PHYSICAL REVIEW B VOLUME 9, NUMBER 5 1 MARCH 1974

Ferromagnetic properties at high fields and high pressures of nickel-platinum alloys near the critical concentration for ferromagnetism

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(Received 30 April 1973)

The magnetic properties of ferromagnetic Ni-Pt alloys containing 42.9, 45.2, 47.6, and 50.2 at.% Ni were measured in magnetic fields up to 60 kOe, at temperatures between 4 and 60 K, and at pressures up to 7 kbar. The results are presented in terms of Arrott and Mathon plots whose essential straightness over a broad range of alloying contents and external influences provides evidence for the close applicabilities of weak-ferromagnetic theory based on the itinerant-electron model. This model is used to discuss the results quantitatively and the pressure dependence of the critical concentration is estimated, in good agreement with observation. The effective interaction U is found to be 0.44 eV for these alloys, while the ratio of U to the bare interaction is found to be 0.65. The results are compared quantitatively with those for other similar materials, in particular $ZrZn_2$.

I. INTRODUCTION

The itinerant-electron model of ferromagnetism in weakly ferromagnetic materials has been applied in a number of instances in recent years [Fe-Ni (Invar) alloys, Ni_3Ga and Ni_3Al, $ZrZn_2$, etc.]. Following previous work of Besnus and Herr[1] and Gillespie and Schindler[2] we present here data in high fields and at high pressures for Ni-Pt alloys near the critical concentration c_F where ferromagnetism appears in the disordered alloys. Earlier evidence points to these alloys being more homogeneous in their magnetization than Ni–Cu and other nickel alloys, so that the model has a greater chance of success in this case. The pressure experiments are believed to be more extensive in the present work than in previous investigations on nickel alloys. They were partly aimed at testing the constancy of the Landau parameter B under pressure and partly to establish any analogies between these nickel alloys and other materials already known to have Invar characteristics.

In Sec. II the magnetic properties of the ferromagnetic alloys at normal pressures are represented in the form of Arrott plots (M^2 vs H/M) and Mathon plots (saturation magnetization squared, Curie temperature squared, and inverse high-field susceptibility varying with concentration). The linear relationships observed give evidence for the applicability of the model. In Sec. III the pressure data are analyzed in the same way. The results are interpreted on the basis of the model in Sec. IV and a number of important parameters are estimated numerically, in particular, the effective interaction and the bare interaction energies for the itinerant electrons.

II. MAGNETIC PROPERTIES AT NORMAL PRESSURES

Platinum and nickel can be mixed in any proportions to form a disordered solid solution having fcc structure. The disordered Ni-Pt alloys of composition close to $Ni_{0.75}Pt_{0.25}$ and $Ni_{0.5}Pt_{0.5}$ can, however, be ordered by a suitable heat treatment.[3] The disordered Ni-Pt alloys rich in nickel are ferromagnetic. Their Curie temperature and spontaneous magnetization increase with the atomic concentration c of nickel (Fig. 1). The critical concentration for the appearance of ferromagnetism is close to 0.42 according to Besnus and Herr[1] and close to 0.44 according to Gillespie and Schindler.[2] The alloys used in this study have concentrations c in nickel of 0.429, 0.452, 0.476 and 0.502. They were supplied by Besnus and Herr. They have been prepared by high-frequency melting in an alumina crucible, in a purified argon atmosphere with 99.99% pure Pt and Ni rods. After heat treatment at 1000 °C for 12 h they were water quenched to room temperature. The Fe content was examined in the $Ni_{0.452}Pt_{0.548}$ sample. It was found to be 140 ppm. The influence of iron atoms on the magnetic properties of the four alloys under consideration will be neglected.

A. Predictions of homogeneous-weak-ferromagnetism theory

Edwards and Wohlfarth[6] and Mathon[7] have applied the Landau theory of second-order phase transitions[8] to the magnetic properties of very weak ferromagnets (WFM). It was assumed that the magnetization was spatially homogeneous. The difference between the free energy of the ferromagnetically ordered and disordered states is then

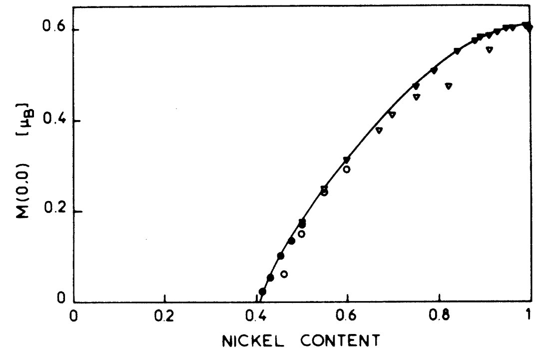

FIG. 1. Variation with atomic nickel concentration of the spontaneous magnetization $M(0,0)$: Marian (Ref. 4) ▽; Fisher and Besnus (Ref. 5) ▼; Gillespie and Schindler (Ref. 2) ○; present work ●.

$$\Delta F = \tfrac{1}{2}A(T)M^2(H,T) + \tfrac{1}{4}B(T)M^4(H,T) - HM(H,T) \tag{1}$$

disregarding, for the time being, the effects of magnetostriction; M is the magnetization at field H and at temperature T. In the case of a WFM when $M(H,T)$ is much smaller (in Bohr magnetons) than the number of holes per atom, expression (1) applies not only at the phase-transition temperature (Curie temperature) but also at low temperatures, when the Landau theory is more likely to be correct.[7] Minimizing ΔF leads to the implicit equation for M:

$$[A(T) + B(T)M^2(H,T)]M(H,T) = H. \tag{2}$$

If one calls $\chi(H,T) = [\partial M(H,T)/\partial H]_{H,T}$ the high-field susceptibility of the WFM state, then $A(T)$ and $B(T)$ can be expressed as

$$A(T) = -[2\chi(0,T)]^{-1}$$

and (3)

$$B(T) = [2M^2(0,T)\chi(0,T)]^{-1},$$

where A thus changes its sign at the Curie temperature. We will take the Stoner law[6]

$$M^2(0,T) = M^2(0,0)[1 - T^2/T_C^2]$$

and further (4)

$$\chi(0,T) = \chi(0,0)[1 - T^2/T_C^2]^{-1}$$

to describe the thermal variation of $M(0,T)$, and $\chi(0,T)$, so that, in line with Landau theory, B is independent of temperature. Then

$$\left[\frac{M(H,T)}{M(0,0)}\right]^2 = 1 - \left(\frac{T}{T_C}\right)^2 + \frac{2\chi_0 H}{M(H,T)}, \tag{5}$$

where $\chi_0 = \chi(0,0)$, which extends the Arrott plots[9] to a wider temperature range. Thus WFM theory predicts a linear dependence of $M^2(H,T)$ as a function of $H/M(H,T)$ with a slope $1/B$ independent of temperature. At the Curie temperature $T = T_C$, $M^2(H,T)$ is simply proportional to $H/M(H,T)$ [Fig. 2(a)].

