Behavior of an Ising model with randomly mixed classical and quantal spins

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The observation that deuterons tunnel much less than protons in tunneling mediated phase transitions suggests that it might be useful to look at disordered quantum-spin models in which the disorder resides entirely in the size of the quantal fluctuations. As an example, we consider a system of randomly mixed classical and quantum-mechanical (tunneling) spins. The mean-field theory phase diagram shows that sufficiently dilute quantal spins cannot lead to zero-temperature disorder. In addition, our analysis of the infinite transverse field limit suggests that the critical behavior must differ from that of the pure classical problem.

There are numerous examples of hydrogen-bonded solids that undergo order-disorder phase transitions involving significant proton tunneling. Ferroelectric materials of the KDP(KH_2PO_4) class are typical in this regard.[1] One knows that it is, in fact, tunneling that controls the proton rearrangement in these substances largely because of two experimental observations: First of all, by increasing the pressure the order-disorder transition can be made to occur at zero temperature[2]—indicating the primary role of quantum effects.[3] Second, at any lower pressure the transition temperature can be increased dramatically simply by replacing all the mobile protons with deuterium—a result consistent with the finding that the relevant deuterium tunneling frequencies are typically orders of magnitude less than the proton ones.[1] This last observation, however, is sufficiently intriguing in its own regard that it prompts one to wonder what happens to the phase transition with a *quenched random mixture of H and D.* Indeed, it is this question which motivates this Brief Report.

A rigorous treatment of KDP-like crystals is somewhat involved, both because of the ice-rule correlations and because the elementary fluctuating degrees of freedom are not really single protons.[1] Nonetheless, it is revealing to study the problem in the context of a commonly used model which ignores such subtleties: the Ising model in a transverse field,[4] defined by the Hamiltonian

$$H=-J\sum_{\langle i,j\rangle}\sigma_i^z\sigma_j^z-\sum_j K_j\sigma_j^x \ , \qquad (1)$$

where σ^z and σ^x are Pauli matrices, J is the interaction between "spins," K_j is the transverse field at lattice site j, and the first sum is over nearest-neighbor lattice sites. Normally, all the K_j's are set equal to a single tunneling integral K, which is then taken to describe the tunneling process which switches the proton between two possible states at a lattice site, flipping the spin. If this K were zero, we would recover the classical Ising model, but as K is increased from zero, it is known that the critical temperature decreases until $T_c=0$ for $K=K_c$.

The version of the model we propose is an *Ising model in a random transverse field.* That is, the values of K at each site are now distributed according to a probability distribution:

$$P(K_j)=(1-x)\delta(K_j)+(x)\delta(K_j-K) \ , \qquad (2)$$

with x the concentration of quantal spins (protons with tunneling integral K) and $1-x$ the concentration of classical spins (deuterons with tunneling integral 0).[5] Hence, we are considering a quenched disordered system in which the randomness resides entirely in the quantal part of the problem, a situation which, itself, automatically raises some intriguing questions. For our model in particular, we would like to know if there will continue to be a critical K value for *all* concentrations of quantal spins or only for sufficiently large x. And, if the latter, what is it that determines the critical x value?

Before we try to answer these questions, it is worth noting some easily recognizable limiting behaviors. For example, for both $x=0$ (no quantal spins), and $K=0$ (no tunneling), the system reduces to a pure, classical Ising model. When $x=1$ the system becomes a pure quantal Ising model. An even more interesting behavior is obtained by letting $K\rightarrow\infty$. Physically, this limit corresponds to increasing the tunneling frequency of quantal spins, which, presumably ought to make then effectively less coupled to the nontunneling (classical) spins.[6] We can make this idea more precise by considering the partition function for the system:

$$Z=\mathrm{Tr}\exp(-\beta H) \ .$$

The (partial) trace over the quantal spins can easily be carried out in the limit $K\rightarrow\infty$ by splitting the Hamiltonian as

