Two Successive Transitions in Uniaxially Anisotropic Spin-Glasses

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Longitudinal ($\parallel c$) and transverse ($\perp c$) ac susceptibilities have been measured on single crystals of the Ising-type spin-glass ZnMn and the *XY*-type spin-glass CdMn. A clear cusp is observed in each direction at different temperatures for ZnMn with 270, 390, and 600 ppm Mn. These behaviors indicate that two successive transitions occur in these systems as was suggested by mean-field theories for anisotropic spin-glasses. The phase diagram obtained is in accordance with the theoretical one.

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Magnetic properties of spin-glasses with single-ion uniaxial anisotropy have been extensively studied since Albrecht *et al.*[1] showed that in dilute hexagonal single crystals, depending on the crystal-field parameter D, there exists Ising-type behavior in ZnMn ($D > 0$), *XY*-type behavior in CdMn ($D < 0$), while MgMn orders in a Heisenberg-type manner ($D \approx 0$). In these studies and successive investigations on rare-earth (RE) systems[2] freezing occurred only in the easy direction, longitudinal (L) for $D > 0$ and transverse (T) for $D < 0$. On the other hand, theoretical investigations[3-5] for m-component vector spin-glasses predicted that in the presence of random exchange $\sum J_{ij} S_i S_j$ with a Gaussian probability distribution $P(J_{ij})$ and local uniaxial anisotropy $-D\sum(S_{iz})^2$ there should be two successive transitions, from the paramagnetic (P) state to the L state and then at lower temperatures to a mixed phase (LT) when D/J is smaller than a critical value $(D/J)^+$, and a sequence of P-T-LT transitions for $0 > D/J > (D/J)^-$ in the *XY* case. Recently, transverse ordering of the Ising-type spin-glass Fe_2TiO_5 was discussed by Yeshurun and Sompolinsky.[6] They suggested that the transverse ordering is included at the longitudinal-ordering temperature, because of weak coupling between L and T components due to random anisotropy. Yet, L-T coupling can always be expected to exist in such a concentrated system and the occurrence of longitudinal and transverse freezing at the same temperature even for moderate D/J values is thus not a disproof of the theoretical predictions.[3-5] A search for the double spin-glass transition was also done by Adachi *et al.*[7] on $Mn_{1-x}Ni_xSb$ alloys. These authors, however, studied polycrystalline samples and thus could not find differences between the longitudinal and transverse susceptibilities $\chi_\parallel$ and $\chi_\perp$.

In the present paper we restudied the uniaxial systems ZnMn and CdMn, but with much higher impurity concentrations than in the previous investigation[1] in order to obtain values of D/J small enough to stay below the limit $|D/J| < |(D/J)^{\pm}|$. The ac susceptibility as measured parallel ($\parallel$) and perpendicular ($\perp$) to the c axis on single-crystalline samples ZnMn with 270, 390, and 600 ppm Mn shows clear cusps at different temperatures $T_{g\parallel}$ and $T_{g\perp}$ ($T_{g\parallel} > T_{g\perp}$), respectively, and no anomaly in $\chi_\perp$ at $T_{g\parallel}$ or $\chi_\parallel$ at $T_{g\perp}$. A CD + 488-ppm-Mm sample is very likely just on the edge of the critical $(D/J)^-$ value. The results thus give clear evidence for the theoretical predictions within the mean-field model.[3-5] Furthermore, we are able to show that the experiments are even quantitatively in accordance with the theoretically predicted phase diagram.

Single crystals are grown by the Bridgman technique. The crystallographic axes are determined from x-ray analysis. Thin sample rods ($10\times1\times1$ mm^3) are spark cut parallel and perpendicular to the c axis. The ac susceptibility is measured by means of a Hartshorn bridge circuit with a typical ac field of 10 Oe at a frequency of 80 Hz in a dilution refrigerator from 50 mK to 4.2 K. Static fields of up to 1600 Oe can be applied parallel or perpendicular to the c axis in addition to the measuring ac field.

