Heisenberg, *XY*, and Ising Spin-Glass Behavior in Hexagonal Metallic Systems

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dc magnetization measurements at millikelvin temperatures on dilute ZnMn, CdMn, and MgMn single crystals show that in accordance with the sign of the single-site anisotropy D, ZnMn is "Ising-like," i.e., the Mn moments lie parallel to the c axis. CdMn is "X-Y-like" with the easy axis in the basal plane. MgMn is isotropic, "Heisenberg-like." Dominance of spin-orbit scattering leads to a qualitative understanding of the anisotropy.

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It is an open question whether the existence of a magnetic anisotropy is necessary for the appearance of a susceptibility cusp in a spin-glass. Besides very recent calculations[1] most theoretical models do not take any anisotropy into account. Experimentally, early measurements[2,3] on cubic single crystals of CuMn failed to show the existence of an anisotropy associated with a certain lattice direction. Field-induced unidirectional properties were independent of crystallographic directions. In recent work[4,5] on polycrystalline CuMn a single macroscopic anisotropy field associated with the remanence was found. Spin-orbit scattering was the physical reason for this anisotropy, and Fert and Levy[6] tried to explain this theoretically. It remains unclear, however, how to link a microscopic anisotropy—of what origin so ever (crystal-field splitting, dipole-dipole interaction, or spin-orbit scattering)—to an experimentally observed macroscopic anisotropy behavior, e.g., to a displaced hysteresis loop[3,4] or an anisotropy of the susceptibility like in this work. This means that significance and understanding of the anisotropy in spin-glasses has still to be worked out.

In the present paper we will show that susceptibility maxima are only found in certain lattice directions of macroscopic single-crystalline metallic *hexagonal* spin-glass samples. The behavior is such that—in accordance with the sign of the isolated ion anisotropy D—ZnMn is "Ising-like," i.e., the Mn moments lie parallel to the c axis; CdMn is "X-Y-like," i.e., the Mn moments lie preferentially in the basal plane; whereas MgMn is "Heisenberg-like," isotropic. The three experimental examples therefore represent what can be considered as typical model systems for comparison with theory.

There have been earlier investigations on quasimetallic $(Ti_{1-x}V_x)_2O_3$ (Ref. 7) and insulating Fe_2TiO_3 (Ref. 8) single crystals revealing spin-glass maxima in $\chi(T)$ only in certain lattice directions. In contrast to our dilute-limit case these systems are relatively concentrated, and therefore direct dipole-dipole interactions might cause the anisotropy.

Single crystals are prepared by the Bridgman technique, oriented with x rays, and spark cut into sample rods parallel ($\parallel$) and perpendicular ($\perp$) to the c axis. The Mn concentrations are determined by atomic absorption spectroscopy. The dc magnetization of the samples is measured in a dilution refrigerator by a SQUID technique, using a double-gradient coil system. A resistance-heatable Nb cylinder allows freezing of fixed magnetic fields of values between 0.004 and ~20 mT. For the very small fields the earth field is shielded by a Mumetal cylinder. The field values are determined from the coil current and/or by comparison with a calibrated sample. The sensitivity of the setup for a field of 0.1 mT is $\Delta M/H \lesssim 8 \times 10^{-9}$. Absolute values of the susceptibility are determined at higher temperatures (1.5–50 K) in a commercial SQUID susceptometer. Details are given elsewhere.[9] We define the temperature-dependent contribution of the Mn impurities to the measured susceptibility χ_{tot} as $\Delta\chi(T) = \chi_{tot} - \chi_0$, where χ_0 is the temperature-independent contri-

