Hydrogen-induced changes in magnetic susceptibility of (Zr68Fe32)1-xHx metallic glasses

Kokanović, Ivan; Leontić, Boran; Lukatela, Jagoda

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Hydrogen-induced changes in magnetic susceptibility of $(Zr_{68}Fe_{32})_{1-x}H_x$ metallic glasses

I. Kokanović and B. Leontić
Department of Physics, Faculty of Science, University of Zagreb, Zagreb, Croatia

J. Lukatela
Institute of Physics, P.O. Box 304, Zagreb, Croatia
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The magnetization of hydrogen-doped $(Zr_{68}Fe_{32})_{1-x}H_x$ metallic glasses has been measured in the temperature range 1.7–100 K for various dopant concentrations. For hydrogen concentrations $x<0.1$ the samples are paramagnetic with magnetic susceptibilities that are only weakly temperature dependent down to about 35 K, below which a slight increase can be observed. For larger hydrogen concentrations the magnetic susceptibilities become strongly temperature dependent (and show Curie-Weiss behavior). It is found that $\chi(100\ \mathrm{K})$ increases upon hydrogenation. This is explained by an increase in the contribution of the Fe $3d$-electronic states to the density of states at the Fermi level, due to Zr-H bond formation, which leads to an enhancement of spin fluctuations and the formation of magnetic moments on the Fe-atom site at hydrogen concentrations $x>0.1$. The form and magnitude of the observed temperature dependence of the magnetic susceptibility are well accounted for by the sum of the Curie-Weiss term and quantum corrections to the susceptibility.
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I. INTRODUCTION

Zr-Fe amorphous alloys can be formed over a wide composition range and thus provide a means for the study of their properties as a function of compositions. The alloys exhibit superconductivity in Zr-rich compositions,[1] but form various magnetic phases in some Fe-rich compositions.[2] The interplay between the superconductivity and magnetism has been clearly demonstrated (Refs. 1 and 2), the superconducting transition temperature being reduced by the presence of spin fluctuations. In Zr_xFe_{1-x} superconductivity is destroyed for concentrations $x<0.71$. $Zr_{68}Fe_{32}$ metallic glass is a good matrix for examining spin fluctuations and the formation of magnetic moments in a highly disordered system, since the level of spin fluctuations and the formation of magnetic moments can be varied by hydrogen doping in varying concentrations. The system is characterized by a high room-temperature resistivity ($\rho\approx173\ \mu\Omega\ \mathrm{cm}$) and is not a superconductor, although it has a higher density of states at the Fermi level compared with other $Zr_{68}3d_{32}$ (Cu, Ni, Co), metallic glasses.

Our previous results[3] show that in the $Zr_{68}Fe_{32}$ metallic glass hydrogen doping produces a positive anomaly in the temperature dependence of the electrical resistivity with a maximum around 25 K. Above that temperature, the curve resumes a monotonic decrease with temperature and the temperature coefficient of the resistivity is enhanced by the hydrogen dopant. The magnetoresistivity results[4] of $Zr_{68}Fe_{32}$ metallic glass doped with hydrogen show positive anomalous values which increase with the hydrogen concentration. The magnetoresistivity enhancement and the positive anomaly in the temperature dependence of the electrical resistivity together with its maximum in $(Zr_{68}Fe_{32})_{1-x}H_x$ systems have been attributed to the increase of the Stoner factor $(1\text{-}I)^{-1}$ and of the spin-scattering rate[3,4] τ_s^{-1} due to the enhancement of the spin fluctuations with hydrogen.

In this paper we present the results and a detailed quantitative analysis for magnetic susceptibilities of the $(Zr_{68}Fe_{32})_{1-x}H_x$ metallic glasses. We compare these with our magnetic susceptibility results for $(Zr_{67}Ni_{33})_{1-x}H_x$ (Ref. 5) and $(Zr_{67}Co_{33})_{1-x}H_x$ (Ref. 6), which also show a strong influence of the hydrogen dopant on the magnetic properties and electronic structure in Zr $3d$ ($3d$ Ni, Co) metallic glasses. For instance, the room-temperature magnetic susceptibilities of hydrogen-doped Zr-Ni and Zr-Co (and, hence, the density of states at E_F) decrease with increasing hydrogen concentration.

