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Effects of internal exchange fields on magnetization steps in diluted magnetic semiconductors

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We present a new model of the high-field magnetization in dilute ($x \leq 0.05$) $A^{\mathrm{II}}_{1-x}\mathrm{Mn}_x B^{\mathrm{VI}}$ alloys (A^{II}=Zn, Cd, Hg; B^{VI}=S, Se, Te) which includes the effects of internal effective fields due to distant-neighbor exchange interactions. The model represents an extension of a nearest-neighbor-only cluster model used previously for these materials to explain the observation of "magnetization steps" associated with Mn nearest-neighbor pairs. The presence of internal fields causes a broadening and shift of these steps, in agreement with experiment. The distributions of such fields for fcc and hcp magnetic sublattices are modeled by considering exchange interactions out to third and fourth neighbors, respectively. Published data for $\mathrm{Cd}_{1-x}\mathrm{Mn}_x\mathrm{Te}$ and $\mathrm{Cd}_{1-x}\mathrm{Mn}_x\mathrm{Se}$ are reinterpreted using this approach to provide a more accurate determination of nearest-neighbor exchange constants as well as approximate values of more-distant interactions. All are found to be antiferromagnetic. The internal-field corrections reduce the magnitude of the nearest-neighbor exchange constant by 18% in the telluride and 9% in the selenide. The more-distant exchange is found to be larger than previously assumed but consistent with other known magnetic properties.

I. INTRODUCTION

The magnetic properties of diluted magnetic semiconductors (DMS's) such as $\mathrm{Cd}_{1-x}\mathrm{Mn}_x\mathrm{Te}$ and $\mathrm{Cd}_{1-x}\mathrm{Mn}_x\mathrm{Se}$ are currently being extensively investigated. The *very dilute* regime ($x \leq 0.1$) is particularly attractive for study because the short-ranged exchange interactions allow one to separate the magnetic response of the system into the sum of responses of small clusters (singles, pairs, triples, etc.)[1] In recent experiments on $\mathrm{Cd}_{0.95}\mathrm{Mn}_{0.05}\mathrm{Se}$ and $\mathrm{Zn}_{0.95}\mathrm{Mn}_{0.05}\mathrm{Se}$ the pair magnetization showed an abrupt increase (a step) as a function of external field at about 10 T.[2,3] A similar step and a second step at almost twice this field has also been observed in other properties dependent on the magnetization, such as the spin splitting of band-edge excitons in $\mathrm{Cd}_{1-x}\mathrm{Mn}_x\mathrm{Te}$.[4] These steps provide a direct local probe of the magnetic interactions in DMS's and are also important as a means of testing whether or not the magnetic ions are randomly distributed.

To date, step experiments have been analyzed using a cluster model including only nearest-neighbor (NN) antiferromagnetic Heisenberg interactions with random Mn site occupation.[2-4] A consequence of this model is that the second step should occur at exactly twice the field of the first, but experimentally the ratio seems to be somewhat less than 2. The observed widths are also not well accounted for by thermal broadening in this model. In this paper we show that both of these discrepancies can be resolved by taking into account the magnetic interactions of more distant neighbors with the pair. The NN-only model is generalized to treat the effects of such interactions on the high-field magnetization of zinc-blende and wurtzite DMS's. The usefulness of this approach is illustrated through a re-analysis of existing data for $\mathrm{Cd}_{1-x}\mathrm{Mn}_x\mathrm{Te}$ and $\mathrm{Cd}_{1-x}\mathrm{Mn}_x\mathrm{Se}$, which allows the extraction of more accurate values of the NN exchange constants and approximate values for second- and third-neighbor exchange constants. We hope that this work will stimulate further experiments on these materials in the very dilute regime.

II. NEAREST-NEIGHBOR CLUSTER MODEL

Before describing our generalization, it is first necessary to review the basic features of the NN-only cluster model. As described in Ref. 2 this model is similar to that introduced by Kreitman *et al.* for the analysis of low-field susceptibility data.[5] The model assumes that the probability of a magnetic ion occupying a given site is independent of the occupation of other sites. The probabilities of various clusters can then be calculated, in principle, given the crystal structure. In zinc-blende $A^{\mathrm{II}}_{1-x}\mathrm{Mn}_x B^{\mathrm{VI}}$ alloys the Mn ions lie on a fcc sublattice, and the probability that a Mn ion is a member of a particular cluster is[6] $P_1=(1-x)^{12}$ for singles, $P_2=12x(1-x)^{18}$ for nearest-neighbor pairs, $P_{3a}=18x^2(1-x)^{23}(7-5x)$ for open triangles, $P_{3b}=24x^2(1-x)^{22}$ for closed triangles, and so on. In wurtzite alloys, where the Mn ions occupy a hcp sublattice, the probabilities are the same except for P_{3b}, but the difference from the zinc-blende value is not significant for $x \leq 0.1$. In very dilute samples of either type, most spins are either singles or members of pairs.

