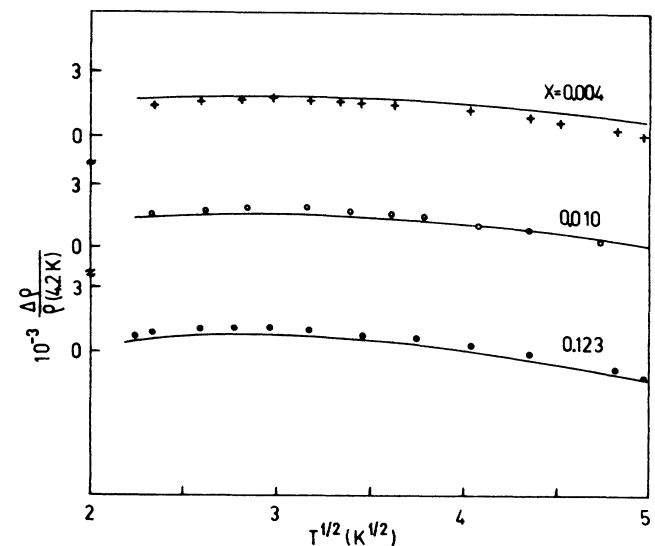


FIG. 6. The change in the resistivity [ $\Delta\rho(T) - \Delta\rho(4.2$  K)] normalized to the value at 4.2 K vs the square root of the temperature below 25 K for  $Zr_{0.67}Ni_{0.33}H_x$  metallic glasses. The lines are the best fits obtained from Eq. (12).

TABLE II. Values of the diffusion coefficient  $D$ , the inelastic scattering time  $\tau_i$ , the spin-orbit scattering time  $\tau_{so}$ , and the superconducting transition temperature  $T_c$  as determined from the temperature dependence of the resistivity.

Sample	$D$ ( $10^{-5}$ m $^2$ s $^{-1}$ )	$\tau_i$ ( $10^{-11}$ s)	$\tau_{so}$ ( $10^{-12}$ s)	$T_c$
Zr $_{0.67}$ Ni $_{0.33}$ H $_{0.004}$	2.94	3	3.4	2.57
Zr $_{0.67}$ Ni $_{0.33}$ H $_{0.01}$	2.91	2.76	3.43	2.57
Zr $_{0.67}$ Ni $_{0.33}$ H $_{0.123}$	2.7	2.33	4.41	2.43

where  $A = e^2/2\pi^2\hbar$  is constant and  $B = 1/D\tau_{so}$ ,  $C = (1/4D\alpha)^{1/2}$ , and

$$S = \frac{1.3}{2} \left[ \frac{4}{3} - 3F^* - \frac{2}{\ln \left( \frac{T_c}{T} \right)} \right]$$

and  $E = (k_B/D\hbar)^{1/2}$  are parameters of the fit. It is assumed that  $\tau_i = \alpha T^{-2}$ . The values of  $D$ ,  $\tau_i$ ,  $\tau_{so}$ , and  $T_c$  as determined from the fit are given in Table II. Their magnitudes and concentration dependence are in good agreement with the ones obtained from the magnetoresistivity data (Table I).

The fit of the experimental data to Eq. (12) (solid lines) is shown in Fig. 6 versus the square root of temperature for temperatures up to 25 K and in Fig. 7 versus temperature for temperatures up to 80 K. The quantum corrections to the resistivity are proportional to  $T^{1/2}\ln^{-1}(T_c/T)$  from 5 to 16 K as predicted by the Coulomb interaction model and to  $-T$  from 16 to 80 K due to electron localization. Below 5 K one should take into account the Aslamazov-Larkin contribution to the conductivity, which is beyond the scope of the present work.