II. EXPERIMENTAL METHODS

Ribbons of $Zr_{68}Fe_{32}$ metallic glass were prepared by rapid solidification of the melt on a single-roll spinning copper wheel in an argon atmosphere. The samples cut from the ribbons were 5–8 mm long, 1.7–2.2 mm wide, and 25–30 μm thick. The hydrogenation was carried out electrolytically. The hydrogen concentrations were determined volumetrically using a McLeod manometer.

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

The magnetic susceptibility was measured in the temperature range 1.7–100 K using Quantum Design's Magnetic Property Measurement System which uses a superconducting quantum interference device (SQUID) amplifier as a sensitive magnetic field detector. It is capable of resolving variations in magnetic moments as small as $10^{-11}\ \mathrm{J\,T^{-1}}$.

III. RESULTS AND DISCUSSION

The change in the electrical resistivities of $(Zr_{68}Fe_{32})_{1-x}H_x$ samples relative to the resistivity of the undoped alloy at 273 K, $\Delta\rho/\rho(273\ \mathrm{K})$, vs the hydrogen concen-

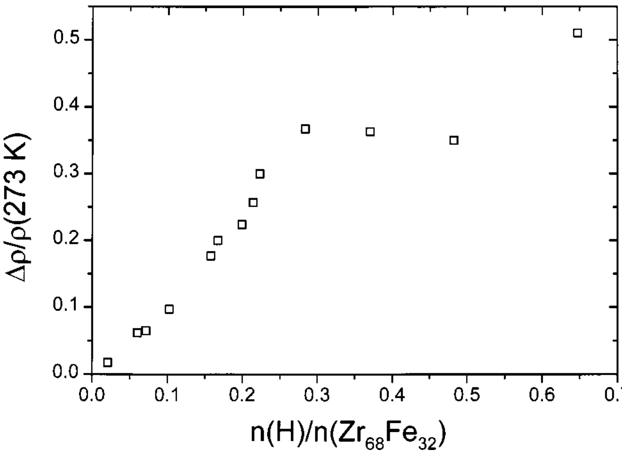

FIG. 1. Change in the electrical resistivity of hydrogen-doped $(Zr_{68}Fe_{32})_{1-x}H_x$ relative to the resistivity of the undoped sample at 273 K, $\Delta\rho/\rho$(273 K), vs the hydrogen concentrations $n(H)/n(Zr_{68}Fe_{32})$.

trations $n(H)/n(Zr_{68}Fe_{32})$, determined volumetrically, are shown in Fig. 1. The relationship between the $\Delta\rho/\rho$(273 K) and the hydrogen concentration in the sample is linear for $n(H)/n(Zr_{68}Fe_{32})<0.13$ and then saturates at $\Delta\rho/\rho(Zr_{68}Fe_{32})\approx 0.35$ for $0.28<n(H)/n(Zr_{68}Fe_{32})<0.48$. For larger hydrogen content the $\Delta\rho/\rho(Zr_{68}Fe_{32})$ increases again.

The intensity of first broad diffraction halo in XRD spectra of $(Zr_{68}Fe_{32})_{1-x}H_x$ metallic glasses ($x=0$, 0.18, 0.32, 0.40) vs the scattering angle 2α is shown in Fig. 2. The position of the first maximum is shifted to smaller values of 2α with increasing hydrogen concentration. Thus the nearest-neighbor distance increases from $r=(0.245\pm 0.001)$ nm in $Zr_{68}Fe_{32}$ to $r=(0.257\pm 0.001)$ nm in $(Zr_{68}Fe_{32})_{0.60}H_{0.40}$. These values correspond to the Zr-Fe nearest-neighbor distance.[7]

