Impure Ferromagnetic Nanoparticles: Scale Free Avalanches during Decay from Metastable States

Pablo I. Hurtado*, J. Marro* and Pedro L. Garrido*

*Institute 'Carlos I' for Theoretical and Computational Physics, and Departamento de Electromagnetismo y Física de la Materia, University of Granada, 18071, Granada, Spain.

Abstract.

Metastability is observed in fluids, solids, plasmas and other systems, and it often determines their behavior. A better microscopic understanding of this ubiquitous natural phenomenon, which is mathematically challenging,[1] is therefore of great practical and theoretical interest. In particular, metastability is relevant to the behavior of magnetic storage devices. Here one needs in practice to create and to control fine grains, i.e., magnetic particles whose size ranges from mesoscopic to atomic levels, namely, clusters of 10^4 to 10^2 spins, and even smaller ones. The underlying physics is much less understood than for bulk properties. In particular, one cannot assume that such particles are neither *infinite* nor *pure.* That is, they have free borders, which results in a large surface/volume ratio inducing strong surface effects, and impurities. The *microscopic* nature of the latter, which shows up in actual specimens as spin, bond and/or lattice disorder and other inhomogeneities, quantum tunneling,[2] etc., suggests they might dominate the behavior of near−microscopic particles; in fact, they are known to influence even macroscopic systems. An interesting issue is therefore understanding the formation of a new phase inside a metastable cluster which is finite, small and contains impurities.

Following recent efforts, we study in this paper the simplest possible model of this situation, namely a $2d$ Ising ferromagnet with free boundaries that we endow with a weak dynamic perturbation competing with the thermal spin-flip process, which impedes equilibrium. Consider then the Hamiltonian function $\mathscr{H}(\vec{s}) = -J\sum_{\langle i,j\rangle} s_i s_j - h\sum_{i=1}^{N} s_i$, where $J > 0$ (ferromagnetic interactions), $s_i = \pm 1$ stands for the two possible states of the spin at site i of the square lattice, $i = 1, \ldots, N$, and the first sum is over any pair $\langle i, j\rangle$ of nearest−neighbor sites. The system configuration, $\vec{s} \equiv \{s_i\}$, is let to evolve in time due to superposition of two canonical drives. That is, we chose the transition probability per unit time for a change of $\vec{s}$ to be

$$\omega(\vec{s} \to \vec{s}^{\,i}) = p + (1-p)\frac{\mathrm{e}^{-\frac{1}{T}\Delta\mathscr{H}_i}}{1+\mathrm{e}^{-\frac{1}{T}\Delta\mathscr{H}_i}} \qquad (1)$$

(*Glauber rule*). Here $\vec{s}^{\,i}$ stands for $\vec{s}$ after flipping the spin at i, and $\Delta\mathscr{H}_i \equiv \mathscr{H}(\vec{s}^{\,i}) - \mathscr{H}(\vec{s})$. The Boltzmann constant is set $k_B \equiv 1$ in this paper. One may interpret that this rule describes a spin−flip mechanism under the action of two competing heat

