Spontaneous Hall effect and resistivity of Fe-Co-Ni-base glasses

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The spontaneous Hall effect and dc resistivity have been measured at room temperature in the glassy alloy series $Fe_{80-x}Co_xB_{20}$ ($0 \leq x \leq 80$ at.%) and $Fe_{80-x}Ni_xB_{20}$ ($0 \leq x \leq 60$ at.%) and in $Co_{40}Ni_{40}B_{20}$ glass. The density-derived thicknesses used herein give smaller values of R_s and ρ than generally reported for metallic glasses. Because of the relatively large resistivities of metallic glasses, $\rho \simeq 10^{-4}$ Ωcm, the nonclassical side-jump mechanism is expected to dominate the spontaneous Hall effect. The magnitude of the side jump in $Fe_{80}B_{20}$ glass is comparable to that in crystalline Fe and Fe-base dilute alloys, viz. 10^{-8}cm. The spontaneous Hall conductivity γ_{Hs} shows a proportionality to the magnetostriction λ_s as predicted theoretically with a slope that is close to that observed in crystalline Fe-Ni alloys. The compositional dependence of γ_{Hs} is interpreted in terms of a split-band model in which charge is transferred from boron to the transition-metal d states in the glassy alloys. The applicability of the split-band model to these data implies that an intrinsic spin-orbit interaction involving the itinerant d electrons is effective here and not a spin–other-orbit interaction.

I. INTRODUCTION

The Hall effect (HE) in ferromagnetic materials contains not only the ordinary component due to the Lorentz force, but has also a much larger, spontaneous contribution due to the scattering of itinerant electrons, through spin-orbit interaction, from impurities and phonons. The Hall resistivity may be written[1]

$$\rho_H = R_o B + 4\pi R_s M_s , \tag{1}$$

where $B = H + 4\pi M$ and R_o and R_s are the ordinary and spontaneous Hall coefficients, respectively.

The spontaneous HE has been studied in several isolated transition-metal–metalloid (TM-M) glass compositions[2-6] and, heretofore, in only one complete series of TM-M glasses.[7] The present paper reports and interprets spontaneous HE and resistivity data for the first time in two series of glassy alloys, $Fe_{80-x}Ni_xB_{20}$ ($0 < x < 60$ at.%) and $Fe_{80-x}Co_xB_{20}$ ($0 < x < 80$ at.%), which have already been fully characterized in terms of their saturation moments[8] and magnetostrictions.[4,9,10] Employed herein is a different method of determining sample thickness which is more accurate than that believed to be in widespread use on metallic glasses. It is described, along with other experimental consideration, in Sec. II. In Sec. III, the Hall data and resistivities for the two series of glassy alloys are presented. Section IV discusses these results in terms of the magnitude of the side jump (Sec. IV A), the relation between the spontaneous Hall effect and magnetostriction (Sec. IV B), the occurrence of zeros in the spontaneous Hall effect and magnetostriction (Sec. IV C), and the interpretation of the data in terms of a split-band model (Sec. IV D).

II. EXPERIMENTAL

The metallic-glass samples were rapidly quenched (~10^5 K sec^{-1}) from the liquid phase to form continuous, noncrystalline ribbons.[11] The ribbon samples (typically 35 μm × 2 mm in cross section) were mechanically polished to improve surface flatness. Four-point resistivity and Hall resistivity were measured using a constant-current source and a digital voltmeter. Pressure contacts with copper wires were found to give accurate and reproducible results. The Hall voltages were recorded as a function of applied field up to ±20 kOe for both current directions. Because $R_s \gg R_o$ and because demagnetization gives $H = 4\pi M$ for $H \ll 4\pi M_s$, we have from Eq. (1) $\rho_H \approx R_s H$. Therefore, $R_s = (\partial \rho_H / \partial H)_{H=0}$.