$$H=H_{\mathrm{cl}}+H_{\mathrm{qm}} \ ,$$

where H_{cl} includes interactions between classical spins, whereas H_{qm} contains the interactions between different quantal spins and that between classical and quantal spins as well as the transverse fields acting on quantal spins. Because spin operators at different sites commute, and since $H_{\rm cl}$ contains only σ^z matrices, $H_{\rm cl}$ and $H_{\rm qm}$ also commute, so that the partition function can be written as

$$Z = \mathrm{Tr}_{\rm cl}[\exp(-\beta H_{\rm cl}) Z_{\rm qm}] \ ,$$

where

$$Z_{\rm qm} = \mathrm{Tr}_{\rm qm} \exp(-\beta H_{\rm qm}) \ .$$

Note that $Z_{\rm qm}$ is, for general K, still a function of classical spins; however, as $K \rightarrow \infty$, it is easy to see that $Z_{\rm qm}$ reduces to the constant $[2\cosh(\beta K)]^{xN}$. Therefore, in the extreme quantum limit our problem reduces to a classical (site) diluted Ising model. As x is increased, the critical temperature is depressed, until $T_c = 0$ for $1-x = 1-x_c$, the site percolation threshold for classical spins.

Based on this evidence, we can expect a phase diagram resembling that presented in Fig. 1. In order to get the quantitative results for this phase diagram (which are actually presented in the figure) we perform a simple mean-field-theory calculation. The most elementary version of mean field theory[4,6] is obtained by replacing the fluctuating variables corresponding to the neighbors of a tagged spin by a constant molecular field m, which is subsequently identified with the average magnetization of the system. A self-consistency equation is then obtained by requiring that the expectation value of the tagged spin be equal to m. However, in problems involving quenched disorder, one has to resort to slightly more sophisticated methods[7,8] in order not to miss phenomena such as percolation.

One of the most efficient methods is based on an approximate application of the Callen identities[9,10] to quantal spin systems.[11] In this approach, one still calculates the average of the tagged spin $\langle \sigma_0 \rangle$ while fixing the values of all the other spins. However, instead of replacing the neighboring spins by a constant, an improvement is obtained by replacing them with *classical* fluctuating spin variables $S_j = \pm 1$, where $j = 1, \ldots, z$ indexes the nearest neighbors of the spin. The problem is then reduced to calculating $\langle \sigma_0 \rangle$ for a single quantal spin in a fixed, external (longitudinal) field $-J\sum S_j$. The straightforward result is

$$\langle \sigma_0^z \rangle = \frac{\tanh\left[\left[\left(\beta J \sum_{j=1}^{z} S_j\right)^2 + (\beta K)^2\right]^{1/2}\right]}{\left[\left(\beta J \sum_{j=1}^{z} S_j\right)^2 + (\beta K)^2\right]^{1/2}} \left(\beta J \sum_{j=1}^{z} S_j\right) \ . \qquad (3)$$

At this stage one can make use of the nontrivial algebra of classical spin variables. The idea is to rearrange the right-hand side (RHS) of the equation (without making any approximations) into the form[10]

$$\mathrm{RHS} \equiv R(\{S_j\}) = A(\beta J, \beta K) v^{(1)} + B(\beta J, \beta K) v^{(2)} + \cdots \ , \qquad (4)$$

where

$$v^{(1)} = \sum_j S_j, \ v^{(2)} = \sum_{i<j<k} S_i S_j S_k, \ldots \ . \qquad (5)$$

This process can be carried out systematically by observing that general functions of z spin variables form a Hilbert space provided we define an inner product by

$$(f,g) = \sum_{\{S_j\}} f(\{S_j\}) g(\{S_j\}) \ . \qquad (6)$$

It is easy to show that $v^{(1)}, v^{(2)}, \ldots$, are orthogonal functions with respect to this inner product, so that we obtain

$$A = (R, v^{(1)})/(v^{(1)}, v^{(1)}) \ , \qquad B = (R, v^{(2)})/(v^{(2)}, v^{(2)}), \ldots \ . \qquad (7)$$

