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Spin-glass ordering in three-dimensional Heisenberg systems

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We argue that semimagnetic semiconductors are Heisenberg systems, exhibit spin-glass ordering at finite temperature, and obey power-law scaling with universal critical exponents. A theoretical argument is presented that supports spin-glass ordering in three-dimensional Heisenberg spin glasses with short-range antiferromagnetic interactions.

I. INTRODUCTION

A number of theoretical studies[1-5] predict the absence of spin-glass ordering in three-dimensional (3D) Heisenberg systems with short-range interactions, but no rigorous proof has been reported so far. To our knowledge, there is no experimental support of this prediction either. In contrast, there is at least one argument[6,7] that suggests that the lower critical dimension of Heisenberg spin glasses is as low as that of Ising spin glasses, which is presently believed[5] to be less than 3. In this paper, we first briefly discuss the models supporting the absence of transition in 3D Heisenberg spin glasses. Then we develop both theoretical and experimental arguments that support the occurrence of spin-glass order in 3D Heisenberg systems at finite temperature.

II. THEORY

A theoretical argument against 3D Heisenberg and XY spin-glass ordering has been reported by Schuster[8] who gives a tentative proof based on the Bogoliubov inequality. An improved version was given later by Ozeki and Nishimori.[4] Their conclusion, however, is not entirely convincing either, for reasons given by the authors themselves, including the use of their replica trick which is not safe.

Monte Carlo methods give a valuable hint that the transition temperature is lower for a Heisenberg spin glass than for an Ising spin glass with identical coupling constants. This may be the indication that free-energy barriers are scarce and low, but does not rule out a transition. The main weakness of the unbiased Monte Carlo method is that it can miss the absolute minimum of the free energy. Thus, it seems more convincing to force the system into two different states to be compared[1] (with periodic and antiperiodic boundary conditions respectively), and to determine the energy difference by the Monte Carlo method. In practice, however, such studies offer no decisive proof either, since the sample size is always limited in the numerical works.

Real-space renormalization-group methods[3,9] are also questionable in the case of spin glasses, because the choice of the pertinent variables that are taken into account is rather arbitary. In particular, the pseudospin variables (see below) have not been introduced in the renormalization scheme.

Let us now discuss an argument in favor of the 3D ordering in the systems of interest, namely classical models described by the Heisenberg Hamiltonian

$$\mathcal{H} = -\sum_{i,j} J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j \ , \tag{1}$$

where the random coupling constants J_{ij} between the three-dimensional spins S_i and S_j localized at sites i, and j are competing and thus imply frustration.[10] A consequence of the frustration is that, at low temperature, spins try to use the dimension of the space and form canted structures[6,11,12] associated with metastable states. A mathematical translation of this statement for 3D spins is that, if $\mathbf{S}_i$ $\mathbf{S}_j$, and $\mathbf{S}_l$ are three neighboring spins, the mixed product

$$\zeta_{ijl} = \mathbf{S}_i\cdot(\mathbf{S}_j\times\mathbf{S}_l) \tag{2}$$

is not zero in the ground state, but can take two opposite values. This is easily checked in the case of a tetrahedron of four spins if all the interactions are, for instance, antiferromagnetic and satisfy appropriate inequalities. Such a unit will be called a *highly frustrated tetrahedron* (HFT). Now imagine a Heisenberg system consisting of such HFT's embedded in a ferromagnetic or antiferromagnetic matrix. It is assumed that the HFT concentration is small, so the HFT's do not touch each other, and the exchange coupling J in the matrix is much weaker than the coupling inside the HFT's. In each HFT (labeled by λ or μ) a triangle (ijl) is chosen, and the corresponding Ising variable $\sigma_\lambda=\sigma_{ijl}=\zeta_{ijl}/|\zeta_{ijl}|=\pm 1$ is defined. We start with $J=0$ and arbitrary values of the σ_λ's and then switch on J at temperature $T=0$. It is assumed that the system loses its energy progressively without being able to jump over potential barriers. Then, because of the weakness of J, it goes into a state where the σ_λ's have the same values as before. However, because of J, this state is generally not the ground state. The ground state is generally nondegenerate and corresponds to well-defined values of the pseudospins σ_λ, except that reversal of *all* pseudospins does not change the energy. The energy of the metastable states is a function of the pseudospins σ_λ and can approximately be written as a biquadratic expression, namely