The Landau theory of WFM has been used by Mathon[7] to discuss the effects of concentration in binary alloys near the critical concentration for the appearance of ferromagnetism. Within Landau theory there exists a close analogy between what happens for a fixed concentration when the temperature is varied and what happens for a fixed temperature when the concentration is varied. One thus has

$$\begin{aligned} A(T,c) &= a(T)[c_F(T) - c], \\ B(T,c) &= B, \end{aligned} \tag{6}$$

where c_F is the critical concentration, at temperature T, where the alloy becomes ferromagnetic.

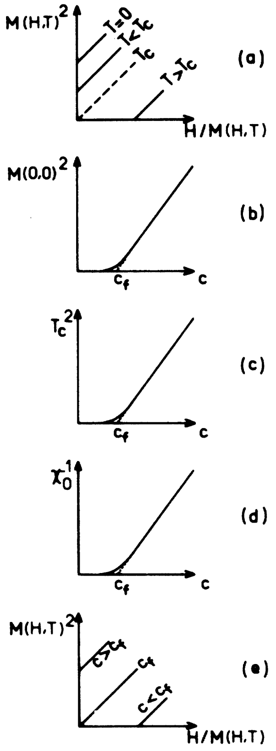

FIG. 2. Results of weak-ferromagnetism theory (normal pressures). (a) Variation of the square of the magnetization $M(H,T)$ as a function of $H/M(H,T)$; H is the applied field and T is the temperature (Arrott plot). (b) Variation of the square of the spontaneous magnetization $M(0,0)$ at $T = 0$ K, as a function of concentration. (c) Variation of the square of the Curie temperature T_C as a function of concentration. (d) Variation of the inverse of the initial susceptibility χ_0 as a function of concentration. (e) Variation of the square of the spontaneous magnetization $M(H,T)$ as a function of $H/M(H,T)$ for constant temperature T and various concentrations.

Thus one has a linear dependence of $M^2(0,0)$, T_C^2, and χ_0^{-1} on $c_F(0)-c$, which has been called Mathon plots [Figs. 2(b), 2(c), and 2(d)]. The validity of these relationships is limited to values of $c-c_F(0)$ large enough so that the susceptibility is smaller than a constant value which depends on the Landau parameters A and B[10] and small enough so that A depends linearly on temperature and B can be considered as a constant.

The constancy of B as a function of the concentration c should be related, in Arrott plots, to the slopes of such plots being independent of concentration c [Fig. 2(e).

B. Experimental results

The magnetization of the ellipsoidally shaped samples, weighing about 1 g has been measured in a superconducting magnet between 4 and 60 K, at fields up to 60 kOe. The experimental techniques have been described elsewhere.[11,12] Some magnetization data have been obtained in fields up to 150 kOe, in a water-cooled solenoid. The results obtained are extensions of those of Besnus and Herr[1] and Gillespie and Schindler.[2] In the first work, fields up to 28 kOe at 4.2, 20, and sometimes 1.4 K were used, whereas Gillespie and Schindler used fields up to 87 kOe but at 1.5 and 4.2 K only.

The magnetization of $Ni_{0.429}Pt_{0.571}$, as determined in the supraconducting magnet, is plotted in Fig. 3 as an Arrott plot. Equation (5) is well obeyed in the experimental temperature range, except at the lowest magnetic fields. The larger the concentration c, the smaller is the field from which a linear variation of $M^2(H,T)$ as a function of $H/M(H,T)$ is observed. No deviation from linearity for $M^2(H,T)$ as a function of $H/M(H,T)$ has been observed from 20 to 150 kOe, between 4.2 and 77 K. The Landau parameter B can thus be considered as a constant, as indicated by the parallelism of the Arrott plots for the various temperatures under consideration. The intercept $I(T)$ of the straight lines with the $M^2(H,T)$ axis is given in Fig. 4; $I(T)$ is a linear function of T^2, in good agreement with the Stoner law [Eq. (4)]. The Arrott plots allow the determination of T_C, $M(0,0)$, and χ_0. Their values are given in Table I. Such quantities as $M(0,0)/T_C$ or $\chi_0 M^2(0,0)$ are reasonably constant, in agreement with the theory. We have reported, Fig. 5, the Arrott plots, at 4.2 K for the various alloys under consideration. Thus the prediction according to which B is independent of concentration is confirmed. The closer the critical concentration, the larger the influence of magnetic or chemical clustering, to which should be attributed the observed low-field deviations, as it can be observed (Fig. 3) in the nearly critical $Ni_{0.429}Pt_{0.571}$ alloy.

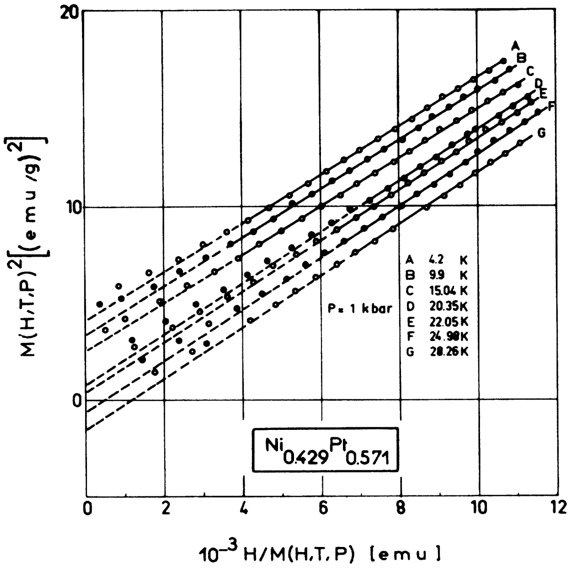

FIG. 3. Arrott plots for $Ni_{0.429}Pt_{0.571}$.

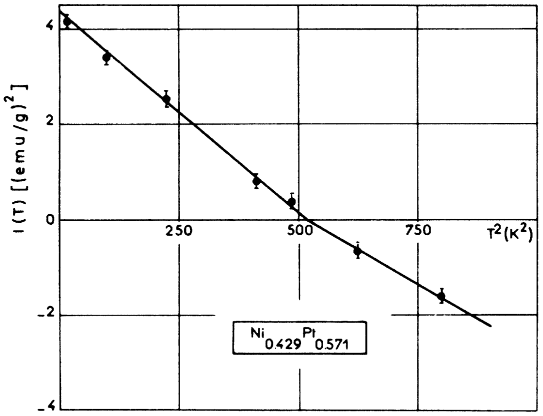

FIG. 4. Intercept $I(T)$ (see text) as a function of the square of the temperature in $Ni_{0.429}Pt_{0.571}$.

