Magnetization of $Fe_{65}(Ni_{1-x}Mn_x)_{35}$ alloys in fields up to 350 kOe

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The field and temperature dependences of the magnetizations of disordered fcc $Fe_{65}(Ni_{1-x}Mn_x)_{35}$ alloys in the ferromagnetic concentration range ($0 < x < 0.3$) have been investigated at temperatures between 77 and 360°K in pulsed magnetic fields up to 350 kOe. It is shown that the theory of very weak itinerant ferromagnetism developed by Wohlfarth gives a good description of the magnetic properties of these alloys. It is found that the molecular field approximation indicates a substantial difference in the behavior of the lines of equal magnetization for localized ferromagnets and for very weak itinerant ferromagnets. This difference can be used to classify ferromagnets according to the type of statistics (Maxwell–Boltzmann or Fermi–Dirac) obeyed by the magnetic-moment carriers.

It was shown in [1-3] that many properties of disordered fcc alloys of composition $Fe_{65}(Ni_{1-x}Mn_x)_{35}$ can be explained on the basis of the theory of very weak itinerant (collectivized) ferromagnetism. The low-temperature specific heats of these alloys were investigated in [1]. A peak was found in the concentration dependence of the electronic specific heat and the presence of this peak was explained in terms of the theory of very weak itinerant ferromagnetism. This theory has also been used to explain the results of measurements of the magnetization, the Curie point and the susceptibility under pressure [2], and the temperature dependence of the spontaneous bulk magnetostriction [3] of $Fe_{65}(Ni_{1-x}Mn_x)_{35}$ alloys. It would accordingly seem of interest also to test other conclusions concerning these alloys that can be drawn from the theory of very weak itinerant ferromagnetism.

We have investigated the temperature and field-strength dependences of the magnetization of polycrystalline specimens of $Fe_{65}(Ni_{1-x}Mn_x)_{35}$ alloys with $0 < x < 0.3$. In this concentration range ferromagnetic ordering sets in when the temperature is lowered sufficiently [4].

Wohlfarth's theory [5,6] of very weak itinerant ferromagnetism gives the following equation in the molecular field approximation for the magnetization as a function of field strength and temperature:

$$\left[\frac{\sigma(H,T)}{\sigma(0.0)}\right]^3-\frac{\sigma(H,T)}{\sigma(0,0)}\left[1-\left(\frac{T}{T_c}\right)^2\right]=\frac{2\chi_0 H}{\sigma(0,0)}. \qquad (1)$$

Here $\sigma(H, T) = \sigma$ is the magnetization per unit mass, $\sigma(0, 0) = \sigma_0$ is the magnetization in zero field at the absolute zero of temperature, T_c is the Curie point, and χ_0 is the initial susceptibility per unit mass at the absolute zero of temperature. For very weak itinerant ferromagnets we have $\chi_0 \neq 0$ [5,6].

Equation (1) is satisfied when $T \ll T_F$ and $\zeta \ll 1$; here T_F is the effective degeneracy temperature and $\zeta = \sigma/[nN\mu_B]$, where μ_B is the Bohr magneton, N is the number of atoms per unit mass, and n is the number of magnetically active electrons per atom.

Equation (1) can be rewritten in the form [5,6]

$$\sigma^2=\sigma_0^2\left[1-\left(\frac{T}{T_c}\right)^2\right]+2\chi_0\sigma_0^2\frac{H}{\sigma}. \qquad (2)$$

From this it follows that for a very weak itinerant ferromagnet the $\sigma^2 = f(H/\sigma)$ curves for various fixed temperatures in the range $0 < T \ll T_F$ (this range may include T_c) will constitute a family of parallel straight lines. Similar relationships were derived in [7,8] from Landau's theory of second order phase transitions, but (in contrast to the result derived from Wohlfarth's theory) only for a small neighborhood of the Curie point.

It is evident from Eq. (2) that the slope of the $\sigma^2 = f(H/\sigma)$ lines is $2\chi_0\sigma_0^2$. The line for $T = T_c$ passes through the origin of coordinates. The intercept q(T) on the σ^2 axis of the $\sigma^2 = f(H/\sigma)$ line for $T < T_c$ is equal to the square of the spontaneous magnetization: σ_s^2. The temperature dependence of q(T) is obtained by setting H = 0 in (2):

$$q(T)=\sigma_0^2\left[1-\left(\frac{T}{T_c}\right)^2\right]=\sigma_0^2(1-t^2). \qquad (3)$$

Thus, the experimental values of q(T) for a very weak itinerant ferromagnet should lie on a straight line in the q, T^2 plane. Extrapolation of this line to T = 0 gives the value of $q(0) = \sigma_0^2$, and the line crosses the T^2 axis at T_c^2. The physical meaning of the intercept q(T) for a temperature $T < T_c$ is the square of the spontaneous magnetization, $\sigma_s^2(T)$, at the given temperature T. Knowing the slope of the lines $\sigma^2 = f(H/\sigma)$, which is equal to $2\chi_0\sigma_0^2$, and also knowing σ_0, we can find χ_0.