The calculation of sums over spins involved in evaluating the inner products can be further simplified if the functions depend only on $v^{(1)} = \sum S_j$. In this instance we can sum over distinct values of $v^{(1)}$, weighting the terms by appropriate combinatorial factors. In general,

$$\sum_{\{S_j\}} f\left(\sum S_j\right) = \sum_{k=0}^{z} \binom{z}{k} f(2k-z) \ . \qquad (8)$$

To finish the calculation, we have to average Eq. (3) over both the spin configurations and the disorder (transverse fields). Given the form of Eq. (4) though, this averaging reduces to averaging $v^{(1)}, v^{(2)}, \ldots$, over spins, and $A, B, \ldots$, over disorder. The mean-field theory is then obtained by factoring the spin moments

$$\langle v^{(1)} \rangle = zm, \ \langle v^{(2)} \rangle = z(z-1)(z-2)m^3/6, \ldots,$$

so the self-consistency condition reads

$$m = \overline{zA} m + \overline{z(z-1)(z-2)B} m^3/6 + O(m^5) \ , \qquad (9)$$

where the overbar indicates an average over the disorder. The final equation for the critical surface is

$$1 = \overline{zA} \ , \qquad (10)$$

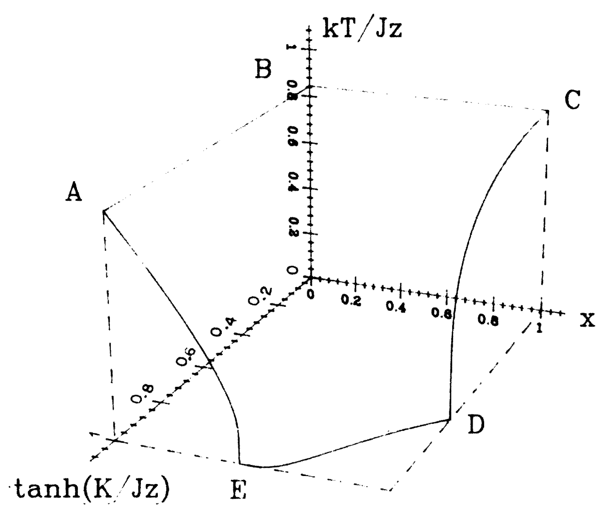

FIG. 1. Temperature-concentration-tunneling integral phase diagram for an Ising model with a fraction x of quantal spins and fraction $1-x$ of classical spins. The results shown are for coordination number $z=6$. Note that the tunneling integral is plotted as $\tanh(K/Jz)$ in order to map the whole $0 \le K \le \infty$ range into (0,1). The ferromagnetic phase resides under the critical surface *ABCDE* and the paramagnetic phase sits above the surface.

which, with the aid of Eqs. (3) and (5)–(8) can be written as

$$1=2^{-z}\sum_{k=0}^{z}\binom{z}{k}(2k-z)\left[(1-x)\tanh[\beta J(2k-z)]+x(2k-z)\frac{\tanh\{[\beta J(2k-z)]^2+(\beta K)^2\}^{1/2}}{[(2k-z)^2+(K/J)^2]^{1/2}}\right]. \tag{11}$$

This algebraic equation can easily be solved numerically, and the results for $z=6$ are shown in Fig. 1. Our results confirm the expected behavior, giving in particular a zero-temperature (percolation) transition at $x_c=0.47$ for $K/Jz=\infty$. As the transverse field is reduced, the zero-temperature critical concentration is increased until for $x=1$ (the pure transverse Ising model) we obtain a critical transverse field of $K_c/Jz=0.78$. For both $x=0$ and $K/Jz=0$ (the pure classical Ising model) the critical temperature is $T_c/Jz=0.85$. We can get an idea of the relative accuracy of our approximations by comparing these figures with known series results:[12] For a simple cubic lattice $(z=6)$, $(x_c)^{\text{series}}=0.69$, $(K_c/Jz)^{\text{series}}=0.85$, $(T_c/Jz)^{\text{series}}=0.75$. The quantitative accuracy of the results can be further increased by using a larger spin clusters; alternatively one can employ path-integral methods to provide a better treatment of the quantal spins on the exterior of the cluster.[6]

