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FERROMAGNETIC PROPERTIES OF Fe AND Ni IN RELATION TO THEIR BAND STRUCTURE

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Résumé. — De nombreuses propriétés de Fe et Ni peuvent être reliées les unes aux autres au moyen d'une théorie empirique supposant que le moment atomique magnétique n'est pas constant dans les métaux de transition mais est supposé dépendre dans ces cas de la température, du volume et du champ magnétique externe.

Les propriétés en question sont, parmi d'autres, la magnétostriction en volume dépendant du champ, la variation de magnétisation et du point de Curie avec la pression et les déviations par rapport aux courbes de Brillouin de la magnétisation à saturation en fonction de la température.

Les variations du moment atomique magnétique sont estimées en incorporant les moments magnétiques localisés dans la théorie des bandes pour les électrons 3d.

Abstract. — Several properties of the elements Fe and Ni can be correlated with each other by means of an empirical theory, which assumes that the atomic magnetic moment does not have a constant value in transition metals, but is thought to depend in these cases on temperature, volume, and external magnetic field.

The properties in question are, among others, the field-dependent volume magnetostriction, the change of magnetization and Curie point with pressure, and the deviations of the temperature dependence of the saturation magnetization from the appropriate Brillouin curves.

Variations of the atomic magnetic moment are estimated by incorporating localized magnetic moments into the electron band theory of the 3d-electrons.

Some of the physical properties of iron metal display unexplained anomalies. We found [1] that the field-dependent volume magnetostriction, h'_0 , does not disappear at low temperatures, but remains approximately constant. Moreover, the temperature dependence of the saturation magnetization deviates more than 10 % from the theoretically expected Brillouin curve [2], while the value for the exchange energy as found from spin wave theory, differs considerably from the value, calculated from the experimentally determined Curie point [3]. In other ferromagnetics these properties give a better fit with theory.

These difficulties can be removed by considering the localized atomic magnetic moment, σ , of the atoms in a magnetic metal as a variable. This is in contradistinction to the moments in magnetic salts, which have fixed values that have been calculated. We assume that σ is a function of tempe-

rate, volume, etc. For a certain value of σ , the energy will be a minimum; we assume a quadratic dependence of the energy upon σ in the neighbourhood of this equilibrium value.

If we define σ as the magnetic moment per atom expressed in Bohr magnetons, and ω as a relative change in volume of the lattice, then the terms of the free energy per atom, F , which depend on σ or ω , can be written as :

$$F = \frac{1}{2} A(\sigma - \sigma_p)^2 - B(\sigma - \sigma_p) \omega + \frac{1}{2} (k/N) \omega^2 - \frac{1}{4} J^*(1 - \xi\omega) \gamma\sigma^2 - (\mu_0/N) M \cdot H + (1/N) P\omega + \text{entropy term.} \quad (1)$$

The effects of lattice vibrations, causing the thermal expansion are omitted here. The first term gives the quadratic dependence of F on σ , which we have already mentioned; the second

term is a cross-term, necessary for understanding the observed dependence of magnetization on volume. The third term gives purely elastic energy; k is the modulus of compression and N is the number of atoms per unit volume. The fourth term denotes the ferromagnetic exchange energy; γ is a parameter describing the spin-ordering on neighbouring atoms; $\gamma\sigma^2 = \overline{\sigma_1 \cdot \sigma_2}$. The factor, $1 - \xi\omega$, expresses the dependence of the exchange energy on volume to the first order. The two following terms denote the effect of an external magnetic field, H , and of an external pressure, P . At saturation at 0 °K, $|M| = (N\hbar/2m)\sigma$. The last term denotes effects connected with entropy changes and is not written out; we will consider here only those cases in which the dependence on σ and ω of this term is relatively unimportant, i.e., $T = 0$ °K and $T \gg T_c$. By differentiating equation (1) with respect to σ and ω it is now possible to calculate variations of these quantities provided that the various constants are known. As an example we present here the calculation of the variation of σ and ω in the neighbourhood of the Curie point. Far above this point, in magnetic field zero and pressure zero, we find: $\sigma = \sigma_p$ and $\omega = 0$. σ_p is therefore the moment in a certain state, high enough above the Curie point that no spin ordering exists, but at a temperature low enough that the derivatives of the entropy term can be neglected. At 0 °K, we, however, find:

$$\sigma = \sigma_0 = \frac{A}{A - \frac{1}{2}J^*} \sigma_p,$$

$$\text{and } \omega = \omega_0 = 3h_0 = \frac{\frac{1}{2}NJ^*\sigma_0}{k} \left(\frac{B}{A} - \frac{1}{2}\sigma_0\xi \right),$$

if some small approximations are made ($\xi\omega \ll 1$, $B^2 \ll Ak/N$, which are found to be allowed by the experimental data).

