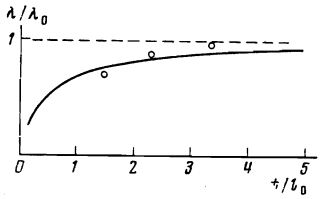

FIG. 3. Change of the ratio of the thermal-conductivity coefficient of aluminum films to the bulk value as a function of the normalized thickness.

thickness (the quartz-vibrator method, the calorimetric method) and the geometric thickness (multiple-wave interferometry) offer evidence that the film density at the investigated thicknesses, within a measurement accuracy 3–5%, coincides with the density of the bulk metal.[18–20] In this case the specific heat of the films does not depend on the thickness and coincides, within the ~ 10% accuracy limit of the measurement procedure, with the data for the bulk metal. It follows also that the thermal conductivity is the main cause of the change in the diffusivity of thin films.

Thus, the experimental data on the thermal conductivity and diffusivity of thin films of aluminum, with structure close to equilibrium, is well described by the classical theory of the size effect. A deviation of the specific heat of the films from the value for the bulk metal can be expected only at low temperatures, where the size factor can greatly influence the value of c.[2]

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Thermodynamics of disordered Heisenberg ferromagnets near the threshold concentration (percolation threshold)

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The temperature and concentration dependences of the specific heat and magnetization for a disordered Heisenberg ferromagnet in which the concentration x of magnetic atoms is close to the percolation threshold x_c are calculated with the aid of a scaling hypothesis formulated earlier for the percolation problem. The dependence of the Curie temperature on $x - x_c$ is also obtained.

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In solid solutions of magnetic and nonmagnetic materials, in the framework of a model in which nearest magnetic neighbors interact, macroscopic magnetic order arises only if the concentration x of magnetic atoms exceeds the threshold value x_c determined by percolation theory. For $x < x_c$ the probability of existence of an infinite connected cluster of magnetic atoms is equal to zero, while for $x > x_c$ this probability is nonzero and so macroscopic magnetic order appears at sufficiently low temperatures. The thermodynamics of such systems, both with Ising and with Heisenberg interactions of the localized spins, has been investigated repeatedly with the aid of high-temperature and concentration expansions.[1] However, near the percolation threshold, for $|x - x_c|/x_c \ll 1$, because of the poor convergence of the corresponding series, these methods have been found ineffective: it has not been possible to obtain by means of them the temperature and concentration dependences of the thermodynamic quantities.

In the present paper the thermodynamics of disor-

dered Heisenberg ferromagnets near the percolation threshold is investigated with the aid of a scaling hypothesis formulated for the percolation problem.[2-4] The point is that the appearance of an infinite cluster as x is raised above the threshold value x_c occurs in a way that is analogous in many respects to a second-order phase transition with respect to the temperature. The analog of the order parameter is the concentration of magnetic atoms in an infinite cluster, which varies from zero for $x \leqslant x_c$ to unity for $x=1$. Correlation functions, a correlation length and the analog of the thermodynamic potential are also introduced for the percolation problem. The dependences of the characteristic quantities on $x-x_c$ are assumed to be power dependences; relations between the critical indices are established.

The numerous computer calculations that have by now been carried out have confirmed the validity of the scaling theory for the percolation problem. Using the scaling theory we have obtained the energy dependence of the density of states of the magnetic excitations, and this has made it possible to determine the dependence of the Curie temperature T_c on $x-x_c$ and also the temperature and concentration dependences of the magnetization and specific heat. It is interesting that the temperature dependences are described by the Bloch law, not right up to a temperature of the order of T_c, as in ordered ferromagnets, but in a much narrower temperature interval. Outside this interval these dependences are found to be power laws, but the powers are equal to 1 and 0.73, respectively, and not $\frac{3}{2}$.

1. THE DENSITY OF STATES

We shall consider first a solid solution with a concentration of magnetic atoms greater than x_c. At zero temperature the spins of the atoms in each cluster have parallel orientations. The density of states of the magnetic excitations in an infinite cluster, $\rho_{\text{inf}}(\omega)$, can, at zero temperature, be introduced in the following way:

$$\rho_{inf}(\omega)=\frac{1}{\pi S}\int \operatorname{Im} G^{+-}(k,\omega)\frac{d^3k}{(2\pi)^3},$$
$$G^{+-}(k,\omega)=-i\int e^{i\mathbf{kr}-i\omega t}\langle \overline{TS_r^+(t)S_0^-(0)p_rp_0}\rangle\, d^3r\,dt, \quad (1)$$
$$S^{\pm}=S^x\pm iS^y.$$

Here $\mathbf{S}$ is the spin operator and the quantity $p_{\mathbf{r}}$ is equal to unity if a magnetic atom belonging to an infinite cluster is found at the site $\mathbf{r}$, and equal to zero otherwise. A superior bar denotes averaging over the configuration of impurities and the z axis is parallel to the magnetization. The density of states in finite clusters can be introduced in an analogous way.

