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ELECTRONIC PROPERTIES OF HYDROGEN-DOPED $(Zr_{80}3d_{20})_{1-x}H_x$ (3d = Fe, Co, Ni) METALLIC GLASSES

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Dedicated to Professor Boran Leontić on the occasion of his 70th birthday

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The electrical resistivities of hydrogen-doped $(Zr_{80}3d_{20})_{1-x}H_x$ (3d = Fe, Co, Ni; $x \leq 0.11$) metallic glasses have been measured at temperatures between 2 K and 290 K. The increase of the room-temperature resistivity and its temperature coefficient are explained as a consequence of increased disorder due to hydrogen-doping. The temperature dependence of the resistivity has been analysed using theoretical models of weak-localisation and electron-electron interaction in disordered three-dimensional conductors. The hydrogen dopant is found to reduce effective electron diffusion constant, D, the spin-orbit scattering rate, τ_{so}^{-1}, the superconducting transition temperature, T_c, and broadens the superconducting transition region. We discuss the effects of different hydrogen environments on T_c of $(Zr_{80}3d_{20})_{1-x}H_x$ metallic glasses.

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Keywords: electrical resistivity, metallic glasses, $(Zr_{80}3d_{20})_{1-x}H_x$ (3d = Fe, Co, Ni; $x \leq 0.11$), hydrogen-doped, disordered three-dimensional conductors, electron diffusion constant, spin-orbit scattering, superconducting transition temperature

1. Introduction

Metallic glasses offer an excellent matrix in which to study the behaviour of electrons in highly disordered systems. It has been shown that the diffusive motion of electrons in two-dimensional as well as in three-dimensional disordered systems entails quantum corrections to the resistivity and the magnetoresistivity. There are

two principal sources of quantum corrections, known as weak localisation, WL, [1,2] and Coulomb interaction, CI, [3,4]. Both of these corrections are important when the mean free path becomes short so that electron propagation between scattering events is no longer free-electron-like but diffusive. At low enough temperatures, where the elastic scattering time is a few orders of magnitude shorter than the inelastic scattering time, the quantum corrections arising from the interference of the electronic partial waves are very important. They lead to an anomalous dependence of the resistivity on temperature, sample dimension, and external fields. Although the WL and CI theories have been developed for free electrons, their applicability to transition-element metallic glasses, with dominant d-band conductivity, has been successful.

In the present paper we report on the resistive properties and the effect of increased disorder on T_c resulting from doping $(Zr_{80}3d_{20})_{1-x}H_x$ (3d = Ni, Co, Fe) metallic glasses with hydrogen. $(Zr_{80}3d_{20})$ metallic glasses are characterised by high room-temperature resistivities, they are paramagnetic and become superconducting at temperatures below 4 K. We have demonstrated [5] that hydrogen can be used as an atomic probe to study quantum interference at defects and to gain insight into the way the atomic microenvironment influences the electronic properties of a disordered system.

2. Experimental

Ribbons of $(Zr_{80}3d_{20})$ metallic glass were prepared by rapid solidification of the melt on a single-roll spinning copper wheel in an argon atmosphere. The samples, 1 – 2 cm long, 1.5 mm wide and 25 μm thick were then cut from the ribbon. The hydrogenation was carried out electrolytically. The amount of absorbed hydrogen was determined volumetrically [5,6]. The as-quenched and hydrogenated samples were examined by X-ray diffraction, using CuK radiation to verify that they were amorphous.

The sample resistance was measured by a low-frequency (23.2 Hz) four-probe AC method in the temperature range 2 – 290 K. The precision of these measurements extends to a few parts in 10^6. The samples were mounted on a copper holder and placed in a cryostat. The temperature range from 2 – 40 K was covered using liquid-helium cooling, while, above 40 K and up to 290 K, the measurements were carried out in a conventional Cryodyne Refrigerator System.

3. Theoretical models

In the case of a superconductor, the fluctuating conductivity dominates over the localisation and CI conductivity [7,8] in the vicinity of the superconducting transition temperature, T_c. This excess electrical conductivity consists of two terms:

$$\Delta\sigma = \sigma_{\mathrm{AL}} + \sigma_{\mathrm{MT}}\,, \tag{1}$$

where $\sigma_{\rm AL}$ is the Aslamazov-Larkin, (AL) [7], term which originates from the virtual Cooper pairs created by thermal fluctuations and $\sigma_{\rm MT}$ is the Maki-Thompson, (MT) [8], term coming from the interaction of normal conducting electrons and the superfluid.