$$\tilde{\mathcal{H}}=-\sum_{\lambda,\mu} g_{\lambda\mu}\sigma_\lambda\sigma_\mu \;, \tag{3}$$

restoring an Ising picture out of the Heisenberg model, The g's are a function of the geometry. Terms with four or more pseudospins are small, because they are of higher order in J. Equation (2) ignores spin rotations and is only appropriate to describe the relative minima of Eq. (1). Spin rotations can easily be introduced through usual spin-wave variables, which are obviously continuous. Equation (2) has been derived at $T=0$, but is still correct as a Landau free-energy functional at low temperature T. The g's depend on T because of the effect of spin waves.

We have thus derived, in an *ad hoc* system of Heisenberg spins, the existence of metastable states characterized by Ising pseudospins and described by the effective Hamiltonian of Eq. (3). It would be surprising that such two-level systems did not exist in other more physical-frustrated Heisenberg systems. Indeed, numerical models did show their existence.[13] While we have used the language of pseudospins, the existence of two-level systems can be equally inferred using the language of local Onsager random anisotropy, which, contrary to the usual crystalline anisotropy, does not vanish in Heisenberg spin glasses.[14] Note that it makes irrelevant the concept of isotropic Heisenberg spin glass with the absence of irreversibilities.[15] Moreover, in a suitably modified XY model which, in contrast with our *ad*+*hoc* Heisenberg model, is of the Edwards-Anderson-type, Eq. (2) can be explicitly derived.[6,12] In that case, the σ_λ's denote vector products of two spins instead of mixed products of three spins. Equation (2) implies the order of the σ_λ's at $T=0$. This order has no reason to be ferromagnetic or antiferromagnetic, and actually we prove in the Appendix that a careful investigation of *ad hoc* models reveals the existence of competing interactions between the σ_λ's. Therefore, an order of the spin glass type is expected. The reader will perhaps wonder whether this order of strange pseudospins implies the order of ordinary spins. In other words, is the knowledge of the pseudospins sufficient to determine a relative minimum of Eq. (1) when the J's are known as well as the orientation of two spins? One can easily convince oneself that the answer is yes for finite systems, for XY systems, and presumably in all cases $T=0$.

The above argument does not necessarily imply the existence of Heisenberg spin glasses since thermal fluctuations might destroy the order. Even in the XY case where Eq. (2) is clearly valid, the pseudospin Hamiltonian is not of the Edwards-Anderson-type, and there are no computational or other theoretical studies supporting (or infirming) a transition at finite temperature. However, it would be surprising that certain frustrated random systems have spin-glass order at finite temperature, while others have none. Thus, we speculate that Heisenberg spin glasses with short-range interactions have the same lower critical dimension as Ising spin glasses with short-range interactions. If the present belief about the latter is correct, Heisenberg spin glasses should have a phase transition in three dimensions.

III. EXPERIMENTS

To experimentally confirm an ordering in Heisenberg spin glasses, one must first consider materials where the crystalline anisotropy is sufficiently small. In metals, this situation is always questionable, because the spin-orbit scattering of the free carriers by nonmagnetic impurities results in large anisotropic fields.[16] Among insulating spin glasses $Eu_{1-x}Sr_xS$ has been investigated with particular care, but then the spin-glass ordering temperature T_g is so small that anisotropic dipole interactions cannot be neglected.[17] Similar objections hold for many other investigated insulating spin glasses. For instance, in manganese fluorophosphate glasses, Tg is only a few Kelvin, while the local anisotropy is expected to be large.[18] In $CsNiFeF_6$ where $T_g \simeq 5$ K,[19] the spin-orbit interaction, although small, is not negligible, so Dzyaloshinki-Moriya (DM) interactions,[16] for example, might be important to establish anisotropy.

