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TECHNICAL REPORT

THE ORIGIN OF THE "INVAR" EFFECT

by

R. J. WEISS

MATERIALS RESEARCH LABORATORY
U. S. ARMY MATERIALS RESEARCH AGENCY
JUNE 1963

WATERTOWN 72, MAS

Electronic structures

Metals, transition elements

Crystal lattices

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R. J. Weiss

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ABSTRACT

The ideas of two electronic configurations of iron atoms in a f.c.c. lattice are extended to the iron-nickel alloy system. With a reasonable variation of the energy difference of these two configurations with nickel concentration it is shown that "invar" originates from the thermal excitation of the configuration with lower atomic volume in opposition to the normal anharmonic source of expansion. The anomalous pressure dependence of the Curie temperature and the variation of Curie temperature with concentration for alloys containing 30-60% Ni are also shown to follow.

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THE ORIGIN OF THE "INVAR" EFFECT

INTRODUCTION

In previous work on the thermodynamics of alpha (b.c.c.) and gamma (f.c.c.) iron (Tauer and Weiss (1961); Kaufman, Clougherty and Weiss (1963)) it was pointed out that gamma iron has two electronic structures (γ_1 and γ_2) that are separated by ~0.0355eV in energy. If either of these two forms could be completely retained its properties obtained from alloy extrapolation appear to be

TABLE I

The approximate values of lattice parameter, spin per atom, and Curie or Néel temperature of the two electronic structures of gamma iron obtained by extrapolation from alloy data (γ_1 - FeMn and FeN, γ_2 - FeNi, FePt and FePd).

	Crystal Structure	Lattice Parameter T = 290°K	Magnetic Structure	Spin Per Atom	Curie or Néel Temp.
γ_1 (lower level)	f.c.c.	$3.54\ A^{\circ}$	antiferro-magnetic	$\sim 0.5\ \mu_B$	~80°K
γ_2 (upper level)	f.c.c.	$3.64\ A^{\circ}$	ferro-magnetic	$\sim 2.8\ \mu_B$	~1800°K

In pure γ iron the antiferromagnetic (low volume) state is the ground state (T = 0°K) although the upper level can be excited thermally. Since the volume change between the two is only ~8% γ_1 does not convert to γ_2 as a first order transition but rather as an internal electronic excitation. It can be visualized as a shift of one electron from one half of the 3d band to the other half. Such a reversal in spin would account for a difference of 2.0 μ_B while the observed difference of ~2.3 μ_B per atom (Table I) may be associated with a small additional orbital contribution or differences in exchange polarization of the bands. The thermodynamics of the transition has been treated as a Schottky effect (Kaufman, Clougherty and Weiss (1963)) although a more sophisticated treatment will be required to account for details of the changes in the energy bands with temperature.

The basic ideas of this note are:

1. That when iron is alloyed with sufficient nickel, palladium, or platinum the order of the levels can be reversed with the ferromagnetic γ_2 (high volume) level being stabilized (T = 0°K).

2. That in the cases where the γ_2 level is stabilized thermal excitation of the γ_1 level decreases the atomic volume in *opposition* to the normal anharmonic source of expansion and depending on the energy difference of the two levels can yield an 'apparent' expansion coefficient that is markedly decreased or even zero (invar effect).

3. That in iron-rich alloys in which the γ_1 level is lower thermal excitation of the γ_2 level increases the lattice parameter. When this increase is added to the normal anharmonic source of expansion it gives rise to an anomalously high 'apparent' coefficient of expansion.

4. That in the ferromagnetic alloys (γ_2 lower) thermal excitation of the γ_1 level (antiferromagnetic) leads to a marked decrease in the Curie temperature because of the reversed sign of the exchange interaction of the γ_1 state.

5. Finally, that the application of pressure in the ferromagnetic alloys tends to stabilize the γ_1 (low volume) state and this can cause an anomalously large reduction in the Curie temperature under pressure.

We wish to show that with a reasonable variation of the energy separation of the two levels with concentration one can obtain quantitative agreement in the Fe-Ni alloy system with

1. The measured temperature dependence of the lattice parameter;
2. The variation of the Curie temperature with concentration;
3. The variation of the form of the magnetization curves with concentration;
4. The variation of the saturation magnetization (T = 0°K) with concentration, and
5. The variation with concentration of the pressure dependence of the Curie temperature.

