Anomalies of magnetic properties of dilute $Mn_{1-x}Zn_xF_2$ and $Ni_{1-x}Mg_xF_2$ antiferromagnets below the percolation threshold

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It has been observed that with lowering of temperature the magnetic moments of $Mn_{0.2}Zn_{0.8}F_2$ and $Ni_{0.2}Mg_{0.8}F_2$ single crystals acquire a nonlinear dependence on the applied magnetic field and that the law governing the temperature dependences of their magnetic susceptibilities changes from $\chi(T) \propto 1/(T+\theta)$ to $\chi(T) \propto 1/T$. These anomalies are due to formation of magnetic states with finite correctional radii (magnetic clusters) in these single crystals. The difference between the anomalies of the magnetic properties of $Mn_{0.2}Zn_{0.8}F_2$ and $Ni_{0.2}Mg_{0.8}F_2$ is caused by the presence in the latter of a Dzyaloshinskiĭ interaction acting in the (001) plane and comparable with the exchange interaction at these magnetic-ion concentrations.

The properties of $M_{1-x}Zn_xF_2$ (M = Mn, Co, Ni) single crystals are studied at present by various methods.[1–4] These investigations have shown that at a magnetic-ion concentration higher than the percolation threshold $1-x_c = 0.25$ there are produced in $M_{1-x}Zn_xF_2$ antiferromagnetic states corresponding to the antiferromagnetic states of MF_2 but with a lower Néel temperature than in the latter. The anomalies of these states are governed by the random distribution of the interacting magnetic ions in $M_{1-x}Zn_xF_2$. Investigations of $M_{1-x}Zn_xF_2$ samples with the aid of neutron scattering[4,5] have shown that the anomalies of their antiferromagnetic properties are due to the onset of magnetic states with finite correlation radii. At magnetic-ion concentrations lower than the percolation threshold $1-x<1-x_c$ no antiferromagnetic states are produced in $M_{1-x}Zn_xF_2$, but the states produced when the temperature is lowered differ strongly from the ordinary paramagnetic states.

The purpose of the present study was an investigation of the magnetic properties of dilute $Mn_{1-x}Zn_xF_2$ and $Ni_{1-x}Mg_xF_2$ antiferromagnets in the absence of antiferromagnetic states, i.e., at $1-x<1-x_c$. The difference between the chosen substances is that the Dzyaloshinskiĭ interaction that leads to weak ferromagnetism in antiferromagnets is absent in $Mn_{1-x}Zn_xF_2$ but is present in $Ni_{1-x}Mg_xF_2$. We have previously[3,6] investigated $Mn_{1-x}Zn_xF_2$ and $Ni_{1-x}Mg_xF_2$ single crystals near the percolation threshold $1-x_c$, but with $x<x_c$, i.e., in the antiferromagnetic state. In the investigation of the $Ni_{1-x}Mg_xF_2$ single crystals it was observed that in the antiferromagnetic state the antiferromagnetism vector **L** is oriented along the [100] or [010] axis and a weak ferromagnetism σ_D due to the Dzyaloshinskiĭ interaction is produced and is oriented perpendicular to **L** along the [010] or [100] axis, respectively. It was shown that the Dzyaloshinskiĭ interaction H_D is independent of the concentration of the nonmagnetic ions, in contrast to the average exchange interaction H_E. At nonmagnetic-ion concentrations $x \approx x_c$ it was found that $H_D \leqslant H_E$ in $Ni_{1-x}Mg_xF_2$. An investigation of the magnetic properties of $Mn_{1-x}Zn_xF_2$ and $Ni_{1-x}Mg_xF_2$ makes it possible to determine the anomalies produced by the Dzyaloshinskiĭ interaction in the magnetic properties of dilute antiferromagnets.

We investigated the dependences of the magnetic moments of the samples on the applied magnetic field at various temperatures. We used a magnetometer with vibrating sample, magnetic fields up to 65 kOe, and temperatures up to 100 K (Ref. 7).