Let us now argue that the best examples of Heisenberg spin glasses are semimagnetic semiconductors such as $Hg_{1-x}Mn_xTe$, and $Cd_{1-x}Mn_xTe$, for intermediate Mn concentrations $0.2<x<0.6$.[20] First, the transition temperatures are large: $T_g=8.4$ K in $Hg_{0.7}Mn_{0.3}Te$, while $T_g=6.45$ K and 12.9 K in $Cd_{1-x}Mn_xTe$ for $x=0.3$ and $x=0.4$, respectively.[21,22] In $Cd_{1-x}Mn_xTe$, it has been argued that the dominant magnetic exchange interaction is the antiferromagnetic (AF) superexchange coupling between the Mn^{2+} ions.[23] This is also presumably the case in $Hg_{0.7}Mn_{0.3}Te$.[24] This interaction is short range: the superexchange coupling between next-nearest neighbors J_2 is antiferromagnetic like the coupling constant J_1 between nearest neighbors (NN), but it is 1 order of magnitude smaller than J_1,[23] and thus does not play any significant role.

On the other hand, the main anisotropic exchange interaction is a DM-like interaction arising from a virtual coupling between the d states of the Mn^{2+} ions and the electron states of the valence band.[23] This anisotropic interaction, just as the isotropic exchange, is short range, and it can thus be restricted to the NN coupling constant D_1 only. It is, however, 1 order of magnitude smaller than J_1. For example, in $Cd_{1-x}Mn_xTe$, $|D_1/J_1|=0.054$, independent of x.[23] The same order of magnitude is expected in $Hg_{1-x}Mn_xTe$, although D_1 has not been explicitly calculated in such compounds.[24] Therefore, this anisotropic part cannot be responsible for the spin-glass freezing process. Another source of anisotropy is the dipolar interaction. Since this dipolar coupling $d=0.013$ K (Ref. 23) is even smaller than D_1, it *a fortiori* cannot produce any order at T_g. However, contrary to the previous anisotropic- and anisotropic-exchange contributions discussed above, the dipolar coupling is long range, and one can wonder whether a combination of the exchange and dipole interactions might not be effective.[25] For instance, the isotropic exchange might be the dominant mechanism at length scales smaller than some value ξ, while dipole interactions would be effective for larger lengths. However, this scheme does not work because the effective dipole interaction between cells of size ξ^3 should be of order $\gamma\xi^{-3}(\xi^{3/2})^2=\gamma$. Here, γ is the strength of microscopic dipole interactions (of the order of 1 K), $\gamma\xi^{-3}$ is the dipole interaction at distance ξ, and $\xi^{3/2}$ is the average total spin of a cell of size ξ^3. Thus, the effective dipole interaction is expected to be independent of the scale, and negligible at all scales. Note this argument is wrong in the presence of ferromagnetic short-range order, since the cell spin might be larger than $\xi^{3/2}$. Such an order, which exists for example in $Eu_{1-x}Sr_xS$, is absent in both $Hg_{1-x}Mn_xTe$ and $Cd_{1-x}Mn_xTe$ considered here.

We thus decide to consider $Hg_{1-x}Mn_xTe$ and $Cd_{1-x}Mn_xTe$ $(0.2<x<0.6)$ as Heisenberg spin glasses, in which the spin-glass freezing occurs from purely antiferromagnetic short-range interactions. Those are, in essence, archetypes of the spin glasses envisioned in Ref. 11. With this regard, it may be desirable to discuss the value of T_g with respect to J_1 in these materials. An estimation of the magnetic energy per magnetic ion, in the mean-field approximation (MFA), is $E_m=-zJ_1S^2$, where $z=12x$ is the average coordination number. The reduction of T_g by the quantum-spin fluctuations, not taken into account in the MFA, can be characterized by the ratio

$$r_n=T_g/E_m=T_g/(z|J_1|S^2),$$

indexed by the number of spin components n (three for Heisenberg systems). Since $J_1=-5.7$ K in $Cd_{1-x}Mn_xTe$,[23] we find $r_3=0.05$ and 0.08 for $x=0.3$ and 0.4, respectively. Note the x dependence of r_3 comes from the nonlinear variation of T_g as a function of x, a consequence of the short-range nature of the exchange interaction.[26] To our knowledge, no theoretical estimate of r_3 is available. On the other hand, Ogielski and Morgenstern[27] have determined that $T_g/|J_1|=1.22$ for the case of Ising spins on a square lattice ($S=1$, $z=6$), hence $r_1=0.2$. We thus find $r_1/r_3=2.5$ and 4 for $x=0.3$, and 0.4, respectively.[28] Therefore, although r_3 is significantly smaller than r_1, in agreement with the Monte Carlo predictions,[1] both parameters have the same order of magnitude as is expected from our theoretical arguments and Eq. (2) in the preceding section.