The measured magnetizations of $(Zr_{68}Fe_{32})_{0.60}H_{0.40}$ metallic glass vs magnetic field at different temperatures ($T=1.8$, 5, 10, 20, 40, 60, 80, and 100 K) are shown in Fig. 3. The magnetization is linear with magnetic field up to 3 T for temperatures $T>10$ K. The linear behavior of the magnetization vs magnetic field was obtained for all measured samples of $(Zr_{68}Fe_{32})_{1-x}H_x$ metallic glasses ($x=0$, 0.05, 0.07, 0.10, 0.18, 0.32, 0.40). The values of the magnetic susceptibilities were determined from the linear part of the measured magnetization in the magnetic fields up to 2 T. The magnetic susceptibilities of $(Zr_{68}Fe_{32})_{1-x}H_x$ metallic glasses vs temperature below 100 K are shown in Fig. 4. The solid lines are the best fits of the experimental data to Eq. (5). The systems are paramagnetic, with magnetic susceptibilities that are only weakly temperature dependent down to about 35 K for hydrogen concentration $x<0.1$, below which a small increase is observed, whereas for larger hydrogen concentrations the magnetic susceptibilities are strongly temperature dependent. The values of the magnetic susceptibility at 100 K increase with hydrogen concentrations. At 100 K, $\chi_{\rm expt}=(323\pm 1)\times 10^{-5}\,{\rm J\,T^{-2}\,mol^{-1}}$ for $Zr_{68}Fe_{32}$, whereas for $(Zr_{68}Fe_{32})_{0.60}H_{0.40}$, $\chi_{\rm expt}=(824\pm 1)\times 10^{-5}\,{\rm J\,T^{-2}\,mol^{-1}}$ (where ''mol'' refers to 1 mol of metallic atoms, 68% Zr and 32% Fe) (Table I).

The experimental magnetic susceptibility is given as

$$\chi_{\rm expt}=\chi_P^s+\chi_P^d+\chi_{\rm ion}+\chi_{\rm orb}+\chi_{\rm CW}, \tag{1}$$

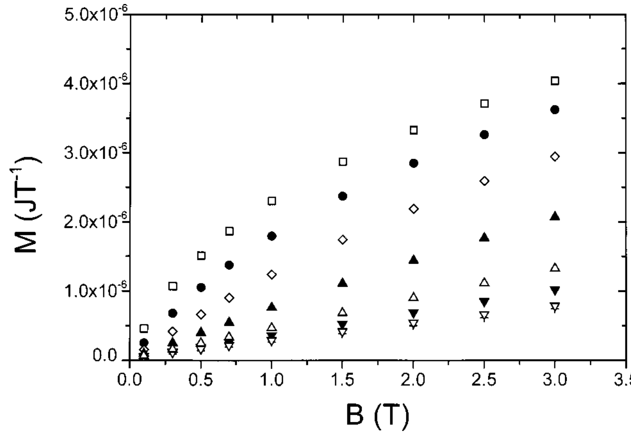

FIG. 3. Measured magnetizations of $(Zr_{68}Fe_{32})_{0.60}H_{0.40}$ metallic glass vs magnetic field at different temperatures [$T=1.8$ K (□), 5 K (●), 10 K (◇), 20 K (▲), 40 K (△), 60 K (▼), 80 K (▽), and 100 K (+)].

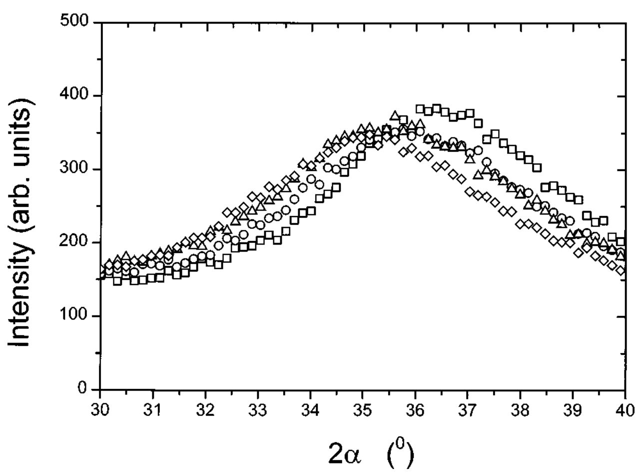

FIG. 2. First broad diffraction halo in XRD spectra of $(Zr_{68}Fe_{32})_{1-x}H_x$ metallic glasses [$x=0$ (□), 0.18 (○), 0.32 (△), and 0.40 (◇)] vs the scattering angle 2α.