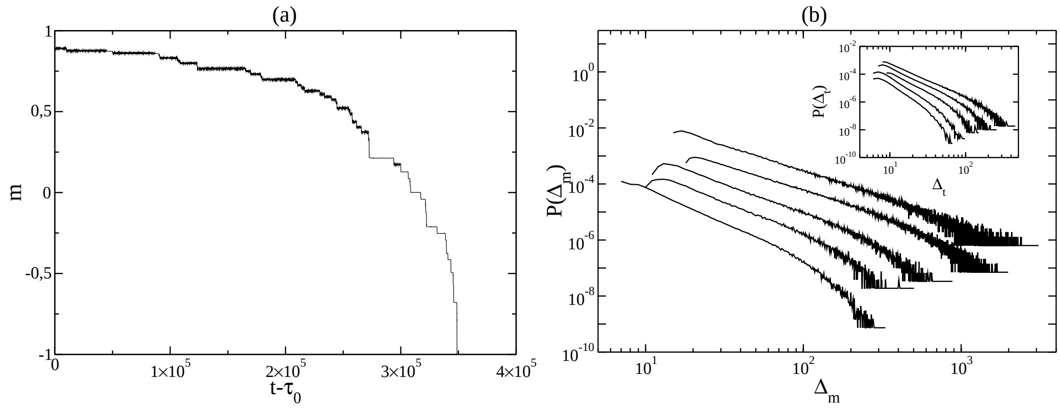

FIGURE 1. (a) Decay from a metastable state for a $R = 30$ particle. Avalanches are seen by direct inspection. Time is in Monte Carlo Steps per Spin (MCSS). Here $\tau_0 \sim 10^{30}$ MCSS. (b) Large avalanche size distribution $P(\Delta_m)$ for the circular magnetic nanoparticle and varying R. From top to bottom, $R = 120, 84, 60, 42$ and 30. Curves have been shifted in the vertical direction for visual convenience. The inset shows $P(\Delta_t)$ for the same system sizes.

baths: with probability p, the flip is performed completely at random ($\vec{s}$ is assumed in contact with a heat bath at 'infinite' temperature), while the change is performed at temperature T with probability $1-p$. For $p = 0$, eq. (1) corresponds to the equilibrium Ising case, which exhibits for $h = 0$ a critical point at $T = T_C \approx 2.2691J$. Otherwise, the conflict in (1) impedes canonical equilibrium, and the system evolves with time towards a non−equilibrium steady state whose nature essentially differs from the Gibbs state for T. It is assumed that this kind of stochastic, non−canonical perturbation for $p > 0$ may actually occur in nature due to microscopic disorder or impurities, etc.[2] Motivated also by the experimental situation, we choose to study a finite, relatively small two−dimensional system subjected to open circular boundary conditions. This system is defined on a square lattice, where we inscribe a circle of radius R. Sites outside this circle do not belong to the system and hence contain no spins. We mainly report here on a set of fixed values for the model parameters, namely, $J = 1$, $h = -0.1$, $T = 0.11T_C$ and $p = 10^{-6}$. This choice is dictated by simplicity and also because (after exploring the behavior for other cases) we came to the conclusion that this corresponds to an interesting region of the system parameter space, where the effects of p and T are comparable and clusters are compact and hence easy to analize. We believe that we are describing here typical behavior of our model, and the chances are that it can be observed in actual materials. The lattice is set initially with all spins *up*, $s_i = +1$ for $i = 1, ..., N$. Under negative field $(h = -0.1)$, this ordered state is metastable, and it eventually decays to the stable state which, for $T = 0.11T_C$, corresponds to $m \equiv N^{-1} \sum_i s_i \simeq -1$. Finally, the simulations reported here required in practice using the $s-1$ Monte Carlo algorithm with *absorbing Markov chains*,[3, 4] and the *slow forcing limit approximation*.[5]

Under these conditions, the system, after spending a long time wandering around the metastable state, finally relaxes towards the stable one. As it is clear in Fig. 1.a, the relaxation of magnetization occurs via a sequence of well−defined abrupt jumps. That is, when the system relaxation is observed after each MCSS, which corresponds to a

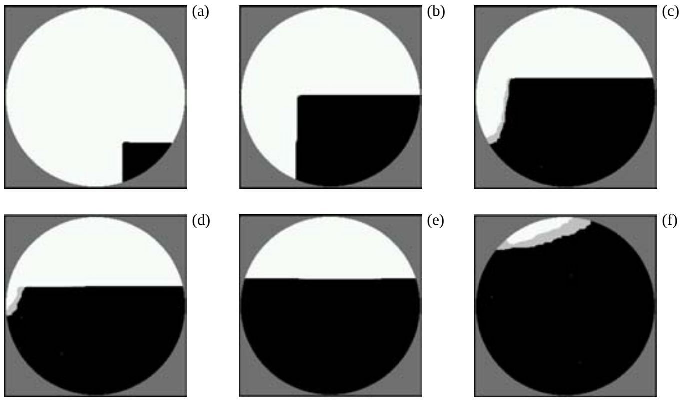

FIGURE 2. Some snapshots of a particular decay of a circular nanoparticle of radius $R = 120$. Avalanches are plotted in grey. Notice that large avalanches appear only for curved domain walls.