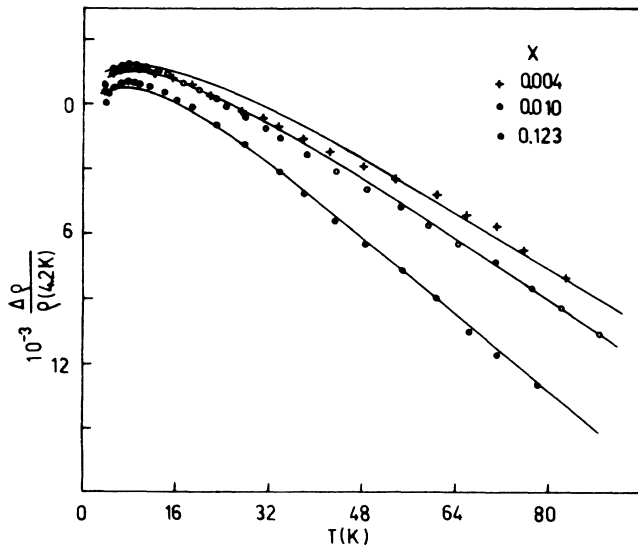


FIG. 7. The change in the resistivity [ $\Delta\rho(T) - \Delta\rho(4.2 \text{ K})$ ] normalized to the value at 4.2 K as a function of temperature below 80 K.

### Magnetic susceptibility

The temperature dependence of the magnetic susceptibility data for Zr $_{0.67}$ Ni $_{0.33}$ H $_x$  ( $x=0,0.13,0.33$ ) and Zr $_{0.60}$ Cu $_{0.40}$ H $_x$  ( $x=0,0.11$ ) samples are plotted in Fig. 8 versus the square root of temperature. The values of the susceptibilities are field independent up to 9.4 kOe, thus confirming the absence of magnetic impurities. Both systems are paramagnetic with susceptibilities that are only weakly temperature dependent down to about 30 K, below which a slight increase is observed. A shallow minimum exists between 60 and 115 K. The susceptibility is lowered upon hydrogenation, the low-temperature upturn is increased and the position of the minimum is shifted to higher temperatures. The enhancement of the magnetic susceptibility at low temperatures proportional to hydrogen concentration has been also observed<sup>38</sup> in Zr $_2$ PdH $_x$  and Zr $_3$ RhH $_x$  and in amorphous Zr $_2$ Pd as compared with the crystalline one, but without an explanation of the effect. At room temperature  $\chi_{\text{exp}} = 128 \times 10^{-5} \text{ J T}^{-2} \text{ mol}^{-1}$  for Zr $_{0.67}$ Ni $_{0.33}$  whereas for Zr $_{0.67}$ Ni $_{0.33}$ H $_{0.33}$  it is  $\chi_{\text{exp}} = 105 \times 10^{-5} \text{ J T}^{-2} \text{ mol}^{-1}$ . Again for the Zr $_{0.60}$ Cu $_{0.40}$  system the effects are more pronounced; thus  $\chi_{\text{exp}}$  decreases from its room temperature value of  $93 \times 10^{-5} \text{ J T}^{-2} \text{ mol}^{-1}$  in the undoped sample to  $\chi_{\text{exp}} = 76 \times 10^{-5} \text{ J T}^{-2} \text{ mol}^{-1}$  for Zr $_{0.60}$ Cu $_{0.40}$ H $_{0.11}$ .

The temperature-independent magnetic susceptibility is generally given as

$$\chi_{\text{exp}} = \chi_{\text{el}}^P + \chi_{\text{ion}} + \chi_{\text{orb}}, \quad (13)$$

where  $\chi_{\text{ion}}$  and  $\chi_{\text{orb}}$  are the ionic core diamagnetism and orbital paramagnetism with the Landau contribution included, respectively.