In three dimensions, the temperature-dependent quantum correction to the conductivity due to WL in the presence of inelastic, $\tau_{\rm i}^{-1}$, and spin-orbit, $\tau_{\rm so}^{-1}$, scattering is given by Fukuyama and Hoshino [1]:

$$\sigma_{\rm WL} = \frac{e^2}{2\pi^2\hbar}\left[3\left(\frac{1}{D\tau_{\rm so}}+\frac{1}{4D\tau_{\rm i}}\right)^{1/2} - \left(\frac{1}{4D\tau_{\rm i}}\right)^{1/2}\right], \qquad (2)$$

where D is the diffusion coefficient. $\tau_{\rm so}$ is temperature independent and $\tau_i = \alpha_i T^{-p}$, with $2 \leq p \leq 4$ at temperatures $T < \Theta_D$ (Θ_D is the Debye temperature), at which the dominant contribution to the inelastic scattering comes from the electron-phonon interaction.

The correction for the electron-electron interaction in the diffusion and the Cooper channels takes the following form [3,4]:

$$\sigma_{e-e} = \frac{1.3\,e^2}{4\pi^2\hbar}\left(\frac{4}{3} - 3F^* - \frac{2}{\ln(T_0/T)}\right)\left(\frac{kT^{1/2}}{D\hbar}\right)^{1/2}, \qquad (3)$$

where T_0 is equal to T_c, if the alloy is a superconductor. Otherwise, T_0 is taken to be of the order of the Fermi temperature, T_F. $F^* = F - \lambda$, where F is the averaged screened Coulomb potential ($0 \leq F \leq 1$, depending on the screening length) and λ is the electron-phonon coupling constant.

4. Results and discussion

The change in the temperature-dependent electrical resistivity, relative to its value at 290 K, $\Delta\rho/\rho(290K)$, of hydrogen-doped $(Zr_{80}Ni_{20})_{1-x}H_x$ (x = 0, 0.03, 0.05, 0.11) is shown in Fig. 1 for temperatures 3 K< T < 290 K. The temperature coefficients of the resistivity (TCR) of the hydrogen-doped $(Zr_{80}3d_{20})_{1-x}H_x$ (3d = Ni, Co, Fe) metallic glasses are negative in the temperature range 10 – 295 K and their absolute values increase with the hydrogen concentration [9, 10]. The change in the temperature-dependent electrical resistivity, relative to its value at 290 K, $\Delta\rho/\rho(290K)$, of the $(Zr_{80}Ni_{20})_{1-x}H_x$ (x = 0, 0.03, 0.05, 0.11) samples and the fit to the sum of Eq. (2) and Eq. (3) (solid lines) are shown in Fig. 2 as a function of temperature up to 110 K. The temperature-dependent part of the electrical resistivity relative to its value at 4.2 K, $\rho(T)/\rho(4.2K)$, of $(Zr_{80}Ni_{20})_{1-x}H_x$ metallic glasses (x = 0, 0.03, 0.05, 0.11), in the vicinity of T_c is shown in Fig. 3. Doping with hydrogen reduces T_c. The superconducting transition region broadens when hydrogen enters into the Zr-Ni matrix and "steps" appear in the resistance curve at the transition for hydrogen concentration x = 0.11. These steps can be associated with inhomogeneities due to different environments of H-atoms in the matrix.