'macroscopic' time scale, *stricktly monotonic changes of* $m(t)$ can be identified that we shall call *avalanches* in the following. To be precise, consider the avalanche beginning at time t_a, when the system magnetization is $m(t_a)$, and finishing at t_b. We define its *size* and *lifetime* or *duration*, respectively, as $\Delta_m = |m(t_b) - m(t_a)|$ and $\Delta_t = |t_b - t_a|$. Our interest is on the histograms $P(\Delta_m)$, $P(\Delta_t)$ and $P(\Delta_t|\Delta_m)$.

Fig. 1.b shows the avalanche size distributions $P(\Delta_m)$ for different sizes R of the magnetic nanoparticle, once the trivial *extrinsic noise*[6] has been substracted[7]. A power law behavior, followed by an (exponential) cutoff is clearly observed. The measured power law exponents, $P(\Delta_m) \sim \Delta_m^{-\tau(R)}$, show size-dependent corrections to scaling. Similar corrections have been also found in real experimental systems.[8] These corrections are compatible with a functional dependence of the form $\tau(R) = \tau_\infty + a/R^2$, where $\tau_\infty = 1.71(4)$. Analogously, the lifetime distributions also show power law behavior, $P(\Delta_t) \sim \Delta_t^{-\alpha(R)}$. The inset of Fig. 1.b shows $P(\Delta_t)$ (once substracted the extrinsic noise) for the same system sizes. Again, the exponents $\alpha(R)$ are compatible with a law $\alpha(R) = \alpha_\infty + a'/R^2$, where $\alpha_\infty = 2.25(3)$. Hence we expect avalanche power law distributions $P(\Delta_m) \sim \Delta_m^{-\tau_\infty}$ and $P(\Delta_t) \sim \Delta_t^{-\alpha_\infty}$, with $\tau_\infty = 1.71(4)$ and $\alpha_\infty = 2.25(3)$, in the Thermodynamic Limit. On the other hand, the power law behavior of both $P(\Delta_m)$ and $P(\Delta_t)$ lasts up to an exponential cutoff Δ_m^c and Δ_t^c, respectively, which depends on system size. We measure these cutoff values fitting an exponential function of the form $exp[-\Delta_{m(t)}/\Delta_{m(t)}^c]$ to the cutoff tails, and find a power law dependence with R, i.e. $\Delta_m^c \sim R^{\beta_m}$ and $\Delta_t^c \sim R^{\beta_t}$, where $\beta_m = 2.32(6)$ and $\beta_t = 1.53(3)$. Analogous power law dependences of cutoff with system size have been found in real magnetic materials. [9] In order to investigate the relation between the size and lifetime of an avalanche, we study the histogram $P(\Delta_t|\Delta_m)$, i.e. the probability of measuring an avalanche with lifetime Δ_t when its size is Δ_m. We find that for each value of Δ_m, the marginal distribution $P(\Delta_t|\Delta_m)$ shows a narrow peak around certain typical value $\langle\Delta_t\rangle_{\Delta_m}$. This means that the relation between the lifetime and the size of an avalanche is rather deterministic in our model system. If we now assume that this relation is of the form $\Delta_m \sim \Delta_t^\gamma$, we are able to measure γ in an indirect way using the cutoff dependence with R. In this way we