Since the dominant contribution to the electronic density of states at the Fermi level in transition metals comes from  $d$  electrons the Pauli spin susceptibility is dominated by the  $d$  band and differs from the free-electron value through the Stoner enhancement factor

$$\chi_{\text{el}}^P = \frac{\mu_B^2 N_0(E_F)}{1 - I_{\text{eff}} N_0(E_F)}, \quad (14)$$

where  $\mu_B$  is the Bohr magneton,  $N_0(E_F)$  is the bare density of states at the Fermi level and  $I_{\text{eff}}$  is the effective exchange integral within the  $d$  band. While the ionic core susceptibility  $\chi_{\text{ion}}$  is relatively small for all the systems investigated (i.e., of the order of  $-20 \times 10^{-5} \text{ J T}^{-2} \text{ mol}^{-1}$ , which is an order of magnitude smaller than the measured values), the orbital magnetic moments of the electrons are not completely quenched for partly filled degen-



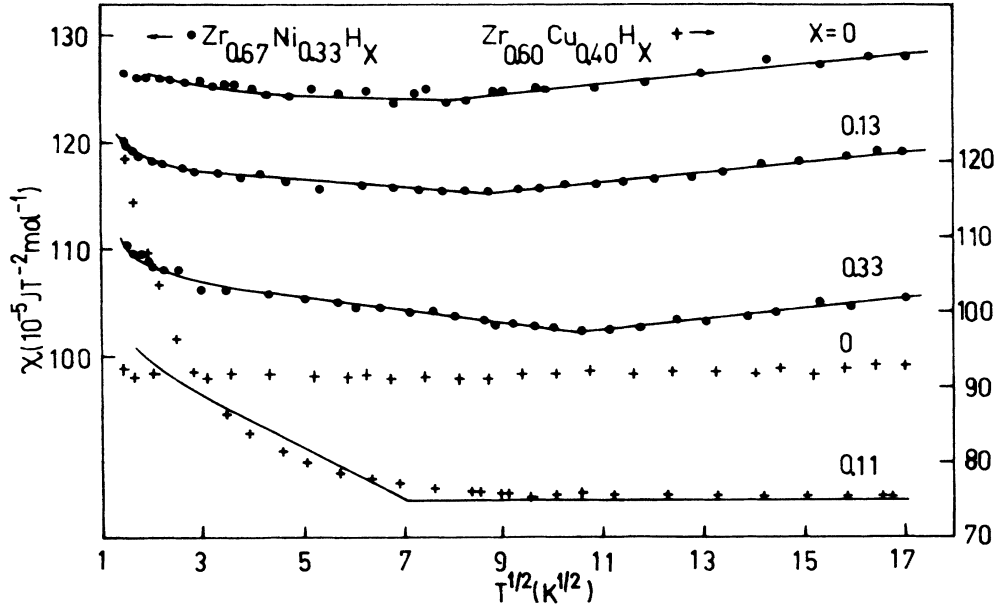


FIG. 8. The magnetic susceptibility of hydrogen-doped  $Zr_{0.67}Ni_{0.33}H_x$  and  $Zr_{0.6}Cu_{0.4}H_x$  metallic glasses as a function of the square root of the temperature. The lines are the best fits obtained from Eqs. (15) and (16).

erate bands (as is the case in Zr-based metallic glasses) and are of the same order of magnitude as the Pauli spin term.

A summary of measured and calculated values is given in Table III. Using for  $N_0(E_F) = 1.3$  states  $eV^{-1}at^{-1}$  the value taken from the measured electronic specific-heat coefficient<sup>32</sup> and  $I_{eff}$  for Zr and Ni as 0.022 Ry and 0.028 Ry, respectively,<sup>39</sup> the Stoner enhancement factor for  $Zr_{0.67}Ni_{0.33}$  turns out to be 1.7 and the Pauli spin susceptibility  $\chi_{el}^P = 73 \times 10^{-5} J T^{-2} mol^{-1}$ . Subtracting  $\chi_{el}^P$  and  $\chi_{ion}$  from  $\chi_{exp}$  gives the orbital susceptibility value of  $75 \times 10^{-5} J T^{-2} mol^{-1}$ . Zirconium gives much higher contributions to the orbital susceptibility than does nickel on account of its different  $d$ -band filling. If we assume that  $\chi_{orb}$  of the alloy is that of crystalline Zr ( $100 \times 10^{-5} J T^{-2} mol^{-1}$ ) and Ni ( $25 \times 10^{-5} J T^{-2} mol^{-1}$ ) scaled to their concentration we obtain a value which is in good agreement with the measured one.