The data obtained can be compared to those obtained for Zn_2Zr by Ogawa and Sakamoto[13]: $M(0,0) = 0.037\mu_B/g$ atom, $\chi_0 = 7.3\times10^{-3}$ emu/g atom Oe^{-1} and $T_C = 16.9$ K. As observed for Zn_2Zr by Edwards and Wohlfarth,[6] the $I(T)$ points as a function of T^2 for the $Ni_{0.429}Pt_{0.571}$ and $Ni_{0.414}Pt_{0.586}$ alloys also fall on two straight lines, apparently meeting at $T=T_C$. This was tentatively ascribed to short-range ordering above T_C, but this interpretation is uncertain and the phenomenon unexplained. The values $M(0,0)^2$, T_C^2 and χ_0^{-1} can be considered approximately as a linear function of concentration c from $c\simeq0.42$ to $c\simeq0.48$ (Fig. 6), The difference between the obtained critical concentrations $c_F=0.42$ and $c_F=0.44$ as given by Gillespie and Schindler[2] can be partly attributed

TABLE I. Variation of the spontaneous magnetization $M(0,0)$, Curie temperature T_c, and initial susceptibility χ_0 with nickel concentration c.

c	0.429	0.452	0.476	0.502
$M(0,0)$ emu/mole	283	579	798	1000
μ_B	0.051	0.104	0.143	0.179
T_C (K)	23.0	54.2	74[a]	100[a]
$10^2\chi_0$ (emu/mole)	1.98	0.625	0.393	0.241
$M(0,0)/T_C$ emu/mole K	12.3	10.7	10.8	10.0
$10^{-2}\chi_0 M(0,0)^2$ emu/mole	16	21	25	24

[a] Besnus and Herr (Ref. 1).

to accuracy of the chemical concentration and partly to small differences between metallurgical treatments. This difference has only very small consequences on the analysis of the actual data.

C. Conclusions

(i) Plots of $M^2(H,T)$ against $H/M(H,T)$ are parallel straight lines over a wide range of temperature, fields, and alloying contents which means that the Landau parameter B is reasonably constant, independant of temperature, fields, and alloying contents.

(ii) The intercept $I(T)$ of the Arrotts plots on the M^2 axes is linear in T^2 above and below the Curie temperature.

(iii) Plots of $M^2(0,0)$, T_C^2, and χ_0^{-1} versus alloying content are approximately linear over a large range of alloying contents.

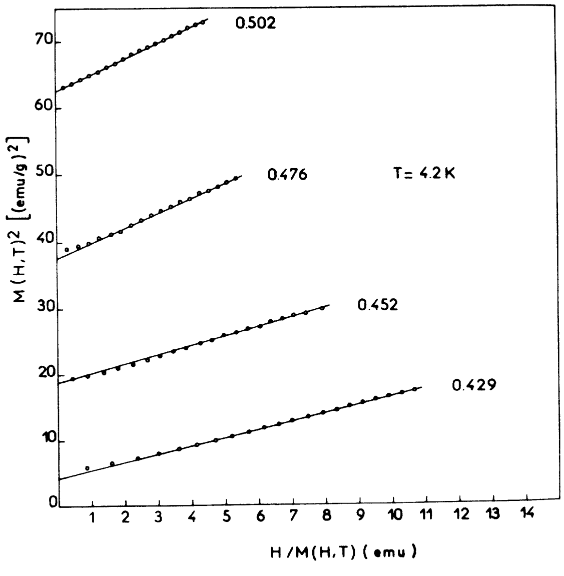

FIG. 5. Arrott plots for various Ni-Pt alloys at 4.2 K.

III. MAGNETIC PROPERTIES AT HIGH PRESSURES

We have shown in Sec. II that the variations with concentration and temperature of the magnetic properties of nickel-platinum alloys at normal pressures are essentially governed by the dependence on concentration and temperature of the Landau parameter A. The Landau parameter B was, at least as a first approximation, independent of con-

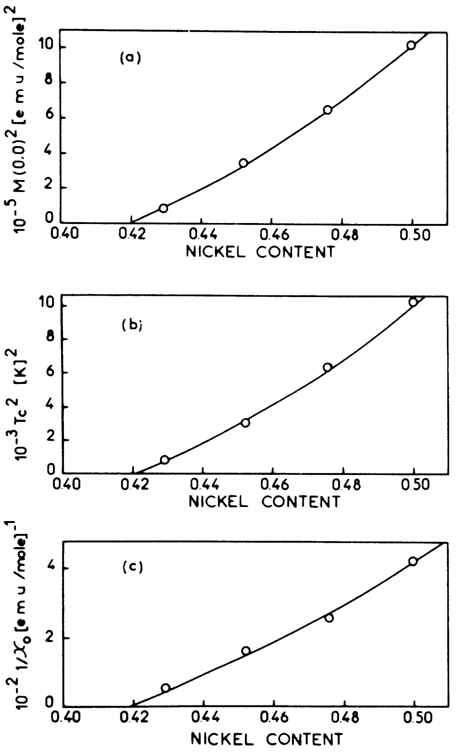

FIG. 6. Variation with atomic nickel concentration of (a) the square of the spontaneous magnetization $M(0,0)$; (b) the square of the Curie temperatures (from Besnus and Herr, Ref. 1); (c) the inverse initial susceptibility χ_0.

centration and temperature. We have undertaken to study their magnetic properties under high pressure in order to study (i) the variation of T_C, $M(0,0)$, and χ_0 with interatomic distances and the correlations between their variations, (ii) the variations of A and B with interatomic distances, and (iii) the pressure dependence of the critical concentration.