To study the anomalies of the relaxation of magnetic states in dilute antiferromagnets, we investigated the dependence of the magnetic susceptibility χ of $Mn_{0.2}Zn_{0.8}F_2$ measured in weak magnetic fields, on the frequency ν of the rf magnetic field. Owing to the difficulty of growing sufficiently large $Ni_{1-x}Mg_xF_2$ single crystals, their $\chi(\nu)$ was not investigated. The $\chi(\nu)$ dependences were measured only by a modulation procedure[8] in a frequency range up to 10^8 Hz at $T = 4.2$ K. The calibrating reference signal at high frequencies was the value of χ of single-crystal MnF_2, whose magnetic susceptibility is independent of ν up to frequencies on the order of 10^{11}–10^{12} Hz.

MEASUREMENT RESULTS

Figure 1 shows the dependences of the magnetic moment on the applied magnetic field for single-crystal $Mn_{0.2}Zn_{0.8}F_2$ at various temperatures. It can be seen that

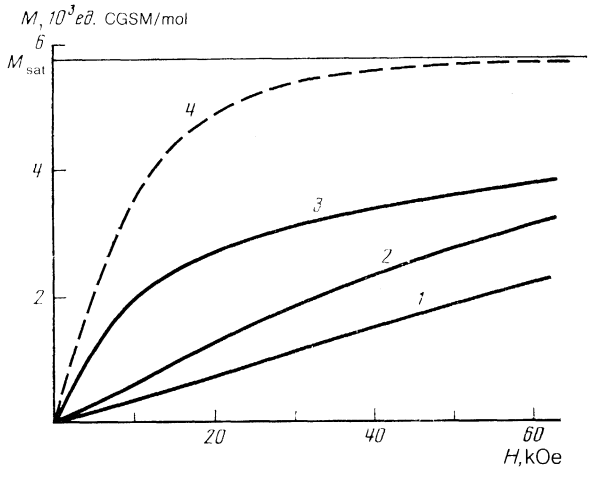

FIG. 1. Dependences of the magnetic moment of $Mn_{0.2}Zn_{0.8}F_2$ on the applied magnetic field at various temperatures T: *1*—11.3, *2*—4.2; *3*—3.1 K; theoretical plot of $M(H) = M_0 B_J(g\mu_B JH/kT)$ for $J = 5/2$ and $T = 2$ K, calculated under the assumption that $Mn_{0.2}Zn_{0.8}F_2$ is in the paramagnetic state; M_{sat} is the magnetic moment of saturated $Mn_{0.2}Zn_{0.8}F_2$.

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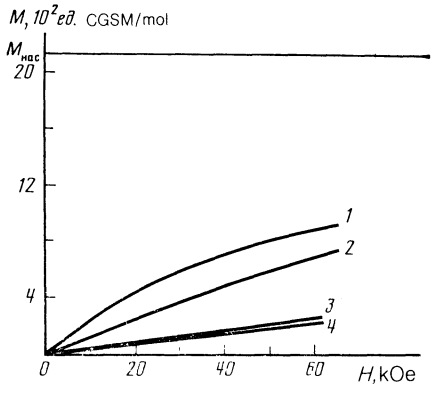

FIG. 2. Magnetic moment of $Ni_{0.2}Mg_{0.8}F_2$ vs the applied magnetic field at various temperatures and orientations of **H** relative to the single-crystal axes: $T = 4.2$ **H**$\|[100]$—curve *1*, $T = 4.2$ K, **H**$\|[001]$—curve *2*, $T = 40$ K, **H**$\|[100]$—curve *3*, $T = 40$ K, **H**$\|[001]$—curve *4*. M_{sat}—magnetic moment at saturation.

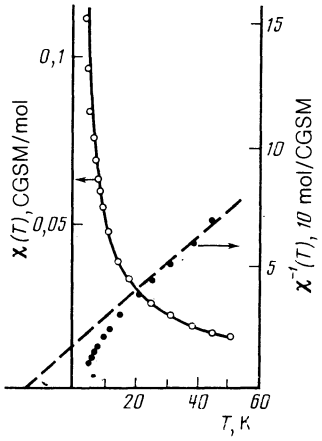

FIG. 3. Temperature dependences of the magnetic susceptibility (○) and of the reciprocal susceptibility (●) of $Mn_{0.2}Zn_{0.8}F_2$, measured in weak magnetic fields $H \to 0$.

