Sign reversal of the extraordinary Hall coefficient in Ni-Fe-Cr and Ni-Fe-V alloys

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The extraordinary Hall effect and dc resistivity have been measured at 300 and 77 K for several alloys in the Ni-Fe-Cr and Ni-Fe-V series. From our data, the line where extraordinary Hall conductivity (γ_{HS}) changes sign has been established for the first time in the ternary diagram for both the systems. An attempt to explain the sign change in terms of the split-band model reveals that the experimental $\gamma_{HS}\simeq 0$ line lies far away from the one predicted by this model, and contrary to expectation, γ_{HS} and λ_s (coefficient of linear magnetostriction) do not change sign simultaneously. Also, the position of the already established $\lambda_s\simeq 0$ line can be explained only when the experimentally observed nonlinear compositional dependence of $\bar{\mu}$ (average number of Bohr magnetons per atom) is empirically incorporated into this model. Though the $\gamma_{HS}\simeq 0$ line is essentially in disagreement with the model, γ_{HS} and λ_s still show a proportional relationship, as predicted by the model. Some possible reasons for the disagreement with the model have been suggested in terms of the slow variation of γ_{HS} with Cr or V concentrations and the existing band-structure calculations for the corresponding binary alloys.

I. INTRODUCTION

It is well known that the Hall effect in a nonmagnetic material is basically a manifestation of the Lorentz force. But in the case of ferromagnets or materials having some kind of magnetic ordering, an additional term is introduced, and the Hall resistivity is given by the sum of these two components:

$$\rho_H = R_0 B + R_s M_s \tag{1}$$

(SI units), where R_0 is the ordinary (Lorentz) Hall coefficient and R_s is the extraordinary (additional) one; other terms have their usual meaning. During the last thirty years there has been a burst of theoretical activity that aims to establish plausible explanation for this extraordinary term. The present understanding is that some kind of spin-orbit interaction is responsible for its origin, and no single theory is capable of explaining all the characteristics of this effect in diverse kind of materials. Under the influence of spin-orbit (henceforth referred to as s.o.) interaction, the scattering of the current carriers by various scattering centers (impurities, phonons, etc.) assumes a left-right asymmetry[1] ("skew scattering") with respect to the plane containing the electron's spin and its incident velocity, or the carriers experience a transverse displacement[2] ("side jump"). Several theories, based both on "itinerant"[3–8] and "localized"[9–12] models, have been proposed so far; however, the quantitative understanding of the phenomenon still remains incomplete in many cases. Good reviews on this account can be found elsewhere.[13,14]

Another aspect of the problem is the sign of the extraordinary Hall coefficient R_s. It is well known that R_s is positive in Fe and Co and negative in Ni at room temperature. On the other hand, R_s changes sign upon alloying in various Ni- and Fe-based alloys.[15,16] Although the sign reversal of R_s in binary systems has been well studied, little work has been done on the ternary alloy systems. Except for crystalline Ni-Fe-Cu[17] and amorphous $Fe_{80-x}Co_xB_{20}$ ($0\le x\le 80$ at. %), $Co_{40}Ni_{40}B_{20}$, and $Fe_{80-x}Ni_xB_{20}$ alloys,[18] very few ternary systems have been taken up so far. So it was felt to be worthwhile to establish the $R_s\simeq 0$ line experimentally for the two ternary systems, Ni-Fe-Cr and Ni-Fe-V. The special interest in these systems lies in the theoretical prediction for the $R_s\simeq 0$ compositions by Ashworth *et al.*[17] in their "split-band" model (which will be discussed in detail in Sec. II). Of the two, Ni-Fe-Cr system appeared to be of additional interest because of the already existing magnetization data by Men'shikov *et al.*[19] They have reported considerable nonlinearity in $\bar{\mu}$ (the average number of Bohr magnetons per atom) versus concentration curves. But in the split-band model, the inherent assumption is that the number of electrons or holes contributed by one atom of Cr or V, is concentration independent (implying a linear $\bar{\mu}$-versus-concentration curves). With the above ideas in mind, we have measured ρ_H, M_s, and resistivity in several alloys of the two ternary systems at 77 and 300 K, and from these data (as will be presented in Sec. IV) we have been able to establish the $\gamma_{HS}\simeq 0$ ($\gamma_{HS}=R_sM_s/\rho^2$) line. Our data clearly show that the experimentally observed $\gamma_{HS}\simeq 0$ lines in the ternary diagram lie far away from those predicted by the split-band model for both the systems. Some possible reasons for the disagreement with the split-band model will be discussed in Sec. IV.

II. THEORETICAL BACKGROUND

A. Sign of the extraordinary Hall coefficient (R_s)

The first successful theory to account for the sign of R_s in common ferromagnetic materials such as Fe, Ni, and Co was due to Kondorskii.[20] It is well known that both R_s and the ordinary Hall coefficient R_0 have the same sign in Fe and Ni (both are positive in Fe and negative in

Ni at room temperature). It was earlier assumed that it is only the carrier type which determines the sign of both R_s and R_0. But in the case of Co, R_s changes from the low-temperature negative value to a room-temperature positive value at around 220 K, whereas R_0 remains negative throughout.[21] Later measurements on purer single crystals of Fe also show that R_0 changes sign around 77 K (from negative to positive high-temperature values), although R_s remains positive throughout.[22] These facts along with the data for R_0 and R_s in some alloys [e.g., Ni-Fe, where R_s changes from negative to positive values around 18 at.% Fe whereas R_0 changes sign only around 95 at.% Fe (Ref. 23)] clearly show that R_0 and R_s can have different signs, in general. Guided by these, Kondorskii[20] suggested a theory which involves a detailed knowledge of the Fermi surface of the metal to predict the sign of R_s.

According to Kondorskii, in ferromagnets, due to the exchange splitting of the spin-up and spin-down bands, the occupation of the two subbands are different. The contribution to R_s by the carriers in different subbands (i.e., with different spins) will be different; the carrier type (electronlike or holelike) in a particular subband will be decided by the topology of that part of the Fermi surface. Therefore, the overall sign of R_s will be determined by the dominating carriers and their spins. Accordingly, Kondorskii formulated that R_s will be positive if the dominant carriers are electronlike and are from the up-spin band and negative if they are electronlike and are from the down-spin band. For holes, the signs will be just the opposite of those for electrons. Using the above criterion and from the knowledge of the Fermi surface of Ni, Kondorskii found that the main contributions in Ni are from electronlike carriers from the unfilled down-spin band and hence R_s is negative. But for iron, the predominant current carriers are electrons from the up-spin band and holes from the down-spin band, both of which contribute to a positive value of R_s.