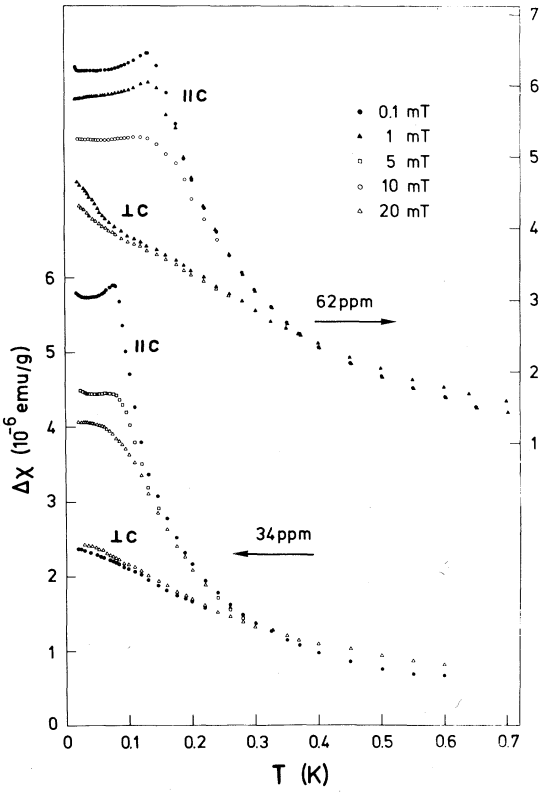

FIG. 1. Impurity contribution to the susceptibility $\Delta\chi$ vs temperature for two ZnMn single crystals as measured in different magnetic field on samples cut parallel and perpendicular to the c axis.

bution of the matrix, evaluated by extrapolating $\chi_{tot}(1/T)$ for $1/T \to 0$.

For temperatures $T \geq 2$ K all samples show a Curie-Weiss type of behavior in $\Delta\chi(T)$. Only small anisotropies are found for ZnMn and CdMn in the Curie constants, the paramagnetic Curie temperatures, the spin, and χ_0. MgMn behaves isotropically.[9] The picture looks completely different in the millikelvin range. Figure 1 shows $\Delta\chi(T)$ for two ZnMn alloys as measured in different cooling fields on samples cut parallel (∥) and perpendicular (⊥) to the c axis. The uniaxial anisotropy behavior is obvious from this plot. There are spin-glass typical maxima only for $\Delta\chi(T) \parallel c$, with $T_g = 78$ mK and $T_g = 137$ mK for the two concentrations, respectively. There is no indication for the occurrence of maxima for $\Delta\chi(T) \perp c$ in both alloys. Note also that already for $T \lesssim 3T_g$, $\Delta\chi \perp c$ is smaller than $\Delta\chi \parallel c$. There is a typical field dependence for the direction $\parallel c$. The maximum is "rounded" and reduced to about ~30% of its absolute value in a field of 10 mT,[10] with constant susceptibility for $T \leq T_g$. On the other hand, $\Delta\chi(T)_\perp$ depends on a field of 10 mT only to within ~5%. The ZnMn system is consequently "Ising-like," with the c axis as easy axis of magnetization and the Mn moments parallel to this direction. The negative sign of the value for the observed energy splitting D of the Mn ion ground state (see Table I below) is in accordance with this.

Figure 2 shows that in a single crystal of CdMn (125 ppm) the properties are just reversed as compared to ZnMn; so is the sign of D. There is now a maximum occurring in the direction ⊥ to the c axis ($T_g = 138$ mK), with an appreciable field dependence of the peak susceptibility. Well above and below T_g, $\Delta\chi(T) \parallel c$ is small as compared to $\Delta\chi(T) \perp c$. Note that $\Delta\chi(T) \parallel c$ shows no anomaly and does not depend on fields between 0.03 and 1 mT to more than ± 3%. The CdMn system therefore is also strongly anisotropic, but "X-Y-like," and the easy directions of magnetization lie in the basal plane.

Finally, Fig. 3 reveals that in MgMn we find maxima in the susceptibility for both lattice directions ∥ and ⊥ to the c axis at equal glass temperatures $T_g^{\parallel} = T_g^{\perp} = T_g = 48$ mK for 186 ppm Mn and $T_g = 90$ mK for 274 ppm. Figure 3 also shows that in MgMn the difference between $\Delta\chi(T)_\parallel$ and $\Delta\chi(T)_\perp$ is small and of the same order of magnitude in the spin-glass as well as in the paramagnetic regime. The MgMn system is therefore "Heisenberg-like" with no preferred lattice direction and no preferred easy axis.

TABLE I. Summary of data for the alloy systems. Change of T_g with Mn concentration dT_g/dc; average spin value $\vec{S}$ as derived from high-temperature $\Delta\chi(T)$ measurements; crystal-field splitting parameter D (see Ref. 12); anisotropy $\chi_A(0)$ for $T \to 0$ [see Eq. (1)], and atomic spin-orbit scattering strength of the matrix λ_{so} (see Ref. 12).

