the orbital character of the Fe moments.

In summary, we have presented the calculation of the DM interaction between impurity spins arising from the spin-orbit scattering of conduction electrons by nonmagnetic transition-metal impurities. We have shown that this mechanism can account for the magnitude of the anisotropy fields measured in $CuMn_x T_x$ spin-glass alloys and for their dependence on the nonmagnetic element T. Experiments are in progress on these alloys to investigate whether the effects of DM interactions are limited to producing magnetic anisotropy or whether they play a more fundamental role in determining spin-glass behavior.[16] We emphasize that the DM interactions we have calculated can be remarkably large and should give important effects in many magnetic systems with low symmetry, e.g., alloys and metallic glasses. Similar DM-type interactions between nuclei may also be at the origin of the unexplained NMR line broadening in cold-worked metals.[17]

We would like to thank A. Blandin, J. Friedel, H. Hurdequint, D. Kim, P. Monod, J. J. Préjean, R. W. Richardson, E. J. Robinson, L. Rosenberg, and S. J. Williamson for very useful discussions. This work was supported in part by the National Science Foundation under Grant No. DMR 78-25008 and by the Centre National de la Recherche Scientifique.

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Hierarchy of Exchange Interactions in a Disordered Magnetic System

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(Received 29 January 1980)

A simple and very physical model to explain the magnetic properties of heavily doped, n-type semiconductors below the metal-nonmetal transition is proposed. Based on the wide distribution of exchange interactions between donors, this model establishes a hierarchy of exchange interactions accounting for the existence of the low-lying energy levels which determine the magnetization of the donor system at low temperature, and explain the absence of a transition to an ordered antiferromagnetic state.

PACS numbers: 75.30.Hx, 71.70.Gm, 75.50.Kj

Heavily doped, n-type semiconductors undergo a transition from insulating to metallic state, when the donor concentration N_D is increased above a critical value N_C.[1] On the insulating side of the transition, the electrons are localized on randomly distributed donors and the semiconduc-

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tor may be considered as an amorphous antiferromagnet.[2,3] We propose a simple explicit model to account for the magnetic behavior of this system. Based on the wide distribution of the exchange interactions, this model considers separate pairs chosen in order of decreasing interactions. The corresponding interaction "hierarchy" is shown to dominate the magnetic properties of the donors.

These properties have recently received a renewed interest with the electron spin susceptibility and magnetization data obtained from Faraday-rotation experiments in CdS[2,3] and Si.[4] Below the transition, the susceptibility χ follows approximately a Curie-Weiss law

$$\chi = \frac{N_D \mu_B^2}{\rho k_B (T+\Theta)}, \quad (1)$$

where μ_B is the Bohr magneton, ρ the the material density, T the temperature. The Curie-Weiss temperature Θ is positive, corresponding to antiferromagnetic exchange. This confirms previous ESR results.[5] However, Faraday-rotation experiments have revealed a definite departure from the Curie-Weiss behavior at low temperature: In opposition with many spin-glasses, no ordering transition to an antiferromagnetic state has been found,[2-4] even at temperatures one order of magnitude lower than Θ. No slope discontinuity or minimum was observed in the dependence of the inverse susceptibility χ^{-1} with temperature, but a slight bending to low χ^{-1} as $T \to 0$ (Fig. 1). A close analysis of this curvature has revealed the existence of a large amount of low-lying magnetic states.[3] The magnetization was also shown to saturate less rapidly than a Brillouin function.[2,3]

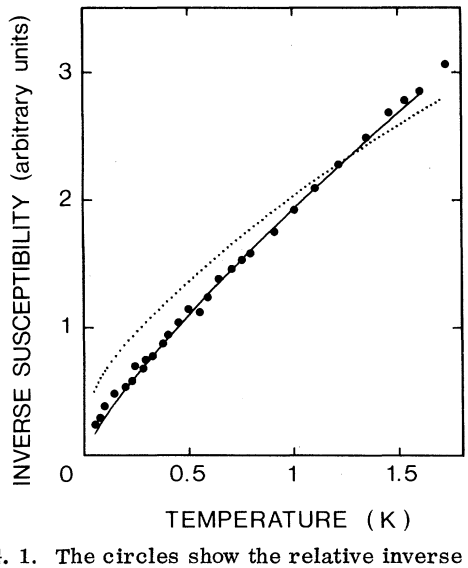

FIG. 1. The circles show the relative inverse susceptibility data obtained from Faraday-rotation experiments in CdS doped with 8×10^{16} cm^{-3} In (from Ref. 2). The solid line is the result of the calculation with use of the interaction hierarchy approximation presented in this paper. The dotted line is obtained from the nearest-neighbor approximation.