Though successful in explaining the signs of R_s in Fe and Ni, the application of this kind of theory requires detailed knowledge of the Fermi surface, which is often not available, especially for alloys. To explain the sign change of R_s with composition in various Ni- and Fe-based alloys, Ashworth *et al.*[17] proposed a split-band model, which we will discuss in detail in the next section.

B. Split-band (SB) model and the sign change of R_s in alloys

It was discovered long ago[15] that the extraordinary Hall constant R_s changes sign in Ni-Fe and Ni-Co alloys almost for the same electron-to-atom ratio ($\sim$27.7). This led to a theory,[15] under the rigid-band approximation, in which the sign change of R_s was associated with the Fermi level crossing some degeneracy in the Ni band. However, subsequently it was found in Ni-Fe-Cu alloys that no such correlation exists between R_s and the electron-to-atom ratio. To explain these results, Ashworth *et al.*[17] suggested an extreme model, known as a split-band (SB) model, where all the three constituents have distinctly separate subbands in the ternary alloy with the bands for Cu lying at the bottom and those for Fe at the top. The sign change of R_s was identified with the Fermi level crossing the top of the Ni spin-down band (or where the spin-down bands of Ni and Fe meet). This model was further extended for Ni-Fe-M (M=Cr, V, Ti, W, Mo, etc.) alloys, and the band picture suggested for them is as shown in Fig. 1. Of the three constituents, the bands for Ni are at the bottom since it is most attractive to electrons. On the other hand, due to the large valence difference between M and Ni, the bands for M are split from the host and are formed on top. We call T the point where the spin-down subbands for Fe and Ni meet. According to Berger's model, the sign change of R_s is associated with the Fermi level (E_F) crossing T. Now the total number of states in a given $3d$ subband is 5 times the concentration of the corresponding atoms. Hence the Fermi-level crossover will take place when the total number of holes in the ternary alloy system is equal to $5C_{\mathrm{Fe}}$ (C_{Fe} equals the concentration of Fe atoms); i.e., when

$$5C_{\mathrm{Fe}}=0.55+2C_{\mathrm{Fe}}-(10+Z)C_M\ ,$$

or

$$3C_{\mathrm{Fe}}+(10+Z)C_M=0.55\ , \tag{2}$$

where 0.55 is the number of holes per atom in Ni, C represents concentration, and Z is the valence difference between M and Ni (e.g., $Z=-4$ for Cr and -5 for V). It is to be noted that for counting the contribution of electrons or holes by Cr or V atoms, instead of simply taking the valence difference (Z) between Ni and Cr or V, a factor $10+Z$ has been used. Because of the position of the Cr or V bands above E_F, they empty out all electrons into the Ni spin-down band, the number of which is given by $10+Z$.[24] But instead of Ni-Fe-Cr or Ni-Fe-V alloys, if one considers Ni-Fe-Cu system, then the subbands for Cu will lie below those of Ni. In that case, band filling will be guided by the valence difference ($Z=+1$ in this case) only and so the sign change in the Ni-Fe-Cu system will be determined by Eq. (2) with the term $(10+Z)C_M$ replaced by simply C_{Cu}.

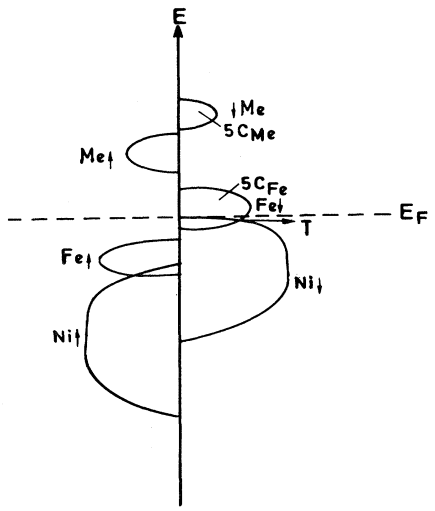

FIG. 1. Schematic density of states for ternary Ni-Fe-M (M=V, Cr, etc.) alloy systems according to the split-band model.

The reason for the sign change in R_s is the following.[16] We recall that some s.o. interaction is necessary for the origin of the extraordinary Hall effect. Now the s.o. coupling parameter, in contrast to that of free electrons ($\lambda_{\text{s.o.}}=\hbar^2/2m^2c^2$), for $3d$ band electrons is given by[25]

$$\lambda_{\text{s.o.}}(E_F)=A_{\text{s.o.}}Xd^2\sum_n \frac{|\mathcal{M}_n|^2}{E_n-E_F}, \tag{3}$$

where $A_{\text{s.o.}}$ is the atomic s.o. parameter for $3d$ electrons, X ($\simeq 0.1$) is an overlap integral between nearest-neighbor atomic $3d$ states, d is the nearest-neighbor distance of atoms, E_n is the energy of a band state n, and $\mathcal{M}_n$ are matrix elements. As can be seen from Eq. (3), the sign of $\lambda_{\text{s.o.}}$ and hence R_s will be determined by the quantity in the denominator. When the Fermi level E_F is in the upper half of the band, the states at E_F mostly mix up with those of lower energy, thus rendering the denominator E_n-E_F effectively negative. The case is reversed when E_F is at the lower half of a band. Hence R_s should change sign when E_F crosses the boundary between the two subbands.

Such a change of sign has been experimentally observed in various binary and ternary alloys. According to Eq. (2) the sign change in binary Ni-Fe alloys should occur when $3C_{\text{Fe}}=0.55$, i.e., around 18 at.% Fe, which is in close agreement with the experimental findings.[26] In Fe-V alloys, although a sign change is not expected since both the spin-up and spin-down bands are partially full, still some anomaly was observed[27] around the composition predicted by the SB model. The other systems where the SB model has been found successful are Ni-Fe-Cu,[17] amorphous $Fe_{80-x}Co_xB_{20}$ ($0\le x\le 80$ at.%), $Co_{40}Ni_{40}B_{20}$, $Fe_{80-x}Ni_xB_{20}$ ($60\le x\le 0$),[18] etc.