The suggested variation of ΔE, the energy separation of the two levels in the Fe-Ni system, is shown in Figure 1. The value of -0.0355 eV for pure gamma iron was determined from the PVT curve for the alpha-gamma transition in iron (Kaufman, Clougherty and Weiss (1963)) while the concentration of nickel for which ΔE passes through zero and becomes positive is derived from the saturation magnetization curves of Crangle and Hallam (1963) and Kondorski and Fedotov (1952) Figure 2, as well as the hyperfine field of the f.c.c. Fe-Ni alloys determined by Mössbauer measurements (Johnson, Ridout, and Cranshaw (1963)), Figure 3. At about 29 atomic per cent nickel there is a sudden loss of ferromagnetism since the antiferromagnetic γ_1 level is stabilized below 29% (T = 0°K). The remainder of the ΔE curve is made consistent with the calculations that follow although it is at least quite reasonable to expect the curve to level off at high nickel concentrations (>85%) where the iron atoms are no longer nearest neighbors.

1. *Calculation of the temperature dependence of the lattice parameter:*

The lattice parameter of the Fe-Ni f.c.c. alloys is calculated assuming Vegard's law to hold i.e. the volume occupied by each nickel atom and the γ_1 and γ_2 iron atoms are identical in the alloy as in the pure element. For nickel this corresponds to a room temperature lattice parameter of 3.517 A°. The calculation follows the procedure in Kaufman, Clougherty and Weiss (1963) with one additional consideration. In treating a Schottky two level excitation the ratio of the degeneracy of the two levels must be included. While the degeneracy of the two levels, γ_1 and γ_2, are approximately given by their values of 2J + 1 (J = spin) in a completely disordered spin system ($T >> T_c$), the degeneracy of the levels is identical in a ferromagnet below the Curie temperature and to a certain extent in the region above the Curie temperature where short range order predominates. In pure gamma iron the ratio of the degeneracy of the upper level to the lower level (for $T >> T_c$) was determined to be 1.79 from a knowledge of the entropy difference between alpha and gamma iron. While the calculated ratio is ~1.9 (i.e. the lower level must have at least a degeneracy of two since the spins can point up or down) if the orbital moment is completely quenched ($J = S$), some orbital contribution can account for the difference. We shall thus use 1.79 for the ratio of the degeneracies below 29 atomic per cent nickel since calculations will be confined to $T >> T_c$ and unity above 29% nickel since calculations above 29% will be made below or not too far above the Curie temperature. The lattice parameter for X atomic fraction of nickel and for $T \geq 290°K$ is thus:

$$a_o = 3.54\left(1 - 0.0075X + \frac{0.0293(1 - X)}{1 + g/1.79}\right)\left(1 + \alpha(T - 290)\right);\ X<0.29 \qquad (1)$$

$$a_o = 3.64\left(1 - 0.0339X - \frac{0.0269(1 - X)}{1 + g}\right)\left(1 + \alpha(T - 290)\right);\ X>0.29 \qquad (2)$$

$$g = \exp(\Delta E/kT) \qquad \begin{array}{l} a = \text{coefficient of linear expansion} \\ \cong 14 \times 10^{-6}(1 + 6 \times 10^{-4}(T - 290)) \end{array}$$

Figure 4 shows the calculated and observed lattice parameters for pure gamma iron at T = 290°K (see Kaufman, Clougherty, and Weiss (1962)) and for 23.4, 34.3, 57.0, 72.8 and 100% nickel as a function of T. The "invar" effect i.e. a zero 'apparent' coefficient of expansion is seen near room temperature in the 34.3 per cent alloy. The 'apparent' average coefficient of expansion of ~25 x 10^{-6}/deg. for the 23.4% alloy over the temperature range 300-400°K is anomalously high.

Considering that we assumed Vegard's law to hold and that the coefficient of expansion was taken to be that of pure nickel over the entire alloy range the agreement appears quite good. It is also clear that the invar effect has little to do with effects at the Curie temperature as shown by the smooth variation of the lattice parameter through the Curie temperature, denoted T_c, in the 34.3, 57.0, 72.8, and 100% alloys in Figure 4. (In the 34.3% alloy the minimum coefficient of expansion occurs some 200 degrees below T_c.)

