Spin Glass Model Based on the Onsager Reaction Field

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Changes in the Onsager reaction field are used to account quantitatively for aging (the decrease in the magnetic susceptibility when cooling in zero field is halted below the glass temperature) and rejuvenation (the disappearance of aging phenomena on further cooling only to reappear at T_w on heating) that characterize spin glasses. These effects must be caused by interactions between the spins since, absent the interactions, the magnetic properties of N spins are just N times the magnetic property of a single spin that cannot display aging. A spin introduced at an empty site with a nonzero field becomes polarized, and the polarized spin in turn polarizes its neighbors, thereby changing the local field. This additional field is the Onsager reaction field. Ma's theory for the reaction field in spin glasses [PRB **22**, 4484 (1980)] has been extended to provide a spin-glass model that can account for the experimental data.

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There has been an enormous theoretical effort devoted to spin glasses in the last three decades. Virtually all the work has been devoted to the droplet or the replica symmetry breaking models (for a recent review, see Kawashima and Rieger [1]). Interestingly, comparison with experiment has been minimal, and since 1999, when rejuvenation was discovered neither the droplet nor the replica symmetry breaking model has been able to account for it. Ma's approach does not fit easily into either of these two categories and has not received the same degree of attention, but as will be shown below, it can be used to account for aging and rejuvenation.

Ma's theory will now be summarized (the details can be found in [2]): a random spin distribution interacts via the dipole or RKKY interaction to provide a fluctuating field at lattice sites whose average is zero. The field distribution for a random array of Heisenberg spins has been calculated by Walker and Walstedt [3], and Ma [2], and is

$$P_0(h) \propto (h_g^2 + h^2)^{-2}. \tag{1}$$

While Eq. (1) yields the field at an empty lattice site, the presence of a spin at the site modifies the field because the spin is polarized, and this in turn polarizes its neighbors and thereby alters the field. This is the Onsager reaction field [4]. From Eq. (1) the most probable fields are small fields around zero. Inclusion of the Onsager field changes this, developing a hole or cavity in the probability near zero field.

Ma [2] has calculated the reaction field and its effect on the decay of a magnetization produced by cooling in a small magnetic field. Ma's approach can be summarized as follows: in addition to the single spins there are ''nuclei'' of two or more spins that consist of spins that are strongly coupled because they are very close to each other. The concentrations of nuclei can be considerable: the concentration of single spins that have no nearest neighbors is $c_1 = c(1-c)^r$, where c is the concentration of magnetic material and r is the number of nearest neighbors; so the fraction of the magnetic material that is in clusters of two or more spins is $1-(1-c)^r$. For instance, for a CuMn alloy with 10% Mn, only 28% of the Mn are spins with no neighbors, and 72% of the Mn are in multispin nuclei.

The nuclei have a net spin and an anisotropy energy that separates an equilibrium state in a magnetic field from a metastable equilibrium with the nucleus reversed. It should be noted that (Ma [2] and Nozieres [5]) the anisotropy arises from the interaction of the coupled spins with their neighbors. Thus there are two fields to consider: a mean field, h, from all the other spins in the sample, and an anisotropy field, h^*, that must be overcome for the nucleus to reverse.

Ma uses a pair to estimate the properties of the excited states of the nucleus whose Hamiltonian is $H_{12} = -J\mathbf{s}_1 \cdot \mathbf{s}_2 - \mathbf{h}\cdot(\mathbf{s}_1+\mathbf{s}_2) - \mathbf{h}^*\cdot(\mathbf{s}_1-\mathbf{s}_2)$. For a strongly coupled pair there will be singlet and triplet states. Ignoring the singlet states, for $J>0$, the energy of the ground state in zero field is $-h^{*2}/J$. In a field, $h<h^{*2}/J$, it is $-h^{*2}/J - Jh^2\sin^2\phi/h^{*2}$. The anisotropy energy is h^{*2}/J, and because $\langle \mathbf{s}^*\rangle = \langle \mathbf{s}_1 - \mathbf{s}_2\rangle = 2\mathbf{h}^*/J$ is frozen along $\mathbf{h}^*$ it is related to the barrier height Λ (the precise value of Λ depends on the behavior of the neighbors), the average spin of the pair is $\langle \mathbf{s}\rangle = 2J[\mathbf{h} - (\mathbf{h}^*\cdot\mathbf{h})\mathbf{h}^*/h^{*2}]/h^{*2} \approx J\mathbf{h}/h^{*2} = \mathbf{h}/\Lambda$. Ma then extends the analysis to include strongly and ferromagnetically coupled trios and larger clusters, and derives a distribution,

$$\rho(\Lambda) \approx 2\alpha\sqrt{\alpha\Lambda/\pi}e^{-\alpha\Lambda}, \tag{2}$$

where α is a constant, proportional to the concentration of magnetic material.

