Demagnetization of spin systems at low temperature

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We report on analytical and numerical results for the time evolution of a (lattice) model of ferromagnetic particles after field inversion. Relaxation from the metastable phase occurs by superposition of two independent spin-flip processes: with probability $1-p$, the flip obeys the Metropolis rule at finite temperature, T, while the flip is performed at random, as for $T\rightarrow\infty$, with probability p. For small p, the latter process mimics quantum tunneling or any other sort of *impurity* that would induce both additional randomness and a nonequilibrium steady-state condition asymptotically. Critical *avalanches* and constant magnetic viscosity at low T ensue as two key features of the relaxation for any $p\neq 0$. This has some implications for experiments on demagnetization and mesoscopic quantum coherence. [S0163-1829(97)08437-3]

Many phenomena in nature, including some that bear technological relevance, concern a system that relaxes from a metastable state.[1–3] Therefore, investigating universal features of this process is interesting. Some important, interrelated questions are the physical origin for the lack of any length scale that has been observed under certain conditions,[4] and how scale invariance develops during time relaxation far from standard tunable critical points. The observation that complex systems may organize themselves into a *minimally stable* nonequilibrium state[5] has attracted much attention; cf. Refs. 6 and 7, for instance, and references therein. The system is then very sensitive to perturbations, and avalanches of all sizes occur that fit a power law. It seems that the state has spontaneously became critical, and remarkably correlated, which reflects on "$1/f$ noise." Indeed, fluctuations with power spectrum $S(\Delta)\sim\Delta^{-\alpha}$, $\alpha>0$, i.e., more correlated than white noise, have been reported, e.g., the *excess* low-frequency noise in electrical conductors under a bias, acoustic emission during martensitic transformations, gravity-driven motion of sand and rice piles, traffic jams, and migration phenomena.[8–12] However, the consequences and range of validity of this picture are not yet well enough understood; cf. Ref. 13 and references therein.

Studies of such questions in magnets are scarce (see, however, Ref. 14) even though these systems are more familiar to physicists and suited for experimental and theoretical analysis than, e.g., granular media. Consider demagnetization, which has a great practical interest.[15–18] It has been intensively studied looking for quantum phenomena at the mesoscopic scale. That is, the electronic spins in certain magnetic particles (small ferromagnetic domains, antiferromagnetic protein cages, Mn_{12} acetate molecules$\cdots$) tend to behave coherently at low temperature, as locked together into an ordered state, e.g., predominantly aligned for ferromagnetic materials. The resulting magnetic vector can rotate, with two (or more) low-energy directions. Therefore, quantum tunnelling (QT) may occur between the minima,[19,20] which is expected to influence the magnetization $m(t)$ of an appropriate ensemble of fine particles (while it relaxes after the field, h, is either suppressed or inverted). Experiments on different materials confirmed the prediction[21,1] that $m(t)\approx m(t_0)-\nu(h,T)\ln(t/t_0)$ within a wide time range, where the *magnetic viscosity* $\nu(h,T)$ is observed to become independent of temperature for $T<T_Q$, typically a few kelvins. Together with other facts, this is taken as evidence that *all* the spins within the particle behave coherently, which is known as *quantum coherence*; as a matter of fact, agreement between theory and experiments (cf. Refs. 20, 22, 23), together with some technical objections (e.g., Refs. 24–30, and references therein) have been reported.

We have studied demagnetization in a microscopic magnetic model. While our results are consistent with the belief that QT has been observed in the laboratory, they suggest that observations might be affected by microscopic details, namely, further sources of randomness not considered by existing theory. On the other hand, our model exhibits avalanches and $\Delta^{-\alpha}$ ($\alpha>0$) noise, which suggests that time-scale invariance might characterize a wide range of magnetic relaxation phenomena in nature. The model and its possible laboratory implementations are simple enough to help testing and developing further theory.

Consider binary spins, $\sigma_i=\pm 1$, at lattice sites $i=1,\ldots,N$, with the Ising energy, $H(\{\sigma_i\})=-J\Sigma'_{ij}\sigma_i\sigma_j+h\Sigma_i\sigma_i$, where the first sum is over nearest-neighbor pairs (J and Boltzmann's constant are set to unity hereafter). In addition to mean-field results for $N\rightarrow\infty$, we report here (cf. Ref. 31 for further details) on computer simulations for the square lattice of side L and for a circle of radius 30, both with free boundaries. One may endow this system (named *cluster* hereafter) with different interpretations, as discussed below in detail. The *cluster* is initially ordered, i.e., $\sigma_i=+1\ \forall i$, with small h (typically $|h|=0.1$) pointing in the other direction. Time evolution from this metastable state proceeds according to competing dynamics. That is, we consider flipping the spin i (selected at random) according to the Metropolis rule at infinite or finite temperature with probability p or $1-p$, respectively. In other words, the probability per unit time of flipping σ_i is $c(\{\sigma_i\},i)=p+(1-p)\min\{1,\exp(-\Delta H/T)\}$ for *cluster* configuration $\{\sigma_i\}$, where ΔH is the change of energy brought about by the attempted flip. Implementing here the kinetic Ising model with such competing dynamics has a definite motivation. First, such dynamics impedes detailed balance so that, in particular, the system cannot be at equi-

