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Hall Effect and Resistivity of Amorphous $Fe_{83-x}Zr_7B_{10}Nb_x$ Alloys

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Abstract-The effect of small addition of Nb on the electrical resistivity and Hall coefficient of the amorphous $Fe_{83}Zr_7B_{10}$ alloy and annealed ones below the crystallization temperature were investigated, which has been considered to be suitable for high frequency core material. At room temperature, their resistivities ρ and the spontaneous Hall coefficients R_s are ~1.6 μΩm and ~3 x 10^{-8} m^3/As, respectively. R_s and ρ are decreased with increasing temperature from 100 K to room temperature. Side-jump effect was adopted to analyze the effect of the small variation of concentration and annealing. The quantity of R_s/ρ^2 at room temperature, which is directly related to the electronic structure of the mother alloy, remained almost a constant except as quenched one as it can be predicted from the side-jump effect. The unexpected temperature dependence of R_s/ρ^2 measured at low fields much below T_C is left as a question.

I. INTRODUCTION

Fe-based amorphous alloys containing about 20 at.% metalloid elements have large magnetic induction and small core loss at low frequencies (50 - 400 Hz). These alloys, however possess inferior soft magnetic properties in the high frequency range over 1 kHz, thus partial crystallization [1,2] or reduction of the magnetostriction by adding the Nb, Mo, Cr etc. [3] were previously tried to improve the high-frequency characteristics. Moreover, the metal-metal type amorphous alloy based on Fe (Co, Ni)-10 at.% Zr composites was reported as a potential soft magnetic core material [4]. However, no further reports on the high-frequency core properties for this alloy system was found.

With an aim to develop a new amorphous alloy system for high frequency use, we investigated the composition dependence of effective permeability(μ_e) in the amorphous Fe-Zr-B ternary alloys with various Zr and/or B content. As a result, very large μ_e more than 2 x 10^4 was achieved in the composition range of 6-9 at.% Zr and 9-12 at.% B, and especially the $Fe_{83}Zr_7B_{10}$ alloy was found to have optimum magnetic property [5]. If this sample was annealed below the crystallization temperature, then better μ_e would be obtained [5].

In this paper we present a study of the resistivity and Hall effect as a function of temperature from 100 K to room temperature in the metallic glass $Fe_{83-x}Zr_7B_{10}Nb_x$ (x=0, 0.5, 1.0, 1.5 at.%) which are annealed below the crystallization temperature. The experimental data for Hall coefficients will be discussed on the basis of

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side-jump effect.

II. EXPERIMENT

Ribbon samples of amorphous $Fe_{83-x}Zr_7B_{10}Nb_x$(x=0, 0.5, 1.0, 1.5 at.%) alloys about 1.5 - 2.0 mm width and 20 - 30 µm thickness were prepared by a single-roller melt spinning method in an argon atmosphere and confirmed to be amorphous by an x-ray diffractometer. The alloy was wounded into toroidal core with 21 mm inner diameter and subsequently annealed at 450 °C below crystallization temperature for 1 hr in a vacuum of about 10^{-4} Torr.

Electrical contacts were made by using silver paint or indium. The electric resistivities were measured by a standard dc four probe method using a constant current power supply. A commercial temperature controller with calibrated Pt and carbon-glass thermometer was used.

Room temperature measurements of the Hall effect were carried out with the low frequency ac method using phase sensitive lock in amplifier and digital voltmeter. The maximum applied magnetic field was 2.5 T.

For studying the temperature variation of the Hall effect we used the ac double frequency method [6]. This method uses an alternating current $I = I_o \cos\omega_1 t$, and an alternating magnetic field $B = B_o \cos\omega_2 t$ at a different frequency ω_1. In this experiment we chose $\omega_1 = 5$ Hz, $\omega_2 = 3$ Hz respectively. The amplitude corresponding to the frequency , $\omega_1 + \omega_2$, is due to the Hall voltage. The signal were detected using a lock-in amplifier. The temperature was varied over the range 100 - 300 K by a temperature control unit named Cryogenic Microminiature Refrigerator.