These results are in excellent agreement with a simulation calculation performed on computer "samples".[2] In this calculation, each sample is divided into clusters consisting of donors coupled by exchange interactions larger than a critical value J_C, defined by $J_C/k_B = 0.15$ K. The cluster Hamiltonian is solved exactly for clusters with up to eight donors. The exchange interactions between donors belonging to different clusters are taken into account in a molecular-field approximation, but they are found to bring minor corrections to the calculated susceptibility and magnetization. The calculation shows that relatively small clusters have a magnetic behavior similar to that of the overall spin system, and low-lying states have been found for a particular eight-spin cluster.[3]

Walstedt *et al.* have postulated that the physical reason for such magnetic properties, could be the broad distribution of exchange interactions J between donors, created by the rapid decrease of J with interdonor distance.[3] We take this wide distribution, implying large local fluctuations of the exchange interactions, as the starting point of the model. A detailed analysis of four-donor clusters will first give an illustration of this model and outline its limitations.

The usual Hamiltonian for an isolated cluster with n spins $\frac{1}{2}$ is

$$\mathcal{H} = \mathcal{H}_Z + \mathcal{H}_J, \quad (2)$$

where

$$\mathcal{H}_Z = -\sum_{i=1}^{n} g\mu_B \vec{H}_0 \cdot \vec{S}_i$$

is the Zeeman term in the magnetic field H_0, and $\mathcal{H}_J = -\sum_{i<j} J_{ij} \vec{S}_i \cdot \vec{S}_j$ is the exchange interaction for the pair i,j ($J_{ij} < 0$). The magnitude of J_{ij} decreases quasiexponentially with interdonor distance at a range of several Bohr radii.[2,3,6]

Because of the commutation of $\mathcal{H}_Z$ and $\mathcal{H}_J$ with each other and with the total spin $\vec{S}$, the eigenstates of a four-spin cluster may be separated in one quintuplet, three triplets and two singlets.

In general when the interactions J_{ij} are larger than $g\mu_B H_0$, the ground state is a singlet. Clus-

ters with one donor coupled to the others by interactions of the same order of magnitude have a triplet ground state at low magnetic field, but because of the wide distribution of exchange interactions, such a "star" configuration is not very likely. Using a simulation calculation to generate clusters[7] with four donors distributed at random, we found only 10% with this configuration.

In the general case, one of the interactions, suppose J_{12} is very probably much larger than the others: We then calculate the eigenstates of the cluster in the framework of a perturbation theory. Starting from the singlet and the triplet states of the pairs 1,2, and 3,4 (Fig. 2 inset), we assume that, in the zeroth-order approximation, the energies of the states of the cluster are the sum of the energies of these pairs. As J_{12} is much larger than J_{34}, the energy levels are separated into two groups (Fig. 2): If S_{ij} and T_{ij} refer to the singlet and triplet states of the pair i,j, respectively, we find in the lower group one singlet $S_{12}S_{34}$ and one triplet $S_{12}T_{34}$, separated by J_{34}. The upper group consists in the quintuplet, two triplets and one singlet. The energy levels of the two groups are separated approximately by J_{12}.

We take the other terms of $\mathcal{H}_J$ as a perturbation of the form $-\sum_{i<j}{}'J_{ij}\vec{S}_i\cdot\vec{S}_j$, where the prime indicates that the couples 1,2 and 3,4 are not included in the summation. These terms have no matrix elements in the lower group and are only responsible for second-order corrections, of the order of $\sum_{i<j}{}'J_{ij}^2/J_{12}$ on these energy levels, which determine the magnetic behavior of the cluster when J_{12} is much larger than $k_B T$.

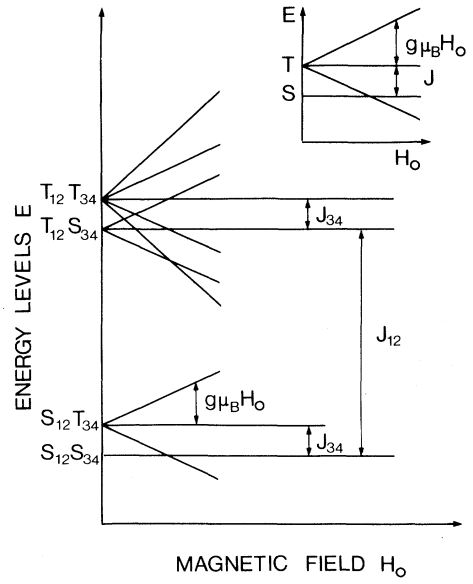

FIG. 2. Energy levels for a pair (inset) and for a four-spin cluster with J_{12} much larger than J_{34}: In the zeroth-order approximation shown here, the other interactions are neglected.

The extension of this method to the infinite spin system is straightforward: We arrange the donors in a collection of separate pairs ordered by decreasing interactions (or increasing distances). This is shown on a two-dimensional example in Fig. 3(a) and compared with the nearest-neighbor approximation in Fig. 3(b).

