Investigation of the disordered state of two-dimensional ferromagnetic substances

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The magnetic properties are studied of two-layered compounds, of $NiCl_2$ imbedded in graphite and of $(CH_3NH_3)_2CuCl_4$ at temperatures above that for the transition to the magnetic-ordered state. For both compounds an exchange ferromagnetic interaction between the magnetic ions in the layers is observed which exceeds by several orders of magnitude the interaction between adjacent layers. It is shown that at low temperatures the magnetization curves may be described by the spin-wave theory. In this case two magnons with approximately equal wave-vector values form bound states. The binding energy of such states is determined from the experimental data.

The physical properties of one-dimensional and two-dimensional magnetic systems differ considerably from the properties of ordinary three-dimensional magnets. Such systems have a large number of peculiarities, many of which are connected with the anomalously large influence of the short-range order. In three-dimensional magnets, short-range order is observed in a narrow temperature interval near the phase-transition point. On the other hand, in one-dimensional and two-dimensional magnets the transition to the magnetically-ordered state is frequently observed at temperature much lower than zJ/k, where J is the exchange integral and z is the number of neighbors. Therefore in a sufficiently broad temperature interval, from the transition temperature up to zJ/k, a short-range order should be observed in such magnets, so that the motion of the neighboring spins is correlated, whereas at sufficiently large distances there is no such correlation. The present paper is devoted to an investigation of this state of magnets.

A large number of recently reported compounds can be regarded in first-order approximation as one-dimensional and two-dimensional magnets. These include two classes of layered compounds, in which the magnetic ions are coupled by ferromagnetic interaction. The first class consists of complex compounds of copper chloride, with a common formula $(C_nH_{2n+1}NH_3)_2CuCl_4$, where n = 1, 2, ..., 10. The divalent copper ions are arranged in the form of flat layers[1], between which the relatively long organic molecules are located. In each layer, the copper ions are connected with chlorine bridges and form an almost regular quadratic lattice. Exchange ferromagnetic interaction is observed between the neighboring magnetic ions within each layer[2,3]. The magnitude of this interaction is approximately the same for all the investigated compounds, and amounts to J/k ≈ 20° K. At the same time, antiferromagnetic interaction is observed between neighboring layers[4]. The magnitude of the interaction depends strongly on the organic molecules with which the complex copper-chloride compounds are formed, but for this entire class of compounds the ferromagnetic interaction inside the layer is larger by several orders of magnitude than the antiferromagnetic interaction between layers. Owing to this interaction, compounds of this class go over at sufficiently long temperatures into the magnetically ordered state. Above the phase-transition point, however, the magnetic properties of these compounds can be described by the Heisenberg two-dimensional model with isotropic exchange interaction, while at low temperatures it is necessary to take into account also the weak interaction between layers[2-4]. The second class of two-dimensional ferromagnets consists of layered compounds of graphite with transition-metal chlorides, in which single layers of the chlorides are separated by either one or several layers of graphite. The interaction between the magnetic ions of neighboring layers separated by one layer of graphite decreases in magnitude, but retains the same sign[5,6]. In compounds in which the paramagnetic layers are separated by two or more graphite layers, it was impossible to observe magnetic interaction between them. The properties of the compounds of graphite with $FeCl_2$ and $FeCl_3$ agree with the Ising two-dimensional model, while the compounds with $NiCl_2$ and $CoCl_2$ can be described in first order by the Heisenberg model[7,8].

We have investigated two compounds pertaining to different classes of two-dimensional ferromagnets. The complex compound of copper chloride with $(CH_3NH_3)_2CuCl_4$ has a face-centered rhombic unit cell with parameters $a_0 = 7.30$ Å, $b_0 = 7.54$ Å, and $c_0 = 18.55$ Å[1]. The copper ions in layers parallel to the ab plane are coupled by exchange ferromagnetic interaction with J/k = 19.2° K, while the interaction between two ions of neighboring layers is weaker by approximately four orders of magnitude[2]. We investigated single-crystal samples in the form of square plates measuring 2.5 × 2.5 × 0.3 mm, as well as spherical samples made up of such plates, with approximate diameter 2 mm. The crystals were grown from a solution of this compound in ethyl alcohol by the method of evaporation at an approximate temperature 50° C.

