mated by the interpolation scheme of Hodges, Ehrenreich, and Lang (Ref. 1)] at 505 points in the reduced zone. Agreement between the two estimates was excellent. We employ the results of Goodings and Harris because their more exhaustive sampling of the zone should yield a more realistic estimate of the structure and a more accurate placement of the Fermi level. We note numerical differences between our estimates of the radial matrix elements and theirs. These differences are not large, and probably arise from their different choice of core wave functions.

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Spin-Disorder Scattering in Iron- and Nickel-Base Alloys

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The electrical resistivities of iron-base alloys with Cr, W, Mn, Ru, Co, Ni, Si, or Ge solutes, and of nickel-base alloys with Cr, Fe, Cu, or Pd solutes were measured from 4.2 °K to above the Curie temperatures θ. The solute resistivities $\rho_r(T)[=\rho_{\text{alloy}}(T)-\rho_{\text{host}}(T)]$ were found to be strongly temperature dependent. Above ~ 300 °K, the temperature dependence of $\rho_r(T)$ reflects changes in the electronic structure or in the scattering processes associated with the solutes and is related to changes in the magnetic order. The solute resistivities above 300 °K were found to be in reasonable agreement with a spin-disorder model which explicitly involves the moments localized at the solute sites. Agreement with the well-known Mott band model for ferromagnetic effects in transport properties was not as satisfactory. An essential feature of the analyses of the data involved comparing resistivities of alloy and host corresponding to the same degree of magnetic order. This was accomplished by assuming the ferromagnetic effects in the resistivity scaled as T/θ. The experimental results support the validity of this assumption.

I. INTRODUCTION

It is well known that for metals the electronic transport properties are closely related to the magnetic state. In particular the resistivities of the ferromagnetic transition elements iron and nickel exhibit unusually strong temperature dependences in the temperature ranges in which the magnetic order changes significantly (cf. Figs. 1 and 2). There have been two general approaches to understanding these phenomena.[1-4] In the first approach, a band model, the unusual temperature dependence is associated with increases in the electron d states available for scattering as the spontaneous magnetization decreases. In the second approach, disordered magnetic moments are considered to act as additional scattering centers for conduction electrons, and the temperature dependence of the resistivity reflects changes in the degree of magnetic order.

A means of determining whether spin-disorder scattering or the band mechanism is more appropriate for describing the resistivity of a ferromagnetic metal on the basis of qualitative experimental features is provided by studies of the resistivities of dilute alloys.[1-4] The following argument by Coles[1] illustrates the difference in the effects of solutes on the resistivity for the spin-disorder and band models. In a "crude but useful approximation," the resistivity is written as

$$\rho = (P_a + P_T + P_{sd}) F \ , \tag{1}$$

where P_a, P_T, and P_{sd} represent the perturbations produced by atomic, thermal, and spin disorder, and F is a factor depending on the electron configuration and involving the density of states to which conduction electrons can make transitions. The band model for the temperature dependence of the magnetic effects on the resistivity corresponds to $P_{sd} = 0$ and to F being a monotonic-increasing function of temperature. Above the Curie temperature, F should become temperature independent. All the anomalous temperature dependence of the resistivity is associated with F, which in turn results from changes in the density of d states available as "sinks" $[\eta_d^+(\epsilon_F), \ \eta_d^-(\epsilon_F)]$. The spin-disorder model for the resistivity corresponds to F being a constant and the perturbation P_{sd} being an increasing function of temperature, which for the localized case becomes constant above the Curie temperature.

The addition of solute atoms in dilute quantities will increase P_a and the solute resistivity, defined by

$$\rho_r(T) = \rho_{\text{alloy}}(T) - \rho_{\text{host}}(T) \ , \tag{2}$$

will be $P_a F$. In the band model, ρ_r will be strongly temperature dependent through the factor F; ρ_r will increase with temperature in a manner similar to the resistivity of the pure metal – apart, of course, from the temperature dependence due to the thermal perturbation P_T. By contrast, the prediction of the spin-disorder model is that ρ_r will be temperature independent. Coles performed the indicated experiments using dilute alloys *Ni*Pd and *Fe*Ru. The reported results are that the value of the solute resistivity of the *Ni*Pd alloy was larger above θ_0 than at 4.2 °K: whereas, for the *Fe*Ru alloy, $\rho_r(T)$ had the same value above θ_0 and at 4.2 °K. These observations have been used to support the thesis that the resistivity of nickel reflects changes in the density of d states associated with changes in the spontaneous magnetization as postulated by Mott, and that the ferromagnetic effects in the resistivity of iron arise from spin-disorder scattering.[1-4]

In this paper we report results of recent measurements of several dilute iron- and nickel-base alloys from 4.2 °K to above the Curie temperatures. (A preliminary report of the resistivities of some iron-base alloys has been given elsewhere.[5]) Particular emphasis is given to the solute resistivity $\rho_r(T)$ in the temperature range in which the spontaneous moment decreases most significantly, i.e., $T > \frac{1}{2}\theta_0$. The solutes in the iron-base alloys include Cr, W, Mn, Ni, Co, Si, Ge, and Ru, while those in the nickel-base alloys include Cr, Fe, Cu, and Pd. Several compositions for each system were measured. For all the alloys, with the exception of the *Ni*Pd alloys, the solute resistivity $\rho_r(T)$ was strongly temperature dependent. In the iron-base alloys, $\rho_r(T)$ generally decreased with temperature from ~400 °K to above the Curie temperature, and the values of $\rho_r(T)$ above θ_0 and at 4.2 °K were significantly different. By contrast, $\rho_r(T)$ for the *Ni*Pd alloys showed relatively little change from 4.2 to 900 °K, well above the Curie temperature. These results not only differ from those previously reported, but also appear contradictory to both the spin-disorder and the band models.

However, if the localized spin-disorder model is generalized to allow for different moments on solute and host sites, the solute resistivity is predicted to be temperature dependent. [In Eq. (1) this corresponds to changes in P_{sd} associated with the addition of solutes.] This modification of the spin-disorder model can be used to explain the observed sign of the difference between the solute resistivities in the ferromagnetic and paramagnetic states, as well as the variation of the magnitude of this change with solute moment for both the iron- and nickel-base alloys.

Thus, it is suggested that above $\frac{1}{2}\theta_0$, the resistivities of iron and nickel, at least with regard to the effects of dilute amounts of solutes, exhibit behavior expected for a ferromagnetic metal with localized magnetic spins and that these resistivities are not greatly influenced by any changes in the densities of d states at the Fermi energy which may accompany the decrease of the spontaneous moment.

II. EXPERIMENTAL PROCEDURE

The majority of the iron-base alloys were originally used in a previously reported study of low-temperature resistivities.[6] The iron alloys containing Cr (1.5 at.%), Co (1.5 at.%), and Ru (0.5, 1, and 3 at.%) and all the nickel-base alloys were prepared by arc-melting high-purity chips (Ni and Fe of nominal purity[7] 99.997%) in an argon atmosphere. The arc-melted samples were cold rolled 60% and annealed at 1000 °C for one week. For these anneals the samples were sealed in quartz capsules; the iron-base alloys were in a helium atmosphere; the nickel-base alloys were in a nitrogen atmosphere. The samples were machined to right cylinders with diameters ~0.25 cm and lengths ~3.8 cm, and reannealed for 2 h; the annealing temperatures for the iron-base alloys were below the α-γ transformation and for the nickel-base alloys were ~800 °C.

