Magnetic susceptibility of the dilute antiferromagnet $Mn_{0.2}Zn_{0.8}F_2$

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The magnetic susceptibility of $Mn_{0.2}Zn_{0.8}F_2$ single crystals as measured in weak fields is found to depend on the frequency of the modulating field at temperatures $T < T_f = 6.5 \pm 0.2$ K. Analysis of the experimental data shows that in the absence of magnetic field a "transient" spin-glass state arises in this compound at $T < T_f$.

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It has been shown[1-4] that the replacement of the magnetic ions Mn^{++} by Zn^{++} in the crystal lattice of $Mn_{1-x}Zn_xF_2$ progressively lowers the temperature of the transition to the antiferromagnetic state, until at concentrations $x > x_c = 0.75 \pm 0.02$ this transition is no longer observed. It was shown by Petrov and the author[2] that as the concentration x of Zn^{++} ions approaches x_c from below ($x < x_c$) in $Mn_{1-x}Zn_xF_2$ compounds, there is a temperature region $T_N < T < T_f$ in which the magnetic moment $M(H)$ is observed to display a nonlinear behavior which is independent of the orientation of **H** in the crystal, indicating that a magnetic state of the crystal different from either the ordinary paramagnetic or the antiferromagnetic state exists in the crystal in this temperature range.

The present study was undertaken to investigate the magnetic moment and magnetic susceptibility of the system $Mn_{0.2}Zn_{0.8}F_2$ ($x > x_c$). Studies of $Mn_{1-x}Zn_xF_2$ systems with Zn^{++} concentrations in this range are of definite interest, inasmuch as the substitution of the nonmagnetic Zn^{++} ions for the magnetic ions Mn^{++} gives rise to a random distribution of the interacting magnetic ions, with a random distribution of " $-$ " or " $+$ " exchange interactions, the " $-$ " sign resulting from the exchange interaction of Mn^{++} ions belonging to different sublattices in the original MnF^2 and a " $\pm$ " sign from the exchange interaction of Mn^{++} ions belonging to the same sublattice. Such a distribution of the exchange interaction together with the dipole interaction is typical of the spin glasses that are so much under study these days.[5,6]

The experiments to measure the dependence of the magnetic moment on the applied magnetic field were done on a vibrating-sample magnetometer.[7] The experiments on the frequency dependence of the magnetic susceptibility were carried out on an apparatus designed for this purpose with the usual[8] two compensated measuring coils (the sample was inserted into one of them in the course of the experiment) and a coil for modulating the magnetic field. As is shown in Ref. 8, the signal in the measuring coils is proportional both to the frequency of the modulating field and to the magnetic susceptibility of the system under study. The amplitude of the alternating field was 15 Oe.

The sensitivity of this apparatus in reference to the magnetic susceptibility is $\chi \sim 10^{-4}$ cgs emu. The $Mn_{0.2}Zn_{0.8}F_2$ single crystals[1)] were oriented in an x-ray appara-

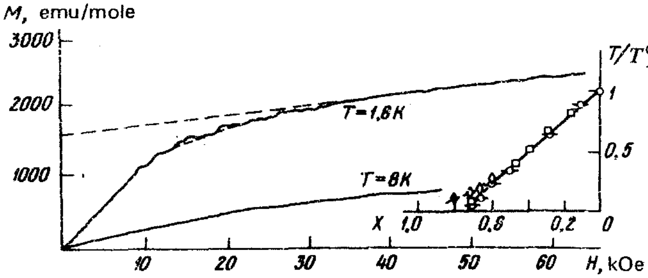

FIG. 1. Magnetic moment as a function of the applied magnetic field at different temperatures. The inset shows the influence of the concentration of Zn^{++} ions on the reduced temperature T_N/T_N^0 at which the antiferromagnetic properties of $Mn_{0.2}Zn_{0.8}F_2$ appear (the points O are the refined data of Ref. 1, the points □ are the data of Ref. 4) and the reduced temperature T_x/T_N^0 at which the peculiar magnetic properties of $Mn_{0.2}Zn_{0.8}F_2$ appear (the points ◇ are the data of Ref. 3, the point ◆ is the datum of the present work).

tus. The axes of the single crystal were determined to within 2–3°. The concentration of Mn^{++} ions in the $Mn_{0.2}Zn_{0.8}F_2$ was determined from the room-temperature magnetic susceptibility[2] to within 10%.