The split-band model is just an extension of the virtual bound-state model,[28] applied to fairly concentrated alloys. This has good theoretical support in the coherent-potential approximation (CPA) calculations for the band structure of various Ni- and Fe-based alloys by Hasegawa and Kanamori.[29–31] According to the CPA calculations, distinctly separate bands for constituents A and B are possible in a binary alloy $A_{1-x}B_x$, if the average energy of an electron at site A (E_A) and site B (E_B) differs by more than the bandwidth W of the host metal.[32] Usually this condition is fulfilled if the valency difference between the constituents is sufficient (≥ 2). But in the case of ferromagnetic materials the energy at a site i ($i=A,B$) is made of (i) electrostatic and (ii) exchange parts. Therefore, large valency differences between the host and the impurity may not always ensure complete splitting since the two energies may be of different signs. For the same reasons the difference E_A-E_B will also be different for spin-up and spin-down electrons. As for example in Ni-Fe alloys, $E_{\text{Fe}}-E_{\text{Ni}}=-0.06$ for the spin-up electrons, and for the spin-down electrons it is 0.56, given in units of half the bandwidth.[30] Therefore, due to the addition of Fe in Ni, the spin-up band remains almost unaltered, whereas the spin-down band gets considerably deformed and almost splits up into two different subbands. In this context, the band picture shown in Fig. 1 appears to be somewhat extreme since the spin-up bands for Fe and Ni have been shown to be separate. However, since the spin-up bands are full and lie below E_F, it is of little importance in the present case whether they are split or remain as a single band.

There are fairly good direct experimental evidences in support of such band splittings. Ultraviolet photoelectron spectroscopy (UPS) and reflectivity data for Cu-Ni alloys[33] show a rapid increase in the number of states between the top of the Cu d band and the Fermi level with the addition of Ni, signifying the growth of a separate band for Ni. Later measurements on the same system[34] not only confirm the above findings, but also show a qualitative agreement with the CPA calculations.[35] UPS studies even in amorphous Cu-Zr and Pd-Zr (Ref. 36) alloys show the splitting of the d bands, although the shapes of the d bands are completely different from those of the constituent elements.

In addition to R_s, the following parameters are also expected to show some anomaly around $R_s\simeq 0$ compositions.

(i) A minimum in the electronic specific heat coefficient is expected as the Fermi level crosses a minimum in the density of states and has been experimentally observed in Ni-Fe (Ref. 37) and Fe-V (Ref. 27) (a quasiminimum) alloys.

(ii) The linear coefficient of magnetostriction (λ_s) should also change sign at the same composition since according to the deformation-potential theory[38] of magnetostriction, λ_s depends on the average z component of the orbital angular momentum [$\langle L_z(E_F)\rangle$], which changes sign with the Fermi level crossing the boundary between the two subbands. Experimental evidence[16] for a number of binary and ternary alloys, both crystalline and amorphous, lends support to this hypothesis.

(iii) A maximum in the ferromagnetic anisotropy of resistance has also been observed in several binary[15] and a few ternary[17] alloys and a suitable explanation in the framework of the SB model is found.[17]

III. EXPERIMENTAL

About a dozen alloys of both the ternary series (Ni-Fe-Cr and Ni-Fe-V) were prepared by induction-melting of the required amount of constituent elements of "Specpure" grade, obtained from Johnson-Mathey, Inc. (England). A few samples (viz sample numbers 9, 24, and 29 in the Cr series and 18, 19, 20, and 30 in the V series) were obtained from A. Sinha. The furnace-cooled ingots were homogenized at 1150 °C for 48 h under argon atmosphere. Through swaging and cold rolling, samples of the required shapes and sizes were obtained which were finally annealed at 900 °C for 24 h and water-quenched. Final compositions of the alloys were determined by using spectroscopic methods and also in a few cases by chemical analysis.

A special cryostat was designed and fabricated for the measurements of Hall effect and resistivity from 77 K to about 473 K. In order to remove the uncertainty in thickness measurements associated with normal soldered contacts, a special type of pressure contact was used. This also enabled us to reduce the resistive voltage, which is a factor due to the misalignment of the Hall probes, to a low-level ($\sim\mu$V), thereby improving the resolution of

measurement without the use of any special circuitry. Details of the cryostat and sample holder can be found elsewhere.[39]

Standard four-probe dc technique was used for the present measurements, and special attention was given to maintain low-noise level ($\sim$nV). The transverse voltage was measured by a model no. 148 nanovoltmeter (Keithley Instruments Inc.) and the corresponding Hall voltage was found by reversing both the current and magnetic field directions and taking their average with proper regard to signs. Measurements in magnetic fields up to 18 kOe (provided by a 15-in. Varian Associate electromagnet) enabled us to extract the extraordinary part from the normal one by taking the intercept of the high-field straight-line part of the Hall voltage, extrapolated to zero field, as is customarily followed. The sign of the Hall voltage was determined with respect to a standard Ni sample whose Hall coefficient is negative.

The results for R_sM_s presented in this paper are the averages over several measurements of the same piece of a given sample and where necessary on different pieces. The scatter of the results were typically around $\pm 2\%$ and the total error in R_sM_s was estimated to be less than 4% (main contribution being through the thickness measurement).

IV. RESULTS AND DISCUSSION

A. Sign reversal of R_s in Ni-Fe-Cr alloys

Table I shows the data for R_sM_s, resistivity ρ, and the extraordinary Hall conductivity $\gamma_{HS}=R_sM_s/\rho^2$ for the various alloys studied. As has been pointed out by Berger,[2] since in concentrated alloys the side-jump mechanism is more important (implying $R_s \propto \rho^2$), it is more meaningful to communicate in terms of the extraordinary Hall conductivity γ_{HS} rather than R_s, since γ_{HS} is almost temperature independent except for a weak dependence through M_s. Accordingly, γ_{HS} is a better parameter when a comparison is necessary among various compositions. The error in resistivity mostly comes from dimension measurements, of which the smallest quantity, i.e., the thickness, contributes the maximum. In most of the cases, the total error in ρ was of the order of 3%. On the other hand, γ_{HS}, being a function of ρ^2, inherits maximum uncertainty through ρ; the typical error in γ_{HS} was about 10%. It can be seen from this table that out of the fifteen alloys studied, seven have positive Hall conductivity. For some of the alloys, the values of γ_{HS} are slightly different from the results presented[40] at the initial stages of this investigation. The results for sample nos. 9 and 24, within experimental uncertainty, are in agreement with the data of Sinha and Majumdar.[41] For sample no. 50, since the temperature of measurement was very close to T_c, except for the sign, not much significance should be attached on its absolute value of γ_{HS}.