Solute concentrations, except for the *Fe*Ru series, were determined by chemical analysis. For the *Fe*Ru series the reported concentrations are based on the weights of the two constituents prior to melting. In general, for all the alloys studied, the solute concentration and $\rho_{4.2}$ were consistent with values of the appropriate resistivity increments reported in the literature.[6,8,9]

A nickel standard was obtained by centerless grinding a section of high-purity rod to a diameter of ~1.8 mm. Two iron standards were prepared by swaging zone-refined iron (supplied by Turkdogan of this laboratory) to diameters of ~1.8 mm. The values of the resistivities at 4.2 °K were 0.024 $\mu\Omega$ cm for the nickel standard and ~0.04 $\mu\Omega$ cm for the iron standards.

The average cross-sectional area of each sample was deduced from the weight, length, and density; the density was derived using the chemical analyses and the tabulated values of the change in lattice parameter for that solute concentration. Current and potential leads (nickel or iron wire depending on the solvent) were spot welded to each sample, and the gauge length was measured with a traveling microscope. The determination of the geometrical factors is expected to be the largest source for error in the determination of the solute resistivities $\rho_r(T)$ $(=\rho_{\text{alloy}}-\rho_{\text{std}})$. Uncertainties in the geometrical factors are believed to be less than 0.2%, and errors in $\rho_r(T)$ are certainly less than 0.5% of the resistivity of the standard.

Subambient resistivity measurements were made using procedures described in connection with a study of the resistivities of *Fe*Si alloys.[10] For the high-temperature measurements, a sample and appropriate standard were mounted within a noninductively wound furnace in thermal proximity of each other and of a thermocouple well. An atmosphere of He 2% H_2 flowed past the samples during the high-temperature measurements. Temperature was monitored using a calibrated Pt-Pt 10% Rh couple. A dc current of 0.5 A (maintained constant to 1 part in 10^5) was used, and standard reversing techniques were employed to eliminate thermal emf's.

The simultaneous measurement of the resistivities of a sample and standard obviated a precise determination of the temperature. However, the Curie temperatures of the iron and nickel standards and the alloys (taken to be the temperature at which the slope of the resistivity is maximum) and the temperatures of the α-γ transitions were observed to occur within a few degrees of the accepted values.

The measurements were made "quasistatically" with the temperature increasing at a constant rate of ~1 °C/min. The voltage was amplified with a Keithley model 140 dc amplifier (rise time 0.05 sec) and measured with a digital voltmeter. The input impedance of this system was large enough that the effect of lead or contact resistance was negligible. Five significant figures were obtained, and a complete set of measurements (sample and standard potentials for both current directions) could be made within a temperature change of $\lesssim 0.2\,^\circ$C.

For the various samples and standards, values of the resistivity at 4.2 °K measured after a high-temperature run showed little change from the values measured before the run. Several high-temperature runs, including temperatures above the α-γ transition for the iron alloys, were made for some samples, and data from the various runs showed no significant differences, indicating that changes in substructure had no measurable effect.

III. ANALYSIS OF DATA

A. Change in Curie Temperature

Of primary interest is this study in the effect of various solutes on the scattering of conduction electrons. Since in the high-temperature ranges the solute resistivity of dilute alloys is only a few percent of the total resistivity, the solute resistivity $\rho_r(T)$ can most accurately be determined as the difference between the resistivity of the alloy and the resistivity of the pure solvent. However, as described in the Introduction, the resistivity of a ferromagnetic metal is closely related to its magnetic state as well as to its temperature. Significant values of $\rho_r(T)$ will be obtained only if the standard and the alloy are in the same magnetic state. The magnetic state at temperature T is often described in terms of the parameter (T/θ), and it is reasonable to expect the magnetic effects in the resistivity to depend on T/θ also.[11] Because the addition of most solutes change the Curie temperatures θ of iron and nickel, the alloy and the pure solvent at the same temperature T are not in the same magnetic state, i.e., $(T/\theta_0)\neq(T/\theta_a)$, where θ_0 and θ_a are the Curie temperatures of the pure solvent and alloy, respectively.

Specific scaling rules can be formulated only when a particular model has been selected to describe the ferromagnetic contribution to resistivity. For the *spin-disorder model*, the ideal resistivity of the pure solvent is $\rho_0(T;\theta_0)=\rho_{sd}(T/\theta_0)+\alpha_{ph}T$, where α_{ph} describes the contribution to the resistivity at high temperatures from electron-phonon interactions. The ideal resistivity corresponding to the ordering temperature θ_a in terms of the measured resistivity of a high-purity standard $\rho_{\text{std}}(T)$ is

$$\rho_0(T;\theta_a)=\rho_{\text{std}}(T\tau)-\alpha_{ph}T(\tau-1)\,, \qquad (3)$$

where $\tau = (\theta_0/\theta_a)$. For the *Mott band model* the ideal resistivity is $\rho_0(T;\theta_0) = (\alpha_{\text{ph}}T)\,F(T/\theta_0)$, where the temperature dependence of F primarily reflects changes in the density of d-state "sinks" at the Fermi surface. The scaled resistivity is

$$\rho_0(T;\theta_a) = \rho_{\text{std}}(T\tau) + (1/\tau - 1)\,\rho_{\text{std}}(T\tau)\,. \tag{4}$$

The form in which Eqs. (3) and (4) have been written indicates the manner in which the scaling was accomplished. To find values of τ, published reports of changes in Curie temperature for the appropriate alloy ststems were interpolated at the solute concentrations of the samples. The values of the resistivity of the standard which were measured with the particular sample were used as a table for $\rho_{\text{std}}(T\tau)$ with $\tau = 1$. This table was interpolated to find $\rho_{\text{std}}(T\tau)$ with the τ appropriate to the sample.[12] Corrections for the phonon scattering effects corresponding to the second term in Eqs. (3) and (4) were then made. It is necessary to introduce explicit values for α_{ph} only for the spin-disorder model.

B. Temperature Dependence of Solute Resistivity

As described in the Introduction, the simple prediction for the spin-disorder model was that the solute resistivity will be temperature independent; whereas, in the Mott band model the solute resistivity will be an increasing function of temperature with a change of slope at the Curie temperature. These predictions resulted from the implicit assumption that the solutes had the same spin and d-state characteristics as the host. In the following subsections the effect of solute spins and d states on the temperature dependence of the solute resistivity are considered within the framework of the spin-disorder and Mott band models, respectively.

