

Ferromagnetism of disordered systems

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This review concerns disordered ferromagnets in which the magnetic atoms are randomly located on sites of a crystal lattice. Attention is given principally to magnets with strong spatial fluctuations of the exchange interaction. Naturally the magnetism of such systems differs substantially from the properties of ordinary ferromagnets; and for them, as for strongly disordered nonmagnetic systems, the methods of perturbation theory are inapplicable. The general concepts of the theory of disordered magnetic systems are presented with application to two models: disordered ferromagnets with interaction between nearest neighbors in the lattice, and ferromagnets in which the exchange interaction depends exponentially on the distance between the magnetic atoms. An example of ferromagnets of the second type is provided by dilute alloys of palladium with iron, cobalt, and manganese. The principal experimental results for these alloys are presented, and it is shown that the theory of disordered magnetic systems describes their properties well.

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I. INTRODUCTION

Disordered magnets—that is, systems in which the magnetic atoms do not form a regular crystalline lattice—have been intensively investigated since the 1960's. Development in this direction was stimulated by two facts. First, there was general interest in the problem of disordered condensed systems. Second, over the last 10 to 15 years there have been obtained disordered magnets of extremely diversified types:

amorphous magnets,¹ spin glasses,¹⁻³ ferromagnetic alloys with small concentrations of magnetic atoms,^{1,4-6} etc. Their properties are peculiar, and an approach to them on the basis of the usual ideas about ordered magnets often proves unsound.

The number of papers devoted to disordered magnets is enormous. We have not attempted to give a review of all the existing material. Our aim is to present, in the simplest cases, the principal concepts of the theory of

disordered magnetic systems, and to apply it to the analysis of the properties of several specific materials.

We shall consider ferromagnets with localized magnetic moments, in which the magnetic atoms are distributed at random (that is, without correlation) over the sites of a crystalline lattice. Depending on the type of potential, two opposite situations are possible.

1. Only nearest neighbors in the lattice interact. In this case, magnetic order exists only if the concentration x of magnetic atoms exceeds a critical value x_c (the percolation threshold); that is, there occurs in the system, along with a temperature phase transition when $x > x_c$, also a concentration phase transition at zero temperature.

2. There is a nonvanishing exchange interaction for an arbitrary distance between the magnetic atoms. In this case, ferromagnetism exists at an arbitrarily small concentration x . We shall suppose that the exchange potential falls off exponentially with distance: $V(r) \sim e^{-r/R}$.

Common to these systems is the decisive role of spatial fluctuations in the most interesting range of concentrations: in the first case, when the concentration x is close to the singular point, the percolation threshold x_c ; in the second system, when the mean distance between magnetic atoms is larger than the radius R of the exchange potential, and therefore the fluctuations of the exchange interaction are large. Naturally the magnetism of such systems differs strongly from the properties of ordinary ferromagnets; and for them, as for strongly disordered nonmagnetic systems, the methods of perturbation theory are inapplicable.⁷

Ferroelectrics in which some of the magnetic atoms have been replaced by nonmagnetic ones could become materials of the first type. Although at present only diluted ferri- and antiferromagnets have been obtained by this method,⁸ the diluted-ferroelectric model is the best studied theoretically, and it is for it that the basic concepts of the theory of disordered ferromagnetism have been successfully formulated and that certain general results have been obtained.

The classic example of a ferromagnet of the second type is dilute alloys of palladium with iron, cobalt, and manganese. The unusual properties of these alloys have been known for some time, but they remained understood for a long time because the spatial fluctuations of the exchange energy were not taken into account.

The theory of ferromagnetic alloys of the PdFe type is presented in the third section of the review. There, also, are presented the principal experimental results and their interpretation.¹⁾ We emphasize that dilute alloys of the PdFe type are so far the only real strongly disordered magnetic system whose properties can be understood fairly completely.

¹⁾ More detailed information about the experimental aspect of the matter is presented in the review by Nieuwenhuys⁹ published in 1975.

II. DILUTE FERROMAGNETS WITH NEAREST-NEIGHBOR INTERACTION

1. Concentration phase transitions. Basic concepts

We consider a system of localized spins whose interaction is described by the Ising or Heisenberg model:

$$\mathcal{H} = -\frac{1}{2} \sum_{i,j} V_{ij} [\alpha (S_i^z S_j^z + S_i^y S_j^y) + S_i^x S_j^x] - \mu H \sum_i S_i^z; \quad (1.1)$$

here \mathbf{s}_i is the spin localized at the site \mathbf{r}_i of the lattice, H is the external magnetic field, μ is the effective magneton, and $V_{ij} = V(\mathbf{r}_i - \mathbf{r}_j)$ is the energy of exchange interaction of nearest neighbors in the lattice. For $\alpha = 1$ the Hamiltonian (1.1) corresponds to the Heisenberg model, for $\alpha = 0$ to the Ising model.

The disorder in this system is most simply produced in two ways. First, it is possible to replace magnetic atoms by nonmagnetic ones, in a random manner, at the lattice sites. In this case $V_{ij} = V_0 p_i p_j$, where $p_i = 1$ if the site is occupied by a magnetic atom, $p_i = 0$ if by a nonmagnetic one. Second, it is possible to fracture bonds in a random manner; that is, to count certain V_{ij} as equal to zero. It is in principle possible to produce such a situation in ferromagnets with indirect exchange via nonmagnetic ions. Removal of the latter or replacement of them by ions that do not take part in indirect exchange corresponds to the breaking of a bond between spins. Accordingly, in (1.1) and everywhere hereafter the summation is assumed to be only over occupied sites or unbroken bonds.

At temperature zero, all spins coupled by exchange interaction are oriented parallel. If the concentration x of magnetic atoms or of unbroken bonds is small, then the magnetic atoms form mutually isolated clusters, whose dimensions do not change with increase of the volume of the crystal. We shall call such clusters finite. The mean magnetic moment of the whole crystal is in this case zero.

With increase of x , the characteristic dimension of the finite clusters increases; and at a certain critical concentration x_c , there appears a cluster the number of particles in which is proportional to the volume of the system. We shall hereafter call such a cluster infinite.

When $x > x_c$, the magnetic moment $M(x)$ of the crystal is nonzero and is equal to

$$M(x) = M(1) P(x), \quad (1.2)$$

where $M(1)$ is the moment of the ordered crystal, and where $P(x)$ is the concentration of magnetic atoms (per lattice site) that fall within the infinite cluster. Thus in the disordered ferromagnet at $T=0$, a phase transition from the paramagnetic state to the ferromagnetic occurs when the concentration $x = x_c$.

The problem of a concentration phase transition, i.e., of the appearance of an infinite cluster, is studied in percolation theory.¹⁰⁻¹³ In accordance with the two methods considered above for producing disorder in the lattice, it is customary to distinguish the "site problem" and the "bond problem." The critical concentration x_c is called the "percolation threshold." The values of x_c depend on the type of lattice and are different for

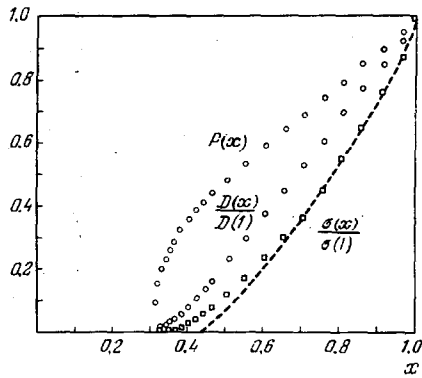


FIG. 1. Variation of the probability $P(x)$, the spin-wave stiffness coefficient $D(x)/D(1)$, and the conductivity $\sigma(x)/\sigma(1)$ with the concentration x of magnetic atoms, for a simple cubic lattice.^{11,20} The dashed curve is a calculation of the conductivity by the effective-medium theory.

the site problem and for the bond problem. They have been obtained as a result of computer experiments. For cubic lattices, in the bond problem $x_c = 0.243$ (bcc), 0.195 (fcc); in the site problem $x_c = 0.178$ (bcc), 0.120 (fcc). A detailed table giving x_c for various lattices is given in the review by Essam.¹³

The value of $P(x)$ varies from 0 at $x = x_c$ to 1 at $x = 1$. A graph of the function $P(x)$, obtained by computer calculations,¹¹ is shown in Fig. 1.

2. Properties of a ferromagnet far from the percolation threshold

a) *Spin waves in the Heisenberg model.* In the ground state, the spins of the magnetic atoms contained in the infinite cluster are parallel, and the saturation magnetic moment is determined by the number of atoms that fall within it. The deviation of the magnetic moment from saturation at finite temperatures is caused by magnetic excitations within the infinite cluster.

The question arises of the character of the elementary excitations in such a disordered system. In particular, it is not obvious *a priori* that the low-frequency excitations constitute a weakly attenuating hydrodynamic spin-wave mode, analogous to spin waves in an ordered ferromagnet. In general, the low-frequency magnetic excitations in a disordered ferromagnet might be localized, like electrons in the field of a random potential.⁷ We shall now show that at least in an isotropic Heisenberg ferromagnet, the low-frequency excitations actually constitute weakly attenuating spin waves with a quadratic dispersion law.

In fact, the equation of motion for the transverse spin component $S^+ = S^x + iS^y$ has the form

$$\frac{dS_m^+}{dt} = i \sum_l V_{lm} (S_l^+ - S_m^+). \quad (2.1)$$

The indices l and m identify sites occupied by magnetic atoms and belonging to the infinite cluster. The operator $\hat{V}\varphi = \sum_l V_{lm}(\varphi_l - \varphi_m)$ has an eigenfunction (normalized to unity) $\varphi_{01} = 1/\sqrt{P(x)N}$, corresponding to the eigenvalue $\omega = 0$; N is the total number of sites in the crystal. Therefore it is to be expected that the eigenfunctions of

the low-frequency excitations, which we shall describe by a "wave vector" q , can be represented in the form

$$\varphi_{qm} = u_{qm} e^{i(qr_m)}, \quad (2.2)$$

where u_{qm} can be expanded as a power series in q . Then the equation for the eigenvalues ω_q of the operator \hat{V} has the form¹⁴

$$\sum_m V_{ml} (u_{qm} - u_{ql} e^{iqx_{lm}}) = \omega_q u_{ql}; \quad (2.3)$$

here the x axis is directed along q , and $x_{lm} = x_l - x_m$. Taking into account that $\omega_0 = 0$, we get to the first order in q

$$\sum_l V_{ml} (u_{0l}^1 - u_{0l}^0 - i u_{0l} x_{lm}) = \omega_0^1 u_{0m}. \quad (2.4)$$

On summing (2.4) over m and interchanging m and l on the left side of the equation, we get

$$\omega_0^1 \sum_m u_{0m} = 0, \quad (2.5)$$

that is, $\omega_0^1 = 0$. To the second order in q , we have

$$\sum_l V_{ml} (u_{0m}^2 - u_{0l}^2 - 2i u_{0l} x_{lm} - u_{0l} x_{lm}^2) = \omega_0^2 u_{0m}. \quad (2.6)$$

On summing over m , interchanging m and l , and using (2.3), we get

$$\omega_0^2 = \frac{\sum_{m,l} V_{ml} (u_{0l}^2 u_{0m}^2 - |u_{0l}^2 - u_{0l}^1|^2)}{\sum_n u_{0n}^2}. \quad (2.7)$$

Consequently the spin-wave spectrum has the form

$$\omega_q = D(x) q^2 + O(q^3), \quad (2.8)$$

where $D(x) = \omega_0^2$ is a real, positive quantity.

It is evident from (2.8) that the damping of magnons is proportional to a higher power of q than is their energy.²⁾ We emphasize that this conclusion is based solely on the fact that the Heisenberg Hamiltonian conserves the total spin of the system; it does not depend on the specific form of the potential $V(r)$ or on the nature of the spatial distribution of impurities.

The further problem consists of the determination of the concentration dependence of the magnon "stiffness" D . Here, as was first shown by Kirkpatrick,¹⁷ it is convenient to use the relation that exists between the concentration dependences of $D(x)$ and of the conductivity $\sigma(x)$ of the equivalent lattice of resistors:

$$\frac{D(x)}{D(1)} = \frac{\sigma(x)}{P(x)\sigma(1)}. \quad (2.9)$$

The equivalent resistor lattice is constructed from the magnetic lattice as follows: with the magnetic bonds V_{ij} are associated conductances σ_{ij} , distributed according to the same rule as are the V_{ij} . The relation (2.9) can be derived¹¹ by use of the analogy between the right side of the equation of motion (2.1) and Kirchhoff's law for the equivalent resistor lattice:

$$\sum_m \sigma_{lm} (\varphi_l - \varphi_m) = 0, \quad (2.10)$$

where φ_l are the potentials at sites within the volume.