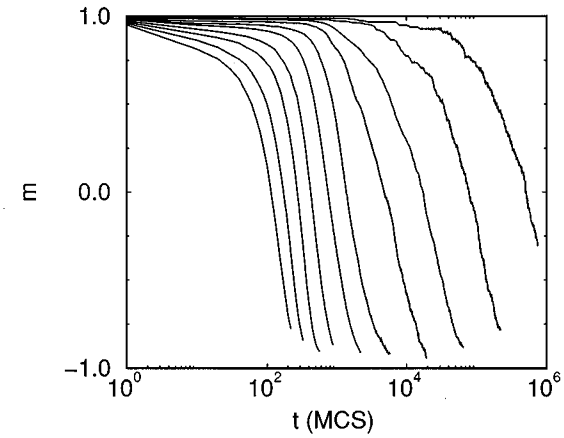

FIG. 1. Magnetization vs ln t (t in units of MC steps per lattice site) for different temperature, $0.4<T<0.9$ (in units of $T_O=2.2691$; k_B, $J=1$), decreasing from left to right, as obtained in computer simulations of an ensemble of 1500 independent 32×32 clusters. (The final part of the evolutions, where statistics are poor, is not shown here.)

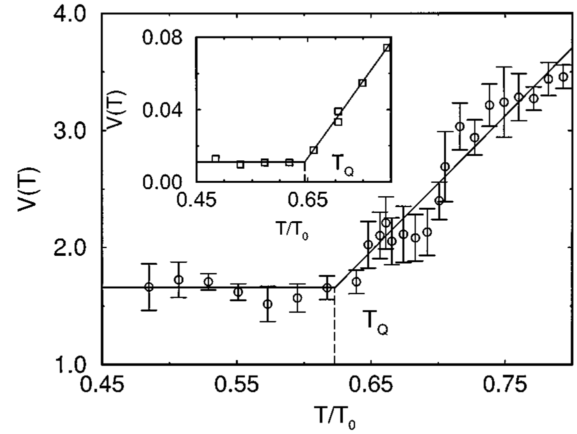

FIG. 2. Magnetic viscosity (arbitrary units) as a function of T for the late (main graph) and early (inset) regimes illustrated in Fig. 1. Lines are a guide to the eye.

librium in general, even if it asymptotically reaches a steady state; cf. Ref. 33. This is important because it is likely that both temporal criticality and constant magnetic susceptibility—as described, respectively, in the two first paragraphs above—can only occur under nonequilibrium conditions. On the other hand, the infinite-T process, occurring with probability p, is intended as an oversimplified representation of that sort of *impure* (dynamic) behavior which is an intrinsic property of all natural systems; it can mimic quantum tunnelling, for example, as argued below, but one may think of it as a more general source of randomness or disorder in the system. Our data here are for $p=10^{-3}$, 10^{-6}, and $0.1T_O<T<0.9T_O$, where $T_O=2.2691$ is the Onsager critical temperature. That is, only small disturbances from the familiar thermal relaxation process are considered. Evolution is followed until the minimum (negative) value of the magnetization, $m=N^{-1}\Sigma_i\sigma_i$, corresponding to the stable phase for given values of T and h, is reached. We typically average data over $\mathcal{N}=1500$ independent computer runs. Varying $|h|$, $\mathcal{N}$, and the shape of the *cluster* does not seem to modify our qualitative conclusions below.[31]