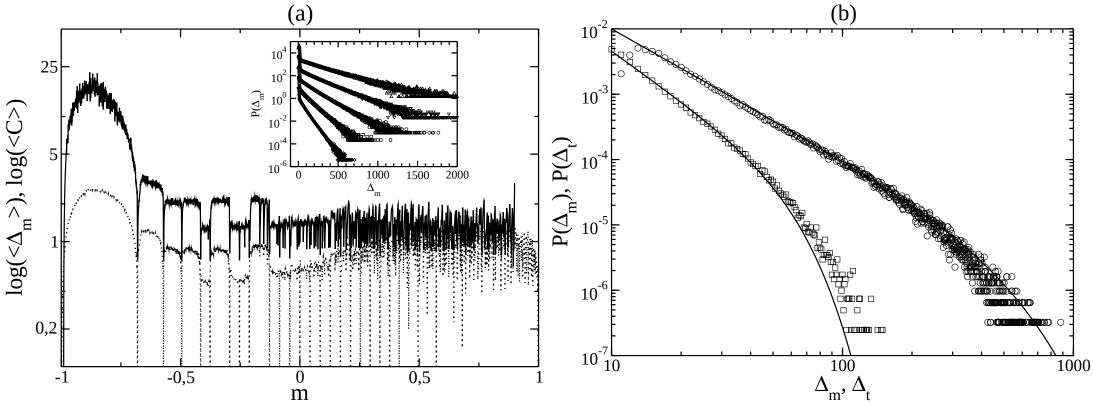

FIGURE 3. (a) The mean avalanche size $\langle \Delta_m \rangle$ and the mean curvature $\langle \mathscr{C} \rangle$ as a function of magnetization for a particle with $R = 30$ after averaging over $N_{exp} = 3500$ runs. The inset shows a semilog plot of the avalanche size distributions for domain walls with constant nonzero curvature. Curvature increases from bottom to top. (b) Avalanche size and lifetime distributions as obtained from eq. (2), for $N = 200$, $A_{min} = 0.007$ y $A_{max} = 1$. For comparison we also show the measured $P(\Delta_m)$ and $P(\Delta_t)$ for the $R = 60$ particle.

obtain $\gamma = \beta_m / \beta_t$ which yields $\gamma = 1.52(5)$ using the measured values of β_m and β_t. The observed avalanche power laws strongly depend on the presence of both the free borders and the impurity p. This *apparent* scale-free behavior disappears if the magnetic nanoparticle shows no free borders, or if we make $p = 0$ in eq. (1).

The reported results apparently support the existence of an underlying continuous phase transition responsible of the observed scale invariance. However, as we will see below, the observed power law behavior is *effective*, in the sense that the system is not really critical. Instead, we will show that a finite (but large) number of different, gap−separated typical scales appear superposed in such a way that the global distributions shows several decades of power law behavior. Fig. 2 shows some snapshots of the demagnetization process. Due to the low temperature, the evolution of the system is determined by the interface and its interplay with the open boundaries. In fact, we observe that large avalanches are associated to curved regions of the domain wall. Interfacial curvature appears due to the faster growth of the domain wall near the concave open borders, and it gives rise to large avalanches towards configurations with less interfacial energy. In order to clarify this relation we study now the mean avalanche size $\langle \Delta_m \rangle$ and the mean interface curvature $\langle \mathscr{C} \rangle$ (measured as the number of step-like up interfacial spins) as a function of magnetization. Fig. 3.a shows both curves measured in a $R = 30$ particle after averaging over 3500 runs. Here we observe that there is an high correlation between avalanche size and domain wall curvature. High curvature involves large avalanches and reversely. Therefore the typical avalanche size is perfectly determined by the curvature of the interface when this avalanche starts. Moreover, we can measure the probability of finding an avalanche of a given size Δ_m if the domain wall has certain constant curvature. In order to do that we design an (unrealistic) modification of our system where a domain wall with constant curvature evolves indefinitely.[7] This evolution proceeds also via avalanches. The inset of Fig. 3.a shows a semilog plot of the avalanche size distribution obtained in this system for different curvatures $\mathscr{C}$. Apart from the trivial (exponential) extrinsic noise, a stretched exponential regime for large avalanches is observed, $P_{large}(\Delta_m) \sim \exp[-(\Delta_m/\bar{\Delta}_m)^\eta]$, with $\eta \approx 0.89$ and where $\bar{\Delta}_m$ depends in an exponential fashion on $\mathscr{C}$. Hence, a domain wall with constant curvature $\mathscr{C}$ is characterized by avalanches with a *typical* size $\bar{\Delta}_m(\mathscr{C})$. On the other hand, interfacial curvature in the magnetic nanoparticle takes a wide range of different values as the particle demagnetizes (see Figs. 2 and 3.a). Thus, the power law avalanche distributions observed in the circular particle are just finite superpositions of distributions with well defined typical sizes.