We investigated also the magnetic properties of a layered compound of graphite with $NiCl_2$. The nickel

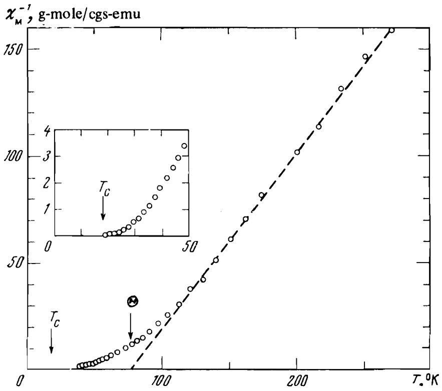

FIG. 1. Temperature dependence of the reciprocal susceptibility of a compound of $NiCl_2$ with graphite (per g-mole of $NiCl_2$) in a direction parallel to the layers.

chloride forms with graphite only a compound of degree II, in which the chloride layers are separated by two graphite layers. We investigated both polycrystalline samples with large chloride content (up to 50% by weight), and samples obtained by penetration of nickel chloride into oriented pyrolytic graphite. In the latter case, the chloride content in the sample was approximately 2% by weight. The investigated samples were spherical, of 3 mm diameter. The arrangement of the magnetic ions in the layer is not known for this compound, but by analogy with the investigated compounds of $FeCl_3$ and $MoCl_5$[9,10] it can be assumed that the structure of the single layer in $NiCl_2$ and in its compound with graphite is the same. The Ni^{+2} ions form a flat hexagonal lattice (z = 6) and the distances between them are 3.84 Å.

RESULTS

The magnetic susceptibility in a zero external field was measured by a bridge method at 175 Hz, and the amamplitude of the magnetic field at the sample did not exceed 2 Oe. At these values of the field, the susceptibility is independent of the magnitudes of the field and can be regarded as the limiting value as $H \to 0$. The sample temperature was measured with a thermocouple consisting of Au + 0.07 at.% Fe paired with Cu[11], one junction of which was in thermal contact with the sample and the other was immersed in liquid helium. The magnetization curves were plotted by a ballistic method, and fields from 0.5 to 60 kOe were produced by a superconducting solenoid. It is difficult to obtain controllable fields lower than 500 Oe with such a solenoid, in view of the residual fields produced under these conditions and due to the frozen-in flux, and the magnetic field is not a single-valued function of the current through the solenoid. Weak fields were therefore produced with a copper solenoid. The sample temperature was measured with an analogous thermocouple.

Figure 1 shows the temperature dependence of the magnetic susceptibility of a compound of $NiCl_2$ with graphite. At temperatures above 50° K, the susceptibility is isotropic, but actual anisotropy sets in at lower temperatures and the susceptibility parallel to the layers exceeds the susceptibility in the perpendicular direction. The anisotropy increases with decreasing temperature, so that at the critical temperature $T_c = 18.1°K$, corresponding to the maximum of the susceptibility, the values of $\chi_\parallel$ and $\chi_\perp$ differ by approximately one order of magnitude. For a polycrystalline sample, the molar susceptibility (per g-mole $NiCl_2$) in the entire temperature interval (18.1—300° K) is equal to the average value of the paramagnetic susceptibilities for the oriented sample: $\chi = (\chi_\perp + 2\chi_\parallel)/3$. The temperature of the transition to the magnetically-ordered state ($T_c = 18.1°K$) is also the same for both investigated samples. Thus, the magnetic properties of the compound of $NiCl_2$ with graphite did not depend on the amount of the penetrated $NiCl_2$ and are determined completely by the magnetic interaction of the Ni^{+2} ions inside the layer.