A. Predictions of thermodynamics and WFM theory

On the same footing as B was earlier considered as independent of temperature and concentration in the neighborhood of the critical concentration, we will assume and then verify that B is also independent of pressure P, at least of moderate pressures. Taking into account a possible variation of A with pressure, Eq. (1) becomes

$$\Delta F(P) = \tfrac{1}{2}A(T,P)M^2(H,T,P) + \tfrac{1}{4}BM^4(H,T,P) - HM(H,T,P) \tag{7}$$

with the corresponding Stoner equation

$$M^2(0,T,P) = M^2(0,0,P)[1 - T^2/T_c^2(P)] \,. \tag{8}$$

The coefficient A can be written as

$$A(T,P) = A(T,0) + \frac{\partial A(T,0)}{\partial P}P \,, \tag{9}$$

where $\partial A(T,0)/\partial P$ can be considered as temperature independent. Thus the product $\chi_0 T_C^2$ is pressure independent since, from (3) and (4),

$$\frac{\partial A(T,0)}{\partial P} = \frac{1}{2\chi_0^2}\;\frac{\partial \chi_0}{\partial P} + \frac{T^2}{2}\;\frac{\partial}{\partial P}\left(\frac{1}{\chi_0 T_C^2}\right) = \frac{\partial A(0,0)}{\partial P} \,. \tag{10}$$

Hence

$$\frac{\partial \log T_C}{\partial \log V} = -\frac{1}{2}\;\frac{\partial \log \chi_0}{\partial \log V} \,. \tag{11}$$

From the relation obtained from (3),

$$M^2(0,T,P) = -A(T,P)/B \,, \tag{12}$$

we deduce

$$\frac{\partial \log M(0,T,P)}{\partial \log V} = \frac{\frac{1}{2}\partial \log|-A(T,0)|}{\partial \log V} \,, \tag{13}$$

where V is the volume.

From the relation, which generalizes (3),

$$B = [2M^2(0,T,P)\,\chi(0,T,P)]^{-1} \tag{14}$$

and the constancy of B, it follows that

$$\frac{\partial \log \chi(0,T,0)}{\partial \log V} = \frac{-2\partial \log M(0,T,0)}{\partial \log V} \,. \tag{15}$$

In conclusion,

$$\frac{-\partial \log \chi_0}{\partial \log V} = \frac{2\partial \log M(0,0,0)}{\partial \log V} = \frac{2\partial \log T_C}{\partial \log V} = \frac{\partial \log|-A(0,0)|}{\partial \log V} \,. \tag{16}$$

For moderate pressures P the relative variations of $M(0,0,P)$ and $T_C(P)$ with pressure are identical. Their variation is equal to half of that of $\chi_0(P)$ but has the opposite sign. These predictions of thermodynamics and WFM theory are represented in Fig. 7.

The variation of the magnetization with pressure is related thermodynamically to the variation of the volume with applied field, through the relation

$$\left(\frac{\partial M(H,T,P)}{\partial P}\right)_{H,T} = -\left(\frac{\partial V(H,T,P)}{\partial H}\right)_{P,T} \,. \tag{17}$$

Introducing the elastic energy $V^2/2K$ in Eq. (7) and minimizing the free energy leads to

$$\Delta V_I(H,T) = -\tfrac{1}{2}K\left(\frac{\partial A(0,0)}{\partial V}\right)_{H,T} \times [M^2(H,T,0) - M^2(0,T,0)] \,, \tag{18}$$

where K is the compressibility.

The spontaneous magnetostriction, that is, the difference between the volume of the ferromagnetic sample and that of a (hypothetical) disordered one, is then

$$\Delta V_s(0,T) = -\tfrac{1}{2}K\left(\frac{\partial A(0,0)}{\partial V}\right)_{H,T} M^2(0,T,0) \,. \tag{19}$$

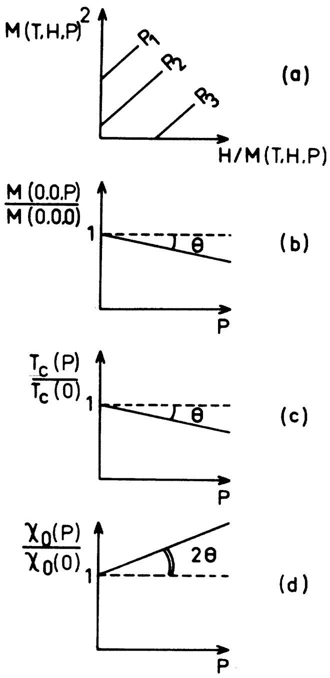

FIG. 7. Results of weak-ferromagnetism theory (pressure effects). (a) Arrott plot for constant temperature T, and various pressures P. (b) Pressure dependence of the spontaneous magnetization $M(0,0)$. (c) Pressure dependence of the Curie temperature T_C. (d) Pressure dependence of the susceptibility χ_0.

The above treatment is in agreement with that of Belov[14] and Wohlfarth.[15]

B. Experimental results

The high-pressure magnetization of the Ni-Pt alloys has been measured in the superconducting coil used for the measurements at zero pressure. The sample is located in a thick-walled beryllium-copper cell, connected through a capillary to a high-pressure intensifier. The pressure is applied to the sample at a temperature where helium is fluid. When low-temperature measurements are needed, the sample holder containing pressurized helium is cooled in a temperature gradient in such a way that the solidification of helium begins at the bottom of the cell. This happens at the sample level and is achieved in the capillary; the solidification is thus isobaric. Once helium is solid, the temperature is monitored to the desired value. During cooling the volume occupied by the solid helium can be considered as constant and thus the pressure is correctly determined from the *PVT* relations for solid helium. It is also determined from the value of the resistance of strain gauges located on the outside of the high-pressure beryllium-copper cell. The measurement of the magnetization is performed using an extraction technique. Corrections associated with the small negative magnetization of the beryllium-copper cell have been made.[11,12] Typical results obtained at 4.2 K are given in Figs. 8 and 9. The slopes of the Arrott plots do not depend on pressure, which confirms that B is independent of pressure.

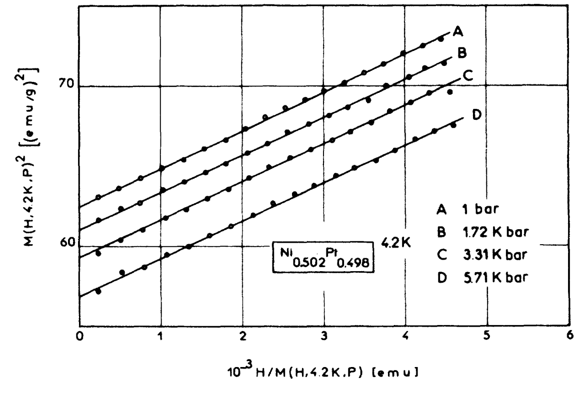

FIG. 8. Arrott plot for $Ni_{0.502}Pt_{0.498}$ at 4.2 K.