Figure 1 shows the temperature dependence of the longitudinal ($\chi_\parallel$) and transverse ($\chi_\perp$) susceptibilities for ZnMn with 390 ppm and 600 ppm Mn. As temperature is lowered from the paramagnetic state, $\chi_\parallel$ shows a cusp at $T_{g\parallel}$, while $\chi_\perp$ still increases smoothly through $T_{g\parallel}$. On further decrease of the temperature, $\chi_\perp$ also shows a sharp maximum at $T_{g\perp}$. We find $T_{g\parallel} = 0.64$ K and $T_{g\perp} = 0.17$ K for ZnMn(390 ppm), and $T_{g\parallel} = 0.77$ K and $T_{g\perp} = 0.29$ K for ZnMn(600

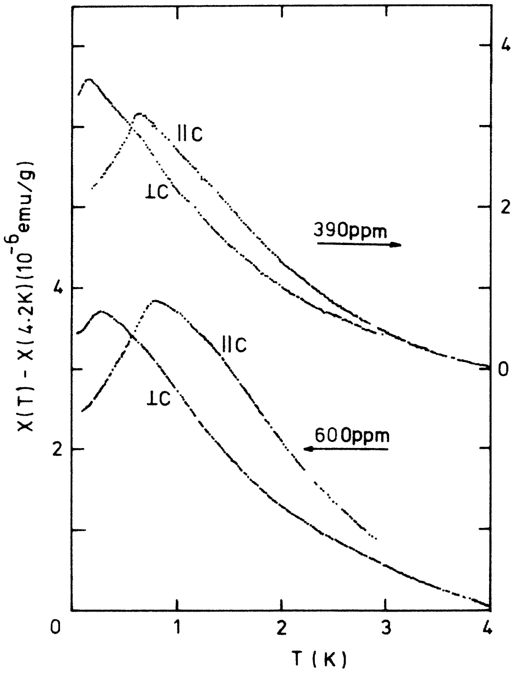

FIG. 1. Temperature dependence of the ac susceptibility $\chi(T)$, with $\chi(4.2\ \mathrm{K})$ subtracted, in each direction parallel ($\parallel c$) and perpendicular ($\perp c$) to the c axis for two ZnMn single crystals.

ppm). The occurrence of two independent cusps obviously suggests that the longitudinal and transverse components of Mn spins freeze independently.[8] Furthermore, we have measured the static-field dependence of ac $\chi(T)$ for all samples. Results for the 390-ppm-Mn and 600-ppm-Mn samples are given in Figs. 2 and 3, respectively. It can be seen that in both alloys in both directions the maxima become smaller and broader with increasing field. Similar results have been found for the ZnMn(270 ppm) sample. From the data we can determine the nonlinear susceptibility $\chi_{\mathrm{nl}}=\chi(0)-\chi(H)$ as a function of T. As shown recently by Yeshurun, Ketelson, and Salamon[9] the critical behavior of χ_{nl} provides a crucial test for the occurrence of a spin-glass transition. Results of $\chi_{\mathrm{nl}}(T)$ for $\chi\perp c$ ($H \perp c$) of the 390- and 600-ppm alloys are respectively shown in the insets of Figs. 2 and 3. The presence of a maximum at $T_{g\perp}$ gives clear evidence for a spin-glass transition at this temperature and thus provides further support for the presence of two successive transitions in ZnMn. We have also measured the $H_\perp$ dependence of $\chi_\parallel$ and $H_\parallel$ dependence of $\chi_\perp$ and observed a negative nonlinear contribution to the susceptibility around $T_{g\parallel}$ and $T_{g\perp}$, as well.

The temperature dependence of $\chi_\parallel$ and $\chi_\perp$ for CdMn (488 ppm) is shown in Fig. 4. Since this alloy orders in an XY-type manner, $\chi_\perp$ is larger than $\chi_\parallel$ and shows a maximum at $T_{g\perp}=0.88$ K. $\chi_\parallel$ shows a weak and broad maximum at ~ 1 K, roughly equal to $T_{g\perp}$. However, $\chi_\parallel$ increases again for $T\rightarrow 0$. Possibly $\chi_\parallel$ includes some contribution of $\chi_\perp$ due to the presence of some misorientated grains or precipitations of Mn. Therefore, we suppose that the P-T sample in this occurs at $T_{g\perp}$ but that a second transition T-LT transition for this concentration is not to be expected. Higher concentrations of Mn in Cd are not possible because of metallurgical problems (segregation).