In the temperature region 110—270° K, the variation of the susceptibility can be described by the Curie-Weiss law with $\Theta = 75 \pm 5°$. It should be noted that $\Theta = 70°$ for pure $NiCl_2$[12]. This confirms the previously advanced hypothesis that the chloride layer has the same structure in pure $NiCl_2$ as in its compounds with graphite. Knowing Θ_c, we can determine the exchange integral with the nearest neighbors. In accordance with the molecular-field theory, we have

$$J/k = 3\Theta/2zS(S+1), \qquad (1)$$

whence J/k = 9.4° K. The magnetic susceptibility of the compound $(CH_3NH_3)_2CuCl_4$, was measured earlier[2]. From an analysis of the temperature dependence of the susceptibility at $T > T_c$, using high-temperature expansion, yielded the value J/k = 19.2°.

At sufficiently high temperatures, the magnetic moment of either investigated compound increases linearly with the increasing magnetic field. At low temperatures, however, the linear dependence is preserved only in weak fields, while in strong fields the dependence of the moment on the field is close to logarithmic. Figures 2 and 3 show the corresponding curves. Curve 1 of Fig. 3 was plotted in the magnetically order state at $T < T_c$. The antiferromagnetic state is destroyed in very weak fields, so that at H > 100 Oe the interaction between the layers can be neglected in first order. It should also be noted that according to our measurements the compound $(CH_3NH_3)_2CuCl_4$ goes over into the magnetically ordered

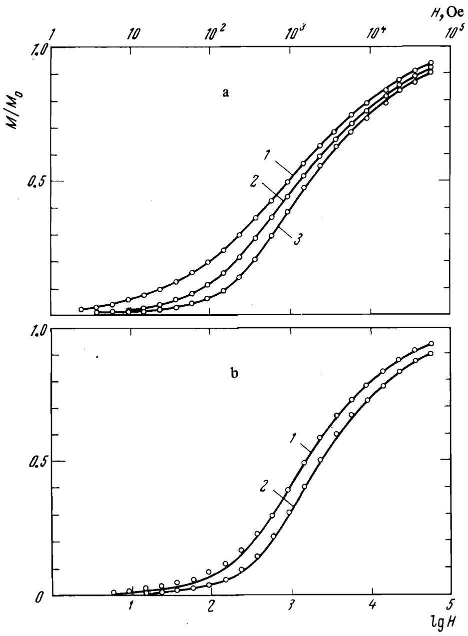

FIG. 2. Plots of the relative magnetization against the field for the compound of $NiCl_2$ with graphite at different temperatures: a) curve 1—18.1, 2—19.5, 3—21.0°K, magnetic field applied parallel to the layers; b) curve 1—18.1, 2—21.0°K, field perpendicular to the layers; circles—experimental results, the curves are drawn in accordance with formula (3).

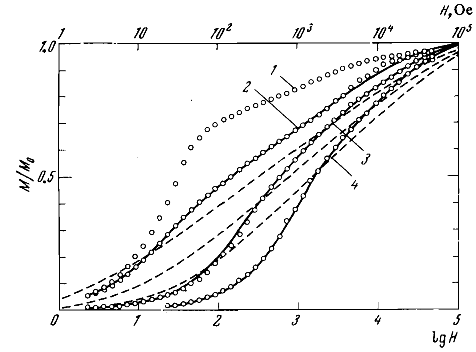

FIG. 3. Dependence of the relative magnetization on the field for the compound $(CH_3NH_3)_2CuCl_4$ at different temperatures (1—6.11, 2—8.61, 3—11.0, 4—11.35°K), magnetic field applied parallel to the layers; circles—experimental results, dashed curves—plots of (9) and (10), J/k = 14.5°K; solid lines—plots of (13) and (14) values of J/k given in Fig. 4, α = 80.

state at $T_c = 8.49°K$, whereas the susceptibility reaches a maximum at $8.91°K$[2]. This discrepancy can be due to the fact that first, different thermometers were used, and second, the transition temperature was determined by different methods. In our experiments we determined T_c by plotting M(T) in an external field from 1 to 30 Oe. For each value of the field we determined the temperature above which the magnetic moment decreased. The sequence of temperatures obtained in this manner was extrapolated to zero external field, and the obtained value was taken to be the transition temperature.

