Nonuniqueness of $H^{2/3}$ and H^2 field-temperature transition lines in spin-glasses

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Including the magnetic field dependence on the *superparamagnetic* relaxation time τ, "transition" lines in the H-T plane are obtained for constant τ. These lines follow the relation $T_H - T_0 \propto H^{\nu}$ where $\nu \sim \frac{2}{3}$ except for $H \rightarrow 0$ which shows a crossover to $\nu = 2$. Thus a power law similar to that derived from mean-field models of spin-glasses is obtained, based strictly on a superparamagnetic relaxation-time approach. This questions the conclusion that experimental observations of H-T lines are solely the result of a mean-field phase transition.

Much theoretical work has been devoted to the description of the transition from the paramagnetic to the frozen spin-glass state and to the nature of this frozen state. Originally, the frozen state was described either in terms of a broad spectrum of relaxation time[1] analogous to the Néel picture of superparamagnetism or within the concept of a phase transition based on the Edwards-Anderson (EA) model.[2] The former description has lost much of its appeal because the gradual freezing process predicts larger time (relaxation) effects than experimentally observed, and as a phenomenological model it is not an especially challenging theoretical approach. The latter model, however, resulted in a new type of transition which provided considerable theoretical stimuli. This model has been extended to infinite range by Sherrington and Kirkpatrick (SK)[3] and has required several years of extensive calculations to obtain viable solutions.

de Almeida and Thouless[4] showed that in the presence of a magnetic field, the SK model for classical Ising spins exhibited an instability for purely random interactions which has been subsequently interpreted as a phase transition. Toulouse and Gabay[5] have extended this calculation for an m-component spin system, resulting in the so-called AT line of the following form:

$$1 - T_{AT}(H)/T_{AT}(0) = [(m+1)(m+2)/8]^{1/3} h^{2/3} , \tag{1}$$

where $h = g\mu_B SH/k_B T_{AT}(0)$, T_{AT} is the transition temperature, and m is the spin dimensionality ($m=1$ for the Ising case and $m=3$ for the Heisenberg case). In the course of their calculation, Toulouse and Gabay found another transition line corresponding to the onset of a freezing in the transverse degrees of freedom of the spins. The equation of this transition line (the GT line) is given by

$$1 - \frac{T_{GT}(H)}{T_{GT}(0)} = \frac{h^2}{4} \frac{m^2+4m+2}{(m+2)^2} , \tag{2}$$

where h and m have identical meanings as for Eq. (1). Obviously, in zero field $T_{GT}(0) = T_{AT}(0)$ and the notion of transverse freezing for an Ising system is to be excluded. Consequently, the prediction of these two transition lines in the H-T plane has created a widespread experimental search for such behavior, even though the precise experimental definitions of T_{GT} and T_{AT} are uncertain.[6]

In several recent experimental papers, evidence has been presented supporting the existence of either the AT line ($h^{2/3}$ dependence) or the GT line (h^2 dependence); however, to date, no single experiment has been able to observe both field dependences. The first evidence for the $h^{2/3}$ behavior was from dc magnetization results,[7–10] but the experimental value of the prefactor in Eq. (1) was approximately seven times larger than the theoretical prediction. A similar field dependence was observed in an insulating spin-glass $Eu_{0.4}Sr_{0.6}S$,[11,12] but with a strong dependence upon the time scale t_m of the measurement. Again the observed prefactor was 5–9 times larger. More recently, the onset of strong irreversibilities in torque measurements[13] on CuMn and AuFe have also shown a field-temperature dependence which follows the AT behavior of $h^{2/3}$. Previously, an h^2 behavior has been suggested from the magnetic field dependences of the heat capacity in CuMn,[14] with the coefficient of h^2 being 2.5 smaller than the theoretical value. The apparent success of observing these field dependences has given much support to the validity of the SK model in explaining the spin-glass properties and the reality of a phase transition.