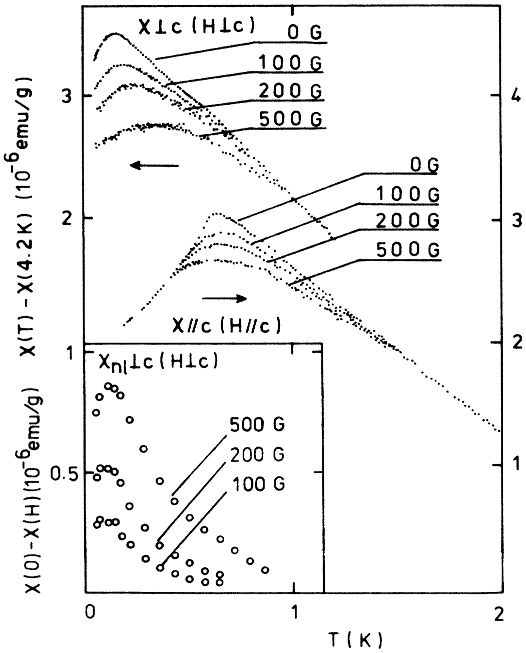

FIG. 2. Temperature dependence of the longitudinal and transverse susceptibilities of ZnMn(390 ppm) at various fields parallel to the ac field. Inset: the nonlinear susceptibility $\chi_{\mathrm{nl}}(H)=\chi(0)-\chi(H)$ for the hard direction ($\perp c$).

To obtain the experimental D/J-T/J phase diagram and compare it with the theoretical results we use the following procedure. According to Hedgcock *et al.*[10] the crystalline-field parameters $D\sim 0.078$ K in ZnMn and $D\sim -0.16$ K in CdMn do not depend on the Mn concentration as long as the alloys are dilute. Therefore, we assume that the D values in all the present samples are the same as the ones described above. It is not easy to estimate the variance J in the presence of random exchange interactions. We use the approximation $J\sim kT_{g\parallel}-2D$ for $|D/J| \ll 1$ as given for Ising-type spin-glasses by the mean-field theories in order to determine the J value from $T_{g\parallel}$ and D in ZnMn with 270, 390, and 600 ppm Mn. For ZnMn with Mn concentrations less than 270 ppm, J might be of the same order of magnitude as D, so that it is impossible to use the above approximation. Yet it is well known[11-13] that the internal fields of dilute alloys due to $1/r^3$-type Ruderman-Kittel-Kasuya-Yosida interactions have a Lorentzian distribution with a distribution

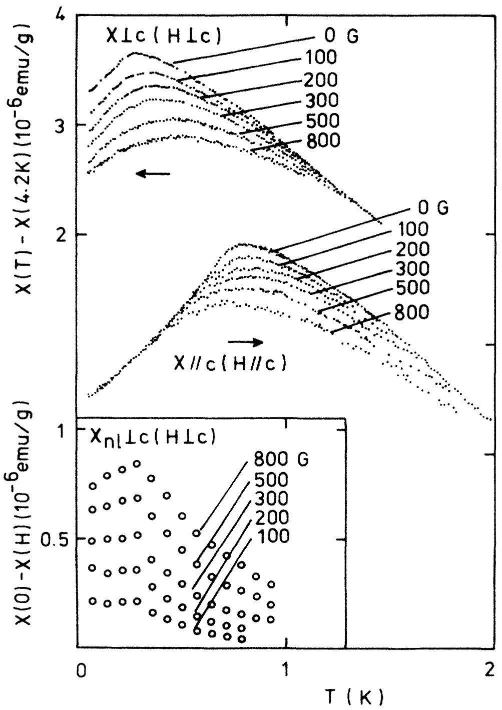

FIG. 3. Temperature dependence of the longitudinal and transverse susceptibilities of ZnMn(600 ppm) at various fields parallel to the ac field. Inset: the nonlinear susceptibility for the hard direction ($\perp c$).

width proportional to the impurity concentration. Assuming $J(c) \propto c$ (c is the impurity concentration) we obtain the J values for ZnMn with 150 and 62 ppm Mn on the basis of the ZnMn(390 ppm) value: $J(c) = J(390\ \mathrm{ppm}) \times c/(390\ \mathrm{ppm})$ (data for 62 ppm are given by Albrecht *et al.*[1]). For CdMn(488 ppm), we determine J from the equation $J = kT_{g\perp} + D$ for $|D/J| \ll 1$ in XY-type spin-glasses, assuming that the Mn concentration is high enough to fulfill the condition $|D/J| \ll 1$. All experimental data are collected in the D/J-T/J phase diagram in Fig. 5. The figure shows that our experimental results are fully consistent with the theoretical predictions. This means that the mean-field theories describe well the uniaxially anisotropic spin-glasses ZnMn and CdMn.