²⁾ Results of microscopic investigation of alloys with a small concentration of nonmagnetic atoms¹⁵ or with a large radius of interaction of the spins¹⁶ allow us to suppose that the damping of spin waves, in the alloys considered, is proportional to q^5 .

An alternative derivation of (2.9) is based on the analogy between (2.1) and the kinetic equation that describes the hopping conductivity of electrons for which the probability of a transition from site i to site j is V_{ij} .¹⁴

b) *The effective-medium method.* Far from the percolation threshold ($x > x_c$), most of the magnetic atoms are contained in the infinite cluster. If the number of nonmagnetic atoms $1 - x$ is not small, then the number of magnetic atoms in the finite clusters is comparable with the number of them in the infinite cluster, and a systematic analytical theory of these systems cannot be developed. Therefore such systems are investigated either by various interpolation methods or by numerical calculations on a computer.

Interpolation methods have been found useful in the calculation of the concentration dependence of the spin-wave stiffness $D(x)$. Simplest and most graphic is the effective-medium method, which is equivalent to the coherent-potential method.^{18,19} We turn now to an exposition of this method and an analysis of the results obtained with it.

In accordance with the relation (2.9), instead of calculating $D(x)$ it is sufficient to determine the concentration dependence of the conductivity $\sigma(x)$ of the equivalent resistor network.

The idea of the effective-medium method consists in replacement of the random quantities σ_{ij} by a certain mean conductance σ_m in such a way that the change of voltage $\Delta\varphi$ due to the deviation of the conductivity of a single bond from σ_m shall on the average be zero:

$$\int g(\sigma) \Delta\varphi(\sigma) d\sigma = 0; \quad (2.11)$$

here $g(\sigma)$ is the distribution function of the conductance σ . In particular, for the bond problem discussed above

$$g(\sigma) = x\delta(\sigma - \sigma_0) + (1 - x)\delta(\sigma). \quad (2.12)$$

The change of potential $\Delta\varphi(\sigma)$ is calculated differently for the bond problem and for the site problem.

In the first case, we have (Fig. 2) for a resistor lattice connected to an external circuit, in the fixed-current mode,

$$\sigma_m \frac{\Delta\varphi}{2} (z - 2) + \sigma_m \varphi_m = \sigma (\varphi_m - \Delta\varphi), \quad (2.13)$$

where z is the number of nearest neighbors; Fig. 2 shows schematically a plane lattice for which $z = 4$.

From (2.11) and (2.13) follows

$$\int g(\sigma) \frac{\sigma_m - \sigma}{\sigma + [(z/2) - 1]\sigma_m} d\sigma = 0. \quad (2.14)$$

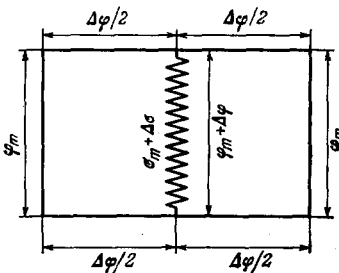


FIG. 2. Voltage drop near a conductance $\sigma_m + \Delta\sigma$.

On substituting (2.12) in (2.14), we find

$$\sigma_m = \sigma_0 \frac{x - x'_c}{1 - x'_c}, \quad (2.15)$$

where $x'_c = 2/z$ is the value of the critical concentration within the framework of the effective-medium theory.

For the site problem the calculation is somewhat more complicated, since removal of a single site corresponds to correlated fracture of z bonds. The result is analogous to (2.15) with replacement of $(z/2) - 1$ by $(2t_p - 1)^{-1}$,²⁰ where

$$t_p^{-1} = 1 - \frac{2}{Nz} \sum_{\mathbf{k}} \frac{\sin^2 k_x d}{1 - \gamma_{\mathbf{k}}}, \quad (2.16)$$

$$\gamma_{\mathbf{k}} = \frac{1}{z} \sum_{\mathbf{d}} e^{i(\mathbf{k}\mathbf{d})},$$

d is the lattice constant. According to Ref. 20, $t_p = 1.265$ (simple cubic), 1.19 (bcc), 1.15 (fcc).

Numerical calculations on a computer, by the Monte Carlo method,²⁰ have shown that formula (2.15) describes the concentration dependence of the conductivity well everywhere except for a narrow range of x near x_c (see Fig. 1). The inapplicability of the effective-medium method to a description of this critical range is natural, since this method is a variant of the theory of the self-consistent field. The concentration dependence of $D(x)$ follows from (2.15) by use of the relation (2.9). For small concentrations of the defects ($1 - x \ll 1$), the expression for $D(x)$ obtained by the effective-medium method agrees with the result of Izyumov.²¹

It must be especially noted that in disordered ferromagnets, spin waves determine the density of states only at low frequencies. At high frequencies, local excitations in the infinite and finite clusters also become important. The density of states of disordered two- and three-dimensional ferromagnets has been determined over the whole frequency range by numerical solution of the equations of motion (2.1) on a computer.^{22,23}

c) *High-temperature expansions.* The concentration dependence of the Curie temperature, $T_C(x)$, and the critical phenomena, for various models of disordered ferromagnets, have been studied for many years by the method of high-temperature expansions,²⁴⁻³⁰ which had been successfully applied to the calculation of T_C and of the critical indices in ordered ferromagnets.³¹ It was found that T_C decreases with x according to a law not too much different from a linear one, and that

$$\frac{d}{dx} \left(\frac{T_C(x)}{T_C(1)} \right)_{x=1} \begin{cases} = 1 & \text{(Ising model)}, \\ > 1 & \text{(Heisenberg model)}. \end{cases} \quad (2.17)$$

It is impossible to determine by this method the $T_C(x)$ relation for x close to x_c , since in this concentration range the series converge poorly. If we extrapolate the $T_C(x)$ data to the small- x range, we find that $T_C(x)$ vanishes at values of x very close to x_c . Then the intersection of the $T_C(x)$ curve with the axis of abscissas lies slightly to the left of x_c for the Ising model and to the right for the Heisenberg model. The critical indices $\gamma_T(x)$ of the susceptibility and $\nu_T(x)$ of the correlation radius increase with decrease of x (Fig. 3).^{27,28,30} The dependence of the critical indices on the concentration

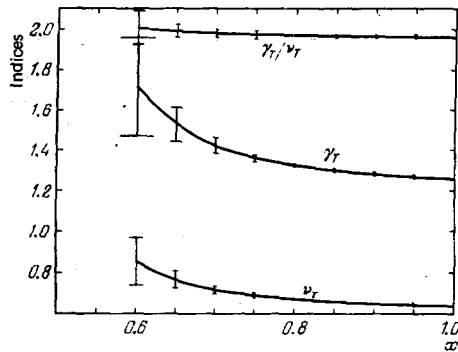


FIG. 3. Variation of the critical indices γ_T and ν_T and of their ratio γ_T/ν_T with the concentration x of magnetic sites, for a face-centered cubic lattice.³⁰

of impurities contradicts the universality hypothesis. At the same time, the ratio $\gamma_T(x)/\nu_T(x)$ is independent of x within the limits of accuracy of the calculations³⁰ (see Fig. 3); that is, the concentration dependence of the indices agrees with the hypothesis of weak universality proposed by Suzuki.³² The hypothesis advanced by Suzuki is that what is universal is the dependence of physical quantities not on $T - T_c$ but on the correlation radius ξ . Therefore what should vary with concentration is the ratio $\gamma_T(x)/\nu_T(x)$ and not these indices themselves.

3. The concentration range near the percolation threshold

a) *Concepts of scaling theory for a concentration phase transition.* As we have already mentioned, at $T=0$ there occurs in a system of magnetic atoms a concentration phase transition between paramagnet and ferromagnet. The concentration $P(x)$ of atoms in the infinite cluster plays the role of order parameter; as is seen from Fig. 1, the variation of $P(x)$ with the concentration of magnetic atoms is similar to the variation with temperature of the magnetization of a ferromagnet.

The magnetic susceptibility near the percolation threshold also behaves analogously to the susceptibility of a ferromagnet near the Curie point. The character of the variation of the susceptibility as $x - x_c$ can be understood as follows.

Let n_l be the number of clusters (per lattice site) containing l magnetic atoms. If the temperature is small in comparison with the exchange-interaction energy, then the spins within each cluster are oriented parallel. For the Ising model, which we shall consider in this subsection, the free energy of each cluster in an external magnetic field H , directed along the z axis, is $-T \ln(e^{\beta \mu H S^l} + e^{-\beta \mu H S^l})$, and the free energy of the whole system is

$$F = -T \sum_l n_l \ln(e^{\beta \mu H S^l} + e^{-\beta \mu H S^l}), \quad \beta = T^{-1}. \quad (3.1)$$

Hence the susceptibility is

$$\chi = -\frac{\partial^2 F}{\partial H^2} \Big|_{H=0} = \frac{\mu^2 S^2}{T} \sum_l n_l l^2. \quad (3.2)$$

The quantity $\bar{n} = \sum_l n_l l^2$ represents the mean number of particles in finite clusters.

At concentration less than the critical, all the mag-

netic atoms are in finite clusters, so that $\sum_l n_l l = x$. With approach to the critical concentration, merging of the finite clusters occurs; their mean size \bar{n} , and with it also the susceptibility (3.2), increase without bound.

On the basis of the analogy between phase transitions of the second kind and the appearance of an infinite cluster in percolation theory, in recent years a scaling hypothesis has been formulated³³⁻³⁵ for the description of a concentration phase transition, and it has subsequently been corroborated by numerous computer calculations³³⁻³⁷ and calculations by the renormalization-group method.³⁸⁻⁴⁰

Within the framework of scaling theory, the singular part of the free energy (3.1) can be represented in the form³⁴

$$F_{\text{sing}}(x, H, T) = -T |x - x_c|^{2\Delta - \nu \Phi} \left(\frac{\mu H}{T |x - x_c|^\Delta} \right), \quad (3.3)$$

where Δ is a "gap" index.

In a weak magnetic field, the part of the free energy that is due to the field should be proportional to H^2 in the paramagnetic phase and to H in the ferromagnetic phase. This determines the variation of $\Phi(z)$ with z at small z .

Hence it follows that the susceptibility is

$$\chi \sim |x - x_c|^{-\nu}, \quad (3.4)$$

and that the spontaneous moment is

$$M \sim (x - x_c)^\beta, \quad \beta = \Delta - \nu. \quad (3.5)$$

Along with the order parameter and the susceptibility, an important characteristic of the system near the phase-transition point is the correlation radius L , which in the paramagnetic phase ($x < x_c$) describes the distance over which the spins may be considered coupled.

The degree of coupling of the system can be studied by means of the spin correlation function

$$G(i, j) = \langle S_i^z S_j^z \rangle - \langle S_i^z \rangle \langle S_j^z \rangle, \quad (3.6)$$

where the angular brackets denote a thermodynamic average. It is easily seen that in zero magnetic field

$$G(i, j) = \langle S_i^z S_j^z \rangle, \quad (3.7)$$

if i and j belong to the same finite cluster, while $G(i, j) = 0$ in all other cases. In scaling theory, the correlation radius is the only characteristic quantity with the dimensions of length; therefore the correlator $G(r) = \overline{G(i, j)}$, where the bar denotes a configurational average, can be written in the form

$$G(r) \sim (x - x_c)^{-\nu + 3\nu} G_0 \left(\frac{r}{L} \right). \quad (3.8)$$

Since the homogeneous susceptibility $\chi \sim \int G(r) d^3 r$, it follows from (3.4) and (3.8) that

$$L \sim (x - x_c)^{-\nu}. \quad (3.9)$$

As is seen directly from (3.7), the correlation radius L for $x < x_c$ describes the geometric size of a finite cluster. For $x > x_c$, L , as the only scale with the dimensions of length, determines also the characteristic size of an inhomogeneity of the infinite cluster.

According to Refs. 35-37, $\gamma=1.8$, $\beta=0.39$, and $\nu=0.85$. Above, we have always assumed that the spins in each cluster are oriented parallel. Therefore the quantities F_{sing} , χ , and G introduced in this subsection, and also the critical indices, essentially describe the topology of a disordered system of magnetic atoms.

