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journal or publication title	Science reports of the Research Institutes, Tohoku University. Ser. A, Physics, chemistry and metallurgy
volume	26
page range	214-224
year	1976
URL	http://hdl.handle.net/10097/27851

Hall Effect and Magnetoresistance in Ferromagnetic Amorphous Fe-Co and Fe-Ni Alloys*

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(Received December 25, 1976)

Synopsis

The magnetoresistance over the temperature range from 77 K to the crystallization temperature and the Hall resistivity at room temperature were measured for the rapidly quenched amorphous alloys $(Fe_{1-x}Co_x)_{78}Si_{10}B_{12}$ and $(Fe_{1-x}Ni_x)_{78}Si_{10}B_{12}$. The anisotropic magnetoresistance ratio $\Delta\rho/\rho$ in both systems was roughly one order of magnitude smaller than that of crystalline Fe, Co and Ni metals and their alloys, and the normal and anomalous Hall coefficients R_0 and R_s were roughly one or two order of magnitude greater than those of the crystalline metals and alloys. The values of $\Delta\rho/\rho$ and R_0 and R_s monotonically changed with composition x at room temperature. The changes in $\Delta\rho/\rho$ and R_s with composition were compared with those for the Ni-based crystalline alloys on the basis of Berger's theory.

I. Introduction

Recently there has been considerable interest in the peculiar phenomena on the electrical resistance and the galvanomagnetic effects in the metal-metalloid amorphous alloys. The most significant experimental results reported on the literature are the large absolute value of electrical resistivity, the very small temperature coefficient of the resistivity and the nearly temperature independent anomalous Hall coefficient(1,2). The absence of the periodical atomic arrangement in the amorphous structure has been thought to be mainly responsible for these characteristics, but the detailed mechanisms of which are not yet to be made clear.

On the other hand, the Fe, Co or Ni based amorphous alloys have soft magnetic nature(3,4). We previously examined the magnetic properties of Fe-Co based amorphous alloys, and found that the magnetostriction contributes greatly to the appearance of finite values of the magnetic anisotropy and the hysteresis energy in these alloys(5). With regard to this significant behavior of magnetostriction, another problem of interest is the magnetoresistance of these amorphous alloys.

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* The **1669th** report of the Research Institute for Iron, Steel and Other Metals.

(1) S.C.H. Lin, J. appl. Phys., **40** (1969), 2175.
(2) T. Shirakawa, T. Kodama, K. Okamoto, S. Matsushita and Y. Sakurai, 7th Annual Conf. on Mag. in Japan (1975), 5pA–1 (in Japanese).
(3) T. Egami, P.J. Flanders and C.D. Graham, Appl. Phys. Letters, **26** (1975), 128.
(4) M. Kikuchi, H. Fujimori, Y. Obi and T. Masumoto, Japan. J. appl. Phys., **14** (1975), 1077.
(5) H. Fujimori, Y. Obi, T. Masumoto and H. Saito, Mater. Sci. Eng., **23** (1976), 281.

It appears that the anomalous magnetoresistance depends upon the magnetic domain structure associated with stress-magnetostriction. The magnitude of anisotropic magnetoresistance may depend upon the electronic mechanism associated with amorphous structure which is related to the characteristics of magnetostriction. However, the available data on the magnetoresistance of the amorphous alloy[6,7] are not sufficient to discuss the above problem.

In order to clarify the characteristics of the electrical transport phenomena associated with ferromagnetism in amorphous alloys, we have investigated the temperature and composition dependences of the magnetoresistance and the Hall resistivity in the amorphous alloys $(Fe_{1-x}Co_x)_{78}Si_{10}B_{12}$ and $(Fe_{1-x}Ni_x)_{78}Si_{10}B_{12}$.

II. Experimental procedure

The amorphous alloys $(Fe_{1-x}Co_x)_{78}Si_{10}B_{12}$ and $(Fe_{1-x}Ni_x)_{78}$-$Si_{10}B_{12}$ in the form of ribbons, about 20 μm$\times$2 mm in cross section were produced by a rapid quenching technique of the rotating cylinder type[8]. All the as-quenched specimens were confirmed to be in the amorphous state by the X-ray diffraction method.

The magnetoresistance was measured on specimens about 27 mm long by the standard four-point potentiometric technique over the temperature range from 77 K to the crystallization temperature. For measurements of the longitudinal and transverse magnetoresistances, external magnetic fields up to 14 kOe were applied parallel to the long axis and across the width, while the current of 10 mA was only applied parallel to the long axis.

The Hall effect measurement was performed by the standard dc-method using about 70 mm long specimens. Figure 1 illustrates the block diagram of the apparatus. All the Pt-lead wires (0.1 mm in diameter) were spot-welded to the edge of the specimens. The direction of the applied magnetic field was perpendicular to the specimen surface. When the field was not applied the helicalohm R, shown in the diagram was adjusted such that the Hall voltage became zero. The dc-current used was 100 mA and the field strength was up to about 20.5 kOe.