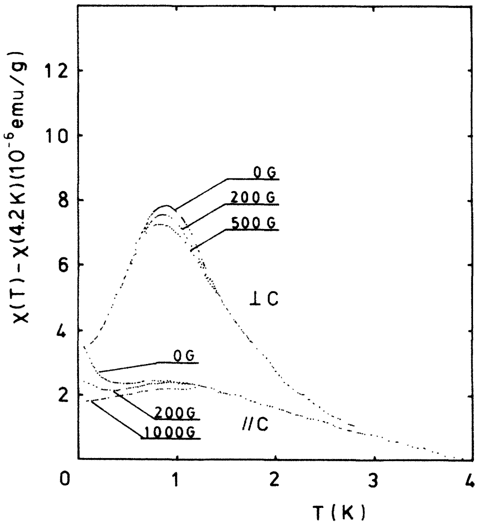

FIG. 4. Temperature dependence of the longitudinal and transverse susceptibilities of CdMn(488 ppm) at various coaxial fields.

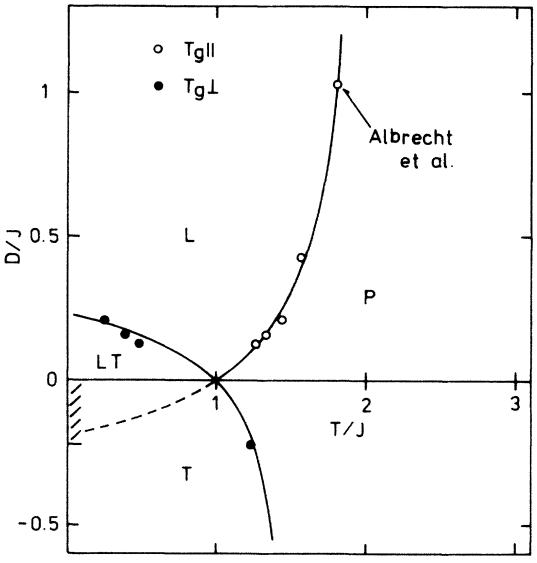

FIG. 5. The experimental D/J-T/J phase diagram for ZnMn and CdMn obtained from the cusps in $\chi_{\parallel}$ and $\chi_{\perp}$ including the results of Albrecht *et al.* The solid lines serve only to guide the eyes.

From the field dependence of the susceptibility in Figs. 2 and 3 it is found that the temperature of the maximum T_m in $\chi_{\parallel}$ shifts to lower temperatures when applying $H_{\parallel}$, whereas the temperature of the maximum in $\chi_{\perp}$ shifts to higher temperatures with $H_{\perp}$. In Fig. 6 we plot T_m versus the applied field for the 270-ppm-Mn sample (filled triangles for $\chi_{\parallel}$ and open circles for $\chi_{\perp}$) including the $H_{\perp}$ dependence of $\chi_{\parallel}$ and the $H_{\parallel}$ dependence of $\chi_{\perp}$ (filled circles for $\chi_{\parallel}$ and open triangles for $\chi_{\perp}$). In the case of $H_{\parallel}$, the maximum temperatures of both $\chi_{\parallel}$ and $\chi_{\perp}$ decrease when applying a field. This indicates that the temperatures for the P-L and L-LT transitions decrease with rising coaxial field $H_{\parallel}$ in ZnMn, which is in accordance with theoretical predictions by Elderfield and Sherrington.[14] In the case of $H_{\perp}$, $\chi_{\perp}$ is much more affected by the field than $\chi_{\parallel}$. The temperature of the maximum in $\chi_{\perp}$ increases with $H_{\perp}$ as shown in Figs. 2, 3, and 6. This remarkable change of $\chi_{\perp}$ can be qualitatively explained as follows. The transverse components of Mn spins induced by $H_{\perp}$ couple with the frozen longitudinal