when the sample temperature is lowered a nonlinear $M(H)$ dependence sets in, describable in the case of strong magnetic field by the expression $M(H) = M^* + \chi H$, where χ is different from zero. A linear dependence $M(H) = \chi^*(T)H$ is observed in weak magnetic fields. The nonzero magnetic susceptibility χ measured in strong magnetic fields indicates that the sample is not in the usual paramagnetic state. Figure 1 shows $M(H,T)$ plots with the magnetic field applied along a binary axis of the $Mn_{0.2}Zn_{0.8}F_2$ crystal. Investigations have shown that the form of the $M(H,T)$ dependence is almost unaffected by variation of the orientation of **H** relative to the $Mn_{0.2}Zn_{0.8}F_2$ crystal axes.

Figure 2 shows $M(H,T)$ plots for single-crystal $Ni_{0.2}Mg_{0.8}F_2$ at various orientations of **H** relative to the crystal axes. It is seen from the figure that when the temperature is lowered a nonlinear $M(H)$ dependence arises only when **H** is perpendicular to the tetragonal axis [001]; the shape of the curve does not depend on the **H** orientation in a plane perpendicular to this axis. At low temperatures the curves for magnetization along and perpendicular to the [001] axis differ greatly from one another, whereas in high temperatures there is no such difference. At high temperatures the dependences are linear, $M(H,T) = \chi(T)H$, and are practically independent of the orientation of **H**. It is seen from Fig. 2 that at low temperatures the $M(H)$ dependence is linear, when **H** is oriented along the [100] axis, only in magnetic fields $H < 10$ kOe, beyond which $M(H)$ is nonlinear. With **H** oriented along the tetragonal [001] axis, $M(H)$ is almost linear. A small deviation of $M(H)$ from linearity at **H**$\|[001]$ ($\Delta M/M \approx 8\%$) is observed only at $H > 45$ kOe.

With increase of the magnetic-ion concentration, a deviation from a linear $M(H) = \chi(T)H$ dependence for **H** oriented in the (001) plane takes place in stronger fields. The linearity of $M(H,T)$ is hardly disturbed at **H**$\|[001]$.

Figure 3 shows for single-crystal $Mn_{0.2}Zn_{0.8}F_2$ the temperature dependences of the magnetic susceptibility and of the reciprocal of the magnetic susceptibility measured in weak magnetic fields. It is seen from Fig. 2 that when the

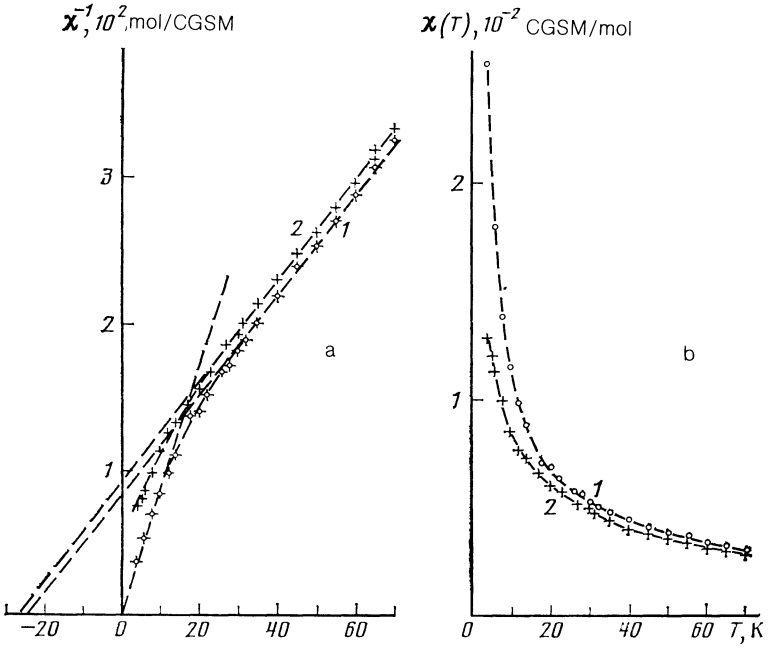

FIG. 4. a) Temperature dependences of the reciprocal magnetic susceptibilities, measured in weak magnetic fields, for $Ni_{0.2}Mg_{0.8}F_2$ at orientation **H**$\|[100]$ (curve 1) and **H**$\|[001]$ (curve 2); b) temperature dependences of magnetic susceptibilities measured in weak magnetic fields for $Ni_{0.2}Mg_{0.8}F_2$ at orientations **H**$\|[100]$ (1) and **H**$\|[001]$ (2).

