Spin fluctuations in metallic glasses $Zr_{75}(Ni_xFe_{1-x})_{25}$ at ambient and higher pressures

F. Hamed, F.S. Razavi, S.K. Bose, and T. Startseva

Department of Physics, Brock University, St. Catharines, Ontario, Canada L2S 3A1

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The superconducting properties of metallic glasses $Zr_{75}(Ni_xFe_{1-x})_{25}$ ($x = 0, 0.25, 0.50, 0.75, 1.0$) were studied both at ambient pressure and under quasi-hydrostatic pressure up to 8 GPa (80 kbar). To obtain information on the spin fluctuation parameter λ_{SF} the following measurements were carried out: the dc magnetic susceptibility, the upper critical field at the ambient pressure superconducting transition temperature $T_c(P{=}1$ bar), mass density, and room temperature resistivity. From the pressure dependence of the superconducting transition temperature $T_c(P)$ in $Cu_{25}Zr_{75}$ glass, we estimate the volume derivative of the electron-phonon (e-ph) coupling parameter $\lambda_{e\text{-ph}}$ in this alloy using the McMillan equation. This is used as a reference value for the volume derivative of $\lambda_{e\text{-ph}}$ in the $Zr_{75}(Ni_xFe_{1-x})_{25}$ alloys. Using the modified McMillan equation, which includes spin fluctuation effects and the above reference value for the volume derivative of the e-ph coupling parameter, we determine $\lambda_{e\text{-ph}}$, the electron-spin fluctuation coupling parameter λ_{SF} and the rate of suppression of spin fluctuations with the pressure ($d \ln \lambda_{SF}/d \ln V$) as a function of Fe concentration in these alloys. Both spin fluctuations and the rate of suppression of spin fluctuations with pressure ($d \ln \lambda_{SF}/d \ln V$) are found to increase with higher Fe concentration in these alloys.

I. INTRODUCTION

Systematic variations of physical conditions such as spin fluctuations can be achieved in amorphous alloys more easily than in crystals, by gradually varying the concentration of various components.[1] Fe- and Ni-based zirconium amorphous alloys exhibit superconductivity as well as ferromagnetism depending on the concentration of the magnetic components. The superconducting transition temperature T_c in $Fe_{100-x}Zr_x$ and $Ni_{100-x}Zr_x$ glasses decreases with increasing concentration of Fe and Ni.[1,2] The decrease in T_c has been attributed to spin fluctuations.[2,3] It has been shown that spin fluctuations are suppressed under pressure.[4] The effect of quasihydrostatic pressures up to 8 GPa (80 kbar) on T_c in both $Ni_{100-x}Zr_x$ and $Fe_{100-x}Zr_x$ alloys has been reported previously.[4,5] Mahini *et al.*[5] observed that T_c increases as a function of pressure for $Ni_{100-x}Zr_x$ alloys. This increase can be accounted for by the change in the Fermi level density of states (DOS) $N(E_f)$ and, consequently, the change in the electron-phonon mass enhancement parameter $\lambda_{e\text{-ph}}$. Hamed *et al.*[6] reported a rapid increase of T_c in $Fe_{20}Zr_{80}$ and $Fe_{25}Zr_{75}$ alloys with pressure. They observed that $T_c(P)$ increases at a faster rate in $Fe_{25}Zr_{75}$ than in $Fe_{20}Zr_{80}$ alloy. Their results were interpreted as a consequence of suppression of spin fluctuations under high pressure.

In this study we extract, from measured T_c and $T_c(P)$, the spin fluctuation parameter and the effect of pressure on spin fluctuation in $Zr_{75}(Ni_xFe_{1-x})_{25}$ alloys. Our goal is to examine the effect of a systematic increase in Fe concenteration on both spin fluctuations and their pressure dependence keeping the Zr concentration constant. The remainder of this paper is organized as follows. In Sec. II we briefly describe the experimental procedure. In Sec. III A we present our results for T_c as a function of pressure and concentrations of Ni and Fe. Based on these results and the modified McMillan equation[7] we derive values of the spin fluctuation coupling parameter and its volume dependence as a function of Fe concentration. The values of the spin fluctuation coupling parameter and its volume derivatives, other physical parameters extracted from experiments, and a summary of our results and conclusions are presented in Sec. III B.