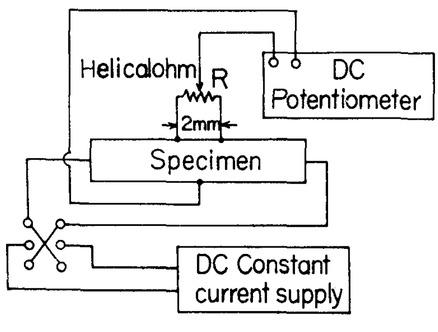

Fig. 1. Block diagram for Hall voltage measurement.

(6) M.R. Bennett and J.G. Wright, Phys. Letters, **38A** (1972), 419.
(7) L. Berger, A.I.P Conf. Proc., **29** (1975), 165.
(8) T. Masumoto *et al.*, unpublished.

The sensitivity of the potentiometer was within the limit of 0.01 μV. No hysteresis effects are observed in the Hall voltage *vs.* field curves. All the measurements were carried out at room temperature.

III. Results and discussion

Magnetoresistance

Figure 2 shows the magnetoresistance *vs.* magnetic field curves measured at room temperature and at 77 K for the as quenched $(Fe_{0.3}Ni_{0.7})_{78}Si_{10}B_{12}$ alloy, where $\Delta\rho_{/\!/}/\rho_0(=\rho_{/\!/}/\rho_0-1)$ and $\Delta\rho_{\perp}$ $(=\rho_{\perp}/\rho_0-1)$ represent the longitudinal and the transverse magnetoresistance-ratios, respectively, and $\rho_{/\!/}$ is the longitudinal magnetoresistance, $\rho_{\perp}$ the transverse magnetoresistance, and ρ_0 the specific electrical resistivity at each temperature. $\Delta\rho_{/\!/}/\rho_0$ (or $\Delta\rho_{\perp}/\rho_0$) increases (or decreases) sharply with increasing field in the low field region. In the high field region, $\Delta\rho_{/\!/}/\rho_0$ and $\Delta\rho_{\perp}/\rho_0$ decrease linearly with increasing field, having the same gradient. This means that the value of $\rho_{/\!/}-\rho_{\perp}$ reaches saturation at a higher field. Such a saturation in $\rho_{/\!/}-\rho_{\perp}$ occurs corresponding to the saturation of magnetization. The linear decrease in $\Delta\rho_{/\!/}/\rho_0$ or $\Delta\rho_{\perp}/\rho_0$ against field is characteristic of ferromagnetic materials and is called the forced effect. Similar magnetoresistance curves are obtained in all other composition alloys.

Since the value of $\rho_{/\!/}-\rho_{\perp}$ is nearly constant against field in the saturation

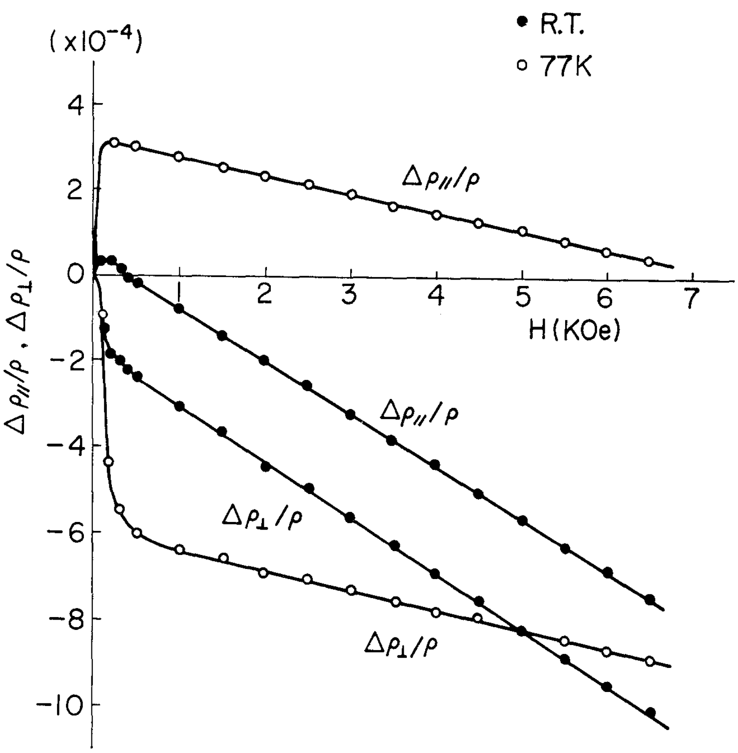

Fig. 2. The longitudinal and the transverse magnetoresistance ratio $\Delta\rho_{/\!/}/\rho$, $\Delta\rho_{\perp}/\rho$ *vs.* applied magnetic field at room temperature and 77 K in the $(Fe_{0.3}Ni_{0.7})_{78}Si_{10}B_{12}$ amorphous alloy.