2. *Calculation of the Curie temperature of the f.c.c. Fe-Ni alloys.*

In order to calculate the Curie temperatures of the f.c.c. alloys we assume that the spin on each nickel atom is 0.6 μ_B for all concentrations and that the spins of the γ_1 and γ_2 states of the iron atom are given as in Table I. We employ the semi-empirical expression (Tauer and Weiss (1955)) for the Curie temperature of transition metals and alloys

$$T_c \cong 113.5|Z_\uparrow - Z_\downarrow|\ell n(2s + 1)$$

where $Z_\uparrow$ is the number of nearest neighbors with spin aligned in the direction favored by the sign of the exchange integral and $Z_\downarrow$ is the number whose spin is opposed. Table II shows that equation 3 gives reasonably good values of T_c over a wide range of values of 2s and Z particularly for the ferromagnets.

TABLE II

The calculated (equation 3) and observed Curie temperature of various transition elements and alloys. The values of $\overline{2s}$ are determined from magnetization measurements for the ferromagnetics and from neutron diffraction for the antiferromagnetics

Substance		$\lvert Z_\uparrow - Z_\downarrow \rvert$	2s	T_c(eqn.3)°K	T_c(observed)
Ni	F	12	0.6	640	636
$Ni_{0.5}Cu_{0.5}$	F	12	0.1	129	120
Co	F	12	1.73	1368	1400
$Ni_{0.5}Co_{0.5}$	F	12	1.17	1055	1095
αFe	F	8	2.22	1061	1040
$Fe_{0.5}Cr_{0.5}$	F	8	0.9	582	580
γ_2Fe	F	12	2.8	1820	~1800
γ_1Fe	AF	8-4=4	0.5	153	~ 80
γMn	AF	8-4=4	2.55	575	~ 550
Cr	AF	8	0.4	306	312

In the case of alloys we use average values for 2s and $\lvert Z_\uparrow - Z_\downarrow \rvert$.

For the iron-nickel series the pertinent parameters of equation 3 are

$$\overline{2s} = \left[0.6X + \frac{2.8(1 - X)g}{1 + g} + \frac{0.5(1 - X)}{1 + g}\right] \tag{4}$$

$$\lvert \overline{Z_\uparrow - Z_\downarrow} \rvert = \left[12\left(1 - \frac{2(1 - X)}{1 + g}\right)\left(1 - \frac{1 - X}{1 + g}\right)\right] \tag{5}$$

evaluated at $T = T_c$

which yields the curve in Figure 5, evidencing good agreement with the data of Crangle and Hallam (1963).

3. *The variation in the form of the magnetization curve with concentration.*

While pure nickel shows a magnetization curve which follows more or less the standard 'Brillouin function' the curves steadily depart from this shape as iron is added to nickel The basic reason is that the excitation of the low spin 'antiferromagnetic' γ_1 level reduces $\overline{2s}$ more than due to thermal misalignment of spins. Assuming the pure nickel magnetization curve to account for the thermal misalignment of spins, equation 4 yields an additional factor reducing the magnetization curve when taken relative to the value of equation 4 at T = 0°K.

This is shown in Figure 6 for the 32.3%, 50% and 100% nickel alloys evidencing good agreement with the data of Crangle and Hallam (1963).

4. *The variation of the saturation magnetization with composition $T = 0°K$.*

This is merely given by equation 4 at T = 0°K ($g = \infty$) i.e.

$$\overline{2s} = 0.6X + 2.8(1 - X);\ X > 0.29 \qquad (6)$$

and is plotted as the solid line in Figure 2.

5. *The variation with composition of the pressure dependence of the Curie temperature.*

The application of pressure introduces a $P\Delta V$ term in the energy separation of the γ_1 and γ_2 levels favoring the γ_1 (low volume) level. Thus in the ferromagnetic region $x > 0.29$ the application of pressure reduces the magnitude of ΔE by 0.0082 eV per 10 k bars since the value of ΔV is known (Table I). Substituting the $P\Delta V$ term into equations 3, 4, and 5 (i.e. $g = \exp[(\Delta E + P\Delta V)/kT]$) we find the pressure dependence of the 36% and 68% alloys as given in Figure 7. A small correction (+ 0.8% per 10 k bars) was applied to these alloys since there is an intrinsic pressure dependence of + 0.8% per 10 k bars for pure nickel. The agreement with the data of Patrick (1954) is quite good.