The anisotropy of the magnetic properties of the investigated compounds can be described by introducing a demagnetizing field. The distances between the magnetic ions within the layer are much smaller than the distances between layers, so that layered compounds can be regarded as sets of plane-parallel magnetic plates. The demagnetizing factors of a thin plate are equal to zero and 4π for parallel and perpendicular fields, respectively. If the field is parallel to the plane of the plate, then the internal field coincides with the external one, but if it is perpendicular to the plane, then the internal field is smaller than the external one by an amount $4\pi m$, where m is the density of the magnetic moment. For a single layer of magnetic ions, it is meaningless to introduce the density of the magnetic moments, and it is simpler to introduce the demagnetizing field H_d. We shall assume that the field inside and outside the layer are equal for parallel orientation, and in the perpendicular direction the internal field is $H - (M/M_0)H_d$.

Such an assumption is well confirmed experimentally. For a layered compound of graphite with $NiCl_2$, the magnetization curves were plotted at equal temperatures and different orientations of the magnetic field (Fig. 2). Assume that at a definite temperature the dependence of the moment on the field applied in the plane of the layer is given by a certain function:

$$M_{\parallel}(H) = f(H). \tag{2}$$

Then, for the same temperature, the dependence of the moment in the perpendicular direction should be expressed by the same function but with a different value of the argument:

$$M_{\perp}(H) = f\left(H - \frac{M}{M_0} H_d\right). \tag{3}$$

The curves in Fig. 2b were plotted in accordance with formula (3), the function f(H) is specified in Fig. 2a, and H_d was chosen equal to 1.20 kOe. A similar reduction of the data for $(CH_3NH_3)_2CuCl_4$ shows that in this case, too, the anisotropy of the magnetic properties at temperatures above T_c can be described by a demagnetizing field $H_d = 0.82$ kOe. In this case, however, the agreement between the calculated magnetization curve and the experimentally observed one is somewhat worse, particularly at low values of the moment. The possible reason is that the magnetic layers are separated here much less than in the compound of $NiCl_2$ with graphite.

DISCUSSION

As follows from Fig. 1, for two-dimensional ferromagnets, short-range order is observed in a wide range of temperatures, from $T = T_c$ all the way to $T = \Theta$. The range is from 18.1 to 70°K for the compound of $NiCl_2$ with graphite, and from 8 to 35°K for $(CH_3NH_3)_2CuCl_4$. Indeed, the thermal-motion energy becomes comparable with the energy that binds the individual magnetic ion with its own neighbors at $T = \Theta$. In three-dimensional ferromagnets, a phase transition takes place at $T = \Theta$, and at $T < \Theta$ these ferromagnets are in the ordered state. While no phase transition is observed at $T = \Theta$ in two-dimensional systems, the probability of the single-particle spin excitations decreases rapidly with decreasing temperature, and these excitations can be neglected at $T < \Theta$. The principal excitations that determine the properties of the system are the spin waves.