magnetization region the quantity defined as[9]

$$\Delta\rho/\rho = (\rho_{/\!/}-\rho_{\perp})/(\rho_{/\!/}/3+2\rho_{\perp}/3) \qquad (1)$$

is an important value to represent the anisotropic magnetoresistance ratio. Figure 3 shows the temperature dependence of $\Delta\rho/\rho$ measured in the temperature range 77 K to the crystallization temperature T_{cry} for some of the as-quenched Fe-Co amorphous alloys. As seen in this figure, $\Delta\rho/\rho$ decreases monotonically with increasing temperature, except for the $\Delta\rho/\rho$ in $Co_{78}Si_{10}B_{12}$ which shows a slight increase with increasing temperature between 200° and 300°C. According to Kondo's theoretical consideration[10], $\Delta\rho/\rho$ in the crystalline ferromagnets goes to zero at 0K and at the Curie temperature, and hence takes a maximum

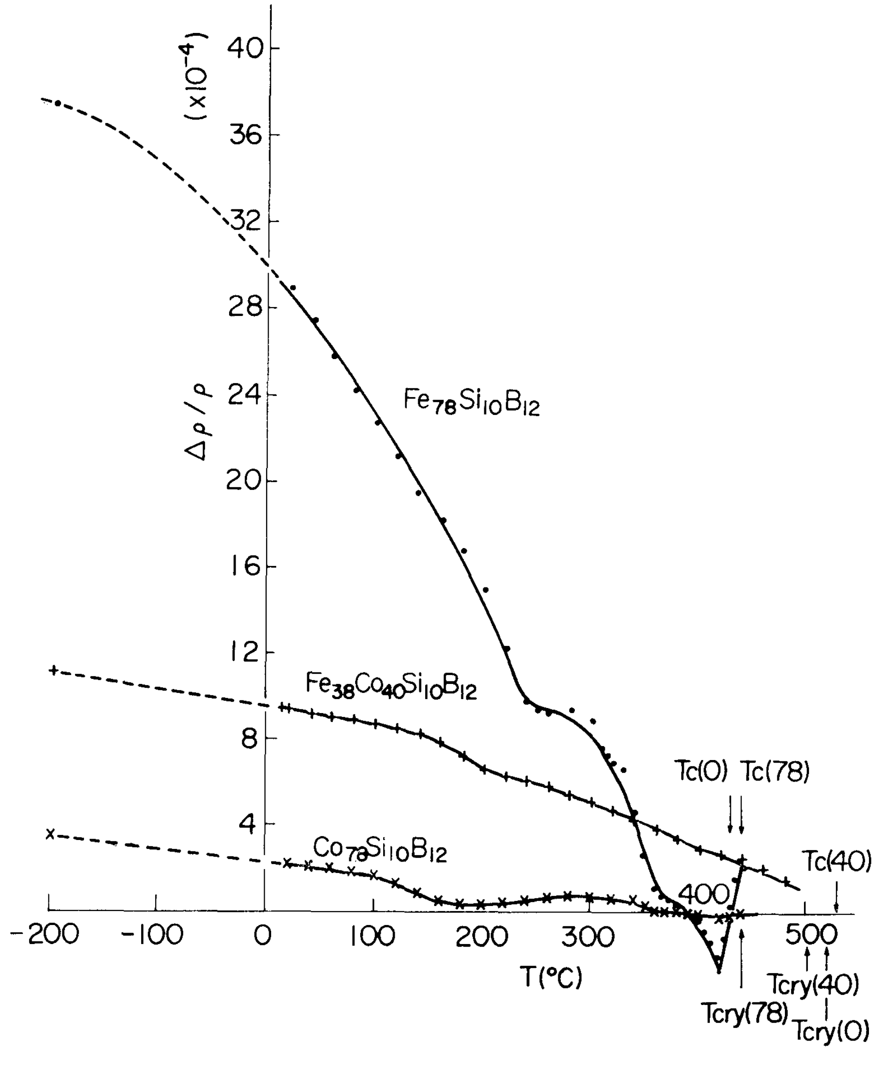

Fig. 3. The temperature dependence of anisotropic magnetoresistance ratio $\Delta\rho/\rho$ for the systems $Fe_{78}Si_{10}B_{12}$, $Fe_{38}Co_{40}Si_{10}B_{12}$ and $Co_{78}Si_{10}B_{12}$. Values in parenthesis 0, 40 and 78 indicate the Co composition.