During cooling, starting from a temperature high enough for all the nuclei to be superparamagnetic they will initially be able to maintain their equilibrium distribution. Eventually, however, the relaxation rate becomes too slow for this to be possible, and the grains become blocked. The temperature at which this occurs, the blocking tem-

perature, can be defined as that temperature below which a nucleus flips its magnetization no more than once before a measurement has been completed. The rate at which the spin of the nucleus flips is $\tau^{-1} = \omega e^{-(\Lambda/T)}$, where ω is an attempt frequency. At the end of the cooling process, if the rate of cooling is $\frac{dT}{dt} = \gamma$, $\gamma\tau^{-1}dt$ flips will occur in time dt, and the condition becomes $1 = \int_T^{T_b} dT\gamma^{-1}\omega e^{-(\Lambda_b/T)} = \omega\gamma^{-1}[T_b E_2(\frac{\Lambda_b}{T_b}) - TE_2(\frac{\Lambda_b}{T})]$, where T_b is the blocking temperature, and $E_2(x)$ is the exponential integral function of order 2. Using the asymptotic expansion of E_2 [6], $\frac{\omega}{\gamma} \times (\frac{T_b^2}{\Lambda_b} e^{-(\Lambda_b/T_b)} - \frac{T^2}{\Lambda_b} e^{-(\Lambda_b/T)}) \cong 1$, neglecting the second term for $T \ll T_b$, $\frac{\Lambda_b}{T_b} + \ln\frac{\Lambda_b}{T_b} \cong \ln\frac{\omega}{\gamma} T_b$, ω is between about 10^9 and 10^{13} Hz; so the value of T_b does not have much effect on the right-hand side. If the time taken to cool the sample from T_b to a low temperature is ~1000 s, $\frac{\Lambda_b}{T_b} + \ln\frac{\Lambda_b}{T_b}$~23, and $\frac{\Lambda}{T_b(\Lambda)} = b$ is roughly constant with a value lying between about 24 and 34, depending on ω, and the time taken to cool.

Using an ingenious approach Ma then calculates the contribution of the blocked nuclei to the reaction field, with the field distribution becoming $P(h) = P_0(h)C(h)$, where $C(h)$ is the cavity factor $= \exp[h^2/2\Lambda T_b(\Lambda) - \Lambda/2T_b(\Lambda)]$. Ma assumes that all nuclei block at T_g. This is clearly a drastic oversimplification and will not be made here. Furthermore, on heating the blocking temperatures of the nuclei that have relaxed are replaced by the sample temperature, and the cavity factor expression becomes:

$$C(h,T) = \exp\left[\frac{h^2}{2\Lambda T_b(\Lambda)} - \frac{\Lambda}{2T_b(\Lambda)}\right]\exp\left[-\omega t\exp\left(-\frac{\Lambda}{T}\right)\right] + \exp\left[\frac{h^2}{2\Lambda T} - \frac{\Lambda}{2T}\right]\left\{1 - \exp\left[-\omega t\exp\left(-\frac{\Lambda}{T}\right)\right]\right\}. \quad (3)$$

If the sample has been cooled to a low temperature in zero field, and a small field, h_a, is applied, the cavity factor becomes $C(h - h_a, T)$ and the sample acquires a moment that increases as it is heated, and the nuclei unblock. If it has been heated to a temperature T for a time t, the unblocked nuclei and the n neighbors will acquire a moment. The time dependent moment acquired after zero field cooling to a temperature T_0, and heating to T for a time t, becomes

$$M \approx a\frac{h_a}{T}\int_{\Lambda_b(T_0)}^{\Lambda(T_g)} d\Lambda\rho(\Lambda)(1 - e^{-\omega te^{-(\Lambda/T)}}) \times \int_0^{\Lambda} h^2\left(\frac{h^2}{\Lambda^2} + \frac{n}{4}\right)dhP_0(h)C(h,T), \quad (4)$$

