

**THE CHANGE OF ATOMIC MAGNETIC MOMENTS IN FERROMAGNETIC METALS  
UNDER HYDROSTATIC COMPRESSION**

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A study was made of the change of magnetic moment  $\sigma$  and electrical resistance  $R$  under hydrostatic compression for iron, nickel, iron-nickel alloys with nickel contents of 34% or more, and nickel-copper alloys containing up to 48% copper; the study was made at helium temperatures using magnetic fields up to 8600 oe. It was established that, in iron-nickel alloys with nickel contents between 34 and 45% (Invar alloys), the change of  $\sigma$  and  $R$  under pressure was approximately an order greater than in the other alloys and metals studied. In all the metals and alloys studied the magnetic moment decreased under hydrostatic compression. The electrical resistance decreased under hydrostatic compression for iron and nickel, and increased for Invar alloys. Possible reasons are considered for the changes of  $\sigma$  and  $R$  which were found, and it is shown that the comparatively large changes of these quantities under pressure and the other peculiarities of Invar alloys can be explained by the existence of "latent" anti-ferromagnetism, existing due to the negative sign of the exchange integral between atoms of iron in the face-centered lattice.

THE authors have previously shown<sup>1</sup> that the quantity  $(1/\sigma_0)\Delta\sigma/\Delta p = K_\sigma$  (where  $\sigma_0$  is the saturation magnetic moment of unit mass) does not tend to zero at  $T = 0^\circ\text{K}$  for Invar alloys. It was also established that the change of magnetization under hydrostatic compression cannot be completely attributed to a displacement of the Curie point.

For pure ferromagnetic metals  $K_\sigma$  is much smaller than for Invar alloys. The values of  $K_\sigma$ , quoted in the literature by various authors for nickel and iron at room temperature, differ greatly from one another. This is associated with experimental difficulties arising because of the smallness of the effect sought for ( $\Delta\sigma/\sigma_0 \approx 10^{-4}$  at 1,000 atm<sup>2-4</sup>). With regard to data on the behavior of this quantity at low temperatures, there has been only one attempt<sup>5</sup> to determine  $(1/\Psi)\Delta\Psi/\Delta p$  for iron at liquid nitrogen temperature ( $\Psi$  is the magnetic flux through the specimen).

In the present work we have studied the effect of hydrostatic compression on the saturation magnetization of nickel, iron, a single crystal of meteoric iron, and iron-nickel and copper-nickel alloys, at temperatures of 20.4° and 4.2° K. It was also of interest to find what connection  $K_\sigma$  had with the effect of hydrostatic compression on the electrical resistance of these metals, i.e., the quantity  $(1/R_0)\Delta R/\Delta p = K_R$ .

#### MEASUREMENT PROCEDURE AND RESULTS

The arrangement for measuring the quantity  $(1/\Psi)\Delta\Psi/\Delta p$  is shown schematically in Fig. 1. A bomb of beryllium bronze containing the specimen, 1, is situated in the field of a water-cooled solenoid, 3. A pressure of approximately 150 atm was provided by gaseous helium which was supplied to the bomb from a gas cylinder through a capillary, 4. The measuring coil, 2, disposed outside the bomb, had two sections, an internal and an external. The sections were connected in series in opposite sense. The number of turns in each section was chosen so that, on reversing the magnetic field in the solenoid with no specimen present, the signals arising in each section cancelled one another out. The change of magnetic moment of the specimen was recorded because of the different number of turns in the inner and outer sections; the difference in the number of turns in our case was 7643. The measuring coil was connected to a photoelectric fluxmeter with sensitivity 5 maxwells/division. In the experiment, the movement of the fluxmeter pointer, due to the change of magnetic flux through the specimen on applying and removing the pressure, was measured directly. The error of the measurements, referred to unit cross-sectional area of the specimen  $\delta [(1/4\pi s)\Delta\Psi/\Delta p]$ , was