SUMMARY

In summary, we believe that the basic idea of Tauer and Weiss (1961) and Kaufman, Clougherty and Weiss (1963) that the iron atom in a f.c.c. lattice can exist in two different electronic structures closely spaced in energy, provides a good working hypothesis to the unusual properties of gamma iron and its alloys with Ni, Pd and Pt. While the "invar" effect has been observed for Fe-Pt alloys and should, we believe, also be observed in f.c.c. FePd alloys insufficient data preclude any calculations.

A possible method of studying some of the details of the $\gamma_1 - \gamma_2$ transition is to use inelastic scattering of neutrons. Since the transition is from antiferromagnetic to ferromagnetic coupling it may yield some insight as to the reason for the change in sign of the exchange integral.

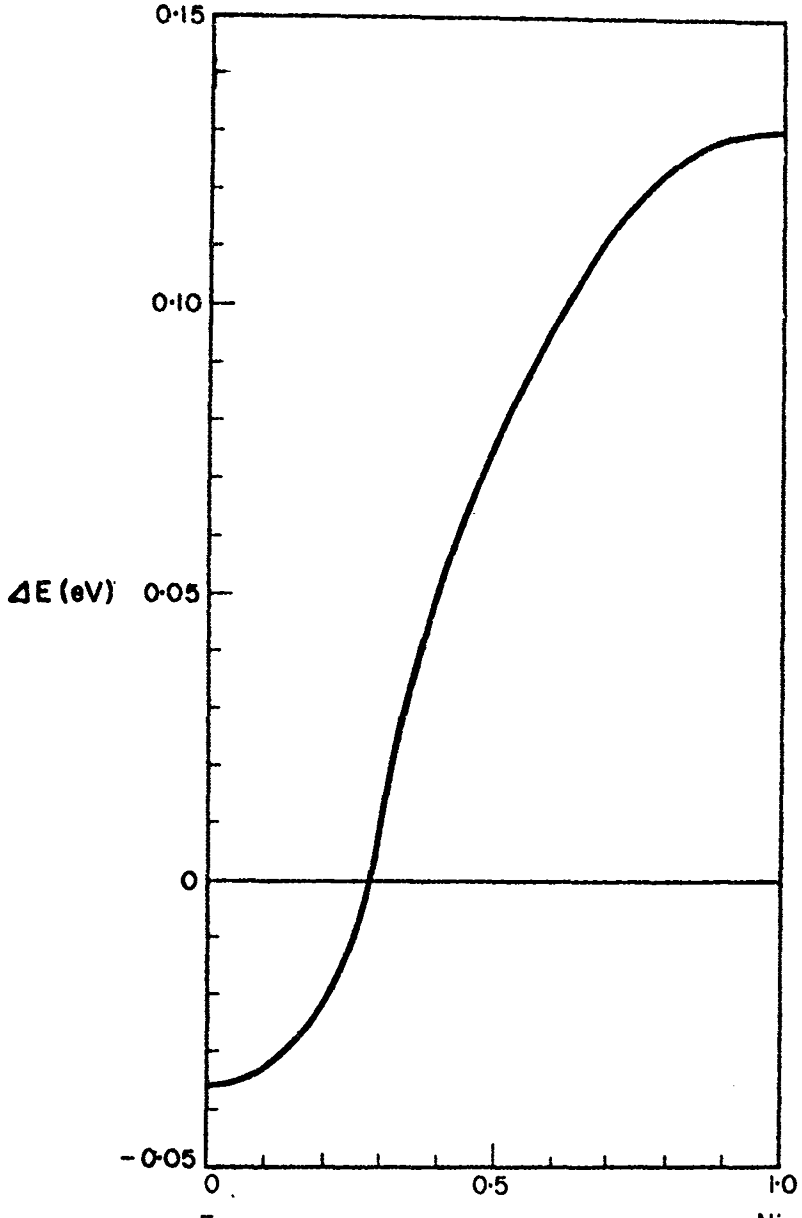

Figure 1. The energy separation in eV of the γ_1 and γ_2 levels of the iron atom in a f.c.c. lattice as a function of nickel concentration. Negative values of ΔE indicate the γ_1 level as the lower level (see Table I).