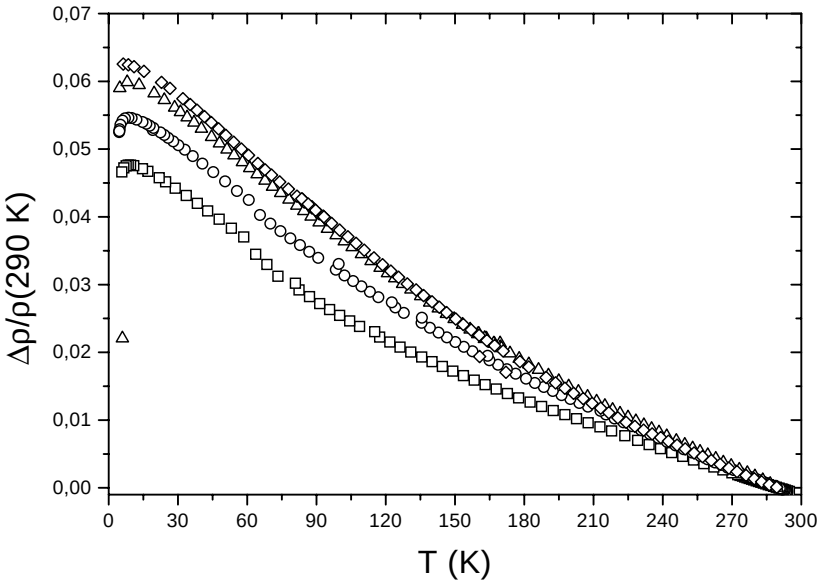

Fig. 1. Change of the temperature-dependent electrical resistivity, relative to its value at 290 K, *of hydrogen doped* $(Zr_{80}Ni_{20})_{1-x}H_x$ *metallic glasses.* $x = 0$ (□), 0.03 (○), 0.05 (△), 0.11 (◇).

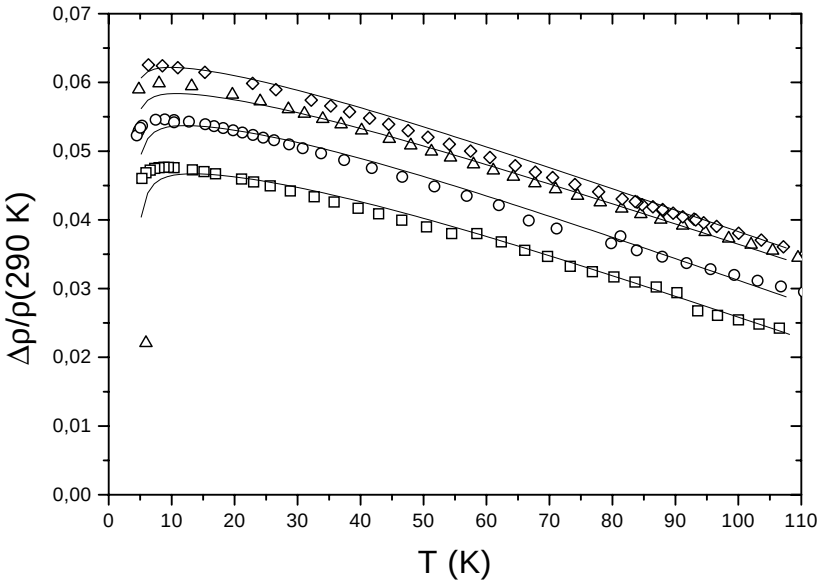

Fig. 2. Change of the temperature-dependent electrical resistivity, relative to its value at 290 K, *of hydrogen doped* $(Zr_{80}Ni_{20})_{1-x}H_x$ *metallic glasses.* $x = 0$ (□), 0.03 (○), 0.05 (△), 0.11 (◇). *The lines are the best fits of the sum of Eqs. (2) and (3).*

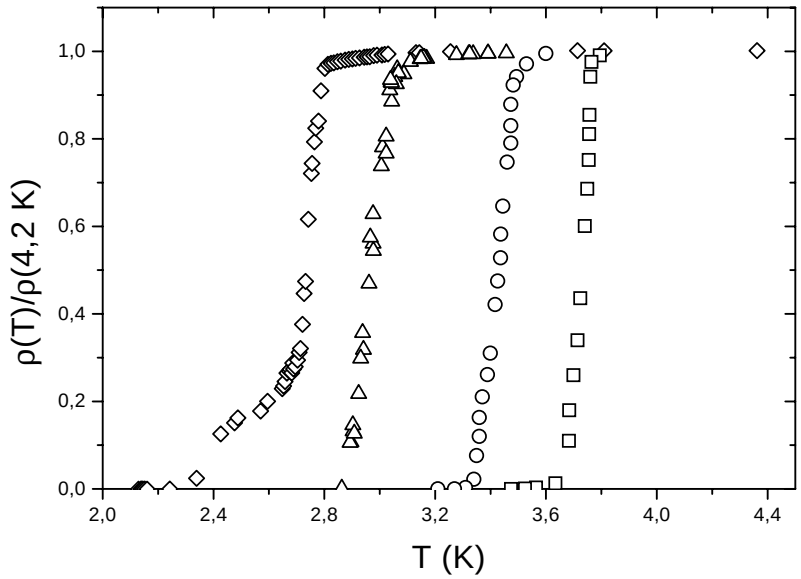

Fig. 3. The temperature-dependent part of the electrical resistivity relative to its value at 4.2 K, $\rho(T)/\rho(4.2\mathrm{K})$, *of* $(Zr_{80}Ni_{20})_{1-x}H_x$ *metallic glasses* $x = 0$ (□), 0.03 (○), 0.05 (△), 0.11 (◇).