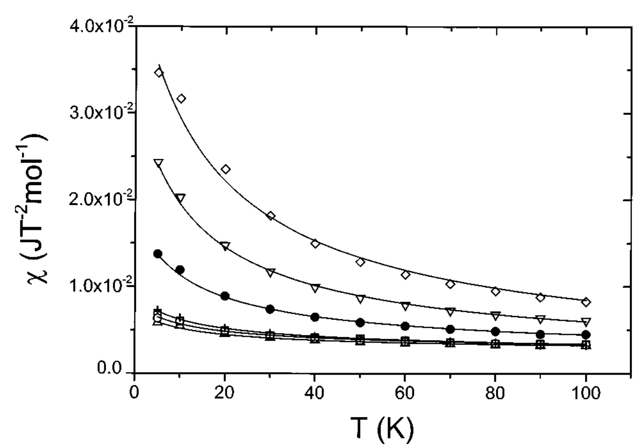

FIG. 4. Magnetic susceptibilities of $(Zr_{68}Fe_{32})_{1-x}H_x$ metallic glasses vs temperature below 100 K [$x=0$ (△), 0.05 (○), 0.07 (□), 0.1 (+), 0.18 (●), 0.32 (▽), and 0.40 (◇)].

TABLE I. Magnetic susceptibility data for $(Zr_{68}Fe_{32})_{1-x}H_x$ metallic glasses. The experimental magnetic susceptibility χ_{expt}, the Curie-Weiss susceptibility χ_{CW}, the Pauli spin susceptibility $\chi_0^P=\mu_B^2\, N_0^d(E_F)$, the enhanced Pauli spin susceptibility χ_{el}^P (where ''mol'' refers to 1 mol of metallic atoms, 68% Zr and 32% Fe), the Stoner factor $(1\text{-}I)^{-1}$, and the electronic density of states at the Fermi level $N_0(E_F)$.

Sample $(Zr_{68}Fe_{32})_{1-x}H_x$ x ± 0.01	χ_{expt} $\pm 1\times10^{-5}$ $(J\,T^{-2}\,mol^{-1})$	χ_{CW} $\pm 0.5\times10^{-5}$ $(J\,T^{-2}\,mol^{-1})$	χ_0^P $\pm 0.5\times10^{-5}$ $(J\,T^{-2}\,mol^{-1})$	χ_e^P $\pm 0.5\times10^{-5}$ $(J\,T^{-2}\,mol^{-1})$	$(1-I)^{-1}$ ± 0.05	$N_0(E_F)$ ± 0.05 (states/eV at.)
0	323×10^{-5}	40.5×10^{-5}	53.1×10^{-5}	181.6×10^{-5}	3.4	1.6
0.05	335×10^{-5}	45.0×10^{-5}	51.4×10^{-5}	192.6×10^{-5}	3.74	1.55
0.07	334×10^{-5}	50.4×10^{-5}	51.1×10^{-5}	186.6×10^{-5}	3.65	1.54
0.10	329×10^{-5}	54.1×10^{-5}	50.1×10^{-5}	182.6×10^{-5}	3.64	1.51
0.18	449×10^{-5}	152×10^{-5}	46.8×10^{-5}	191.6×10^{-5}	4.09	1.41
0.32	606×10^{-5}	323×10^{-5}	42.5×10^{-5}	171.6×10^{-5}	4.03	1.28
0.40	824×10^{-5}	548×10^{-5}	39.8×10^{-5}	157.6×10^{-5}	3.95	1.2

where χ_{ion} is the ionic-core diamagnetism, χ_{orb} is the orbital paramagnetism with the Landau contribution included, and χ_{CW} is the Curie-Weiss susceptibility, $\chi_{CW}=C_1/(T+\Theta)$. The Pauli paramagnetism of s electrons is

$$\chi_P^s=\mu_B^2 N_0^s(E_F), \tag{2}$$

where μ_B is the Bohr magneton and $N_0^s(E_F)$ is the bare density of states of s electrons at the Fermi level. The Pauli paramagnetism of the d band, χ_P^d, which is enhanced over the free-electron value due to the Stoner exchange interaction, is given by

$$\chi_P^d=\frac{\mu_B^2 N_0^d(E_F)}{1-I_{eff}N_0^d(E_F)}, \tag{3}$$

where I_{eff} is the effective exchange integral within the d band and $N_0^d(E_F)$ is the bare density of states of d electrons at the Fermi level.