A characteristic feature of the Heisenberg Hamiltonian is the existence of the conservation law for the total spin. A consequence of this conservation law is the existence, in disordered Heisenberg ferromagnets, of weakly damped long-wavelength spin waves with a quadratic dispersion law[5]

$$\omega=Dk^2. \quad (2)$$

In this respect there is an analogy between the magnetic and elastic properties of disordered systems: it is well known that the invariance of the equations of atomic dynamics with respect to a uniform translation ensures the existence of weakly damped acoustic phonons.[6]

According to Kirkpatrick,[7]

$$D\sim\sigma/P, \quad (3)$$

where P is the ratio of the number of sites in an infinite cluster to the number of sites in the crystal, σ is the conductivity of the lattice (in which all the links between nearest magnetic neighbors possess a conductance equal to unity and the other links do not conduct current). We note that the relation (3) was obtained without any model assumptions about the topology of the infinite cluster.

The density of states in the spin-wave region is

$$\rho_{inf}(\omega)=\int\delta(\omega-Dk^2)\frac{d^3k}{(2\pi)^3}=\frac{1}{4\pi^2}\frac{\omega^{1/2}}{D^{3/2}}. \quad (4)$$

According to the scaling hypothesis,[2-4] P and σ have power-law dependences on $x-x_c$:

$$P\sim(x-x_c)^{\beta},\ \sigma\sim(x-x_c)^{t}.$$

Numerous computer calculations confirm the existence of such dependences and lead to the following values of the indices: $\beta=0.33$,[7] $t=1.72$.[8] Thus, in the spin-wave region, $\rho_{\text{inf}}(\omega)\sim\omega^{1/2}(x-x_c)^{-2.1}$.

The dispersion law (2) is valid so long as the wavelength of the magnons is greater than the characteristic length determining the topology of the infinite cluster, since only in this case does the wave "average" the spatial fluctuations existing in the structure of the infinite cluster, so that the damping of the wave is small. According to the scaling hypothesis,[3,4] near the percolation threshold the system is characterized by one parameter with the dimensions of length ($L\sim(x-x_c)^{-\nu}$), which, by analogy with second-order phase transitions, is called the correlation length. As follows from the definition of the correlation length,[3,4] it has a simple meaning: the quantity L is of the order of the mean size of the finite clusters, and also, therefore, of the order of the characteristic size of the infinite cluster which "flows around" the finite clusters. Thus, the relations (2) and (4) are true only for $\omega\ll\omega_0=DL^{-2}$. The critical index ν is equal to 0.8–0.9.[4]

In order to determine $\rho_{\text{inf}}(\omega)$ for $\omega>\omega_0$, we use the method, well-known from the theory of phase transitions, of matching the long-wavelength hydrodynamic mode with the so-called critical mode, for which the wavelength is less than the correlation length.[9] Here, the most important thing for us will be the fact that, since the characteristic length for the critical excitations is less than L, their energy and density of states per magnetic atom, $\tilde{\rho}_{\text{inf}}(\omega)$, should depend only on the geometrical properties of the infinite cluster over distances less than L. But the geometrical characteristics of the system at such distances do not depend on $x-x_c$; the parameter $x-x_c$ determines only the topology of the system over distances greater than L.

Thus, we have arrived at the conclusion that $\tilde{\rho}_{\text{inf}}(\omega)$

for $\omega \gg \omega_0$ does not depend on $x - x_c$, i.e., $\tilde{\rho}_{inf}(\omega) \sim f(\omega/V_0)$ (V_0 is the exchange integral between nearest magnetic neighbors). Consequently,

$$\rho_{inf}(\omega) \sim P f(\omega/V_0). \tag{5}$$

Matching the expressions for $\rho_{inf}(\omega)$ determined by formulas (4) and (5) at

$$\omega = \omega_0 \sim (x - x_c)^{t+2\nu-\beta}$$

we obtain

$$\rho_{inf}(\omega_0) \sim \frac{\omega_0^{3/2}}{D^{3/2}} \sim \omega_0^{(\nu+\beta-t)/(t+2\nu-\beta)} \sim P f\left(\frac{\omega_0}{V_0}\right). \tag{6}$$

Since

$$P \sim (x - x_c)^{\beta} \sim \omega_0^{\beta/(t+2\nu-\beta)},$$

it follows from (6) that $f(y) = y^{-z}$, where $z = (t-\nu)/(t + 2\nu - \beta)$. Therefore, the density of states for $\omega \gg \omega_0$ is equal to

$$\rho_{inf}(\omega) = \frac{\gamma P}{V_0 \Omega_0}\left(\frac{V_0}{\omega}\right)^z. \tag{7}$$

Here Ω_0 is the volume of the unit cell and γ is an unknown numerical coefficient. From the values given above for the indices it follows that $z = 0.27$.