1. *Spin-Disorder Model*

The interaction of conduction electrons spin S_c with localized moments spin S is considered to take the form $H_I \cong G\vec{S}\cdot\vec{s}_c$. The scattering cross section per isolated spin (and, consequently, the resistivity in the magnetically disordered state, ρ^0_{sd}) is proportional to $G^2S(S+1)$.[13] For a random substitutional alloy with solvent spin S_0 and solute spin S_i, the lattice perturbation at low temperatures in the magnetically ordered state will be proportional to $|S_i - S_0|$, and the contribution to the resistivity will be

$$\rho^{\text{ord}}_{\text{sd}} = \{\rho^0_{\text{sd}}[c/S_0(S_0+1)]\}\,(|S_i - S_0|)(|S_i - S_0| + 1)\,, \tag{5a}$$

where c is the atomic fraction. In the disordered state a magnetic atom of scattering cross section $\sim S_0(S_0+1)$ has been replaced by one with scattering cross section $\sim S_i(S_i+1)$. Thus, the contribution to the resistivity in the magnetically disordered state will be

$$\rho^{\text{dis}}_{\text{sd}} = \{\rho^0_{\text{sd}}[c/S_0(S_0+1)]\}\,[S_i(S_i+1) - S_0(S_0+1)]\,. \tag{5b}$$

The difference in the value of the solute resistivity in the ordered and disordered states will be

$$\delta\rho_r^{\text{ord-dis}} = \rho_r^{\text{dis}} - \rho_r^{\text{ord}} = -2c\rho^0_{\text{sd}}[1 - (S_i/S_0)]\,, \qquad S_i < S_0 \tag{5c}$$

$$\delta\rho_r^{\text{ord-dis}} = \rho_r^{\text{dis}} - \rho_r^{\text{ord}} = -2c\rho^0_{\text{sd}}\left(\frac{S_0}{S_0+1}\right)\left(1 - \frac{S_i}{S_0}\right), \qquad S_i > S_0 \tag{5d}$$

where c is the atomic fraction of solutes and $\rho^0_{\text{sd}} \cong G^2S_0(S_0+1)$ is the spin-disorder contribution to the resistivity of the pure solvent in the disordered state. In obtaining Eq. (5), it has been assumed that the exchange constant G has the same value for solute and solvent atoms.[14] In addition to these spin-disorder contributions, there will also be contributions from ordinary potential scattering. The density of d states at the Fermi surface will be important in determining the magnitude of both these contributions to the resistivity. However, in the spin-disorder model the density of d-state sinks is assumed to not change significantly with temperature.

2. *Mott Band Model*

In this model the resistivity is considered to be the product of perturbation terms (P_A due to solutes plus P_T due to phonons) and an electronic structure factor F [see Eq. (1)]. The function F contains all the temperature dependence associated with changes in the magnetic state and, in particular, reflects changes in the density of d states at the Fermi energy. Consequently, the solute resistivity in the band model will be

$$\rho_r = cP_AF\left[1 + \left(\frac{d\ln F}{dc}\right)\right] + cP_TF\left(\frac{d\ln F}{dc}\right). \tag{6}$$

The second term takes account of phonon-induced transitions to impurity d states. The usual approximation is that $d\ln F/dc$ is small and, therefore, apart from the linear temperature dependence of the phonon perturbation, the solute resistivity $\rho_r(\sim P_AF)$ has a temperature dependence similar to that of the pure solvent, ρ_{id}.

Many recent studies of transition-metal alloys have indicated that localized near the solute atoms there is a considerable perturbation of the lattice and, in particular, of the density of d states. Consequently, it is not reasonable to expect $d\ln F/dc$ to be small near the solutes. In order to relate

$d\ln F/dc$ to localized d states, specific assumptions must be made concerning the relative importance of s-s and s-d scattering and the extent to which the subbands conduct in parallel. The simplest approach is (i) to neglect s-s scattering and (ii) to assume spin mixing to be complete.

The consequence of the first assumption is the enhancement of the effect of changes in d states on the solute resistivity. With regard to the validity of assumption (ii), recent studies of deviations from Matthiessen's rule below 300 °K indicate that spin mixing may be complete at room temperature.[8,15,16] Above the Curie temperature where the subbands are equivalent assumption (ii) is not necessary. With these assumptions, F is simply proportional to the total density of d states at the Fermi surface $\eta_d(\epsilon_F)$.

In the Mott band model the temperature dependence of F arises from an exchange splitting of the d bands which varies with the spontaneous magnetization and, consequently, the density of d states at the Fermi energy $\eta_d(\epsilon_F)$ varies with temperature in each subband. The d states associated with the solutes are intimately related to the density of d states of the host and to the position of the Fermi energy within the band. Therefore, under the assumptions of the Mott band model, it is expected that $d\eta_d(\epsilon_F)/dc$ will also vary with temperature.

IV. EXPERIMENTAL RESULTS

In Figs. 1 and 2 the resistivity for the iron and nickel standards are shown along with the first derivative of the resistivity. The spin-disorder components ρ_{sd}, shown as dashed lines, were obtained by subtracting phonon contributions $\alpha_{ph}T$ with $\alpha_{ph} = 0.03$ and 0.025 $\mu\Omega$ cm/°K for iron and nickel, respectively. These values of α_{ph} are the same as those found by Weiss and Marotta.[17]

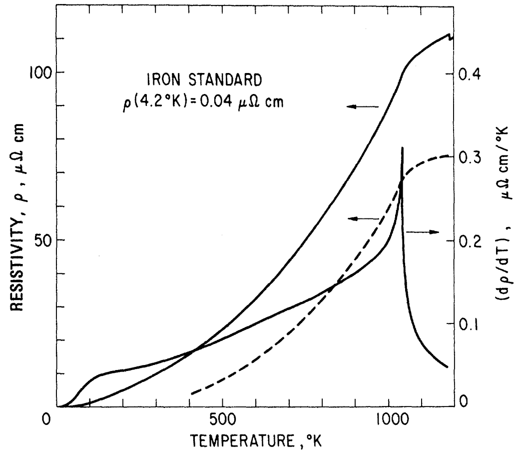

FIG. 1. Resistivity ρ and first derivative $d\rho/dT$ of iron standard, solid curves. Spin-disorder resistivity, dashed curve.

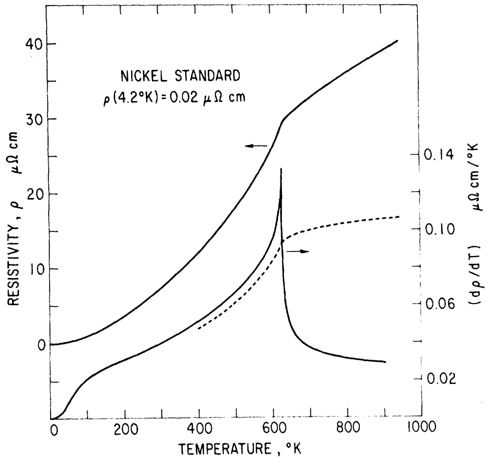

FIG. 2. Resistivity ρ and first derivative $d\rho/dT$ of nickel standard, solid curves. Spin-disorder component, dashed curve.