Let us now characterize T_g as an ordering temperature. The best experimental test of the spin-glass *transition* is the observation of scaling laws. Critical activated dynamic scaling has been recently proposed for semimagnetic semiconductors.[29] Such a scaling is plausible in random-field systems where the transition is believed to be governed by a zero-temperature fixed point. In disordered antiferromagnets, however, the random field vanishes in the limit of small applied magnetic fields where the scaling properties are investigated. Therefore, the semimagnetic semiconductors we are considering here are not random-field systems. Nevertheless, there is no proof that activated dynamics should not occur in spin glasses; therefore, we found it desirable to test the activated dynamics in our materials. Although the experimental data can be interpreted in terms of activated scaling for each sample separately, the critical exponents obtained from such a scaling were found to be different in various samples such as $Hg_{0.7}Mn_{0.3}Te$, $Cd_{0.7}Mn_{0.3}Te$,[21] and $Cd_{0.6}Mn_{0.4}Te$.[29] This inconsistency is at odds with the expected universality of the critical exponents, which rules out the activated-dynamic scaling in these materials. To the contrary, we have found that the experimental data can be interpreted in terms of power-scaling laws with universal exponents for these various samples. In dynamic scaling, the imaginary part of the ac magnetic susceptibility $\chi''(\omega)$ at frequency ω has been found to satisfy a law of the form[21]

$$\chi''(\omega,T)=t^{\beta}F(\omega\tau_0\tau^{-z\nu}) , \tag{4}$$

where ν is the critical exponent for the correlation length ξ, z is the dynamic exponent relating ξ and the relaxation time $\tau\propto\xi^z$, and τ_0 is a microscopic-relaxation time. t is the reduced temperature: $t=T/T_c-1$, with T_c the zero-field static spin-glass-ordering temperature. The values of $z\nu$ and β for $Hg_{0.7}Mn_{0.3}Te$ and $Cd_{0.7}Mn_{0.3}Te$ obtained from this scaling are reported in Table I. Moreover, the same function F has been obtained for both materials, as expected for universality. In the limit $\omega\tau\ll1$, the asymptotic form of Eq. (4) can be written

$$\chi''(\omega,T)=\omega\tau_0t^{-z\nu+\beta} . \tag{5}$$

This dynamic scaling has also been tested in the same range of observation times 10^{-5} s $<t_{\mathrm{obs}}=2\pi/\omega<1$ s for both samples. The values of $z\nu-\beta$ yielded by this scaling, also reported in Table I, are consistent with the values of $z\nu$ and β derived from the full scaling of χ'' according to Eq. (4). To explore the scaling law given by Eq. (5) closer to T_c, an investigation of $\chi''(\omega,T)$ has been achieved up to longer observation times, allowing a test of the validity of Eq. (2) in $Cd_{0.6}Mn_{0.4}Te$, in the full range 10^{-5} s $<t_{\mathrm{obs}}<3\times10^3$ s, corresponding to an explored critical range of reduced temperatures $7\times10^{-3}<t<0.2$.[22] This study also yields an exponent $z\nu-\beta$ in agreement with the other values in Table I, including results obtained in $Cd_{1-x}Mn_xTe$, for different manganese concentrations.[30] (An overestimated value of $z\nu$ has been reported in Ref. 29, for reasons which have been discussed elsewhere.[31] How good the fits of the data to Eq. (4) and (5) can be appreciated in Fig. 2 of Ref. 21 and Fig.

TABLE I. Critical exponents in $Cd_{1-x}Mn_xTe$ and $Hg_{1-x}Mn_xTe$ Heisenberg spin-glasses. The uncertainties of the exponents are for the entire range of values quoted in the text.