Thus, from the experimental data on the temperature and field-strength dependences of the magnetization one can evaluate all three of the phenomenological constants σ_0, T_c, and χ_0, which determine the magnetic properties of a very weak itinerant ferromagnet.

It follows from Eq. (1) that the temperature dependence of the initial susceptibility has the form [5,6]

$$\frac{1}{\chi}=\frac{1}{\chi_0}\left[1-\left(\frac{T}{T_c}\right)^2\right] \quad \text{for } T<T_c; \qquad (4)$$

$$\frac{1}{\chi}=\frac{1}{2\chi_0}\left[\left(\frac{T}{T_c}\right)^2-1\right] \quad \text{for } T_F\gg T>T_c. \qquad (5)$$

The $Fe_{65}(Ni_{1-x}Mn_x)_{35}$ alloys used in the present study were prepared by induction fusion. The compositions of the alloys are given in the table in weight percent. The ingots were subjected to a homogenizing anneal at 1000°C for 10 hr and were then quenched in water. The alloys obtained in this manner are homogeneous disordered solid solutions with an fcc lattice all the way down to liquid nitrogen temperatures.

Cylindrical specimens 10 mm long and 1.4 mm in diameter weighing about 120 mg, were made from the ingots. The magnetization of the $Fe_{65}(Ni_{1-x}Mn_x)_{35}$ alloys was measured at temperatures from 77 to 360°K in a pulsed magnetic field (pulse length 0.01 sec, pulse height up to 350 kOe). The magnetization measurements were

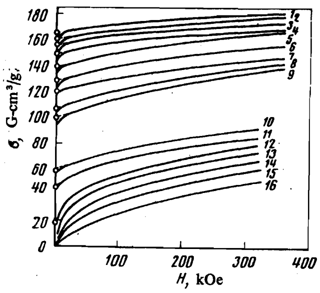

FIG. 1. Magnetization isotherms for $Fe_{65}(Ni_{1-x}Mn_x)_{35}$ alloys. Curves 1–9 are for alloys containing no manganese (x = 0) at the following temperatures (°K): 1–79, 2–113, 3–155, 4–190, 5–229, 6–625, 7–308, 8–337, 9–358; curves 10–16 are for alloys containing 5 wt. % manganese (x ~ 0.14) at the following temperatures (°K) 10–80, 11–163.5, 12–225, 13–250, 14–290, 15–329, 16–368.

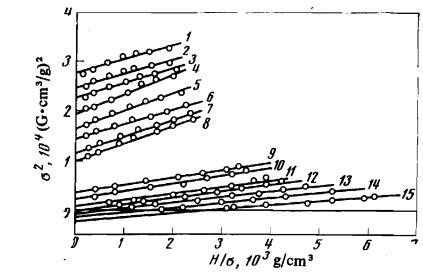

FIG. 2. σ^2 vs H/σ plots for $Fe_{65}(Ni_{1-x}Mn_x)_{35}$ alloys. Curves 1–8 are for alloys containing no manganese (x = 0) at the following temperatures (°K): 1–79, 2–155, 3–190, 4–229, 5–265, 6–308, 7–337, 8–358; curves 9–15 are for alloys containing 5 wt. % manganese (x ~ 0.14) at the following temperatures (°K): 9–80, 10–163.5, 11–225, 12–250, 13–290, 14–329, 15–368.

made by an induction method [9]. The errors in measuring the magnetization and the magnetic field strength amounted to 8%. The random errors (spread of the experimental values) were 3% for the magnetic field strength and 5% for the magnetization. The magnetizing process was adiabatic. The maximum change in the temperature of the specimens due to the magnetocaloric effect reached 8° near T_c in the strongest field. Then the values of $(\partial\sigma/\sigma T)_H$ were less than or equal to 0.25 G·cm³/g·°K. Thus, near T_c and in the strongest fields, the measured magnetization adiabats differ from isotherms by only 2 G·cm³/g at a magnetization of ≈ 150 G·cm³/g, i.e., the difference amounts to only 1.5%. The difference is even smaller in weaker fields and farther from T_c. As a good approximation, therefore, the experimental magnetization curves obtained in the present study can be regarded as isotherms.