III. RESULTS AND DISCUSSIONS

Extra ordinary Hall resistivity of ferromagnetic materials are considered to have two main mechanisms; side-jump effect and skew scattering [7-11]. The latter part is to be dominant if the resistivity of the material is quit low, since this mechanism produces a Hall coefficient that is proportional to resistitivity , ρ, while that due to the side-jump effect is proportional to ρ^2. Since this condition is not even close to the characteristics of our samples, we simply neglect the skew scattering mechanism and concentrate on the side-jump effect to analyze our data. Theoretical equations for the side-jump effect can be summarized as

$$\begin{aligned}\rho_H^{SJ} &= \rho^2\sigma_{xy}^{SJ} \qquad \cdots \quad (1)\\ &= \sum_i \frac{2\rho^2 N_i q_i^2}{\hbar}\lambda_{SO}^{SJ}\langle S_z\rangle\end{aligned}$$

$$\text{with } \lambda_{SO}^{SJ} = \sum_n \frac{|\langle n|\vec{L}|0\rangle|^2}{\varepsilon_n - \varepsilon_F} I\, l^2 A_{SO} ,$$

where i denotes carrier type, q_i denotes charge of i-type carrier, λ_{SO}^{SJ} denotes the effective spin-orbit parameter, A_{SO} is the atomic spin-orbit parameter, I is the overlap integral and l is the distance between the scatterers.

And the abnormal Hall resistivity by the side-jump effect with the direction of normal Hall resistivity due to the Lorentz force can be written as [7],

$$\begin{aligned}\rho_H &= \frac{V_H \cdot d}{I}\\ &= \mu_o R_o(H_a - NM) + \mu_o R_1 M\\ &= R_o B_a + \mu_o M(R_1 - R_o N)\\ &= R_o B_a + \mu_o M R_s \qquad \cdots \quad (2)\end{aligned}$$

where V_H is the Hall voltage, d the thickness of specimen, I the electric current, H_a the applied magnetic field intensity, B_a $(= \mu_o H_a)$ the applied magnetic induction, M the magnetization, N the demagnetization factor (assumed equal to 1 for our geometry), R_o and R_1 are the ordinary and extraordinary Hall coefficients, respectively. $R_s(=R_l-R_0)$ is the spontaneous Hall coefficient. Numerically, there is not much difference between R_l and R_s in high resistivity ferromagnets. The expression in the first brackets is the internal magnetic field. At the fields well below the saturation, $\mu_0(H_a- NM) \cong 0$. The initial slope in Fig. 1, is essentially R_l. At high field, $M = M_s$, the saturation magnetization, the slope will be R_o. Where the two straight lines (extrapolated) intersect, the abscissa, B_a, equals to the saturation magnetization $\mu_0 M_s$. The fitting data of Fig. 1 and other information on these samples are given in Table 1.

Table 1. Ordinary and spontaneous Hall coefficients, the electric resistivities and R_s/ρ^2 values for amorphous $Fe_{83-x}Zr_7B_{10}Nb_x$(x=0, 0.5, 1.0, 1.5 at.%) alloys at room temperature.

sample	ρ (μΩm)	R_s (m^3/As)	R_0 (m^3/As)	$\mu_0 M_s$ (T)	R_s/ρ^2
x=0 as quenched	1.71	3.32x10^{-8}	0.26x10^{-8}	0.68	1.13x10^4
x=0 anneaned	1.53	3.33x10^{-8}	0.12x10^{-8}	0.84	1.42x10^4
x=0.5 annealed	1.51	3.29x10^{-8}	0.12x10^{-8}	1.00	1.44x10^4
x=1.0 annealed	1.70	4.04x10^{-8}	0.11x10^{-8}	0.91	1.40x10^4
x=1.5 annealed	1.63	3.90x10^{-8}	0.12x10^{-8}	0.85	1.47x10^4

Since the magnetic saturation was not reached within the limit of our experiment, the evaluated values of the ordinary Hall coefficient ($R_0 \cong 1 \times 10^{-9}$m^3/As) of the amorphous $Fe_{83-x}Zr_7B_{10}Nb_x$ alloys were ten times larger than those of the other previously reported ferromagnetic amorphous alloys [7]. Thus, the value of R_0 can be regarded only as an upper limit of the ordinary Hall coefficient.

R_s in the equation (2) can be written as

$$R_s/\rho^2 = \beta\lambda_{SO} \qquad \cdots (3)$$

with the equation (1), where β is a numerical constant. It is known that λ_{SO} of d-electrons is about 1000 times larger than that of a free electron case ($\lambda_{SO}^{free} = a_0^2 \alpha^2$) [8,11,12]. As shown in the equation (1), λ_{SO} is sensitive to the location of the Fermi level in the electronic structure of each sample, and it is impossible to calculate it unless all informations about the band structure is calculated in detail. Some critical concentrations of the sign change of the R_s for rather simple alloys can be calculated on the basis of available number of electrons of each elements [8]. The variation of λ_{SO} for Zr based Zr-Ni, Zr-Co and Zr-Cu alloys with the change of concentration of early transition metals was studied recently [12]. The authors showed that the change of λ_{SO} is dramatic for binary Zr-early transition metal alloys when the concentration varies from the Zr rich region to late transition metal rich region [12]. However, when Al or metalloid is doped to the binary alloys, doping did not change the λ_{SO} at all [13,14].