This may be surprising since several calculations[15] on spin-glass models with short-range interactions predict a lower critical dimension of 4, thus no phase transition in the real world of three dimensions. To provide a possible explanation for this apparent success, Kinzel and Binder[16] have shown by Monte Carlo simulations of an Ising-spin nearest-neighbor EA model that several experimental features could be determined without the existence of a phase transition. In particular, irreversible behavior on the time scale t sets in for a magnetic field following an AT line, although no static freezing temperature existed. Simultaneously, Young[17] investigated the temperature and field dependence of the spatial correlations and relaxation times by a similar Monte Carlo simulation of a two-dimensional EA model. His results also showed the following. (i) There is no phase transition in zero field but an average relaxation time τ and correlation range ξ that increase smoothly with temperature. (ii) In a field the energy barrier height ($E = T \ln\tau$) and the correlation length tend to finite values as $T \rightarrow 0$. (iii) Lines of constant τ in the H-T plane seem to vary by a similar power law as the AT line of Eq. (1). Thus the concept of relaxation times reappears in both simulations without the necessity of a phase transition.

In light of these computer simulations, we felt it would be instructive to return to the original superparamagnetic relaxation-time approach of independent thermal activation over energy barriers in order to establish whether "transition" lines similar to those of Eqs. (1) and (2) exist. The effect of a magnetic field on the superparamagnetic relaxation time τ was calculated by Brown[18] in 1963 for a high-energy-barrier approximation as

$$\tau^{-1}=C\alpha^{3/2}(1-h^2)\{(1+h)\exp[-\alpha(1+h)^2] + (1-h)\exp[-\alpha(1-h)^2]\} \quad , \quad (3)$$

where C is a numerical constant, $\alpha=E/k_BT$, $h=\mu H/2E$, E is the energy barrier in zero field, and μ is the superparamagnetic moment. Expanding Eq. (3) to order H^2 and replacing the preexponential factor with a constant for simplicity, one obtains

$$\tau=\tau_0\exp(E/k_BT)[1-\tfrac{1}{2}(\mu H/k_BT)^2+\cdots], \quad \mu H \ll E \ . \quad (4)$$

For a constant τ one can determine an H-T relationship

$$\exp(E/T_0-E/T_H)=1-\tfrac{1}{2}(\mu H/k_BT_H)^2 \ ,$$

where T_0 and T_H are characteristic "freezing" temperatures in zero-field and an applied field, respectively, for an observation time $t_m \sim \tau$. Since $T_H \sim T_0$, an expansion of the exponential results in

$$1-T_H/T_0=\frac{\mu^2}{2Ek_BT_0}H^2 \ .$$

The preceding result was determined in the limit $\mu H \ll E$ and we further wish to find the H-T relationship for constant τ for higher fields.[19] Since a simple analytical expression was not derivable, a numerical calculation of Eq. (3) was performed. The results for a constant τ are plotted as $H^{2/3}$ vs T for Fig. 1, where E is a constant ($=10$ K) for all temperatures. Note the reasonable agreement with an $H^{2/3}$ behavior over a wide range of magnetic fields and the crossover to an H^2 behavior as $H \rightarrow 0$. If a temperature-dependent E is included such as of the form $a+b/T$ suggested by Young's simulations,[17] the results (see Fig. 2) still show an $H^{2/3}$ dependence until the lower fields where again an H^2 dependence predominates. In fact, these latter results appear to be in very good qualitative agreement with Fig. 4 of Young's work.[17] From the slope of the $H^{2/3}$ lines, a coefficient of the $H^{2/3}$ term can be deduced and compared with that of the AT line. For $2<\ln\tau<10$ in Fig. 1, a ratio of 0.31 to 0.14 $(\mu/g\mu_B)^{2/3}$ is determined. For $Eu_{0.4}Sr_{0.6}S$ with strong ferromagnetic clustering,[20] the superparamagnetic moment $\mu \sim g\mu_BSN$ so that $\sim$20–40 Eu^{3+} spins would be correlated in a cluster. This value seems quite reasonable for the above Eu concentration.