The resistivities of the alloys are analyzed and presented as solute resistivities $\rho_r(T)$ defined as the difference between the resistivity of the alloy and the resistivity of the host metal, i.e., the resistivity of the appropriate standard. "As-measured" values for $\rho_r(T)$ were obtained by subtracting the measured values of the resistivity of the standard without making adjustments for differences in Curie temperatures. Results for nickel-base alloys are presented in Figs. 3–6 and for

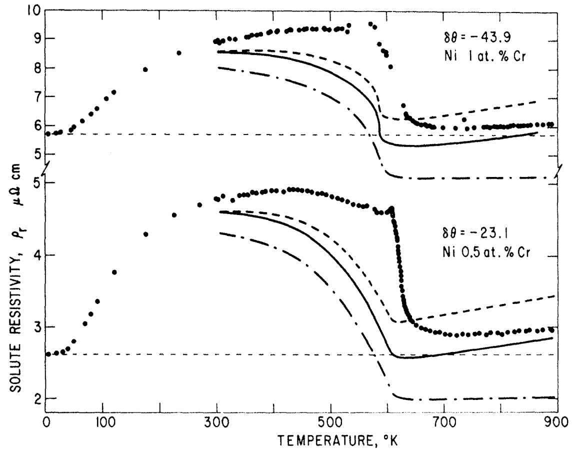

FIG. 3. Solute resistivity for NiCr alloys: uncorrected, closed circles; FM scaling, dash-dot curve; SD scaling, solid curve; MB scaling, dashed curve (see text). Horizontal dashed lines indicate resistivity at 4.2 °K.

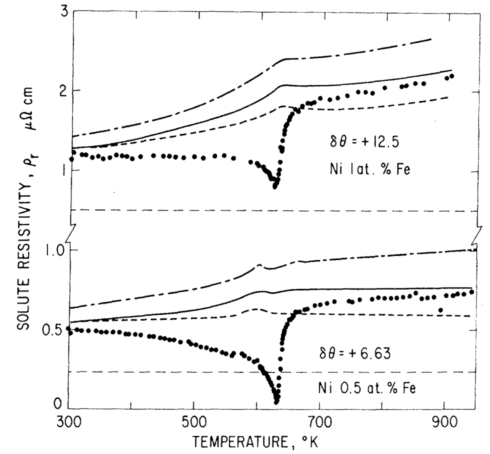

FIG. 4. Solute resistivity for *Ni*Fe alloys: uncorrected, closed circles; FM scaling, dash-dot curves; SD scaling, solid curves; MB scaling, dashed curves (see text). Horizontal dashed lines indicate resistivity at 4.2 °K.

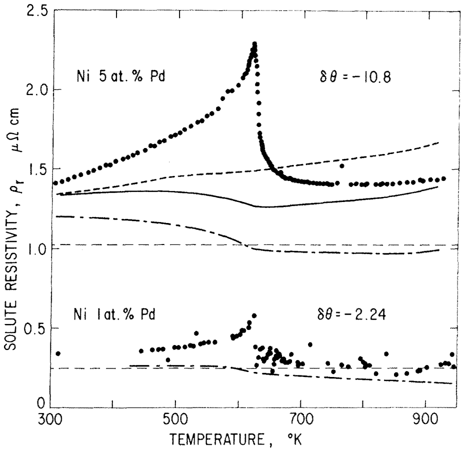

FIG. 6. Solute resistivity of *Ni*Pd alloys: uncorrected, closed circles; FM scaling, dash-dot curve; SD scaling, solid curves; MB scaling, dashed curves (see text). Horizontal dashed lines indicate resistivity at 4.2 °K.

iron-base alloys in Figs. 7–12. In all cases the full circles indicate the as-measured values of the solute resistivities. The general features of $\rho_r(T)$ are an increase from its value at 4 °K to a maximum or plateau near 300 to 400 °K, and large variations with temperature near θ.

The temperature dependence of the solute resistivity can be described as deviations from Mattthiessen's rule Δ defined as

$$\Delta = \rho_{\text{alloy}}(T) - \rho_{\text{alloy}}(0) - \rho_{\text{host}}(T) = \rho_r(T) - \rho_r(0) . \qquad (7)$$

Deviations from Matthiessen's rule for ferromagnetic alloys in the low-temperature range ($T \lesssim 300$ °K) have been studied by several authors.[6,8,15,16] The experimental results are in agreement with the general predictions of two-current models for transport properties. These models predict that the deviations Δ will approach a constant positive value at high temperatures, and the experimental results indicate that this maximum value is nearly obtained at 300 °K. On the basis of these remarks, it is expected that the increases in solute resistivity below ~ 300 °K may be understood in terms of multicurrent models for de-

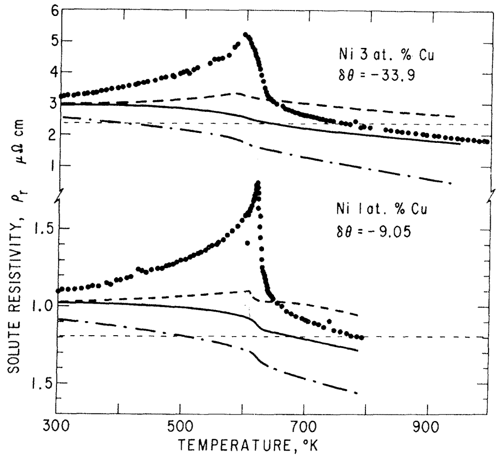

FIG. 5. Solute resistivity of *Ni*Cu alloys: uncorrected, closed circles; FM scaling, dash-dot curve; SD scaling, solid curves; MB scaling, dashed curve (see text). Horizontal dashed lines indicate resistivity at 4.2 °K.

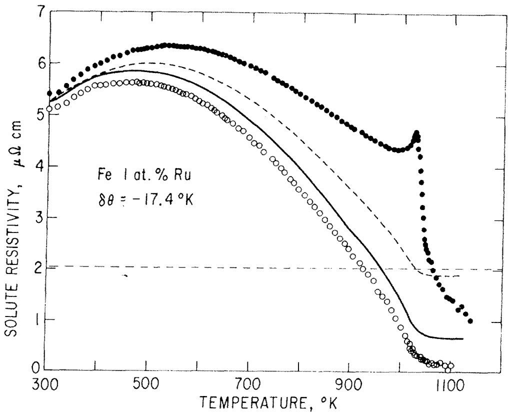

FIG. 7. Solute resistivity for Fe 1-at. % Ru: uncorrected, closed circles; FM scaling, open circles; SD scaling, solid curves; MB scaling, dashed curves (see text). Horizontal dashed line indicates resistivity at 4.2 °K.

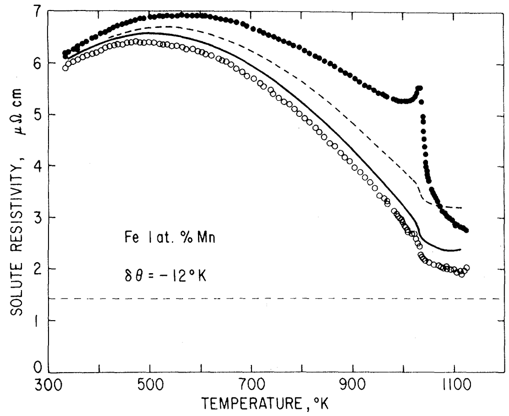

FIG. 8. Solute resistivity for Fe 1-at.% Mn: uncorrected, closed circles; FM scaling, open circles; SD scaling, solid curves; MB scaling, dashed curves (see text). Horizontal dashed line indicates resistivity at 4.2 °K.