temperature is lowered the law governing the temperature dependence changes from $\chi(T) \propto 1/(T+\theta)$ (where $\theta = 15 \pm 2$ K) observed at $T > 30$ K to $\chi(T) \propto 1/T$ observed at $T < 6$ K. The shapes of the $\chi(T)$ and $\chi^{-1}(T)$ curves for $Mn_{0.2}Zn_{0.8}F_2$ are practically independent of the magnetic-field orientation. A change in the law governing the temperature dependence of the magnetic susceptibility $\chi(T)$ is observed in all the investigated dilute antiferromagnets.

The measured temperature dependences of the magnetic susceptibility and of the inverse magnetic susceptibility of $Ni_{0.2}Mg_{0.8}F_2$ in weak magnetic fields $H < 5$ kOe at various magnetic-field orientations relative to the crystal are shown in Fig. 4. It is seen from Fig. 4a that the $\chi^{-1}(T)$ dependences for $Ni_{0.2}Mg_{0.8}F_2$ single crystals at the orientations $\mathbf{H} \| [100]$ and $\mathbf{H} \| [001]$ differ qualitatively. For magnetic-field orientations in the (001) plane, the temperature dependence of the magnetic susceptibility changes when the temperature is lowered, from $\chi^{-1}(T) \propto (T+\theta)$ (where $\theta = 25 \pm 2$ K) at $T > 30$ K to $\chi^{-1}(T) \propto T$ at $T < 8$ K. At $T < 8$ K and $\mathbf{H} \perp [001]$ a $\chi^{-1}(T) \propto T$ dependence is observed. With the magnetic field oriented along the [001] axis, lowering the temperature causes the temperature dependence to deviate from the $\chi^{-1}(T) \propto T(+\theta)$ law (where $\theta = 27 \pm 2$ K) observed at high temperature; no transition to the $\chi^{-1}(T) \propto T$ law, however, is observed. When the concentration of the nonmagnetic ions in $Ni_{1-x}Mg_xF_2$ is increased at an orientation $\mathbf{H} \| [001]$, the deviation from the $\chi^{-1}(T) \propto [T+\theta(x)]$ law becomes more appreciable.

DISCUSSION OF RESULTS

In investigations of the magnetic properties of single-crystal $Mn_{1-x}Zn_xF_2$ or $Ni_{1-x}Mg_xF_2$ it is necessary to monitor the concentration of the magnetic ions in the samples and the uniformity of their distribution in the volume. The concentration of the magnetic ions in $M_{1-x}Zn_xF_2$ crystals can be determined by comparing the slopes of the $\chi^{-1}(T)$ plots of $M_{1-x}Zn_xF_2$ and MF_2 samples at high temperatures $T \gg T_N$. The concentration of the Ni^{2+} ions in $Ni_{0.2}Mg_{0.8}F_2$ was determined also by x-ray structure analysis from the change of the lattice constants of the investigated samples comapred with NiF_2 and MgF_2.[1)]

The homogeneity of the magnetic-ion distribution in the samples was estimated from the value of the constant θ in the Curie–Weiss law $\chi(T) \propto 1/(T+\theta(x))$ for the magnetic susceptibility, since θ depends on the distribution of the magnetic ions in the crystals. The most perfect crystals are apparently those in which the probability of finding a magnetic ion at a given crystal-lattice site is the same for all sites and is equal to the concentration x of the magnetic ions. The probability of the magnetic ion having $n \leqslant 8$ nearest neighbors is then determined by the binomial distribution

$$P(n) = \frac{8!}{(8-n)!\,n!} x^n (1-x)^{8-n}.$$

With this distribution it is possible to calculate the concentration dependence of the quantity θ in the Curie–Weiss law for $\chi(T)$; we obtain

$$\theta = \sum_{n=1}^{\infty} np(n)\theta_2,$$

where θ_2 is a quantity determined by the exchange interaction of only two neighboring magnetic ions. The summation is over eight nearest neighbors. The dependence of θ on the magnetic-ion concentration is determined here by the formula $\theta(x) = x\theta_0$, where θ_0 is the value of θ for MF_2.