According to the structural analyses of hydrogen doped Zr-3d metallic glasses [11] H-atoms tend to occupy preferentially tetrahedral holes (TH) surrounded by four Zr-atoms, while TH defined by three Zr-atoms and one 3d-atom begin to be occupied at higher hydrogen concentration (3d = Co, $x = 0.048$; 3d = Fe, $x = 0.071$ and 3d = Ni, $x = 0.11$). If we compare the results for $(Zr_{80}3d_{20})_{1-x}H_x$ with those in our previous paper for the $(Zr_{67}Co_{33})_{1-x}H_x$, metallic glasses [12], we conclude that in the system with higher zirconium concentration, the "steps" appear at higher hydrogen concentration. Hydrogen atoms migrate to the Zr-rich sites where their s-electron hybridise with Zr d-band. This results in a reduction of the Zr 4d-density of states at E_F. The same effect has been observed in the specific heat measurements at low-temperature of hydrogen-doped Zr-Ni metallic glasses [13]. The value of the superconducting transition temperature of the doped sample relative to the superconducting transition temperature of the undoped sample, $T_{c\,H}/T_{c\,0}$, versus the hydrogen dopant concentration, H (%), of $(Zr_{80}3d_{20})_{1-x}H_x$ (3d = Ni, Co, Fe) metallic glasses is shown in Fig. 4. It can be seen from Fig. 4 that the data of $(Zr_{80}3d_{20})_{1-x}H_x$ (3d = Ni, Co, Fe) metallic glasses fall close to one curve for hydrogen dopant concentrations $x \leq 0.08$. $T_{c\,H}/T_{c\,0}$, versus the change in the resistivity, $(\rho_{\max} - \rho(290\mathrm{K}))/\rho(290\mathrm{K})$, (where $\rho_{\max}$ is the maximal value of the temperature dependent resistivity) is shown in Fig. 5. It can be seen from Fig. 5, that for the undoped samples $(\rho_{\max} - \rho(290\mathrm{K}))/\rho(290\mathrm{K})$ increases with the substitution of Fe with Ni and Co, which indicates that this substitution increases disorder. In addition to this, it is obvious that hydrogen increases disorder in $((Zr_{80}3d_{20})_{1-x}H_x$ (3d = Ni, Co, Fe) metallic glasses. $T_{c\,H}/T_{c\,0}$ decreases linearly

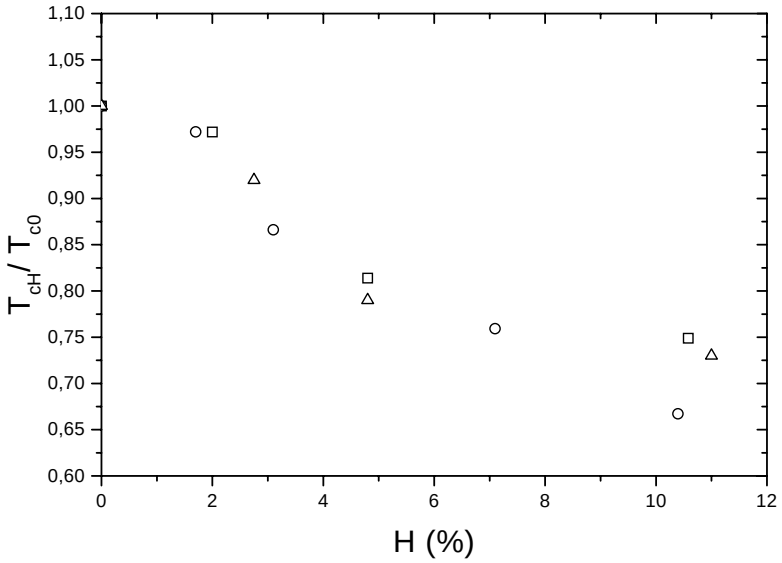

Fig. 4. The value of the superconducting transition temperature of the doped sample relative to the superconducting transition temperature of the undoped sample, $T_{c\,H}/T_{c\,0}$, versus the hydrogen dopant concentration, H *(%), of* $(\mathrm{Zr_{80}3d_{20}})_{1-x}\mathrm{H}_x$, 3d = Ni (△), Co (□), Fe (○) *metallic glasses.*