where a is a constant, and T_g is the temperature of the maximum in the magnetization acquired on heating in h_a. The integral over h cuts off at Λ because if h is larger than Λ the moment of the cluster will reverse immediately. The double exponential changes *very* rapidly with $\frac{\Lambda}{T}$ and a good approximation, that is often used [2], is that $(1 - e^{-\omega te^{-(\Lambda/T)}}) \cong 1$ if $\frac{\Lambda}{T} \leq \ln\omega t$ and 0 otherwise (but note that rejuvenation takes place over a small temperature range; so this approximation cannot be used when discussing it), with Eq. (1)

$$M \approx a\frac{h_a}{T}\int_0^{T\ln\omega t} d\Lambda\rho(\Lambda)H(\Lambda), \quad (5)$$

where $H(\Lambda) = \int_0^{\Lambda} h^2(\frac{h^2}{\Lambda^2} + \frac{n}{4})dh(h_g^2 + h^2)^{-2} \times e^{[h^2/2\Lambda T_b(\Lambda)] - [\Lambda/2T_b(\Lambda)]}$.

If M is measured while heating, t becomes an effective time at the measurement temperature. Using arguments identical to those in the blocking temperature discussion $t \approx \frac{T}{\beta}$, where β is the heating rate.

The local field distribution has been measured by de Vegvar and Fulton [8] in CuMn, who obtain $P(h) = (4\sqrt{w}/\pi)(h/h_g)^2[w + (h/h_g)^2]^{-2}$, where $h_g = kT_g/\mu$, with k Boltzman's constant, and μ the Mn magnetic moment. w is an adjustable width; de Vegvar and Fulton choose $w = 2$ in order to fit their data. h_g can be very large, in de Vegvar and Fulton's samples, with 0.1% Mn, T_g was 1.8 K, and h_g was 0.84 T. For the materials considered here T_g is an order of magnitude greater, and h_g is much larger than any anisotropy fields; so h can be neglected in comparison with h_g, and the integral over h becomes

$$H(\Lambda) \cong (4\sqrt{w}/\pi)h_g^{-2} \times \int_0^{\Lambda} h^2\left(\frac{h^2}{\Lambda^2} + \frac{n}{4}\right)dhe^{[h^2/2\Lambda T_b(\Lambda)] - [\Lambda/2T_b(\Lambda)]}, \quad (6)$$

with $\Lambda/T_b(\Lambda) = b \approx$ constant, and $h/\Lambda = x$, $H(\Lambda) \cong (4\sqrt{w}/\pi)h_g^{-2}\Lambda^3 e^{-(b/2)}\int_0^1 x^2(x^2 + \frac{n}{4})dxe^{(b/2)(x^2-1)}$. The integrals are integral representations of the confluent hypergeometric function [6], and $H(\Lambda) \cong (4\sqrt{w}/\pi)h_g^{-2}\Lambda^3\frac{e^{-(b/2)}}{2}[\frac{\Gamma(1.5)}{\Gamma(2.5)\Gamma(1)}M(2.5, 1.5; \frac{b}{2}) + \frac{n}{4}\frac{\Gamma(0.5)}{\Gamma(1.5)\Gamma(1)} \times M(1.5, 0.5; \frac{b}{2})]$. The recurrence relations for these functions [7] can be employed to obtain $H(\Lambda) \cong (4\sqrt{w}/\pi)h_g^{-2}\Lambda^3\frac{b}{8}$.

With Eq. (2), the magnetization becomes

$$M \approx A\frac{h_a}{T}\int_{\Lambda_b(T_0)}^{\Lambda_b(T)} d\Lambda\Lambda^{7/2}e^{-\alpha\Lambda} = A\frac{h_a}{T}\int_{bT_0}^{T\ln\omega\frac{T}{\beta}} d\Lambda\Lambda^{7/2}e^{-\alpha\Lambda}, \quad (7)$$

where A is a constant, and T_0 is the temperature at which h_a is turned on and heating started. Performing the integration we obtain

$$M \approx A\alpha^{3/2}\frac{h_a}{T}\left[\gamma\left(\frac{9}{2}, \alpha T \ln\frac{\omega T}{\beta}\right) - \gamma\left(\frac{9}{2}, \alpha b T_0\right)\right] \quad \text{for } T < T_g,$$
$$M \approx A\alpha^{3/2}\frac{h_a}{T}\left[\gamma\left(\frac{9}{2}, \alpha T_g \ln\frac{\omega T_g}{\beta}\right) - \gamma\left(\frac{9}{2}, \alpha b T_0\right)\right] \quad \text{for } T > T_g, \tag{8}$$

where $\gamma(n, x)$ is the incomplete γ function. Equation (7) is compared with results obtained by Kenning *et al.* [9] in Fig. 1.