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The model further assumes that only the NN exchange constant J_1 is appreciable, all other exchange constants being much smaller. In Mn-based DMS's J_1 is of order 10 K and antiferromagnetic. The Mn ions are assumed to have spin $S=\frac{5}{2}$, and g factor $g=2$, and to be coupled by Heisenberg interactions. The Hamiltonian for a pair is thus

$$\mathscr{H}_{\text{pair}}=-2J_1\mathbf{S}_1\cdot\mathbf{S}_2+g\mu_B(S_{1z}+S_{2z})H\ . \tag{1}$$

This Hamiltonian has energy levels

$$E(S_T,m;H)=-J_1[S_T(S_T+1)-\tfrac{35}{2}] +g\mu_B mH,\ \ 0\le S_T\le 2S \tag{2}$$

with $m=-S_T,-S_T+1,\dots,S_T$. The ground state of the pair for small H has $S_T=m=0$ and does not contribute to the magnetization. At a magnetic field $H^{(1)}$ given by the solution of $E(0,0;H^{(1)})=E(1,-1;H^{(1)})$, the $S_T=1$ state becomes the ground state, and the first step occurs at $g\mu_B H^{(1)}=2\,|J_1|$. In all, the pair magnetization $\delta m(H,T)$ has five steps with $H^{(n)}=nH^{(1)}$, $n=1,2,3,4,5$, each of height $g\mu_B$. In all DMS's examined, these steps occur at fields high enough that singles are almost completely saturated. Measurements of the steps thus provide a way of determining the NN exchange constant. The closed and open triangles with $S_T=\frac{1}{2}$ and $\frac{5}{2}$ ground states, respectively, also generate a series of steps starting at $3\,|J_1|$. Because of the small probabilities P_{3a} and P_{3b}, the triples are much less important than the pairs, and will be considered only as a small correction to the analysis of experimental data.

In addition to predicting the existence of steps, the NN-only model also explains[2] why the apparent or "technical" saturation observed at fields below the first step is always found to be less than the saturation magnetization for a *paramagnet* of the same x. This behavior results from the fact that clusters larger than singles have a small magnetization per spin in their low-field ground states, due to the antiferromagnetic coupling within each cluster. The model predicts that the technical saturation magnetization should be the sum of the magnetizations of all clusters *in their low-field ground states.* The agreement with experiment, both for this prediction and for the height of the first step, is particularly important because it provides evidence that the Mn ions in these samples are indeed randomly distributed.[2]

III. GENERALIZED CLUSTER MODEL

Despite these successes the NN-only model cannot account for the experimentally observed relationship $H^{(2)}<2H^{(1)}$. Moreover, the addition of thermal broadening in this model is not sufficient to explain the observed step widths.[3,4] We now show how both of these discrepancies can be resolved by considering the effects of distant-neighbor exchange interactions. Again assuming Heisenberg interactions,[7] the Hamiltonian for a pair $(\mathbf{S}_1,\mathbf{S}_2)$ interacting with other spins is

$$\mathscr{H}=\mathscr{H}_{\text{pair}}+\mathscr{H}_I+\mathscr{H}'\ , \tag{3}$$

where

$$\mathscr{H}_I=-2\sum_{r\ge 3}J_{(1,r)}\mathbf{S}_1\cdot\mathbf{S}_r-2\sum_{s\ge 3}J_{(2,s)}\mathbf{S}_2\cdot\mathbf{S}_s \tag{4}$$

describes each member of the pair interacting with other spins in the crystal (the sums are taken over Mn-occupied sites in a given realization of the alloy), and

$$\mathscr{H}'=g\mu_B H\sum_{n\ge 3}S_{nz}-\sum_{p,q\ge 3}J_{(p,q)}\mathbf{S}_p\cdot\mathbf{S}_q\ , \tag{5}$$

where $J_{(p,q)}$ is the exchange between spins at sites p and q, describes interactions of the other spins, excluding the chosen pair. We assume that the magnetic field is large enough, or the temperature low enough, so that $g\mu_B H\gg k_B T$. This assumption means that singles are nearly field aligned and spin fluctuations are strongly suppressed. We can then make a mean-field approximation for $\mathscr{H}_I$, leading to an effective pair Hamiltonian

$$\mathscr{H}_{\text{eff}}=\mathscr{H}_{\text{pair}}+g\mu_B h_{1z}S_{1z}+g\mu_B h_{2z}S_{2z}\ , \tag{6}$$

where

$$g\mu_B h_{1z}=-2\sum_r J_{(1,r)}\langle S_{rz}\rangle\ ,$$

and similarly for h_{2z}; the effective fields are parallel to the external field since only isotropic exchange is present. We assume that distant-neighbor exchange constants $J_{n>1}$ are weak compared to J_1; here $J_n=J_{(p,q)}$ when p and q are nth neighbors. This means that self-consistency is unnecessary to lowest order in the interactions in $\mathscr{H}_I$ and therefore that thermodynamic averages can be computed with respect to the Hamiltonian $\mathscr{H}'$.