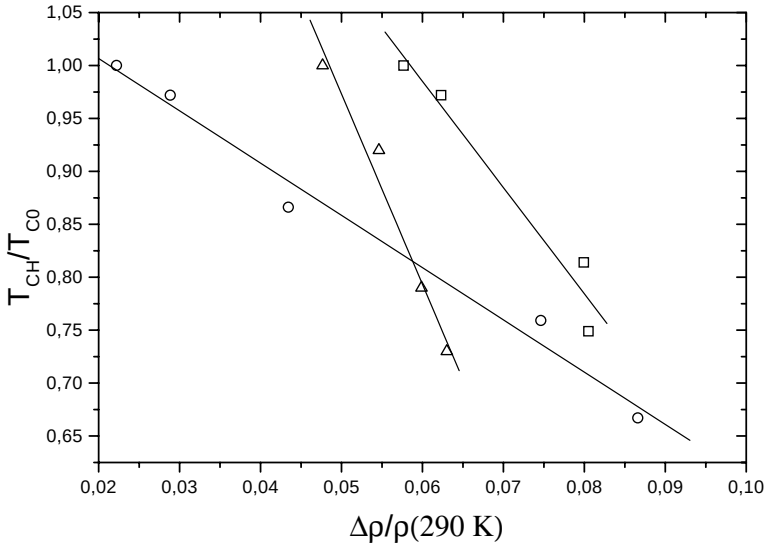

Fig. 5. The value of the superconducting transition temperature of the doped sample relative to the superconducting transition temperature of the undoped sample, $T_{c\,H}/T_{c\,0}$, versus the change in the resistivity, $(\rho_{\max} - \rho(290\mathrm{K}))/\rho(290\mathrm{K})$, (where $\rho_{\max}$ is the maximal value of the temperature dependent resistivity) of $(\mathrm{Zr_{80}3d_{20}})_{1-x}\mathrm{H}_x$, 3d = Ni (△), Co (□), Fe (○) *metallic glasses.*

with $(\rho_{\max} - \rho(290\text{K}))/\rho(290\text{K})$, and the slope of the curve is the smallest for Fe and the largest for Ni. The suppression of T_c with hydrogen-dopant is caused partly by the decrease of $N(E_\text{F})$ and partly by enhancement of the disorder.

It can be seen from Table 1 that the value of T_c is the highest for $(Zr_{80}Co_{20})$ ($T_c = 3.98$ K) whereas it is the smallest for $Zr_{80}Fe_{20}$ ($T_c = 3.28$ K). The substantial decrease of T_c in $Zr_{80}Fe_{20}$ [14] has been explained as due to the influence of spin fluctuations and the formation of localised magnetic moments at the Fe-sites. One important factor to consider in deciding whether a given Fe atom will form a magnetic moment in metallic glasses is the variety of local atomic environments available for the Fe atom to occupy. Even if the average local atomic environment is unfavourable for moment formation, it may still be possible for some Fe atomic sites to satisfy the necessary requirements. Hence the disordered atomic structure

TABLE 1. Values of the hydrogen concentration, x, the diffusion constant, D, the inelastic scattering time, $\tau_\text{i}(4.2\text{ K})$ the spin-orbit scattering time, τ_so, the superconducting transition temperature, T_c, the electrical resistivity, $\rho(290\text{K})$ and the screening parameter F^*.