By applying these concepts, we shall in the next subsection study the thermodynamics of a Heisenberg ferromagnet for $x \approx x_c$.

b) *The thermodynamics of a Heisenberg ferromagnet.* In the Heisenberg model, the thermodynamics of the system is determined by the fluctuations of the transverse components of the spin. We shall calculate the density of states of the corresponding excitations.^{41,42} The density of states $\rho_{\text{inf}}(\omega)$ of the low-frequency excitations in the infinite cluster is determined by spin waves; that is,

$$\rho_{\text{inf}}(\omega) = \frac{1}{4\pi^2} \frac{\omega^{1/2}}{D^{3/2}}. \quad (3.10)$$

According to the results of computer calculations, σ near the threshold varies with $x - x_c$ according to a power law,^{11,12,20,43}

$$\sigma \sim (x - x_c)^t, \quad t = 1.72, \quad (3.11)$$

so that $D \sim (x - x_c)^{t-\beta}$. Consequently

$$\rho_{\text{inf}} \sim \omega^{1/2} (x - x_c)^{-(3/2)(t-\beta)} \sim (x - x_c)^{-2.0}.$$

The quadratic law is correct as long as the wavelength of the magnons is larger than the correlation radius, i.e., if

$$\omega < \omega_0 = DL^{-2} \sim (x - x_c)^{t+2\nu-\beta}, \quad (3.12)$$

since it is only in this case that the wave "averages" the spatial fluctuations that occur in the structure of the infinite cluster and that, therefore, its damping is small.

In order to determine $\rho_{\text{inf}}(\omega)$ for $\omega > \omega_0$, we shall use the method, known from the theory of phase transitions, of matching a long-wave hydrodynamic mode with a so-called critical mode, for which the wavelength is less than the correlation radius.⁴⁴ Here the important fact for us will be that, because the characteristic scale of the critical excitations is less than L , their energy and their density of states per magnetic atom $\rho_{\text{inf}}(\omega)$ must depend only on geometric properties of the infinite cluster at distances less than L . But the geometric characteristics of the system at such distances are independent of $x - x_c$; the parameter $x - x_c$ determines only the topology of the system at distances larger than L .

Thus $\tilde{\rho}_{\text{inf}}(\omega)$ for $\omega \gg \omega_0$ is independent of $x - x_c$; that is, $\tilde{\rho}_{\text{inf}}(\omega) \sim f(\omega/V_0)$. Consequently

$$\rho_{\text{inf}}(\omega) \sim P(x) f\left(\frac{\omega}{V_0}\right). \quad (3.13)$$

On matching, at $\omega = \omega_0$, the expressions for $\rho_{\text{inf}}(\omega)$ determined by formulas (3.10) and (3.13), we get

$$\rho_{\text{inf}}(\omega_0) \sim \frac{\omega_0^{1/2}}{D^{3/2}} \sim \omega_0^{(v+\beta-t)/(t+2\nu-\beta)} \sim P f\left(\frac{\omega_0}{V_0}\right). \quad (3.14)$$

Since $P(x) \sim (x - x_c)^\beta$, it follows from (3.14) that $f(y) \sim y^{-\xi}$, where $\xi = (t - \nu)/(t + 2\nu - \beta)$. Therefore the density of states for $\omega \gg \omega_0$ is

$$\rho_{\text{inf}}(\omega) = \frac{AP(x)}{V_0 \Omega_0} \left(\frac{V_0}{\omega}\right)^\xi, \quad (3.15)$$

where A is an unknown numerical coefficient and Ω_0 is the volume of the elementary cell. From the values of the indices given above, it follows that $\xi = 0.27$.

Thus the density of states has a maximum at energies of order ω_0 and decreases quite slowly with increase of the energy for $\omega \gg \omega_0$. The principal contribution to the normalization integral comes from energies of order V_0 ; the number of states in the spin-wave range is proportional to $L^{-3} \sim (x - x_c)^{2.6}$, i.e. very small in comparison with the total number of states. Knowing the density of states, we can calculate in the usual way the deviation of the magnetization from saturation:

$$\Delta M = M_0 - \langle M^2 \rangle = \mu \int \frac{\rho_{\text{inf}}(\omega)}{e^{\omega/T} - 1} d\omega. \quad (3.16)$$

In the spin-wave range $T \ll \omega_0$, we have

$$\Delta M \sim \left(\frac{T}{D}\right)^{3/2} \sim (x - x_c)^{-2.9}. \quad (3.17)$$

The deviation of the magnetization from saturation is small over the whole spin-wave range, up to $\omega \sim \omega_0$. In fact, at $\omega \sim \omega_0$

$$\frac{\Delta M}{M_0} \sim \frac{1}{PL^3} \sim (x - x_c)^{3\nu-\beta} = (x - x_c)^{2.2} \ll 1. \quad (3.18)$$

Hence it is evident that the Curie temperature is much higher [according to the parameter $(x - x_c)^{-1}$] than the energy ω_0 at which the spin-wave part of the spectrum ends. This deviation from the properties of an ordered ferromagnet, in which these two characteristic quantities coincide, is due to the already mentioned smallness of the number of states in the spin-wave range.

If $T \gg \omega_0$, then the principal contribution to the integral (3.16) comes from $\omega \sim \omega_0$; therefore

$$\Delta M \sim \rho_{\text{inf}}(\omega_0) T. \quad (3.19)$$

Since $\Delta M/M_0 \sim 1$ at a temperature of order T_c , it follows that $T_c \sim P^{-1}(x)_{\text{inf}}(\omega_0)$, and by use of (3.15) we find³⁾

$$T_c = BV_0 \left(\frac{x - x_c}{x_c}\right)^{t-\nu}; \quad (3.20)$$

here B is an unknown numerical coefficient. The exponent in this formula is $t - \nu \approx 0.9$.

The vanishing of T_c when $x \rightarrow x_c$ is due to the fact that the magnetic order is destroyed by thermodynamic fluctuations with energy of order ω_0 and characteristic scale L , and their density of states increases rapidly on approach to the percolation threshold.

Since $T_c/\omega_0 \sim (x - x_c)^{\beta-3\nu} \gg 1$, there is a broad range of temperatures in which $\Delta M \sim T$.

The specific heat C_M at $T \ll \omega_0$ is determined, like ΔM , by spin waves; that is, $C_M \sim (T/D)^{3/2}$. At $T \gg \omega_0$, C_M is determined by excitations with energy $\omega \gg \omega_0$, localized principally in finite clusters. The specific heat is proportional to⁴²

$$C_M \sim \left(\frac{T}{V_0}\right)^{t-\xi'}, \quad \xi' = \frac{t-\beta-\nu}{t+2\nu-\beta}. \quad (3.21)$$

Since the specific heat at $T \gg \omega_0$ is determined by exci-

³⁾Qualitative considerations leading to (3.20) were first enunciated by Shender and Shklovskii.⁴⁵ This relation was also obtained by Stauffer.⁴⁶ We remark that the density of states that he determined for $\omega > \omega_0$ is incorrect, since he omitted the factor $P(x)$ in the matching equation (3.13).

tations in finite clusters, the expression (3.21) is correct both for $x < x_c$ and for $x > x_c$. In other words, the specific heat is insensitive to the concentration phase transition (here we are not discussing critical anomalies near T_c). At $T \approx V_0$ the specific heat reaches a maximum.

4. The Bethe lattice

A system of spins randomly distributed on a Bethe lattice constitutes an exactly solvable model of a disordered ferromagnet. Many results that in real models have no completely rigorous basis or have been obtained as a result of numerical calculations on a computer can be obtained rigorously and comparatively easily for the Bethe lattice.

In the Bethe lattice, z bonds go out from each site, producing nonintersecting branches (Fig. 4). We shall assume that magnetic atoms are randomly distributed on the sites of a Bethe lattice, that their concentration (per lattice site) is x , and that their interaction is described by the Hamiltonian (2.1). We shall find an expression for the mean magnetic moment of the lattice at $T \ll V_0$. Let R be the probability that a branch going out from a certain fixed site (site 0 in Fig. 4) is finite; that is, that the magnetic atoms in it do not form an infinite cluster. A branch can be finite if the site next to the zeroth site in the given branch (site 1) is unoccupied (probability $1 - x$) or if this site is occupied, but the $z - 1$ branches going out from it are finite (probability xR^{z-1}). Consequently, R satisfies the equation⁴⁷⁻⁵⁰

$$R = 1 - x + xR^{z-1}. \quad (4.1)$$

The simplicity of this equation is due to the basic property of the Bethe lattice, nonintersection of the branches.

The mean moment is

$$M(x) \sim P(x) = 1 - R^z. \quad (4.2)$$

Equation (4.1) always has the trivial solution $R = 1$, corresponding to $M = 0$. Nontrivial solutions $R \neq 1$ appear when $x = x_c = (z - 1)^{-1}$; x_c is the percolation threshold in the Bethe lattice. Near the threshold, where $(x - x_c)/x_c \ll 1$, we have from (4.1)

$$\frac{M(x)}{M(x_c)} = \frac{2z}{z-2} \frac{x - x_c}{x_c}. \quad (4.3)$$

Thus for the Bethe lattice the critical index is $\beta = 1$.

The magnetic moment in an external field H at $T \ll T_c$ can be found from the expression (4.1) for the free

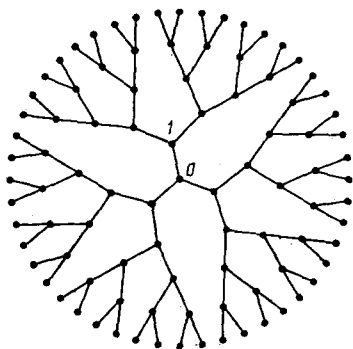


FIG. 4. A Bethe lattice with $z = 3$.⁴⁷

energy. Restricting ourselves for simplicity to spin $S = 1/2$, we have

$$\frac{M(x, H, T)}{M(x)} = P(x) + \sum_l n_l \frac{1 - \Lambda^l}{1 + \Lambda^l}, \quad (4.4)$$

where $\Lambda = e^{-\mu H / T}$. For the quantity

$$\Psi^z(\Lambda, x) = \frac{1}{\Lambda} \sum_l n_l \Lambda^l \quad (4.5)$$

one can derive an equation analogous to (4.1)⁵⁰:

$$\Psi^z(\Lambda, x) = 1 - x + x\Lambda\Psi^{z-1}. \quad (4.6)$$

In a weak magnetic field, near the percolation threshold, $\Lambda \approx 1$ and $\Psi^z(\Lambda, x) \approx 1$, so that equation (4.6) is easily solved, and the denominator in (4.4) can be replaced by a field-independent number of order unity. As a result we get

$$M(x_c, H, T) = \sqrt{1 - \Lambda} \sim \sqrt{H}. \quad (4.7)$$

The coefficient of proportionality was calculated in Ref. 49. It follows from (4.7) that in the Bethe lattice the index δ is 2. For the Bethe lattice it is also possible to investigate the properties of the temperature phase transition for any concentration $x > x_c$. In particular, Harris,⁵¹ Bell,⁵² and Young⁴⁹ have shown that for the Ising model with spin $1/2$

$$\text{th} \frac{2V_0}{T_c} = \frac{x_c}{x}. \quad (4.8)$$

Spin waves in the Bethe lattice were first studied by Stinchcombe⁴⁷ by use of the relation (2.9). He determined the conductivity by fixing the difference of potential between the origin (point 0 in Fig. 4) and the boundary. It was found that for the Bethe lattice, the effective-medium method is correct for all values of the parameter z^{-1} except a region near the threshold, for which the conductivity is proportional to $(x - x_c)^2$. De Gennes pointed out⁵³ that the macroscopic conductivity should be determined for a fixed external field in the whole specimen. Then near the threshold, the conductivity varies according to the law⁵³

$$\sigma \sim (x - x_c)^3, \quad (4.9)$$

that is, for the Bethe lattice the index t is 3. It follows from (2.9), (4.3), and (4.9) that $D \sim (x - x_c)^2$. We see that the exact results for the Bethe lattice agree completely with the scaling hypothesis near the percolation threshold.

III. IMPURITY FERROMAGNETISM OF STRONGLY PARAMAGNETIC METALS

5. Model of an impurity ferromagnet

Early in the 1960's, a striking phenomenon was discovered by Crangle⁴ and by Bozorth and coworkers⁵: the paramagnetic metal palladium becomes ferromagnetic when very slightly alloyed with iron or cobalt. It was later shown that a spontaneous moment occurs even at concentrations x of iron and cobalt of order 10^{-4} ,^{54,55} and also in alloys of palladium with manganese,⁵⁶ of platinum with iron and cobalt,⁵⁷ and of Ni_3Ga with iron.^{58,59}

The effective magnetic moment per magnetic impurity was found to be anomalously large: in the alloys

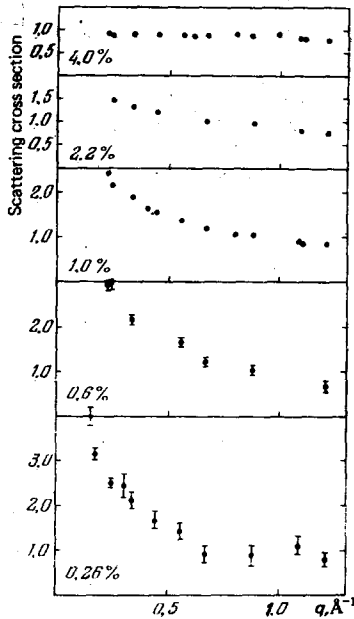


FIG. 5. Cross section for diffuse magnetic scattering as a function of the scattering vector $q = 4\pi \sin(\theta/\lambda)$ (2θ is the scattering angle), for PdFe alloys.⁶⁰ The iron content is indicated on the figure.