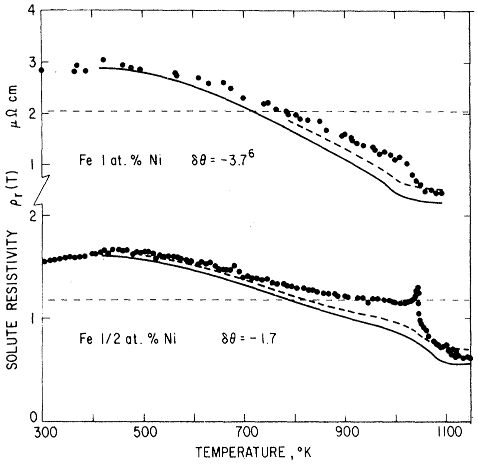

FIG. 10. Solute resistivity for *Fe*Ni alloys: uncorrected, closed circles; SD scaling, solid curve; MB scaling, dashed curves (see text). Horizontal dashed lines indicate resistivity at 4.2 °K.

viations from Matthiessen's rule. On the other hand, the temperature dependence of the solute resistivity above ~400 °K will reflect changes in the electronic properties and should be compared with the predictions outlined in Secs. I and III above. Since it is the latter phenomena with which this paper is primarily concerned, subsequent discussion will relate only to resistivities at temperatures greater than 300 °K. In view of the previous remarks, the value of the solute resistivity at ~300 °K becomes the reference value for discussing the temperature dependence of the solute resistivity. This reference value is considered to be a good approximation to the value of the solute resistivity of an alloy in a well-ordered magnetic state but with the multicurrent effects of deviations from Matthiessen's rule completely saturated.

The large variations in $\rho_r(T)$ near θ are due to the alloy and the standard having different degrees of magnetic order (see also Schwerer and Cuddy[5]). Corrections for this effect were made using the procedures described in Sec. III A. As emphasized above, the procedures for eliminating the effect

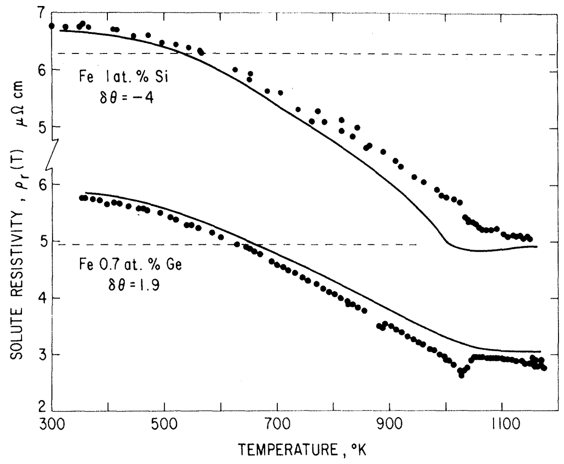

FIG. 9. Solute resistivity for Fe 1-at.% Si and Fe 0.7-at.% Ge: uncorrected, closed circles; SD scaling, solid curve. Horizontal dashed lines indicate resistivity at 4.2 °K.

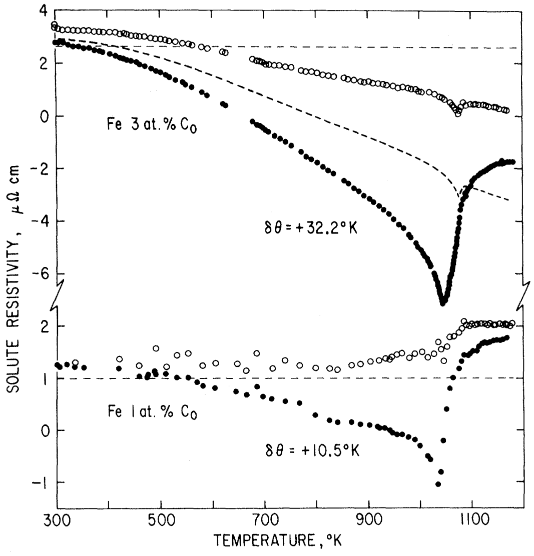

FIG. 11. Solute resistivity of *Fe*Co alloys: uncorrected, closed circles; SD scaling, open circles; MB scaling, dashed curves (see text). Horizontal dashed line indicates resistivity at 4.2° K.

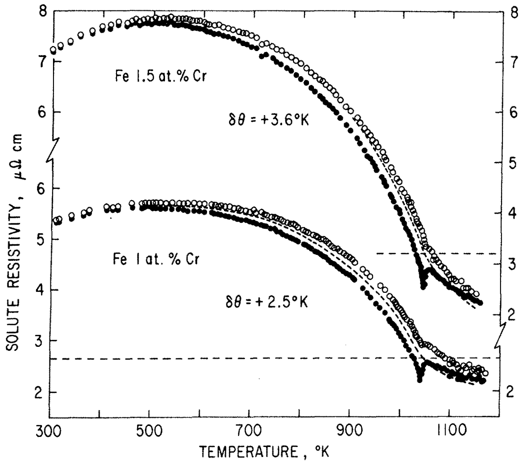

FIG. 12. Solute resistivity for *Fe*Cr alloys: uncorrected, closed circles; SD scaling, open circles; MB scaling, dashed curves (see text). Horizontal dashed line indicates resistivity at 4.2 °K.

of changes in the Curie temperatures depend on the model adopted to describe the ferromagnetic contributions to the resistivity. Accordingly, the data were corrected in three ways corresponding to the following assumptions: (i) The resistivity is all of ferromagnetic origin, i.e., the first term on the right-hand side of Eq. (3) or (4). (ii) Magnetic and lattice contributions to the resistivity may be described by a spin-disorder model [Eq. (3)]. (iii) The resitivity may be described by the Mott band model [Eq. (4)]. The results of applying these corrections are shown in Figs. 3–12 and are identified in the captions by the initials, FM, SD, and MB corresponding to the three different assumptions for the resitivity. The values of $\delta\theta (= \theta_a - \theta_0)$ used[18–20] are listed in Tables I and II.

It is obvious from Figs. 3–12 that this procedure eliminates much of the structure in the data and, in particular, the large rapid variations near the Curie temperatures. On the basis of a comparison of the qualitative features of the corrected solute resistivities, it is not possible to determine which of the three models for the resistivity is most appropriate; however, the results do support treating the ferromagnetic contributions to the resistivity as functions only of T/θ.

For some of the alloys, structure still remains near the Curie temperature. One reason for this is that the scaling parameters τ $(= \theta_0/\theta_a = 1 - \delta\theta/\theta_a)$ were not chosen to optimize the corrections and could be in error either due to uncertainties in the chemical analyses or in the interpretation of the published data. Near the Curie temperature the corrected results are quite sensitive to the choice of τ ($d\rho/dT$ is large near θ). At higher and lower temperatures, however, the solute resistivity was not nearly so sensitive, and adjustments of τ to remove this structure would not significantly affect the quantitative values presented below. The remaining deviations from a smooth temperature dependence could also arise from experimental uncertainties which are not properly averaged out by the simple numerical techniques used to interpolate the measured values of the resistivity of the standard. A more sophisticated treatment of the data from both the alloys and the standards is probably required.

In connection with the choice of τ (or $\delta\theta$), two observations are perhaps of interest. One is that the present data for the Fe 1-at.% Si alloy suggest a value of $\delta\theta \approx -4$ °K, which is more consistent with the early data of Fallot[20] than with more recent data.[19] Secondly, the resistivity data indicate a value of $\delta\theta \sim 2.5$ °K for the Fe 1-at.% W alloy in contrast to the tabulated data,[18] which indicate no change in θ upon addition of *W*.