Ribbon thicknesses were determined in two ways: with a micrometer (yielding a thickness t' and thus values for the resistivity and Hall coefficient of ρ' and R_s') and from the weight, width, length, and independently measured densities[8] (yielding a thickness t and thus ρ and R_s). The former method leads to greater experimental error than the latter, probably because of sample thinness and residual surface irregularity. This is evidenced by greater scatter in the ρ' and R_s' data than in the ρ and R_s data. It is important to note that for each sample $t/t' = 0.70 \pm 0.05$. ρ' and R_s' are generally found to be in better agreement than are ρ and R_s with values in the literature for comparable glassy alloys (See Sec. III). However, ρ, R_s, and γ_{Hs} are believed to be more accurate because of the greater reliability of the thickness measurement upon which they depend. Moreover, the γ_{Hs} values show a proportionality to λ_s (Ref. 12) (measured on the

same compositions)[4,9] that tends to support their accuracy rather than that of the γ'_{Hs} data (See Sec. III A). The values ρ, R_s, and γ_{Hs}, arising from density-derived thickness measurements, are reported herein.

III. RESULTS

The density-thickness-derived Hall coefficients and resistivities are displayed in Fig. 1 for the Fe-Co- and Fe-Ni-base glasses. One Co-Ni-base glass, $Co_{40}Ni_{40}B_{20}$, was measured and it showed $R_s = 0.16 \times 10^{-9}$ Ω cm/G and $\rho = 101$ μΩ cm. The quantities in Fig. 1 are generally smaller than values reported for comparable metallic glasses (Table I) because of the method used here (believed to be more accurate) for determining sample thickness. Because[2,5,6] R_s and[14,15] ρ are observed to be essentially independent of temperature in metallic glasses, the data of Fig. 1 are expected to be within 5% of their low-temperature values. $Ni_{80}B_{20}$ glass cannot presently be fabricated by rapid quenching, however, the values $R_s(o) \cong 0.18 \times 10^{-9}$ Ω cm/G and $R_s(290\text{ K}) = 0$ (because $T_c \approx 135$ K)[16] can be estimated from Fig. 1 by extrapolation.

It is often more instructive to discuss the Hall conductivity[17]

$$\gamma_H \equiv \rho_H/\rho^2, \quad \gamma_{Hs} = 4\pi R_s M_s/\rho^2. \tag{2}$$

Again, because of the temperature independence of R_s and ρ, we can calculate either $\gamma_{Hs}(0)$ or $\gamma_{Hs}(290\text{ K})$ from R_s by using the known values of either $M_s(0)$ or $M_s(290\text{ K})$[8] in Eq. (2).

The compositional dependence of $\gamma_{Hs}(290\text{ K})$ is shown in Fig. 2 for both series of glasses and for $Co_{40}Ni_{40}B_{20}$ glass. For both series of glasses, γ_{Hs} decreases with the addition of Co or Ni to $Fe_{80}B_{20}$ and in neither case does γ_{Hs} appear to change sign before the end of the series. These trends are consistent with preliminary results previously reported on five members of the Fe-Co-B series[4] and with recent measurements by Obi *et al.* on $(FeCoNi)_{78}B_{12}Si_{10}$ glasses.[7]

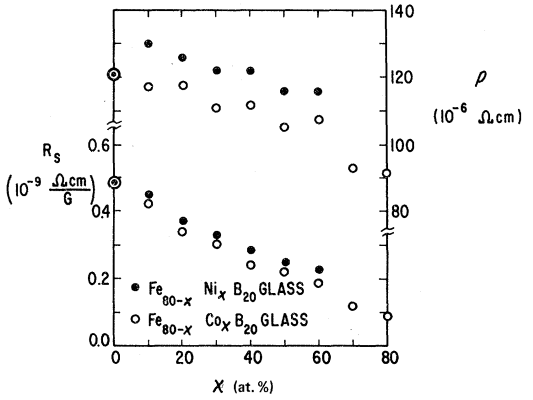

FIG. 1. Spontaneous Hall coefficients R_s and resistivities ρ at 290 K for $Fe_{80-x}Ni_xB_{20}$ glasses ($0 \leq x \leq 60$ at. %) and for $Fe_{80-x}Co_xB_{20}$ glasses ($0 \leq x \leq 80$ at. %). Those $(FeNi)_{80}B_{20}$ glasses in the range $65 \lesssim x \lesssim 80$ are not currently fabricable by rapid quenching.