If cooling is halted for a time t_w at a temperature T_w the distributions of grains with blocking temperatures between T_w and $T_w b^{-1}\ln\omega t_w$ will be reset to the equilibrium distribution corresponding to T_w, and Eq. (4) becomes, with $C_w(h, T)$ equal to Eq. (3) with T_b replaced by T_w

$$M_w = a\frac{h_a}{T}\int_{\Lambda_b(T_0)}^{\Lambda_b(T_w)} d\Lambda\rho(\Lambda)(1 - e^{-\omega t e^{-(\Lambda/T)}})\int_0^{\Lambda} h^2 dh P_0(h)\left(\frac{h^2}{\Lambda^2} + \frac{n}{4}\right)C(h, T) + a\frac{h_a}{T}\int_{\Lambda_b(T_w)}^{T_w \ln\omega t_w} d\Lambda\rho(\Lambda)(1 - e^{-\omega t e^{-(\Lambda/T)}})$$
$$\times \int_0^{\Lambda} h^2 dh P_0(h)\left(\frac{h^2}{\Lambda^2} + \frac{n}{4}\right)C_w(h, T) + a\frac{h_a}{T}\int_{T_w \ln\omega t_w}^{\Lambda_b(T_g)} d\Lambda\rho(\Lambda)(1 - e^{-\omega t e^{-(\Lambda/T)}})\int_0^{\Lambda} h^2 dh P_0(h)\left(\frac{h^2}{\Lambda^2} + \frac{n}{4}\right)C(h, T) \tag{9}$$

When M_w is subtracted from M, the first and third integrals in Eq. (9) cancel, and the moment is decreased by $\Delta M = a\frac{h_a}{T}\int_{\Lambda_b(T_w)}^{T_w \ln\omega t_w} d\Lambda\rho(\Lambda)(1 - e^{-\omega t e^{-(\Lambda/T)}})H_w$, where $H_w = \int_0^{\Lambda} h^2 dh P_0(h)(e^{[h^2/2\Lambda T_b(\Lambda)] - [\Lambda/2T_b(\Lambda)]} - e^{(h^2/2\Lambda T_w) - (\Lambda/2T_w)})e^{-\omega t e^{-(\Lambda/T)}}$. Following the previous procedure we obtain $H_w \approx \frac{\Lambda - bT_w}{8T_w} e^{-\omega t e^{-(\Lambda/T)}} \frac{4}{\pi}\sqrt{w}\frac{\Lambda^3}{h_g^2}$, and

$$\Delta M \approx A\frac{h_a}{T}\int_{bT_w}^{T_w \ln\omega t_w}\left(\frac{\Lambda}{bT_w} - 1\right)\Lambda^{(7/2)} e^{-\alpha\Lambda}(1 - e^{-\omega t e^{-(\Lambda/T)}})e^{-\omega t e^{-(\Lambda/T)}}. \tag{10}$$

Integrating by parts, $\Delta M \approx A\alpha^{3/2}\frac{h_a}{T}\left[\frac{1}{\alpha b T_w}\gamma\left(\frac{11}{2}, x\right) - \gamma\left(\frac{9}{2}, x\right)\right](1 - e^{-\omega t e^{-(x/T)}})e^{-\omega t e^{-(x/T)}}\Big|_{\alpha b T_w}^{\alpha T_w \ln\omega t_w} - A\alpha^{3/2}\frac{h_a}{T} \times \int_{\alpha b T_w}^{\alpha T_w \ln\omega t_w}\left[\frac{1}{\alpha b T_w}\gamma\left(\frac{11}{2}, x\right) - \gamma\left(\frac{9}{2}, x\right)\right]\frac{\partial}{\partial x}\left[(1 - e^{-\omega t e^{-(x/\alpha T)}}) \times e^{-\omega t e^{-(x/\alpha T)}}\right]$. Numerical evaluation reveals that the remaining integral makes a negligible contribution, and

$$\Delta M \approx A\alpha^{3/2}\frac{h_a}{T}\left[g\left(1 - e^{-\omega t e^{-(T_w \ln\omega t_w/T)}}\right) \times e^{-\omega t e^{-(T_w \ln\omega t_w/T)} - (1 - e^{-\omega t e^{-(bT_w/T)}})} e^{-\omega t e^{-(bT_w/T)}}\right], \tag{11}$$