Determination of $\theta(x)$ of single-crystal $Mn_{1-x}Zn_xF_2$ and $Ni_{1-x}Mg_xF_2$ and comparison with the linear law $\theta(x) = x\theta_0$ (Ref. 9) can serve as the criterion for classifying single crystals by the magnetic-ion distribution in them. Deviation of the experimental θ from the linear $\theta(x)$ law indicates both the presence of magnetic-ion clusters and the presence, in excess over the binomial distribution, of free magnetic ions surrounded only by Zn^{2+} or Mg^{2+} ions. In the investigated crystals, the deviations from the calculated $\theta(x)$ were insignificant.

The main anomalies of the magnetic properties of the investigated single crystals are the onset, when the temperature is lowered, of nonlinear dependences of the magnetic moments $M(H,T)$ on the applied magnetic field, which have in strong magnetic fields (Figs. 1 and 2) a nonzero magnetic susceptibility, and also the change of the temperature dependence of the magnetic susceptibilities measured in weak magnetic fields, from $\chi(T) \propto 1/[T+\theta(x)]$, observed at high temperatures to $\chi(T) \propto 1/T$ at low ones.

The dependence of the magnetic susceptibility on temperature and magnetic field for a system of interacting magnetic ions was obtained in Ref. 10, and is of the form

$$\chi(H,T) = \frac{1}{NkT}\sum_{i,j}[\langle S_iS_j\rangle_T]_{av} - \left\{\frac{3H^2}{N(kT)^3}\sum_{i,j,k,l}\left[\langle S_iS_j\rangle_T\langle S_kS_l\rangle_T - \frac{1}{3}\langle S_iS_jS_kS_l\rangle_T\right]_{av}\right\}, \quad (1)$$

where $\langle \ldots \rangle_T$ denotes averaging over the temperature and $[\ldots]_{av}$ averaging over the configurations. The onset of a linear $\chi^{-1}(T)$ dependence (Figs. 3 and 4) and its deviation from $\chi^{-1}(T) \propto (T+\theta)$ when the temperature is lowered, as well as the onset of a nonlinear $M(H,T)$ dependence, are due to the onset of nonzero correlation functions of the magnetic ions.[1] Using the following expansion of the magnetic moment $M(H,T)$ in powers of H

$$M(H,T) = \chi_1(T)H - \chi_3(T)H^3 + \chi_5(T)H^5,$$

we can determine the contributions to the magnetic susceptibilities from the various correlators contained in Eq. (1).

Let us examine the onset of which states of the investigated magnetic systems can account for the observed features of the magnetic properties of $M_{1-x}(Zn, Mg)_xF_2$. To verify the assumption that all the magnetic ions in $Mn_{0.2}Zn_{0.8}F_2$ can be paramagnetic, we have plotted the magnetic moment vs the magnetic field at $T = 2$ K:

$$M^*(H,T) = M_0B_J\left(\frac{g\mu_BJH}{kT}\right) \quad (2)$$

for $J = 5/2$ (Mn^{2+} ions), as shown by curve 4 of Fig. 1. It is seen from this figure that the plot of $M^*(H,T)$ as given by Eq. (2) differs greatly from the experimental plot, indicating that the Mn^{2+} ions in $Mn_{0.2}Zn_{0.8}F_2$ are not purely paramagnetic. A plot of Eq. (2) for $Ni_{0.2}Mg_{0.8}F_2$ at orientation

$\mathbf{H}\|[100]$ and $J=1$ (Ni^{2+} ions) deviates significantly from the experimental plot.