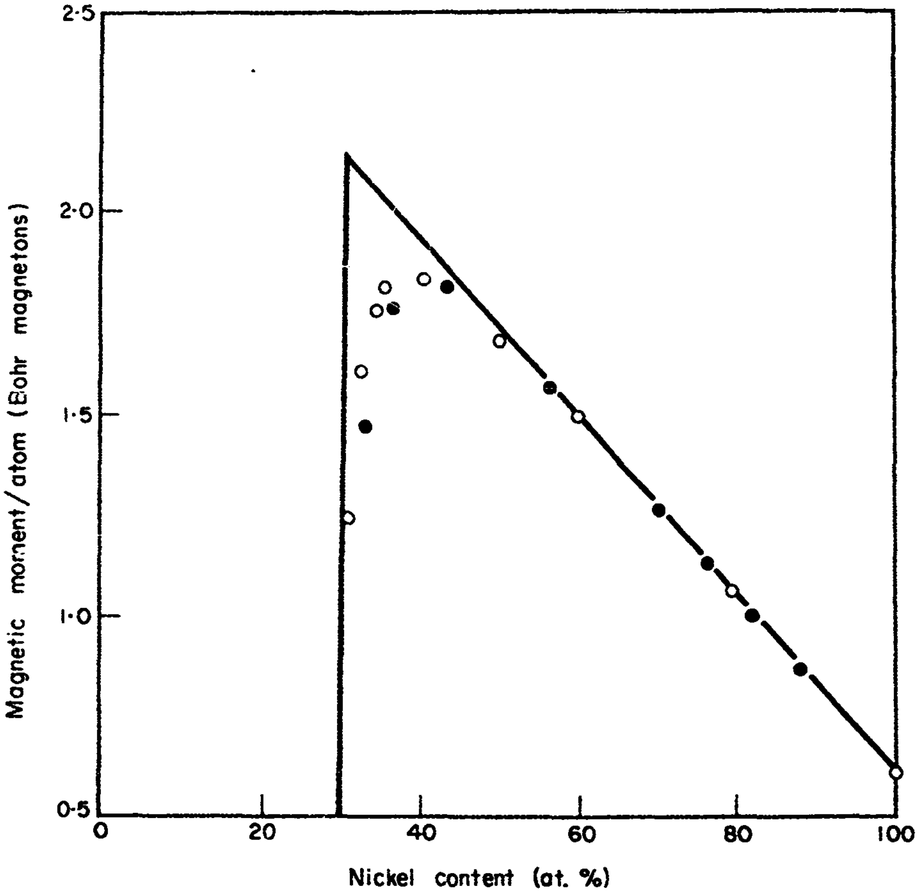

Figure 2. The magnetic moment per atom at T = 0°K versus concentration for the ferromagnetic f.c.c. iron-nickel alloys. The sudden decrease in the vicinity of 29 atomic per cent nickel is due to the transition from ferromagnetism to antiferromagnetism.