Figure 1 shows our measured magnetization isotherms for alloys containing 0 and 5% manganese (the full curves). The circles on the σ axis marks the values of the spontaneous magnetization σ_s determined from the equations of Wohlfarth's theory. The dashed lines at the weak-field ends of the curves show the extrapolation used to find the initial susceptibility of the paraprocess. The alloy containing no manganese (curves 1—9) is ferromagnetic over the entire investigated temperature interval, the spontaneous magnetization ranging from 100 to 160 G-cm³/g. The susceptibility of the paraprocess is of the order of 10^{-4} cm³/g and depends strongly on the field strength.

The alloy containing 5% manganese (curves 10—16) is ferromagnetic at the low-temperature end of the investigated temperature interval and paramagnetic at the high-temperature end. The spontaneous magnetization at 80°K is 60 G-cm³/g. The susceptibility of the paraprocess in the ferromagnetic region and the paramagnetic susceptibility are very large (10^{-4}—10^{-3} cm³/g) and strongly field dependent. The magnetization in the 330 kOe field at a temperature 100°C above the Curie point (curve 16 of Fig. 1) is greater than the spontaneous magnetization at a temperature almost 100°C below the Curie point (curve 11 of Fig. 1).

The magnetization curves of the other investigated alloys look much like those shown in Fig. 1.

Figure 2 shows σ^2 vs H/σ plots for alloys containing σ and 5% manganese. It is evident that the experimental points for the alloy containing no manganese (plots 1—8) lie close to straight lines. The slopes of these lines are all equal within 10%. The σ^2 vs H/σ plots for the alloys containing 1.5, 3, and 8% manganese exhibit similar characteristics. These alloys were investigated in a temperature interval that lay either entirely below or entirely above the Curie point. The alloy containing 5% manganese was investigated both above and below the

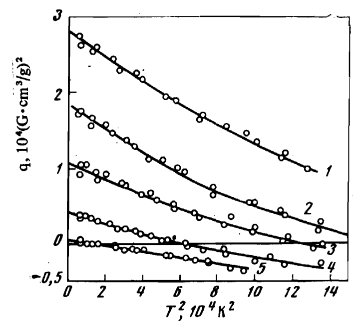

FIG. 3

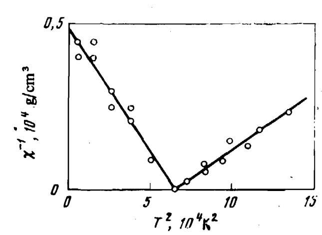

FIG. 4

FIG. 3. Intercepts q on the σ^2 axis of the σ^2 vs H/σ lines (Fig. 2) vs temperature squared for $Fe_{65}(Ni_{1-x}Mn_x)_{35}$ alloys containing the following amounts of manganese (wt. %): 1–0, 2–1.5, 3–3, 4–5, 5–8.

FIG. 4. Reciprocal of the initial susceptibility vs temperature squared for the $Fe_{65}(Ni_{1-x}Mn_x)_{35}$ alloy containing 5 wt. % manganese.

Curie point. The $\sigma^2 = f(H/\sigma)$ lines for this alloy are parallel only for $T < T_c$. For $T > T_c$ the slope of the lines decreases with increasing temperature, the change in the slope being too great to be attributed to experimental errors; this behavior is in conflict with the predictions of Wohlfarth's theory.

Figure 3 shows the quantity q (see Eq. (3)) as a function of the square of the temperature for all the investigated alloys. The $q(T^2)$ curve for the alloy containing 8% manganese, which is in the paramagnetic state throughout most of the investigated temperature interval, is a straight line, in accordance with the predictions of the theory of very weak itinerant ferromagnetism (Eq. (3)). The experimental $q(T^2)$ curve for the alloy containing 5% manganese can be represented by two straight lines, one below the Curie point and another above it. Deviation from the linear relationship can be discerned in a neighborhood of the Curie point. A similar relationship was found in [10] for $ZrZn_2$. According to [6], such a deviation from the theoretical relationship might be due to the presence of short-range order at $T > T_c$. It is also possible that the presence of short-range order might be responsible for the temperature dependence of the slope of the $\sigma^2 = f(H/\sigma)$ lines for this alloy at $T > T_c$. The $q(T^2)$ curves for the alloys containing 3 and 1.5% manganese deviate from straight lines at $T < T_c$. This deviation may be due to the fact that the magnetization of these alloys is fairly high, so that the condition $\zeta \ll 1$ for the applicability of the theory of very weak itinerant ferromagnetism may not be sufficiently well satisfied for these alloys.