Since the dominant contribution to the electronic density of states at the Fermi level in the transition metals comes from the d electrons, χ_P^s is an order of magnitude smaller than the values of χ_P^d. The core susceptibilities of the two elements are small ($\chi_{ion}=-20\times10^{-5}\,J\,T^{-2}\,mol^{-1}$ for Zr and $\chi_{ion}=-29\times10^{-5}\,J\,T^{-2}\,mol^{-1}$ for Fe), and we estimate $\chi_{ion}=-23\times10^{-5}\,J\,T^{-2}\,mol^{-1}$ of the $(Zr_{68}Fe_{32})_{1-x}H_x$. The orbital magnetic moments of the electrons are not completely quenched for partly filled degenerate bands, and their contribution to the paramagnetic susceptibility is estimated from the values[8] $\chi_{orb}(Zr)\approx150\times10^{-5}\,J\,T^{-2}\,mol^{-1}$ and $\chi_{orb}(Fe)\approx73\times10^{-5}\,J\,T^{-2}\,mol^{-1}$. Thus the value of the orbital susceptibility for the $Zr_{68}Fe_{32}$ system is estimated to be $\chi_{orb}(Zr_{68}Fe_{32})=125.4\times10^{-5}\,J\,T^{-2}\,mol^{-1}$. The Curie-Weiss susceptibility $\chi_{CW}=C_1/(T+\Theta)$ was calculated using the parameters of the fit C_1 and Θ given in Table II. Subtraction of χ_{ion}, χ_{orb}, and χ_{CW} from χ_{expt} gives the Pauli spin susceptibility $\chi_P=(181.6\pm0.5)\times10^{-5}\,J\,T^{-2}\,mol^{-1}$ for the $Zr_{68}Fe_{32}$ and $\chi_P=(157.6\pm0.5)\times10^{-5}\,J\,T^{-2}\,mol^{-1}$ for the $(Zr_{68}Fe_{32})_{0.60}H_{0.40}$.

A summary of measured and calculated values is given in Table I. The enhancement of the $\chi_{expt}(100\,K)$ magnetic susceptibility upon hydrogenation can be explained as due to the influence of hydrogen on the formation of localized magnetic moments, the enhancement of spin fluctuations, the electronic density of states at the Fermi level, and/or its possible effect on the orbital moments. The conditions for forming a localized moment are that the intra-atomic Coulomb energy is greater than the width of the $3d$ subband and that the position of the centroid of the $3d$ element must be less than a bandwidth away from E_F.[9]

The UPS data,[10] soft-x-ray spectroscopy (SXES) measurements,[11] and band-structure calculations[12] 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). The valence-band spectra of Zr $3d$ metallic glasses are characterized by a varying d-band splitting and the shift of the d states of the $3d$ metal to higher binding energies on alloying. Replacing Cu by Ni, Co, and Fe, i.e., going to the left in the first series of transition metals, the separation of the two band peaks decreases and the contribution from the $3d$ metal increases.