The nonzero magnetic susceptibility observed in strong magnetic fields both for $Mn_{0.2}Zn_{0.8}F_2$ and for $Ni_{0.2}Mg_{0.8}F_2$ (see Figs. 1 and 2), and the fact that the sample magnetization in fields $H\approx 60$ kOe is still far from saturation, indicate that the samples magnetic-moment saturation should take place in magnetic field much stronger than in the case of paramagnetic state. The assumption that one part of the ions are antiferromagnetically ordered and another paramagnetically is likewise not in accord with the experimental data for either $Mn_{0.2}Zn_{0.8}F_2$ or $Ni_{0.2}Mg_{0.8}F_2$ inasmuch as ordering in $Mn_{0.2}Zn_{0.8}F_2$ should lead to a maximum on the $\chi(T)$ plot if the magnetic field is oriented along a tetragonal axis, and to different magnetic susceptibilities at $\mathbf{H}\| C_4$ and $\mathbf{H}\perp C_4$, whereas in $Ni_{0.2}Mg_{0.8}F_2$ there should be observed a weak ferromagnetism $\sigma_{D\perp}$ (Ref. 3).

The anomalies of the magnetic properties of $M_{1-x}(Zn, Mg)_xF_2$ might be attributed to the presence of Mn^{2+} or Ni^{2+} magnetic ions surrounded only by nonmagnetic Zn^{2+} or Mg^{2+} ions, the number of which can be determined from the binomial distribution. The magnetic susceptibility of such Mn^{2+} (Ni^{2+}) ions is $\chi(T)\propto 1/T$. This assumption, however, does not accord with the experimental results for $Ni_{0.2}Mg_{0.8}F_2$ which are shown in Fig. 2 and in Figs. 4a and 4b, since a strong difference is observed between the $M(H,T)$ and $\chi(T)$ dependences in the cases $\mathbf{H}\|[100]$ and $\mathbf{H}\|[001]$ at low temperatures. In the case of $Mn_{0.2}Zn_{0.8}F_2$ an interpretation of the experimental results (Figs. 1 and 3) on the basis of this assumption leads to the conclusion that the investigated samples have an appreciable (up to 50%) content of free Mn^{2+} ions, which does not agre with the observed value of the constant θ in the Curie–Weiss law.

The shapes of the magnetization curves $M(H,T)$ at low temperatures (Figs. 1 and 2) as well as the change of the $\chi(T)$ dependences when the temperature is lowered (Figs. 3 and 4), can be explained by assuming that at lower temperatures there are produced magnetic states with finite dimensions and correlation radii— magnetic clusters—which have a "superparamagnetic" behavior. According to the picture of the distribution of the magnetic ions in the crystals, the investigated samples contain up to 16% of free magnetic ions surrounded only by Zn^{2+} or Mg^{2+} ions, up to 30 magnetic ions having only one nearest magnetic ion, and up to 30% magnetic ions having two nearest magnetic ions; the remaining 24% correspond to magnetic ions having 3, 4, or 5 nearest magnetic neighbors. Magnetic ions having more than one nearest magnetic neighbor can form magnetic clusters of finite size, since the concentration of the magnetic ions is lower than the percolation threshold.

Such a distribution of the magnetic ions governs the distribution of the exchange interaction over the volume in crystals, which determines in turn the observed average value of θ in the Curie–Weiss law for $\chi(T)$. The formation, when the temperature is lowered, of noninteracting magnetic clusters having superparamagnetic properties causes the temperature dependence of the magnetic susceptibility to change from $\chi^{-1}\propto T+\theta$ to $\chi^{-1}\propto T$, since the magnetization of a superparamagnet is described by a Brillouin function. The sample nonzero magnetic susceptibility observed in strong magnetic fields (see Figs. 1 and 2) is determined by the magnetic susceptibility of the clusters if they are magnetized along the applied magnetic field. The cluster susceptibility is determined by the average exchange interaction $\langle H_E\rangle$ within the magnetic clusters. Saturation of the magnetization curves should be observed in this case in fields $H\approx\langle H_E\rangle$. For $Mn_{0.2}Zn_{0.8}F_2$ and $Ni_{0.2}Mg_{0.8}F_2$ we have $\langle H_E\rangle\approx 120$ and ≈ 200 kOe, respectively.