Moreover, we can calculate[7] the result of a finite superposition of exponential distributions, each one with a well-defined typical rate. Let's $\mathscr{P}(x|A) = A\exp(-Ax)$ be the probability of finding an event of size x when the system is characterized by certain observable taking the value A. Now $Q(A)$ is the probability of finding the system in a state characterized by A. If we assume that the observable A can take a finite number $N+1$ of equally spaced values in the interval $[A_{min}, A_{max}]$, $A_n = A_{min} + n\Delta$, where $\Delta = (A_{max} - A_{min})/N$, and that all these values are equally probable, $\mathscr{Q}(A) = \text{constant}$, then the probability of finding an event of size x can be written as[7],

$$\mathscr{P}(x) = \Delta e^{-A_{min}x}\left[\frac{A_{min}}{1-e^{-\Delta x}} - A_{max}\frac{e^{-(N+1)\Delta x}}{1-e^{-\Delta x}} + \Delta\frac{e^{-\Delta x}(1-e^{-N\Delta x})}{(1-e^{-\Delta x})^2}\right] \tag{2}$$

Fig. 3.b shows the event size distribution $\mathscr{P}(x)$ obtained from eq. (2) for $N = 200$, $A_{min} = 0.007$ and $A_{max} = 1$. The curve follows power law behavior up to an exponential cutoff given by $\exp(-A_{min}x)$. Fig. 3.b also shows for comparison the avalanche size distribution measured for a circular particle with $R = 60$. The agreement is very good. Moreover, assuming that eq. (2) represents the avalanche size distribution, and using the relation $\Delta_m \sim \Delta_t^\gamma$ between the size and the lifetime of an avalanche, we can obtain the avalanche lifetime distribution via the conservation of probability, $P(\Delta_m)d\Delta_m = P(\Delta_t)d\Delta_t$. Thereby, if $\mathscr{P}(\Delta_m)$ is the probability of finding an avalanche with size Δ_m, the probability of finding an avalanche with lifetime Δ_t is $\gamma\Delta_t^{\gamma-1}\mathscr{P}(\Delta_t^\gamma)$. This curve, also shown in Fig. 3.b, agrees with the measured avalanche lifetime distribution for the $R = 60$ magnetic particle when we use the previously measured value $\gamma = 1.52(5)$. This agreement confirms the measured value for the exponent γ, and on the other hand it also strengthens our conclusion about the origin of the scale invariance in this problem. Hence the superposition of a finite (but large) number of exponential distributions with different typical rates, which we observe for avalanches in our magnetic particle model, results in a global distribution which shows several decades of power law behavior, together with an exponential cutoff corresponding to the slowest exponential typical rate.

