Effect of magnetic field on the transition temperature of the antiferromagnet $Mn_{0.5}Zn_{0.5}F_2$

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Neutron diffraction is used to investigate the temperature dependence of the intensity of magnetic scattering from (100) planes in the crystal $Mn_{0.5}Zn_{0.5}F_2$ in the absence of a field and in the presence of a magnetic field parallel to the axis of easy magnetization. In a field $H = 20$ kOe there is a decrease in the transition temperature by an amount 1.3 ± 0.3 K, which is evidently a manifestation of the effect of a random magnetic field.

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There has been growing interest of late in both the theoretical and experimental study of magnetic systems with random interactions. Imry and Ma[1] have shown that the random field in such systems substantially alters the state of the magnet at $T < T_N$. One of the ways of creating such systems is that proposed by Fishman and Aharony.[2] Those authors showed that if an anisotropic Ising antiferromagnet diluted by nonmagnetic impurities is placed in a static and uniform magnetic field directed along the easy axis, then the field will act like a random magnetic field in a nondiluted ferromagnet. Because the number of magnetic ions in each sublattic varies along the sample, the total magnetic moment should also fluctuate. When an external magnetic field is applied, a competition arises between the antiferromagnetic ordering of the spins and the separation of the system into regions with a total moment turned along H. Thus the magnetic field leads, in particular, to a disruption of the long-range order for $T < T_N$.

We have measured the intensity of the diffraction peaks corresponding to elastic scattering of neutrons from the (100) and (200) planes of a $Mn_{0.5}Zn_{0.5}F_2$ single crystal as a function of the temperature in the absence of magnetic field and in a field of 20 kOe parallel to the C_4 axis. The indicated Mn and Zn concentrations correspond to the relative molar content of the initial components from which the crystals were grown. The crystal structure of MnF_2 is such that the (100) reflection is purely magnetic. The measurements were made by the time-of-flight method at a Bragg reflection angle of 9° on a diffractometer based on the microtron at the Institute of Semiconductor Physics of the Academy of Sciences of the USSR.[3] A sample with dimensions of $12 \times 7 \times 3$ mm was placed in a vacuum chamber inside a helium cryostat; the sample was heated by current coils wound onto the aluminum crystal holder. The field was produced by superconducting Helmholtz coils and was directed along the tetragonal axis of the crystal. Each point was measured over a time ~ 30 min; the temperature was held stable to within 0.1 K.

Figure 1 shows the temperature dependence of the relative intensity of the magnetic reflection from the (100) plane. A neutron flux monitor was used to normalize the curves. The incoherent-scattering contribution was estimated from the results of

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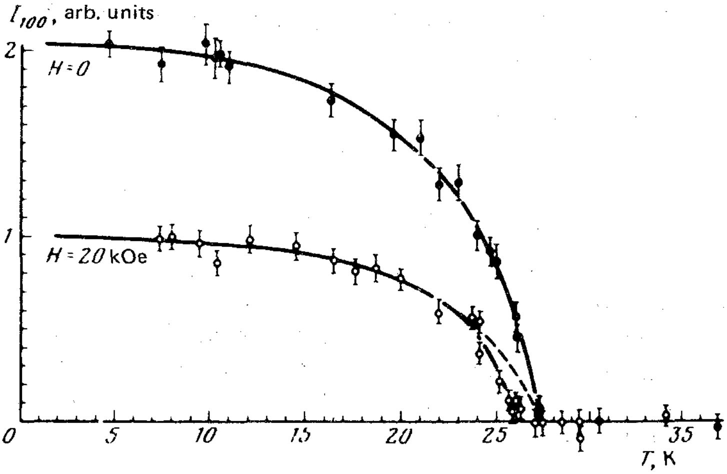

FIG. 1. Temperature dependence of the intensity of the magnetic reflection from the (100) plane at $H=0$ and $H=20$ kOe. The dashed part of the curve for $H=20$ kOe shows the function $I_{100}(T)$ for $H=0$.