The magnitudes of the spontaneous Hall conductivities $\gamma_{Hs}(295\text{ K})$ are displayed in Table II for crystalline Fe, Co, and Ni as well as for the related glassy alloys studied here. The noncrystalline alloys show comparable but consistently smaller values of γ_{Hs} compared to the crystalline materials. For $Ni_{80}B_{20}$ glass $\gamma_{Hs}(295\text{ K}) = 0$ because $T_c \approx 135$ K.[16]

IV. DISCUSSION

A. Side jump

It is observed that, for a given band structure, the variation of R_s with resistivity generally follows the expression[1]

$$R_s = a\rho + A\rho^2. \tag{3}$$

TABLE I. Comparison of present results with spontaneous Hall coefficients reported for other noncrystalline alloys.

This work		Other metallic glasses		
Alloy (at. %)	R_s (290 K) (10^{-9} Ω cm/G)	Alloy (at. %)	R_s (290 K) (10^{-9} Ω cm/G)	Ref.
$Fe_{80}B_{20}$	+0.49	$Fe_{80}P_{13}C_6$	+0.6	2
		$Fe_{80}B_{20}$	+0.7	4
		$Fe_{78}Si_{10}B_{12}$	+0.62	7
$Fe_{40}Ni_{40}B_{20}$	+0.29	$Fe_{40}Ni_{40}P_{14}B_6$	+0.335	3
		$Fe_{40}Ni_{40}B_{20}$	+0.35	6
		$Fe_{40}Ni_{40}Si_{10}B_{12}$	+0.41	7
$Co_{80}B_{20}$	+0.09	$Co_{80}B_{20}$	+0.12	4
		$Co_{78}Si_{10}B_{12}$	+0.26	7
$Ni_{80}B_{20}$	~0	Ni	−0.1	13
		$Ni_{78}Si_{10}B_{12}$	~0	7

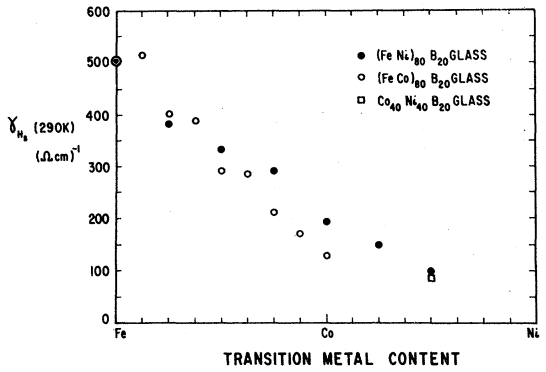

FIG. 2. Spontaneous Hall conductivites γ_{H_s} (290 K) for Fe-Co-Ni-B glasses as a function of TM content.

The first term describes classical asymmetric scattering of charge carriers from impurities and is characterized by a spontaneous Hall angle ϕ_s:

$$\tan\phi_s = 4\pi R_s M_s/\rho = 4\pi M_s/a\,. \tag{4}$$

The second term in Eq. (3) describes the nonclassical side-jump mechanism[19-21] and is characterized by a finite lateral displacement Δy of the conduction electron trajectory. For a single band[21]

$$\Delta y = 4\pi M_s A\left(\frac{\hbar k_F}{ne^2}\right) = \frac{4\pi R_s M_s}{\rho 2}\left(\frac{\hbar k_F}{ne^2}\right)\,, \tag{5}$$

where the symbols in parentheses have their usual meanings. The side-jump mechanism is expected to dominate the spontaneous HE when the ratio of the Fermi wavelength λ_F to the mean-free path Λ becomes appreciable.[19] This is the case for metallic glasses. However, the ρ^2 relation is not easily confirmed experimentally in metallic glasses because of the nearly temperature independence of[2,5,6] R_s and[14,15] ρ and because small variations in impurity content have little effect on the resistivity which is large due to the overriding disorder. Nevertheless, Fig. 3 shows that the spontaneous