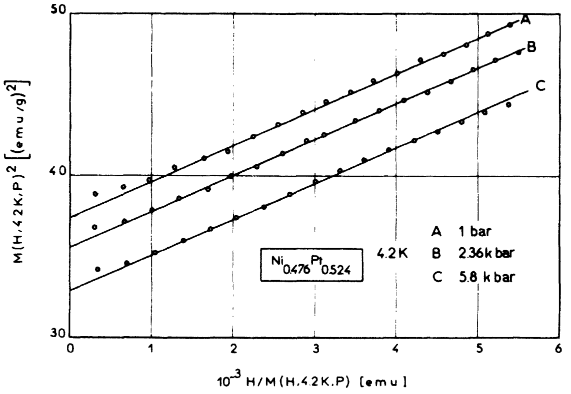

FIG. 9. Arrott plot for $Ni_{0.476}Pt_{0.524}$ at 4.2 K.

From the variation of $M^2(H,T,P)$ vs $H/M(H,T,P)$ determined at various temperatures and various pressures (Figs. 10 and 11) one can determine the variation of T_C, $M(0,0)$, and χ_0 with pressure (Figs. 12 and 13). As observed at ordinary pressures the intercept $I(T)$ of the Arrott line with the M^2 axis (Fig. 14) is a linear function of T^2, in good agreement with Stoner's law [Eq. (8)], although the behavior above T_C differs from that observed below T_C. The variation with pressure of the Curie temperature of $Ni_{0.476}Pt_{0.524}$ and $Ni_{0.498}Pt_{0.502}$ has not been determined due to the limited temperature range of the experimental setup. The data are summarized in Table II. The compressibility has been taken as a linear function of atomic concentration varying from 0.53×10^{-6} bar^{-1} for pure nickel, to 0.36×10^{-6} bar^{-1} for pure platinum. One observes (Figs. 12 and 13), in good agreement with Eqs. (10) and (16), that the relative variations of $M(0,0)$ and T_C with pressure are almost identical and have opposite sign and half the value of the relative variation of χ_0. In the experimental pressure range T_C, $M(0,0)$, and

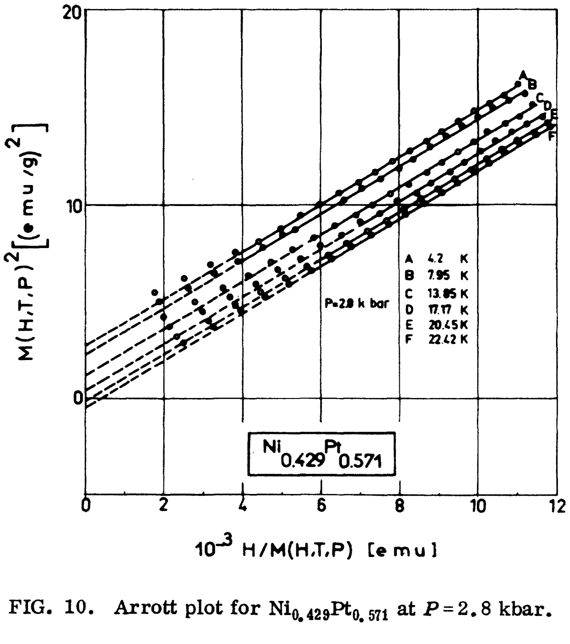

FIG. 10. Arrott plot for $Ni_{0.429}Pt_{0.571}$ at $P=2.8$ kbar.

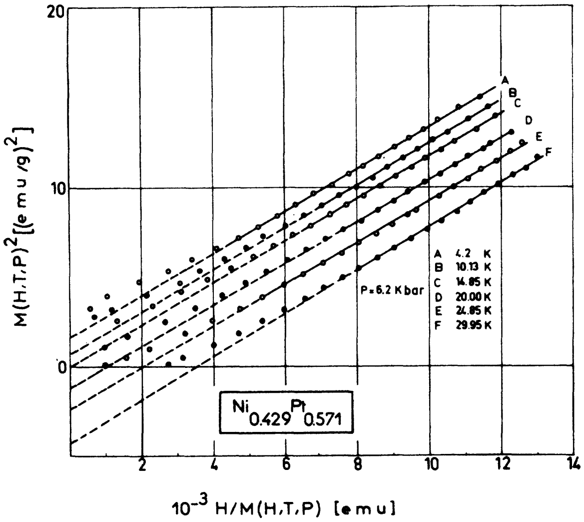

FIG. 11. Arrott plot for $Ni_{0.429}Pt_{0.571}$ at $P = 6.2$ kbar.

χ_0 can be considered as varying linearly with pressure.

C. Pressure variation of the critical concentration

In order to determine the pressure variation of the critical concentration c_F, one can examine the variation of $M(0,0)$, T_C or χ_0^{-1} with concentration. Taking $M^2(0,0)$, T_C^2, and χ_0^{-1} as proportional to $c - c_F$, as shown in Sec. II A, then

$$2\frac{\partial\log}{\partial\log V} = 2\frac{\partial\log T_C}{\partial\log V} = -\frac{\partial\log\chi_0}{\partial\log V}$$

$$= \frac{\partial\log(c-c_F)}{\partial\log V}\,. \tag{20}$$

Values of $\partial\log(c - c_F)/\partial\log V$ as a function of $c - c_F$ are given as a full line in Fig. 15. They have been calculated using the values $c_F = 0.42$ and $dc_F/dp = +1\times10^{-3}$ kbar^{-1}. They are in good agreement with the experimental values of

$$2\frac{\partial\log M(0,0)}{\partial\log V}\,,$$

$$2\frac{\partial\log T_C}{\partial\log V}\,,$$

and

$$\frac{-\partial\log\chi_0}{\partial\log V}\,.$$

These quantities are thus confirmed to be proportional to $(c - c_F)^{-1}$. As $M^2(0,0)$, T_C^2, and χ_0^{-1} are themselves proportional to $c - c_F$, it is then obvious that such quantities as

$$\frac{M^2(0,0)\,\partial\log M(0,0)}{\partial\log V}\,,$$

$$\frac{T_C^2\,\partial\log T_C}{\partial\log V}\,,$$

and

$$\frac{\chi_0^{-1}\partial\log\chi_0}{\partial\log V}$$

should be only slowly varying functions of c. This is in fact observed (Table II). From this table, in particular, the product $-T_C\partial T_C/\partial P$ is almost constant for the two alloys with 42.9 and 45.2-at.% Ni, having the respective values 34.5 and 37.9 kbar^{-1}. The corresponding value for $ZrZn_2$ is surprisingly close to this value, namely, 39.6 kbar^{-1}.[16]

D. Conclusion

(i) Plots of $M^2(H,T,P)$ against $H/M(H,T,P)$ at fixed temperatures are parallel straight lines over a wide range of pressures, which means that B is reasonably independent of pressure.