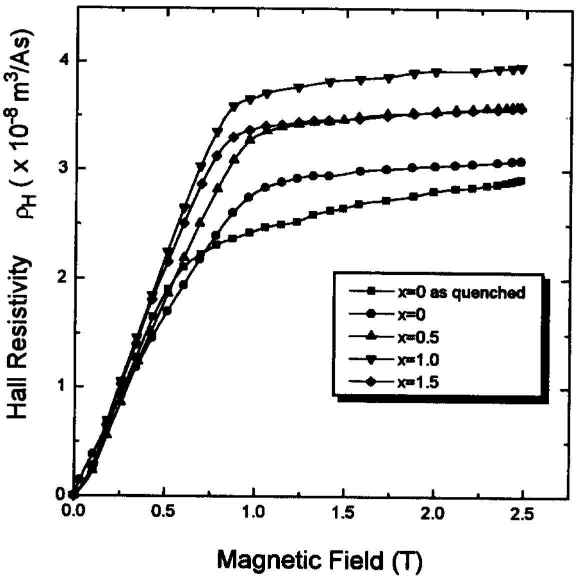

Fig. 1 The Hall resistivity ρ_H as a function of the applied magnetic field for amorphous $Fe_{83-x}Zr_7B_{10}Nb_x$ (x=0, 0.5, 1.0, 1.5 at.%) alloys

In this experiment, the values of λ_{SO} are supposed to be almost a constant even though some Fe is substituted by Nb and heat treated, since the amount of substitution is small and the effect of the heat treatment can not change the electronic structure dramatically even though it could rearrange the ions of the material by releasing the thermal stress of the as quenched states. We have measured d.c. resistivities at room temperature (Table 1) and it is satisfying to note that for the present system also R_s/ρ^2 , is roughly a constant within error range, except only an as quenched amorphous $Fe_{83}Zr_7B_{10}$ alloy. This is perhaps due to the small change of the electronic structure caused by heat treatment rather than substitution of Fe, since all 4 annealed samples maintain small values of R_s. It is well known that the atomic distances in the crystallized state is shorter than those of as quenched state. Therefore, we conclude that the annealing process affected the electronic structure of as quenched alloys and changed the intrinsic properties of λ_{SO}, even though our annealing process did not crystallized samples totally.

The variation of ρ with the Nb content is not pleasing, since it is supposed to be monotonically increasing with Nb content, but 1.0 at. % Nb showed a rather larger value than the other annealed samples. We guess that the sample of 10 at.% Nb was not fully annealed to generate lower resistivity, while the others were.

Fig. 2 and Fig. 3 show the temperature dependence of spontaneous Hall coefficient R_s measured at $H \rightarrow 0$ using the ac method described above, and electric resistivity ρ, respectively, for as quenched amorphous $Fe_{83}Zr_7B_{10}$ alloy and annealed $Fe_{82.5}Zr_7B_{10}Nb_{0.5}$ alloy. It was found that R_s and ρ decrease with increasing temperature. This small negative TCR (Temperature Coefficient of Resistivity) is common in amorphous alloys of high resistivity [15]. Of course the decrease of R_S is partially caused by the decrease of resistivity with temperature.