Using the positive and negative values of γ_{HS} at 77 K one can easily draw the $\gamma_{HS}\simeq 0$ line in the ternary phase diagram of this system. Figure 2 shows such a diagram along with the previously existing zero line for λ_s (Ref. 42) and the one predicted from the SB model [Eq. (2)]. The numbers associated with the solid circles represent the sample numbers as given in Table I and those in the brackets represent γ_{HS} ($10^3\ \Omega^{-1}\,m^{-1}$) values at 77 K with appropriate signs. The experimental $\gamma_{HS}\simeq 0$ line in the regions of low Fe concentration (<5 at. %), shown by the dotted line, has been drawn by mere extrapolation and hence is somewhat uncertain. The alloys in this concentration range, which are expected to show a change in sign, have T_c below 77 K and thus beyond the reach of our present experimental facility. From the nature of our $\gamma_{HS}\simeq 0$ line, it appears that in binary Ni-Cr alloys a sign change in γ_{HS} cannot be observed since ferromagnetism is destroyed before such a composition is reached. This is also consistent with the literature data,[43] where no such sign change has been reported so far. The main points which emerge from Fig. 2 are the following.

(i) The experimental $\gamma_{HS}\simeq 0$ line definitely lies much below the theoretical line and essentially in disagreement with Berger's model. The experimental $\lambda_s\simeq 0$ line is also

TABLE I. ρ, R_sM_s, and γ_{HS} at 77 and 300 K for some Ni-Fe-Cr alloys.

Sample no.	Composition of Ni-Fe-Cr alloys (at. %)	T_c (K)	ρ (10^{-7} Ω cm) 77 K	ρ (10^{-7} Ω cm) 300 K	R_sM_s (10^{-9} Ω m) 77 K	R_sM_s (10^{-9} Ω m) 300 K	γ_{HS} ($10^3\ \Omega^{-1}\,m^{-1}$) 77 K	γ_{HS} ($10^3\ \Omega^{-1}\,m^{-1}$) 300 K
42	78-6-16	185±5	8.4±0.4	8.8±0.4	−(0.71±0.03)	0	−(10±1.0)	0
50	72-8-20	93±3	10.7±0.3	11.1±0.3	+0.02	0	+0.02	0
9	85.5-11-3.5	620±3	5.2±0.15	5.8±0.15	−(6.6±0.2)	−(5.6±0.15)	−(24.4±2)	−(16.6±1.5)
24	81-11-8	470±4	8.8±0.25	9.3±0.3	−(8.0±0.3)	−(4.6±0.2)	−(10.3±0.8)	−(5.3±0.5)
40	73.5-11.5-15	260±5	10.3±0.4	10.7±0.4	−(2.9±0.1)	0	−(2.7±0.3)	0
29	75.1-12.8-12.1	365±3	9.3±0.2	9.8±0.2	−(3.6±0.1)	−(1.20±0.05)	−(4.2±0.3)	−(1.25±0.1)
48	70-12-18	179±2	10.6±0.3	11.0±0.3	+(1.10±0.05)	0	+(1.05±0.1)	0
34	72.5-13.7-13.8	315±7	9.9±0.2	10.2±0.2	−(0.72±0.02)	−0.01	−(0.73±0.05)	−0.01
26	80-16-4	693±3	6.3±0.3	6.9±0.3	−(1.50±0.05)	−(0.73±0.05)	−(3.8±0.3)	−(1.5±0.15)
28	75-17-8	543±3	7.7±0.2	8.3±0.2	−(1.90±0.08)	−(0.84±0.04)	−(3.2±0.3)	−(1.20±0.1)
33	68.1-17.4-14.5	320±6	10.4±0.5	10.8±0.5	+(2.7±0.1)	+(1.25±0.05)	+(2.5±0.2)	+(1.03±0.1)
35	76.8-21.2-2	778±4	4.3±0.1	5.3±0.1	+(0.90±0.04)	+(1.40±0.03)	+(4.9±0.4)	+(5.0±0.3)
27	75.5-20.3-4.2	717±3	6.1±0.2	7.0±0.2	+(1.54±0.04)	+(2.5±0.07)	+(4.1±0.4)	+(5.1±0.4)
51	67-21-12	470±2	10.4±0.3	11.0±0.3	+(5.9±0.1)	+(5.1±0.1)	+(5.4±0.4)	+(4.2±0.3)
32	69.6-22.8-7.6	635±5	9.0±0.15	9.6±0.15	+(4.9±0.1)	+(5.4±0.1)	+(6.0±0.3)	+(5.8±0.3)

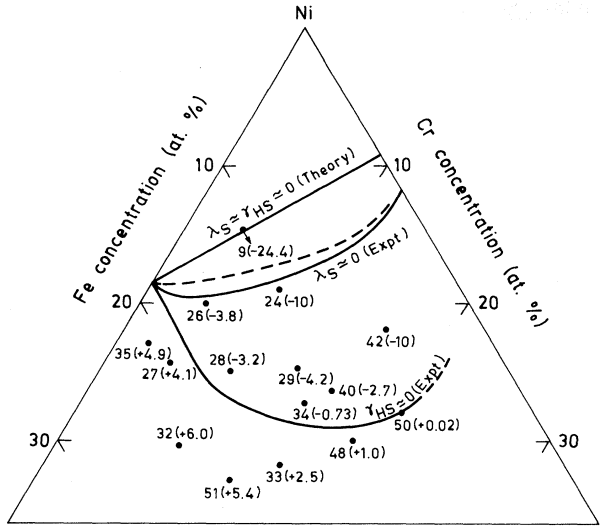

FIG. 2. Ternary phase diagram for Ni-Fe-Cr alloys. Our experimentally obtained $\gamma_{HS} \simeq 0$ line is shown along with the previously established experimental $\lambda_s \simeq 0$ line and the theoretically predicted $\lambda_s \simeq \gamma_{HS} \simeq 0$ line according to the SB model. The numbers in the parentheses are the values of γ_{HS} ($10^3\ \Omega^{-1}\,m^{-1}$) at 77 K. For the dotted line, see text.

at variance with the theoretical line, but to a lesser extent.

(ii) The deviation from the SB model is more pronounced in the Cr-rich region, implying that the presence of Cr is the cause of disagreement with a model, which has so far been found to be useful in several binary and ternary alloys.

(iii) Both the experimental $\gamma_{HS} \simeq 0$ and $\lambda_s \simeq 0$ lines show considerable curvature in contrary to the straight lines expected from the simple SB model.

(iv) λ_s and γ_{HS} are expected to change sign simultaneously. But in this case as one approaches the zero line from the Ni-rich side, λ_s changes sign long before γ_{HS}. Thus if a γ_{HS}-versus-λ_s plot is made, the curve should have a negative intercept on the γ_{HS} axis. This is in contrast with the other crystalline binary and ternary systems[17] as well as some amorphous systems[18] where the intercepts are positive.