Given $\mathscr{H}_{\text{eff}}$, it is straightforward to compute corrections to $E(S_T,m;H)$ in perturbation theory in the (small) effective fields. By dividing the perturbation in Eq. (6) into even and odd terms under spin interchange, one finds that, to lowest order,[8] the even term simply adds $\frac{1}{2}(h_{1z}+h_{2z})$ to the external field, while the odd term has vanishing expectation in the states $|S_T,m=-S_T\rangle$ because they have equal $\hat{z}$ projections on the two spins. The energies of these states then become

$$E(S_T,m;H+\tfrac{1}{2}(h_{1z}+h_{2z}))\ .$$

The first step is thus shifted to a field $H'^{(1)}$, given by

$$E(0,0;H'^{(1)}+\tfrac{1}{2}(h_{1z}+h_{2z})) =E(1,-1;H'^{(1)}+\tfrac{1}{2}(h_{1z}+h_{2z}))\ ,$$

i.e.,

$$g\mu_B H'^{(1)}=2\,|J_1|-g\mu_B\tfrac{1}{2}(h_{1z}+h_{2z})\ . \tag{7}$$

For the nth step,

$$g\mu_B H'^{(n)}=2n\,|J_1|-g\mu_B\tfrac{1}{2}(h_{1z}+h_{2z})\ .$$

All five steps are shifted by the same amount. Since the relationship $H'^{(2)}\le 2H'^{(1)}$ is observed experimentally, we infer that h_{1z},h_{2z} are negative and thus that the non-NN interactions are predominantly *antiferromagnetic.*

At finite temperatures, the steps associated with each pair are symmetrically broadened due to thermal population of the lowest levels. An actual crystal, of course, contains many pairs with different "environments,"

TABLE I. Number of nth neighbors, Z_n, and distances to them in units of a, the nearest-neighbor separation, for fcc and (ideal) hcp lattices. Shown also are exchange constants for each neighbor in the notation of the text.

n	fcc			hcp		
(neighbor)	Z_n	Distance	Exchange constant	Z_n	Distance	Exchange constant
1	12	a	J_1	12	a	J_1
2	6	$a\sqrt{2}$	J_2	6	$a\sqrt{2}$	J_2
3	24	$a\sqrt{3}$	J_3	2	$a\sqrt{8/3}$	J_3'
4	12	$2a$		18	$a\sqrt{3}$	J_3
5	24	$a\sqrt{5}$		12	$a\sqrt{11/3}$	

(h_{1z},h_{2z}), due to statistical (or nonstatistical) composition fluctuations. If $\mathcal{P}(h)$ is the probability distribution of effective fields $h\equiv\frac{1}{2}(h_{1z}+h_{2z})$, the observed pair magnetization is just

$$\delta M(H,T)=\int_{-\infty}^{\infty}\mathcal{P}(h)\delta m(H-h,T)dh \ ,$$

where $\delta m(H-h,T)$ is the magnetization of a pair with effective field h. In practice, we replace this integral with a discrete sum over environments τ characterized by an effective field h_τ and probability $\mathcal{P}_\tau$:

$$\delta M(H,T)=\sum_{\tau}\mathcal{P}_\tau\delta m(H-h_\tau,T) \ . \qquad (8)$$

For $x\leq 0.05$ the contribution of *other* pairs to the effective field is small compared to the contribution of singles: h and $\mathcal{P}(h)$ can therefore be taken to be independent of H. Thus each step has the same *shape*, and each shifts by the same amount. An important consequence is that J_1 can be extracted directly from the *difference* between the second and first steps, independently of $\mathcal{P}(h)$. In addition, since both the shape and shift can be measured from the first two steps, no further information concerning the pairs is gained by going to fields beyond the second step.

The shape of each step can be related to the magnitude of the first few exchange constants by considering an explicit model of the distribution of effective fields in zincblende and wurtzite DMS's. Here we assume (1) random Mn site-occupation probability, consistent with the agreement of step height between theory and experiment; (2) only the singles contribute significantly to h for $x\leq 0.05$. This is reasonable since there are many more singles than larger clusters. In addition, the low-lying states of larger clusters have small magnetization per spin, so they contribute little to h. (3) Only sites which are third- (fourth-) nearest neighbors or closer to either spin in the pair in the fcc (hcp) magnetic lattice need be considered. Numerical analysis shows that more distant sites may be neglected due to the rapid decrease of exchange constants with distance in nonmetallic systems.

Given these additional assumptions it is straightforward to compute $\delta M(H,T)$ for any set of exchange constants, (J_1,J_2,J_3). Here we use the notation for nth-neighbor exchange constants introduced in Table I, which emphasizes the fact that J_3 in both the fcc and hcp magnetic lattices corresponds to the same distance. Details of the calculation of probabilities and effective fields are given in the Appendix. [We assume that $\langle S_{rz}\rangle=\frac{5}{2}$ for an isolated single spin $\mathbf{S}_r$, equivalent to neglecting the second term of Eq. (5)]. For a fcc lattice, the mean effective field $\bar{h}$, giving the mean shift of the step from its NN-only location, is $g\mu_B\bar{h}\approx 4.53J_3+0.89J_2$ at $x=0.05$. This formula shows the importance of taking third neighbors into account. J_3 contributes at approximately 5 times the level of J_2, mostly because of the larger number of third neighbors on a fcc lattice (cf. Table I). For a hcp lattice at $x=0.05$, the situation is similar, if not so pronounced: $g\mu_B\bar{h}\approx 2.89J_3+0.43J_3'+0.89J_2$. Again J_3 is seen to be important.