Since the dopant atoms migrate mainly to the Zr-rich sites where their s electrons hybridize with the Zr d band, they are expected to influence significantly the electronic density of states and hence the Pauli susceptibility. On the other hand, we have assumed that the hydrogen-electron hybridization with the Zr d band does not influence greatly the orbital paramagnetism. The value of the Stoner factor $(1-I)^{-1}=3.4$ for the undoped $Zr_{68}Fe_{32}$ sample is taken from our magnetoresistivity results (Table 1 and Eq. 1 in Ref. 4). We have calculated $N_0(E_F)$ for the doped samples from Eq. (3) with χ_{ion}, χ_{orb}, and the Stoner factor as described above. The obtained values of the electronic density of states at the Fermi level (Table I) decreases as the hydrogen concentration increases: $N_0(E_F)=(1.6\pm0.05)$ states eV^{-1} at.$^{-1}$ for $Zr_{68}Fe_{32}$ and $N_0(E_F)=(1.2\pm0.05)$ states eV^{-1} at.$^{-1}$ for $(Zr_{68}Fe_{32})_{0.60}H_{0.40}$. At the same time the values of the Stoner factor $(1-I)^{-1}$ increase with increasing hydrogen concentrations (Table I) due to the enhancement of spin fluctuations. As a result of these two opposing contributions, the Pauli susceptibility χ_{el}^p remains nearly constant (Table I). This is different from the $(Zr_{67}Ni_{33})_{1-x}H_x$ (Ref. 5) and $(Zr_{67}Co_{33})_{1-x}H_x$ (Ref. 6) where hydrogen reduces the χ_{el}^p through a decrease of both $N_0(E_F)$ and $(1-I)^{-1}$. The corrections to the magnetic susceptibility in a nonsuperconduct-

TABLE II. A, B, C_1, and Θ are the coefficients of the fit of the experimental data to the Eq. (5), and r is the nearest-neighbor distance.

Sample $(Zr_{68}Fe_{32})_{1-x}H_x$ $x\pm0.01$	A ±0.5 $(10^{-5}\,\mathrm{J\,T^{-2}\,mol^{-1}\,K^{-1/2}})$	B ±3 $(10^{-5}\,\mathrm{J\,T^{-2}\,mol^{-1}})$	C_1 ±1 $(10^{-3}\,\mathrm{J\,T^{-2}\,mol^{-1}\,K})$	Θ ±1 (K)	r ±0.001 (nm)
0	3	314	45	11	0.245
0.05	5	345	50	11	0.245
0.07	6	349	54	11	0.245
0.10	7	355	60	11	0.249
0.18	9	384	170	12	0.251
0.32	13	404	365	13	0.255
0.40	17	430	680	16.5	0.257

ing disordered system that are weakly temperature dependent arise from the spin splitting. It has been shown[13] that the interplay of the disorder and the electron-electron interactions results in the spin-diffusion constant being suppressed, which in turn leads to an enhancement of the spin susceptibility at low temperatures.

The quantum correction to the spin susceptibility in the diffusion channel $\Delta\chi_s^d$ in a three-dimensional system is given as[14]

$$\Delta\chi_s^d(T)=\lambda^{j=1}\frac{\zeta(1/2)(g\mu_B)^2\sqrt{k_BT}}{16\sqrt{2}\sqrt{(\pi Dh)^3}}, \tag{4}$$

where $\lambda^{j=1}$ is a dimensionless constant for the electron-hole interaction with $j=1$ ($\lambda^{j=1}<0$ for a repulsive interaction) and D is the diffusion constant. This correction is enhanced for transition metals by the Stoner factor.

We have fitted the temperature-dependent magnetic susceptibility to the relation

$$\chi=\frac{C_1}{(T+\Theta)}-A\sqrt{T}+B, \tag{5}$$

where the first term on the right-hand side is the Curie-Weiss susceptibility and the second term is a correction due to the spin-splitting effect in the diffusion channel. The solid lines in Fig. 4 represent the fit of the experimental data to the Eq. (5).