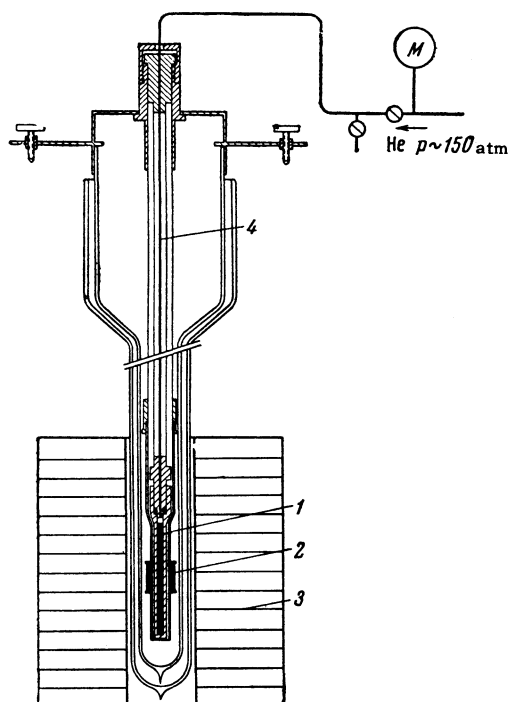


FIG. 1. Diagram of the basic part of the apparatus.

$0.2 \times 10^{-4}$  gauss/atm. The specimens used were in the form of cylinders 112 mm long and 5.9 mm in diameter. The specimens were fabricated from technically pure materials. After fabrication they were subjected to a vacuum anneal for 8 hours at a temperature of  $1,000^\circ\text{C}$  and were subsequently cooled in the furnace.

For the measurements on  $K_R$ , the pressure was created by freezing an aqueous solution of alcohol ( $\sim 3\%$ ) at constant volume. We used the same method as in references 6 and 7.

The main results are shown in Fig. 2. Curves 1 and 2 display, respectively, the dependence of  $(1/\Psi_0)\Delta\Psi_0/\Delta p$  and  $K_R$  on specimen composition. The specimens were at a temperature of  $4.2^\circ\text{K}$ , and a field of 8600 oe was applied, magnetic saturation being obtained. The quantity  $(1/\Psi_0)\Delta\Psi/\Delta p$  had the same value at  $20.4^\circ$  and  $4.2^\circ\text{K}$  within the limits of accuracy of our measurements. The value of  $K_R$  generally depended on the temperature, and for all the ferromagnets studied the dependence was such that  $\partial/\partial T(\Delta R/\Delta p) < 0$ . In the region of Invar alloys where  $(1/\Psi_0)\Delta\Psi/\Delta p$  and  $K_R$  are large, the behavior of curves 1 and 2 is approximately similar, and while the magnetization of the specimen decreased under hydrostatic compression, its electrical resistance increased. The increase of  $(1/\Psi_0)\Delta\Psi/\Delta p$  for nickel-copper alloys with increasing copper content is associated with the decrease of  $\Psi_0$ , as can be seen from the table.  $K_\sigma$  is related to  $(1/\Psi_0)\Delta\Psi/\Delta p$  by the relationship

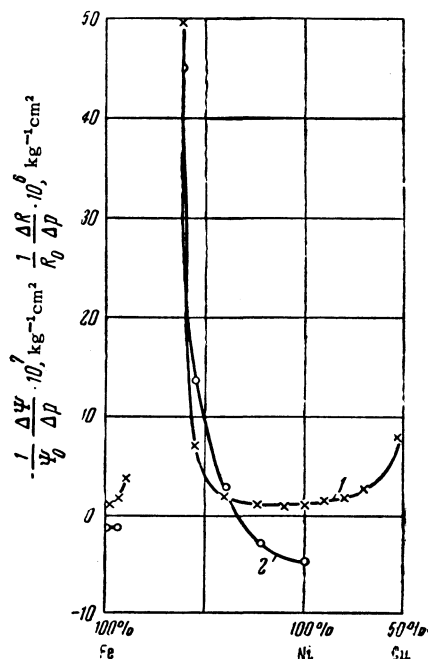


FIG. 2. Values of the quantities  $(1/\Psi_0)\Delta\Psi_0/\Delta p$  (curve 1) and  $(1/R_0)\Delta R_0/\Delta p = K_R$  (curve 2) for iron, nickel, iron-nickel and copper-nickel alloys in the magnetic saturation region.  $\Psi_0$  is the magnetic flux of the specimen obtained by extrapolation to absolute zero,  $R_0$  is the residual resistance.