A formal application of spin-wave theory to the two-dimensional case[13] shows that for any finite temperature there exists in the system an infinite number of excitations. This extremely important conclusion of spin-wave theory proves the absence of ordered states in the two-dimensional Heisenberg model. In order to obtain with the aid of spin-wave theory concrete results that can be compared with experiment, it is necessary to make the following assumptions:

1) A two-dimensional ferromagnet is placed in an external magnetic field **H** is described by the Heisenberg Hamiltonian

$$\mathscr{H} = -2J \sum_{\text{б.с.}} \mathbf{S}_i \mathbf{S}_j - Hg\beta \sum_i S_i^z, \tag{4}$$

where the first sum extends over all the pairs of nearest neighbors, S is the spin-moment operator, g is the electronic g-factor, β is the Bohr magneton, and the direction of the field **H** is chosen to be the z axis. The ground-state energy corresponds to the ferromagnetic case:

$$E_0 = -Ng\beta SH - NzJS^2, \tag{5}$$

where N is the number of spins per unit area.

2) In the absence of a field, at any temperature $T_c < T < \Theta$, the number of excited spin waves is equal to the total spin NS of the system. This allows us to assume that there are no single-particle spin excitations in the system, and the magnetic moment without the field is equal to zero.

3) The excited spin waves obey Bose statistics, and the interaction between them can be initially be disregarded.

Taking the foregoing assumptions into account, the magnetic moment per unit area takes the form

$$\mathbf{M}(\mathbf{H}, T) = -\frac{\partial E_0}{\partial \mathbf{H}} - \frac{1}{4\pi^2} \int_{\mathbf{q}_1}^{\mathbf{q}_0} \frac{\partial \varepsilon(\mathbf{q}) / \partial \mathbf{H} \, d\mathbf{q}}{\exp[\varepsilon(\mathbf{q})/kT] - 1}, \tag{6}$$

where $\epsilon(\mathbf{q})$ is the energy of a spin wave with wave vector $\mathbf{q}$; $\mathbf{q}_0$ and $\mathbf{q}_1$ are the limiting momenta that determine the region of the allowed values of $\mathbf{q}$. The maximum momentum $\mathbf{q}_0$ corresponds to the boundary of the first Brillouin zone, and the minimum momentum $\mathbf{q}_1$ is obtained from the condition 2): M(0, T) = 0.

Let the field **H** be directed parallel to the plane of the layer and let the sample be located at a sufficiently low temperature near T_c. The energy of a spin wave with wave vector $\mathbf{q}$ in a flat quadratic lattice can be written in the form

$$\varepsilon(\mathbf{q}) = g\beta H + 2JSq^2a^2, \tag{7}$$

where a is the lattice constant. Substituting (7) in (6) and neglecting the small terms, we obtain

$$M = Ng\beta S + \frac{Ng\beta kT}{8\pi JS}\left\{\ln\left[\exp\left(\frac{g\beta H + 2JSq_1^2a^2}{kT}\right) - 1\right] - \frac{g\beta H + 2JSq_1^2a^2}{kT}\right\};$$
$$q_1^2a^2 = \frac{kT}{2JS}\exp\left(-\frac{8\pi JS^2}{kT}\right). \tag{8}$$

In weak fields $g\beta H/kT \ll 1$ expression (8) becomes simpler:

$$M=\frac{Ng\beta kT}{8\pi JS}\ln\left(1+\frac{H}{H^*}\right),\quad H^*=\frac{kT}{g\beta}\exp\left(-\frac{8\pi JS^2}{kT}\right). \tag{9}$$

In strong fields $g\beta H \gg 2JSq_1^2a^2$ we have

$$M=Ng\beta S-\frac{Ng\beta kT}{8\pi JS}\ln\left[1-\exp\left(-\frac{g\beta H}{kT}\right)\right]. \tag{10}$$

Comparison with experiment (Fig. 3) shows that the magnetization curves calculated from formulas (9) and (10) agree qualitatively with experiment, and one can hope that the proposed scheme describes the situation correctly in principle. The physical meaning of the assumptions made can be explained in the following manner. We introduce the correlation radius R_c, defined as

$$R_c=\frac{\pi}{q_1}=\pi\left(\frac{2JS}{kT}\right)^{1/2}\exp\left(\frac{4\pi JS^2}{kT}\right). \tag{11}$$