Figure 1 shows curves of the magnetic moment as a function of the applied magnetic field at various temperatures. At temperatures $T > T_f \sim 6 - 6.5$ K, the magnetic moment $M(H)$ is a linear function described by $M(H) = \chi(T)H$. At temperatures $T < T_f$ one observes a nonlinear behavior of the magnetic moment as a function of the applied magnetic field which is independent of the orientation of **H** and can be described in strong magnetic fields by the expression $M(H) = M_0(T) + \chi(T)H$. Figure 1 shows the functions $M(H)$ for $T = 6$ K and $T = 1.8$ K. Figure 2 shows the temperature

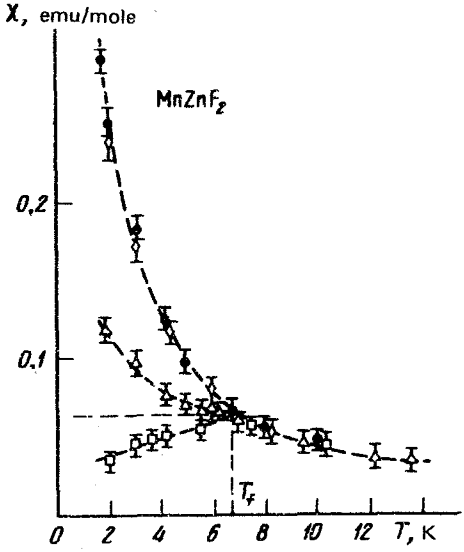

FIG. 2. Temperature dependence of the magnetic susceptibility $\chi_\perp$ measured at various values of the applied magnetic field and various frequencies of the modulating field: (●) $H \to 0$, $\omega \simeq 0$, (◇) $H \to 0$, $\omega < 200$ Hz, (△) $H \to 0$, $\omega > 700$ Hz, (□) $H > 40$ kOe, $\omega \simeq 0$.

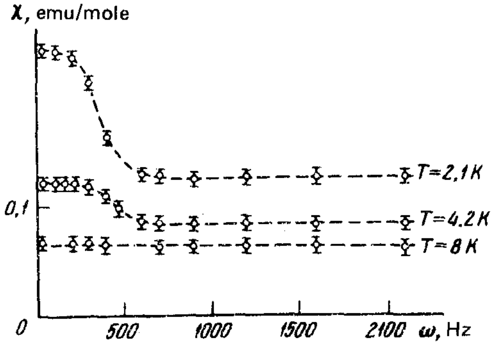

FIG. 3. Magnetic susceptibility measured in weak magnetic fields as a function of the frequency of the modulating field at various temperatures.