(9) T.R. McGuire and R.I. Potter, IEEE Trans. Magnetics, MBG-**11** (1975), 1018.
(10) J. Kondo, Progr. Theor. Phys., **27** (1962), 772.

value in the intermediate temperature region. However the curves of $\Delta\rho/\rho$ *vs.* temperature for the present alloys do not take any significant maximum in the temperature range 77 K to T_c. Moreover, it must be noted that all the curves have a similar anomaly in the neighborhood of 200°C. Since it has been found that the $\Delta\rho/\rho$ changes irreversively against temperature after the specimen is heated beyond about 200°C, the anomaly observed near 200°C is thought to occur as a result of the change in amorphous structure due to the annealing. For instance, the value of $\Delta\rho/\rho$ of the as-prepared $Fe_{78}Si_{10}B_{12}$ was reduced to about 60% by annealing for 60 min at 300°C. The annealing effects on other physical properties are also remarkable between 200°C and T_{cry}(11). But the details of the structural changes involved are still unknown.

The observed results on $\Delta\rho/\rho$ for the amorphous alloys used for the investigation are summarized in Fig. 4 as a function of composition x. The values of $\Delta\rho/\rho$ in some of the alloys are also shown in Table 1. It is to be noted that $\Delta\rho/\rho$ decreases monotonically with increasing x in both alloy systems. In the Fe-Ni system, $\Delta\rho/\rho$ becomes zero at the composition of $x=0.8$ at room temperature and also at the composition of $x=1$ at 77 K, because these two alloys are paramagnetic at those temperatures. The values of $\Delta\rho/\rho$ in the amorphous

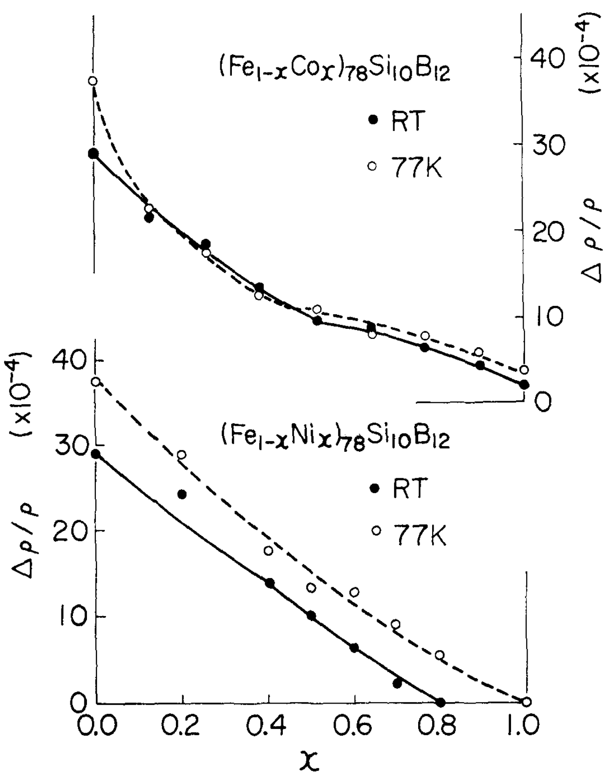

Fig. 4. The compositional dependence of $\Delta\rho/\rho$ in the $(Fe_{1-x}Co_x)_{78}Si_{10}B_{12}$ and $(Fe_{1-x}Ni_x)_{78}$-Si_{10}-B_{12} amorphous alloy systems at room temperature and 77 K.

(11) H. Fujimori, H. Morita, Y. Obi and S. Ohta, Presented at the 2nd International Symposium on Amorphous Magnetism (Troy, 1976). To be published in the Proceedings.

Table 1. Data for $\Delta\rho/\rho$, R_0 and R_s in the present alloys and some of other alloys.

	I_s emu/cm^3	T_c °C	ρ_0 $\mu\Omega$ cm	$\Delta\rho/\rho$ ($\times 10^{-4}$)	R_0 V·cm/A·G ($\times 10^{-12}$)	R_s V·cm/A·G ($\times 10^{-12}$)
$Fe_{78}Si_{10}B_{12}$	1230	430	175	29	33.7	614
$Fe_{38}Co_{40}Si_{10}B_{12}$	1110	~530	155	9.5	12.7	309
$Co_{78}Si_{10}B_{12}$	880	438	150	2.2	3.05	262
$Fe_{39}Ni_{39}Si_{10}B_{12}$	490	330	178	10.1	3.76	428
$Ni_{78}Si_{10}B_{12}$	—	—	117	0	−1.17	0
$Fe_{80}P_{13}C_7$		313			30[1]	600[1]
$Fe_{40}Ni_{40}P_{14}B_6$	694	227	154		6.6[20]	335[20]
Fe	1717	1043		20[31]	0.23[32]	7.22[32]
Co	1424	1389		190[9]	−0.84[32]	0.6 [32]
Ni	484	631		202[9]	−0.46[32]	−6.05[32]
$Fe_{50.7}Ni_{49.3}$			35.2	~50[21]	0.53[23]	30.0 [23]
$Fe_{13.8}Ni_{86.2}$				~460[13]	~−1.68[23]	~0 [23]
$Co_{50}Ni_{50}$				505[13]	~−1.96[24]	~2.55[24]

alloys are roughly one or two orders of magnitude smaller than the crystalline ferromagnets, as seen in Table 1. The main reason for the small $\Delta\rho/\rho$ in the amorphous alloys is thought to be related to the large electrical resistivity of the amorphous alloys.