For each point on the plane of a two-dimensional ferromagnet there exists a region with radius R_c within which the motion of the spins is correlated, and the pair correlators for all the spins spaced by more than R_c apart are equal to zero. In such a system, the magnons can have a maximum wavelength $2R_c$, and magnons with longer wavelength have no physical meaning. We note that magnons with maximum possible wavelength of order $2R_c$ exert the major influence on the thermodynamic characteristics of the two-dimensional ferromagnet, since the energy of such excitations is minimal and approaches the ground state of the ferromagnet.

When the temperature is lowered, the correlation radius R_c increases exponentially (see (11)), and magnons with ever-increasing wavelengths are produced in the system. However, R_c becomes infinite only at T = 0, and spin waves with q = 0 are possible only at T = 0. Thus, long-range order appears only at T = 0, and the system is in the disordered state at any finite temperature. Real magnets, on the other hand, have finite dimensions, so that at a definite temperature T_c the correlation radius becomes comparable with the dimension of the investigated sample, and a phase transition should be observed in the system. In the compounds investigated by us, however, the transition to the magnetically ordered state occurs at temperatures much higher than T_c. This is caused by the difference between the real paramagnets and the idealized two-dimensional Heisenberg model. In one of them ($(CH_3NH_3)_2CuCl_4$), an important role is apparently played by weak interaction between magnetic layers, whereas the phase transition in the other compound ($NiCl_2$ with graphite) is due to the presence of weak hexagonal anisotropy in the plane of the layer.

In the derivation of formulas (9) and (10) it was assumed that the spin waves do not interact with one another. The agreement between the calculated magnetization curves and those obtained experimentally can be improved greatly by suitably accounting for this interaction. The behavior of the spin waves in the two-dimensional case differs in principle from the three-dimensional case. In the two-dimensional case, the lower excited states correspond to bound states of two magnons[14], which occur at any value of the wave vector **q**, whereas in the three-dimensional case there are no bound states at all at small values of **q**[13,15]. The energy of the bound state of two magnons can be expressed in general form as follows:

$$\varepsilon(\mathbf{q}',\mathbf{q}'')=\varepsilon(\mathbf{q}')+\varepsilon(\mathbf{q}'')-\Delta(\mathbf{q}',\mathbf{q}'',H,T),$$

where $\epsilon(\mathbf{q}')$ and $\epsilon(\mathbf{q}'')$ are the energies of the interacting magnons and Δ is the binding energy, which depends not only on the wave vectors $\mathbf{q}'$ and $\mathbf{q}''$ but also on the temperature and on the external magnetic field. The form of the function $\Delta(\mathbf{q}', \mathbf{q}'', H, T)$ is not known for a two-dimensional ferromagnet, but the experimental results can be described by a relatively simple method. It was necessary for this purpose to assume that the magnons are excited in a two-dimensional system in pairs, and the energy of such a pair in a flat quadratic lattice is equal to

$$\varepsilon_2=2g\beta H+4JSq_1^2a^2-4\alpha JSq_1^2a^2M/M_0, \tag{12}$$

where α is a constant. The first two terms here are equal to double the energy of a magnon with wave vector **q** (7), and the last term is the binding energy. Expression (12) is apparently a rather crude approximation, valid only for the lowest-lying energies of the spin waves with wave vectors near the limiting momentum q_1. This is precisely why the binding energy is proportional to the expression $q_1^2a^2$ given in (8). The linear dependence of the binding energy on the magnetic moment is also understandable, since the interaction of two magnons comes into play when they have close values of **q** and propagate in phase with each other over a considerable distance $R > R_c$; the relative moment M/M_0 is a measure of the coherence length. The meaning of formula (12) becomes quite clear if it is assumed that bound states are realized in a two-dimensional ferromagnet for magnons with equal but opposite wave vectors **q** and −**q**. Such an assumption, however, calls for a theoretical analysis and for experimental verification.