(ii) The relative variations of $M(0,0,P)$ and $T_C(P)$ with pressure are identical. Their variation is equal to half that of $\chi_0(P)$, but has the opposite sign.

(iii) The critical concentration $c_F = 0.42$ varies with pressure at a rate of $+1\times10^{-3}$ kbar^{-1}. This last value is sufficient to account for the pressure

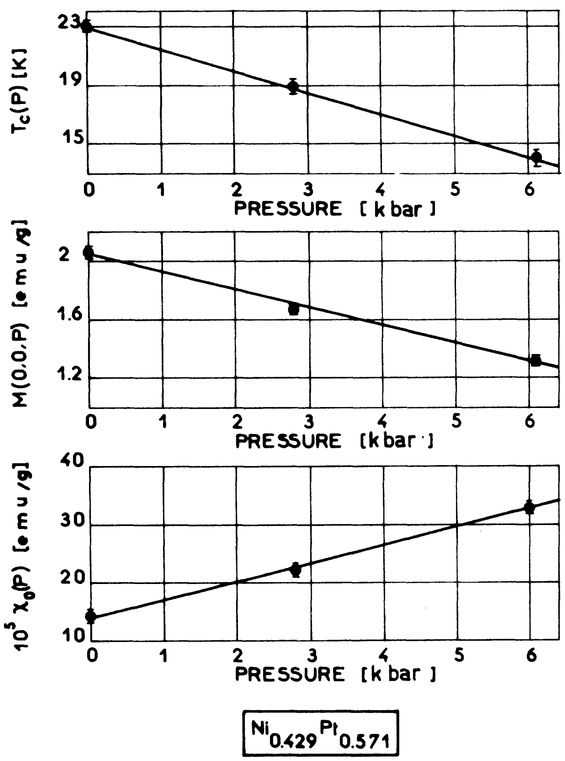

FIG. 12. Variation with pressure of the Curie temperature T_C, spontaneous magnetization $M(0,0)$, and initial susceptibility χ_0 of $Ni_{0.429}Pt_{0.571}$.

variation of $M(0, 0P)$, $T_C(P)$, and $\chi_0(P)$ in the alloying range under consideration (see Fig. 15).

IV. ANALYSIS OF EXPERIMENTAL RESULTS

The earlier discussions on the magnetic properties of Pt-Ni alloys near their critical concentration have led to the following statements.

(i) Below $c = c_F$, the disordered alloys appear to be exchange-enhanced paramagnets. Their magnetic susceptibility increases strongly as the critical composition is approached.[2,3] Contrary to most ordered-disordered magnetic systems, atomic order in Ni-Pt is associated with magnetic disorder.[17]

(ii) A simple uniform exchange enhancement has been proposed[18] to account for the variation of the Curie temperature with composition. According to this model, the short-range effective interatomic Coulomb repulsion $\overline{U}$ between d electrons of opposite spin is the same throughout the solid. This Coulomb interaction $\overline{U}$ was written $\overline{U}_{\mathrm{Pt}} + c\Delta$, where $\overline{U}_{\mathrm{Pt}}$ is the Coulomb interaction appropriate to platinum and c is the nickel concentration; Δ is the increase of the Coulomb interaction resulting from the nickel impurities. Such an assumption gave

$$T_C = T_{c,\mathrm{Ni}}\left(\frac{c - c_F}{1 - c_F}\right)^{1/2} ,$$

whereas a local model would lead to a Curie temperature T_C proportional to c.

(iii) From resistivity, specific heat, and magnetic measurements Gillespie, Mackliet, and Schindler[17] concluded that clustering in Pt-Ni alloys was small, in contrast, for instance, with Ni-Cu alloys near their critical concentration.

(iv) The homogeneity of Pt-Ni alloys has also been tentatively suggested by Besnus and Herr.[1]

In the present discussion we will follow as a first approximation a uniform enhancement model. We will neglect the rather small clustering effects which appear in the low-field magnetization data. The electronic structure of d electrons will be described by a single band. This approximation is strengthened by the fact that Pt and Ni are isoelectronic. We will assume that the effective Coulomb interaction U between electrons of opposite spins is identical on Ni and Pt sites in the alloys close to the critical concentration, which means that we consider a spatially averaged Coulomb interaction. The density of states $N(E)$ (per atom per spin) and its product with the effective Coulomb interactions $\overline{U} = UN(E_F)$ will thus be considered as a smooth function of concentration c.

A. Normal pressure susceptibility

The spin-orbit coupling contribution to the band structure and magnetic susceptibility of pure platinum is large.[19,20] However, ferromagnetic resonance experiments performed on Pt-Ni alloys show that these alloys have a g factor equal to 1.98 for $\mathrm{Ni}_{0.603}\mathrm{Pt}_{0.397}$ and 2.00 for $\mathrm{Ni}_{0.502}\mathrm{Pt}_{0.498}$.[5] The low-field susceptibility of the paramagnetic alloys with $c \simeq c_F(T)$ will then be the enhanced spin susceptibility

$$\chi(T) = 2NN(E_F)\mu_B^2 / [1 - \overline{U}(T)] , \qquad (21)$$

where the Stoner enhancement factor $1/[1 - \overline{U}(T)]$ results from the high density of states at the Fermi level $N(E_F)$. This density of states is defined per atom per spin. The Pauli susceptibility calculated from the bare band density of states is $2N[N(E_F)\mu_B^2]$; $\overline{U}$ depends on temperature. As its value is close to 1, its thermal variation will be the main reason for the variation of χ with temperature. When $c < c_F$, the alloy remains paramagnetic down to $T = 0$ K; we will call χ_0^p its value at 0 K.