The same analysis may be applied to the  $Zr_{0.60}Cu_{0.40}$  alloy. The Pauli spin susceptibility is found to be  $50.4 \times 10^{-5} J T^{-2} mol^{-1}$  and  $\chi_{orb} = 60.4 \times 10^{-5} J T^{-2} mol^{-1}$  which is again very close to the Zr value scaled to its concentration in the alloy and reflects the fact that Cu atoms have no orbital magnetic moments since the Cu  $d$  band is well below the Fermi level.<sup>16</sup>

The lowering of the room-temperature magnetic-susceptibility values upon hydrogenation can be explained as due to the influence of hydrogen on the electronic density of states at the Fermi level and/or its possible effect on the orbital moments. Since the dopant atoms migrate mainly to the Zr-rich sites where their  $s$ -electrons hybridize with the zirconium  $d$  band, a strong dependence of the electronic density of states and hence

of the Pauli susceptibility on the hydrogen concentration is expected. On the other hand we have assumed that the hydrogen electron hybridization with the Zr  $d$  band does not influence greatly the orbital paramagnetism.

We have calculated  $N_0(E_F)$  for the doped samples from the Eq. (14) with  $\chi_{orb}$ ,  $\chi_{ion}$ , and the Stoner factor as described above. The obtained values (Table III) decrease as the hydrogen concentration increases. [e.g.,  $N_0(E_F) = 1.3$  states  $eV^{-1}at^{-1}$  for  $Zr_{0.67}Ni_{0.33}$  and  $N_0(E_F) = 1.03$  states  $eV^{-1}at^{-1}$  for  $Zr_{0.67}Ni_{0.33}H_{0.33}$ ].

These values are in good agreement with the ones obtained from the electronic specific-heat measurements<sup>32</sup> which indicate that our observation of susceptibility suppression can be wholly attributed to a change in the Pauli term.

Again, the hydrogen influence is stronger in the Zr-Cu system since copper is in the  $d$  10 state in all its alloys and no hydrogen (for the concentration used), migrates to Cu-rich sites.

The temperature-dependent magnetic susceptibility has been fitted for temperatures  $T_c < T < T_{min}$  (where  $T_{min}$  corresponds to the minimum in the susceptibility) to the relation

$$\delta\chi = -AT^{1/2} - BT^{1/2} \ln^{-1} \left[ \frac{T_c}{T} \right], \quad (15)$$

where the first term on the right-hand side is a correction due to the spin-splitting effect in the diffusion channel and the second one are the corrections in the particle-particle channel due to the orbital effect and to the spin susceptibility.

The results could not be successfully fitted above  $T_{min}$

TABLE III. Magnetic susceptibility data for  $Zr_{0.67}Ni_{0.33}H_x$  and  $Zr_{0.6}Cu_{0.4}H_x$  metallic glasses.  $\chi$  s are in units of  $10^{-5} \text{ J T}^{-2} \text{ mol}^{-1}$ .

Sample	$\chi_{\text{exp}}$	$\chi_{\text{ion}}$	$\chi_0^P$	$\chi_{\text{el}}^P$	$\chi_{\text{orb}}$	$N_0$ (states/eV at.)
$Zr_{0.67}Ni_{0.33}$	128	-20	42	73	75	1.3
$Zr_{0.67}Ni_{0.33}H_{0.13}$	120	-20	39.2	65	75	1.21
$Zr_{0.67}Ni_{0.33}H_{0.33}$	105	-20	33.3	50	75	1.03
$Zr_{0.6}Cu_{0.4}$	93	-17.8	31.5	50.4	60.4	0.98
$Zr_{0.6}Cu_{0.4}H_{0.11}$	76	-17.8	20.9	33.4	60.4	0.65

to the relation (15), which is why for temperatures  $T > T_{\text{min}}$  the data have been fitted to the relation

$$\delta\chi = -BT^{1/2}\ln^{-1}\left[\frac{T_c}{T}\right] - C, \quad (16)$$

where  $A$  and  $B$  are the temperature-independent parameters of the fit and  $C = AT_{\text{min}}^{1/2}$ . This is because the inelastic relaxation time  $\tau_i$  becomes comparable to  $\tau$  at about 100 K ( $\tau_i \sim 5 \times 10^{-15}$  s at 100 K and  $\tau \sim 10^{-15}$  s from a diffusion constant) and localization effects, mainly confined to the diffusion channel, become negligible.