IV. RESULTS AND DISCUSSION

Using this model we have reanalyzed existing experimental data[3,4] for $Cd_{0.95}Mn_{0.05}Te$ and $Cd_{0.95}Mn_{0.05}Se$. The data was fitted by numerical minimization of χ^2, the weighted sum of squares of deviations, as a function of the exchange constants and the pair probability P_2. It is not meaningful to directly compare our fit with previous NN-only fits since the latter allowed $H^{(2)}\neq 2H^{(1)}$, for which there is no theoretical justification. In either model the second step is completely determined once $H^{(1)}$ and $\mathcal{P}(h)$ are known. In our model $H^{(1)}$ still determines the $T=0$ *onset* of the first step, because there are still some pairs with $h=0$ although most are shifted to higher fields. Therefore, $H^{(2)}=2H^{(1)}$ holds for the onset of each step, but not for the *center*.

Since the fit is based on numerical minimization, there is no guarantee that the *absolute* minimum of χ^2 has been found, but we have searched considerable regions of parameter space in producing our fit. In addition, we require that our parameters be consistent with other known magnetic properties. Within the context of the Heisenberg Hamiltonian, these values should therefore be physically unique. The parameter errors should be considered approximate, both because of uncertainty in the experimental error and because there exists no unique definition of the error for a nonlinear regression. We adopted a widely used error convention based on a quadratic approximation to χ^2 near the minimum.[9]

To improve the fit in the region of the second pair step, it was found necessary to include the closed-triangle step. This step is expected to have about 8% of the height of a pair step and an onset at $\frac{3}{2}H^{(1)}$, and it was inserted at a range of fields between the onset and $2H^{(1)}$. The quality

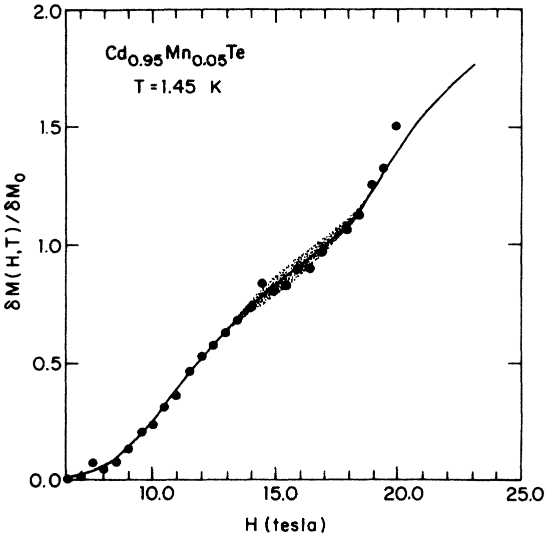

FIG. 1. Pair magnetization of $Cd_{0.95}Mn_{0.05}Te$, scaled to $\delta M_0 = g\mu_B P_2$ (the random Mn step height) as a function of magnetic field at 1.45 K. Solid circles are optical data of Ref. 3. The curve is derived from the model detailed in the text, with $J_1 = -6.3$ K, $J_2 = -1.9$ K, and $J_3 = -0.4$ K. The shaded region reflects uncertainty regarding the broadening of the small step due to closed triangles.

of the fit was insensitive to its location in this range.

Figure 1 shows the best fit for $Cd_{0.95}Mn_{0.05}Te$ to the $T = 1.45$ K optical data of Ref. 4. Shading indicates the range of curves for different locations of the closed-triangle step. Figure 2 shows a similar fit for $Cd_{0.95}Mn_{0.05}Se$ to the $T = 1.8$ K pulsed-field magnetization data of Ref. 3. In each case, $\delta M(H,T)$, scaled by $\delta M_0 \equiv g\mu_B P_2$, is plotted versus magnetic field.

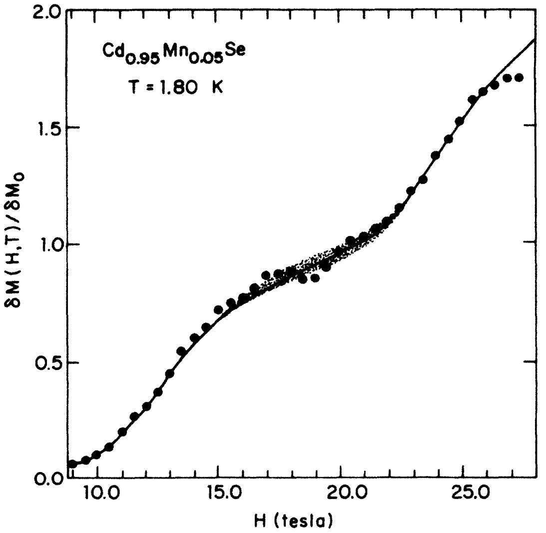

FIG. 2. Pair magnetization of $Cd_{0.95}Mn_{0.05}Se$, scaled to $\delta M_0 = g\mu_B P_2$ (the random Mn step height) as a function of magnetic field at 1.80 K. Solid circles are pulsed-field magnetization data of Ref. 2. The curve is derived from the model detailed in the text, with $J_1 = -7.9$ K, $J_2 = -1.6$ K, $J_3 = J_3' = -0.2$ K. The shaded region reflects uncertainty regarding the broadening of the small step due to closed triangles.