$$K_\sigma = (1/\Psi_0)\Delta\Psi/\Delta p + \frac{1}{3}\Delta V/V\Delta p.$$

Since both the terms on the right-hand side have negative signs,  $|K_\sigma| > |(1/\Psi_0)\Delta\Psi/\Delta p|$ . The calculated values of  $K_\sigma^*$  are given in the table.

The values of  $K_R$  for pure electrolytic nickel ( $R_{300^\circ}/R_{4.2^\circ} = 250$ ) and for technically pure nickel ( $R_{300^\circ}/R_{4.2^\circ} = 6$ ) are equal.

The variation of  $(1/\Psi_0)\Delta\Psi_0/\Delta p$  with magnetic field  $H$  at  $20.4^\circ\text{K}$  is given in Fig. 3. Curve 1 refers to a specimen of polycrystalline nickel, curve 2 to a specimen of single-crystal meteoric iron (5.5% Ni) cut in the direction of the  $[100]$  axis, i.e., along the direction of easy magnetization. In Fig. 4 are given the curves showing the variation of the electrical resistance of nickel with magnetic field at a temperature of  $4.2^\circ\text{K}$  for two pressures, atmospheric (curve 1) and 1380 atm (curve 2). The curves cross at low magnetic fields. This is associated with the increase of  $(1/\Psi_0)\Delta\Psi/\Delta p$  on reducing the magnetic field  $H$ .

## DISCUSSION OF RESULTS

The authors have previously<sup>1</sup> enumerated the possible reasons for the changes of specific spontaneous magnetization  $\sigma_0$  and residual electrical resistance  $R_0$  close to absolute zero under hydro-

\*We took the values of  $\frac{1}{3}\Delta V/V\Delta p$  for  $T = 300^\circ\text{K}$  from reference 2.

Ni content in NiFe alloy specimens %	Saturation magnetization $I_0$ , gauss	Specific electrical resistance $\rho_0$ at $T = 4.2^\circ\text{K}$ $\mu\text{ohm cm}$	$\frac{1}{4\pi s} \frac{\Delta\Psi}{\Delta p} \cdot 10^4$ , gauss $\text{kg}^{-1} \text{cm}^2$	$\frac{1}{\Psi_0} \frac{\Delta\Psi}{\Delta p} \cdot 10^7$ , $\text{kg}^{-1} \text{cm}^2$	$K_G \cdot 10^7$ , $\text{kg}^{-1} \text{cm}^2$	$K_R \cdot 10^4$ , $\text{kg}^{-1} \text{cm}^2$
0	1751*	2.58	-1.9	-1.1±0.2	-3.1	-1.4±0.3
10.0	1750	—	-6.7	-3.8±0.4	-5.9	—
34.7	1250	50	-131	-105±15	-109	+38±3
38.0	1360	31.2	-68	-50±8	-54	+45±3
44.9	1380	18.8	-9.66	-7±1.5	-11	+13.5±2
59.9	1100	10.4	-2.03	-1.9±0.2	-4.4	+2.6±0.5
76.0	948	6.15	-1.01	-1.1±0.25	-3.6	-2.9±0.5
90.2	680	—	-0.57	-0.85±0.3	-3	—
100	512	0.034***	-0.56	-1.1±0.4	-2.9	-4.7±1.5
Cu content in NiCu alloy specimens %						
10.2	415	—	-0.63	-1.5±0.5	—	—
19.6	350	—	-0.60	-1.7±0.6	-4.3	—
29.0	235	—	-0.64	-2.7±0.8	-5.6	—
48.4	82.5	—	-0.65	-7.9±2.5	—	—

\*Value for  $I_0$  for iron from reference 8.  
 \*\*For electrolytically pure nickel.  
 \*\*\*For technically pure nickel.