$\text{Pd}_{1-x}\text{Fe}_x$, it was $12\mu_B$ at $x=0.0028$.⁵⁷ With increase of the degree of alloying, the magnetic moment decreases; at concentration 10 at. % iron in Pd, it is $6\mu_B$. The large value of the magnetic moment per impurity in palladium alloys is due to the cloud of polarized d electrons of the matrix, surrounding the impurity spin. The existence of such clouds in dilute alloys of palladium with iron and cobalt has been confirmed by neutron-diffraction measurements of Low, Holden, and Hicks.⁶⁰ These authors studied the cross section for diffuse scattering of neutrons in palladium alloys, which is proportional to the magnetic form factor of the localized moments. Figure 5 shows the relation that they obtained between the scattering cross section and the momentum transferred. In dilute alloys, the cross section has a sharp peak for forward scattering; this indicates a slow falling off of the polarization with increase of distance from the impurity. With increase of the impurity concentration, the peak in the forward scattering is lowered; and at 4 at. % concentration of iron or cobalt, the scattering cross section is completely independent of the momentum transferred; that is, the alloy is magnetized almost uniformly.

The occurrence of impurity ferromagnetism of paramagnetic metals is caused by indirect interaction of the impurity spins through strongly correlated electrons in the narrow d band.^{61,62}

The interaction of an impurity spin S_0 , localized at lattice point 0, with the spins $\sigma/2$ of the band electrons of the matrix is described by the Hamiltonian

$$\mathcal{H}_J = -J(S_0 \sigma_0). \quad (5.1)$$

This interaction leads to polarization of the electrons of the matrix near the impurity. The moment density of the polarized electron cloud, to the first order in the parameter $JN(\epsilon_F)$, where $N(\epsilon_F)$ is the density of elec-

tronic states on the Fermi surface, is

$$m(r) = S_0 \frac{2\Omega_0 J}{g\mu_B} \int \chi(k) e^{ikr} \frac{d^3k}{(2\pi)^3}, \quad (5.2)$$

where g_e is the g factor of the d electrons and where μ_B is the Bohr magneton.

According to Landau's theory of the Fermi surface, the homogeneous static susceptibility is⁶³

$$\chi(0) = \frac{\chi_p}{1+\Gamma}, \quad (5.3)$$

where χ_p is the Pauli susceptibility of free electrons, and where Γ is the exchange scattering amplitude of the electrons.

It is evident from (5.3) that the condition for stability of the paramagnetic state is the inequality $1+\Gamma > 0$. Within the framework of the Hartree-Fock approximation, this corresponds to Stoner's criterion. If $1 \gg 1 + \Gamma > 0$, then $\chi(0) \gg \chi_p$ (a "strong paramagnet"). This is precisely the situation in the paramagnetic metals enumerated above, in which a slight impurity of paramagnetic atoms causes a transition to a ferromagnetic state. For example, in palladium $1+\Gamma \approx 0.1$,^{9,64} and in Ni_3Ga $1+\Gamma \approx 0.03$.⁵⁹ Having such metals in mind hereafter, we can express $\chi(k)$ at small k in the form⁶⁵

$$\chi(k) = \frac{\chi_p}{1+\Gamma+a^2k^2}, \quad (5.4)$$

where $a \approx k_F^{-1}$, k_F is the Fermi momentum of the electrons. At large momenta, $k \gg a^{-1}$, electronic correlation does not affect the susceptibility appreciably, so that $\chi(k)$ coincides with the free-electron susceptibility.

Thus $m(r)$ can be represented as the sum of two parts, $m_1(r) + m_2(r)$; $m_1(r)$ is due to the contribution of small k to the integral (5.2) and, according to (5.4), falls off with distance from the impurity like a screened Coulomb potential:

$$m_1(r) = g\mu_B S_0 \frac{\Omega_0 J N(\epsilon_F)}{4\pi a^2 r} e^{-r/R}, \quad R = \frac{a}{\sqrt{1+\Gamma}} \gg a. \quad (5.5)$$

According to data of the papers of Low, Holden, and Hicks cited above, for PdFe and PdCo alloys the radius R is, within the limits of accuracy of the experiment, 3 to 5 Å. The moment $m_2(r)$ is due to the contribution of large k and is determined by the usual RKKY (Ruderman-Kittel-Kasuya-Yosida) formula⁶⁶; that is, for $r \gg k_F^{-1}$

$$m_2(r) = g\mu_B S_0 \frac{\Omega_0 k_F^3}{16\pi^3} \frac{J}{e_F} \frac{\cos(2k_F r)}{(k_F r)^3}. \quad (5.6)$$

Since $R^2/a^2 = (1+\Gamma)^{-1} \gg 1$, at not too large distances $m_1(r) \gg m_2(r)$.

Thus an impurity spin is surrounded by a cloud of electrons polarized, depending on the sign of J , along or opposite to the moment of the impurity. The oscillations of the electron moment that are usual for weakly paramagnetic metals begin only at large r , where the polarization itself is already small. Therefore the principal contribution to the total magnetic moment of the polarized electron cloud comes from $m_1(r)$. On integrating (5.2) over r , we get for the effective moment of the impurity

$$\mu = g_i \mu_B S \left(1 + \frac{g_e}{g_i} \frac{JN(\epsilon_F)}{1+\Gamma} \right), \quad (5.7)$$

where g_i is the g factor of the impurity.

Because of the smallness of the denominator $1 + \Gamma$ in (5.7), the effective magneton of the impurity considerably exceeds μ_B if the exchange integral is not too small.

Overlap of the polarized electron clouds surrounding an impurity leads to indirect exchange interaction between impurities. The potential of the indirect exchange is

$$V(r) = \frac{4J^2}{g_i^2 \mu_B^2} \chi(r). \quad (5.8)$$

Like the moment $m(r)$, the potential $V(r)$ can be expressed as the sum of two parts: a ferromagnetic,

$$V_\phi(r) = U_0 \frac{R}{r} e^{-r/R}, \quad U_0 = J^2 N(e_F) \frac{\Omega_0}{2\pi a^3 R}, \quad (5.9)$$

and an oscillatory,

$$V_{\text{RKKY}} = V_1 \frac{\cos(2k_F r)}{(k_F r)^3}, \quad V_1 = \frac{J^2 N(e_F) k_F^3 \Omega_0}{8\pi}, \quad (5.10)$$

at not too large distances, $V_f(r) \gg V_{\text{RKKY}}$. Therefore the impurity spins are ordered ferromagnetically. Only at very small concentrations of the impurity (in $\text{Pd}_{1-x}\text{Fe}_x$ apparently at $x < 10^{-4}$), when the mean distance r_c between impurities is sufficiently large, does the oscillatory potential V_{RKKY} exceed the ferromagnetic one, so that a spin glass is formed in the alloy in analogy to alloys of the CuMn type.¹⁻³ Hereafter, unless there is an explicit statement to the contrary, we shall have in mind magnetic-impurity concentrations for which the effect of the potential (5.10) may be neglected.

In the ferromagnetic phase, the magnetic moment of the localized spins magnetically biases the electrons of the matrix. The mean polarization of the electrons is of the order of $J\chi(0)x$. The treatment carried out above is obviously correct as long as the mean polarization of the electrons is small, i.e.,

$$J\chi(0)x \sim \frac{JN(e_F)x}{1+\Gamma} \ll 1. \quad (5.11)$$

As has already been mentioned, the polarization becomes uniform at $x \approx 0.04$ (see Fig. 5). When $x \leq 0.02$, the polarization of the electrons is highly nonuniform; that is, the criterion (5.11) is satisfied.

Thus in the case of small concentrations [in the sense of the inequality (5.11)], with which we shall be concerned hereafter, the magnetic properties of the alloy can be described by the following model.

Over the sites of the lattice are randomly distributed magnetic atoms with a concentration $x \ll 1$. Their exchange interaction is described by the Heisenberg Hamiltonian (1.1) with the potential (5.9).

In the following sections, the properties of such a disordered ferromagnet will be investigated in detail; the concentration dependence of the Curie temperature will be found; and the spectrum of temperature excitations and the temperature and concentration dependence of thermodynamic quantities will be studied.

In parallel with the exposition of the theory, we shall present the results of experimental investigations of the properties of alloys of palladium with iron, cobalt,

and manganese impurities and shall show that the magnetism of alloys of the PdFe type is well described within the framework of this model.

6. Concentration dependence of the Curie temperature

The properties of a magnetic system described by the model formulated above are completely determined by the ratio of two characteristic lengths: the radius R of the potential and the mean distance $r_c \approx n^{-1/3}$ between impurities (n is the number of spins per unit volume). If $R \gg r_c$, then a given spin on the average interacts effectively with a large number of magnetic atoms $\nu_R = (4/3)\pi n R^3 \gg 1$. Therefore the energy of exchange interaction of each spin with its surroundings depends only slightly on the specific configuration. In this case, of course, T_C is proportional to the mean exchange-interaction energy; that is, $T_C \sim x$.¹⁶

An entirely different situation arises when $R \ll r_c$, i.e., $\nu_R \ll 1$. This is the inequality that practically always holds in the alloys under consideration when the criterion (5.11) is satisfied. Now the energy of exchange interaction of a given spin with the other spins depends very strongly on the specific configuration. In fact, for the majority of spins the nearest neighbor is located at a distance of the order of r_c . It is sufficient to change the distance between them by an amount Δr such that $R < \Delta r \ll r_c$ in order that the interaction energy shall change exponentially strongly. Thus even insignificant fluctuations in the location of the spins lead to a substantial change of the exchange-interaction energy.

The concentration dependence of T_C for $R \ll r_c$ was first found in one of our papers.⁶⁷ Here we shall find this dependence by use of concepts of percolation theory.⁶⁸

Since the characteristic distances of the problem considerably exceed R , the exponential plays the principal role in (5.9). Therefore we shall for simplicity at first omit the factor r^{-1} and find T_C for a system of randomly distributed spins whose exchange interaction is

$$V(r) = V_0 e^{-r/R}. \quad (6.1)$$

We introduce a length $r(T)$ defined by the equation $V(r) = T$; that is,

$$r(T) = R \ln \frac{V_0}{T}. \quad (6.2)$$

Because of the exponential variation of the exchange energy with r , all spins the distance between which is less than $r(T)$ may be considered, at the given temperature, to be oriented parallel. Spins distant from each other by a distance $r > r(T)$ may be considered to be uncorrelated. In other words, all bonds divide into two groups: rigid [$r < r(T)$] and broken [$r > r(T)$]. The rigid bonds join magnetic atoms into clusters, in each of which the spins are oriented parallel. The system is ferromagnetic if connected spins form an infinite cluster. Thus the determination of the Curie temperature reduces to the following geometric problem:

In a system of randomly distributed points, all bonds between points separated from each other by a distance $r < r_{\text{max}}$ are switched on. When $r_{\text{max}} \ll n^{-1/3}$, the system

contains only finite clusters of coupled atoms. When $r \gg n^{-1/3}$, almost all atoms are coupled into an infinite cluster. At what value of r_{\max} does the infinite cluster first appear?

This is one of the problems considered in percolation theory, the so-called continuum problem, which has been solved in many papers (a detailed bibliography has been given¹²). The critical value of r_{\max} is⁶⁹

$$r_0 = (0.87 \pm 0.01) n^{-1/3}. \quad (6.3)$$

At high temperatures, $r(T) < r_0$; that is, the system is paramagnetic. The temperature of the ferromagnetic transition is determined by the condition $r(T) = r_0$, i.e.,

$$T_C = V_C \exp\left(-\frac{0.87}{Rn^{1/3}}\right), \quad (6.4)$$

where $V_C \approx V_0 S^2$. The Curie temperature is of the order of the interaction energy of spins located at a distance of the order of the mean distance (but it is not equal to the mean interaction energy, which is proportional to n). Therefore allowance for the pre-exponential factor in the interaction potential gives

$$T_C = U_C \frac{R}{r_0} \exp\left(-\frac{0.87}{Rn^{1/3}}\right), \quad (6.5)$$

where $U_C \approx U_0 S^2$.