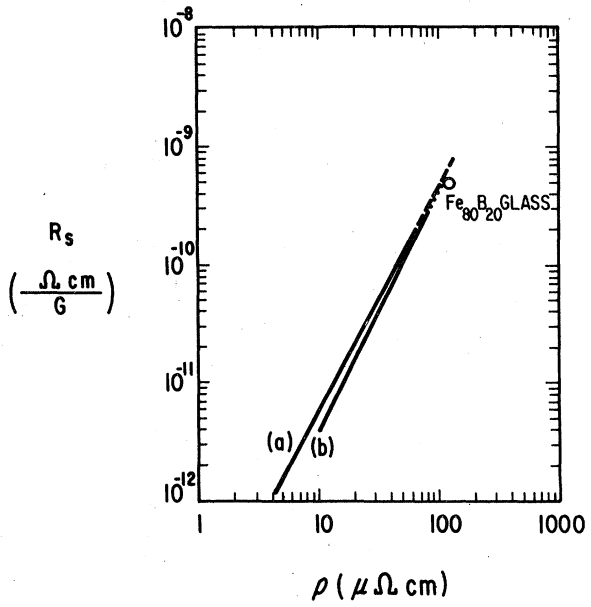

FIG. 3. Spontaneous Hall coefficient vs resistivity (log scales) for crystalline Fe and Fe-base dilute alloys [(a) Ref. 22 and (b) Ref. 18] and for $Fe_{80}B_{20}$ glass at 290 K.

Hall coefficient for noncrystalline $Fe_{80}B_{20}$ lies close to the ρ^2 dependence observed in crystalline Fe and Fe-base dilute alloy.[18,22]

From Fig. 3 it is evident that the magnitude of the side jump, $\Delta y \propto R_s/\rho^2$ [Eq. (5)] in metallic glasses is comparable to that observed in crystalline TM alloys, viz., $\Delta y \approx 10^{-8}$ cm. Specifically, for $Fe_{80}B_{20}$ glass, $4\pi M_s A = \gamma_{Hs} = 506\ (\Omega\,\mathrm{cm})^{-1}$ and assuming $k_F \approx 10^8\ \mathrm{cm}^{-1}$ and $n \approx 2\times 10^{22}\ \mathrm{cm}^{-3}$, Eq. (5) gives $\Delta y = 1\times 10^{-8}$ cm. The agreement of this value with that observed in crystalline TM alloys is not unexpected because of the predicted insensitivity of the side jump to the strength and range of the scattering potential.[19,20]

TABLE II. Spontaneous Hall conductivities γ_{Hs} (290 K) for three of the metallic glasses from the present work compared with those for related crystalline transition metals.

This work		Crystalline transition metals		
Metallic glass	γ_{Hs} (290 K) $(\Omega^{-1}\mathrm{cm}^{-1})$	Metal	γ_{Hs} (290 K) $(\Omega^{-1}\mathrm{cm}^{-1})$	Footnote
$Fe_{80}B_{20}$	+506	Fe	+1200	(a)
			+800	(b)
$Co_{80}B_{20}$	+98	Co	+700	(a)
			+500	(b)
$Ni_{80}B_{20}$	~0	Ni	−500	(c, d)

[a] F. P. Beitel and E. M. Pugh, Phys. Rev. 112, 1516 (1951).
[b] Reference 18.
[c] S. Foner and E. M. Pugh, Phys. Rev. 91, 20 (1953).
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B. Spontaneous Hall effect and magnetostriction