The change in volume near the Curie point has been known for a long time; it is the spontaneous volume magnetostriction. The small change in atomic magnetic moment near the Curie point, has not been discussed before as far as we know, it can however, explain the deviation in the curve of saturation magnetization vs. temperature, as is shown in figure 1.

It is supposed here that the Curie point, T_c , depends on σ , according to: $T_c \sim J^*\sigma(\sigma + 2)$. We admit that the Brillouin curves, which are derived with the molecular field theory, are a bad approximation even if σ is constant, and that the method of P. R. Weiss [4] should give better results. No one has, however, done calculations on this.

At low temperatures the decrease of M_s is best described with the spin wave theory. For a

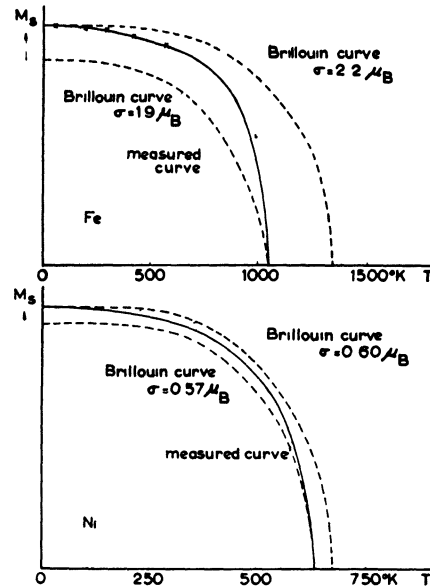


Fig. 1. — Saturation magnetization of iron and nickel as a function of temperature. The measured curve can be understood if it holds true that the magnetic moment per atom decreases with temperature.

b. c. c. lattice with spins 1, as in the case with iron, it has been calculated that [3]:

$$M_s(T) = M_s(0) \{ 1 - 0.178 (T/T_c)^{3/2} - \dots \}.$$

If the Curie point of 1 380 °K is taken, which value can be read from figure 1, for the case that σ would stay equal to σ_0 at high temperatures, this equation is in good agreement with the experimentally found values. This agreement was poor with the real Curie point of 1 043 °K.

In the following table, the values used for the different constants are given, together with six different quantities which can now easily be calculated by differentiating equation (1) with respect to σ and ω .

A consequence of our hypothesis is now, that $\partial\sigma/\partial H$, $\partial\sigma/\partial P$ and h'_0 are not necessarily strongly temperature dependent at low temperatures. h'_0 does not need to tend to zero at 0 °K, which is indeed observed by us [1]. The agreement between theory and experiment is rather good, since many approximations have been made, in particular that the various constants in equation (1) are thought to be temperature independent. Only two constants, B and ξ , are fitted to the experimental values.

The constant A can be connected with the density of states of the 3d-electron bands. We suppose that a force exists in atoms situated in a solid, which tries to make the resulting spin moment as large as the Pauli principle will permit. This corresponds to Hund's rule, which is valid for free atoms; it can be expressed very roughly by a

TABLE

	Fe	Ni		Fe	Ni
A	0.38 eV	1.7 eV	J^*	0.144 eV	0.240 eV
B	0.43 eV (*)	— 0.14 eV (*)	ξ	1.15 (*)	0.6 (*)
$k \left\{ \begin{array}{l} 300^\circ\text{K} \\ T_c \end{array} \right.$	12.5 eV	13.0 eV	N	$8.5 \times 10^{28}/\text{m}^3$	$9.15 \times 10^{28}/\text{m}^3$
\bar{N}	6.5 eV	12.3 eV	σ_0	2.2	0.6

(*) Best fit to observed data below.

Numerical values of constants, used in equation (1).

	Fe _{calc}	Fe _{obs}	Ni _{calc}	Ni _{obs}	
$(\sigma_0 - \sigma_p)/\sigma_p$	0.49	agrees	0.07	agrees	—
h_0 near T_c	2.6×10^{-4}	2.6×10^{-4}	-4.7×10^{-4}	-3.7×10^{-4}	—
$\partial\sigma/\partial H$	2×10^{-8}	—	0.4×10^{-8}	—	oe ⁻¹
$1/3 \partial\omega/\partial H = h'_0$	1.7×10^{-10}	1.8×10^{-10}	-0.2×10^{-10}	$0 \pm 0.3 \times 10^{-10}$	oe ⁻¹
$\partial\sigma/\partial P$	-5.0×10^{-7}	-6.2×10^{-7} [5]	0.4×10^{-7}	0.5×10^{-7} [5]	atm ⁻¹
ΔT_c (1 000 atm)	+ 0.1	0 ± 0.1 [6]	+ 0.25	+ 0.35 [6]	°K

Physical quantities derived with equation (1), using the values given above.

term in the energy, $E_H = -\frac{1}{2} W\sigma^2$. Owing to the band structure, in a solid an opposing force exists (cf. fig. 2).