The essential features of the corrected solute resistivities are a smooth variation with increasing temperature – the temperature dependence

TABLE I. Nickel-base alloy.

Solute (nom. at.%)	C (at.%)	$\delta\theta$ (°K)	$\delta\rho^{\text{ord-dis}}/C$ ($\mu\Omega$ cm/at.%)	S_i (μ_B)	$d\mu/dc$ (μ_B)	$\delta\rho_{\text{calc}}$ ($\mu\Omega$ cm/at.%)	$\delta\rho$ (obs) $\delta\rho$ (calc)
Pd(1)	1.00	− 2.24	(< 0.05)	...	0	0	...
(5)	4.76	−10.79	(< 0.02)				
Cu(1)	0.89	− 9.0	−0.18	0	−1.0	−0.3	1.7
(3)	2.76	−33.9	−0.18				
Cr(1/2)	0.54	−23.06	−3.9	+0.6	−6.0	0 (−3)[a]	1.2[a]
(1)	0.99	−43.85	−3.5				
Fe(1/2)	0.55	+ 6.6	+0.5	+2.8	+2.2	+0.37	1.8
(1)	1.06	+12.50	+0.7				

[a] Based on $d\mu/dc$.

TABLE II. Iron-base alloys.

Solute (nom. at %)	C (at.%)	$\delta\theta$ (°K)	$\delta\rho^{\text{ord-dis}}/C$ ($\mu\Omega$ cm/at.%)	S_i (μ_B)	$d\mu/dc$ (μ_B)	$\delta\rho_{\text{calc}}$ ($\mu\Omega$ cm/at.%)	$\delta\rho$(obs) $\delta\rho$(calc)
Co(1)	0.99	+10.5	+0.76	···		···	···
(1.5)	1.42	+17.0	−0.16	2.1	+1.2	−0.07	2.3
(3)	2.94	+32.2	−0.99	1.7		−0.37	2.7
Ni(1/2)	0.44	− 1.7	−2.3	0.9	+0.6	−0.95	2.6
(1)	0.94	− 2.8	−2.5				
Mn(1)	0.99	−12.0	−4.2	0	−2.0	−1.6	2.6
Ge(1)	0.71	+ 1.9	−3.7				
(2)	2.25	+ 1.2	−4.5				
Si(1)	0.97	− 4.0	−3.9		−2.2		
Cr(1)	1.06	+ 2.5	−3.1	−0.7	−2.4	−2.1	1.7
(1.5)	1.4	+ 3.6	−3.6				
(3)	3.11	+ 7.4	−3.6				
W(1)	0.94	+ 2.5	−3.7		−2.0		
Ru(½)	0.54	− 7.8	−5.1	0.9	−1.5	−0.95	5.6
(1)	1.04	−17.4	−4.9				

being greatest just below the Curie temperature. Above the Curie temperature the variation with temperature is relatively small.

V. DISCUSSION

A. Spin-Disorder Model

In this section the results are discussed in terms of a spin-disorder model. Consequently, the data referred to have been corrected for changes in θ using Eq. (3). The temperature dependence of the solute resistivities of the iron-base alloys is illustrated in Figs. 13 and 14, where the change in solute resistivity $\delta o_r(T)$ from its value in the reference state has been plotted with the data normalized to correspond to concentrations c of 1 at.%. The total change in resistivity $\delta\rho_r^{\text{ord-dis}}$ on going from the ordered ($T \approx 300\,°\text{K}$) to the disordered ($T \gtrsim \theta$) state is listed in Table II. Also shown are values of the magnetic moment S_i localized at the solute site.[21–23] For the *Fe*Co, *Fe*Ni, *Fe*Mn, *Fe*Si, and *Fe*Ge alloys, decreases of solute resistivity as well as a correlation of the relative mag-

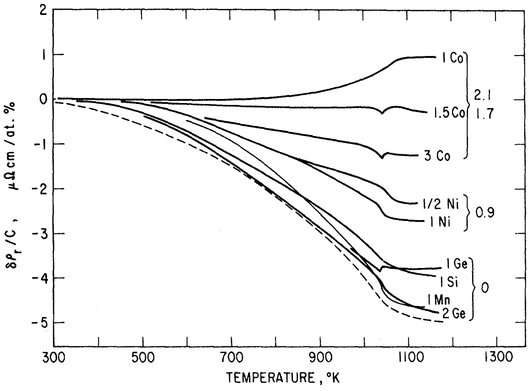

FIG. 13. Changes in solute resistivity for iron-base alloys (SD scaling) normalized to concentrations of 1 at.% and referred to maximum value (see text). Numbers beyond brackets indicate local moments in μ_B. Curves are labeled by approximate solute contents. Dashed curve is the negative of the spin-disorder contribution to the resistivity of iron in arbitrary units.

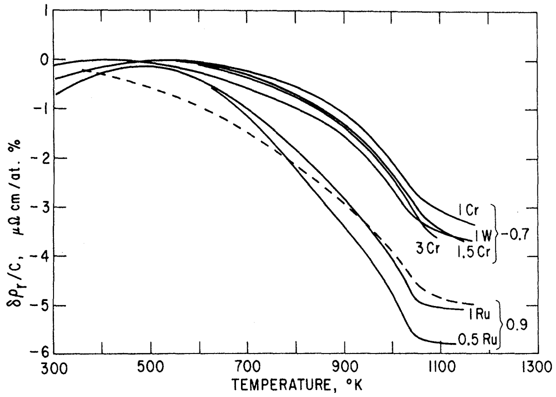

FIG. 14. Changes in solute resistivity for iron-base alloys (SD scaling) normalized to concentrations of 1 at.% and referred to maximum value (see text). Numbers beyond brackets indicate local moments in μ_B. Curves are labeled by approximate solute contents. Dashed curve is the negative of the spin-disorder contribution to the resistivity of iron in arbitrary units.

nitude of the decrease with solute moment are observed in agreement with Eq. (5). The Fe 1-at.% Co alloy represents an exception and is discussed below. It should be emphasized that despite large differences in their effects on other properties [e.g., the residual resistivity $\rho(4.2\,^\circ\mathrm{K})$ or low-temperature deviations from Matthiessen's rule Δ], those solutes which act as simple dilutents (i.e., $S_i \cong 0$), including both transition and nontransition elements, have nearly the same value for the change in solute resistivity $\delta\rho_r^{\text{ord-dis}}$. However, for all these solutes, Co, Ni, Mn, Si, and Ge, the observed changes $\delta\rho_r(\text{obs})$ are larger than the calculated values $\delta\rho_r(\text{calc})$ by a factor of ~2.6. (For iron-base alloys the values $S_0 = 2.2\ \mu_B$ and $\rho_{\text{sd}}^0 = 80\ \mu\Omega\,\text{cm}$ were used in these calculations.)