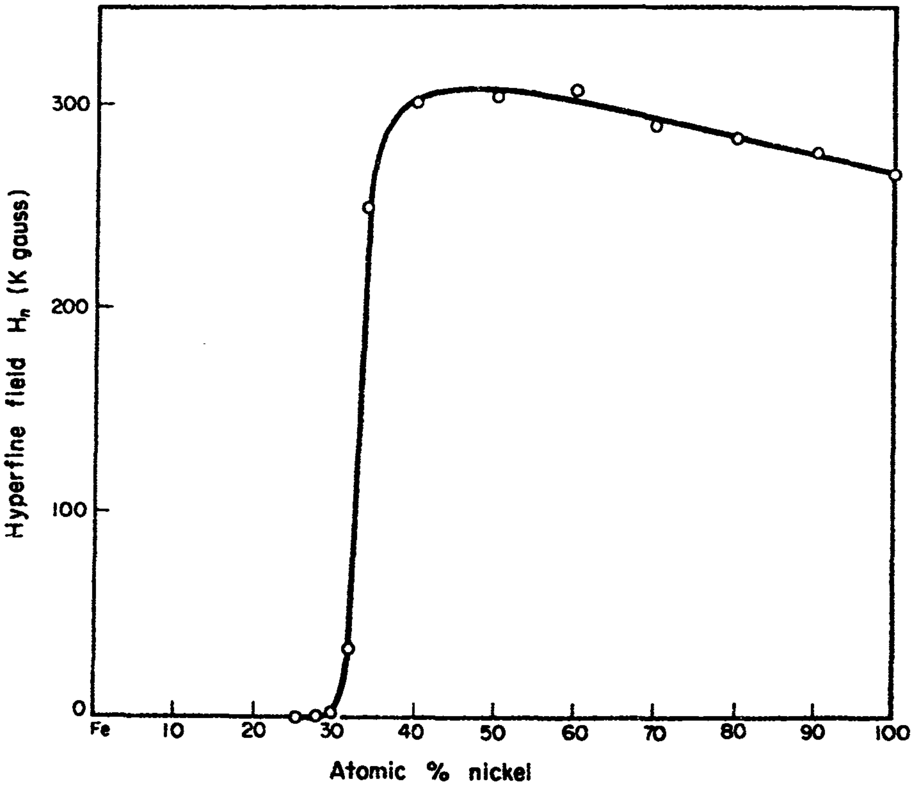

Figure 3. Mössbauer hyperfine field in kilogauss versus concentration for the ferromagnetic f.c.c. iron-nickel alloys. The sudden decrease in the vicinity of 29 atomic per cent nickel is due to the transition from ferromagnetism to antiferromagnetism.

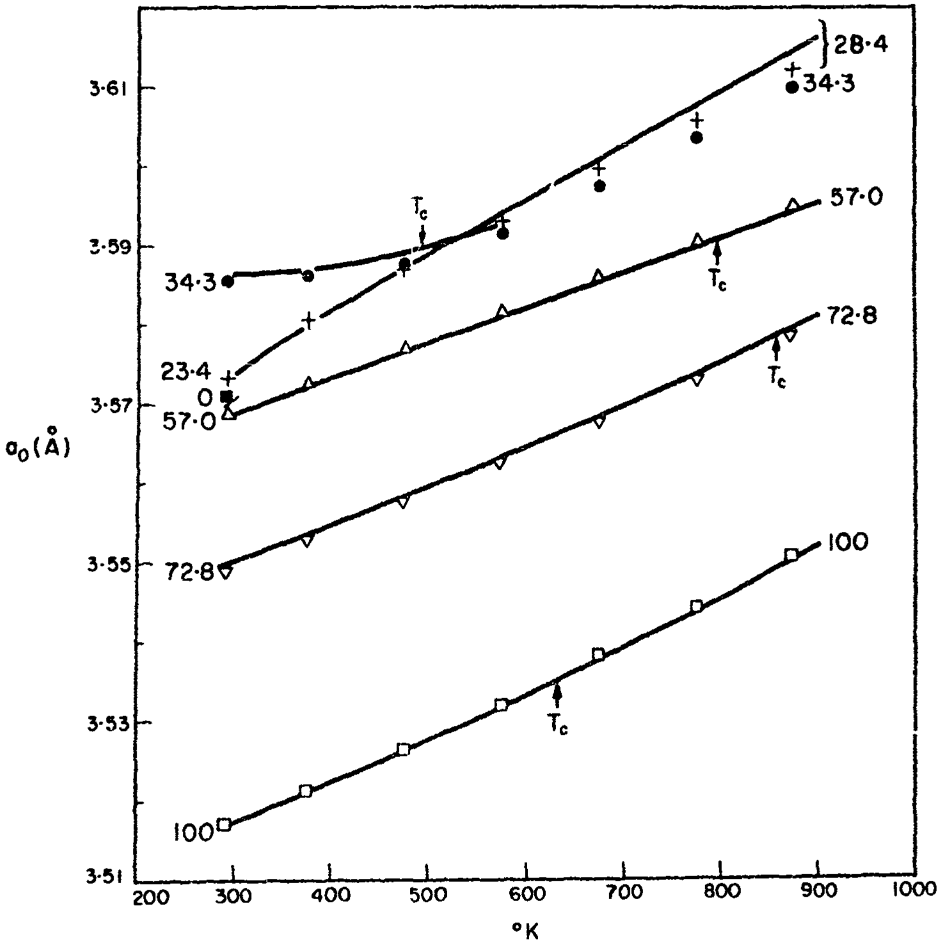

Figure 4. The lattice parameters in A° of the 23.4, 34.3, 57.0, 72.8 and 100 at % nickel alloys as a function of temperature in °K calculated from equations (1) and (2) and Figure 1. The observed lattice parameters (Pearson (1958)) are shown as well as the ferromagnetic Curie temperatures. The calculated and observed (Pearson (1958)) lattice parameter at 290°K for pure gamma iron are also given. The 'invar effect' i.e. ~zero coefficient of expansion is seen to occur near room temperature for the 34.3 at % alloy.