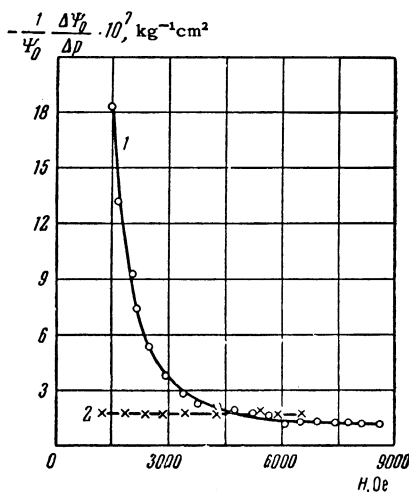


FIG. 3. Variation of the relative change of magnetic flux under pressure on magnetic field at  $T = 20.4^\circ\text{K}$ : 1 - nickel, 2 - single-crystal meteoric iron (magnetic field directed along the [100] axis).

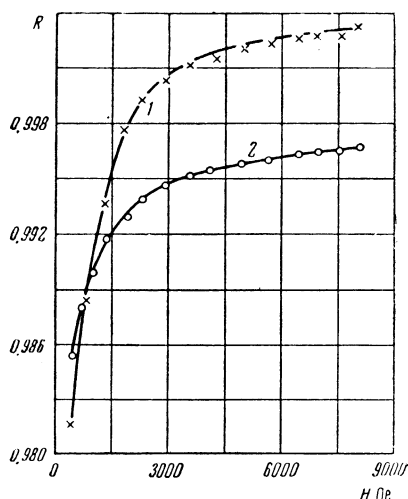


FIG. 4. Variation of the electrical resistance of nickel ( $R$  in relative units) with magnetic field at  $T = 4.2^\circ\text{K}$ : 1 - at atmospheric pressure, 2 - at pressure of 1380 atm.

static compression.\*

1. A change in the value of the d-d exchange integrals  $J_{dd}$  on changing the interatomic distances under pressure. If some of these integrals are negative ("latent" antiferromagnetism), the change of  $J_{dd}$  involves a change in the number of anti-parallel spins in the lattice; consequently, there is a decrease (or increase) of the spontaneous magnetization. Therefore the electrical conductivity decreases (or increases); this can occur due to the change in degree of magnetic order and the change in the value of the spontaneous magnetization.

2. A change in the values of the s-d exchange integrals  $J_{sd}$  on changing the interatomic distances. Changing  $J_{sd}$  alters the molecular field acting on the conduction electrons, which affects their contribution to the spontaneous magnetization.<sup>9</sup> In this case the electrical conductivity can either decrease or increase, because the degree of scattering of the conduction electrons at inhomogeneities of the magnetic moment depends on the quantities  $J_{sd}$ ,<sup>10</sup> and because there are changes in the effective mass and mean free path of the conduction electrons associated with their polarization in the molecular field.<sup>9</sup>

3. The transfer of electrons from the s into the d band or vice versa under lattice compression. Here the number of unfilled sites in the d band changes, as well as the number of s electrons, which alters the magnetic moment and the electrical conductivity. In this case the electrical

\*The order in which these are enumerated is different from that in reference 1.

conductivity changes because the number of carriers alters and because there is an increase or decrease of the number of "holes" and the degree of scattering of electrons at "holes."

Electrical conductivity changes can also be due to a "smoothing" of irregularities in the potential — which is possible under hydrostatic compression.