For $Cd_{1-x}Mn_xTe$ the best fit corresponds to $J_1 = -6.3 \pm 0.3$, $J_2 = -1.9 \pm 1.1$, and $J_3 = -0.4 \pm 0.3$ K, with a χ^2 of 0.8 per degree of freedom.[10] The optimum value of P_2 is $(90 \pm 5)\%$ of the value expected for random Mn site occupation.[11] For comparison, the NN-only analysis[4] produced $J_1 = -7.7 \pm 0.3$ K, which is a substantial overestimate of the magnitude of J_1 resulting from our analysis. The new J_1 is probably the most accurate yet determined for these materials. It is reflected directly in the spacing between steps and has little effect on their shape. The latter feature is determined by the temperature and by J_2 and J_3, which are both found to be antiferromagnetic and larger in magnitude than previously assumed. Despite the large errors in J_2 and J_3, statistical tests showed that both lead to statistically significant improvements in χ^2, and, therefore, both must be included.

The results for $Cd_{1-x}Mn_xSe$ are $J_1 = -7.9 \pm 0.5$, $J_2 = -1.6 \pm 1.5$ and $J_3' \approx J_3 \approx -0.2$ K, with a χ^2 of 0.4 per degree of freedom.[10] The optimum value of P_2 is the same as in the telluride. χ^2 is smaller and parameter errors are larger than in the telluride because of greater experimental uncertainties, due to pulsed fields and attendant sample-heating effects. It was confirmed that the above parameters also give a good fit to optical data for $Cd_{0.95}Mn_{0.05}Se$, which, however, does not extend to high enough fields to see the second step.[3] For comparison, the NN-only theory gave $J_1 = -8.7$, or -8.3 K,[3,2] which are much closer to our results than in the case of the telluride. This occurs both because of smaller J_2/J_1 and J_3/J_1 than in the telluride and because of differences between hcp and fcc environments.[12] Here statistical significance tests showed that J_2 is necessary but $J_3 = J_3' = 0$ cannot be ruled out. The minimum χ^2 does show antiferromagnetic J_3 and J_3', but a definitive answer to the question awaits further experiment.

Because of the uncertainties in J_2, J_3 it is important to check that the values determined above are consistent with other magnetic properties. Θ, the paramagnetic Curie temperature, is proportional to the thermodynamic sum $\sum_n J_n Z_n x$ for random Mn distribution. The experimental Θ extrapolated to $x = 0.05$ is[2,13] -29.2 K for $Cd_{1-x}Mn_xTe$ and -31.5 K for $Cd_{1-x}Mn_xSe$, while we calculate -28.2 ± 3.0 and 31.9 ± 3.5 K, respectively. The agreement in both cases is very good, and is slightly improved over that of the NN-only model [$\Theta = -27.0$ K for $Cd_{0.95}Mn_{0.05}Te$ (Ref. 4) and $\Theta = -30.5$ K for $Cd_{0.95}Mn_{0.05}Se$ (Ref. 2)].

In the case of $Cd_{1-x}Mn_xTe$ additional constraints are provided by the type-III antiferromagnetic (AF) short-range order observed in neutron scattering measurements on concentrated $(0.60 < x < 0.70)$ samples.[14] Minimization of the semiclassical energy per spin leads to stability conditions, for a fully occupied fcc lattice, which exchange constants must satisfy. The same conditions may reasonably be applied in the short-range-ordered case. There are experimental[15] and theoretical[16] indications that the exchange constants do not depend strongly on x, a conclusion supported by the agreement between our value of J_1 (at $x = 0.05$) and that derived from the neutron scattering (at $x = 0.60$).[17] The stability of AF type III requires

TABLE II. Effective fields h_τ, normalized by $\langle S_z \rangle$, and corresponding probabilities $\mathscr{P}_\tau$ of local environment configurations of a nearest-neighbor pair of spins in a fcc lattice.