The Curie temperature of alloys of palladium with iron, cobalt, and manganese has been measured by various methods. A summary of most of the results has been given in Ref. 9. We also point out several other references⁷⁰⁻⁷². In the analysis of these data, it must be kept in mind that most classical methods of determining T_C are unsuitable for a disordered ferromagnet. Thus hyperfine splitting of a Mössbauer line requires only magnetic ordering in large finite clusters, which exist also when $T > T_C$. The maximum in the specific heat may be shifted to temperatures below T_C , since excitations in finite clusters make a significant contribution to the specific heat. And finally, as will be shown in Section 11, the paramagnetic Curie temperature in this case is parametrically larger than T_C . Thus data on the magnetization are the most reliable, if the appropriate measurements are made in a weak magnetic field. These data, and also results of neutron-diffraction investigations, are presented in Table I. The table also includes values of T_C obtained from the break in the electrical resistivity; these are close to the T_C values found from the magnetization.

TABLE I. Curie temperature of palladium alloys

Pd _{1-x} Fe _x		Pd _{1-x} Co _x ^{70, 71}		Pd _{1-x} Mn _x	
x, at.%	T _C , °K	x, at.%	T _C , °K	x, at.%	T _C , °K
0.03	0.115 ⁸⁴	0.1	3.3 ± 0.5	0.15	0.22 ⁹
0.043	0.2 ⁸⁴	0.35	12.5 ± 1	0.2	0.39 ⁹
0.06	0.41 ⁸⁴			0.31	0.76 ⁹
		0.5	24 ± 3	0.4	1.03 ⁹
0.15	2.10 ⁷⁹	0.6	28 ± 3	0.45	1.41 ⁷⁶
0.16	2.8 ⁷⁴	0.7	38 ± 3	0.49	1.55 ⁷⁷
0.23	5.4 ⁷⁴	1.1	60 ± 3	0.5	1.44 ⁹
0.28	9.5 ⁸⁷			0.7	2.48 ⁹
0.41	13.0 ⁷⁴	1.6	88 ± 3	0.96	3.45 ⁷⁷
0.50	18.0 ⁷²			1.0	3.41 ⁷⁸
0.53	23.0 ⁸⁷			1.05	3.9 ⁷⁹
0.54	20.1 ⁷⁴			1.2	4.9 ⁷⁶
0.78	32.6 ⁷⁴			1.3	4.54 ⁷⁷
1.0	39 ⁷⁵			1.8	7.5 ⁷⁶
1.1	36 ⁷³				

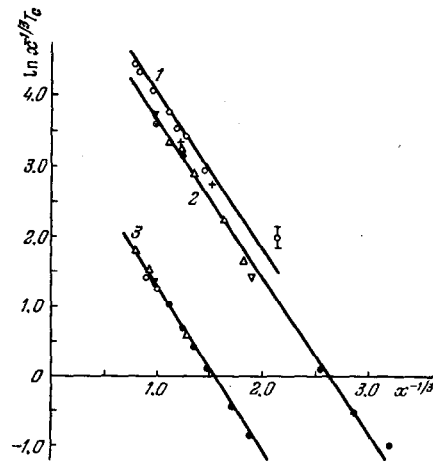


FIG. 6. Concentration dependence of the Curie temperature of alloys. 1, PdCo: O, experimental data from Refs. 70 and 71. 2, PdFe: ●, from Ref. 54; ∇, Ref. 73; Δ, Ref. 74; +, Ref. 7; ○, Ref. 72; √, Ref. 75. 3, PdMn: ●, from Ref. 9; Δ, Ref. 76; ○, Ref. 77; √, Ref. 79.

Over a number of years it has been noted in many experimental papers⁹ that the concentration dependence of T_C is nonlinear: T_C increases with x considerably faster than linearly. At the same time, in theoretical papers on this subject^{80, 81} a linear relation has been obtained. This is due to the fact that the cited authors neglected spatial fluctuations of the molecular field that acts on the localized spins. Therefore the Curie temperature obtained was equal to the mean interaction energy of localized spins, which is determined by pairs of spins located close together (at a distance $r \approx R \ll r_c$) and is proportional to x . It is clear, however, that the interaction in such pairs has no relation to the appearance of a macroscopic moment.

Figure 6 shows the variation of $\ln[x^{-1/3} T_C]$ with $x^{-1/3}$ for Pd(Fe, Co, Mn) alloys. In all three cases, the relation (6.5) is well satisfied over a broad range of concentrations and temperatures. All three straight lines have, as they should, the same slope. The value of the range R of the potential determined by the slope of these straight lines was found to be 4.3 Å; this agrees well with the value $R = 3$ to 5 Å obtained from neutron-diffraction measurements.⁸⁰ By extrapolating the straight lines in Fig. 6 to $x^{-1/3} = 0$, we get $U_0 = 800^\circ\text{K}$ for PdFe and 80°K for PdMn.

7. Spin waves

a) *The stiffness coefficient of spin waves.* In order to determine the concentration dependence of the stiffness D at small concentrations of the magnetic atoms, it is convenient to use the relation between D and the conductivity σ of the equivalent resistor network, which was discussed in Section 2. Generalization of formula (2.9) to a disordered ferromagnet in which the spin-interaction potential varies with distance r according to the law (5.9) requires replacement of the probability $P(x)$ by the number of magnetic atoms whose coupling energy exceeds the energy of a spin wave. As a result, in the limit of low frequencies⁸²

$$D \sim \frac{\sigma}{n}; \quad (7.1)$$

here σ may be regarded as the electron conductivity in a system of randomly located centers, in which the probability of an electron's jumping between centers is determined by formula (5.9). Its variation with concentration is known^{83,84,4)}

$$\sigma \sim r_0^{-(2+\nu)} \exp\left(-\frac{0.87}{Rn^{1/3}}\right), \quad (7.2)$$

where ν is the index of the correlation radius. We recall that $r_0 = 0.87 n^{-1/3}$.

From (7.1) and (7.2) it follows⁸² that

$$D \sim n^{-(1+\nu)S} \exp\left(-\frac{0.87}{Rn^{1/3}}\right). \quad (7.3)$$

The chief concentration dependence of D is determined by the exponential factor in (7.3), which (to within a factor r_0^{-1}) coincides with the interaction energy of spins separated by a distance $r_0 + R$. This seems quite natural, since the infinite cluster in which the spin waves are propagated exists only if bonds with length greater than or equal to r_0 are switched on. Spins in the infinite cluster that are separated by a distance less than r_0 , so that the bond energy is exponentially larger than $\exp(-0.87/Rn^{1/3})$, oscillate in phase with each other and therefore make no contribution to D . Obviously no role is played, either, by weakly coupled spins separated from their nearest neighbors by a distance much greater than r_0 . Thus the spin waves propagate as if, in the infinite cluster, only bonds with lengths $r_1 = r_0 \pm R$ were switched on. Therefore weakly attenuated spin waves exist if their length is larger than a characteristic distance determined by the scale of the spatial inhomogeneities of this cluster. Since $|r_1 - r_0| \ll r_0$, this scale is the correlation radius $L_R = r_0(r_0/|r_1 - r_0|)^\nu$. Consequently, the weakly attenuating spin waves are those with lengths $\lambda \geq L \sim r_0(r_0/R)^\nu$. The maximum spin-wave energy is

$$\omega_0 \approx DL^{-2} \approx \left(\frac{R}{r_0}\right)^{3\nu+1} \exp\left(-\frac{0.87}{Rn^{1/3}}\right). \quad (7.4)$$

It is easily seen that when $T \approx \omega_0$, the deviation of the magnetization from saturation caused by excitation of spin waves is small; that is, $\omega_0 \ll T_C$.

It follows from (6.5) and (7.3) that the concentration dependence of D/T_C contains no exponential but is determined solely by the index ν of the correlation radius:

$$\frac{D}{T_C} \sim n^{-(2+\nu)S}. \quad (7.5)$$

This opens up a tempting possibility for experimental determination of ν .

The theory of spin waves in alloys of the PdFe type was first developed by Doniach and Wohlfarth,⁸⁵ Cole and Turner,⁸⁶ and Long and Turner.⁸⁷ It was assumed that the spin splitting of the d band of palladium is small, i.e., that the inequality (5.11) is satisfied. These authors nevertheless supposed that the splitting of the d band is uniform; this in turn means that the molecular field acting on the localized spins is uniform. In such a treatment, D actually is determined by the mean energy

of indirect interaction of localized spins, to which the chief contribution is made by impurity pairs with a distance of order $R \ll r_c$, although such pairs obviously have no relation to propagation of spin waves. As a result, the incorrect relation $D \sim x$ was obtained.

At such concentrations that the splitting of the d band of palladium is large and the nonuniformity of the magnetization small, the exponential variation of D with x is replaced by a weaker one.⁸²

b) *The density of states and electrical resistivity.*

The density of states of spin waves $\rho_S(\omega)$, normalized to unit volume, is determined by formula (3.10). The respective expressions for the deviation of the moment density from saturation and for the specific heat due to spin waves have the usual form

$$\begin{aligned} \Delta M &= \mu \frac{\zeta(3/2)}{8\pi^{3/2}} \left(\frac{T}{D(x)}\right)^{3/2}, \\ C_S &= \frac{15}{32} \frac{\zeta(5/2)}{\pi^{3/2}} \left(\frac{T}{D(x)}\right)^{3/2}, \end{aligned} \quad (7.6)$$

where $\zeta(y)$ is the Riemann zeta function.

We note that in Ref. 87, because of an error in the calculation of the density of states, a factor x^{-1} was omitted in the expression for $\delta S = S - \langle S^2 \rangle$, which is equal to $\Delta M/\mu n$ [and also in the expression for $S^2 - \langle (S^2)^2 \rangle$].

In contrast to the magnetization and the specific heat, the spin-wave contribution to the electrical resistivity of a disordered system varies with temperature in a manner differing from that in ordered ferromagnets.

In disordered ferromagnets, the temperature-dependent part of the electrical resistivity is produced by two mechanisms: elastic scattering by randomly located spins, with potential $I(S^2)$, and inelastic scattering by spin waves. It is clear from (7.6) that the first part $\Delta\rho_1$ is proportional to $(T/D)^{3/2}$. The spin-wave part $\Delta\rho_2$ of the electrical resistivity also has this same temperature dependence. The point is that in scattering of electrons by spin waves propagating in a disordered system of polarized spins, the law of conservation of quasimomentum is not satisfied.^{16,87} Therefore $\Delta\rho_2$ is simply proportional to the number of spin waves, i.e., to $(T/D)^{3/2}$.

The total resistivity is

$$\Delta\rho = \Delta\rho_1 + \Delta\rho_2 = \frac{\sqrt{2}-1}{2^{3/2}\pi} \zeta\left(\frac{3}{2}\right) \frac{m_e}{n_e e^2} S I^2 \eta(\epsilon_F) \Omega_0 \left(\frac{T}{D}\right)^{3/2}, \quad (7.7)$$

where m_e , n_e , and $\eta(\epsilon_F)$ are, respectively, the mass, concentration, and density of states of the electrons.

We recall that in ordered ferromagnets the spin-wave contribution to the resistivity is proportional to T^2 , since by virtue of the law of conservation of quasimomentum the electrons are scattered only at small angles.

8. Local spin flips

a) *The magnetization and specific heat at low temperatures.*⁸⁷ In ordered ferromagnets at low temperatures, all the thermodynamics is determined by spin waves. In disordered ones there is an additional term due to local flips of spins which, by virtue of the random dis-

⁴⁾In Refs. 83 and 84, the jump probability did not contain the pre-exponential factor r^{-1} . Therefore (7.2) contains an additional factor r_0^{-1} as compared with these papers.

tribution of the magnetic atoms, are separated from their nearest neighbors by a distance larger than the average and are therefore weakly coupled to the main body of magnetic atoms. The molecular field H that acts on such spins is smaller than the molecular field on impurities located at the average distance from each other. Therefore even at low temperatures, inversion of the spin of isolated impurities is possible.

In order to count the number of isolated impurities, we must find the distribution function $W(\omega)$ ($\omega = \mu H$) of the molecular field, which represents the probability per unit $d\omega$ that the energy of a spin in the molecular field lies in the interval $(\omega, \omega + d\omega)$.