After averaging for sufficiently large $\mathcal{N}$, the relaxation exhibits two well-defined main regimes, as illustrated in Fig. 1. First (top of the graph), the initial condition, $m=1$, decreases rather linearly towards $m_T>0$. This *early regime* (under small negative field) does not seem to differ essentially from the relaxation when a large, saturating field is suppressed in experiments. In our case, however, m_T is metastable and, therefore, one observes next an abrupt decay towards the state of negative magnetization. This *late regime* begins later the smaller either T or p is, the influence of p being more dramatic. One may prove[34] that the early, metastable regime lasts (until a critical cluster sets in) a time $\sim\exp(aJ^2|h|^{-1})$ with a of order of unity for $p=0$ at sufficiently low temperature; this is of the same order of magnitude in our simulations with $p>0$. Figure 2 shows the temperature dependence of the viscosities $\nu_{1(2)}(h,T)$, defined as the slope of m versus ln t during the early (late) regime. As in many different experiments (e.g., Refs. 22 and 23), our estimates for both ν_1 and ν_2 decrease linearly with T until they become constant for $T<T_Q$. (The fact that ν_2 is noisier than ν_1 reflects larger error bars in our linear fits to the late regime.) Figure 2 suggests that temperature-independent behavior at low T is an intrinsic property of time relaxation. In fact, our data collapse into a scaling function F, according to $m(t)=m_T-\nu_T F(t/\tau_T)$, where τ_T is a characteristic relaxation time [which we have defined as $m(\tau_T)=0$], and one needs to use two different sets for the parameter ν_T corresponding to early and late regimes, respectively. The crossover temperature T_Q may be defined empirically as in Fig. 2. We have observed that it slowly decreases with p, e.g., $T_Q/T_O\approx 0.6$, 0.5 for $p=10^{-3}$, 10^{-6}, respectively, and we have checked that $T_Q=0$ for $p=0$. (No essential dependence on N was detected.) The observation that T_Q has a slow dependence on p [which is not consistent with the simple estimate $T_Q\simeq -8/\ln p$ for the early regime, obtained after comparing p and the Metropolis probability $\exp(-8/T)$ for $h=0$] is an interesting fact that deserves further study.

Our observations admit a simple interpretation if the *cluster* is assumed to represent the set of independent fine particles in actual experiments (averaging over computer runs then corresponds to the ensemble average in statistical physics). The fact that this interpretation is justified can be made explicit by considering the *cluster* relaxing with the effective rate $c(\{\sigma_i\},i)=p+(1-p)(1-\sigma_i\gamma_i)(1-\sigma_i\gamma_h)$, where $\gamma_h=\tanh(\beta h)$, and $\gamma_i=\tanh(\beta e_i)$ with $e_i=\Sigma_{j_i=1}^{q}\sigma_{j_i}$ (with the sum over q neighbors of i). This shows that the neighborhood of any given spin is weighted by e_i for dynamical purposes, so that e_i formally plays here the role of familiar energy barriers in experiments.[29] Under a mean-field approximation,[32] this case leads to $\partial m/\partial t=-\partial V(m)/\partial m$ with the potential function as given in Fig. 3. This illustrates the effect of parameter p measuring (in this interpretation) the probability of QT relative to thermal flipping of the particle spin. Figure 3 also illustrates the T dependence of the *instant* magnetic viscosity $\nu_\tau\equiv-[\partial m/\partial(\ln t)]_{t=\tau}$ (τ corresponds in Fig. 3 to an arbitrary time during the early regime). Therefore, under the present interpretation, a principal conclusion is that mesoscopic QT should indeed be observable

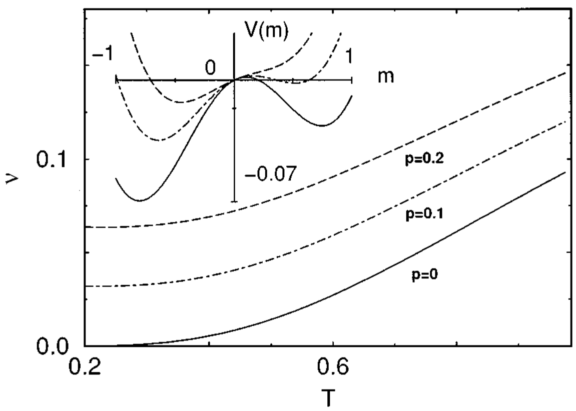

FIG. 3. Typical T dependence of the (instant) magnetic viscosity in the mean-field model for $h=-0.1$, and varying p, as indicated. The inset shows corresponding potential functions $V(m)$ for $T=0.75$ (in units of the associated critical temperature).