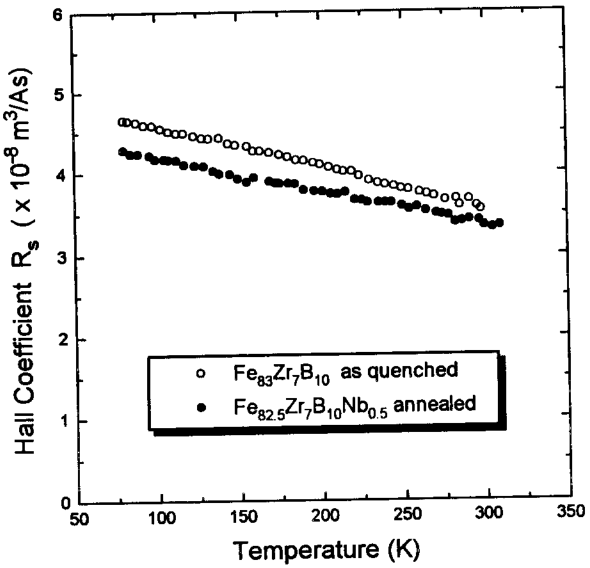

Fig. 2 The temperature dependence of spontaneous Hall coefficient R_s

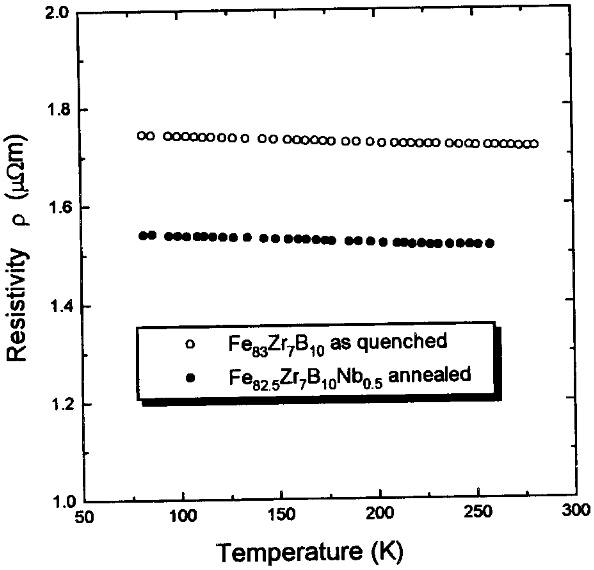

Fig. 3 The temperature dependence of ohmic resistivity ρ

This small TCR, however, is not large enough to explain the temperature dependence of R_S shown in Fig. 2. λ_{SO} in Eq. (1) also is a factor in determining R_S, but it is supposed to be independent of temperature, since λ_{SO} mainly represents the electronic structure around the Fermi level. In order to make it clear, we present the temperature dependence of R_S/ρ^2 (Fig. 4). Recall this value only depends on λ_{SO}. The unexpected temperature dependence of R_S/ρ^2 with the variation of 30~40% from the room temperature (Fig. 4) is quit puzzling, since R_S is independent of temperature well below T_C. One might compare our result to the temperature dependence of R_S of ferromagnetic amorphous Fe-Zr alloys [16] which was almost independent of temperature for alloys well below T_C. Nano-crystalline structure might be the reason for it, but we leave this dilemma as an open question.

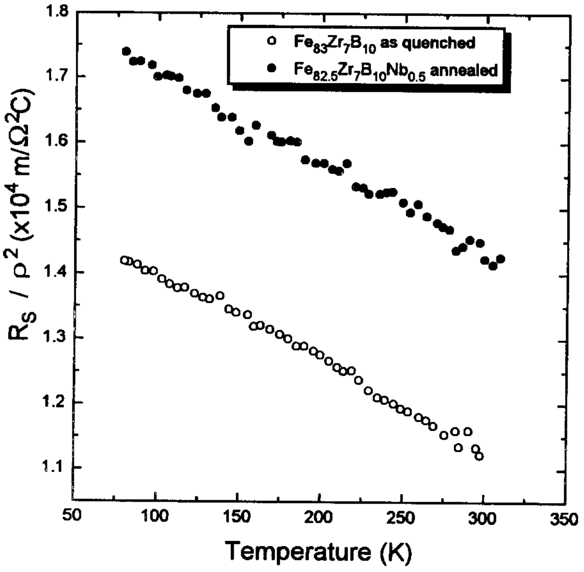

Fig. 4 Correlation between the spontaneous Hall coefficient R_s and the square of electric resistivity ρ^2 ; open circle is an as quenched amorphous $Fe_{83}Zr_7B_{10}$ alloy and solid circle is an annealed $Fe_{82.5}Zr_7B_{10}Nb_{0.5}$ alloy.

IV. CONCLUSIONS

The electronic transport properties of amorphous $Fe_{83-x}Zr_7B_{10}Nb_x$(x=0, 0.5, 1.0, 1.5 at.%) alloys can be characterized as follows;

1) At room temperature their resistivities are ~1.6 μΩm and the spontaneous Hall coefficients are positive and ~3 x 10^{-8} m^3/As.

2) Due to the high field susceptibility, the values of ordinary Hall coefficient ($R_0 \cong 1 \times 10^{-9} m^3/As$) were ten times large than that of the other results.

3) R_s/ρ^2 is roughly a constant within error range at room temperature, except only an as quenched amorphous $Fe_{83}Zr_7B_{10}$ alloy.

4) R_s and ρ are decreased with increasing temperature from 100 K to 300 K.

5) Unexpected temperature dependence of R_s/ρ^2 is observed.

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