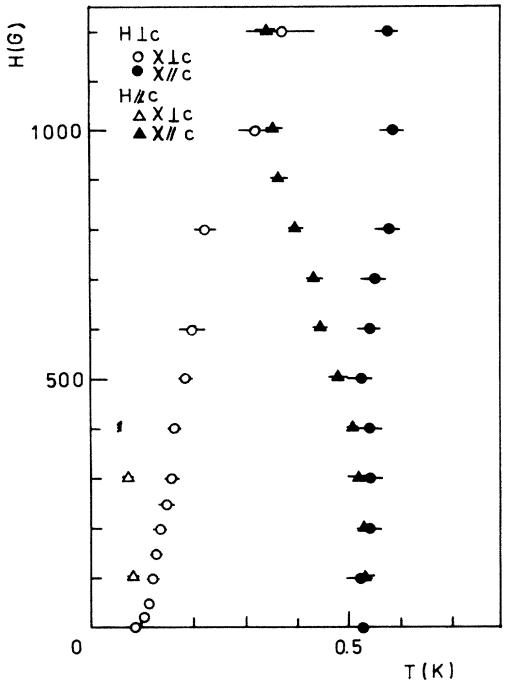

FIG. 6. Longitudinal and transverse field dependences of the temperature of the maximum in $\chi_{\parallel}$ and $\chi_{\perp}$ for ZnMn(270 ppm).

components below $T_{g\parallel}$ as was discussed by Yeshurun and Sompolinsky.[6] Therefore, the induced transverse components order below $T_{g\parallel}$ when $H_{\perp}$ is applied. The maximum temperatures in $\chi_{\perp}$ then increase with $H_{\perp}$ since the induced transverse components—coupled with the longitudinal one—also increase with rising $H_{\perp}$.

In the previous studies[2,6] of anisotropic spin-glasses two successive transitions have not been clearly found because of the existence of random anisotropy.[15] The axis of the local crystalline field in these concentrated spin-glasses[2,6] might be slightly inclined with respect to the crystallographic axis. On the other hand, our anisotropic spin-glasses ZnMn and CdMn are very dilute, so that the local crystalline-field axis is expected to be parallel to the *c* axis (basal plane in CdMn) from a microscopic point of view. Though it is possible that Dzyaloshinsky-Moriya type of random anisotropy[16] exists in these systems, this contribution is considered to be very small since the *S*-state-like Mn impurities give only a very small spin-orbit contribution.[17] The present systems are thus the best examples for really uniaxial spin-glasses.

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[1] H. Albrecht, E. F. Wassermann, F. T. Hedgcock, and P. Monod, Phys. Rev. Lett. **48**, 819 (1982).

[2] K. Baberschke, P. Pureur, A. Fert, R. Wendler, and S. Senoussi, Phys. Rev. B **29**, 4999 (1984).

[3] D. M. Cragg and D. Sherrington, Phys. Rev. Lett. **49**, 1190 (1982).

[4] S. A. Roberts and A. J. Bray, J. Phys. C **15**, L527 (1982).

[5] A. J. Bray and L. Viana, J. Phys. C **16**, 4679 (1983).

[6] Y. Yeshurun and H. Sompolinsky, Phys. Rev. B **31**, 3191 (1985).

[7] K. Adachi, R. Imura, M. Matsui, and H. Sawamoto, J. Phys. Soc. Jpn. **44**, 114 (1978).

[8] S. Murayama, K. Yokosawa, Y. Miyako, and E. F. Wassermann, J. Magn. Magn. Mater. **54–57**, 221 (1986).

[9] Y. Yeshurun, L. J. P. Ketelsen, and M. B. Salamon, Phys. Rev. B **32**, 7425 (1985).

[10] F. T. Hedgcock, J. Appl. Phys. **49**, 1446 (1978); P. L. Li, F. T. Hedgcock, W. B. Muir, and J. O. Ströhm-Olsen, Phys. Rev. Lett. **31**, 29 (1973).

[11] J. Souletie and R. Tournier, J. Low Temp. Phys. **1**, 95 (1969).

[12] R. W. Walstedt and L. R. Walker, Phys. Rev. B **9**, 4857 (1974).

[13] M. Klein, Phys. Rev. B **14**, 5008 (1976).

[14] D. Elderfield and D. Sherrington, J. Phys. C **16**, 4865 (1983).

[15] D. Sherrington, J. Phys. C **17**, L823 (1984).

[16] P. M. Levy and A. Fert, Phys. Rev. B **23**, 4667 (1981).

[17] Y. Miyako, in *Proceedings of the Twelfth International Conference on Low Temperature Physics, Kyoto, 1970,* edited by E. Kanda (Keigaku Publishing Co., Tokyo, 1971), p. 790.