Spontaneous Hall effect is related to magnetostriction because of the common origin of these phenomena in spin-orbit interaction. A band theory for these effects suggests that a material-independent proportionality should exist between γ_{Hs} and the magnetostriction λ_s.[12] Approximately the same ratio, $\gamma_{Hs}/\lambda_s \simeq 20\times10^6$ $(\Omega\,\mathrm{cm})^{-1}$, is observed in three split-band crystalline alloy systems: Fe-Ni, Fe-Ni-Cu, and Ni-Cu. A similar proportionality is observed for the split-band $Fe_{80-x}Ni_xB_{20}$ glasses studied here (Fig. 4). The magnetostrictions for these glasses were reported elsewhere.[4,9] The slope for the density-thickness-derived quantities (solid line, Fig. 4) is approximately $\gamma_{Hs}/\lambda_s \approx 13\times10^6$ $(\Omega\,\mathrm{cm})^{-1}$, whereas micrometer-thickness-derived quantities give $\gamma'_{Hs}/\lambda_s \approx 9\times10^6$ $(\Omega\,\mathrm{cm})^{-1}$. This difference supports the expectation that the former transport properties (λ_{Hs}, R_s, and ρ) are more accurate than the latter (γ'_{Hs}, R'_s, and ρ'). A more general relation would be expected to exist between γ_{Hs} and $G\lambda_s$ (G is the shear modulus). Because the stiffnesses of metallic glasses are generally 10%–20% below those of the related crystalline transition metals[23] the behavior of the Fe-Ni-base glasses would be even closer to that of the crystalline FeNi alloys in such a comparison as compared with Fig. 4. The data for Fe-Ni-B glasses show a positive γ_{Hs} intercept similar to that observed in crystalline Fe-Ni (Fig. 4). That is, if a spontaneous HE zero exists at all in this glassy alloy series, it is expected to be displaced to the high electron-atom side of the $\lambda_s = 0$ composition, as in crystalline Fe-Ni.

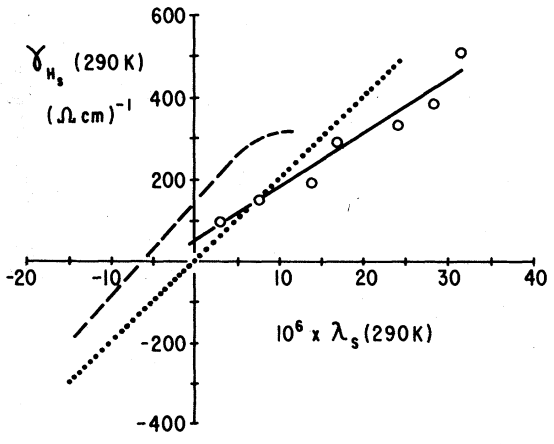

FIG. 4. Spontaneous Hall conductivity vs magnetostriction at 290 K. Data points and unbroken line, Fe-Ni-B glasses; broken line, crystalline Fe-Ni alloys (Ref. 12); dotted line, theory (Ref. 12).

C. Zeros in γ_{Hs} and λ_s

The sign changes in γ_{Hs} and λ_s in crystalline Fe-Ni alloys (Fig. 4) have been considered to be related to similar sign changes which occur in crystalline Co-Ni alloys at approximately the same electron concentration, 27.75 ± 0.05 e/atom, as in the former series.[17] These singularities have limited parallels in the Fe-Co-Ni-B glasses.

The compositional variations of the zeros in γ_{Hs}(290 K) and λ_s for crystalline and noncrystalline TM alloys are shown in Fig. 5. Dashed lines show approximate zeros obtained by extrapolation. Magnetostriction zeros in the iron-rich crystalline alloys have been omitted.

The $\gamma_{Hs} = 0$ line for crystalline alloys was constructed from end-point (binary) data in the literature[18] (see footnotes to Table II). In the glasses, $\gamma_{Hs} = 0$ only at or beyond (higher electron concentration than) the composition $Ni_{80}B_{20}$.

The $\lambda_s = 0$ lines shown for crystalline material were taken from data on thin films[24] and are similar to bulk data with respect to the features of interest here.[25] The course of the $\lambda_s = 0$ line for $(\text{transition metal})_{80}B_{20}$ glasses was taken from Refs. 26 and 27.