The values of the parameters of the fit, A, B, C_1, and Θ, are given in Table II. The best fit gives the values of the parameters $A=(3\pm0.5)\times10^{-5}\,\mathrm{J\,T^{-2}\,mol^{-1}\,K^{-1/2}}$ for the undoped sample, whereas for $(Zr_{68}Fe_{32})_{0.60}H_{0.40}$, $A=(17\pm0.5)\times10^{-5}\,\mathrm{J\,T^{-2}\,mol^{-1}\,K^{-1/2}}$. The parameter A increases upon hydrogenation (Table II) through the lowering of the diffusion constant[4] and the increase of the Stoner factor. The enhancement of the spin susceptibility upon hydrogenation is in agreement with the magnetoresistivity data[4] which show that hydrogen reduces the spin-orbital scattering rate τ_{so}^{-1}, thus reducing the mixing of spin-up and spin-down subbands. 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 magnetoresistivity by the dopant can be taken as evidence that hydrogen atoms migrate mainly to the Zr-rich sites. The strongly temperature-dependent magnetic susceptibility, Fig. 4, and nonlinear behavior of the magnetization, Fig. 3, of $(Zr_{68}Fe_{32})_{1-x}H_x$ metallic glasses for hydrogen concentrations $x>0.1$ and temperatures $T<10\,\mathrm{K}$ are evidence for the existence of localized magnetic moments on the Fe atoms. 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 unfavorable for moment formation, it may still be possible for some Fe atomic sites to satisfy the necessary requirements; hence, the disordered atomic structure of metallic glasses enhances the tendency of localized magnetic moment formation and spin fluctuations. When Zr is alloyed with Fe to form $Zr_{68}Fe_{32}$ metallic glass, the outer electrons of the Zr atoms will hybridize with the $3d$ electrons of the Fe atoms. As shown by Friedel,[15] such hybridization leads to a reduction of the intra-atomic Coulomb interaction between the $3d$ electrons and hence to a reduction in $3d$-band splitting. The best fit yields $C_1=(45\pm1)\times10^{-3}\,\mathrm{J\,T^{-2}\,mol^{-1}\,K}$ for the undoped sample, whereas for $(Zr_{68}Fe_{32})_{0.60}H_{0.40}$, $C_1=(680\pm1)\times10^{-3}\,\mathrm{J\,T^{-2}\,mol^{-1}\,K}$. The increase of C_1 upon hydrogenation (Table II) has been interpreted in terms of an increase in the $3d$-band splitting due to a reduction of contact between the Fe atoms and the neighboring Zr atoms by hydrogen absorption [Zr-Fe distance r increases (Table II)]. These considerations are consistent with structural analyses[16] which reveal that hydrogen atoms preferentially occupy tetrahedral spaces, inherent in a metallic glass structure, surrounded by four Zr atoms. The sites defined by three Zr and one Fe atom are characterized by higher internal energy because Fe atoms are smaller than Zr ones, and therefore their negative contribution to the crystal field at the tetrahedral site will be smaller. That is why these sites begin to be occupied by hydrogen atoms only at higher dopant concentrations and the formation of magnetic moments on Fe sites becomes possible. The increase of the Curie-Weiss temperature for hydrogen concentration $x=0.4$ is not as large as one would expect from the increase of the parameter C_1. It is due to the fact that the distance between the hydrogen-induced magnetic moments on Fe sites is larger than the Fe-Fe nearest-neighbor distance of the undoped sample and the exchange interaction is screened by hydrogen. Hydrogen doping seems thus to influence significantly the collective behavior of magnetic moments located at Fe sites when compared to the situation in an undoped matrix. This has also been observed in our magnetoresistivity measurements[17] where even a small amount of hydrogen concentration ($x=0.017$) changes a small negative magnetoresistivity of the undoped sample to positive one for magnetic fields lower than 0.4 T. The mechanism of this behavior remains to be further investigated.

IV. CONCLUSION

We have analyzed the magnetic susceptibility data as a function of hydrogen concentration in $(Zr_{68}Fe_{32})_{1-x}H_x$ metallic glass. The system is paramagnetic with magnetic susceptibilities that are only weakly temperature dependent down to about 35 K and hydrogen concentration $x<0.1$, below which a slight increase can be observed. For larger concentrations, the magnetic susceptibilities are strongly temperature dependent and show the Curie-Weiss behavior. It has been found that $\chi(100\text{ K})$ increases with the increase of hydrogen content. This behavior is primarily attributed to the increase of the contribution of the Fe $3d$-electronic states to the density of states at the Fermi level and to the formation of spin fluctuations and magnetic moments on the Fe site for hydrogen concentrations $x>0.1$. The observed increase of the magnetic susceptibility upon hydrogenation can be wholly attributed to the increase in the Curie-Weiss susceptibility χ_{CW}.

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