The values of the parameters of the fit together with the superconducting transition temperatures and  $T_{\text{min}}$  are given in Table IV. The corrections to the spin and orbital susceptibility are of the same order of magnitude ( $10^{-5} \text{ J T}^{-2} \text{ mol}^{-1}$ ) which is a few percent of the measured room-temperature value. The corrections due to the magnetic field have been neglected because they are within the experimental error.

The best fit yields  $A = 0.6 \times 10^{-5} \text{ J T}^{-2} \text{ mol}^{-1} \text{ K}^{-1/2}$  and  $B = 4 \times 10^{-5} \text{ J T}^{-2} \text{ mol}^{-1} \text{ K}^{-1/2}$  for the undoped sample. The value of  $B$  calculated from Eq. (7) and from  $\delta\chi_s^C$  part of Eq. (9) using the value of the diffusion coefficient  $D = 2.8 \times 10^{-5} \text{ m}^2 \text{ s}^{-1}$  (Table I) are  $2 \times 10^{-5} \text{ J T}^{-2} \text{ mol}^{-1} \text{ K}^{-1/2}$  and  $1.92 \times 10^{-5} \text{ J T}^{-2} \text{ mol}^{-1} \text{ K}^{-1/2}$ , respectively, and together give  $3.92 \times 10^{-5} \text{ J T}^{-2} \text{ mol}^{-1} \text{ K}^{-1/2}$  in good agreement with the experimental value. The value of  $A$  is also found to agree with the estimated from  $\delta\chi_s^D$  part of Eq. (9) which is  $0.57 \times 10^{-5} \text{ J T}^{-2} \text{ mol}^{-1} \text{ K}^{-1/2}$ .

It can be seen from Fig. 8 and Table IV that the temperature range where the fit to expression (15) is valid (solid line) widens at both ends upon hydrogenation. Since the superconducting transition temperature decreases as hydrogen concentration increases the fit is improved on the low-temperature side, because relation (7)

is valid for  $T \gg T_c$ . At the same time both  $A$  and  $B$  increase (Table IV) through the lowering of the diffusion constant but  $A$  is enhanced relative to  $B$ . A decrease of  $T_c$  reduces the contribution of superconducting fluctuations to the diamagnetic moment through the  $\ln^{-1}(T_c/T)$  term. The hydrogen dopant also reduces the orbital term (which enters coefficient  $B$ ) because it enhances localization by providing additional centers of quasielastic scattering. The enhancement of the spin susceptibility relatively to the orbital part upon hydrogenation below  $T_{\text{min}}$  is in agreement with the magnetoresistivity data (Table I and Fig. 1) which show that hydrogen reduces the spin-orbital scattering rate  $\tau_{\text{so}}^{-1}$  thus reducing the mixing of spin-up and spin-down subbands. The increase of temperature  $T_{\text{min}}$  of the magnetic susceptibility minimum can be related to an increase of the inelastic scattering time [Table I and experimental results in  $\text{PdH}_x$  (Ref. 40)]. This too is consistent with the theoretical model used because the scale of quantum interference is set by the inelastic scattering length [ $L_{\text{Th}} \sim (D\tau_i)^{1/2}$ ].

The low-temperature data pertaining to the Zr-Cu system could not be fitted successfully in the same way as has been done for Zr-Ni system. The reason is as yet not clear. One possibility is the onset of hydrogen paramagnetism at low temperatures due to a different configuration that the dopant takes up in Zr-Cu matrix compared to Zr-Ni. Another explanation may be in terms of the Zr-Cu matrix behaving as a two-phase system. A temperature-dependent diffusion coefficient could explain the low-temperature data in Zr-Cu-H system. A similar phenomenon (namely, temperature-dependent fitting parameters) observed in the resistivity of different systems is also indicative of such an effect.<sup>40,41</sup>

The alternative explanation that the observed temperature dependence of the susceptibility, in both Zr-Ni-H and Zr-Cu-H systems, is the "normal" temperature dependence of the Pauli susceptibility due

TABLE IV. The coefficient of fit of magnet susceptibility data to relations (15) and (16).