	Anisotropy	Easy axis	dT_g/dc (K/at.%)	$\vec{S}$	D (mK)	$\chi_A(0)$	λ_{so} (eV)
ZnMn	"Ising-like"	c axis	22	1.6	−78	−0.7	0.05
CdMn	"X-Y-like"	In basal plane	11	1.8	+160	+1.3	0.2
MgMn	"Heisenberg-like"	Isotropic	3	1.2	+7	−0.09	0.01

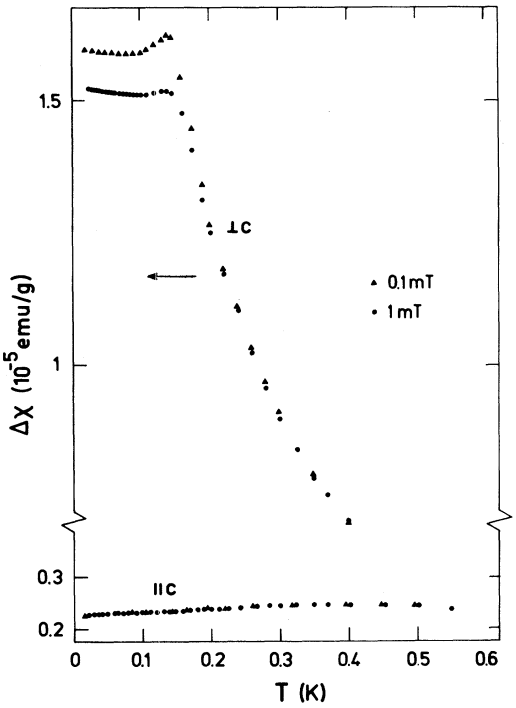

FIG. 2. Magnetic susceptibility $\Delta\chi(T)$ for a CdMn single crystal as measured in different fields parallel and perpendicular to the c axis.

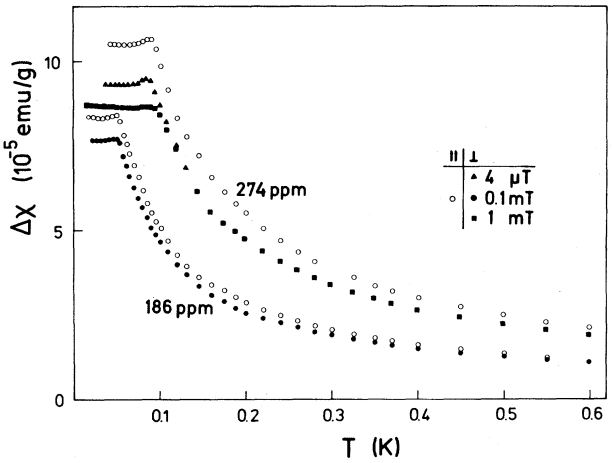

FIG. 3. Magnetic susceptibility $\Delta\chi(T)$ for two MgMn single crystals as measured parallel and perpendicular to the c axis.

We have summarized these features in Table I. Note that in all three systems the Kondo effect is not important, since the change of T_g with concentration, dT_g/dc of the hexagonal systems, is of the same order of magnitude as, e.g., in low-concentration CuMn ($dT_g/dc \approx 9$ K/at.%) or AgMn ($dT_g/dc \approx 4$ K/at.%). Qualitatively, the behavior of our alloys is the following: For ZnMn and CdMn the impurity spins are first constrained in their anisotropy direction (plane) and then the spin-glass order takes place at lower temperatures in this restricted geometry. For MgMn the spins point in the direction in which the cooling field is originally applied, and the anisotropy lies in the same direction.

It is difficult to understand the anisotropy in three systems from an ionic model, since in none of them does Mn show integral or half-integral spin values (S values see Table I), nor is it possible to define an orbital momentum. Thus a Hamiltonian for the level splitting similar to that in Ghatak and Sherrington[11] for the Mn impurity states in a hexagonal environment cannot be used. We do know, however, the sign and the values for the energy splitting parameter D in these alloys (Table I), as determined from torque measurements on single crystals where an anisotropy term varying as T^{-2} was present.[12] Comparing these D values with the measured glass temperatures T_g we can see that for the impurity concentrations studied here the D values for ZnMn and CdMn are of the *same* order of magnitude as the exchange interactions, if one assumes $k_B T_g \sim |\bar{J}_{ij}|$. Therefore, the systems would be moderately well represented by an Ising—or XY—Hamiltonian. In MgMn D is an order of magnitude smaller than the exchange interaction strength, thus defining a Heisenberg situation.