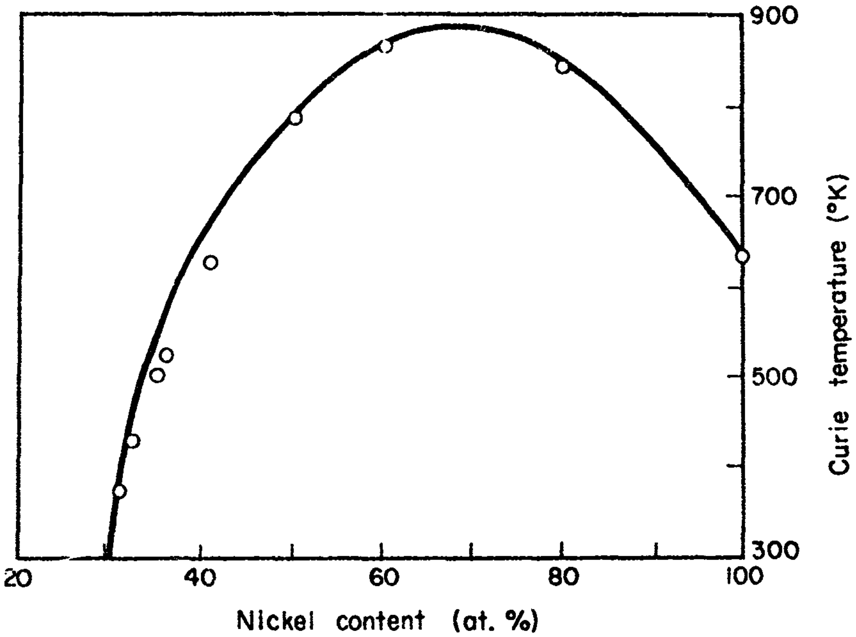

Figure 5. The calculated and observed values of the ferromagnetic Curie temperature versus concentration for the f.c.c. iron-nickel alloys. The rapid fall near 29% nickel is due to the thermal excitation of the 'antiferromagnetic' γ_1 level in these alloys.