Sample	$A$ ( $10^{-5} \text{ J T}^{-2} \text{ mol}^{-1} \text{ K}^{-1/2}$ )	$B$ ( $10^{-5} \text{ J T}^{-2} \text{ mol}^{-1} \text{ K}^{-1/2}$ )	$C_2$ ( $10^{-5} \text{ J T}^{-2} \text{ mol}^{-1}$ )	$T_{\text{min}}$ $T_c$ (K) (K)
$Zr_{0.67}Ni_{0.33}$	0.6	4.0	115.5	60 1.5
$Zr_{0.67}Ni_{0.33}H_{0.13}$	0.85	4.4	106.3	80 0.95
$Zr_{0.67}Ni_{0.33}H_{0.33}$	1.15	4.95	91.7	115 0.85

to the temperature-dependent density of states  $[N(1 - \pi^2 k_B T^2 / 12\mu_0)]$  which shows up at low temperatures should be ruled out because it is two orders of magnitude smaller than the observed correction and goes as  $T^2$ . Because this correction is proportional to the density of states at the Fermi level which is lowered (Table III) in hydrogenated samples the "classical" variation of the Pauli susceptibility would be in the opposite direction with increasing hydrogen content. The same could be said for the Stoner enhancement at low temperatures. [Generally,  $I = I_{\text{eff}} N(E_F)$  is supposed to be temperature independent which is actually not true.  $I$  certainly increases at low  $T$ ]. Since  $I$  appears as a phenomenological interaction in the paramagnon theory it is difficult to know its exact temperature dependence. However, the Stoner enhancement is smaller in Zr-Cu than in Zr-Ni system as well as in the doped samples and the observed corrections are greater in those systems. We assume that at least for H concentrations used  $I_{\text{eff}}$  remains substantially unchanged.

### CONCLUSION

We have analyzed the magnetic and transport properties as a function of disorder in Zr-Ni and Zr-Cu metallic glasses. Disorder was increased by doping the samples with hydrogen.

We find that not only the temperature and the magnetic field dependence of the resistivity but also the anomalous temperature dependence of the magnetic susceptibility of some hydrogen doped  $4d$ - $3d$  metallic glasses can be

accounted for using the theoretical models of weak localization and electron-electron interactions.

Measurements in the magnetic field show positive anomalous magnetoresistance which can be interpreted as being due to WL in the presence of strong spin-orbit scattering and Maki-Thompson (MT) fluctuations. It has been found that both the spin-orbit and the MT contributions are strongly suppressed with increasing hydrogen concentration. At the same time hydrogen enhances quasielastic scattering, thus reducing the effective diffusion constant. This, together with the reduced screening of the Coulomb interaction, leads to an increase of the resistivity and a decrease of the superconducting transition temperature upon hydrogenation. The resistivity is found to saturate at the hydrogen concentration  $x \cong 0.5$ , since the mean free path reaches the Ioffe-Regel limit, and so does the superconducting transition temperature indicating that this is the localization effect. Reasonable values of  $\beta$ ,  $\tau_i$ ,  $\tau_{\text{so}}$ ,  $D$ , and  $F$  have been determined from the analysis of the data.

It has been found that the magnetic susceptibility decreases substantially with increasing hydrogen content. This behavior is primarily attributed to a reduction in the density of Zr  $d$  electronic states at the Fermi level. The rise of the magnetic susceptibility at low temperatures was found to be proportional to the hydrogen concentration, that is to the degree of disorder, as a consequence of the slowing down of the spin diffusion.

An extension of the present investigation into the domain of lower temperatures and other  $4d$ - $3d$  systems is presently being pursued.

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