$h_\tau / \langle S_z \rangle$	$\mathscr{P}_\tau$
0	$(1-x)^{40}$
J_3	$28x(1-x)^{39}$
$2J_3$	$4x(1-x)^{39}+354x^2(1-x)^{38}$
J_2+J_3	$4x(1-x)^{39}+96x^2(1-x)^{38}$
J_2	$4x(1-x)^{39}$
$3J_3$	$96x^2(1-x)^{38}+3276x^3(1-x)^{37}$
$2J_3+J_2$	$116x^2(1-x)^{38}+1512x^3(1-x)^{37}$
$3J_3+J_2$	$16x^2(1-x)^{38}+1960x^3(1-x)^{37}+13\,104x^4(1-x)^{36}$
$4J_3$	$6x^2(1-x)^{38}+1512x^3(1-x)^{37}+20\,475x^4(1-x)^{36}$
$2(J_2+J_3)$	$4x^2(1-x)^{38}+472x^3(1-x)^{37}+2268x^4(1-x)^{36}$
$2J_2+J_3$	$16x^2(1-x)^{38}+168x^3(1-x)^{37}$
$2J_2$	$6x^2(1-x)^{38}$
$4J_3+J_2$	$472x^3(1-x)^{37}+19\,152x^4(1-x)^{36}$
$5J_3$	$168x^3(1-x)^{37}+13\,104x^4(1-x)^{36}$
$3J_3+2J_2$	$232x^3(1-x)^{37}+6720x^4(1-x)^{36}$
$5J_3+J_2$	$24x^3(1-x)^{37}+6720x^4(1-x)^{36}$
$4J_3+2J_2$	$24x^3(1-x)^{37}+4096x^4(1-x)^{36}$
$6J_3$	$4x^3(1-x)^{37}+2268x^4(1-x)^{36}$
$2J_3+3J_2$	$24x^3(1-x)^{37}+688x^4(1-x)^{36}$
$3(J_3+J_2)$	$4x^3(1-x)^{37}+768x^4(1-x)^{36}$
J_3+3J_2	$24x^3(1-x)^{37}+112x^4(1-x)^{36}$
$3J_2$	$4x^3(1-x)^{37}$
$6J_3+J_2$	$688x^4(1-x)^{36}$
$5J_3+2J_2$	$768x^4(1-x)^{36}$
$4J_3+3J_2$	$208x^4(1-x)^{36}$
$7J_3$	$112x^4(1-x)^{36}$
$6J_3+2J_2$	$36x^4(1-x)^{36}$
$2J_3+4J_2$	$36x^4(1-x)^{36}$
$7J_3+J_2$	$16x^4(1-x)^{36}$
$5J_3+3J_2$	$16x^4(1-x)^{36}$
$3J_3+4J_2$	$16x^4(1-x)^{36}$
J_3+4J_2	$16x^4(1-x)^{36}$
$4J_2$	$x^4(1-x)^{36}$
$8J_3$	$x^4(1-x)^{36}$
$4(J_3+J_2)$	$x^4(1-x)^{36}$

$$4J_1+J_2+4J_3<0,\quad J_2<4J_3\ \text{ and } J_1<2(J_2+J_3)\ . \tag{9}$$

These inequalities are satisfied by the exchange constants derived from our fit.

In the case of $Cd_{1-x}Mn_xSe$, to the best of our knowledge, experiments to observe magnetic short-range order have not yet been done. For the hcp lattice (with $J_3'=J_3$), AF type III requires

$$\begin{gathered}4J_1+J_2+3J_3<0,\quad J_2<3J_3\ ,\\ J_1<2J_2+J_3,\ \text{ and } J_1<J_2+2J_3\ .\end{gathered} \tag{10}$$

Since these inequalities are satisfied by the set of exchange constants we derive from our fit, we predict that this material should exhibit AF type-III short-range order, assuming that sufficiently concentrated samples could be prepared in the wurtzite phase.[18] The analysis leading to Eqs. (9) and (10) would be only slightly modified by exchange constants for more distant neighbors for the same reasons that distant-neighbor corrections to effective fields are small.

The present results indicate that $|J_1|$ increases by about 25% from $Cd_{1-x}Mn_xTe$ to $Cd_{1-x}Mn_xSe$. J_2 is significant in both materials, and J_3 is also significant, at least in the telluride. The theory of Larson *et al.*[19] agrees well with these features and provides an explanation for the sign and size of the exchange constants in terms of superexchange. This theory supports the present conclusion that the interactions fall off rapidly with distance, but not so rapidly that second- and third-neighbor exchange constants are negligible.[20] Previous extractions of exchange constants from experiments on DMS's have usually assumed that only J_1, or only J_1 and J_2, are nonzero. Most published values of J_1 are within 50% of those given here, and recent determinations are even closer.[21] The convergence of experimental values for J_1 makes it important to take into account the corrections considered here, which can be 20% of J_1. To ensure accurate results, it will usually be necessary to take at least J_2 and possibly J_3 into account; J_3 may be almost as important as J_2 because of the larger number of third neighbors in these lattices.

The accuracy of the determination of (J_1,J_2,J_3) from high-field magnetization experiments can be improved by obtaining data at lower temperatures. At temperatures small compared to $|J_2|$, interesting structures should start to appear in the steps. Figure 3 shows the predicted first step in $Cd_{0.95}Mn_{0.05}Te$ at $T=0.1$ K for the (J_1,J_2,J_3) given above. At smaller x, the broadening of the steps is reduced, but their height is also reduced, so experiments are more difficult.

At low temperatures it is also possible that some structure might be seen below the first step, in the region usually fitted to a modified Brillouin function

$$M(T)=x_s S B_{5/2}(g\mu_B H/k_B(T+T_0)) .$$

Here $x_s S$ is the technical saturation magnetization, and T_0 is a phenomenological parameter which may depend on both x and T. In our model, T_0 depends on the more distant-neighbor interactions. At temperatures comparable to J_3, the magnetic state of the alloy will be complicated, because even at $x=0.05$ most spins have a third or nearer neighbor; x must be 1% or less for *second- and third-neighbor* singles and pairs to be the dominant clusters. If good samples at these compositions can be prepared and studied at such temperatures, then the second- and third-neighbor pair steps would yield a *direct* determination of J_2 and J_3, thus providing a useful complement to the approach presented here.