In search of a plausible reason for the deviation of our experimental results from those expected from the SB model, it was natural to reflect on the saturation magnetization data at 0 K, since basically both the phenomena are related to band filling. The average number of Bohr magnetons per atom $\bar{\mu}$ should bear the following simple relationship with concentrations, if the band filling really did occur in the manner assumed in Eq. (2),

$$\bar{\mu} = 0.61 + 2C_{\mathrm{Fe}} - (10+Z)C_M \ . \tag{4}$$

But a close look at the data that already exist for $\bar{\mu}$ by Men'shikov *et al.*[19] speaks to the contrary. Their data as well as the value of $\bar{\mu}$ for some of the alloys measured by us (which will be presented elsewhere) confirm considerable nonlinearity in $\bar{\mu}$ versus concentration curves. Experimental values of $\bar{\mu}$ can be fitted to Eq. (4) only if the quantity $10 + Z$ [henceforth termed as $(Z)_{\mathrm{eff}}$] is treated as a variable parameter which is concentration dependent. Also the values of $10 + Z$ from the magnetization data are much less than the expected value of 6 for the Cr alloys.

In order to check whether this could be a reason for the deviation of our $(\gamma_{HS})_{\mathrm{expt}} \simeq 0$ line from the one predicted by the SB model, the following procedure was carried out. A few alloy compositions in the ternary diagram near the experimental $\lambda_s \simeq 0$ line were selected and the corresponding experimental values of $\bar{\mu}$ were obtained from the data of Men'shikov *et al.*[19] The $(Z)_{\mathrm{eff}}$ values for these alloys were estimated by using Eq. (4). Now in turn, the new values of $(Z)_{\mathrm{eff}}$ were substituted in Eq. (2) in place of $10 + Z$ and then the compositions for which a sign change in λ_s and γ_{HS} are expected [according to Eq. (2)] were estimated. Equation (2), thus modified, predicts the dotted curve shown in Fig. 2. Table II shows the data used for such analysis.

The important outcome of this analysis is that when the nonlinearity in $\bar{\mu}$-versus-concentration curves is empirically incorporated into Eq. (2), the theoretical line comes much closer to the experimental $\lambda_s \simeq 0$ line and essentially reproduces its curvature. But the theoretical line still lies far away from the experimental $\gamma_{HS} \simeq 0$ line. From this one could safely conclude that the curvatures of experimental $\lambda_s \simeq 0$ line and possibly also those of the $\gamma_{HS} \simeq 0$ line are the off shoot of the nonlinearity in the $\bar{\mu}$-versus-C curves. In this context, it is also important to point out that for those systems, where SB model could account for

TABLE II. Results of reanalysis of $(\lambda_s)_{\mathrm{expt}} \simeq 0$ data for Ni-Fe-Cr alloys in terms of the SB model, modified to incorporate the compositional dependence of $(Z)_{\mathrm{eff}}$.

Ni-Fe-Cr alloy compositions (at. %) at which $(\lambda_s)_{\mathrm{expt}} \simeq 0$	$(\bar{\mu})_{\mathrm{expt.}}$[a] (units of μ_B)	$(Z)_{\mathrm{eff}}$	Ni-Fe-Cr compositions (at. %) for which λ_s and γ_{HS} should change sign according to modified Eq. (2)
85.7-2-12.3	0.13	4.2	86.3-2-11.7
83.5-5-11.5	0.26	3.9	84.7-5-10.3
81.8-8-10.2	0.39	3.7	83.6-8-8.4
81-10-9	0.47	3.8	83.4-10-6.6
80.4-12-7.6	0.57	3.7	82.9-12-5.1
80-14-6	0.70	3.2	81.9-14-4.1
80-16-4	0.81	3.0	81.7-16-2.3
80.5-18-1.5	0.92	3.3	81.7-18-0.3

[a]Estimated from Ref. 19.

the sign change of γ_{HS} and λ_s, saturation-magnetization data show a linear concentration dependence.

B. Sign reversal of γ_{HS} in Ni-Fe-V alloys

From the measurements of the extraordinary Hall coefficient at 77 and 300 K for 12 alloys in this series, seven have been found to have a positive Hall coefficient. Table III presents the values of ρ, R_sM_s, and γ_{HS} for these alloys. Using these data, the position of the experimental $\gamma_{HS}\simeq 0$ line has been established and shown in Fig. 3, along with the already established $\lambda_s\simeq 0$ line[44] and those predicted by SB model. It is clear from Fig. 3 that for the Ni-Fe-V system the behavior of all the lines (λ_s, $\gamma_{HS}\simeq 0$, etc.) are essentially similar to those of Ni-Fe-Cr and all the observations [(i)—(iv)] made in this connection in Sec. IV A are equally valid for Ni-Fe-V alloys; differences, if any, are merely quantitative. Contrary to the Ni-Fe-Cr system, the experimental $\lambda_s\simeq 0$ and $\gamma_{HS}\simeq 0$ lines are almost coincident in the Fe-rich region of this ternary diagram. However, deviations from the SB model and also from the experimental $\lambda_s\simeq 0$ line increases as V concentration is increased. But the quantitative deviation from the SB model for this sytem is much less than that for Ni-Fe-Cr alloys. This can be expected if one remembers that vanadium is one step further from Ni than Cr in the Periodic Table, and hence band splitting should be more complete in the former than in the latter.

Because of the absence of any systematic data for $\bar{\mu}$ in this ternary system, it could not be checked whether the curvatures of the experimental λ_s and $\gamma_{HS}\simeq 0$ lines are an off shoot of the nonlinearity in the $\bar{\mu}$-versus-concentration curves, as has been observed in Ni-Fe-Cr alloys. However, it should be mentioned that the measurements for $\bar{\mu}$ in some of the ternary V alloys have shown[39] a similar kind of concentration dependence as in the corresponding Cr series (although on a smaller scale). Therefore, there is a distinct possibility that the curvatures of the two lines mentioned (λ_s and γ_{HS}) are due to a reason similar to that for Ni-Fe-Cr alloys.

Thus it appears that the SB model can explain fairly well the position of the $\lambda_s\simeq 0$ line for both the ternary (Cr or V) systems. The apparent discrepancy between the theoretically predicted and experimentally observed lines seems to be due to the nonlinear concentration dependence of saturation magnetization (because this apparent discrepancy, at least in the case of Ni-Fe-Cr alloys, is removed to a large extent when the above-mentioned nonlinearity is empirically incorporated into the SB model). However, the SB model fails to justify the position of the experimental $\gamma_{HS}\simeq 0$ line simply because γ_{HS} and λ_s do not change sign simultaneously, contrary to expectation. The ternary systems studied so far (Ni-Fe-Cu, Cr or V), have all shown that experimental $\lambda_s\simeq 0$ line is always in better agreement with the SB model than the $\gamma_{HS}\simeq 0$ line. This point will be further discussed in the next section.