The existence in the reference state (~300 °K) of either magnetic disorder or incomplete saturation of Δ would reduce the observed values of $\delta\rho_r^{\text{ord-dis}}$. The observed values, however, are all larger than those calculated from Eq. (5). This discrepancy possibly arises in the following way. In deriving Eq. (5), the factor involving the various electronic parameters, the exchange constant G, and the host spin $S_0(S_0+1)$ were set equal to the high-temperature value of the spin-disorder resistivity ρ_{sd}^0. However, at least for spin-$\frac{1}{2}$ systems, the interaction H_I also contains a temperature-independent term. In the pure host, this term is periodic with the lattice and, consequently, will not contribute to the resistivity. (When this term is subtracted from H_I for the pure host, only spin-flip processes remain.) In the alloy, on the other hand, the perturbations due to the solutes are nonperiodic and the full interaction H_I contributes to the resistivity. As an approximation of this effect, ρ_{sd}^0 in Eq. (5) can be multiplied by $\frac{3}{2}(S_0+1)$, where the $\frac{3}{2}$ comes from the spin of the conduction electrons.[24] For the iron-base alloys this would increase the calculated values by a factor of 3, which is close to the observed value of 2.5.

The solutes Cr and W have very similar effects on the resistivity of iron, see Fig. 14. This is in agreement with the spin-disorder model since both have moments at the solute site of $\sim -0.7\ \mu_B$. However, the solute resistivities for Cr and W solutes are still quite temperature dependent above the Curie temperature, and the observed change in the solute resistivity $\delta\rho_r^{\text{ord-dis}}$ is the smallest of the iron alloys ~1.7 times the calculated value.

In Figs. 13 and 14 dashed curves indicate the temperature dependence of the spin-disorder resistivity of pure iron ($-\rho_{\text{sd}}$ is plotted in arbitrary units). The temperature dependence of the solute resistivity for Co, Ni, Si, and Ge solutes is very similar to that of ρ_{sd}. However, for Mn and Ru solutes, the change in resistivity with increasing temperature is slower at low temperatures and becomes more rapid just below the Curie temperature. There is evidence from nuclear-magnetic-resonance studies that the polarization of the Mn moment in iron has a temperature dependence different from that of the bulk magnetization.[25] This would affect the shape of the solute resistivity curve. Similarly, the large temperature dependence of the solute resistivity of the *Fe*Cr and *Fe*W alloys above the Curie temperature could be due to an unusual temperature dependence of the polarization of these antiferromagnetic (negative) moments, and the relatively low value of the observed change $\delta\rho_r^{\text{ord-dis}}$ in solute resistivity could reflect short-range magnetic order associated with the Cr and W solutes which persists to unusually high temperatures.

Although the temperature dependence of the solute resistivities for the *Fe*Ru alloys is similar to that for the *Fe*Mn alloy, the magnitude of the change is larger than the predicted value by a factor of 5.6. The calculated value is based on a solute moment of $0.9\ \mu_B$ for Ru which is close to the value of Ni solutes. However, the effect of Ru on the total moment μ of iron ($d\mu/dc \sim -1.5\ \mu_B$ for Ru)[26] differs greatly from that of Ni ($d\mu/dc \sim +0.6\,\mu_B$).[27] In addition, neutron-diffraction studies[21] indicate that the magnetization disturbance associated with the Ru solutes has a complex spatial dependence and involves many iron neighbors. Consequently, the unusually large value of $\delta\rho_r(\text{obs})/\delta\rho_r(\text{calc})$ indicates that it is necessary to consider the changes in the moments localized at iron sites as well as at the solute. Since Eq. (5) is linear in the local moments, it is tempting to sum the various local moments to obtain the total effect on $\delta\rho_r$. Assuming the entire moment disturbance is in localized states, this summation results in the prediction that $\delta\rho_r/c$ is proportional to $d\mu/dc$. Taking neighboring moments into account in this fashion does not resolve the discrepancies, particularly since comparisons of the observed signs of $\delta\rho_r^{\text{ord-dis}}$, of $(S_i - S_0)$, and of $d\mu/dc$[27] for *Fe*Ni and *Fe*Co indicate a much better correlation of $\delta\rho_r^{\text{ord-dis}}$ with the solute moment S_i than with the total moment disturbance $d\mu/dc$. For Co and Ni solutes, neutron-diffraction studies[21] indicate considerable moment disturbance extending some 8 Å from the solute site. These disturbances are of a single sign and do not show the complex variations associated with Ru solutes. Consequently, for the relatively heavy concentrations studied here, solute spacings ~10 Å, this disturbance due to Co and Ni solutes may approximate a uniform enhancement of the iron moments with a localized spin disturbance at the solute site. On the other hand, the complex spatial variations of the moment disturbance due to the Ru solutes are more localized and will not appear as a uniform background.

Consistent with this interpretation is the observed dependence of the normalized solute resistivity $\delta\rho_r^{\text{ord-dis}}/c$ on the concentration of Co solutes. The solute resistivity $\delta\rho_r^{\text{ord-dis}}$ for the 1.5-at.% alloy is consistent with the localized moment 2.1 μ_B found by Collins and Low[21] for their Fe 1.5-at.% Co sample, while the resistivity for the 3-at.% sample suggests a local moment of 1.7 μ_B, which is closer to that found by workers on more concentrated alloys.[22]

Values of $\delta\rho_{\text{sd}}^{\text{ord-dis}}/c$ for nickel-base alloys are given in Table I. For Pd solutes, there is essentially no change in the solute resistivity associated with the magnetic transition, i.e., $\delta\rho_r^{\text{ord-dis}} = 0$. Additions of palladium to nickel in concentrations to 15 at.% are known[27] to have no effect on the saturation moment of the alloy. Consequently, it is reasonable to assign the value $S_i = S_0$ to the local solute moment for Pd in nickel. The observed values of $\delta\rho_r^{\text{ord-dis}}$ for the two *Ni*Pd alloys are, therefore, in agreement with the predictions of the spin-disorder model.

Neutron diffraction studies indicate a well-localized moment of $+2.8$ μ_B associated with Fe solutes in nickel.[21] Recent studies[28,29] of Cu solutes in nickel suggest there is zero moment at the solute site with diminution of the moment on neighboring nickel atoms. Calculated values for $\delta\rho_r^{\text{ord-dis}}$ for Fe and Cu solutes are listed in Table I. (These calculations are based on the values $S_0 = 0.6$ μ_B and $\rho_{\text{sd}}^0 = 15$ $\mu\Omega$ cm for nickel.[17]) The observed values are about 1.7 times the calculated values. [As described above, if only spin-flip scattering processes contribute to ρ_{sd}^0, then the observed values are expected to be approximately twice those calculated using Eq. (5).] Consequently, the solute resistivities for *Ni*Pd, *Ni*Cu, and *Ni*Fe alloys are in accord with the predictions of the localized spin-disorder model as regards the sign and relative magnitude of the change in resistivity associated with magnetic ordering.

For Cr solutes in nickel, neutron-diffraction studies[21] indicate that the moment at the solute site is essentially the same as at the nickel sites. There is, however, a moment disturbance involving neighboring nickel atoms which accounts for the decrease in bulk moment of 6 μ_B per Cr solute. If the total disturbance is used, the calculated value for $\delta\rho_r$ is -3 $\mu\Omega$ cm, which has the same sign and is approximately 1.2 times less than the observed values. The use of the total moment disturbance $d\mu/dc$ was discussed previously and appears to be justified for the case of Cr solutes in nickel.