II. EXPERIMENTAL SETUP AND PROCEDURE

Samples of amorphous ribbon $Zr_{75}(Ni_xFe_{1-x})_{25}$ were prepared from high purity (99.999%) metallic elements, using the melt spinning technique and then characterized by x-ray diffraction. The mass density of the samples was measured using the Archimedes method with toluene being used as the liquid medium.

The resistivity was measured under quasihydrostatic pressure. The quasihydrostatic pressure cell consisted of a pyrophyllite gasket with soap stone as the pressure transmitting medium.[7] The $T_c(P)$ of lead was used as internal pressure manometer where the Pb manometer was situated near the sample and the $T_c(P)$ of both were measured simultaneously. The temperature was monitored by a calibrated germanium thermometer. A four-point dc-resistivity technique was employed to measure T_c, at midpoint of resistive transition, in the quasihydrostatic cell. The temperature dependence of the upper critical field was measured up to to the field of 5.5 T and temperatures as low as 1.7 K, using a commercial SQUID magnetometer.

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III. RESULTS AND DISCUSSION

A. Experimental results

The measured dc-magnetic suceptibility of $Zr_{75}(Ni_xFe_{1-x})_{25}$ ($x = 0.25, 0.50, 0.75$) showed very weak temperature dependence. As previously reported for Zr-based amorphous alloys,[2] we also observed no evidence of a paramagnetic susceptibility due to magnetic impurities in these three alloys. In the paramagnetic region the measured susceptibility is due to contributions from the χ_ν the dc-magnetic (Pauli spin) susceptibility, $\chi_{\rm core}$ the diamagnetic core susceptibility, and $\chi_{\rm VV}$ the Van Vleck susceptibility. The $\chi_{\rm core}$ is small for all the elements in these alloys and can be neglected without effecting the determination of χ_ν. Batalla *et al.*[2] showed that the contribution of $\chi_{\rm VV}$ (about 115×10^{-6} emu/mol) to measured susceptibility is large and should be considered in calculation of χ_ν. The resulting values for χ_ν for these alloys are given in Table I.

From the dc-magnetization measurements below 4 K (an example is given in Fig. 1) we determined the upper critical field H_{c2} versus temperature (Fig. 2) and the derivative of H_{c2} at T_c, $(\frac{\partial H_{c2}}{\partial T})_{T_c}$, which is given in Table I.

The procedure required to calculate $\lambda_{\rm SF}$ needs some additional measurements, namely, of the mass density d, and the room temperature resistivity ρ, for the $Zr_{75}(Ni_xFe_{1-x})_{25}$ samples. The resistivity of the $Zr_{75}(Ni_xFe_{1-x})_{25}$ ($x = 0.0, 0.25, 0.50, 0.75, 1.0$) amorphous alloys was measured as a function of temperature ranging from 292 K down to 1.1 K under constant applied pressures up to 8.0 GPa. An example of relative resistivity near superconducting transition temperature as a function of temperature and pressure for the $Zr_{75}(Ni_{0.25}Fe_{0.75})_{25}$ amorphous alloy is shown in Fig. 3. The transition width at low pressure is less than 10 mK and at high pressure it is less than 40 mK. The kink that appears at pressures higher than 2.0 GPa may be due to a slight inhemogenity in the sample.[5] In all cases the temperature dependence of the electrical resistivity was linear with a very small negative slope ($\frac{d\ln\rho}{dT} \approx -1.4 \times 10^{-4}$ K^{-1}) up to the superconducting transition temperature. Figure 4 shows the T_c versus pressure for $Zr_{75}(Ni_xFe_{1-x})_{25}$ ($x = 0.0, 0.25, 0.50, 0.75$). From the least squares fit to the $T_c(P)$ data we calculated the T_c's

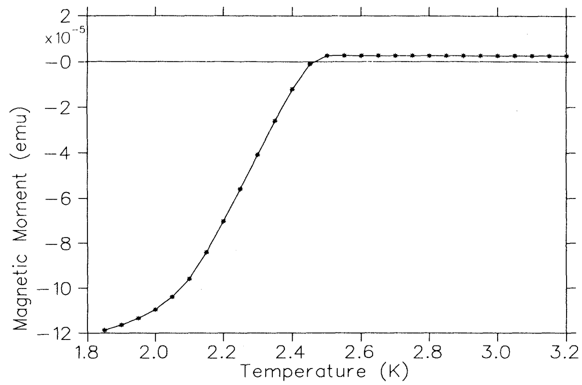

FIG. 1. dc-magnetization measurement of $Zr_{75}(Ni_{0.50}Fe_{0.50})_{25}$ near superconducting transition at the magnetic field of 1.2×10^4 G.