Magnetic characteristics of $Fe_{65}(Ni_{1-x}Mn_x)_{35}$ alloys derived from magnetization measurements via the theory of very weak itinerant ferromagnetism[5,6]

Wt. % Mn	Wt. % Ni	x	T_C, K	σ_0, G·cm³/g	χ_0, 10^{-4} cm³/g*)	
					I	II
0.0	35.0	0,000	475±30	169±14	0.57±0.06	—
1.5	33,5	0.043	415±30	136±11	0.73±0.11	—
3.0	32.0	0.086	360±10	104±8	1,03±0,15	—
5.0	30.0	0.142	255±10	63±5	1,9±0,3	2.2±0.30
8.0	27.0	0.228	125±5	24±5	6.0±1.5	7.4±2.0

*The value of χ_0 given in column I were obtained from the slopes of the σ^2 vs H/σ lines and the temperature trend of σ_S^2, and those in column II, from the temperature dependence of the initial susceptibility.

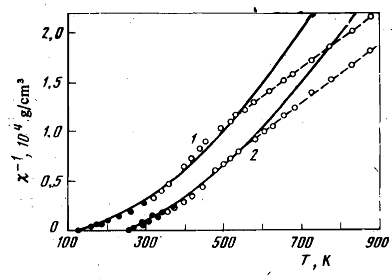

FIG. 5

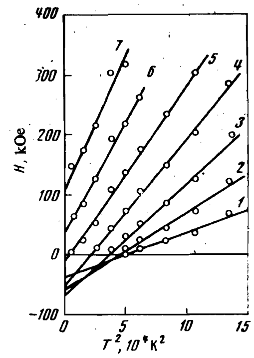

FIG. 6

FIG. 5. Reciprocal of the initial susceptibility vs temperature for $Fe_{65}(Ni_{1-x}Mn_x)_{35}$ alloys. The black circles represent the results obtained in the present study for alloys containing 8 wt. % (curve 1) and 5 wt. % (curve 2) manganese; the open circles represent the results of [4] for alloys containing 7.84 at. % (curve 1) and 5.56 at. % (curve 2) manganese.

FIG. 6. Lines of equal magnetization in the H, T^2 plane for the alloy containing 5 wt. % Mn and the following magnetizations (G·cm³/g): 1–20, 2–30, 3–40, 4–50, 5–60, 6–70, 7–80.

The $q(T^2)$ curve for the alloy containing no manganese also deviates from a straight line. The deviation is not so evident as in the case of the two alloys discussed above; this is due to the fact that the range of reduced temperatures $t = T/T_c$ over which the alloy containing no manganese was investigated ($0 < t^2 < 0.36$) is considerably narrower than that over which the alloys containing 1.5 and 3% manganese were investigated ($0 < t^2 < 1$).

Extrapolating the $q(T^2)$ curve to the q axis gives the value of σ_0^2. The Curie temperature is determined either from the intersection of the experimental $q(T^2)$ curve with the T^2 axis, or by extrapolating this curve to the T^2 axis. The values of σ_0 and T_c obtained in this way are shown in the table for all the investigated alloys; within the experimental errors, they agree with the results obtained in [4].

From the slopes of the experimental σ^2 vs H/σ lines, which are equal to $2\chi_0\sigma_0^2$, and the values of σ_0^2, we evaluated χ_0, the initial susceptibility at the absolute zero of temperature, for all the investigated alloys (table, column I).

Figure 4 shows a plot of the reciprocal susceptibility $1/\chi$ vs the square of the temperature for the alloy containing 5% manganese. For $T < T_c$, the value of χ was obtained from the initial slope of the curve found by extrapolating the magnetization isotherm to the point $\sigma = \sigma_S$, $H = 0$ (Fig. 1); for $T > T_c$, the value of χ was obtained from the slope of the tangent to the magnetization curve at $H = 0$.