From Fig. 5 it is seen that the compositions for which $\gamma_{Hs} = 0$ and $\lambda_s = 0$ in the crystalline Fe-Ni alloys are shifted away from Fe in the corresponding glasses. This shift is probably due to the presence of the metalloid atoms rather than the glassy structure. R_s has been measured in noncrystalline $Ni_{1-x}Au_x$ $(0.15 < x < 0.5)$ alloys and found to be negative for all x in this range as well as for $x = 0$ by

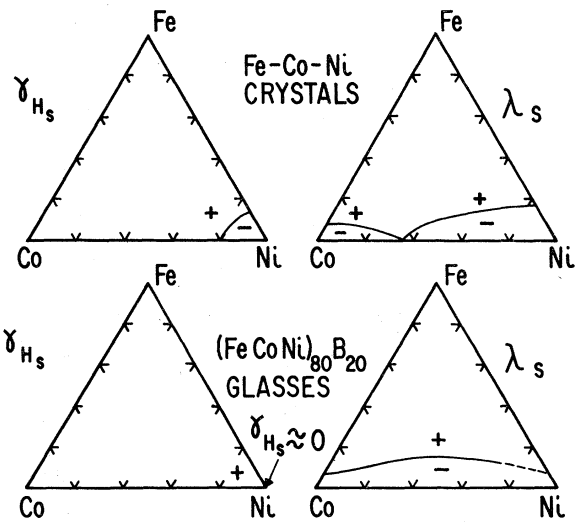

FIG. 5. Triangular diagrams showing γ_{Hs} (290 K) $= 0$ and $\lambda_s = 0$ lines for crystalline and noncrystalline Fe-Ni-Co alloys. Estimated zeros are shown by broken lines.

extrapolation.[13] Magnetic moments[8] and other data[16] on TM-M glasses also suggest an appreciable charge transfer from the metalloid atoms to the TM d bands (e.g., an average of 1.6 e/B atom). The occurrence of $\gamma_{Hs}=0$ at higher electron concentration than $\lambda_s=0$ is seen in Fig. 5 for both crystalline and noncrystalline Fe-Ni-base alloys.

D. Split-band model

The singularities shown in Fig. 5 may be considered to result from the coincidence of the Fermi energy ϵ_F with some feature in the band structure at an energy ϵ_B which causes $\gamma_{Hs}=\lambda_s=0$.[12,17,28] If a rigid-band model were to be used to interpret the shift in the $\gamma_{Hs}=0$ and $\lambda_s=0$ compositions on going from the crystalline to the noncrystalline TM alloys, the direction of charge transfer would have to be *away from* the TM d states in order to explain the direction of the shift [Fig. 6(a)]. On the other hand, a more realistic split-band model[12,29,30] [Fig. 6(b)] interprets the direction of the shift consistently with the direction of charge transfer inferred from other experiments.[8,16]

Such a split-band model, applicable on the Fe-Ni sides of the triangular diagrams in Fig. 5, has been developed by Berger to describe the signs of γ_{Hs} and λ_s and the compositional variations of the $\gamma_{Hs}=\lambda_s=0$ lines. Deformation potential theory[28,30] shows these quantities to be proportional to $\langle L_z(\epsilon_F)\rangle$, the expectation value of the z component of orbital angular momentum at the Fermi energy.[31] In a split-band model, $\langle L_z(\epsilon_F)\rangle=0$, when ϵ_F lies in the gap, or at the boundary between, the top of the Ni $3d\downarrow$ band and the bottom of the Fe $3d\downarrow$ band, ϵ_B [Fig. 6(b)]. At the iron concentration C_{Fe} for which $\epsilon_F=\epsilon_B$, the number of states in the Fe $3d\downarrow$ band is equal to the total number of $3d\downarrow$ holes:

$$5C_{\mathrm{Fe}}=2.55C_{\mathrm{Fe}}+0.55C_{\mathrm{Ni}}\,,\qquad C_{\mathrm{Fe}}+C_{\mathrm{Ni}}=1\,. \tag{6}$$

Equation (6) predicts $C_{\mathrm{Fe}}=0.18$ for $\gamma_{Hs}=\lambda_s=0$ in crystalline Fe-Ni alloys, in excellent agreement with experimental observation.[17,28] Generalizations of this model to other crystalline split-band systems are similarly successful.[12]

In the Fe-Ni-base glasses the effective charge transfer causes Eq. (6) to be generalized to

$$5C_{\mathrm{Fe}}=2.55C_{\mathrm{Fe}}+0.55C_{\mathrm{Ni}}-1.6C_{\mathrm{B}}\,,\qquad C_{\mathrm{Fe}}+C_{\mathrm{Ni}}=0.8,\quad C_{\mathrm{B}}=0.2\,. \tag{7}$$