At small ω , the distribution function is proportional to the probability of fluctuations in which one of the impurities is separated from the rest by a distance larger than the mean. It follows from (6.1)⁵⁾ that in order that the molecular field at a given impurity shall not exceed H , it is necessary that the impurity nearest to it shall be located at a distance not smaller than

$$r(\omega) = R \ln \frac{SV_0}{\omega}. \quad (8.1)$$

A field of order H is obviously produced by each impurity located in a spherical shell of radius $r(\omega)$ and thickness R . If the number of impurities in this shell, which is $4\pi nr^2(\omega)R$, is small, then the molecular field H is produced by a single nearest neighbor, separated from the given impurity by a distance $r(\omega)$. Therefore, using Poisson's formula for the distribution of nearest neighbors, we get

$$W(\omega) = 4\pi nr^2(\omega) \exp\left(-\frac{4\pi}{3} nr^3(\omega)\right) \frac{dr}{d\omega} \\ = \frac{3\nu_R}{\omega} \ln^2 \frac{SV_0}{\omega} \exp\left(-\nu_R \ln^3 \frac{SV_0}{\omega}\right), \quad (8.2)$$

where $\nu_R = (4/3)\pi nR^3$. At small molecular fields, when $3\nu_R \ln^2(SV_0/\omega) \gg 1$, i.e., the number of impurities in a shell of radius $r(\omega)$ and thickness R is large, the molecular field at a given impurity is produced by a larger number of impurities. Therefore in order that the field at a given impurity shall not exceed H , the nearest neighbors to the given impurity must be distant by more than $r(\omega)$. Consequently the function $W(\omega)$ falls off at small ω faster than according to formula (8.2); but it can be shown that the main term in the argument of the exponential is, as before, equal to $\nu_R \ln^3(SV_0/\omega)$. At $\omega=0$ the distribution function $W(\omega)$ of course vanishes.

The deviation of the magnetization from saturation when $\nu_R \ln^2(SV_0/T) \ll 1$, which is produced by local spin flips, is proportional to the number of spins whose energy in the molecular field is $\omega \lesssim T$. By use of the distribution function (8.2), it is easily shown that this number is $\exp[-\nu_R \ln^3(SV_0/T)]$.

Thus at temperatures that satisfy the inequality $\nu_R \ln^2(SV_0/T) \ll 1$ [but $T \ll T_c$, i.e., $\nu_R \ln^3(SV_0/T) > 1$], the deviation of the mean projection of the spin from saturation is

$$\delta S = S - \langle S^2 \rangle = S \exp\left(-\nu_R \ln^3 \frac{SV_0}{T}\right). \quad (8.3)$$

It is interesting that in this temperature range, δS decreases with temperature more slowly than according to a linear law.

It is seen from (8.3) that the second derivative satisfies $d^2\langle S^2 \rangle/dT^2 > 0$ in this temperature range. Thus the character of the temperature variation of the magnetization in the disordered ferromagnets under consideration is qualitatively different from that in ordered ferromagnets: over a broad range of temperature within which the magnetic moment has almost attained saturation, the $M(T)$ curve is concave. This property of the $M(T)$ curve persists up to the transition (see Subsection c).

At low temperature, where $\nu_R \ln^2 T^{-1} SV_0 \gg 1$, the part of δS that is due to local spin flips falls off with temperature faster than according to a power law;⁶⁷ therefore the principal contribution to the departure of the magnetization from saturation must come from spin waves.

The specific heat due to local spin flips varies with temperature, for $\nu_R \ln^2 T^{-1} SV_0 \ll 1$, according to the same law (8.3) as the magnetization (except for the unimportant pre-exponential factor).

But at temperatures such that $\nu_R \ln^2 T^{-1} SV_0 > 1$, the contribution of local spin flips to the specific heat is more important than their contribution to the magnetization. This is explained as follows.

The specific heat of a spin whose energy in the molecular field is ω can, to make an estimate, be written in the form $(\omega/T)^2 e^{-\omega/T}$; that is, it falls off exponentially with increase of ω . On the other hand the distribution function $W(\omega)$, i.e., the number of spins with energy ω , rises rapidly with increase of ω . As a result those fluctuations are optimal for which $\omega/T \approx \nu_R \ln^2 T^{-1} SV_0 \gg 1$, so that the specific heat per impurity is of order

$$C_M \sim (\nu_R \ln^2 T^{-1} SV_0)^2 \exp(-\nu_R \ln^3 T^{-1} SV_0). \quad (8.4)$$

In this formula we have, for simplicity, written only the main term in the argument of the exponential. An exact formula is contained in Ref. 67.

We see that the specific heat (8.4) contains a large pre-exponential factor, which increases with decrease of temperature. Therefore the specific heat decreases with temperature more slowly than does the magnetization, and the role of local spin flips proves more important in the specific heat than in the magnetization.

The variation of the specific heat with temperature for various concentrations of the magnetic atoms is shown in Fig. 7. At not too low temperatures the $C_M(T)$ curves are convex. It follows from (8.3) and (8.4) that the specific heat has this property when $3\nu_R \ln^2 T^{-1} SV_0 < 1$.

b) *Electric kinetic phenomena.* In alloys of the PdFe type, these phenomena are of course determined by the high-mobility s electrons. Exchange scattering of them by magnetic impurities produces the magnetic contribution to the electrical and thermal resistivities, the thermal electromotive force, and other kinetic coeffi-

⁵⁾For simplicity, we use the potential (6.1) instead of (5.9) here and later in this subsection, since for $r \gg R$ the main variation of the potential with r is determined by the exponential.

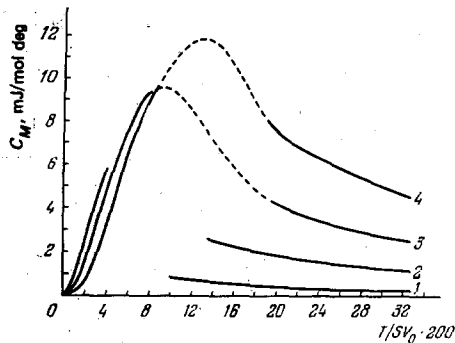


FIG. 7. Specific heat of palladium alloys (theory). The low-temperature parts of the solid curves show the specific heat due to localized spin flips; the high-temperature parts show the specific heat of pairs [formula (11.3)]. The dashed sections of the curves were obtained by extrapolation: $\nu_R = 10^{-2}$ (1), $2 \cdot 10^{-2}$ (2), $3 \cdot 10^{-2}$ (3), and $4 \cdot 10^{-2}$ (4).

icients. In order to calculate the kinetic coefficients in our case, the probability of scattering of s electrons by a localized spin in the molecular field⁶⁸ must be averaged with weight $W(\omega)$. The kinetic coefficients vary with the concentration of magnetic impurities and with the temperature chiefly because of the exponential factor $\exp(-\nu_R \ln^3 T^{-1} S V_0)$.⁶⁷ In addition, they contain large pre-exponential factors, which, as in the case of the specific heat, slow down their decrease with temperature. For example, the thermoelectromotive force α is⁶⁷

$$\alpha \sim \frac{1}{e} \left(\frac{I}{E} \right)^3 (\nu_R \ln^2 T^{-1} S V_0)^2 \exp(-\nu_R \ln^3 T^{-1} S V_0), \quad (8.5)$$

where e is the charge of the electron, and where I and E are, respectively, the constants of exchange and of nonexchange interaction of an s electron with an impurity. It is assumed that $I \ll E$ and that $3\nu_R \ln^2 T^{-1} S V_0 > 1$.

c) *The magnetization at high temperatures.*⁶⁸ In Subsection a) of the present section, we found the temperature variation of the magnetization in the temperature range where the deviation of the magnetization from saturation is small. Now, by using results of percolation theory, we shall find $M(T)$ over a wider temperature interval.

Since the spins in the infinite cluster are oriented parallel, the relative magnetization $M(T)/M(0)$ is equal to the probability $P[r(T)]$ that a given magnetic atom belongs to the infinite cluster [cf. (1.2)]; $r(T)$ is defined by formula (6.2). This probability has been calculated by the Monte Carlo method.⁶⁹ Its variation with

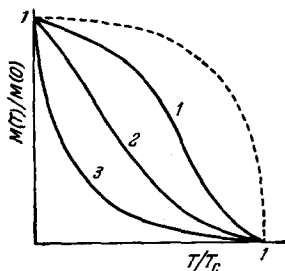


FIG. 8. Temperature dependence of the magnetization: $\nu_R = 5 \cdot 10^{-3}$ (1), $5 \cdot 10^{-4}$ (2), and $5 \cdot 10^{-5}$ (3). For comparison, the dashed curve shows $M(T)$ for pure nickel.

$p = (1/6)\pi r_{\max}^2$ is shown in Fig. 2 of Ref. 89. On using this graph and replacing r_{\max} by $r(T)$, we obtain the variation of $M(T)/M(0)$ with T/T_c that is shown in Fig. 8 for various values of the parameter ν_R . For comparison, the dashed curve in the same figure gives the temperature variation of the magnetization of pure nickel.

A characteristic feature of these curves is the large region of concavity, which increases with decrease of ν_R .

The threshold behavior of the function $P[r(T)]$ is the same as of the function $P(x)$ for disordered ferromagnets with nearest-neighbor interactions¹²; the role of the concentration x is played by the parameter $\pi r^2(T)$. Therefore near T_c (but not in the critical region) the temperature variation of the moment is determined by the relation

$$M(T) \sim \left[\frac{r(T) - r(T_c)}{r(T_c)} \right]^\beta \approx \left(\nu_R \ln \frac{T}{T_c} \right)^\beta, \quad (8.6)$$

where β is the critical index of the probability $P(x)$. It is seen that $M(T)$ increases very slowly (logarithmically) with decrease of T . The phase transition with temperature is found to be extremely extended.

When the probability P approaches 1, the deviation of P from 1 is due to those isolated spins that are separated from their nearest neighbors by a distance greater than $r(T)$. The relative number of such spins is $\exp[-4\pi r^2(T)/3]$; that is,

$$\frac{M(T)}{M(0)} = P(r(T)) = 1 - \exp\left(-\nu_R \ln^2 \frac{S V_0}{T}\right),$$

which agrees with formula (8.2).

d) *Experimental results. Comparison with theory.* Beginning with the 1960's, experimental studies have been made of the temperature and concentration dependence of the specific heat⁹⁰⁻⁹² and of the magnetization and electrical resistivity^{9, 74, 79, 93, 94} of palladium alloys. The measurements were made at magnetic-impurity concentrations larger than 10^{-3} . Figures 9 and 10 show typical temperature variations of the specific heat and electrical resistivity at various magnetic-impurity concentrations in palladium alloys. One notices the abrupt break in the resistivity that is produced by the onset of magnetic order. In this temperature range the specific heat has a broad maximum.

The temperature dependence of the magnetic part of the electrical resistivity for $x < 0.01$ is well described

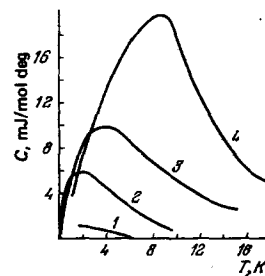


FIG. 9. Temperature dependence of the magnetic specific heat C_M of $\text{Pd}_{1-x}\text{Co}_x$ alloys.⁹ x (at. %) = 0.075 (1), 0.16 (2), 0.24 (3), and 0.35 (4).

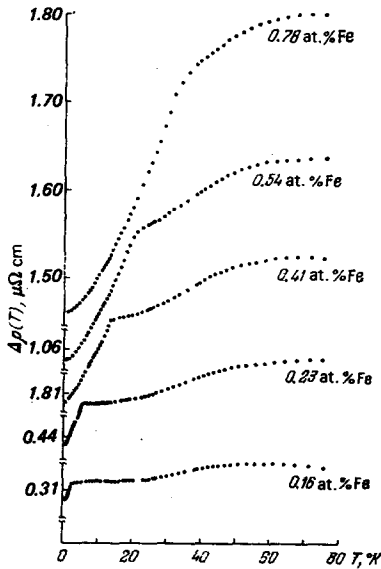


FIG. 10. Temperature dependence of the impurity part of the resistivity, $\Delta\rho(T) = \rho_{\text{alloy}}(T) - \rho_{\text{Pd}}(T)$, of $\text{Pd}_{1-x}\text{Fe}_x$ alloys.¹⁴ The iron content is indicated in the figure.

by the spin-wave $T^{3/2}$ law (Fig. 11). For $x > 0.02$, this law is replaced by the quadratic variation characteristic of an ordered ferromagnet.⁹⁴ The change in the temperature dependence is due to the transition from a strongly disordered system to one that is almost uniformly magnetized.

An attempt has been made⁹⁵ to determine the concentration dependence of the spin-wave stiffness D from the data on electrical resistivity. The authors used, instead of formula (7.7), an expression containing an extra factor x . In addition, an ill-founded assumption was made about variation of the constant I with impurity concentration.