The forced magnetoresistance, $(1/\rho_0)$ $(d\rho/dH)$, measured at room temperature and 77 K for the present amorphous alloys are shown in Fig. 5 as a function of composition x. The values are all negative. The absolute value does not show any remarkable change with composition in the Fe-Co based alloys, but increases sharply with increasing composition in the Fe-Ni based alloys. Such a difference between the forced magnetoresistances of the two alloy systems may be due to the difference between the Curie temperatures of the two alloy systems. The Curie temperatures of the $(Fe_{1-x}Co_x)_{78}Si_{10}B_{12}$ alloys change between 560°C and 430°C[11], but those of $(Fe_{1-x}Ni_x)_{78}Si_{10}B_{12}$ change greatly from 430°C for $Fe_{78}Si_{10}B_{12}$ to 49°C for $(Fe_{0.25}Ni_{0.75})_{78}Si_{10}B_{12}$[12]. Comparisons between the present results and the data on the crystalline materials indicate that the values of $(1/\rho_0)$ $(d\rho/dH)$ in the present Fe-Co amorphous alloys are roughly one order of magnitude smaller than those of Ni-Mn, Ni-Si and Ni-Fe alloys[13].

Hall resistivity

Figures 6 and 7 show the magnetic field dependence of the Hall resistivity, $\rho_H = V_H d/i$, measured at room temperature for both of the amorphous alloy systems,

(12) T. Takahashi, Private Communication.
(13) H.C. Van Elst, Physica, **25** (1959), 708.
(31) Y. Gondo and Z. Funatogawa, J. Phys. Soc. Japan, **7** (1952), 41.
(32) N.V. Volkenshtein and G.V. Fedorov, Sov. Phys. JETP, **11** (1960), 48.

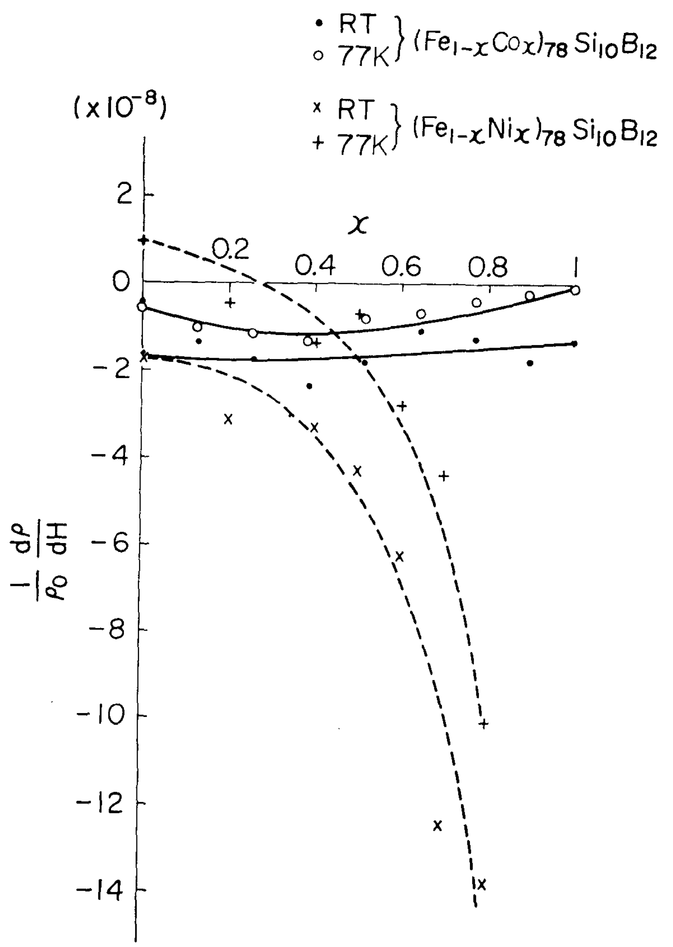

Fig. 5. The compositional dependence of forced magnetoresistance $(1/\rho_0)$ $(d\rho/dH)$ in the $(Fe_{1-x}Co_x)_{78}Si_{10}B_{12}$ and $(Fe_{1-x}Ni_x)_{78}Si_{10}B_{12}$ amorphous alloy systems at room temperature and 77 K.