TABLE I. Values the mass density d, Pauli spin susceptibility χ_ν, the derivative of H_{c2} at T_c $(\frac{\partial H_{c2}}{\partial T})_{T_c}$, room temperature resistivity ρ, "specific heat"density of state $N^\gamma(E_f)$, bare density of states $N(E_f)$, Stoner factor $\bar{I}$, and the electron-spin fluctuation coupling parameter $\lambda_{\rm SF}$ as determined from the experimental results of $Zr_{75}(Ni_xFe_{1-x})_{25}$ alloys.

x	1.0[a]	0.75	0.50	0.25	0
d, $\frac{\rm g}{\rm cm^3}$	6.89	6.87± 0.05	6.85	6.82	6.8
χ_ν, $10^{-6}\frac{\rm emu}{\rm mol}$	150.9	102.4[b]	115.3	126.2	137.3
$\left(\frac{dH_{c2}}{dT}\right)_{T_c}$, $\frac{\rm KOe}{\rm K}$	27.1	35.70± 0.3	35.90	39.27	35.7
ρ, $\mu\Omega\,$cm	166.5	162.0± 2	163.0	169.8	166
$N^\gamma(E_f)$, $\frac{\rm states}{\rm eVatom}$	–	2.33	2.32	2.41	–
$N(E_f)$, $\frac{\rm states}{\rm eVatom}$	–	1.38	1.36	1.41	1.33
$\bar{I}$	–	0.565	0.62	0.64	0.69
$\lambda_{\rm SF}$	–	0.045	0.063	0.070	0.1

[a]Reference 1.
[b]The error in dc-susceptibility measurement is less than 1% however the error in χ_ν is about 5–10 % which is due to estimation of $\chi_{\rm VV}$.

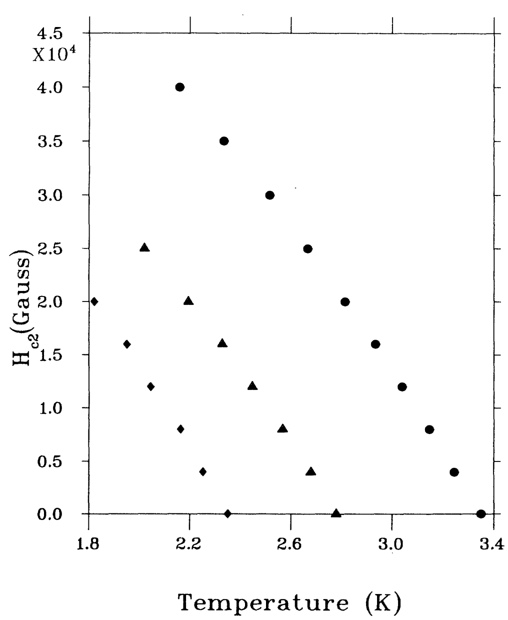

FIG. 2. Upper critical field H_{c2} vs temperature for the $Zr_{75}(Ni_xFe_{1-x})_{25}$ alloys. Solid diamond, $x = 0.25$; solid triangle, $x = 0.50$; solid circle, $x = 0.75$.

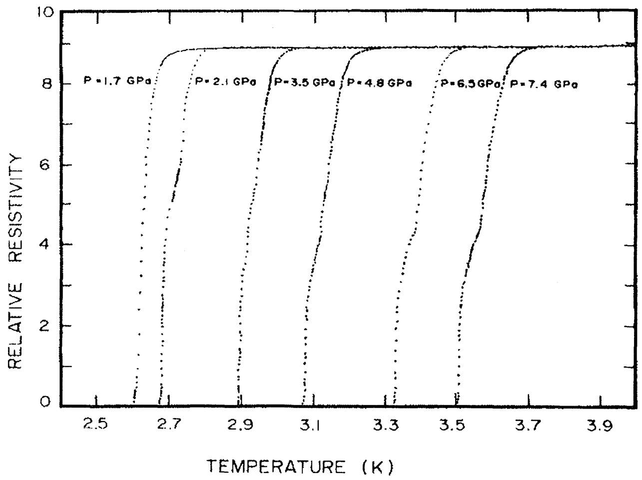

FIG. 3. Normalized resistivity near superconducting transition temperature T_c as a function of temperature and pressure for $Zr_{75}(Ni_{0.25}Fe_{0.75})_{25}$.

at one atmosphere which agree well with the T_c's measured at ambient pressure. The calculated values of $\frac{dT_c}{dP}$ from the least squares fit show an increase in the value with increasing iron concentration in all samples. A plot of $\frac{\partial T_c}{\partial P}$ and $\frac{1}{T_c}\frac{\partial T_c}{\partial P}$ versus Fe concentration is shown in Fig. 5.