What should clearly be the next step once the global phase diagram is known, is to discuss the critical behavior of our system. For pure quantum spin systems, the critical behavior is well understood: although it is true that at zero temperature, the transition becomes a $(d+1)$-dimensional classical one,[13] at any finite temperature quantal fluctuations become irrelevant and the system crosses over to d-dimensional classical behavior.[14] In renormalization-group (RG) language, the system flows away from the zero-temperature "quantal" fixed point $(T=0,\ K=K_c)$, and towards the "classical" fixed point $(T=T_c^{\text{cl}}, K=0)$ under rescaling. A similar behavior has been established for other pure quantum-mechanical models[14] and even for disordered quantal models.[15] As a consequence it is now widely believed that quantum-mechanical effects can be ignored in the study of critical phenomena at finite temperature. For our system, since for both $x=0$ (the pure classical Ising model) and $x=1$ (the pure quantal Ising model), the behavior at finite temperature is indeed classical, one would naively expect the situation to remain unchanged at intermediate concentrations. However, such a conclusion is in direct conflict with a heuristic criterion advanced by Harris[16] which predicts that the disorder alters the critical behavior whenever the specific-heat exponent α for a pure system is positive, as is indeed the case for our system.[17]

Presumably the best way out of this dilemma would be to do an explicit renormalization-group calculation. Although we will not do so in this paper, it is still possible to reconcile these ideas by using a simple stability analysis for the various fixed points. The two fixed points already present in the pure limit are zero-temperature "quantal" fixed point (D in Fig. 1), and the "classical" fixed point (C in Fig. 1). Note that for more general x, the whole critical line ABC corresponds to a single physical system, namely the classical pure Ising model, and should therefore be considered as a single fixed point. When disorder is introduced by mixing classical and quantal spins, a new "percolation" fixed point (E in Fig. 1) appears at $K/Jz=\infty$, corresponding to the fact that our system reduces to the diluted classical Ising model in that limit. Fortunately, the classical diluted Ising model itself has been thoroughly investigated over the years by a variety of RG methods.[18] These studies have been able to confirm the Harris criterion and establish the relevance of disorder for this problem, giving a flow *away* from the pure fixed point (A). Since our proof of the equivalence of our system to a diluted classical Ising model when $K/Jz=\infty$ is exact, we have demonstrated the instability of the pure classical fixed point in at least one direction (which is enough to make it irrelevant). We can therefore conclude that the critical behavior does differ from the classical Ising behavior; randomness, even in the form of quantum fluctuations, represents a relevant perturbation in this system.

To sum up, in this paper we have considered the interplay between quantum mechanics and disorder with an example of randomly mixed classical and quantum-mechanical spins. We verified by explicit construction of the phase diagram that there is a critical concentration for quantal spins and we used the relationship between $K=\infty$ limit and a disordered classical problem to establish the relevance of quantal fluctuations. There are, of course, a number of questions that remain to be answered, both in the framework of our model and beyond. One should certainly perform explicit RG calculations on our systems, both for weak disorder and near percolation, in order to determine the actual critical behavior. From a somewhat broader preceptive, it might also be interesting to look at rather different types of theoretical models with random quantum-mechanical fluctuations, say, for example, a Heisenberg model with spins obeying random commutation relations. Beyond being of theoretical interest, the effects of randomness directly coupled to the size of quantum-mechanical fluctuations are clearly of concern in many experimentally realizable systems, including not only partially deuterated ferroelectrics[19] but also strongly disordered superfluids.[20] One might even expect this kind of randomness to be commonplace for quantal degrees of freedom in glasses.

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