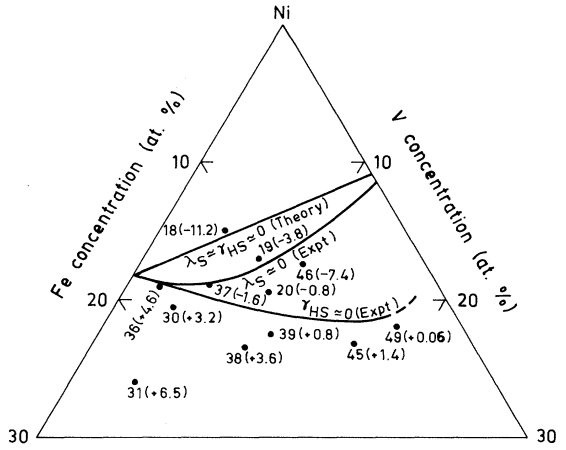

FIG. 3. Same as in Fig. 2, but for Ni-Fe-V alloys.

C. Relationship between λ_s and γ_{HS}

Owing to their common origin in s.o. coupling, λ_s and γ_{HS} have been found to bear a common relationship in the case of various binary systems such as Ni-Fe, Cu-Ni, and also in the ternary Ni-Fe-Cu system[17] and even in Fe-Ni-B metallic glasses.[18] In all the crystalline materials mentioned earlier, an approximate constant ratio of $\approx 2\times 10^9\ \Omega^{-1}\,\mathrm{m}^{-1}$ have been found for γ_{HS}/λ_s; in glassy

TABLE III. ρ, R_sM_s, and γ_{HS} at 77 and 300 K for some Ni-Fe-V alloys.

Sample no.	Composition of Ni-Fe-V alloys (at. %)	T_c (K)	ρ (10^{-7} Ω m) 77 K	ρ (10^{-7} Ω m) 300 K	R_sM_s (10^{-9} Ω m) 77 K	R_sM_s (10^{-9} Ω m) 300 K	γ_{HS} (10^3 $\Omega^{-1}\,\mathrm{m}^{-1}$) 77 K	γ_{HS} (10^3 $\Omega^{-1}\,\mathrm{m}^{-1}$) 300 K
49	78-4-18	90±4	13.2±0.3	13.4±0.3	+(0.1±0.01)	0	+ 0.06	0
46	82.5-7.5-10	362±3	8.2±0.15	8.8±0.15	−(5.0±0.1)	−(2.3±0.05)	−(7.4±0.5)	−(3.0±0.2)
45	76.8-7.2-16	167±5	12.2±0.2	12.3±0.2	+(2.1±0.05)	0	+(1.4±0.07)	0
19	83-10-7	486±2	8.1±0.2	9.0±0.2	−(2.5±0.1)	−(1.20±0.04)	−(3.8±0.3)	−(1.5±0.1)
20	80.5-10.5-9	417±3	10.5±0.3	10.7±0.3	−(0.87±0.03)	− 0.03	−(0.80±0.08)	−0.03
18	85-11-4	609±5	5.4±0.2	6.3±0.2	−(3.3±0.1)	−(3.1±0.1)	−(11.2±1)	−(7.8±0.8)
39	77.4-11.9-10.7	393±1	9.8±0.2	10.6±0.2	+(0.75±0.03)	+(1.09±0.03)	+(0.78±0.06)	+(0.97±0.07)
37	80.9-14-5.1	640±4	6.7±0.2	7.4±0.2	−(0.72±0.03)	−(0.06±0.01)	−(1.6±0.2)	−(0.11±0.02)
38	76.5-14-9.5	462±1	9.3±0.3	10.1±0.3	+(3.1±0.1)	+(3.3±0.1)	+(3.6±0.3)	+(3.2±0.3)
36	81-17-2	746±3	3.4±0.1	4.4±0.1	+(0.53±0.02)	+(0.82±0.03)	+(4.6±0.4)	+(4.2±0.4)
30	79.4-17-3.6	691±2	4.9±0.15	6.0±0.2	+(0.77±0.02)	+(1.30±0.03)	+(3.2±0.3)	+(3.6±0.3)
31	74-22-4	741±3	5.8±0.2	7.1±0.3	+(2.2±0.05)	+(3.2±0.1)	+(6.5±0.6)	+(6.3±0.6)

systems the ratio is slightly lower.[18]

On the basis of the above results, it was natural to check whether any such relationship at all exists between γ_{HS} and λ_s for these two ternary systems, especially when the ternary diagrams in Figs. 2 and 3 show a large separation between the experimental $\lambda_s \simeq 0$ and $\gamma_{HS} \simeq 0$ lines. The problems encountered in such an analysis are the following. The earlier data on linear magnetostriction are mainly confined to near the region of $\lambda_s \simeq 0$ line, and the corresponding data for γ_{HS} are absent. On the other hand, λ_s values corresponding to most of the alloys we have studied do not exist in the literature. Also the earlier data of λ_s were mainly confined to single crystals. Fortunately, λ_s for some of our alloys in both the ternary systems have been recently measured by Majumdar and Greenough.[45] By using their data along with some already existing in the literature,[46] a γ_{HS}-versus-λ_s plot was tried, as shown in Fig. 4. The data used for such an analysis are presented in Table IV. γ_{HS} data for two alloys were taken from Sinha and Majumdar[41] and the rest were measured by us. Except for one alloy ($Ni_{85.5}Fe_{11}Cr_{3.5}$),[46] all the other values of λ_s were taken from Majumdar and Greenough[45] and were measured on the same polycrystalline samples on which γ_{HS} were measured by us. This makes the comparison of γ_{HS} and λ_s more meaningful. Since all the alloys have fairly high resistivity (dominated by impurity scattering), the relationship $R_s M_s \propto \rho^2$ is expected to hold well even at low temperatures. Becuase of this and the lower T_c of some of the alloys, 77-K data were preferred for any such comparison.