Compound	$z\nu$	$z\nu-\beta$	γ	β	$\phi=\gamma+\beta$
$Cd_{0.55}Mn_{0.45}Te$[a]		10 ± 2			
$Cd_{0.6}Mn_{0.4}Te$[b,c]		8.8 ± 0.9	3.3 ± 0.3	0.9 ± 0.2	
$Cd_{0.7}Mn_{0.3}Te$[d]	9.25 ± 0.5	8.85 ± 0.45		0.8 ± 0.1	4 ± 1
$Hg_{0.7}Mn_{0.2}Te$[d]	9.5 ± 0.5	8.5 ± 0.5		0.8 ± 0.1	$3.5 = -1$

[a] Reference 26.
[b] Reference 22.
[c] Reference 28.
[d] Reference 21.

7 of Ref. 22 respectively, for $Hg_{1-x}Mn_xTe$ and $Cd_{1-x}Mn_xTe$ compounds. In an applied field H, in the limit $\omega\tau \ll 1$, the generalized scaling law of Eq. (5) is[21]

$$\chi''=\omega\tau_0[t(H)]^{\beta-z\nu}G[H^{2/\phi}/t(H)] . \tag{6}$$

In-field dynamic measurements[21] at observation times up to 100 s have shown that this scaling law is satisfied in $Hg_{0.7}Mn_{0.3}Te$ and $Cd_{0.7}Mn_{0.3}Te$ in the range $0.06 < t < 0.1$, allowing us to deduce the crossover exponent ϕ (see Table I).

To investigate unambiguously the occurrence of the spin-glass-phase transitions, the most relevant scaling is eventually the static scaling of the nonlinear magnetization:

$$M_s(t,H)=t^{(\pi+2\beta)/2}F(H/t^{\phi/2}) . \tag{7}$$

This scaling has been tested in $Cd_{0.6}Mn_{0.4}Te$, yielding the exponents β and γ (see Table I).[32] Taking the scaling relation $\phi=\beta+\gamma$ into account, the results reported in Table I give evidence that there is a self-consistency of the various and independent determinations of $z\nu$, β, and ϕ or γ in the 3D Heisenberg spin glasses.

IV. CONCLUSION

Through various studies of semimagnetic semiconductors, we conclude that the 3D Heisenberg spin glasses do order. The phase transition has been well characterized by both static and dynamic scaling laws, with universal critical exponents. We note the argument that the Heisenberg systems [Eq. (1)] can be described by the Ising Hamiltonian [Eq. (3)] implies that the critical behavior, i.e., the critical exponents, should be the same in insulating spin glasses, whether they are Ising or Heisenberg spin glasses, with short-range magnetic interactions. Indeed, we note the critical exponents relevant for the Heisenberg systems in Table I are in good agreement with the values determined in other systems mentioned in this paper, such as γ and β in amorphous manganese fluorophosphate and aluminosilicate,[18] and $z\nu$ in $Eu_{1-x}Sr_xS$,[17] in particular, although these systems may not be Heisenberg spin glasses. The present theory does not anticipate that metallic spin glasses with long-range Ruderman-Kittel-Kasuya-Yosida (RKKY) interactions should behave like Heisenberg insulating spin glasses with short-range antiferromagnetic interactions. We can notice, however, that the experimental data reported so far do not reveal the existence of different classes of universality among spin glasses.

APPENDIX: ISING PSEUDOSPINS IN A HEISENBERG MODEL

The best way to demonstrate the possibility of Ising pseudospins in a Heisenberg model is to start with a magnetically ordered periodic system and introduce a single impurity. Then a second impurity can be introduced in order to study the interactions and prove that they are competing. This program will be carried out in this Appendix. Since two impurities certainly do not destroy the periodic order, we will not prove that a spin glass arises.