In Ref. 91 the temperature dependence of the magnetization of $\text{Pd}_{1-x}\text{Fe}_x$ alloys with $x \geq 0.002$ was measured in an external magnetic field. At a fixed field, the temperature variation of the magnetization agrees well with spin-wave theory. The values of D found in Ref. 91 are in satisfactory agreement with results obtained from NMR⁹⁶ and from neutron experiments⁹⁷ (Table II).⁶⁾

The specific heat follows the spin-wave $T^{3/2}$ law only for $x \geq 0.01$. For $x < 0.01$ it increases with T considerably more slowly.^{90,91} From Fig. 9, which shows the experimental variation of the magnetic specific heat of PdCo alloys with temperature,⁹ it is seen that in the region appreciably to the left of the maximum, i.e. for $T \ll T_C$, the $C_M(T)$ curve is convex, and not concave, as it should be according to a spin-wave law. Furthermore, the value of the specific heat is considerably smaller than the spin-wave specific heat (7.6) with D taken from the magnetization. This anomalous behavior of the specific heat apparently means that under the conditions of the experiment, it is determined by fre-

⁶⁾ We note that in Ref. 91 a quite large variation of D with magnetic field was observed. The reason for this variation is not clear.

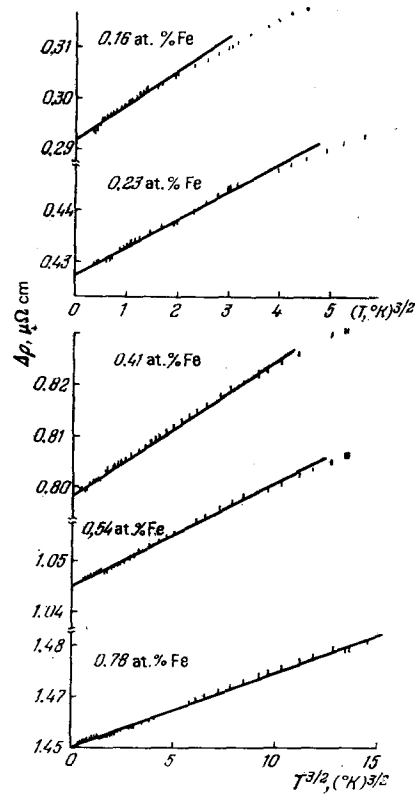


FIG. 11. Variation of $\Delta\rho(T)$ with $T^{3/2}$ for $\text{Pd}_{1-x}\text{Fe}_x$ alloys in the ferromagnetic phase.¹⁴ The iron content is indicated in the figure.

quencies $\omega > \omega_0$. In other words, the principal role is played not by spin waves but by localized excitations with frequencies $\omega < \omega_0$. In particular, this might be local flips of a single spin, for which the density of states in the frequency range under consideration decreases with increase of ω [see (8.2)]. Therefore, as has already been noted, the corresponding specific heat increases with T more slowly than according to a spin-wave law, and the $C_M(T)$ curve is convex.

By use of the value of D from Table II and of T_C from Table I, it is possible to test the correctness of the relation (7.5) between D and T_C . Figure 12 shows, on a log-log scale, the experimental variation of T_C/D with x for $x \leq 0.01$. The experimental points fit a straight line well; that is, in accordance with theory, D and T_C contain the same exponential factor. From the slope of the straight line it is seen that $D/T_C \sim x^{-0.45}$. The exponent is close to the value 0.38 that follows from (7.5) with $\nu = 10.85$.

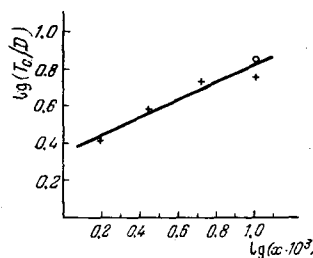


FIG. 12. Variation of $\lg(T_C/D)$ with $\lg x$ for $\text{Pd}_{1-x}\text{Fe}_x$ alloys.

TABLE II. Stiffness coefficient D of spin waves in $\text{Pd}_{1-x}\text{Fe}_x$ alloys.

x , at. %	0.15	0.28	0.50	0.53	1.0	1.7	2.0
D , °K·Å ²	104 ^{*)}	193 ^{*)}	364 ^{*)}	366 ^{*)}	690 ^{*)} , 556 ^{*)} , 630±60 ^{**)}	950 ^{*)}	1020±70 ^{**)}

*)Obtained by analysis of the temperature dependence of the specific heat. All the other values of D , from Ref. 91, were found by analysis of the temperature dependence of the magnetization.
**)The measurements were made in magnetic field of 4 kOe.

9. Effect of an oscillating potential

We have already mentioned that the oscillatory part (5.10) of the exchange potential causes a transition of the alloy, at sufficiently small x , from the ferromagnetic state to a spin-glass state. The transition occurs when, at distances of order r_c , the ferromagnetic potential (5.9) is comparable with the oscillatory part; that is, at concentrations that satisfy the equation

$$\left(\frac{r_c}{a}\right)^2 e^{-r_c/R} \approx \frac{V_1}{V_0} \frac{a}{R}. \tag{9.1}$$

In binary alloys of palladium, no ferromagnetism-spin-glass transition is observed. Such a transition has been observed⁹⁸ in PdFe alloys after hydrogen absorption. By filling the d band of the palladium, the hydrogen decreases the density of states on the Fermi surface and increases $1 + \Gamma$, thus facilitating satisfaction of the criterion (9.1). The transition to a spin-glass state has apparently been observed also in alloys of platinum,⁹⁹ for which the parameter $1 + \Gamma \approx 0.5$,⁹⁴ is larger than in palladium.

But even in the ferromagnetic phase, when at average distances $V_f(r_c) > V_{\text{BKKT}}(r_c)$, the oscillating potential leads to a number of interesting effects. The point is that at distant impurities the oscillating potential may be comparable with the ferromagnetic one or even exceed it. Consequently, the molecular field at them, at $T \ll T_c$, is small or negative (directed opposite to the magnetization); that is, the distribution function $W(\omega)$ of the molecular field is now nonzero for $\omega \leq 0$. Hence it follows, first, that even at $T = 0$ some of the spins are oriented opposite to the magnetization. Their number is

$$n_1 = n \int_{-\infty}^0 W(\omega) d\omega. \tag{9.2}$$

Second, in the specific heat a term appears that decreases linearly with temperature.

In fact, spins whose energy in the molecular field is of order T make a contribution of order 1 to the specific heat (we remind the reader that we are measuring temperature in energy units). At small T , the number of such spins is of order $\int_0^T W(\omega) d\omega \sim W(0)T$. Thus the specific heat per impurity is¹⁰⁰

$$C_M \sim W(0) T. \tag{9.3}$$

Since the spin-wave part of the specific heat $\sim T^{3/2}$, the specific heat (9.3) due to local spin flips becomes dominant at sufficiently low temperatures.

The concentration dependence of $W(0)$ can be found by the optimal-fluctuation method. The probability that

an impurity is separated from its nearest neighbors by a distance $r > r_c$ is proportional to $\exp(-4nr^3/3)$ and decreases with increase of r . At the same time, the probability that the molecular field at the distant impurity is small increases with increase of r .

As a result, it is found that¹⁰⁰

$$W(0) \sim e^{-(4/3)nl^3},$$

where l is the distance to the nearest impurity in the optimal fluctuation: $l \sim R \ln(V_0/V_1 \nu_R)$.

10. Ternary alloys

a) *Alloys with nonmagnetic impurities.* In order to explain the effect of the properties of the matrix on the ferromagnetism of dilute alloys, nonmagnetic impurities (Ag, Sn, Rh, H) have also been introduced into these alloys in a number of investigations.^{9,70,71,98,101} In most cases this leads to an abrupt drop of the Curie temperature. Thus on addition of 6% tin to the alloy $\text{Pd}_{0.98}\text{Co}_{0.02}$, the Curie temperature drops from 94 °K to 20 °K^{70,71} (Fig. 13). At the same time, the paramagnetic susceptibility of the matrix decreases by a considerably smaller amount.

The model formulated in Section 5 enables us to understand these properties of ternary alloys.¹⁰² Nonmagnetic impurities affect the matrix in two ways.

1) Scattering of electrons by impurities and change of their concentration as a result of alloying lead to a change of the density of states of the electrons and of their mean correlation energy;

2) Locally, there is a change of the electron correlation energy at a site occupied by a nonmagnetic impurity.

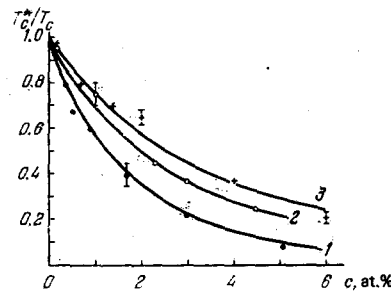


FIG. 13. Dependence of the Curie temperature of PdCoSn alloys on the tin content, for fixed concentrations of cobalt. Experimental points⁷¹: 1, 0.35 at. % Co; 2, 1.1 at. % Co; 3, 2.0 at. % Co. The curves were plotted from formula (10.3) with $\xi = 35$.

We first consider the situation when the local effects can be neglected. Nonmagnetic impurities lead to a renormalization of the value of Γ in formula (5.4) for the susceptibility:

$$\chi(k) = \frac{\chi_p}{1 + \Gamma(1 - \alpha) + a^2 k^2}, \quad (10.1)$$

where c is the concentration of nonmagnetic impurities. Since $1 + \Gamma \ll 1$, we may for small k neglect the influence of the nonmagnetic impurities on χ_p and a . The magnitude and sign of the coefficient α depend on the electron spectrum, the valence difference between the matrix and the impurity, and also the value of the scattering potential of the impurity. At not too strong doping, the value of α can in most cases be estimated in the Hartree-Fock approximation for the rigid-band model. From (5.8) and (10.1) we find the interaction radius R^* in a ternary alloy:

$$R^* = R(1 + \xi c)^{-1/2},$$

$$\xi = -\frac{\alpha\Gamma}{1 + \Gamma} = -\frac{1}{\chi(0)} \left. \frac{d\chi(0)}{dc} \right|_{c=0} \gg 1. \quad (10.2)$$

The Curie temperature of the ternary alloy, in accordance with (6.5), is

$$T_C^* = T_C \exp \left[-\frac{0.87}{n^{1/3}R} (\sqrt{1 + \xi c} - 1) \right], \quad (10.3)$$

where T_C is the Curie temperature of the binary alloy.

Since $n^{1/3}R \ll 1$ and $\xi \gg 1$, T_C^* may differ greatly from T_C even at very small concentrations of nonmagnetic impurities.

If $\xi c \ll 1$, the nonmagnetic impurities produce little change in the susceptibility at saturation and in the potential radius. At the same time, the Curie temperature, which in this case is

$$T_C^* = T_C \exp \left(-\frac{0.87}{n^{1/3}R} \frac{\xi c}{2} \right), \quad (10.4)$$

may change greatly, increasing or decreasing, depending on the sign of ξ . It is evident from (10.4) that at small c

$$\ln \frac{T_C^*}{T_C} \sim n^{-1/3}c. \quad (10.5)$$

The situation is different if the principal role is played by a local change of the Hubbard correlation at a site. Since the perturbation changes the properties of the matrix only at distances of order R from the impurity, the exchange interaction of localized spins located at a mean distance r_c from each other "senses" the nonmagnetic impurity only if the latter falls within a thin cylinder of radius $\sqrt{Rr_c}$ joining the magnetic impurities. The probability of such a configuration is small if the concentration of nonmagnetic impurities (per unit volume) is less than $(Rn^{-2/3})^{-1}$.

The most interesting situation is that of almost magnetic impurities (in the sense of Anderson's¹⁰³ criterion), which have a large magnetic susceptibility. Even a small quantity of these impurities may substantially increase the magnetic susceptibility of the specimen, yet at the same time, as is clear from the preceding, have little effect on the Curie temperature. Furthermore, nonlocal effects, which were discussed above, may even lead to a decrease of T_C for this kind of impurity.

These considerations enable us to understand the experimental results for PdRhMn alloys. In PdRh alloys with rhodium content less than 5%, the susceptibility increases rapidly with increase of c . At the same time, T_C in PdRhMn alloys increases only inappreciably with rhodium content for $c < 1\%$, and then decreases.⁹

The effect of tin impurity, which was studied in Refs. 70 and 71, is primarily due to a decrease in the density of states, which is caused by increase of the number of occupied states in the d band.¹⁰⁴

Formula (10.3) describes well the experimental variation of T_C^* with c , if we take $\xi = 35$ (see Fig. 13). On the other hand, ξ can be found by use of the experimental variation of the susceptibility of PdSn alloy with tin content. Such measurements were made in Ref. 104, and according to these data $\xi = 20$ to 25. The difference between these two values of ξ may be due, first, to the fact that in the derivation of (10.3) no allowance was made for variation of the Fermi energy with the concentration of nonmagnetic impurities; and, second, to the fact that in Ref. 104 the susceptibility was measured at comparatively high concentrations of tin, and therefore the determination of ξ from these data is not altogether reliable.