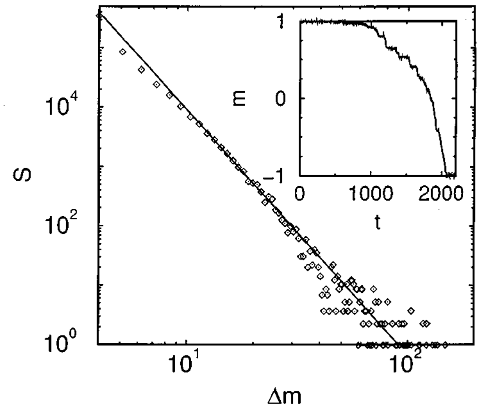

FIG. 4. Log-log plot of the distribution of avalanche sizes in simulations of an ensemble of circular clusters of radius 30, for $p=10^{-6}$ and $T=0.11T_O$; the solid line has slope -3. The inset illustrates $m(t)$, for the same circular cluster with $p=10^{-3}$ and $T=0.6T_O$, in a typical individual history, in which the magnetization steps are evident to the naked eye. (Note that the graphs in Fig. 1 correspond to an average of many, $\mathcal{N}$, individual histories.)

in the laboratory under proper experimental conditions.[23,29]

The same results may be endowed with a different, perhaps less realistic interpretation. That is, the *cluster* may be viewed as an Ising magnet (instead of the set of ''independent'' particles in the previous interpretation) relaxing from a metastable state under superposition of a thermal process and a random one that represents any sort of defect or disorder, e.g., diffusing impurities that would influence the system relaxation.[33] No similarity with the above-mentioned experiments exists in this case, but our simulation would still have some implications on them. That is, it seems to be implied under this interpretation that any disorder, including the possibility of *microscopic* QT occurring very rarely within the particles of experimental interest (or admitting the possibility that, eventually, not all the spins within the particle act coherently), would be enough to cause T independence of ν at low T. In fact, the mean-field model gives $\nu_\tau(T=0)=p\tau m(\tau)$, i.e., one should observe that $\nu_\tau(T)$ goes to a nonzero value for any (even small) $p>0$ for appropriate values of the observation scale τ.

In order to illustrate another interesting feature of the model for $p>0$, the inset in Fig. 4 shows a typical relaxation of an individual *cluster*. This graph (the same behavior is exhibited in the averaged evolutions of Fig. 1, though less evidently in general to the naked eye) clearly reveals discreteness of time evolution which occurs in fact by jumps of the magnetization or avalanches of ''all'' sizes. More precisely, close inspection suggests that such behavior occurs during the late regime only. We mention two possible interpretations of this fact. One is that the system is not *critical* until sufficiently disordered, namely, until a fraction of individual spins or small clusters of spins have already been inverted. The situation would be similar to the one reported for random-field Ising models at $T=0$, in which avalanches occur which are assumed to indicate the existence of a critical condition with both disorder and driving force as tunable parameters.[35] Another possibility is that the cluster of inverted spins is rather compact but exhibits a complex surface whose growth is essentially modified by the random small perturbation. Concluding more definitely about this matter requires more specific effort, e.g., as initiated recently in Ref. 36.

Let Δ_t be the number of consecutive Monte Carlo (MC) steps, each consisting of N attempted flips, elapsed until a minimum variation of m is measured (we remark that m is only evaluated after each MC step); the size of an avalanche is defined as the corresponding jump, $\Delta_m \equiv |m(t)-m(t+\Delta_t)|$. As illustrated in Fig. 4, the distribution of both Δ_t and Δ_m exhibit a power law, e.g., $S(\Delta_m)\sim\Delta_m^{-\alpha}$ (while there is no indication from our analysis that the data can be fitted to a stretched exponential). We have measured $\alpha\approx 6$, 4, 3 for $N\simeq 100$, 1000, 3000, respectively. (Note that $\alpha=0$ for the equilibrium case $p=0$.) That is, although statistical errors prevent us from giving a definite quantitative conclusion (collecting the amount of data needed for that purpose is far beyond our aim here), a rapid tendency towards $1/f$ noise as the *cluster* becomes sufficiently large is clear. (In fact, these preliminary data are roughly consistent with expected finite-size scaling behavior[37,31].) No doubt it would be interesting to investigate experimentally this matter in actual magnets.

Summing up, we have presented an oversimplified model of particle demagnetization that conveniently simulates various interesting processes. The possibility of QT is ideally modeled by means of competing dynamics which causes a nonequilibrium condition. Both analytical and numerical results suggest that macroscopic/mesoscopic QT is experimentally observable, though perhaps some of its effects are in practice added to or obscured by QT of individual spins and/or other possible sorts of microscopic impure behavior. Anyhow, such dynamic disorder produces avalanches in the model during the time relaxation that appear to agree with expectations from self-organized criticality arguments (see, however, Ref. 35). We hope the behavior of this simple model motivates further experimental studies of demagnetization in appropriate substances.

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