x	D	$\tau_\text{i}(4.2\text{ K})$	τ_so	T_c	$\rho(290\text{ K})$	F^*
± 0.001	± 0.02	± 0.02	± 0.02	± 0.02	± 1	± 0.03
–	10^{-5} m^2/s	10^{-11} s	10^{-11} s	K	$\mu\Omega$cm	–
$(Zr_{80}Ni_{20})_{1-x}H_x$						
0	3	0.95	3.5	3.72	165	0.1
0.03	2.97	0.96	4.5	3.42	168	0.1
0.05	2.94	1.27	6.5	2.97	173	0.1
0.11	2.85	1.36	8.0	2.72	183	0.1
$(Zr_{80}Co_{20})_{1-x}H_x$						
0	3	0.79	3.5	3.98	170	0.1
0.02	2.95	0.96	4.5	3.87	173	0.03
0.04	2.9	1.03	5.2	3.24	177	0.03
0.11	2.8	1.08	7.5	2.98	188	0.03
$(Zr_{80}Fe_{20})_{1-x}H_x$						
0	3.5	9.5	2	3.28	160	0.55
0.017	3.47	9	2.05	3.19	163	0.35
0.031	3.45	4.8	2.10	2.84	166	0.15
0.071	3.4	4.45	2.15	2.49	174	0.1
0.104	3.3	5.2	2.2	2.19	180	0.05

of metallic glasses enhances the tendency of localised magnetic moment formation and spin-fluctuations. This has been confirmed by magnetic susceptibility measurements [15] which showed a decrease of the valence susceptibility and an increase of T_c upon thermal annealing of $Zr_{80}Fe_{20}$ metallic glass.

The temperature-dependent change of the resistivity in $(Zr_{80}3d_{20})_{1-x}H_x$ consists of two contributions: weak localisation and electron-electron interaction.

The fits to the experimental data (Fig. 2) are derived from the sum of relations (2) and (3) with the inelastic scattering time, $\tau_{\rm i} = \alpha_{\rm i} T^{-2}$, the spin-orbit scattering time, $\tau_{\rm so}$, the diffusion constant, D, and the screening parameter, F^*, used as the fitting parameters. The values of the parameters are given in Table 1. The values of T_c and $\rho(290\ {\rm K})$ are those determined experimentally. T_c is determined as the midway point on the electrical resistivity vs. temperature transition curve. The influence of the changes in the value of parameters to the fitting procedure has been described in our earlier work [5]. Thus, for example, a change of 2 % in only one of the parameters D, $\tau_{\rm i}$ and $\tau_{\rm so}$ gives a fit which deviates considerably from the experimental data. The values of parameters (Table 1) are the same as those obtained from our magnetoresistivity data [9].

It can be seen from Table 1 that the diffusion constant, D, decreases with hydrogen concentration in all systems. The inelastic scattering time, $\tau_{\rm i}$, increases in $(Zr_{80}3d_{20})_{1-x}H_x$ (3d = Ni, Co). If we calculate the inelastic phase coherence length, $L_i = \sqrt{D \cdot \tau_{\rm i}}$, we find that it increases from $L_{\rm i}({\rm Ni}) = 1.69\ 10^{-8}$ m, $L_{\rm i}({\rm Co}) = 1.54\ 10^{-8}$ m in the undoped samples $Zr_{80}3d_{20}$ (3d = Ni, Co) to $L_{\rm i}({\rm Ni}) = 1.97\ 10^{-8}$ m, $L_{\rm i}({\rm Co}) = 1.74\ 10^{-8}$ m in the doped samples $(Zr_{80}3d_{20})_{0.89}H_{0.11}$. This implies that the weak-localisation correction increases with hydrogen, while the electron-electron contribution is constant ($F^*({\rm Ni, Co}) \approx 0.1$ in Table 1). In $(Zr_{80}Fe_{20})_{1-x}H_x$, L_i decreases from $L_{\rm i}({\rm Fe}) = 5.77\ 10^{-8}$ m to $L_{\rm i}({\rm Fe}) = 4.14\ 10^{-8}$ m, with hydrogen doping. The large value of $F^* = 0.55$ in the undoped $Zr_{80}Fe_{20}$ metallic glass indicates the presence of spin-disorder scattering as described above. Our magnetoresistivity results [16] for the undoped $Zr_{68}Fe_{32}$ metallic glass have shown a small negative magnetoresistance for fields lower than 0.4 T at $T = 4.2$ K. This can be taken as evidence of the presence of the spin-disordered scattering. In samples doped with hydrogen, this negative contribution disappears, and the observed magnetoresistance is positive even for the smallest concentration. The hydrogen "freezes out" these localised magnetic moments [16] and thus lowers the value of F^* to 0.05. At the same time, the positive contribution of the electron-electron interaction to the resistivity is lowered, thus the net change of the temperature coefficient of the resistivity is higher and the slope of the curve of $T_{c\,{\rm H}}/T_{c\,0}$, $T_{c\,H}/T_{c\,0}$ versus $(\rho_{\rm max} - \rho(290{\rm K}))/\rho(290{\rm K})$ in Fig. 5 is smaller for $(Zr_{80}Fe_{20})_{1-x}H_x$ than for $((Zr_{80}3d_{20})_{1-x}H_x$ (3d = Ni, Co) metallic glasses.