Thus, the density of states has a maximum at energies of the order of ω_0 and falls off rather slowly with increasing energy for $\omega \gg \omega_0$. The main contribution to the normalization integral is given by energies of the order of V_0; the number of states in the spin-wave region is proportional to $L^{-3} \sim (x - x_c)^{2.6}$, i.e., is extremely small compared with the total number of states.

For $|x - x_c|/x_c \ll 1$ most of the magnetic atoms are connected in finite clusters, the mean size of which is of the order of L.[3,4] Since the geometrical properties of finite clusters over distances smaller than L are the same as those of an infinite cluster, the density of states $\rho_{fin}(\omega)$ in finite clusters for $\omega \gg \omega_0$ differs from $\rho_{inf}(\omega)$ only by a normalization factor:

$$\rho_{fin}(\omega) = \frac{\gamma}{V_0 \Omega_0}\left(\frac{V_0}{\omega}\right)^z. \tag{8}$$

Naturally, formula (8) is valid both above and below the percolation threshold.

2. THERMODYNAMIC FUNCTIONS

All the results obtained for the density of states are valid not only at zero temperature $T = 0$ but also at temperatures much lower than the Curie temperature T_c, when the deviation of the magnetization from saturation is small. Assuming this condition to be fulfilled, we can, in the usual way, express the deviation ΔM of the magnetization from saturation and the specific heat C in terms of the density of states introduced in (1):

$$\Delta M = M_0 - \langle M^z \rangle = \mu \int_0^\infty \frac{\rho_{inf}(\omega)}{e^{\beta\omega} - 1} d\omega,$$

$$C = \int_0^\infty (\rho_{inf}(\omega) + \rho_{fin}(\omega)) \frac{(\beta\omega)^2 e^{\beta\omega}}{(e^{\beta\omega} - 1)^2} d\omega. \tag{9}$$

Here μ is the effective magnetic moment of the atom, $\beta = T^{-1}$, and the saturation magnetization $M_0 = \mu P \Omega_0$.

In the spin-wave region $T \ll \omega_0$ the magnetization and specific heat obey the Bloch law

$$\Delta M = \mu \frac{\zeta(3/2)}{8\pi^{3/2}}\left(\frac{T}{D}\right)^{3/2}, \quad C = \frac{15}{32}\frac{\zeta(5/2)}{\pi^{3/2}}\left(\frac{T}{D}\right)^{3/2}, \tag{10}$$

where $\zeta(x)$ is the Riemann zeta-function. It can be seen from the formulas (10) that ΔM and C are proportional to $(x - x_c)^{2.1}$.

The formulas (10) also give a correct estimate of the corresponding quantities for $T \sim \omega_0$. It is easy to convince oneself that

$$\left.\frac{\Delta M}{M_0}\right|_{T=\omega_0} \sim \frac{1}{PL^3} \ll 1. \tag{11}$$

It follows from (11) that the Curie temperature is much larger (in the parameter $(x - x_c)^{-1}$) than the energy ω_0 at which the spin-wave part of the spectrum terminates. This difference from the properties of an ordered ferromagnet, for which these two characteristic quantities coincide, is connected with the aforementioned paucity of states in the spin-wave region.

We now calculate ΔM and C for $T \gg \omega_0$. The integral for ΔM is determined in this case by energies $\omega \sim \omega_0$, and therefore

$$\Delta M \sim \rho_{inf}(\omega_0) T. \tag{12}$$

Since $\Delta M/M_0 \sim 1$ at temperatures of the order of T_c, it follows from this that $T_c \sim P\rho_{inf}^{-1}(\omega_0)$, and by means of (4) or (6) we find

$$T_c = \alpha V_0 \left(\frac{x - x_c}{x_c}\right)^{t-\nu}. \tag{13}$$

Here α is an unknown numerical coefficient. The power in (13) $t - \nu \approx 0.9$.