It is evident that the experimental $1/\chi$ vs T^2 curves can be approximated above and below the Curie point by straight lines that correspond to Eqs. (4) and (5) and intersect at $T^2 = T_c^2$. The values of χ_0 found from the slopes of these lines below and above T_c^2 using Eqs. (4) and (5) are 2.0×10^{-4} cm³/g and 2.2×10^{-4} cm³/g, respectively. Within the experimental errors, these values agree well with one another and with the value given in the table (column I). The temperature dependence of the initial susceptibility for the alloy containing 8% manganese is much like that for the alloy containing 5% manganese. The temperature dependences are not given for the alloys containing smaller quantities of manganese since the experimental data for these alloys spread widely on account of the relatively small values of χ. The values of χ_0 for the alloys with 8 and 5% manganese determined from the temperature dependence of the initial susceptibility above T_c using Eq. (5) are given in the table (column II). Figure (5) shows the temperature dependence above T_c of the reciprocal susceptibility of the alloys containing 5 and 8% manganese (black circles). The open circles show the results obtained in [4] for alloys with nearly the same compositions (5.56 and 7.84 at.% manganese). Our results agree well with those of [4]. It is evident from Fig. 5 that a rather sharp change in the form of the temperature dependence of the susceptibility takes place in the temperature range 500–550°K for both of these alloys. At temperatures below 500°K the reciprocal susceptibility is a quadratic function of temperature, in accordance with Wohlfarth's theory. The full curves in Fig. 5 were calculated with Eq. (5), the necessary values of T_c and χ_0 being taken from the table (χ_0 from column II). Above 550°K, the reciprocal susceptibility is a linear function of temperature. This is in qualitative agreement with the conclusions reached in Stoner's paper [11], where it is shown that an itinerant ferromagnet should obey the Curie-Weiss law at high enough temperatures.

From what was said above it follows that the behavior of the magnetic properties of $Fe_{65}(Ni_{1-x}Mn_x)_{35}$ alloys with $0 < x < 0.3$ in the investigated temperature interval ($T < 360$°K) can be approximately described by Eqs. (1)–(5) of the theory of very weak itinerant ferromagnetism. The best agreement between theory and experiment is found for values of x close to 0.3. As the manganese content decreases ($x \to 0$) the observed temperature dependence of the spontaneous magnetization begins to deviate from the theoretical prediction, although qualitative agreement between theory and experiment is still found. This behavior may be attributed to the fact that as the manganese content decreases, the magnetization of the alloy increases, so that the condition $\zeta \ll 1$ for the applicability of the theory becomes less well satisfied.

Figure 6 shows our measured lines of equal magnetization on the H, T^2 plane for the alloy containing 5% manganese. Within the experimental errors, these lines are straight (they would be parabolas on the H, T plane), in accordance with the theory of very weak itinerant ferromagnetism (Eq. (1)). The lines of equal magnetization for the other alloys look much like these.

The intercepts $H(\sigma, T = 0)$ on the field axis of the equal magnetization lines for $\sigma > \sigma_0$ are positive and increase with increasing σ. This fact is of fundamental importance since under certain conditions it makes it possible to determine whether the magnetic-moment carriers in an investigated ferromagnet are collectivized or localized. We must therefore examine the behavior of the function $H(\sigma, 0)$ in more detail. Let us first introduce the following dimensionless variables:

$$y=\frac{\sigma}{\sigma_0},\quad t=\frac{T}{T_c},\quad h(y,t)=\frac{2\chi_0}{\sigma_0}H(\sigma,T). \tag{6}$$

In terms of these variables Eq. (1) assumes the form

$$h(y,t)=y^3-y(1-t^2). \tag{7}$$

Since Eq. (7) contains no physical characteristics of any specific material, h(y, t) is a universal function describing the temperature and field-strength dependence of the magnetization of any very weak itinerant ferromagnet.

The curves in Fig. 7 represent the function h(y, t) as calculated from Eq. (7) for the fixed values 0, 1, and $\sqrt{2}$ of the reduced temperature t. In the figure we have also plotted the experimental values of h(y, t) for the same three fixed values of t, the experimental values being derived from our observed lines of equal magnetization for alloys containing various amounts of manganese, using Eqs. (6) and the values of σ_0, T_c, and χ_0 from the table. The values of h(y, 0) were obtained by extrapolating the lines of equal magnetization to T = 0.

It is evident that the experimental values of h(y, t) for different alloys but for the same fixed reduced temperature $t = T/T_c$ lie on a single curve, which, moreover, is close to the calculated curve. The deviations of the experimental values of h(y, t) from the calculated curves are within the limits of experimental error. Figure 7 also shows that h(y, 0) is not a monotonic function of y, so that $H(\sigma, 0)$ is not a monotonic function of σ either: h(y, 0) is negative when $0 < y < 1$ $(0 < \sigma < \sigma_0)$, has an extremum at $y = \sqrt{1/3}$, changes sign at y = 1, and is positive and increases with increasing y when $y > 1$.