The magnetically ordered system will be chosen such as to make the proof as easy as possible. It is convenient to choose a magnetic order that has already spin components in two space directions. Thus the appearance of a third component will give at once a nonvanishing value of the mixed product ξ_{ijl} defined by Eq. (2). For this reason, we choose a two-dimensional triangular lattice (large circles in Fig. 1) with antiferromagnetic interactions J between nearest neighbors. This argument will be

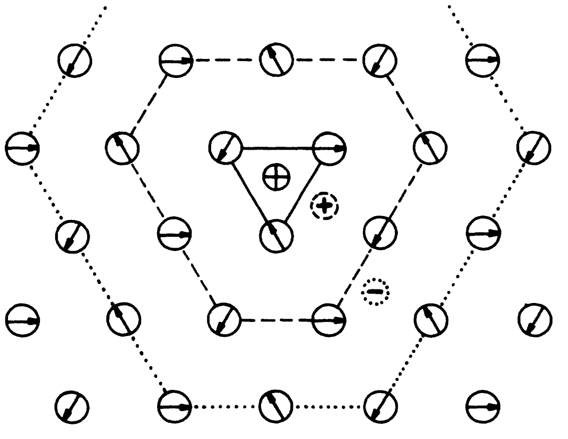

FIG. 1. Interstitial impurities (small circles) antiferromagnetically coupled with an antiferromagnetic triangular lattice. The S_z components of the solid and dashed small circles are ferromagnetically coupled. Those of the solid and dotted small circles are antiferromagnetically coupled.

given at $T=0$ so thermal fluctuations do not destroy long-range order. The ground state consists of three sublattices with magnetizations making angles $2\pi/3$ in a plane, hereafter called the (S_x, S_y) plane.

Disorder is then introduced through additional spins randomly distributed at interstitial positions at the center of the triangles (small circles in Fig. 1). These spins interact through antiferromagnetic interactions J' with the neighboring big circles. It will be first assumed there is only one interstitial impurity (small circle drawn on solid line in Fig. 1). Its magnetization will be directed along the S_z axis, e.g., with a positive sign. Thus, the mixed product ξ_{ijl} is now nonvanishing if i, j, and l designate, for instance, the three neighbors of the impurity. These three spins form what will be called the "first shell" around the impurity (solid line triangle in Fig. 1). Their magnetization has a negative S_z component (due to J'). The spins of the "second shell" (dashed line) that are common neighbors to two spins of the first shell have a positive S_z component with an absolute value smaller than the spins of the first shell. Presumably, the corner spins of the second shell also have a positive S_z, because they have a neighbor in the first shell and one in the second shell; the latter has a positive S_z and the former a negative S_z with a larger absolute value. A similar argument suggests that the spins of the third shell (dotted line) have a negative S_z, at least the ones that are not at the corner. This qualitative discussion might be replaced by an exact argument for classical spins, but the mathematics, although elementrary, would be rather tedious, even in the linear-spin-wave approximation, which can be used only if $J'J \ll 1$.

The interactions between two impurities can now be discussed. If an additional impurity is introduced at the dashed small circle in Fig. 1, it tends to have its S_z opposite to its neighbors, which are mostly on the first shell, i.e., with a negative S_z. Thus, it has a positive S_z. If the second impurity is introduced, not at the dashed small circle, but at the dotted small circle instead, then the neighbors are mostly on the second shell and therefore likely to have a positive S_z. To summarize, the z components of two impurities at nearest-neighbor positions (solid and dashed small circles in Fig. 1) are ferromagnetically coupled while those of two impurities at fourth-neighbor positions (solid and dotted small circles in Fig. 1) are antiferromagnetically coupled. It is easily deduced that the Ising pseudospins defined by $\xi_{ijl}/|\xi_{ijl}|$ for each triangle surrounding an impurity are coupled antiferromagnetically for nearest neighbors and ferromagnetically for fourth neighbors. The interactions between second, third, and fifth neighbors might be discussed in the frame of a serious calculation, which, as stated above, would be rather complicated. These interactions are expected to be weaker, since there is one neighbor in the first shell, and two in the second shell; two is greater than one, but the second shell is less magnetized than the first shell.

Thus, the existence of competing interactions between Ising pseudospins is established for the model of Fig. 1. It is expected that such interactions are still there for any concentration of impurities, so the minima of the energy can be obtained by minimizing a Hamiltonian function of these pseudospins. This Hamiltonian can be represented by the first term of Eq. (3), plus three-spin terms, four-spin terms, etc., which presumably do not play an essential role. The absolute minimum will presumably be a spin glass for appropriate impurity concentrations. It is not forbidden to assume some correlations in the impurity distribution, favoring or not nearest-neighbor pairing.

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