In this way we propose in this paper a new mechanism to obtain power law distributions not related to any underlying critical dynamics.[10] However, the deep insight derived from this analysis comes when we extrapolate the conceptual framework here developed to the understanding of Barkhausen Noise in particular and $1/f$ Noise in general. Barkhausen Noise is the noise by which an impure ferromagnet responds to a slowly varying external magnetic field. This response is not continuous, but burst-like. In particular, magnetization jumps are observed as a function of the applied field which are called *avalanches*. These avalanches exhibit size and lifetime power law distributions. Moreover, this measured scale-free behavior appears without any need of fine tuning. For many years theoretical physicist have been wondering about the origin of this *spontaneous* or *self-organized* scale invariant behavior. Different theoretical approaches have been proposed as explanation of Barkhausen Effect.[11] Most of these approaches are incompatible among them[6], and all of them are based on assuming the existence of an underlying critical point, responsible of the observed scale invariance. This assumption implies that *universality* must be observed in Barkhausen experiments. However, this is not observed in practice. Furthermore, the power of most practical applications of Barkhausen Noise is based on the sensibility of Barkhausen emission to microstructural details in the material.[12] Such sensibility is incompatible with the concept of universality derived from a critical point. On the other hand, the exponents we have obtained for the avalanches in the magnetic nanoparticle are almost equal to those measured by Spasojević et al[6] in Barkhausen experiments on quasi-bidimensional VITROVAC. Furthermore, our system shows *reproducibility*, i.e. large avalanches always appear at the same stages of evolution, independently of the observed experiment (see Fig. 3.a). The same property has been observed in Barkhausen experiments with Perminvar and a Fe-Ni-Co alloy.[13] In addition, our avalanche size and lifetime distributions show exponential cutoffs which depend algebraically on system size. This behavior has been also reported in real materials[9]. Finally, the measured exponents τ and α show finite size corrections similar to those found in experiments with avalanche systems[8]. All these similarities, together with the fact that experimental observations do not support the existence of universality in Barkhausen Noise, led us to suspect that Barkhausen Noise might also come from the superposition of more elementary events with well-defined typical scales. In fact, the $1/f$ noise behavior in this case is assumed to reflect topological rearrangements of domain walls,[14] which result in practice in a series of jumps between different metastable states, which is the basic process in our model.

The chances are that our observation that scale invariance originates in a combination of simple events, which we can prove in our model cases, is a general feature of similar phenomena in many complex systems. This should explain why distributions exhibiting power law, exponential or stretched exponential behavior have been identified in different but related experimental situations and in different regimes of the same experiment.

ACKNOWLEDGMENTS

We acknowledge M.A. Muñoz for useful comments. P.I.H. wishes to thank S. Zapperi for a fruitfull discussion about Barkhausen Noise, and M.A. Novotny for sharing unpublished information about MCAMC algorithms.

REFERENCES

1. O. Penrose and J.L. Lebowitz, in *Fluctuation Phenomena* (2nd edition), edited by E. Montroll and J.L. Lebowitz, North−Holland, Amsterdam 1987.

2. J. Marro and J.A. Vacas, *Phys. Rev. B* **56**, 8863 (1997).
3. M.A. Novotny, *Phys. Rev. Lett.* **74**, 1 (1994).
4. A.B. Bortz et al, *J. Compt. Phys.* **17**, 10 (1975).
5. M. Kolesik, M.A. Novotny and P.A. Rikvold, *Phys. Rev. Lett.* **80**, 3384 (1998).
6. D. Spasojević et al, *Phys. Rev. E* **54**, 2531 (1996).
7. Pablo I. Hurtado, *Dynamics of Nonequilibrium Systems: Metastability, Avalanches, Phase Separation, Absorbing States and Heat Conduction*, Ph.D. thesis (2002).
8. V. Frette et al, *Nature* **379**, 49 (1996).
9. M. Bahiana et al, *eprint cond-mat/9808017* (1998). BUSCAR REFERENCIA PUBLICADA.
10. D. Sornette, *eprint cond-mat/0110426* (2001). BUSCAR REFERENCIA PUBLICADA.
11. J.P. Sethna et al, *Nature* **410**, 242 (2001); G. Durin and S. Zapperi, *Phys. Rev. Lett.* **84**, 4705 (2000).
12. L. Sipahi, *J. Appl. Phys.* **75**(10), 6978 (1994); L. Sipahi et al, *J. Appl. Phys.* **75**(10), 6981 (1994).
13. J.S. Urbach et al, *Phys. Rev Lett* **75**, 4694 (1995).
14. See, for instance, X. Che and H. Suhl, *Phys. Rev. Lett.* **64**, 1670 (1990); K.L. Babcock and R.M. Westervelt, *Phys. Rev. Lett.* **64**, 2168 (1990).