The low-field susceptibility $\chi(T)$ is then related to the Landau parameter $A(T)$ through

$$\chi(T) = 1/A(T) , \qquad (22)$$

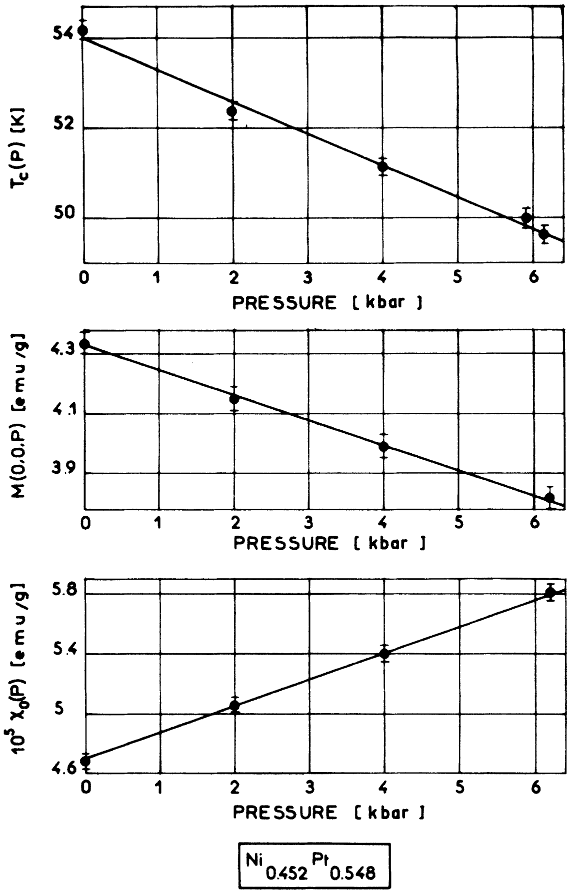

FIG. 13. Variation with pressure of the Curie temperature T_C, spontaneous magnetization $M(0, 0)$, and initial susceptibility χ_0 of $\mathrm{Ni}_{0.452}\mathrm{Pt}_{0.548}$.

since $M(H=0, T)=0$; $A(T)$ is positive for a paramagnetic alloy.

The $\bar{U}$ parameter increases with increasing nickel concentration; $c_F(0)$ corresponds to $\bar{U}(T=0)=1$. For values of $\bar{U}$ slightly larger than 1, corresponding to concentrations slightly greater than $c_F(0)$, the nickel-platinum alloys are weakly ferromagnetic. The susceptibility $\chi(0, T)$ in the WFM state is given by Eq. (3) and is equal to $-1/2A(T)$, where $A(T)<0$. Thus

$$\chi_0=\frac{N[N(E_F)\mu_B^2]}{\bar{U}-1} \tag{23}$$

is the zero field, zero temperature susceptibility of the WFM.

The experimental values of the susceptibilities χ_0^p and χ_0 do not allow a simultaneous determination of $N(E_F)$ and $\bar{U}$, and one needs other experimental information, in particular information derived from low-temperature specific-heat experiments. For a usual paramagnet at very low temperatures the major contribution to the specific heat is the electronic contribution $c=\gamma_0 T$, with

$$\gamma_0=\tfrac{2}{3}\pi^2k^2N[N(E_F)] . \tag{24}$$

Spin fluctuations can lead to an increase of the bare γ_0. The enhancement factor has a rather uncertain value, although much smaller than was first stated.[21-23] The experimental value γ increases from 6.5 mJ/mole K^2 for pure platinum to 14.61 mJ/mole K^2 for disordered $Ni_{0.435}Pt_{0.565}$. These values can be compared to 12.81 and 11.61 mJ/mole K^2 obtained for the $Ni_{0.50}Pt_{0.50}$ alloy in the disordered (ferromagnetic) and ordered (paramagnetic) states.[18]

As the susceptibility of the ordered (paramagnetic) phase is, at a first approximation, independent of concentration c, for $c\simeq c_F$, in the $0.3\leqslant c\leqslant 0.6$ range,[3] we will take $\gamma_0=11$ mJ/mole K^2 in order to have an estimate of $N(E_F)$ in the neighborhood of the critical concentration c_F. The value obtained is $N(E_F)\simeq 2.3$ states/eV atom spin. From χ_0 (Table I), one can deduce the values $\bar{U}=1.004$, 1.012, 1.020, and 1.033 for $c=0.429$, 0.452, 0.476, and 0.502. The value of U is therefore close to 0.44 eV. This value can be compared to 0.62 eV for Ni[24] and 0.35 eV for Zn_2Zr.[6] In the latter case, the density of states is close to 2.9 states/eV atom spin.

B. High-pressure behavior

As the variations with pressure of T_C, $M(0,0)$, and χ_0 are closely connected [Eq. (20)], then in order to analyze their pressure variation, it is sufficient to study that of χ_0. From Eq. (23) one deduces

$$\frac{d\log\chi_0}{d\log V}=\left(\frac{d\log N(E_F)}{d\log V}-\frac{\bar{U}}{1-\bar{U}}\,\frac{d\log\bar{U}}{d\log V}\right), \tag{25}$$

TABLE II. Derivatives for Pt-Ni alloys.

c	0.429	0.452	0.476	0.502
$dM(0,0)/dp$ (emu/mole kbar)	−16.5	−11	−8.6	−8.1
dT_C/dp (K/kbar)	−1.5	−0.7		
$10^5 d\chi_0/dp$ (emu/mole kbar)	420	23	7.4	4.2
$\frac{d\log M(0,0)}{d\log V}$	135	44	24	18
$\frac{d\log T_C}{d\log V}$	150	29		
$\frac{d\log\chi_0}{d\log V}$	−490	−84	−43	−39
$10^{-6}M^2(0,0)\frac{d\log M(0,0)}{d\log V}$ (emu/mole)	11	15	15	18
$10^{-4}T_C^2\frac{d\log T_C}{d\log V}$ (K)	8.0	8.6		
$10^{-4}\chi_0^{-1}\frac{d\log\chi_0}{d\log V}$ (emu)	−2.5	−1.3	−1.1	−1.6

which for a WFM, with $\bar{U} \simeq 1$, can be written

$$\frac{d\log\chi_0}{d\log V}=\frac{1}{1-\bar{U}}\,\frac{d\log\bar{U}}{d\log V}=-2\,\frac{d\log M(0,0)}{d\log V}$$

$$=-2\,\frac{d\log T_C}{d\log V}\;. \qquad (26)$$

Values of $d\log\bar{U}/d\log V$ obtained from $\bar{U}$ and the derivatives of χ_0, $M(0,0)$, and T_C are close to 1.1 for all the alloys under consideration. This value is nearly identical to 1.15, the value obtained for Zn_2Zr.[25]

Following Heine,[26] the width W of the d band depends only on the interatomic distance R at a rate given by

$$\frac{d\log W}{d\log R}=-5,$$

with

$$d\log R=\tfrac{1}{3}d\log V.$$

According to the uniform scaling approximation,[27] $WN(E)$ is independent of pressure. Kanamori[28] has given the following estimation for the effective Coulomb interaction:

$$U=\frac{U_b}{1+\gamma U_b/W}\,, \qquad (27)$$

where U_b is the bare Coulomb interaction and γ is a constant independent of pressure, according to the above approximation. Equation (27) is particularly well adapted to Pt-Ni alloys which have a small number of holes in their d band.