where $g = \frac{\frac{1}{\alpha b T_w}\gamma(\frac{11}{2}, \alpha T_w \ln\omega t_w) - \gamma(\frac{9}{2}, \alpha T_w \ln\omega t_w)}{\frac{1}{\alpha b T_w}\gamma(\frac{11}{2}, \alpha b T_w) - \gamma(\frac{9}{2}, \alpha b T_w)}$. Figure 2 compares Eq. (11) with experimental results published by Mathieu *et al.* [10]. The value of g was calculated approximately to be 0.1, and ω was set at 10^{12} Hz. Mathieu *et al.* [10] remark that it is important to make the cooling and heating rates equal, i.e., $b = \ln\omega t$; however, if this was done the minimum in the moment difference was ~1 K off; in order for it to occur at T_w it was necessary to make the cooling rate 1.2% slower than the heating rate. Such a sensitivity to the difference in the two rates could be a subject of future research.

Assemblies of magnetic nanoparticles can also display the spin-glass effects of aging and rejuvenation [11–13] and have been called "superspin glasses". The analysis presented here can also be applied to them, and this is underway at present. However, there is an important dif-

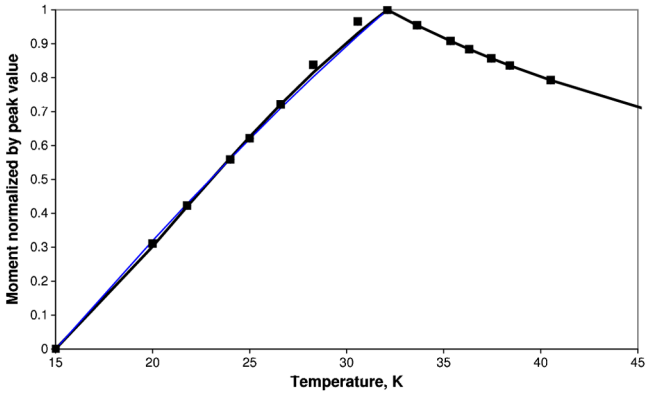

FIG. 1 (color online). ■ mark the magnetization in a 50 G field for $Cu_{0.94}Mn_{0.06}$ taken from measurements by Kenning *et al.* [9], compared with Eq. (8).

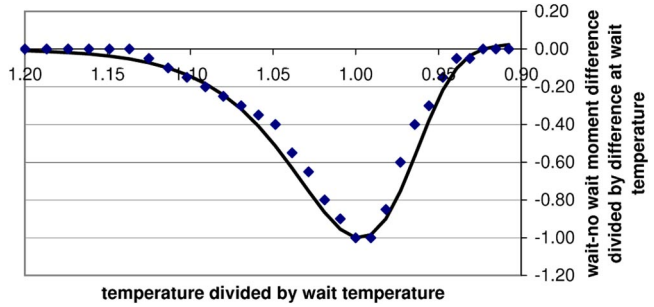

FIG. 2 (color online). ■ mark the difference between the magnetization measured on heating a sample of Ag-11% Mn after cooling in zero field and that for which a wait of 9000 s took place at 27 K plotted against the measurement temperature divided by 27 K. From Mathieu *et al.* [10]. The line was calculated using Eq. (11).

ference: whereas Ma's nuclei and neighbors form the clusters with anisotropy barriers that are responsible for the spin-glass phenomena, in the other case the clusters are the particles themselves. The energy barrier for nanomagnets is proportional to their volume; so the barrier height distribution for nanoparticles depends on their size distribution which has a maximum of many nanometers and depends on how the particles were produced. The nanoparticle size distribution leads to a much broader minimum in the plot of moment difference against temperature for the rejuvenation data.

Finally, while Ma's calculation [2] was specifically for Heisenberg spins, a similar approach yields similar results for Ising spins, and was, in fact, used by Cyrot [14] in the first application of the Onsager theory to spin glasses.

In conclusion, a theory has been described that uses the Onsager reaction field to account for aging and rejuvenation effects in spin glasses. The Onsager reaction field is produced by blocked clusters in metastable states. The number of clusters in metastable states is frozen in at their blocking temperatures during cooling. If the cooling is interrupted for a time at an intermediate temperature T_w a small number of clusters frozen during cooling to T_w can relax. When cooling is resumed their blocking temperature will now be T_w, and the altered distribution will lead to an altered reaction field and susceptibility. However, this change is confined to the clusters blocked during cooling to T_w that have been able to relax during the waiting time, and this corresponds to a narrow temperature interval around T_w; at temperatures below and above this interval the susceptibility is not affected.

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