dependence of the magnetic susceptibility of the single-crystal samples as measured in weak magnetic fields (χ *—the points ⧳) and strong magnetic fields (χ—the points ☐). A characteristic feature of the curves is that the weak-field magnetic susceptibility χ * increases according to a $\sim 1/T$ law as the temperature is lowered, while the strong-field susceptibility χ is nonzero at temperatures $T < T_f$ (although for an ordinary paramagnet saturation should have been reached at these temperatures) and depends only slightly on the temperature. Subsequent experiments were undertaken to study the dependence of the weak-field magnetic susceptibility of the $Mn_{0.2}Zn_{0.8}F_2$ system on the frequency of the modulating field. Figure 3 shows the frequency dependence of the magnetic susceptibility χ * as measured at various temperatures. It is seen in Fig. 3 that for $T < T_f$ there is a rapid change in the susceptibility in the frequency range $\omega \sim 400$ Hz, from a large value χ' ($\omega < 200$ Hz) to a value χ'' ($\omega > 900$ Hz) which, at $T = 4.2$ K, is smaller than χ' by a factor of 1.5. The temperature dependence of $\chi'(T)$ coincides with that of the static susceptibility measured in weak fields. The susceptibility χ'' has a much weaker temperature dependence. As the temperature is lowered, the low-frequency region in which χ' is independent of the frequency becomes slightly smaller. To check the linearity of the apparatus we studied the frequency dependence of the magnetic susceptibility $\chi_\perp$ of a single-crystal sample of MnF_2 at $T = 4.2$ K. The studies on the MnF_2 crystal, just as the studies on the $Mn_{0.2}Zn_{0.8}F_2$ single crystal for $T > T_f$, showed that the signal obtained from the measuring coils was proportional to the frequency to within 5%, and, hence, the magnetic susceptibility is frequency independent to this accuracy. The diamond-shaped points ◊ in Fig. 2 give the temperature dependence of the magnetic susceptibility measured at low frequencies $\omega < 200$ Hz. It is seen that this temperature dependence coincides with that of $\chi^*(T)$ measured earlier in the magnetometer, where the frequency of the corresponding measurements was $\omega \simeq 0$. The triangular points △ in Fig. 2 give the temperature dependence of the magnetic susceptibility χ'' measured at frequencies $\omega > 900$ Hz. It is seen in the figure that although χ'' does exhibit a certain growth as the temperature is lowered, there is a kink in the curve at $T_f = 6.5 \pm 0.5$ K. Above T_f, as can be seen in Fig. 3, the magnetic susceptibility χ * is not observed to depend on the frequency. It is possible that the growth in the high-frequency magnetic susceptibility with decreasing temperature is due to the small (2–3%) content of Mn^{++} ions which are surrounded only by Zn^{++} ions. The vast majority of Mn^{++} ions in the $Mn_{1-x}Zn_xF_2$ lattice at temperatures below T_f are found in a state which differs from an ordinary paramagnetic state and is characterized by the observed frequency dependence of the magnetic susceptibility χ^*, with a characteristic relaxation "frequency" $\omega \simeq 400$ Hz. It can be supposed that this state is in fact a highly degenerate transient spin-glass state.[6] At temperatures $T < T_f$ the $Mn_{0.2}Zn_{0.8}F_2$ single crystal undergoes a transition to a set of energetically equivalent states which differ in respect to the local orientation of the magnetic moments of the system. In the experiments at frequencies $\omega < 200$ Hz the degeneracy causes a paramagnetic growth in the magnetic susceptibility, while at frequencies $\omega > 900$ Hz one measures the magnetic susceptibility of the system in one of the states. This conjecture is also supported by the measurements of the magnetic moment and magnetic susceptibility as functions of the applied magnetic field (Fig. 1).[1,2]

As the magnetic field is increased, the system goes into a steady, stable state with a distribution of the magnetic moments of the ions around the applied magnetic field. Here, according to the experiment (Fig. 1), the field dependence is $M(H) = M_0 + \chi H$. In all probability, the magnetic properties of the dilute antiferromagnets $Mn_{1-x}Zn_xF_2$ in the region $x < 0.7$ (Refs. 1 and 2) are also characterized by a frequency dependent χ^*. The inset in Fig. 1 shows the concentration dependence of the phase-transition temperatures obtained in Ref. 2 for the $Mn_{1-x}Zn_xF_2$ system at $x < x_c$ in a weak magnetic field (curve 1) and in a strong magnetic field (curve 2). The point ⧳ in this inset indicates the temperature T_f of our $Mn_{0.2}Zn_{0.8}F_2$ crystal as obtained from the emergence of the feature on the frequency curve of χ^*. It can be assumed that for concentrations $x < x_c$ and temperatures $T_N(x) < T < T_f$ an unusual new paramagnetic state occurs in these crystals, and T_f depends rather weakly, if at all, on the magnitude of the applied magnetic field. In future studies I hope to resolve these questions by investigating the frequency dependence of the magnetic susceptibility of $Mn_{0.2}Zn_{0.8}F_2$ at various temperatures and fields.

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