where V_H is the Hall voltage, d the specimen thickness, and i the current. As seen in the figures, ρ_H increases linearly with respect to the applied field and then reaches saturation at a higher field. For ferromagnets, the Hall resistivity can be expressed as[14]

$$\rho_H = R_0 B + 4\pi R_s M \,, \tag{2}$$

where R_0 and R_s are the normal and the anomalous Hall coefficients, respectively, M is the magnetization, and B the magnetic induction. Furthermore, ρ_H can be expressed using the applied field H as follows;

$$\rho_H = R_0 H + 4\pi R_1 M \tag{3}$$

and

$$R_1 = \left(1 - \frac{N}{4\pi}\right) R_0 + R_s \,, \tag{4}$$

(14) C.M. Hurd, *The Hall Effect in Metals and Alloys*, Plenum Press, New York-London, (1972), P. 154.

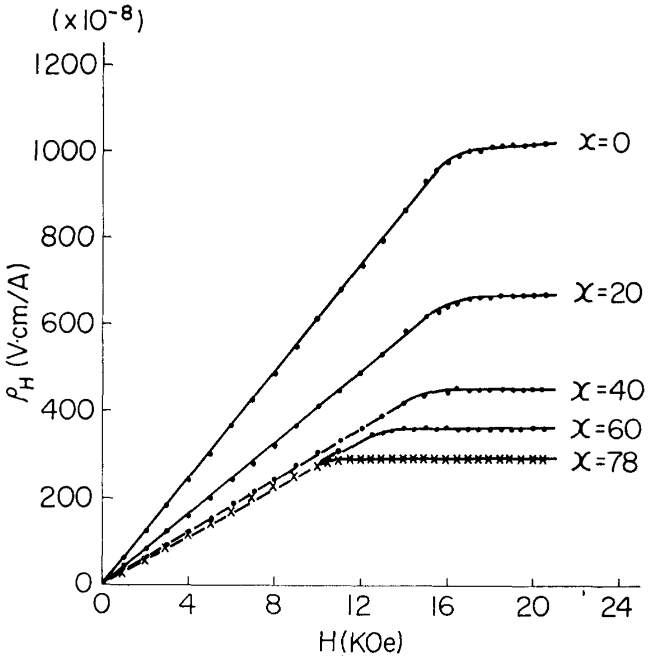

Fig. 6. Magnetic field dependence of Hall resistivity in the $Fe_{78-x}Co_xSi_{10}B_{12}$ amorphous alloy system at room temperature.

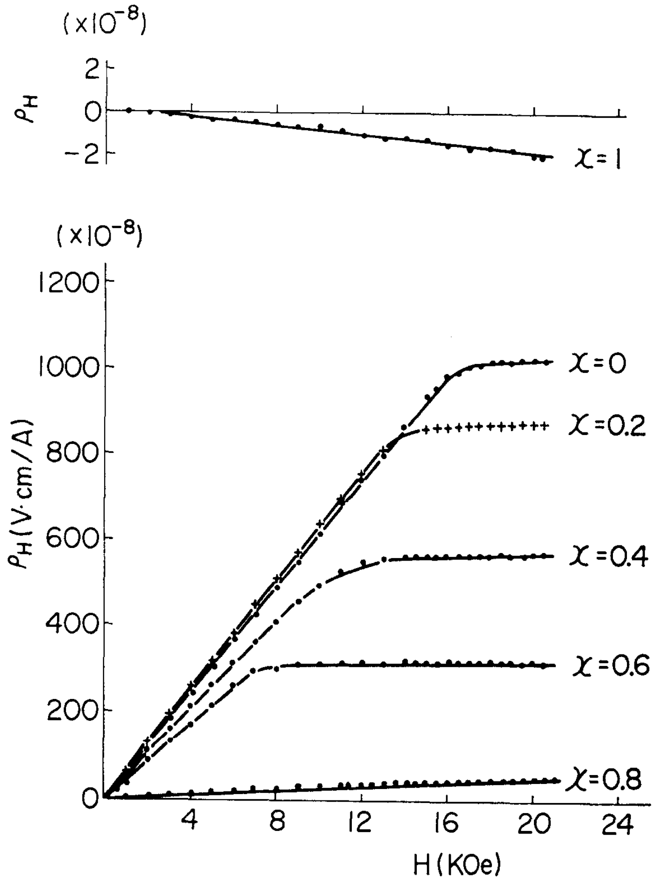

Fig. 7. Magnetic field dependence of Hall resistivity in the $(Fe_{1-x}Ni_x)_{78}Si_{10}B_{12}$ amorphous alloy system at room temperature.

where N is the demagnetization factor normal to the specimen surface. Accordingly the gradient of ρ_H against H at higher fields gives rise to the value of R_0, while the value of $\rho_H(H=0)$ obtained by extrapolating linearly the ρ_H *vs.* H curve at the higher fields to $H=0$ is equal to the value of $4\pi R_1 M_s$, where M_s is the saturation magnetization. Putting the values of R_0 and R_1 into equation (4), we can finally obtain the value of R_s.