B. Analysis of experimental results and discussion

The values of $\lambda_{e\text{-ph}}$, $\lambda_{\rm SF}$, $N(E_f)$ the density of states at the Fermi energy and $\bar{I}$ [$\bar{I} = IN(E_f)$, where I is the Stoner parameter or $[1-\bar{I}]^{-1}$ is the Stoner enhancement factor for the Pauli susceptibility] were determined by the following procedure.

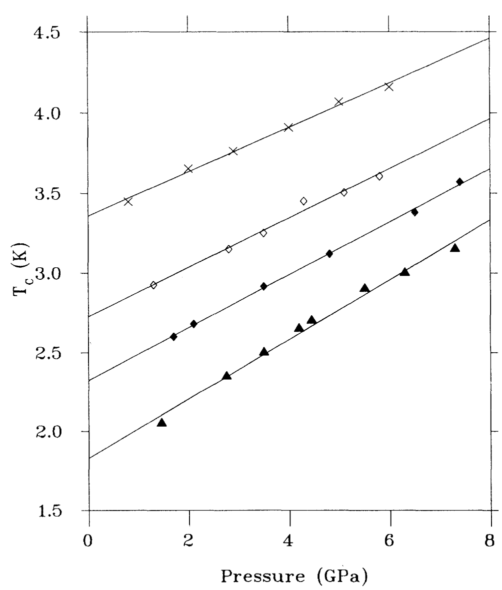

FIG. 4. Dependence of the critical temperature on pressure for the $Zr_{75}(Ni_xFe_{1-x})_{25}$ alloys: solid triangle, $x = 0$; solid diamond, $x = 0.25$; open diamond, $x = 0.50$, cross, $x = 0.75$.

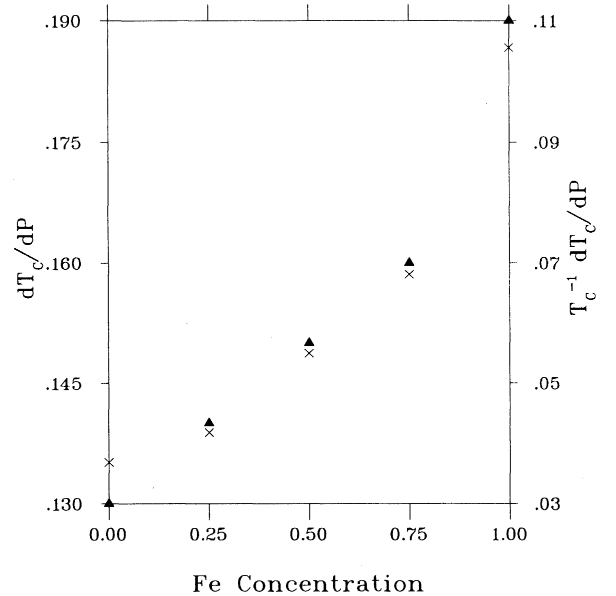

FIG. 5. The variation of $\frac{dT_c}{dP}$ (in K/GPa) and $\frac{1}{T_c}\frac{dT_c}{dP}$ (in GPa^{-1}) with respect to Fe concentration in $Zr_{75}(Ni_xFe_{1-x})_{25}$ alloys. Triangular sign, $\frac{dT_c}{dP}$ dependence; cross sign, $\frac{1}{T_c}\frac{dT_c}{dP}$.