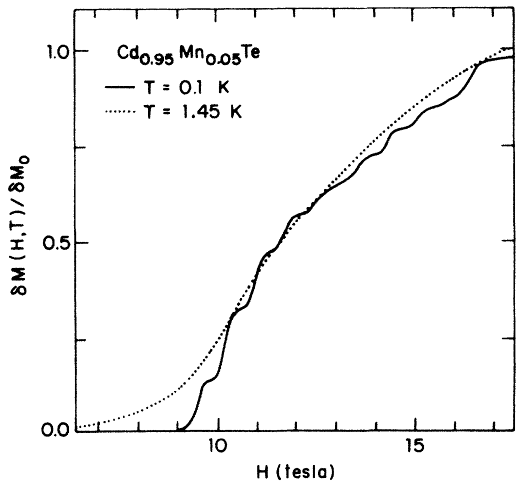

FIG. 3. Theoretical pair magnetization of $Cd_{0.95}Mn_{0.05}Te$ at $T=0.1$ K, as a function of magnetic field, for $J_1=-6.3$ K, $J_2=-1.8$ K, and $J_3=-0.4$ K. Shown for comparison is the theoretical curve of Fig. 1 for the same parameters at $T=1.45$ K.

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TABLE III. Same as Table II, but for a hcp lattice.

$h_\tau/\langle S_z\rangle$	$\mathscr{P}_\tau$
0	$(1-x)^{34}$
J_3	$20x(1-x)^{33}$
$2J_3$	$2x(1-x)^{33}+172x^2(1-x)^{32}$
J_2+J_3	$4x(1-x)^{33}+68x^2(1-x)^{32}$
J_2	$4x(1-x)^{33}$
J_3'	$4x(1-x)^{33}$
$2J_3+J_2$	$80x^2(1-x)^{32}+760x^3(1-x)^{31}$
$3J_3$	$32x^2(1-x)^{32}+1140x^3(1-x)^{31}$
J_3+J_3'	$80x^2(1-x)^{32}$
$3J_3+J_2$	$8x^2(1-x)^{32}+920x^3(1-x)^{31}$
$2J_3+J_3'$	$8x^2(1-x)^{32}+760x^3(1-x)^{31}$
$J_2+J_3+J_3'$	$16x^2(1-x)^{32}+320x^3(1-x)^{31}$
$2J_2+J_3$	$16x^2(1-x)^{32}+120x^3(1-x)^{31}$
$2(J_2+J_3)$	$4x^2(1-x)^{32}+332x^3(1-x)^{31}$
$4J_3$	$x^2(1-x)^{32}+380x^3(1-x)^{31}$
J_2+J_3'	$16x^2(1-x)^{32}$
$2J_2$	$6x^2(1-x)^{32}$
$2J_3'$	$4x^2(1-x)^{32}$
$2J_3+J_2+J_3'$	$352x^3(1-x)^{31}$
$4J_3+J_2$	$164x^3(1-x)^{31}$
$3J_3+J_3'$	$160x^3(1-x)^{31}$
$3J_3+2J_2$	$152x^3(1-x)^{31}$
J_3+2J_3'	$120x^3(1-x)^{31}$
$2J_2+J_3+J_3'$	$64x^3(1-x)^{31}$
$3J_3+J_3'+J_2$	$32x^3(1-x)^{31}$
J_2+2J_3'	$24x^3(1-x)^{31}$
$2J_3'+J_2+J_3$	$24x^3(1-x)^{31}$
$2J_2+J_3'$	$24x^3(1-x)^{31}$
$2(J_2+J_3)+J_3'$	$24x^3(1-x)^{31}$
$3J_2+J_3$	$24x^3(1-x)^{31}$
$3J_2+2J_3$	$24x^3(1-x)^{31}$
$5J_3$	$20x^3(1-x)^{31}$
$2(J_3+J_3')$	$12x^3(1-x)^{31}$
$4J_3+2J_2$	$12x^3(1-x)^{31}$
$3J_3'$	$4x^3(1-x)^{31}$
$4J_3+J_3'$	$4x^3(1-x)^{31}$
$3J_2$	$4x^3(1-x)^{31}$
$3(J_2+J_3)$	$4x^3(1-x)^{31}$
$5J_3+J_2$	$4x^3(1-x)^{31}$

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APPENDIX: CALCULATION OF EFFECTIVE-FIELD DISTRIBUTIONS

The effective-field distributions for zinc-blende and wurtzite lattices are calculated using the model described in the text. A key feature of the model is that only near-neighbor exchange constants need be considered, since exchange constants decrease rapidly with distance.[22] While it is clear that the mean effective field $\bar{h}$ is then determined by nearby spins, this is also true of the *fluctuations* in h because the relative fluctuations in a small sample of sites are stronger than in a large sample. The choice of third- (fourth-) nearest neighbors as the cutoff is motivated by examination of Z_n, the number of nth neighbors in the fcc (hcp) lattice (cf. Table I). For the fcc lattice there are 4 times as many third neighbors as second, so they may contribute significantly to h even with a smaller exchange constant. Inclusion of third neighbors of either spin of the pair leads to an environment of 40 sites (plus the 18 NN sites which must be empty to define the pair). For the hcp magnetic lattice, inclusion of fourth neighbors leads to an environment of 34 sites, again excluding 18 NN sites.