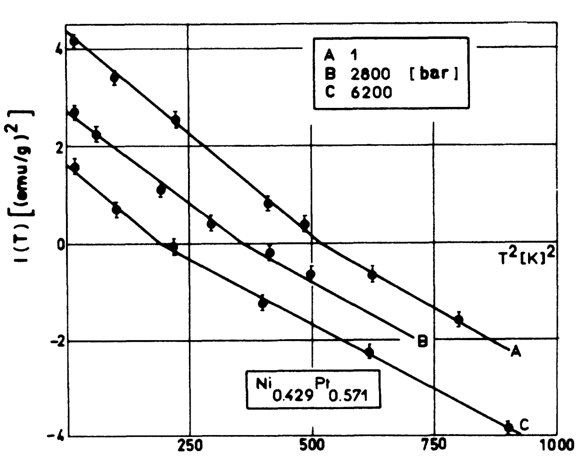

FIG. 14. Intercept $I(T)$ (see text) as a function of the square of the temperature at various pressures for $Ni_{0.429}Pt_{0.571}$.

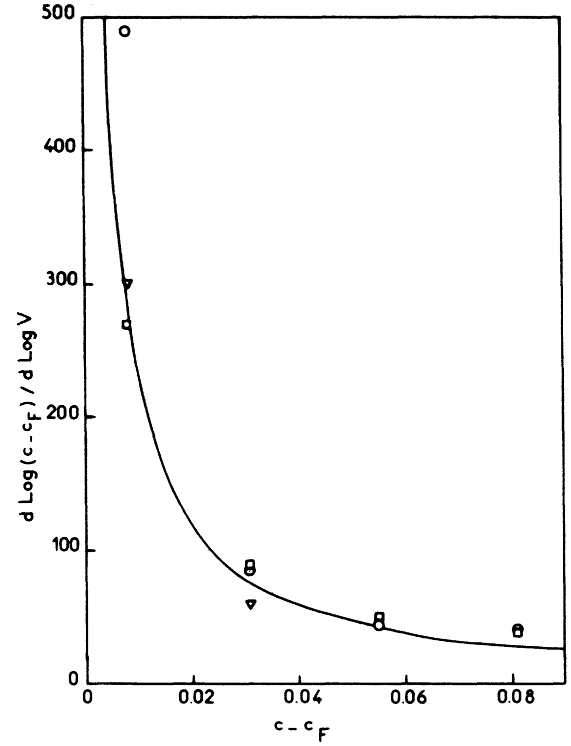

FIG. 15. Calculated values of $d\log(c-c_F)/d\log V$ (see text) and experimental $2d\log T_C/d\log V$, ∇; $2d\log M(0,0)/d\log V$, □; and $-d\log\chi_0/d\log V$, O; as a function of $c-c_F$.

Thus, for a constant number of d electrons, and U_b independent of pressure[29]

$$\frac{d\log\bar{U}}{d\log V}=\frac{5}{3}\left(\frac{U}{U_b}\right) \qquad (28)$$

and

$$\frac{d\log\chi_0}{d\log V}=\frac{5}{3}\left(\frac{U}{U_b}\right)\frac{1}{1-\bar{U}}\,. \qquad (29)$$

From $d\log\bar{U}/d\log V=1.1$ one deduces $U/U_b=0.65$ for the alloys under consideration. This value is, however, only indicative due to the difficulty in the determination of $\bar{U}$ from the specific heat and susceptibility experiments. Some physical quantities can, however, be determined independently of specific-heat experiments. One can thus obtain directly the values of U^2/U_b. From Eqs. (29) and (23), one obtains

$$\frac{d\log\chi_0}{d\log V}=-\frac{5}{3}\,\frac{U^2}{U_b}\,\frac{\chi_0}{N\bar{U}\mu_B^2}\,. \qquad (30)$$

For a WFM, $\bar{U}\simeq 1$, and

$$\frac{U^2}{U_b}=-\frac{3}{5}\,\frac{N\mu_B^2}{\chi_0}\,\frac{d\log\chi_0}{d\log V}\,. \qquad (31)$$

Experimental values of χ_0 and $d\log\chi_0/d\log V$ lead to $U^2/U_b\simeq 0.28$ eV, which is in self-consistent agreement with $U\simeq 0.44$ eV and $U/U_b\simeq 0.65$.

Another problem arises from the approximation according to which the number of d electrons is independent of pressure. In fact, s-d transfer occurs as indicated by a nonzero dependence of the $T=0$ magnetization of pure nickel, for instance, which is a strong ferromagnet.[27,30,31]

Along the critical line, at $T=0$ K, $\bar{U}=1$. The variation of $\bar{U}$ with concentration is compensated by the variation of $\bar{U}$ with pressure, so that

$$\frac{dc_F}{dp}=-\left(\frac{\partial\bar{U}}{\partial P}\right)_c\bigg/\left(\frac{\partial\bar{U}}{\partial c}\right)_p=K\left(\frac{\partial\log\bar{U}}{\partial\log V}\right)_c\bigg/\left(\frac{\partial\bar{U}}{\partial c}\right)_p\ , \tag{32}$$

with $K=4.3\times10^{-7}$ bar^{-1} and $\partial\log\bar{U}/\partial\log V=1.1$. The value $\partial\bar{U}/\partial c$ can be estimated to be 0.65, assuming a linear variation of $\bar{U}$ from 0.58 for Pt to 1.23[24] for Ni. From these values one obtains $dc_F/dp = +0.75\times10^{-3}$ kbar^{-1} in good agreement with the experimental value $dc_F/dp=+1\times10^{-3}$ kbar^{-1}.

C. Conclusion

The experimental results have been analyzed rather successfully using a thermodynamical treatment of weak ferromagnetism. The variation of the critical concentration with pressure arises from the variation with concentration and pressure of the averaged Coulomb interaction $\bar{U}$. Analysis of the experimental data, disregarding possible s-d transfer under pressure, leads to $U/U_b\simeq0.65$ with $U=0.44$ eV which corresponds to Coulomb interactions probably weaker in the alloys under consideration than in pure platinum, associated with a smaller bare Coulomb interaction. Such measurements on materials which have, according to the evidence, a fairly homogeneous magnetization, thus provide valuable evidence for the continuing applicability of the basic itinerant model as well as giving numerical estimates for some important parameters of this model.[32]

ACKNOWLEDGMENTS

We are grateful to C. Vettier for his help in carrying out these measurements and to J. Meyer and M. J. Besnus for valuable discussions. One of us (H. L. A.) wishes to thank D. Bloch for the hospitality with which he was received in the group "High Pressures" at the C.N.R.S. (Grenoble). He also acknowledges financial aid from the South African C.S.I.R. which made his stay at the C.N.R.S. possible.

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