We simultanously analyzed χ_ν along with the superconducting transition temperature T_c and the temperature dependence of the upper critical field H_{c2}, using the equations

$$\chi_\nu = \frac{\mu_b^2 N(E_f)}{1-\bar{I}}, \tag{1}$$

$$\lambda_{\rm SF} = 4.5\bar{I}\ln\left[1+\left(\frac{P_1^2}{12}\right)\left(\frac{\bar{I}}{1-\bar{I}}\right)\right], \tag{2}$$

the modified McMillan equation[7]

$$T_c \simeq \frac{\Theta_D}{1.45}\exp\left(-\frac{1+\lambda_{e\text{-ph}}+\lambda_{\rm SF}}{\lambda_{e\text{-ph}}-\lambda_{\rm SF}-\mu^*}\right) \tag{3}$$

and

$$\left(\frac{dH_{c2}}{dT}\right)_{T_c} = -1.058\times 10^9\frac{\rho d}{N}(1+\lambda_{e\text{-ph}}+\lambda_{\rm SF}) \times N(E_f), \tag{4}$$

where Θ_D is the Debye temperature, and P_1^2 is the momentum cutoff factor for spin fluctuations, proportional to E_f. It depends on the square of the ratio p_1/p_F, where p_1 is the momentum cutoff used by Doniach and Engelsberg[8] in evaluating the one-electron self-energy resulting from spin fluctuations and p_F is the Fermi momentum. Following Altounian and Ström-Olsen[1] and Batalla *et al.*[2] we considered a value of $\frac{1}{6}$ to be appropriate for P_1^2 in Zr-based amorphous alloys.

The results of calculation are shown in Table I. $N^{\gamma}(E_f)$ is the "specific heat" density of states, larger than the bare density of states $N(E_f)$ by the mass enhancement factor $(1 + \lambda_{e\text{-ph}} + \lambda_{SF})$.

To understand the effect of pressure on T_c, and to determine the volume derivative $\frac{\partial \ln \lambda_{SF}}{\partial \ln V}$ we follow the same procedure as described in our previous publication.[6] We first measure the pressure dependence of T_c for samples of $Zr_{75}Cu_{25}$, which do not show spin fluctuations (λ_{SF}=0).[1] For determining the $\frac{\partial \ln \lambda_{e\text{-ph}}}{\partial \ln V}$ of $Zr_{75}Cu_{25}$, we used the McMillan equation[9]

$$T_c \simeq \frac{\Theta_D}{1.45} \exp\left(-1.045 \frac{1 + \lambda_{e\text{-ph}}}{\lambda_{e\text{-ph}} - \mu^*(1 + 0.62\lambda_{e\text{-ph}})}\right). \quad (5)$$

Using $\mu^* = 0.13$, we obtain the logarithmic volume derivative of T_c which can be related to the Grüneisen parameter and the logarithmic volume derivative of $\lambda_{e\text{-ph}}$ by

$$\frac{\partial \ln T_C}{\partial \ln V} = -\gamma_G + \frac{1.097\lambda_{e\text{-ph}}}{(0.919\lambda_{e\text{-ph}} - 0.13)^2} \frac{\partial \ln \lambda_{e\text{-ph}}}{\partial \ln V}, \quad (6)$$

where

$$\gamma_G = \frac{3\alpha B_T}{C_V} = -\frac{\partial \ln \Theta_D}{\partial \ln V} \quad (7)$$

is the Grüneisen parameter. α is the coefficient of linear expansion, B_T is the bulk modulus, and C_V is the specific heat. We calculate the Grüneisen parameter for the alloys from the bulk modulus, specific heat and the coefficient of linear expansion as estimated from the pure component values using Vegard's law. The effective electron-electron Coulomb interaction parameter appearing in the modified McMillan equation[7] is taken as 0.13 for $Zr_{75}Cu_{25}$ as well as for all $Zr_{75}(Ni_xFe_{1-x})_{25}$ alloys considered. We obtain a value of -0.42 for the logarithmic volume derivative of $\lambda_{e\text{-ph}}$ for $Zr_{75}Cu_{25}$. We assume that $\frac{\partial \ln \lambda_{e\text{-ph}}}{\partial \ln V}$ for $Zr_{75}(Ni_xFe_{1-x})_{25}$ ($x = 0.0, 0.25, 0.50, 0.75, 1.0$) samples are the same as $\frac{\partial \ln \lambda_{e\text{-ph}}}{\partial \ln V}$ for $Zr_{75}Cu_{25}$. This assumption, combined with the modified McMillan equation,[7] allows us to determine $\frac{\partial \ln \lambda_{SF}}{\partial \ln V}$ for the $Zr_{75}(Ni_xFe_{1-x})_{25}$ alloys. All parameters obtained from calculation, measurement, and subsequent analysis as described above are listed in Table II.