B. Mott Band Model

In the Mott band model the temperature dependence of the solute resistivity is contained in the function F and reflects changes in the density of d states at the Fermi energy. An analysis by Birss and Dey[30] of the resistivity of pure nickel in terms of this model indicates that the value of F in the paramagnetic state F_p is twice the value in the ferromagnetic state F_f. Estimates based on theoretical band calculations and the assumption that F is proportional to the total density of states at the Fermi energy gives values for F_p/F_f of ~2.4 for iron[31,32] and ~2.1 for nickel.[33,34] Owing to the approximations involved, little significance should be attached to the numerical values, although it is reasonable to expect F_p/F_f to be significantly greater than unity for both iron and nickel. This means that the value of the solute resistivity will be appreciably larger (by approximately a factor of 2) in the paramagnetic region than in the low-temperature ferromagnetic region. Behavior of this type was observed only for the Ni 1-at.% Fe and the Fe 1-at.% Co samples (see Figs. 3–12). It is clear that if the Mott band model describes the temperature dependence of the solute resistivity, the temperature dependence of the localized states associated with the solutes must be taken into account. The data indicate that these changes in $\eta_d(\epsilon_F)$ are such that the relevant density of states with regard to solute scattering is greater in the ferromagnetic than in the paramagnetic state [see Eq. (6)]. The exceptions are Pd and Fe solutes in nickel. The data for the *Ni*Pd alloys indicate that the density of states at the Fermi energy is the same in the ferromagnetic and paramagnetic states.

Empirical estimates of the effect of solute additions on F can be based on measurements of electronic specific heat γ if assumptions are made regarding the extent to which the effects are localized at the solute site. The large negative values of $d\ln\gamma/dc$ obtained for several solutes in iron[35] and nickel[36] indicate that the entire change in γ cannot be ascribed to changes in the density of states for the solute atom. On the other hand, if the change in density of states is uniform throughout the sample, then the effect on F is small and ρ_r is still expected to increase with temperature.

A more detailed comparison of these resistivity data with the Mott band model must be deferred until theoretical calculations of the density of states at the Fermi level in the ferromagnetic and paramagnetic states are available for solutes in iron and nickel.

VI. SUMMARY

Experimental determinations of the effect of various solutes on the electrical resistivity of iron and nickel have been presented. The solute resistivities for both the iron- and nickel-base alloys were in general agreement with a crude development of a localized spin-disorder model. Insofar as comparisons of the results with the Mott band model were possible, the agreement was not satisfactory. The principal facts in support of the spin-disorder interpretation are as follows: (a) For all the solutes studied, in both iron and nickel, the spin-disorder model predicts the correct sign for the change in solute resistivity $\delta\rho_r^{\text{ord-dis}}$. (b) For the solutes Co, Ni, Si, Ge, and Mn in iron and Pd, Fe, and Cu in nickel, the relative magnitude of $\delta\rho_r^{\text{ord-dis}}$ is in agreement with the spin-disorder model and with the moment at the solute site as determined by neutron-diffraction experiments. (c) Cr and W solutes in iron have the same value of the local moment ($-0.7\ \mu_B$) and the solute resistivities show similar behavior. (d) Those solutes which act as dilutents in iron ($S=0$), both transition and nontransition atoms, have similar effects on the solute resistivity. (e) Pd solutes in nickel, with presumably the same moment as the solvent, gave no change in solute resistivity, in agreement with the spin-disorder model and in striking contrast to the Mott band model.

Certainly many features of the experimental results are not described by the simple spin-disorder model used. In particular, the model neglects spin disorder on sites other than the solute site. This neglect is particularly serious for the Cr solutes in Ni and for the complex spin structure associated with the Ru solutes in iron. No attempt has been made to investigate the contribution to the low-temperature resistivity from spin-flip scattering by solutes. However, we feel that the agreement between the experimental results and the spin-disorder model described here strongly suggests that a spin-disorder scattering mechanism is dominant in the high-temperature solute resistivities of iron- and nickel-base alloys.

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Resistive Behavior of Dilute *Pd* Fe Alloys about the Curie Temperature*

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We have analyzed the behavior of the magnetic part of the electrical resistivity ρ_m for four dilute samples of *Pd*Fe(0.1, 0.25, 0.50, and 1.0 at. % of Fe) in the vicinity of the Curie temperature. The ferromagnetic critical temperature for these samples was determined by the location of the maximum in $d\rho_m/dT$ and was found to vary with concentration as c^n, with $n = 1.65 \pm 0.05$. The "singularity" of $d\rho_m/dT$ was found to be much stronger than logarithmic with the exponents varying over a wide range of values. Also, we find that particularly in one sample, $Pd_{0.995}Fe_{0.005}$, the long-range nature of the short-range spin fluctuations plays a significant role in determining the behavior of $d\rho_m/dT$.

I. INTRODUCTION

The magnetic properties of dilute magnetically ordered alloys have been a subject of extensive study in recent years. In such a study, one looks for the basic mechanism responsible for the formation of localized moments, their interaction with the matrix, and the nature of the transition to the ordered state. The understanding of the nonequilibrium properties, such as the transport coefficients, in these systems has left much to be desired. Even though widely different systems do behave fairly similarly with regard to their equilibrium properties near the critical temperature, the transport coefficients do show striking dissimilarities. These differences in the critical behavior of the transport coefficients are expected to shed a great deal of light on the various interaction mechanisms in these systems.[1]

The palladium-iron alloys have the characteristic property of forming a well-defined ferromagnetic state even for very low concentrations of iron. In fact, in the case of *Pd* Fe, there exist distinctly different regions of concentration-dependent behavior. For example, the ordering temperature increases linearly with concentration in the concentration range 1.0 to about 4 at.% of iron, and in the concentration range 4–12 at.% of iron but with a slope which is smaller by a factor of about 3.[2] Above 1 at.% of Fe concentration, the palladium matrix shows an increasingly homogeneous magnetic polarization, and at about 3 at.% of iron, where the neutron-scattering cross section becomes effectively independent of the scattering vector, the polarization is uniform.[3] On the other hand, in the case of lower concentrations (1% and below), the ordering temperature varies more strongly than linearly with concentration. For concentrations less than 1 at.% of Fe the neutron-scattering data[3] have shown that the polarization of the palladium matrix extends out from an iron atom with a range of about 10 Å. This giant moment (as large as 10 μ_B for $\frac{1}{4}$% Fe) in these alloys is the result of contributions from a large number of palladium atoms in the matrix affected by a single iron atom.

Theoretical studies of various properties for these alloys have been carried out by many workers. In these studies, both spin-wave and cluster models have been used with reasonable success.[4–8] In particular, Coles and Turner,[7] and Doniach and Wohlfarth[6] have demonstrated that damped spin waves give an enhanced contribution to the specific heat, which is in fair agreement with the experimental results of Veal and Rayne.[9] Also, the studies of the temperature dependence of the electrical resistivity of dilute *Pd* Fe alloys by Williams and Loram[10] are explained on the basis of scattering contributions from the spin waves.

The critical behavior of these alloys to the ordered state is of particular interest, since, quite evidently, in this behavior the matrix plays a very significant role.[11–13] Our previous work[14] has emphasized the high-concentration regime, thereby limiting the information to be gained about the effects of the long-range part of the polarization which we expect to be more important at low concentrations. In an effort to better understand the dynamic aspect of the spin ordering in the lower-concentration range, we have studied the critical behavior of the electrical resistivity of four samples of dilute *Pd* Fe alloys in the vicinity of the or-