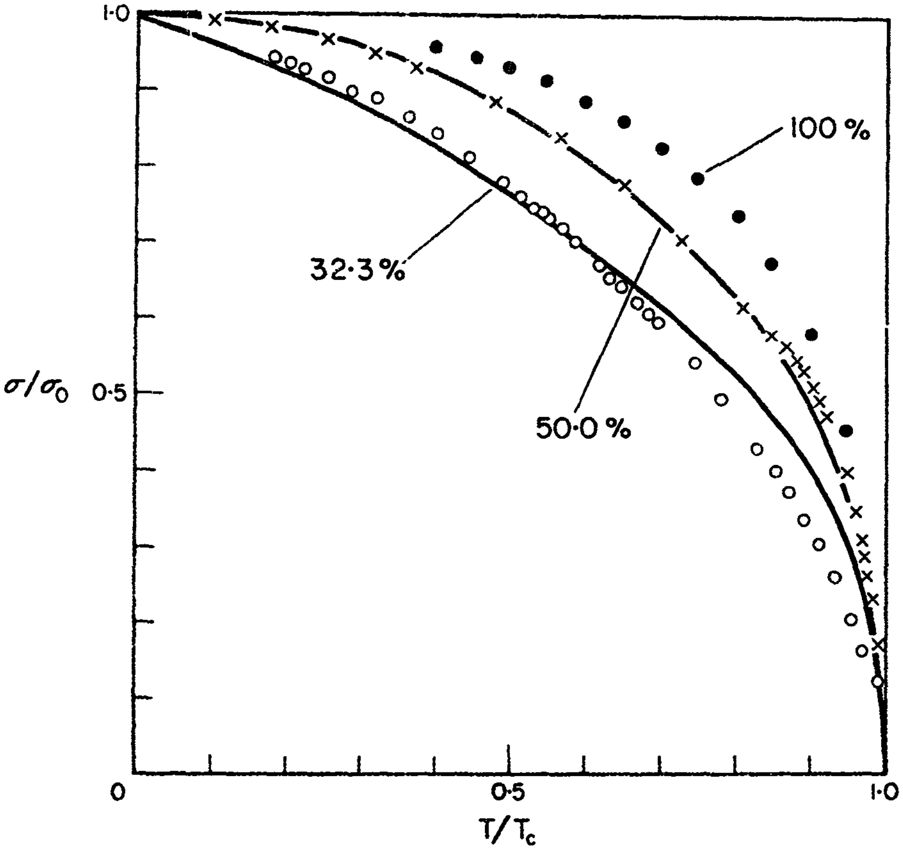

Figure 6. The calculated and observed magnetization curves versus reduced temperature for the 32.2 and 50.0 at % f.c.c. ferromagnetic iron-nickel evidencing appreciable departure from the 'standard Brillouin curve' of pure nickel (100 at %).

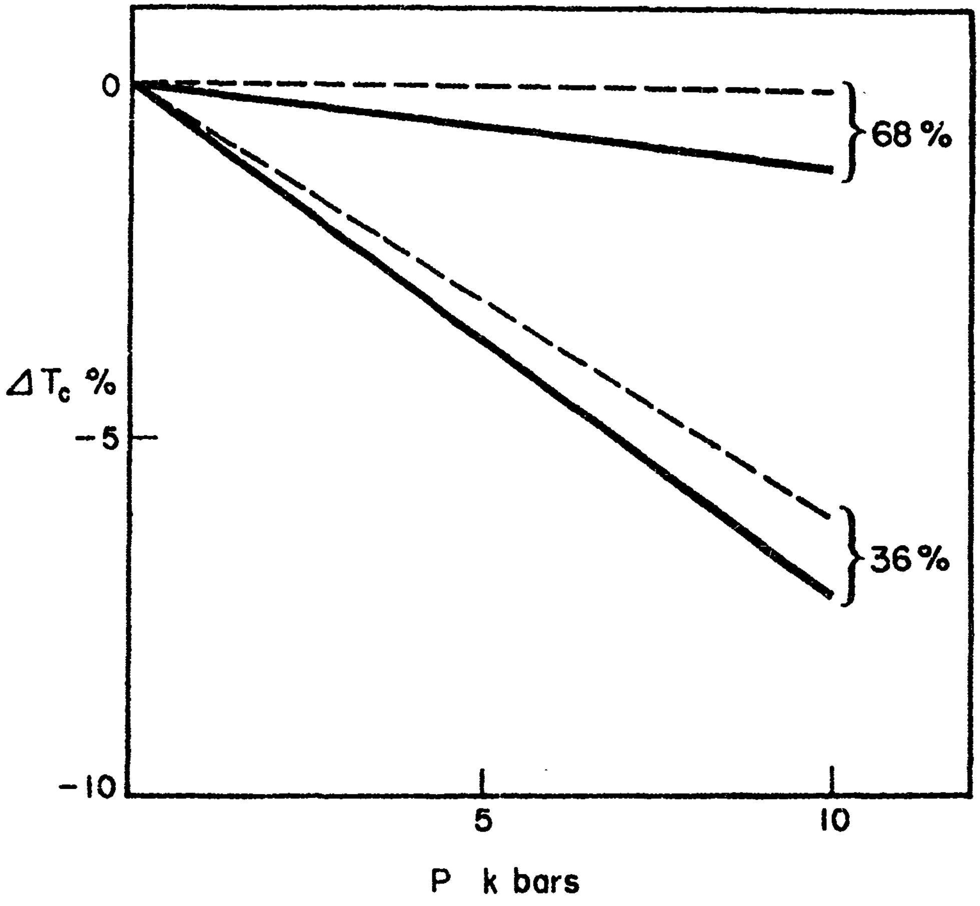

Figure 7. The percentage reduction in the ferromagnetic Curie temperature of the 36 and 68 at % iron-nickel alloys as a function of pressure. The large reduction in the 36 at % Ni alloys is due to the pressure tending to stabilize the 'antiferromagnetic' low volume γ_1 level.

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