For a fixed Zr concentration, λ_{SF}, i.e., spin fluctuation effects, increase as we go from the Ni-rich to the Fe-rich alloy (Fig. 6). This is consistent with the conclusion reached by Altounian and Ström-Olsen,[1] based on their work on M-Zr (M =Cu,Ni,Co, Fe) metallic glasses. The results also indicate that the $\frac{\partial \ln \lambda_{SF}}{\partial \ln V}$ is increasing as the concentration of Fe increases in Ni-Zr matrix (Fig. 6). This shows clearly the reduction in spin fluctuation under pressure hence the faster increase of T_c with presure for higher concentration of Fe in these alloys.

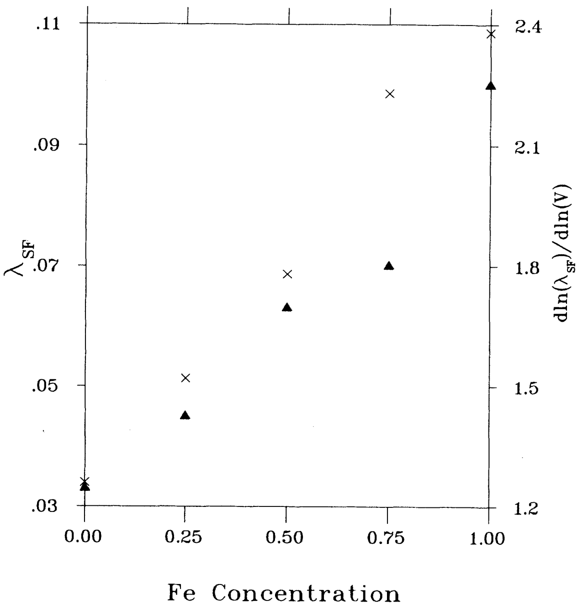

FIG. 6. The variation of λ_{SF} and volume derivative with respect to Fe concentration in $Zr_{75}(Ni_xFe_{1-x})_{25}$ alloys. Triangular sign :λ_{SF} dependence; cross sign, $\frac{d \ln \lambda_{SF}}{d \ln V}$.

In conclusion the superconducting properties of the metallic glasses $Zr_{75}(Ni_xFe_{1-x})_{25}$ have been studied for $x = 0, 0.25, 0.50, 0.75, 1.0$, both at ambient and higher quasihydrostatic pressure up to 8 GPa. We find that the spin fluctuations must be taken into account to properly describe the concentration and pressure dependence of the superconducting transition temperatures in these alloys. We have demonstrated that the modified McMillan equation[7] can correctly describe the spin fluctuations in these alloys. Based on the experimental results we have extracted the values of the spin fluctuation coupling

TABLE II. Summary of results, where, α is the coefficient of linear expansion, B_T is the bulk modulus, C_V is the specific heat, γ_G is the Grüneisen parameter, T_c is the superconducting transition temperature at ambient pressure, $\lambda_{e\text{-ph}}$ is the electron-phonon coupling parameter, λ_{SF} is the electron-spin fluctuation coupling parameter.

Alloy	α (10^{-6}) K^{-1}	B_T (10^{10}) $\frac{N}{m^2}$	C_V $(\frac{cal}{cm^3 K})$	γ_G	T_c K	$\frac{dT_c}{dP}$ (10^{-9}) $\frac{K}{GPa}$	$\lambda_{e\text{-ph}}$	λ_{SF} 10^{-2}	$\frac{d \ln \lambda_{SF}}{d \ln V}$
$Zr_{75}Ni_{25}$	7.6	9.67	0.56	0.936	3.53	0.13	0.63	3.3	1.26
$Zr_{75}(Ni_{0.75}Fe_{0.25})_{25}$	7.54	9.63	0.552	0.943	3.35	0.14	0.6 4	4.5	1.52
$Zr_{75}(Ni_{0.50}Fe_{0.50})_{25}$	7.48	9.60	0.546	0.943	2.73	0.15	0.6 4	6.3	1.78
$Zr_{75}(Ni_{0.25}Fe_{0.75})_{25}$	7.42	9.56	0.54	0.942	2.35	0.16	0.64	7.0	2.23
$Zr_{75}Fe_{25}$	7.36	9.52	0.533	0.938	1.8	0.19	0.64	10	2.38

parameter λ_{SF} and its volume derivative $\left(\frac{d\ln\lambda_{SF}}{d\ln V}\right)$ as a function of concentration. Both spin fluctuations and the rate of suppression of spin fluctuations with pressure are found to increase with higher Fe concentration in these amorphous alloys.

ACKNOWLEDGMENT

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