The calculation of the distribution of interdonor distances in these pairs is very similar to that used in the nearest-neighbor approximation[8]: The probability that there exists a pair with a distance R is the product of (i) the probability for a donor to have a neighbor at a distance R and (ii) the probability that both donors do not belong to a pair with a shorter r. The density of probability $P(R)dR$ then satisfies

$$P(R)dR = \frac{3R^2}{R_D{}^3}\left[1 - \int_0^R P(r)dr\right]^2 dR, \qquad (3)$$

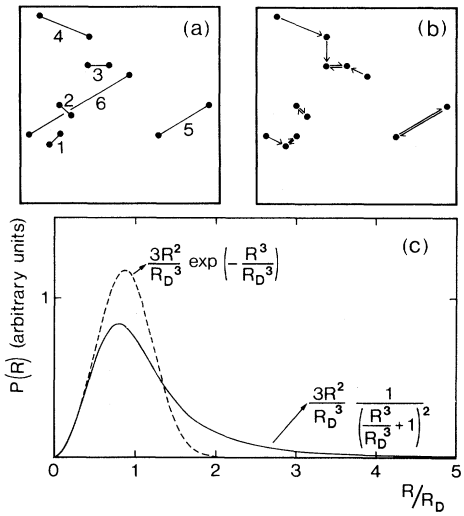

FIG. 3. (a) A system of twelve points placed at random considered as a collection of separate pairs classified by order of decreasing interactions (or increasing distances). (b) The same system considered in the nearest-neighbor approximation: The arrows show the nearest neighbor for each point. (c) The distribution functions of interdonor distances for these two approximations: The solid curve corresponds to the interaction hierarchy model proposed in this paper [3(a)], and the dashed curve corresponds to the nearest-neighbor model [3(b)].

where R_D is defined by $4\pi R_D^3/3 = N_D^{-1}$.

We find

$$P(R) = (3R^2/R_D^3)[1 + R^3/R_D^3]^{-2}. \quad (4)$$

We show in Fig. 3(c) a comparison between the function $P(R)$ (solid line) and the distance distribution in the nearest-neighbor model [dashed line; this corresponds to the distribution $P(J)$ of exchange coupling in Ref. 3].

The magnetization of one isolated pair is

$$M_P(R) = \mu_B \frac{2\sinh(g\mu_B H_0/k_B T)}{1 + \exp(-J/k_B T) + 2\cosh(g\mu_B H_0/k_B T)}. \quad (5)$$

Following the perturbation framework previously presented for four-spin clusters, we assume that the overall magnetization of the donor system is the sum of the magnetizations of the pairs defined above. We neglect interactions between these pairs, assuming that these interactions have little contribution to the lowest-energy levels which dominate the magnetic properties at low temperature. Hence, among all the interactions between donors we select a certain "hierarchy" relevant to the magnetization.

We thus take for the overall magnetization M,

$$M = \frac{N_D}{2}\int_0^\infty P(R) M_P(R) dR \quad (6)$$

from which we deduce $\chi = M/\rho H_0$.

We have compared[9] our calculated results with the Faraday-rotation data of Kummer *et al.*[2] and found a very good agreement, similar to that obtained in the cluster calculation.[2]

As an example we show in Fig. 1 a comparison between the inverse susceptibility obtained from our model (solid line) and the Faraday-rotation data for an In-doped CdS sample, with $N_D = 8\times10^{16}$ cm^{-3} (closed circles, from Ref. 2). This fit is obtained for a magnetization at saturation corresponding to a Faraday rotation $\varphi_{sat} = 1.95\times10^4$ deg/mm, very close to the result $\varphi_{sat} = 2.2\times10^4$ deg/mm of the cluster calculation.[2] This is the only adjustable parameter that we need to fit the Faraday-rotation results. Moreover, a comparison with the susceptibility data obtained by ESR in ^{5}Si shows that the absolute results of our calculation agree with experiments within the experimental error of 1.5 (essentially because of uncertainties in the determination of the donor concentration[5]).

We have used in our calculation for CdS the dependence $J(R)$ proposed by Kummer *et al.*[2] and verified that the sensitivity to the parameters that determine J (the effective Rydberg and the Bohr radius of the impurity) is the same for both calculations.

The model presented above predicts the low-lying states which dominate the magnetic properties at low temperature. This appears in the variation of $P(R)$ at high R/R_D [Fig. 3(c)]: $P(R)$ has a large tail, decreasing as R^{-4}, thus involving pairs with very small interactions. On the contrary, because of the quasiexponential decrease of the Poisson's law, the nearest-neighbor approximation underestimates the susceptibility at low temperature (Fig. 1, dotted line). At high N_D, this results in a minimum in the χ^{-1} vs T curve. Our model neglects inversions of energy levels as calculated for four-spin clusters in a "star" configuration, however, as already mentioned, these are very few: Their contribution to the magnetization, relative to other four-spin clusters, would only predominate for $H_0 \lesssim 1$ G in the CdS sample with $N_D = 8\times10^{16}$ cm^{-3}.

The "interaction hierarchy" model presented here provides a simple and physical description of the disordered spin system of the donor electrons in heavily doped semiconductors below the metal-nonmetal transition. This approximation selects the interactions, distributed over a wide range, which dominate the magnetic properties of the semiconductor. In particular it accounts for low-lying energy levels which determine the variation of the susceptibility at low temperature and the related absence of a transition to an ordered antiferromagnetic state.

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