As can be seen from (13), T_c falls off to zero as $x \to x_c$. This falloff is not connected with the decrease in the energy of interaction of the spins with their surroundings, since, irrespective of the value of $x - x_c$, this energy is not less than V_0. As can be seen from (12), the magnetic order is destroyed by thermodynamic fluctuations with energy of the order of ω_0 and characteristic length L. As $x - x_c$ decreases the density of states of these excitations grows, and this leads to decrease of T_c. The destabilizing role of the excitations with $\omega \sim \omega_0$ is possibly connected with the one-dimensional character of the structure of an infinite cluster over distances shorter than the correlation length.[10]

We note that qualitative arguments about the existence of the relation (13) were put forward by Shklovskiĭ and the author[11]; however, previously[11] we succeeded only in obtaining the inequality

$$T_c \leqslant \alpha V_0 \left(\frac{x - x_c}{x_c}\right)^{t-\nu}.$$

Inasmuch as

$$T_c/\omega_0=(x-x_c)^{-3\nu+\beta}=(x-x_c)^{-2.2},$$

there is, even for not too small values of $x-x_c$, a broad region of temperatures in which $\Delta M \sim T$. The Bloch law is fulfilled at much lower temperatures $T \ll \omega_0 \approx V_0(x-x_c)^{3\nu}$, and, as can be seen from (11), even at $T \sim \omega_0$ the deviation of the magnetization from saturation is extremely small:

$$\Delta M/M_0 \sim (x-x_c)^{2\nu}.$$

The long-range magnetic order disappears at the Curie point, but it is easy to see that, so long as $T \ll V_0$, there is short-range magnetic order over distances shorter than L but much longer than the lattice constant. The magnetization in a region of size r can be calculated by replacing the lower limit of the integration in (9) by $\omega(r)$, where $\omega(r)$ is the minimum energy of excitations in a ferromagnetic region of size r. Short-range magnetic order over a distance r obtains if

$$\frac{\Delta M(r)}{M_0}=\frac{T}{V_0}\left(\frac{V_0}{\omega(r)}\right)^{\gamma} \ll 1. \tag{14}$$

In particular, substituting $\omega(r) \sim T$ into (14), we can convince ourselves that in regions whose size is of the order of the characteristic length of the thermal excitations, short-range magnetic order always occurs.

Since it is precisely the thermal excitations that determine the specific heat, this means that the specific heat can be calculated with the aid of formulas (7)–(9) not only for $T \ll T_c$ but also in the entire temperature interval $\omega_0 \ll T \ll V_0$. We obtain

$$C=\frac{(2-z)\gamma}{V_0\Omega_0}\Gamma(2-z)\zeta(2-z)\left(\frac{T}{V_0}\right)^{1-z}, \tag{15}$$

where $\Gamma(x)$ is the gamma-function and $1-z=0.73$. The principal contribution to the specific heat (15) is given by excitations in finite clusters, the density of states of which is greater by a factor of P^{-1} than in an infinite cluster. Therefore, (15), like (8), is valid both above and below the percolation threshold.

For $T \ll \omega_0$ the temperature dependence of the specific heat is substantially different above and below the threshold. Below the threshold ($x < x_c$) the low-temperature specific heat is determined by clusters of size greater than L, since it is precisely in these that excitations with energy less than ω_0 are possible. Since the mean number of such clusters is exponentially small, the specific heat evidently also depends on T by an exponential law. Above the percolation threshold the specific heat for $T \to 0$ obeys the Bloch law. However, because the magnon specific heat at $T=\omega_0$ is a factor of P smaller than the specific heat (15) of the finite clusters, at $T=T_1 \ll \omega_0$ the Bloch law already ceases to be valid for C. In the range of temperatures between T_1 and ω_0 the specific heat is determined by the finite clusters of size greater than L. At not too small values of $x-x_c$ this region of temperatures is fairly narrow because of the small value of the index β.

Note added in proof: In formulas (8) and (15) for $\rho_{\mathrm{fin}}(\omega)$ and C it is necessary to replace z by $z'=z-\beta/(t+2\nu-\beta)$. The value of z' can be found from the matching condition for the function $\rho_{\mathrm{fin}}(\omega) \sim (v_0/\omega)^{\varepsilon'}$: $\rho_{\mathrm{fin}}(\omega_0) \sim \rho_{\mathrm{inf}}(\omega_0)$. The form of this condition is determined by the fact that the concentration of the magnetic atoms in finite clusters of size L is of the order of P. The author thanks B. I. Shklovskiĭ for calling his attention to the error in the text of the article.

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