Peculiar properties are possessed by ternary alloys containing hydrogen. Investigation of them is just beginning.^{71, 98}

b) *Two kinds of magnetic atoms.* There are magnetic impurities whose exchange interaction with the d electrons of the matrix is very small (for example, gadolinium and certain other rare-earth elements¹⁰⁵). In this case, effects caused by indirect interaction of the impurities show up only at very low temperatures. The interaction of such impurities with the matrix is conveniently studied by investigating alloys that also contain magnetic impurities that interact strongly with the matrix; for example, Fe and Co.¹⁰⁶ In such alloys, there is exchange interaction of three types: V_{AA} , V_{BB} , and V_{AB} , where A denotes the strongly bonded impurity and B the weakly bonded one. It is clear that $V_{AA} > V_{AB} > V_{BB}$ (For definiteness we suppose that $V_{AB} > 0$.) Therefore if the concentration n_B is less than or of the order of n_A , the interaction V_{BB} can be neglected, and T_C is determined by the impurities A . The spins of the impurities B may be weakly coupled with the spins A even at $T \ll T_C$. Therefore the spins B may determine the specific heat and the temperature dependence of the magnetization.

The calculation of the thermodynamic functions of such alloys can be carried out by use of the distribution function of the molecular fields, as in Section 8.¹⁰⁶

If

$$T_C > T_0 = V_{AB} S_A \exp \left(-\frac{0.62}{Rn_A^{1/3}} \right),$$

then in the interval $T_0 < T < T_C$ the mean spin slowly increases with decrease of the temperature, according to the law

$$\langle S_B^z \rangle = S_B \frac{4}{3} \pi n_A R^3 \ln^3 \frac{S_A V_{AB}}{T}. \quad (10.6)$$

The specific heat as a function of the temperature has two maxima: one near T_C , where ferromagnetism originates in spin system A , and the second at $T \approx T_0$, where the spins B become magnetized.

11. The paramagnetic phase

Fluctuations in the distribution of impurities produce unusual properties of an impurity ferromagnet, as compared with an ordered one, not only below the transition temperature but also above it. At $T > T_C$ there remain clusters of rigidly coupled spins. Excitations in these clusters, with energy of order T , determine the magnetic specific heat and the processes of inelastic scattering of electrons and neutrons. Rotation of clusters in a magnetic field determines the magnetic susceptibility. In consequence of the exponential falling off of the potential, the interaction energy $V(r) > T$ for all spins with $r < r(T)$; that is, such spins are rigidly coupled. But if $r > r(T)$, such spins may be considered free.

Pairs of rigidly coupled spins behave in a magnetic field like a single spin of value $2S$, so that the susceptibility of a pair is $\mu^2 2S(2S+1)/3T$. The susceptibility of an uncoupled pair of spins is $\mu^2 2S(S+1)/3T$. Thus formation of a rigidly coupled pairs of spins changes the susceptibility by the amount $2\mu^2 S^2/3T$.

The number of spins rigidly coupled into pairs is $(4/3)\pi n r^3(T) = \nu_R \ln^3 T^{-1} S V_0$, and the number of pairs is $\nu_R \ln^3 T^{-1} S V_0 / 2$.

Therefore the correction to the specific susceptibility of free spins resulting from interaction in pairs is

$$\Delta\chi = \frac{S}{S+1} \chi_0 \nu_R \ln^3 T^{-1} S V_0, \quad (11.1)$$

where

$$\chi_0 = \frac{n\mu^2 S(S+1)}{3T}.$$

Formula (11.1) is correct for $T \ll V_0$. If $T \gg V_0$, the interaction energy of the impurities in a pair is less than T for any distance between the impurities. Therefore the correction $\Delta\chi$ to the susceptibility is proportional to $\bar{V}(r)/T$, where $\bar{V}(r)$ is the mean interaction energy.

In this range of temperature, according to Ref. 67,

$$\Delta\chi = \chi_0 S(S+1) \nu_R \int_0^\infty \frac{dr \cdot r^2}{R^3} \frac{V(r)}{T}. \quad (11.2)$$

A general expression for $\Delta\chi$, valid for all $T \gg T_C$, was obtained in Ref. 67 by the method of virial expansion in powers of the concentration.¹⁰⁷ Thus the temperature dependence of the susceptibility has an anomalous character. The Curie-Weiss law is satisfied only when $T \gg V_0$; the paramagnetic Curie temperature, as is seen from (11.2) and (6.1), is

$$\theta = 2S(S+1) \nu_R V_0 \gg T_C. \quad (11.3)$$

Over a wide temperature range $T_C \ll T \ll \theta$, the susceptibility increases with decrease of temperature much more slowly than according to the Curie-Weiss law. The magnetic specific heat in the paramagnetic phase, for $T \gg T_C$, is due to transitions between spin

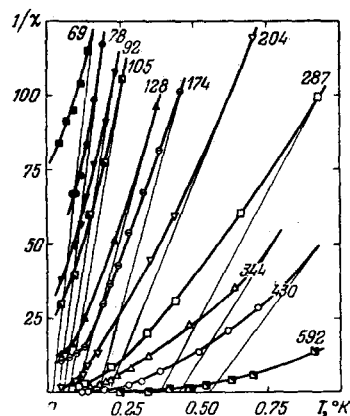


FIG. 14. Inverse susceptibility of $\text{Pd}_{1-x}\text{Fe}_x$ alloys.⁵⁴ The numbers on the curves indicate the iron concentration $x \cdot 10^6$.

levels in pairs. The maximum contribution, of order unity, comes from those pairs in which the splitting of the levels is of order T . The distance between impurities in these pairs obviously lies in the interval $r(T) < r < r(T) + R$; the number of such pairs is of order $\nu_R \ln^2 T^{-1} S V_0$. Consequently the specific heat per impurity is

$$C_M = A_S \nu_R \ln^2 T^{-1} S V_0. \quad (11.4)$$

The coefficient A_S depends on the spin:

$$A_{1/2} = \frac{3}{2} \ln \frac{4}{3}, \quad A_1 = 0.86, \quad A_{3/2} = 1.15,$$

In the interval $T_C \ll T \ll S V_0$, the corrections to all quantities dependent on transitions between magnetic levels in a pair have a temperature variation of the type (11.4). At high temperatures, $T \gg V_0$, the specific heat $C_M \approx \nu_R (V_0/T)^2$.⁶⁷

The method of virial expansion can also be used to allow for the effect of pairs on the electronic kinetic coefficients.^{67, 108}

With decrease of temperature, clusters of larger and larger size become important. Because of the exponential character of the potential, the bonds within a cluster may be considered rigid, and those between clusters broken⁷¹ (see Section 6). Therefore the free energy of the finite clusters in a magnetic field is the same for the Ising and for the Heisenberg ferromagnet. If $\tau = [r(T_C) - r(T)]/r(T) \ll 1$, the free energy is determined by formula (3.3) with $x - x_c$ replaced by τ . For the susceptibility we have

$$\chi \sim \frac{n}{T} \left(\ln \frac{T}{T_C} \right)^{-\nu}. \quad (11.5)$$

The paramagnetic susceptibility has been measured in PdFe ⁵⁴ and PdMn ⁷⁷ alloys. From Fig. 14⁵⁴ it is clearly evident that over the whole temperature range in which measurements were made, the susceptibility, in agreement with the theory presented above, disobeys the Curie-Weiss law. Therefore the paramagnetic Curie temperatures found in this paper have no particular meaning.

⁷¹This procedure is justified if we are not interested in quantities dependent on inelastic transitions between levels of the magnetic system, for example the specific heat.

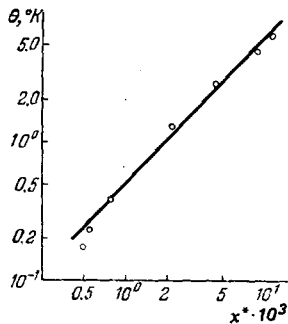


FIG. 15. Paramagnetic Curie temperature θ of $\text{Pd}_{1-x}\text{Mn}_x$ alloys as a function of $x^* = x(1 - 12x)$ (according to Ref. 77).

In the PdMn alloys, the susceptibility was measured at high enough temperatures so that the Curie-Weiss law is satisfied. Figure 15 shows the variation of the paramagnetic Curie temperature θ with $x^* = x(1 - 12x)$ according to the data of Ref. 77 (the factor $(1 - 12x)$ eliminates pairs of Mn atoms that are nearest neighbors in the lattice, and between which there is direct antiferromagnetic exchange). It is seen that this relation is nearly linear. By using the value $R = 3.3 \text{ \AA}^8$) and supposing that $S = 5/2$,⁷⁷ we get from the graph of Fig. 15, according to formula (11.3), $V_0 = 3 \text{ }^\circ\text{K}$, which agrees reasonably well with the value of the Curie temperature. Since, for the same concentration, the Curie temperature in PdFe alloys is about 10 times greater than in PdMn, the Curie-Weiss law should be satisfied in PdFe beginning with a temperature of the order of $30 \text{ }^\circ\text{K}$.

A large magnetic specific heat in the paramagnetic phase of alloys was observed in an early paper of Veal and Rayne.⁹⁰ The most complete data have been obtained by the Leyden group.⁹ It is seen from Fig. 9 that in the paramagnetic range, the specific heat drops slowly with rise of temperature; the smaller the concentration, the longer is the high-temperature "tail" of the specific heat.

The specific heat calculated by formula (11.4) is shown in Fig. 7 (the sections of the curves to the right of the maximum). Qualitatively, the behavior of the specific heat agrees with experiment (see Fig. 9); the absence of experimental data over a wide range of temperature above T_C prevents the carrying out of a quantitative comparison.

By extrapolating the results of calculation of the specific heat from the low- and high-temperature regions to the region of temperatures of order T_C , we get the temperature dependence of the specific heat over the whole range, from temperatures $T \ll T_C$ to the distant paramagnetic range (see Fig. 7). It can be compared with the experimental results in Fig. 9. Of the general qualitative agreement there can be no doubt.

IV. CONCLUSION

We have analysed in detail the properties of ferromagnetic systems with exchange potential of two types.

⁸⁾Formula (11.4) is derived for the potential (6.1), which corresponds to formula (6.4) for T_C . On comparing (6.4) with experiment, we obtain $R = 3.3 \text{ \AA}$.

The model with exchange potential falling off exponentially enabled us to explain the important experimental results on alloys of the PdFe type. We have not discussed thermodynamic systems near the Curie point, which at present have not yet been studied (we can cite only one paper⁷²). Investigations of critical phenomena in such strongly disordered systems would undoubtedly be interesting both theoretically and experimentally. It would also be desirable to determine more reliably the concentration variation of the spin-wave stiffness D . This in turn would, on the one hand, enable us to elucidate the role of local excitations in the thermodynamics of alloys, and, on the other hand, afford a possibility of finding the index of the correlation radius more accurately.

In certain metals and degenerate semiconductors, the characteristic period of the oscillating RKKY potential (5.10) for all the electrons, or for a considerable group of them, may be larger than the mean distance between magnetic atoms. In this case, the potential (5.10) leads to impurity ferromagnetism, which has been observed experimentally in the semiconductor alloys GeTe-MnTe and SnTe-MnTe.^{109,110} Because of the long-range character of the potential, fluctuations do not play as large a role here as in the situations considered above. Nevertheless their effect on the thermodynamic properties of the system may be appreciable.¹¹¹ The properties of such ferromagnets have so far been insufficiently studied experimentally.

The methods discussed in Section II have been found useful for analysis of the properties not only of dilute ferroelectrics, but also ferri- and antiferroelectrics,^{22,112} the experimental investigation of which has begun in recent years.^{8,9)} The study of these and other disordered magnetic systems with complicated magnetic order is at present one of the most interesting problems within the field under consideration.

And finally, we must mention amorphous ferromagnetic metals (Metglas type alloys)—promising materials for practical applications. Interest in them has grown steadily in recent years; it has been possible to obtain metglasses of various contents, whose properties vary over a very wide range. Further experimental investigation of them will without doubt be fruitful.

In the theory, only the very first steps have been taken, and many peculiarities of these materials are not understood either quantitatively or even qualitatively. In contrast to the systems treated in the present review, in amorphous materials a large role is played by correlation in the distribution of magnetic atoms. A correct accounting for the correlation is one of the basic problems of the theory.

⁹⁾In this connection we note that in a recently published paper,¹¹³ in which scattering of neutrons was studied in the anisotropic antiferromagnet $\text{Mn}_{1-x}\text{Zn}_x\text{F}_2$, the authors succeeded in measuring directly the concentration dependence of the correlation radius L near the percolation threshold. It was found that the variation of L with $x - x_c$ is described by a power law with an exponent slightly less than unity.

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