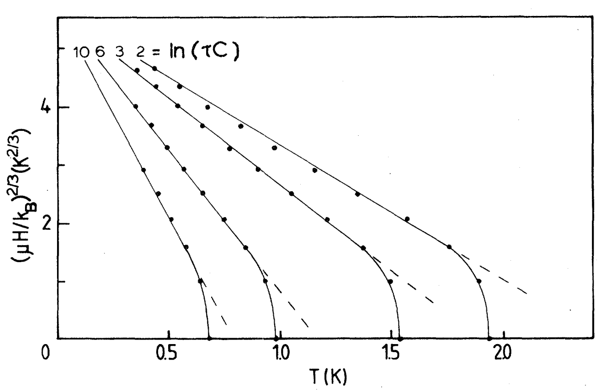

FIG. 1. Points in the H-T plane for constant $\ln\tau$ and a given magnetic field according to Eq. (3) with a constant energy barrier E. The dashed lines show the lower-field extension of a straight-line fit to the $H^{2/3}$ behavior.

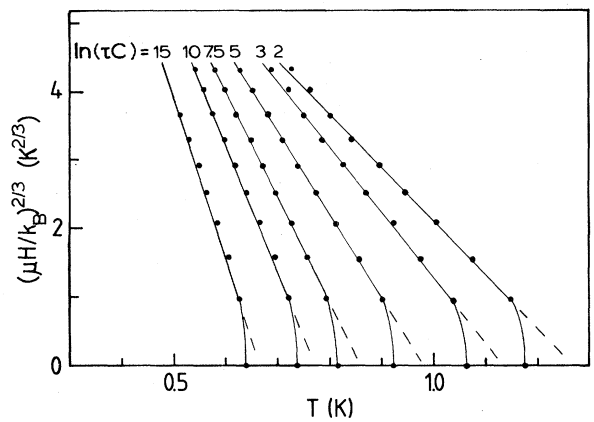

FIG. 2. Points in the H-T plane for constant $\ln\tau$ according to Eq. (3) with an energy barrier $E/k_B=-2+9.5/T$ (see Ref. 17). The dashed lines show the extension of a straight-line fit to the $H^{2/3}$ behavior.

Since most experimental observations show an $H^{2/3}$ transition line, real spin-glasses tend to have a strong Ising-like character. The same $H^{2/3}$ behavior observed in the experiments can be deduced (i) from an Ising (as well as Heisenberg) mean-field approach with a thermodynamic phase transition, (ii) from computer simulations with Ising spins which use the same EA interactions but allow for the dynamics or relaxation of the system (no phase transition), and *now* (iii) from a completely dynamical calculation based on the relaxation of superparamagnetic particles with uniaxial anisotropy which are also Ising-like. In addition, a number of dynamical experiments[11,12] exhibit a low-field crossover exactly as that found from our superparamagnetic model calculation.

Thus we have demonstrated that the H-T dependence determined from the mean-field model of spin-glasses are by no means *unique.* Similar power laws can be determined from the magnetic-field-dependent superparamagnetic relaxation time for a constant τ which can even result in better quantitative agreement with the experimental values, although some arbitrariness is realized in the determination of E and τ. Obviously, a simple relaxation picture without interactions does *not* quantitatively nor qualitatively describe all of the spin-glass freezing properties, nor do we suggest it should. However, relaxation effects and their field dependences, as well as corresponding observation times, are important experimental properties of spin-glasses. Indeed these relaxation effects may be the physical basis for the existence of AT- and GT-type transition lines. Furthermore, careful examination of the experimental data and of possible alternative models is necessitated before asserting that a measurement is more supportive of the mean-field model and a phase transition.

In conclusion, we have shown that an $h^{2/3}$ crossing over to an h^2 field-temperature line can be derived from a purely dynamical (superparamagnetic) treatment without any phase transition. Hence observing such behavior in spin-glasses cannot be used as proof for the validity of the mean-field theory and its corresponding spin-glass phase transition.

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