In Fig. 4 the straight line drawn using the open circles

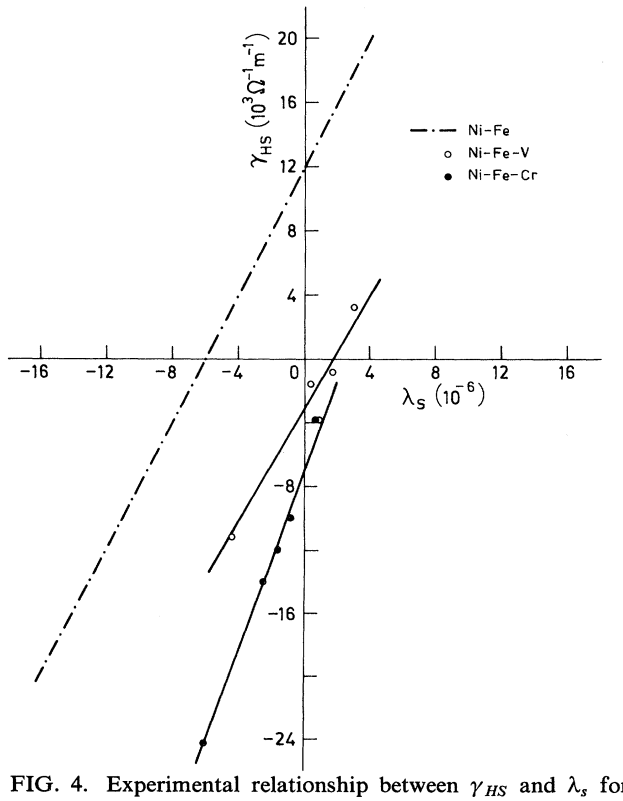

FIG. 4. Experimental relationship between γ_{HS} and λ_s for Ni-Fe, Ni-Fe-Cr, and Ni-Fe-V alloys.

TABLE IV. Data for λ_s and γ_{HS} for some Ni-Fe-Cr and Ni-Fe-V alloys.

Compositions (at. %)	γ_{HS} ($10^3\ \Omega^{-1} m^{-1}$) at 77 K	λ_s (10^{-6}) at 77 K
Ni_{84}-Fe_6-Cr_{10}	−12.0[a]	−1.6[b]
$Ni_{85.5}$-Fe_{11}-$Cr_{3.5}$	−24.4	−6.0[c]
Ni_{83}-Fe_{11}-Cr_6	−14.0[a]	−2.5[b]
Ni_{81}-Fe_{11}-Cr_8	−10.3	−0.75[b]
Ni_{80}-Fe_{16}-Cr_4	−3.8	+0.6[b]
Ni_{85}-Fe_{11}-V_4	−11.2	−4.4[b]
Ni_{83}-Fe_{10}-V_7	−3.8	+0.75[b]
$Ni_{80.5}$-$Fe_{10.5}$-V_9	−0.8	+1.7[b]
$Ni_{80.9}$-Fe_{14}-$V_{5.1}$	−1.6	+0.4[b]
$Ni_{79.4}$-Fe_{17}-$V_{3.6}$	+3.2	+3.0[b]

[a]Reference 41.
[b]Reference 45.
[c]Reference 46.

is for the Ni-Fe-V alloys and the one with solid circles is for the Ni-Fe-Cr alloys. The dotted-dashed line is for Ni-Fe alloys[47,48,26] and has been included for comparison. Although the number of points is not sufficient, it appears that a rough straight line can be drawn for both the systems. However, for Ni-Fe-V alloys, one point (corresponding to $Ni_{83}Fe_{10}V_7$) lies away from the straight line. This deviation is unlikely to be due to experimental error. No comment can be made on it at present, except for the fact that in Ni-Fe-Cu (Ref. 17) system also all the points do not fall on the same straight line but form two separate branches. The main points that can be made from Fig. 4 are the following.

(i) γ_{HS} seems to be proportional to λ_s for both the ternary systems; the ratio γ_{HS}/λ_s is about 2.8×10^9 $\Omega^{-1} m^{-1}$ for Ni-Fe-Cr and 1.8×10^9 $\Omega^{-1} m^{-1}$ for Ni-Fe-V. This is comparable to the value of 2×10^9 $\Omega^{-1} m^{-1}$ for Ni-Fe (Ref. 17) and other systems. However, the value of the slope for Ni-Fe-Cr alloys should be taken cautiously since the diagram is incomplete in the absence of any data for λ_s in the region of positive γ_{HS}.

(ii) Usually the interecept that the line makes on the γ_{HS} axis is positive. But here for both the ternary systems the intercepts are negative, being about -3×10^3 $\Omega^{-1} m^{-1}$ for Ni-Fe-V and about -7×10^3 $\Omega^{-1} m^{-1}$ for Ni-Fe-Cr alloys, in contrast to $+12\times10^3$ $\Omega^{-1} m^{-1}$ for Ni-Fe alloys. According to Berger's theory, the lines should all pass through the origin, which has in fact never been realized in practice.

Thus the fact that the slope and intercept are both nearer to the theoretical value for Ni-Fe-V than Ni-Fe-Cr alloys once again proves that the former is a better candidate to follow the SB model. Another interesting aspect of the problem is that the large deviation of $\gamma_{HS} \simeq 0$ line from $\lambda_s \simeq 0$ line in the ternary diagrams (Figs. 2 and 3) would have led one to expect that even if a proportionality relationship existed between λ_s and γ_{HS}, the intercepts of the straight lines on the γ_{HS} axis would have been sufficiently large compared to other alloys. But on the contrary, we find them even much smaller than in other al-

loys. Physically this means that the change of sign of γ_{HS} with composition is very gradual. A small negative intercept on γ_{HS} axis means that as one approaches the $\lambda_s \simeq 0$ line from the Ni-rich side; first γ_s changes sign, and the γ_{HS} value for the corresponding composition still remains negative but small in magnitude. But to reduce this small negative γ_{HS} to zero, the change of composition necessary is rather large as can be seen from the large separation of the experimental λ_s and $\gamma_{HS} \simeq 0$ lines in the ternary diagrams.

The above statement will be much more clear if one looks at Fig. 5 where γ_{HS} and λ_s have been plotted as a function of increasing Fe + Cr (or V) concentration for a series of ternary alloys whose Fe contents are roughly the same. In this figure, the corresponding quantities for Ni-Fe alloys as a function of Fe concentration are included for comparison. The values of γ_{HS} for Ni-Fe alloys were taken from the data of Jellinghaus and De Andres[26] and λ_s from Bozorth and Walker[47] and Hall.[48] Since magnetostriction data for Ni-Fe alloys in the literature are all for single crystals, they were converted by the relation $\lambda_s = \frac{2}{5}\lambda_{100} + \frac{3}{5}\lambda_{111}$, to compare with our polycrystalline samples. It can be seen from the figure that the rate of change of λ_s with concentration of impurity is hardly affected by the impurity type (Fe,Cr,V). But the rate of change of γ_{HS} with the addition of Cr (or V) in ternary