measurements made above T_N. The error was determined almost entirely by the counting statistics. The solid curves were drawn by hand as an average of the experimental points. These curves do not correspond to the temperature dependence of the sublattice magnetizations, i.e., $I_{(100)}$ is not proportional to $M^2(T)$. We observed analogous discrepancies in undiluted MnF_2 as well and attribute them to the large value of the extinction. The roughly two-fold decrease in the absolute value of the intensity of the reflection in a magnetic field is most likely due to the uncontrollable rotation of the crystal in the field ($\sim 0.5°$), since the intensity of the nuclear reflection from the (200) plane observed at the same Bragg angle decrease by the same factor. Within the error limits the curves reproduce each other for $T<20$ K. Above this temperature the intensity of the reflection from the crystal in the magnetic field falls off more rapidly and goes to zero at a temperature which is lower by 1.3 ± 0.3 K. In undiluted MnF_2 in the same field[5] the temperature shift is $\Delta T = 0.006$ K, and if we assume that this shift is proportional to the concentration of magnetic ions, i.e., $\Delta T = Bh^2 \sim x(H/T)^2$, then we should have $\Delta T \sim 0.19$ K in our crystal, which is significantly smaller than the experimentally observed value. The value of T_N in our crystal was 27.4 K. If we assume that the dependence of T_N on the concentration of Zn ions is given by the expression $T_N(x) = 1.25 T_N(0)(x-0.2)$, in agreement with a number of experimental results,[4] we obtain for our crystal the value $x=0.53$, in good agreement with the initial concentration of the components. We can thus attribute to the action of the random magnetic field a shift ΔT of 1.1 K. Analogous experimental results were recently obtained by Belanger *et al.*,[5] who studied the magnetic-field dependence of the refractive index in $Mn_{1-x}Zn_xF_2$ crystals ($x=0.2$ and 0.35).

According to Fishma and Aharony,[2] the random magnetic field, which is characterized by a reduced field $h = \mu H/kT$, where $\mu = \mu_B gS$, leads to the destruction of the long-range order at a temperature

$$T_c = T_N - B h^2 - T_N (A h^2 / U_0)^{1/\phi},$$

where in accordance with Ref. 5 one has $A \cong x(1-x)(zJ/kT)^2$, and Bh^2 is the shift of the transition temperature in the magnetic field (the shift is also present in undiluted MnF_2). For comparison of our results with the predictions of the theoretical calculations, we made use of the experimental results of Ref. 5, where in a field $H = 20$ kOe the temperature shift was $\Delta T = 0.35$ at $x = 0.2$ and 0.81 K at $x = 0.35$, and a value of the critical exponent $\phi = 1.4$ was obtained. Proceeding from these data, we find that at $x = 0.5$ the shift of the transition temperature should be $T_c - T_N = 1.4$–1.53 K, in agreement with the value obtained by us in the present experiment.

It should be noted that Cowley and co-workers[6,7] have also made neutron studies of the dilute solutions $Mn_{1-x}Zn_xF_2$. In these studies the application of a field along the easy axis was observed to cause a broadening of the diffraction peaks, which is evidence of a decrease in the correlation length. In our experiments the resolution was approximately two orders of magnitude lower than in Ref. 6, and we were therefore unable to draw any conclusions in regard to this matter.

In summary, the presently available data tend to support the theoretical predictions regarding the nature of the effect of magnetic field on the behavior of dilute antiferromagnets. It should be stressed, however, that the theory predicts that the random magnetic field destroys the long-range order at all temperatures below T_N. The experimental situation remains uncertain. For example, some results[6,7] indicate that when $Co_{1-x}Zn_xF_2$ is cooled in the presence of a magnetic field, an unusual state lacking long-range order is formed, but this state may turn out to be metastable. Studies of the state of highly dilute systems, in which the impurity can no longer be considered small, are extremely interesting. In particular, anomalous behavior of $\chi_\perp$ in a strong magnetic field has been observed[8] near the critical impurity concentration.

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