Figure 8 shows the compositional dependence of R_0 and R_s in the as-prepared amorphous alloys. Some of the above data are also shown in Table 1. We may note the following points: i) The signs of R_0 and R_s are always positive over the entire composition range, except for $Ni_{78}Si_{10}B_{12}$ (this alloy is paramagnetic at room temperature).
ii) The magnitude of R_s is roughly one order of magnitude greater than that of R_0 at room temperature in a way similar to the situation in the Hall effect of the ordinary $3d$-ferromagnetic metals and alloys(15).
iii) The values of R_0 and R_s in the Fe-Co system decrease monotonically with increasing composition of Co*. These values in the Fe-Ni system decrease with increasing composition of Ni in the range of $0 \leq x \leq 0.7$ but increase in the range of

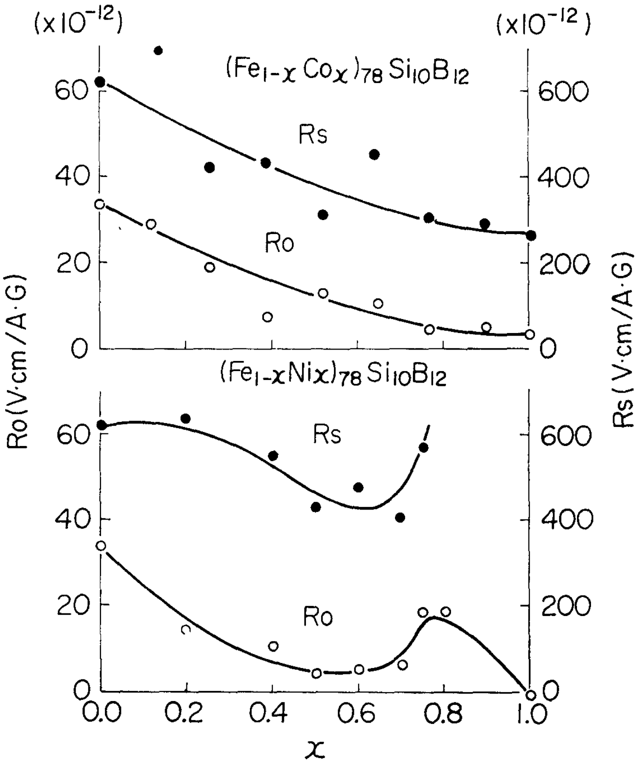

Fig. 8. The compositional dependences of R_0 and R_s in the $(Fe_{1-x}Co_x)_{78}Si_{10}B_{12}$ and $(Fe_{1-x}Ni_x)_{78}Si_{10}B_{12}$ amorphous alloy systems at room temperature.

(15) E.M. Pugh and N. Rostober, Rev. mod. Phys., **25** (1953), 151.

* Recently, almost the same result on R_s was obtained in the $Fe_{80-x}Co_xB_{20}$ amorphous alloys by R.C. O'Handley (the 2nd International Symposium on Amorphous Magnetism Troy, 1976)

$0.7 \leq x \leq 0.8$. The increase of R_s is probably due to the Curie temperature close to room temperature in the range of $0.7 \leq x \leq 0.75$. (Generally, the R_s *vs.* temperature curve often takes a maximum around $T/T_c=0.9$[10,16]).

iv) The values of R_0 and R_s for all the alloys are about one or two order of magnitude greater than those in crystalline Fe, Co and Ni metals and their alloys as seen in the Table. The magnitudes of R_s bear comparison with the extremely large R_s in the alloys such as Invar alloy[17], CrTe[18] and Fe_3Pt[19]. For instance, R_s of 614×10^{-12}V·cm/A.G at $T/T_c=0.42$ for the amorphous $Fe_{78}Si_{10}B_{12}$ is comparable to the absolute value of $R_s=-730\times10^{-12}$V·cm/A.G at $T/T_c=0.47$ for Cr-Te but is larger than the absolute value of $R_s=-4.7\times10^{-12}$V·cm/A.G. at $T/T_c=0.47$ for Ni[18].

v) As seen in the Table, the values of R_0 and R_s for $Fe_{40}Ni_{40}P_{14}B_6$[20] and for $Fe_{80}P_{13}C_7$[1] are nearly equal to those for the $Fe_{39}Ni_{39}Si_{10}B_{12}$ and $Fe_{78}Si_{10}B_{12}$, respectively. This agreement suggests that the different kinds of metalloids have a similar contribution to the Hall resistivity.