Analysis of the nonlinear dependences of the magnetic moment on the field H for the investigated single crystals shows that in magnetic fields up to 20 kOe this dependence can be written in the form

$$M(H,T)=\chi_1(T)H-\chi_3(T)H^3.$$

Plots of $\chi_1(T)$ for the investigated single crystals are shown in Figs. 3 and 4. The determination of the $\chi_3(T)$ temperature dependence is limited by the accuracy of the determination of $\chi_1(T)$ in the magnetic-field range in which the deviation ΔM of $M(H,T)$ from linearity is small: $\Delta M/M\approx 10\%$. The deviation of $\chi_3(T)$ from the $\chi_3(T)\propto 1/T^3$ law can, according to Eq. (1), be determined by the temperature variation of the correlation radii of the produced magnetic clusters. Unambiguous conclusions, however, can be drawn only after performing direct neutron-diffraction investigations of the dependences of the magnetic-cluster correlation radii on the magnetic field and on the temperature. Investigations using small-angle neutron scattering,[5] as already indicated, have shown that when the temperature is lowered there are produced in $M_{1-x}Zn_xF_2$ with $x<x_c$ states with finite correlation radii—magnetic clusters.

With increase of the concentration of the nonmagnetic ions in $Ni_{1-x}Mg_xF_2$ the average exchange interaction decreases, and the influence of the Dzyaloshinskiĭ interaction increases. The random distribution of the magnetic ions in the crystal leads to a disordered orientation of the magnetic moments of Ni^{2+} in the (001) plane perpendicular to the tetragonal axis of the crystal. The Dzyaloshinskiĭ interaction in $Ni_{1-x}Mg_xF_2$ acts in the (001) plane and possibly prevents the magnetic moments from leaving the plane; this would explain the difference between the magnetic properties of $Ni_{1-x}Mg_xF_2$ for $\mathbf{H}\|[001]$ and $\mathbf{H}\perp[001]$. There is no Dzyaloshinskiĭ interaction in $Mn_{1-x}Zn_xF_2$ single crystals. When the temperature is lowered, magnetic clusters can then be formed with arbitrary orientation of the magnetic-ion magnetic moments. In this case the properties of the anomalies of the magnetic properties are practically independent of the orientation of $\mathbf{H}$ relative to the crystal axes, as is in fact observed in experiment.

The formation of magnetic states with finite correlation radii at low temperatures is accompanied by a change of the magnetic-system relaxation. This relaxation determines the frequency dependence of the magnetic susceptibility of the system. We have measured, for $Mn_{0.2}Zn_{0.8}F_2$ samples at $T=4.2$ K, the dependences of the magnetic susceptibility, measured in weak magnetic fields, on the frequency ν of the applied rf magnetic field in the range from 10^2 to 10^8 Hz. This $\chi(\nu)$ dependence is shown in Fig. 5. An investigation of the frequency dependences of the magnetic susceptibilities of several $Mn_{0.2}Zn_{0.8}F_2$ samples grown under various conditions, with different crystal cooling time after growth (corresponding to their annealing) has shown that the $\chi(\nu)$ dependence is governed by the distribution of the magnetic moments in them, and is determined by the prior history of the samples.

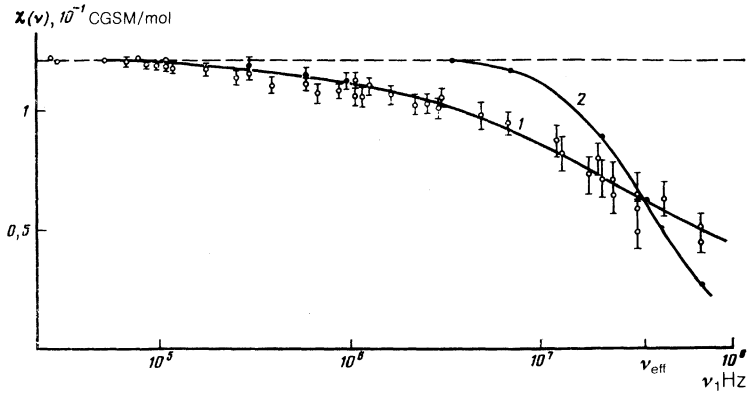

FIG. 5. Magnetic susceptibility of $Mn_{0.2}Zn_{0.8}F_2$, measured in weak magnetic fields, vs the frequency of the rf magnetic field at $T = 4.2$ K (curve 1). Curve 2—calculated $\chi(\nu)$ dependence assuming a relaxation law $M(t) = M(0)\exp[-(t/\tau)]$.