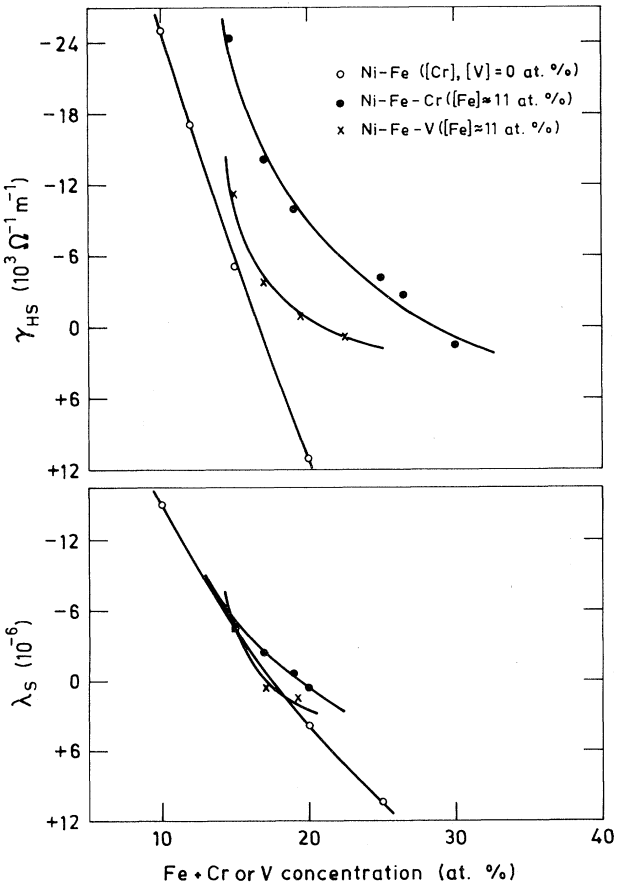

FIG. 5. Variation of γ_{HS} and λ_s with Cr or V concentrations for a series of ternary alloys with approximately the same Fe content.

Ni-Fe-Cr or Ni-Fe-V alloys is much slower near the $\gamma_{HS} \simeq 0$ region than in corresponding binary Ni-Fe alloys. This is as if the transition from negative to positive γ_{HS} is smeared out due to the addition of Cr (or V) in Ni-Fe alloys. In this context we recall that Berger[15] pointed out that if the *d* band has lower-than-spherical symmetry, the transition from positive to negative γ_{HS} could get smeared out. Since transport properties such as γ_{HS} depend only on the electrons near the Fermi level, this effect should be more pronounced in the case of γ_{HS} than λ_s. There seems to be a possibility that the presence of Cr or V alters the *d* band in such a manner as to lower its symmetry which in turn smears out the sign change of γ_{HS}. If this is true, then one should also expect that the maximum in ferromagnetic anisotropy of resistance, found for compositions near the vicinity of the $\lambda_s \simeq \gamma_{HS} \simeq 0$ line, should also become smeared out, being a transport property. Unfortunately, the absence of any such data for the two ternary systems rules out the possibility for a check. It seems that the effect should be more prominent in the case of Ni-Fe-Cr alloys than that of Ni-Fe-V, since the smearing out appears to be less in the latter case.

To summarize, it appears that from all the experimental data available so far, γ_{HS} and λ_s never change sign exactly at the same composition (since γ_{HS} and λ_s plots have never been found to pass exactly through the origin), but approximately at the same compositions. Our systems are also no exception to this rule. However, there is a possibility that the small discrepancy found in other systems in connection with the compositions at which λ_s and γ_{HS} change sign gets magnified many times in these ternary alloys because of the slow variation of γ_{HS} as a function of Cr or V concentration. So the small separation between the $\lambda_s \simeq 0$ and $\gamma_{HS} \simeq 0$ lines in the ternary diagram found in the other systems gets highly magnified in the case of Ni-Fe-Cr and Ni-Fe-V alloys, which gives the impression that no relationship exists between λ_s and γ_{HS} in these systems.

D. Other possible explanations

The problem [the disagreement between the SB model and the $(\gamma_{HS})_{\text{expt}} \simeq 0$ line] can also be looked at from a different point of view. There is a possibility that the band picture shown in Fig. 1 is an oversimplification of the reality. Unfortunately, so far no rigorous calculations have been carried out to give some idea of the band structure of these ternary systems; mainly, computational problems have thwarted such attempts so far. But CPA calculations of band structures for the corresponding binary systems (e.g., Ni-Fe, Ni-Cr, Fe-Cr, etc.) were carried out by Hasegawa and Kanamori.[29–31] Their results show that in the case of Ni-Cr alloys,[30] a distinctly separate band for Cr emerges above the Ni spin-down band, which grows in size with increasing Cr concentration, ultimately merging with the Ni spin-down band. In the case of Fe-Cr alloys,[31] since the energy difference of an electron sitting at the Cr site from the one at the Fe site ($E_{\text{Cr}} - E_{\text{Fe}}$) is very small for the down-spin electrons, upon alloying the down-spin band of Fe is hardy modified. On the other hand, the up-spin band of Fe, consider-

ably deformed, hardly shows any sign of splitting. Under these circumstances, Berger's model for ternary Ni-Fe-Cr alloys seems to be oversimplified. It seems logical to think that the Cr spin-down band does not get separated from the Fe-spin-down band, in contrary to what was suggested by Berger. Also, the Cr spin-up band should not lie far from the Fe spin-down band, and there should be a considerable overlap between the two. With increasing Cr concentration, the Cr subbands should grow in size and move towards the Ni spin-down band. On the other hand, the Fermi level of the system is also pushed up due to electron transfer. Thus at higher Cr concentrations, when the Fermi level is near the edge of Ni spin-down band, the assumption that all spin-up bands in the sytem are full breaks down. In Eq. (2) one must take into account the contribution of states in the spin-up bands also. The major disagreement between our experimentally observed $\gamma_{HS} \simeq 0$ line and the one predicted by the SB model is probably due to this reason. Also, the slow variation of γ_{HS} with Cr concentration near the $\gamma_{HS} \simeq 0$ line seems to be in agreement with this view. If this view is correct, the alloys with increasing Cr content should behave more as itinerant weak ferromagnets simply because part of both up-spin and down-spin states above E_F remains empty. This is consistent with our magnetization measurements,[39] as are planned to be presented elsewhere.

In the case of Ni-Fe-V system, due to the absence of any such band-structure calculations even for binary Ni-V, Fe-V alloys, nothing much could be said. Nevertheless, a similar type of findings for γ_{HS} for this system suggests that similar mechanism could also be responsible for this system. Because of the larger charge difference between Fe and V, band splitting for Fe-V alloys should be more complete, and hence in ternary Ni-Fe-V alloys the overlap between the Fe spin-down and the V spin-up bands should be lesser. The smaller discrepancy between the theoretical and our experimental $\gamma_{HS} \simeq 0$ line probably points to the same direction.

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