According to the way in which  $\sigma_0$  and  $R_0$  change under hydrostatic compression, the metals and alloys studied can be divided into two groups: a) iron-nickel alloys with nickel contents of 34, 38, and 45% (Invar alloys), in which the absolute values of  $K_\sigma$  and  $K_R$  are approximately an order of magnitude greater than in the other materials studied, and in which these quantities have opposite signs ( $K_\sigma < 0$ ,  $K_R > 0$ ); b) iron, nickel, iron-nickel alloys with nickel contents 10, 60, 76, and 90%, and copper-nickel alloys, in which the absolute values of  $K_\sigma$  and  $K_R$  are comparatively small and both are negative —  $K_R$  being approximately the same for specimens with different values of residual resistance. The difference referred to between the quantities  $K_\sigma$  and  $K_R$  shows that the changes of  $\sigma_0$  and  $R_0$  observed under hydrostatic compression in materials of the first and second groups, are due essentially to different factors.

**A. The changes of  $\sigma_0$  and  $R_0$  under hydrostatic compression for materials of the first group (Invar alloys).** It has been remarked above that the absolute values of  $K_\sigma$  and  $K_R$  in Invar alloys are an order of magnitude greater than in other iron-nickel alloys, iron, and nickel. It is well known that Invar alloys also show a number of other anomalies (anomalous coefficient of thermal expansion, high electrical resistance, etc). It is natural to look for a general explanation of all these anomalies. In reference 11 we have shown that such an explanation can be given starting from the hypothesis of a negative exchange interaction between the d electrons of iron in the face-centered lattice. This hypothesis is based on experimental data obtained in reference 12, where it was established that an alloy of iron, nickel and chromium with face-centered lattice, containing 72% iron, becomes antiferromagnetic at a temperature of about 40°K.

As shown in reference 11, small changes in the exchange integral for d electrons on neighboring iron ions displaces the transitional concentration region (i.e., the alloy concentrations lying intermediate between antiferromagnetic and ferromagnetic) towards nickel, and therefore strongly affects  $\sigma_0$ . With decreasing interatomic distance the absolute value of the negative exchange inte-

grals increases more rapidly than it decreases; therefore, under hydrostatic compression a displacement of the transitional region towards the nickel side and, consequently, a decrease of magnetic moment would be expected. Due to the increase in the number of anti-parallel spins, the electrical resistance of the alloys would thus be expected to increase. Therefore, if the hypothesis on the role of antiferromagnetism in iron as an explanation of the anomalies in Invar alloys is true, and if the first of the reasons enumerated above for the change of  $\sigma_0$  and  $R_0$  is the most potent, then the quantities  $K_\sigma$  and  $K_R$  in iron-nickel alloys should attain their greatest absolute values in the same concentration region as that in which the fall of magnetic moment with increasing iron content is observed, and they should have different signs in this region. From the curves given in Figs. 2 and 5, it is seen that the greatest values of  $K_\sigma$  and  $K_R$  do, in fact, correspond to the concentration region referred to, and these quantities have different signs in this region, and  $\Delta\sigma_0/\Delta p < 0$ .

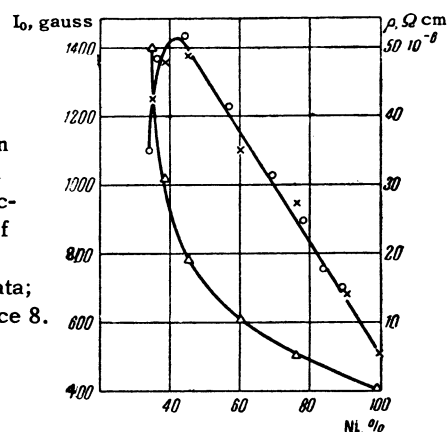


FIG. 5. Saturation magnetization  $I_0$  and residual specific electrical resistance  $\rho_0$  of iron-nickel alloys; x,  $\Delta$  — the authors' data; o — data from reference 8.