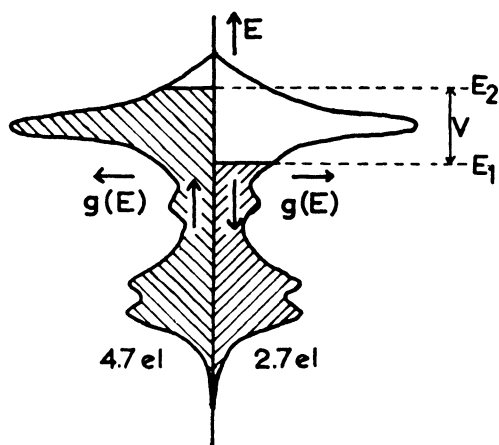


FIG. 2. — 3d-bands of iron, magnetized upwards, for electrons with different spin orientations. Energies connected with the magnetic moment are not presented.

By bringing a virtual amount of electrons from the Fermi level of one semi-band to the other, it can be seen that the condition for equilibrium is : $V = 2W\sigma$. If the shape of the band is known, as is approximately the case for iron [7], the equilibrium value of σ , σ_p , can be calculated if W is known. For iron we need a value of W of 0.17 eV in order to obtain the right σ_p ; this value is in reasonable agreement with data valid for free iron atoms, obtained from atomic spectroscopy, which yields : $W \approx 0.33$ eV.

For small deviations from σ_p , the energy change can be calculated easily. We find :

$$1/2 A(\sigma - \sigma_p)^2 = 1/2 \left\{ \frac{1}{4g(E_1)} + \frac{1}{4g(E_2)} - W \right\} (\sigma - \sigma_p)^2,$$

where $g(E_1)$ and $g(E_2)$ are the densities of states at both Fermi levels in figure 2. In the above equation A is expressed in known constants. Substitution yields the value of $A = 0.38$ eV for iron. In nickel, $g(E_2) = 0$, so that A would become infinite. If we take transitions from the 3d-band to the conduction band into account, we find here that $A \approx 1.7$ eV. This is in good agreement with the several experimental values given in the table. The above derivation is valid for 0 °K. At higher temperatures the magnetization decreases, owing to disorder of the atomic spins. Following Stoner [8], this would result in a movement of both levels in figure 2 towards each other, until the difference becomes zero at the Curie point. We however, are of the opinion that, since localized atomic moments remain, a splitting like that represented in figure 2 also remains above the Curie point. A certain configuration of 3d-electrons stays upon one atom for a comparatively long time. The left-hand side of figure 2 now represents the majority electrons, which are the electrons having their spin parallel to the moment of the atom to which they belong at that instant. The right-hand side represents the minority electrons. If the band of electrons with spin upwards

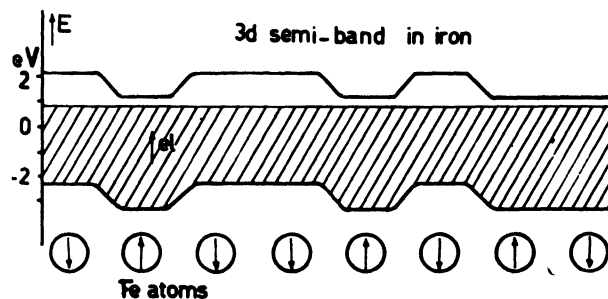


FIG. 3. — Fluctuating band of \uparrow electrons in iron above its Curie point. The complementary band of \downarrow electrons is not drawn.

in iron above the Curie point is drawn, this band would fluctuate through the lattice, as is represented in figure 3.

From the foregoing it follows that A and some of the other constants mentioned above are not strongly dependent on temperature, which agrees with the experimental results. This picture is therefore the justification of the applicability of

equation (1), not only at 0 °K, but at all temperatures. The author wishes to express his gratitude to Prof. Dr. G. W. Rathenau, for many helpful discussions. This work is part of the research program of the " Stichting voor Fundamenteel Onderzoek der Materie " and was made possible by financial support from the " Nederlandse Organisatie voor Zuiver Wetenschappelijk Onderzoek ".

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