The $\chi(\nu)$ dependence obtained by us in Ref. 6 changed after the annealing, and for the investigated samples it has the form shown in Fig. 5. It appears that in the $Mn_{0.2}Zn_{0.8}F_2$ crystal investigated in Ref. 6 the magnetic ions were inhomogeneously distributed, for no such $\chi(\nu)$ dependence was observed in the succeeding measurements. The single crystals investigated in the present study have a more uniform, close to binomial, magnetic-ion distribution. The influence of the prior history on the frequency dependence of the magnetic susceptibility of samples with randomly distributed interacting magnetic ions, of the spin-glass type, was noted in a number of studies, including in the review of Ref. 11. We have investigated $\chi(\nu)$ for $Mn_{0.2}Zn_{0.8}F_2$ up to frequencies of order 10^8 Hz. With our apparatus it is practically impossible to investigate by the modulation method the frequency dependence of the magnetic susceptibility at frequencies higher than 10^8 Hz.

Inasmuch as in real crystals the magnetic clusters produced when the temperature is lowered can have different correlation radii, depending on the magnetic-ion distribution, it is also possible to have in them a distribution of various relaxaiton times $\tau_0,\ldots,\tau_{max}$. It is shown in Ref. 11, for systems having a relaxation-time distribution $\tau_0,\ldots,\tau_{max}$ that the dependence of the physical quantities on the time, in contrast to the usual relaxation law

$$M(t)=M(0)\exp(-t/\tau),$$

shown in Fig. 5, is given by

$$M(t)=M(0)\exp[-(t/\tau)^{\gamma}],$$

where $0<\gamma<1$. Such a relaxation law was observed in spin glasses.

Our computer calculations of the frequency dependence of the Fourier transformation given in Ref. 11

$$\frac{\partial}{\partial t}M(t)=\frac{\partial}{\partial t}\left\{M(0)\exp\left[-\left(\frac{t}{\tau_{\text{eff}}}\right)^{\gamma}\right]\right\}=F(\nu),$$

corresponding to a frequency dependence of the magnetic susceptibility, for different values of τ_{eff} and γ, and a comparison with experiment, have shown that the $\chi(\nu)$ dependence shown in Fig. 5 can be described by the relaxation law

$$M(t)=M(0)\exp\left[-\left(\frac{t}{\tau_{\text{eff}}}\right)^{\gamma}\right],$$

where $\tau_{\text{eff}}\approx 2.5\cdot 10^{-8}$ s and $\gamma\approx 0.4$–0.5. It must be indicated once more, however, that the distribution of the interacting magnetic ions in the crystals and that the dependences of τ_{eff} and of γ on such a distribution as well as on the prior history of the samples[1)] are decisive in such investigations. To study the relaxation it is necessary to investigate directly the magnetic-ion distribution in crystals, a task of considerable difficulty.

We have shown thus that in single-crystal $Mn_{0.2}Zn_{0.8}F_2$ and $Ni_{0.2}Mg_{0.8}F_2$ at magnetic-ion concentrations lower than the percolation limit $1-x_c=0.25$ there are produced, when the temperature is lowered, magnetic states with finite correlation radii—magnetic clusters. These states have superparamagnetic properties. In contrast to $Mn_{0.2}Zn_{0.8}F_2$ single crystals, where the observed anomalies of the magnetic properties are practically independent of the orientation of **H** relative to the crystal axes, in $Ni_{0.2}Mg_{0.8}F_2$ the observed singularities of the magnetic properties depend substantially on the **H** orientation. This difference is due to the presence in the $Ni_{1-x}Mg_xF_2$ single crystals of a Dzyaloshinskiĭ interaction that is comparable at such concentrations of the Ni^{2+} ions with the exchange interaction of magnetic ions.

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