In calculating effective fields for the fcc (hcp) lattice, only configurations of the environments with 4 (3) or fewer spins were considered, not including the two spins in the pair. For $x \leq 0.05$ this approximation accounts for more than 89% of the environments and is made simply for numerical convenience. The neglected configurations have larger effective fields and therefore should not affect the *onset* of the first step, although they are necessary to get the correct overall step height. To correct for this, the missing effective fields were added arbitrarily at the location of the second step.

Following assumption (2), the effective field from a configuration should be computed *ignoring* contributions of any clusters larger than singles in the environment. Environments with three or more spins comprise only 34% of all pair environments at $x = 0.05$, and fewer at smaller x. The proportion of these containing any pairs, triples, etc. is small. Hence we have included the two-spin environments exactly, but treated the three- and four-spin environments as if they included only singles. This approximation becomes even better at smaller x.

Tables II and III show effective fields and associated probabilities for the fcc magnetic lattice and hcp magnetic lattice, respectively. Probabilities are conditional; $\mathscr{P}_\tau$ is the probability, given a pair at (1,2), that the pair experiences effective field h_τ. Thus $\sum_\tau \mathscr{P}_\tau = 1$, and to compute magnetizations the $\mathscr{P}_\tau$ must be multiplied by $g\mu_B P_2$, the height of a pair step. The quantity $h_\tau / \langle S_z \rangle$ effectively counts the number of nth-neighbor bonds from the pair to any spins in the environment. Thus, the second entry in Table I is J_3, corresponding to a single spin occupying one of the 28 sites (in the 40-site environment) which is a third neighbor to one spin in the pair and a fourth or more distant neighbor to the other. The third line, $2J_3$, corresponds to two third-neighbor bonds, which can arise in two ways: (1) two spins are present, each a third neighbor of one pair spin, but not nearest neighbors to each other (354 combinations), or (2) one spin occupies one of the four sites which are third neighbors of *both* pair spins.

Using the tables, one can compute the pair magnetization for any $x \leq 0.05$ and any set of exchange constants. In particular, the mean effective field $\bar{h}$ should give a good measure of the shift of the steps. For a fcc magnetic lattice

$$\begin{aligned} g\mu_B \bar{h} = g\mu_B \sum_\tau \mathscr{P}_\tau h_\tau \Big/ \left[\sum_\tau \mathscr{P}_\tau \right] \approx & \, 5\{J_3[20x(1-x)^3 + 710x^2(1-x)^2 + 14\,820x^3(1-x) + 182\,780x^4] \\ & + J_2[4x(1-x)^3 + 140x^2(1-x)^2 + 2964x^3(1-x) + 36\,556x^4]\} \\ & \times [(1-x)^4 + 40x(1-x)^3 + 710x^2(1-x)^2 + 9880x^3(1-x) + 91\,390x^4]^{-1} \, . \end{aligned} \tag{A1}$$

In the hcp case,

$$\begin{aligned} g\mu_B \bar{h} \approx & \, 5\{J_3[14x(1-x)^2 + 416x^2(1-x) + 7392x^3] + J_3'[2x(1-x)^2 + 64x^2(1-x) + 1056x^3] \\ & + J_2[4x(1-x)^2 + 120x^2(1-x) + 2112x^3]\} \\ & \times [(1-x)^3 + 34x(1-x)^2 + 511x^2(1-x) + 5984x^3]^{-1} \end{aligned} \tag{A2}$$

assuming $\langle S_z \rangle = \frac{5}{2}$ in both cases.

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[8] In fact, since the perturbing term commutes with total z component of angular momentum, it can never mix the states engaged in crossing, to any order in perturbation theory. Thus the step of a single pair for a given configuration (h_{1z},h_{2z}) remains abrupt at $T=0$.

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[10] We take the errors in $\delta M(H,T)/\delta M_0$, for the optical data of Ref. 4 and the pulsed-field data of Ref. 3, to be ± 0.03 and ± 0.05, respectively.

[11] This value does not allow the immediate conclusion that nonstatistical clustering is present, The true step height is not known precisely because the first and second steps overlap; the 90% figure may only reflect the fact that 10% of the pairs experience an effective field large enough to shift their first step into the region of the (majority) second step.

[12] The differences between the two lattices are reflected in the probability that a pair experiences zero effective field. For a fcc environment, this probability is $(1-x)^{40}$, while for a hcp environment, it is $(1-x)^{34}$. Since the hcp environment includes fewer sites, their expected occupation is smaller and the mean effective field is reduced.

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