Taking (12) into account, we obtain for the dependence of the moment on the temperature and on the external field

$$M=\frac{Ng\beta kT}{8\pi JS}\ln\left(1+\frac{2H}{H^*}-\alpha_0\frac{M}{M_0}\right), \tag{13}$$

where H* is determined from (9), and $g\beta H/kT \ll 1$. In strong fields, when $g\beta H \gg 2\alpha JSq_1^2a^2$, we have

$$M=Ng\beta S-\frac{Ng\beta kT}{8\pi JS}\ln\left[1-\exp\left(-\frac{2g\beta H}{kT}\right)\right]. \tag{14}$$

Relations (13) and (14) describe very well the experimental results (Fig. 3). In strong fields, M(H) is a function of one parameter, the exchange integral J. A detailed comparison of (14) with experiment shows that the parameter J increases with decreasing temperature (Fig. 4). Linear extrapolation yields J/k = 18.5 ± 1.5°K at T = 0. The obtained value agrees both with the results of the study of the magnetic susceptibility of $T > T_c$ (J/k = 19.2° K[2]) and with the behavior of the specific

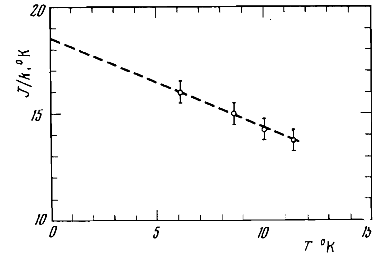

FIG. 4. Temperature dependence of J/k for the compound $(CH_3NH_3)_2CuCl_4$.

heat at $T < T_c$ ($J/k = 17.8°$K[3,16]). The J(T) relation obtained by us is perfectly natural, since we used linear spin-wave theory in the discussion of the results. Allowance for the nonlinear corrections leads to a renormalization of the spin-wave spectrum and to the dependence of their energy on the temperature[17,18]. In purely formal fashion this is described by a decrease of the exchange integral, when the energy of the spin waves becomes comparable with the exchange energy.

For a layered compound of $NiCl_2$ with graphite, the energy of the two bound magnons can be expressed in the form

$$\varepsilon_2 = 2g\beta H + 6JSq^2a^2 - 6\alpha JSq_1{}^2a^2M / M_0, \quad (15)$$

and the dependence of the moment on the field and on the temperature in weak fields, at $g\beta H/kT \ll 1$, takes the form

$$M = \frac{\sqrt{3}\,Ng\beta kT}{24\pi JS}\ln\left(1 + \frac{2H}{H^*} - \alpha\frac{M}{M_0}\right), \quad (16)$$

where

$$H^* = \frac{kT}{g\beta}\exp\left(-\frac{24\pi JS^2}{\sqrt{3}\,kT}\right),$$

and in strong fields $g\beta H \gg 3\alpha JSq_1^2a^2$ we get

$$M = Ng\beta S - \frac{\sqrt{3}\,Ng\beta kT}{24\pi JS}\ln\left[1 - \exp\left(-\frac{2g\beta H}{kT}\right)\right]. \quad (17)$$

Relations (16) and (17) describe well the magnetization curves shown in Fig. 2a, the value of the exchange integral in this case is $J/k = 4.0°$K, and $\alpha = 85$. We call attention to a difference, by more than a factor of 2, between this exchange integral and the value $J/k = 9.4°$K obtained from the temperature dependence of the susceptibility at $T \gg T_c$. One of the possible causes of such a behavior may be the noticeable exchange interaction of the nickel ions with the neighbors from the second coordination sphere.

In conclusion, I am deeply grateful to A. S. Borovik-Romanov for a useful discussion of the results and to Yu. N. Novikov for supplying the samples of the $NiCl_2$ compound with graphite.

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