The spin-orbit scattering time, $\tau_{\rm so}$, is enhanced with increased hydrogen concentration (Table 1). Since most of the spin-orbit scattering takes place on Zr atoms and in the d-band, the reduction of the effective spin-orbit contribution to the resistivity by the dopant can be taken as evidence that hydrogen atoms migrate mainly to the Zr-rich sites. The smaller change of $\tau_{\rm so}$ in Zr-Fe remains to be further investigated by means of magnetoresistivity measurements in high magnetic fields.

The electrical resistivity in hydrogen doped $((Zr_{80}3d_{20})_{1-x}H_x$ metallic glasses increases with hydrogen concentration (Table 1). We have found that sample dimensions remain unchanged during hydrogenation. Thus, the error of the resistivity in Table 1 is in fact the error of the measured resistance and does not include the error due to the geometrical factor. In the case of strong scattering, the electron motion becomes diffusive so that the resistivity is expressed as $\rho = 1/(e^2 N(E_F)D)$. The ultraviolet photoelectron spectroscopy measurements [17] as well as the magnetic susceptibility results [5,18] and specific heat measurements [13] have shown that hydrogen reduces the d-density of states at the Fermi level in Zr-3d metallic glasses. Thus we conclude that the resistivity increase of $(Zr_{80}3d_{20})_{1-x}H_x$ (3d = Fe, Co, Ni) (see Table 1), is partly caused by the decrease of the density of d-states at the Fermi level, $N(E_F)$, and partly by the observed decrease of the electron diffusion constant, D (see Table 1).

5. *Conclusion*

We have analysed the electrical resistivity and superconducting transition temperature as a function of hydrogen concentrations in $(Zr_{80}3d_{20})_{1-x}H_x$ (3d = Fe, Co, Ni) metallic glasses.

The increase of the room-temperature resistivity and its temperature coefficient is explained as a consequence of an increased disorder due to the hydrogen-doping.

The resistivity increase of $(Zr_{80}3d_{20})_{1-x}H_x$ is partly caused by the decrease of the density of d-states at the Fermi level, $N(E_F)$, and partly by the observed decrease of the electron diffusion constant, D (see Table 1).

The temperature dependence of resistivity has been analysed using the theoretical models of weak localisation and electron-electron interaction in disordered three-dimensional conductors. The spin-orbit scattering rate, τ_{so}^{-1} and the diffusion coefficient, D, decrease whereas the behaviour of the inelastic scattering time, τ_i, and of the screening parameter, F^*, is system specific.

The hydrogen dopant lowers T_c and broadens the superconducting transition region in resistivity. The effect of hydrogen on the lowering of T_c is through the decrease of the Zr 4d-density of states at the Fermi level and enhancement of the disorder.

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ELECTRONSKA SVOJSTVA HIDROGENIZIRANIH METALNIH STAKALA $(Zr_{80}3d_{20})_{1-x}H_x$ (3d = Fe, Co, Ni)

Mjerili smo električnu otpornost hidrogeniziranih metalnih stakala $(Zr_{80}3d_{20})_{1-x}H_x$ (3d = Fe, Co, Ni; $x \leq 0.11$) na temperaturama između 2 K i 290 K. Porast otpornosti na sobnoj temperaturi i njen temperaturni koeficijent tumače se kao posljedica povećanog nereda zbog dodavanja vodikovih atoma. Temperaturna ovisnost otpornosti se analizira primjenom teorijskih modela slabe lokalizacije i interakcije među elektronima u neuređenim trodimenzijskim vodičima. Našli smo da hidrogenizacija smanjuje konstantu djelotvorne elektronske difuzije, D, učestalost raspršenja spin-staza, τ_{so}^{-1}, temperaturu supravodljivog prijelaza, T_c, i širi područje supravodljivog prijelaza. Raspravljamo o učinku različitih vodikovih okolina na T_c u metalnim staklima $(Zr_{80}3d_{20})_{1-x}H_x$.