Another fact supporting the explanation given of the effect of pressure on  $\sigma_0$  and  $R_0$  in Invar alloys is obtained by comparing the change of these quantities under hydrostatic compression with their changes during a small decrease of the strong magnetic field which magnetizes the alloys to saturation. If the cause of the changes discussed is the increase of the number of anti-parallel spins, then the ratios  $(\Delta R_0/R_0)$ ,  $(\Delta\sigma_0/\sigma_0)$  in both cases should be approximately equal in magnitude and sign. As the comparison made by us in reference 1 shows, these ratios in fact lie close to one another for Invar alloys.

The considerations given support the idea that the most probable basic reason for the changes of  $\sigma_0$  and  $R_0$  in Invar alloys under hydrostatic compression is the first of the reasons enumerated above. If it were assumed that the basic cause of

these changes lay in the second or third reasons, then it is difficult to explain, without artificial assumptions, why in Invar alloys the variations of  $\sigma_0$  and  $R_0$  under pressure are an order greater than in other iron-nickel alloys and in metals.

B. The changes of  $\sigma_0$  and  $R_0$  under hydrostatic compression in iron, nickel, and alloys belonging to the second group. It was remarked above that in metals and alloys belonging to the second group, both  $K_\sigma$  and  $K_R$  are negative. At the same time it is well known that  $(1/\sigma_0)\Delta\sigma/\Delta H$  and  $(1/R_0)\Delta R/\Delta H$  for these materials in strong fields have different signs. A comparison of the ratio  $K_R/K_\sigma$ , calculated for specimens of the metals according to our data, with the ratio  $[(1/R_0)(\Delta R/\Delta H)]/[(1/\sigma_0)\Delta\sigma/\Delta H]$  at room temperature, derived from data in the literature,<sup>13-15</sup> shows that these ratios in a number of cases differ significantly from one another. The first is insensitive to the quantity  $R_0$ , but the second varies with change of this quantity. Thus, in the materials considered, changes of  $J_{dd}$  cannot be the basic cause of changes in  $\sigma_0$  and  $R_0$  under hydrostatic compression.

It follows from the usual ideas about the relation between the magnetic moments of ferromagnetic metals and the degree of filling of the d band, that on transferring electrons from the d band to the s band, together with a fall of electrical resistance, a rise of magnetic moment should be observed (at least in cobalt and nickel, where the conductivity is mainly due to s electrons). Thus,  $(\Delta\sigma_0/\sigma_0)_{d \rightarrow s} \approx -\alpha\Delta R_0/R_0$ , where  $\alpha$  is the ratio of the number of electrons to the number of unfilled sites in the d band (for nickel  $\alpha = 1$ ). It is seen from the table that for electrolytic nickel  $K_R$  is approximately 15 times greater in absolute terms than  $K_\sigma$ . Consequently, without additional assumptions on the various mechanisms of compensating  $(\Delta\sigma/\sigma_0)_{d \rightarrow s}$ , this assumption contradicts the experimental data on the effect of pressure on  $K_\sigma$  in nickel. The reverse assumption — that, under the influence of pressure, electrons are transferred from the s into the d band, and that this is the basic cause of the fall of  $\sigma_0$  under hydrostatic compression observed in iron and nickel — leads to a quantitative contradiction of the experimental data on the effect of pressure on the electrical resistance of iron.

Thus, the third of the possible reasons enu-

merated above, as well as the first, cannot be the basic reason for the changes of  $\sigma_0$  and  $R_0$  in the metals under hydrostatic compression. There remains the assumption that the basic cause for these changes in the metals is the second one (i.e., the effect of pressure on the values of the  $J_{sd}$  s-d exchange integrals), and that in Invar alloys this effect exceeds the effect of the first one. Unfortunately, it is at present difficult to estimate how the electrical resistance would change during variations of  $\sigma_0$  associated with changes of  $J_{sd}$ .

We note that, if the basic reason for the observed fall of electrical resistance in the metals under pressure is "smoothing" of irregularities of potential, then the quantity  $K_R$  should be insensitive to the value of the residual resistance; this is confirmed by the measurements for pure electrolytic nickel and technically pure nickel.

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