That is, $\epsilon_F=\epsilon_B$ occurs for lower C_{Fe} in the Fe-Ni-B glasses than in the crystalline Fe-Ni alloys. Equation (7) predicts $C_{\mathrm{Fe}}\approx 0.06$ for $\gamma_{Hs}=\lambda_s=0$. λ_s appears to go to zero near this concentration (Fig. 5), however, as in the crystalline alloys, $\gamma_{Hs}=0$ occurs, if at all in the glasses, at slightly higher electron concentrations than $\lambda_s=0$, and apparently outside the range of the Fe-Ni-B glasses studies here (Fig. 5). The slope and intercept of the $\lambda_s=0$ line are well predicted in the Ni-rich Fe-Co-Ni-B glasses by a generalization of Eq. (7).[27]

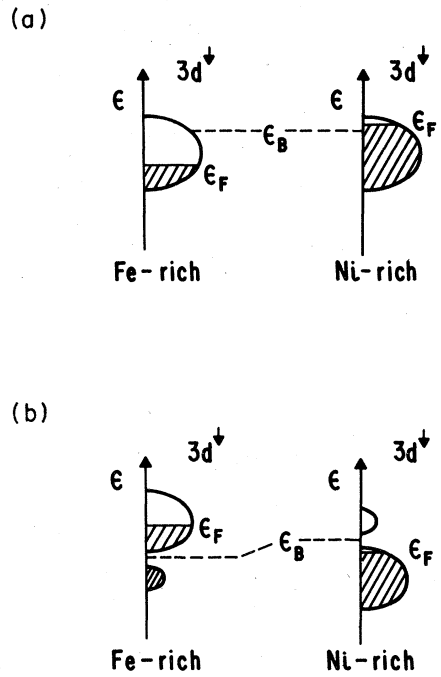

FIG. 6. Band models for TM alloys showing minority-spin ($3d\downarrow$) state densities. (a) Rigid-band model; ϵ_B is an energy at which it is assumed $\langle L_z\rangle=0$. (b) Split-band model; ϵ_B is an energy for which $\langle L_z\rangle=0$.

The applicability of the split-band model to γ_{Hs} and λ_s in Fe-Ni alloys implies that the orbital angular momentum involved in the spin-orbit interaction is that of the $3d$ electrons. Therefore, the possible interactions causing these effects would be $L^{3d}S^{3d}$ and $L^{3d}S^{4s}$ but not the conduction-electron-orbit–localized-spin interaction[32] which obtains in the rare-earth metals.[33] Because the $L^{3d}S^{4s}$ interaction is inherently the weaker of the two possible ones and because the spin polarization of the conduction electrons is a small fraction of that of the $3d$ electeons,[34] the dominant interaction is probably the intrinsic $3d$ spin-orbit interaction,[19,35] $L^{3d}S^{3d}$.

V. CONCLUSION

The spontaneous Hall coefficient and dc resistivity of Fe-Co-Ni-B glasses have been measured. The use of density-derived thickness results in smaller values for R_s and ρ than generally reported for metallic glasses. The values of R_s and ρ for $Fe_{80}B_{20}$ glass agree well with the $\sim\rho^2$ dependence

observed for crystalline Fe and Fe-base dilute alloys. The magnitude of the side jump in metallic glasses is found to be comparable to that in related crystalline transition metals. γ_{Hs} shows a proportionality to λ_s in the Fe-Ni-base glasses that agrees well with theory and with results for crystalline Fe-Ni alloys. The compositional variations of γ_{Hs} and λ_s in the Fe-Ni base glasses support the split-band model for Fe-Ni alloys and are consistent with charge transfer from the metalloid atoms to the TM d states. The applicability of the split-band model to γ_{Hs} and λ_s in the Fe-Ni-base metallic glasses implies the dominance of intrinsic $3d$-spin–$3d$-orbit interaction and not a spin–other-orbit interaction.

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