For comparison let us consider the behavior in the molecular field approximation of the lines of equal magnetization for a ferromagnet whose magnetic moments are localized. In this case the magnetization is given as a function of the field strength and temperature by the formula

$$\frac{\sigma(H,T)}{\sigma(0,0)}=B_J\left\{\frac{\mu_s[H+\gamma\sigma(H,T)]}{kT}\right\}, \tag{8}$$

in which B_J is the Brillouin function, J is the inner quantum number of the magnetically active ion, μ_s is the magnetic moment of the saturated ion, and γ is the molecular-field constant. It follows from Eq. (8) that the lines of equal magnetization for a ferromagnet with localized magnetic moments are straight lines on the H, T plane:

$$H(\sigma,T)=-\gamma\sigma+\left[\frac{k}{\mu_s}B_J^{-1}\left(\frac{\sigma}{\sigma_0}\right)\right]T. \tag{9}$$

Here B_J^{-1} is the inverse Brillouin function. From Eq. (9) we find that $H(\sigma, T = 0) = -\gamma\sigma$, i.e., for the case of localized magnetic moments the intercepts of the equal magnetization lines on the field axis are negative and decrease monotonically with increasing magnetization over the entire magnetization interval $0 < \sigma < \sigma_0$.

Thus, it is possible in principle to distinguish reliably between a ferromagnet with localized magnetic moments and a very weak itinerant ferromagnet on the basis of the shape of the equal-magnetization lines and the dependence of the initial ordinates $H(\sigma, 0)$ of those lines on the magnetization.

The equal magnetization lines of a very weak itinerant ferromagnet are straight lines in the H, T^2 plane (parabolas in the H, T plane). The initial ordinate of the equal magnetization line is a nonmonotonic function of the magnetization; it decreases with increasing magnetization for $\sigma < \sqrt{1/3}\cdot\sigma_0$, increases with increasing magnetization for $\sigma > \sqrt{1/3}\cdot\sigma_0$, and is positive for $\sigma > \sigma_0$.

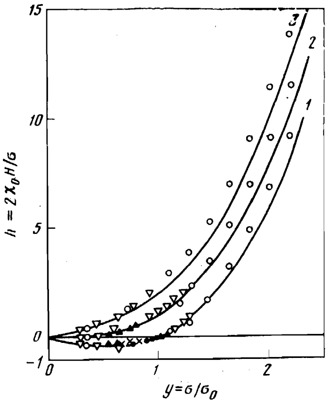

FIG. 7. Wohlfarth's equation in dimensionless variables (Eq. (7) in the text) for the following fixed values of t: 1–t = 0, 2–t = 1, 3–t = $\sqrt{2}$. The curves were calculated with Eq. (7). The points were obtained, using Eqs. (6), from the experimental equal-magnetization lines for the $Fe_{65}(Ni_{1-x}Mn_x)_{35}$ alloys containing the following amounts of manganese (wt. %): black circles–0, crosses–1.5, black triangles–3, open triangles–5, open circles–8.

The equal magnetization lines for a localized ferromagnet are straight lines in the H, T plane. In this case the initial ordinate is negative over the entire magnetization range $0 < \sigma < \sigma_0$ and decreases monotonically with increasing σ. According to Eq. (9), the function $H(\sigma, T)$ does not exist at all for $\sigma > \sigma_0$.

In accordance with the results presented above we should conclude that the investigated alloys are very weak itinerant ferromagnets. We do not feel that this conclusion can be regarded as final, however, since according to the theory of localized magnetic moments, magnetic ordering of a peculiar type, due to the presence of interactions of opposite signs between the atoms of different components and associated with cancelling of the z component of the spin of the crystal, is possible for $Fe_{65}(Ni_{1-x}Mn_x)_{35}$ alloys [12]. The behavior of the magnetization as a function of field strength and temperature that obtains in the presence of magnetic ordering of this type may be of a different kind, generally speaking, than that described by Eq. (8), and it is possible (though not likely) that it might give just as good a description of our experimental results as the theory of very weak itinerant ferromagnetism. Unfortunately the necessary theoretical calculations are not yet available, so the question whether the magnetic-moment carriers in $Fe_{65}(Ni_{1-x}Mn_x)_{35}$ alloys are itinerant or localized must still be regarded as open.

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210