The main reason for the large Hall resistivity as pointed out in (iv) may be related to the large value of specific resistivity due to the amorphous structure.

Finally, let us discuss the dpendences of $\Delta\rho/\rho$ and of R_s on the alloy composition.

For the crystalline 3*d*-ferromagnetic metals and alloys, there is a remarkable correlation between the $\Delta\rho/\rho$ (or the R_s) and the electron per atom ratio. That is, the values of $\Delta\rho/\rho$ for Ni-Fe[21,22], Ni-Co[22], Ni-Cu[22], Ni-Mn[13] and Ni-Fe-Co[13] alloys etc. are found to fall roughly on a curve of $\Delta\rho/\rho$ versus the mean number of $(3d+4s)$ electrons, N_e, and the curve shows a peak near $N_e=9.7$ *e/a*. With the appearance of this maximum in $\Delta\rho/\rho$, the R_s changes sign in the neighborhood of the same electron concentration of 9.7 *e/a*[23,24]. Smit[22], Berger[25,26] and Potter[27] have given a possible explanation for the singularities in the $\Delta\rho/\rho$ and the R_s appeared at 9.7 *e/a* in terms of the rigid band model. They have taken into account the transition probability of the anisotropic scattering of 4*s*-electrons, which abnormaly increases when the Fermi level reaches the energy gap caused by the spin-orbit coupling acting on the degenerate 3*d*-subbands.

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(16) F.E. Maranzana, Phys. Rev., **160** (1967), 421.
(17) E.P. Svirina and Yu. V. Nemchinov, Soviet Phys. Solid State, **9** (1967), 553.
(18) I.K. Kikoin, E.M. Buryak and Yu. A. Muromkin, Soviet Phys. Doklady, **4** (1959), 386.
(19) K.P. Belov and E.P. Svirina, Soviet Phys. Solid State, **8** (1966), 967.
(20) K.V. Rao, R. Malmhall, G. Backstrom and S.M. Bhagat, Solid State Commun., **19** (1976), 193.
(21) R.M. Bozorth, Phys. Rev., **70** (1946), 923.
(22) J. Smit, Physica, **16** (1951), 612.
(23) W. Jellinghaus and M.P. de Andres, Ann. Physik, **5** (1960), 187.
(24) S. Foner and E.M. Pugh, Phys. Rev., **91** (1953), 20.
(25) L. Beger, Physica, **30** (1964), 1141.
(26) L. Beger, Phys. Rev., **138** (1965), A1083.
(27) R.I. Potter, Phys. Rev., **B10** (1974), 4626.

On the other hand, according to Yamauchi *et al.*[28], the change of the mean ferromagnetic moment with N_e in the amorphous Fe, Ni and Co-based alloys seem to be also explicable on the basis of the rigid band model by supposing that the 3*d*-band is additionally filled by the *p*-electrons of metalloids.

If we apply the calculation performed by Yamauchi *et al.* to the present alloy systems, the electrons transferred from the metalloids to the metals is estimated to be 0.41 electrons per metal atom. Therefore, the electron concentration where the singularities in $\Delta\rho/\rho$ and R_s take place may change from 9.7 e/a for the crystalline alloys ($Ni_{85}Fe_{15}$ and $Ni_{70}Co_{30}$ for example) to 9.3 e/a for the amorphous alloy ($(Fe_{0.35}Ni_{0.65})_{78}Si_{10}B_{12}$). However, no singularity can be seen in the present results as shown in Figs. 4 and 8.

The discrepancy mentioned above seems to be caused by the difference between the band structures of the crystalline and amorphous states, even though the rigid band model can be applied to both states. That is, in the crystalline state, the degeneracy of 3*d*-subbands which essentially participate in the singularity of $\Delta\rho/\rho$ and R_s may exist on the symmetry axes or the symmetry planes of the Brillouin zone in the *k*-space[29]. In the amorphous state, the free electron model may be a good approximation for the explanation of the electrical phenomena[30] such as the temperature dependence of the electrical resistance, etc.; this implies that there are no symmetry axes specified in the *k*-space for the amorphous structure. Therefore, the effects of the band structure on the magneto-resistance and the Hall resistivity may mostly be smeared out.

Acknowledgements

The authors are greatly indepted to Professor Y. Nakagawa and Professor T. Masumoto for their support and encouragement in this study. We would also like to thank Mr. T. Takahashi for the supply of the specimens of the amorphous Fe-Ni-Si-B alloys used in this study.

(28) K. Yamauchi and T. Mizoguchi, J. Phys. Soc. Japan, **39** (1975), 541.
(29) J.O. Dimmock, Solid State Physics, Vol. **26** (1971), 184.
(30) G. Bergman, Solid State Commun., **18** (1976), 897.