Although the Fert-Levy[6] mechanism as a possible origin of the anisotropy in the present systems is not obvious, dominance of spin-orbit scattering effects can lead to a qualitative understanding of the observed behavior. We define as a measure of the anisotropy

$$\chi_A(0) = [\chi(0)_\perp - \chi(0)_\parallel]\langle \tfrac{1}{3}\chi(0)_\parallel + \tfrac{2}{3}\chi(0)_\perp\rangle^{-1} \qquad (1)$$

extrapolated for $T \to 0$ from the experiments (for a field of 0.1 mT). As can be seen from Table I these values of $\chi_A(0)$ do correlate with the atomic spin-orbit coupling strength λ_{so} (Ref. 12) for the three matrix metals. From Zn to Cd the anisotropy increases by a factor of about 2, whereas λ_{so} increases by a factor of 4. From Cd to Mg, $\chi_A(0)$ decreases to about $\frac{1}{10}$, as does λ_{so}. This suggests that the anisotropy is roughly proportional to λ_{so}. Further work, however, is necessary to make this point more clear. We finally turn to the question of why the Mn moments point in different but fixed directions in Zn and Cd, while they are isotropically oriented in Mg. This

can be understood qualitatively by considering the Fermi surfaces of the host metals. As pointed out by Harris, Mulimani, and Zuckerman[13] the d-like orbital parts of the Mn impurity wave functions resonate strongly with the d-like parts of the matrix wave functions. There is evidence[14] that in Zn the "needles" of the Fermi surface have a large d component. These "needles" lie parallel to the c axis. The d-d resonance could thus give preferential orientation of the Mn moments parallel to the c axis. In Cd there are no "needles" on the Fermi surface and very likely the "monster" part carries the d character. The "monster" lies mainly perpendicular to the c axis. The Mn moments in CdMn therefore preferentially orient within the basal plane. The Fermi surface of Mg is almost spherical and s like so that the Mn moments find no preferred axis of orientation via the d-d resonance mechanism.

What are further problems and suggestions caused by the present study? First, the dependence of the anisotropy on the impurity concentration should be investigated more closely. Second, a third component with a different spin-orbit scattering cross section should be added to the hexagonal systems to prove whether spin-orbit scattering is fundamental in leading to anisotropy in spin-glasses. We think that the present paper opens the possibility to relate future work on any other anisotropic system to the three "classical" cases presented here. Possibly, our work allows a direct test of the Sherrington-Kirkpatrick[15] model for a mean-field spin-glass in the Ising case.

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[1]R. E. Walstedt and L. R. Walker, Phys. Rev. Lett. 47, 1624 (1981).

[2]J. J. Owen, M. E. Brown, V. Arp, and A. F. Kip, J. Phys. Chem. Solids 2, 85 (1957).

[3]T. Iwata, K. Kai, T. Nakamichi, and M. Yamamoto, J. Phys. Soc. Jpn. 28, 583 (1970).

[4]J. J. Prêjean, M. J. Joliclerc, and P. Monod, J. Phys. (Paris) 41, 427 (1980).

[5]F. Hippert, H. Alloul, and J. J. Prêjean, Physica (Utrecht) 107B, 645 (1981).

[6]A. Fert and P. M. Levy, Phys. Rev. Lett. 44, 1538 (1980).

[7]J. Dumas, C. Schlenker, J. L. Tholence, and R. Tournier, Phys. Rev. B 20, 3913 (1979).

[8]U. Atzmony, E. Gurewitz, M. Melamud, H. Pinto, H. Shaked, G. Gorodefsky, E. Hermon, R. M. Hornreich, S. Shtrikman, and B. Wanklyn, Phys. Rev. Lett. 43, 782 (1979).

[9]H. Albrecht, thesis, Rheinische-Westfälische Technische Hochschule, Aachen, 1981 (unpublished), and to be published.

[10]The small differences in $\Delta\chi$ for $T \lesssim 100$ mK between the two samples are possibly due to their different orientation in the basal plane.

[11]S. K. Ghatak and D. Sherrington, J. Phys. C 10, 3149 (1977).

[12]F. T. Hedgcock, J. Appl. Phys. 49, 1446 (1978).

[13]R. Harris, B. G. Mulimani, and M. J. Zuckermann, Phys. Condens. Matter 19, 269 (1975).

[14]W. B. Muir, G. Stroink, and E. F. Wassermann, Can. J. Phys. 55, 403 (1977).

[15]S. Kirkpatrick and D. Sherrington, Phys. Rev. B 17, 4384 (1978).