Subharmonics and Aperiodicity in Hysteresis Loops

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We show that it is possible to have hysteretic behavior for magnets that does not form simple closed loops in steady state, but cycles multiple times before returning to its initial state. We show this by studying the low temperature dynamics of the 3D Edwards-Anderson spin glass. The specific multiple varies from system to system and is often quite large and increases with system size. The last result suggests that the magnetization could be aperiodic in the large system limit for some realizations of randomness. It should be possible to observe this phenomenon experimentally.

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Hysteresis in magnetic systems [1] is the prototype for understanding history dependent behavior in all complex systems [2–4]. Although the usual example of this is for ferromagnets, it can exist in other magnetic systems such as antiferromagnets [5] or spin glasses [6,7].

For magnetic systems, when one cycles adiabatically between a large positive and negative field H, the magnetization M forms a closed loop in the H-M plane because of saturation. There is an expectation that even if the field were cycled repeatedly between two moderate values, the magnetization would settle down to a closed loop, after some initial transient response. Here we show that this expectation is often wrong. For Ising spin-glass models (short or long ranged) in steady state, many cycles of the external field are necessary to bring the magnetization back to its initial value. Alternatively, with an oscillatory applied field the magnetization shows a subharmonic component. The number of cycles varies from system to system depending on the realization of randomness, grows with system size, and can be very large. Our estimates suggest that this effect should be observable in spin-glass nanoparticles at low temperatures.

This result might appear to be surprising as there are many cases when the strong result of "return point memory" [8,9] can be shown: if a system goes from a field H_1 to H_2, its final state is independent of the time dependence of H as long as H is never outside the interval $[H_1, H_2]$. In the case above this would imply at most one transient cycle before the steady state closed loop was reached. An example of such a system is a random field Ising ferromagnet where an elegant proof of return point memory was given [8]. However, the proof does not apply to cases where antiferromagnetic interactions are also present, such as a spin glass. We show below that in this case there is a much richer class of memory effects.

Earlier work has reported subharmonic response in the magnetization of ferromagnetic materials subjected to oscillating magnetic fields [10]. It is a specific example of the general phenomenon of ferroresonance. However, this occurs when the magnetic material is part of an electrical circuit (e.g., as the core of an inductor) that is driven at finite frequency. Furthermore, the intrinsic response of the ferromagnetic material is expected to show return point memory. From a dynamical systems viewpoint, this is simply the observation that a driven nonlinear oscillator can respond at a frequency different from the driving frequency, possibly depending on the initial conditions of the system. There has also been theoretical work on finite frequency hysteresis loops in ferromagnets [11–13]. In contrast, we consider only H fields that vary sufficiently slowly to be adiabatic and our nonreturn point effect is intrinsic to the material.

We consider the standard Edwards-Anderson spin-glass Hamiltonian [14,15] with Ising spins and nearest neighbor interactions in three dimensions,

$$\mathcal{H} = -\sum_{\langle i,j\rangle} J_{i,j} S_i S_j - H \sum_i S_i. \qquad (1)$$

The coupling strengths $J_{i,j}$ are chosen to be uniform random numbers between -1 and 1. The Ising spin variables S_i take values ± 1. We employ free boundary conditions; other boundary conditions yield similar results. We use single spin-flip dynamics at both zero and nonzero temperatures T, and vary the magnetic field H adiabatically. Thus as the magnetic field is changed, a spin flips only if it is energetically favorable for it to do so [16]. Once a spin flips, it can render several other spins unstable, forming an avalanche. If this happens, we choose which one of the unstable spins to flip with a Monte Carlo algorithm: the change in energy ΔE as a result of a spin flip is calculated for every spin. The probability of a spin flipping is then taken to be $\propto \exp(-\Delta E/T)$, with the spins updated randomly. (Strictly speaking, one would need to use barrier heights for each spin, but this would be difficult to estimate.) The adiabatic nature of the dynamics is implemented by flipping enough spins at one field to ensure that all spins that are unstable have been flipped before changing H. Note that if a spin flip is energetically unfavorable, it is *not* performed, contrary to conventional Monte Carlo procedure. We return to this issue in our estimate of experimental parameters.

At zero temperature, if several spins are unstable, the above algorithm will flip the spin that lowers the energy the most (even if there are several ongoing avalanches). At finite temperature, the order of flips is no longer deterministic and this would be expected to reduce the periodicity of the hysteresis curves. At low temperatures, one might hope to see a clear signature for subharmonic loops. Below we will examine the effects of temperature in detail and show that one should expect to observe subharmonic behavior in experimentally realistic situations.

We start the system at large H with all the spins positive and lower H to $H_{\min}$ adiabatically. The field is then raised to $H_{\max}$. Thereafter, the field is cycled repeatedly between these two extremal values. After an initial transient period, the system reaches steady state. In this steady state we determine the number of cycles of H that are required for the system to repeat its configuration. This varies randomly, depending on the $J_{i,j}$'s.

If $H_{\min}$ and $H_{\max}$ are too large, the magnetization will saturate at the extremal points of the H cycles and the behavior seen is trivial. On the other hand, if the range of H is too small, hysteretic effects are minimal. By trying various values of the extremal fields, we find that it is best to choose them to be about half of the saturation field. Consequently, we work with $H_{\max} = -H_{\min} = 1.4$.

Figure 1 shows an example of a system at zero temperature with linear dimension $L = 8$, i.e., with 8^3 spins, where the steady-state behavior is a two-cycle, i.e., two cycles of the magnetic field correspond to one cycle in the magnetization. Even though the gap between the two halves of the magnetization loop is much smaller than the width of the loop itself, with an oscillatory magnetic field, it would be straightforward to detect the subharmonic response. More complicated cycles are often found, but are not shown for the sake of clarity.

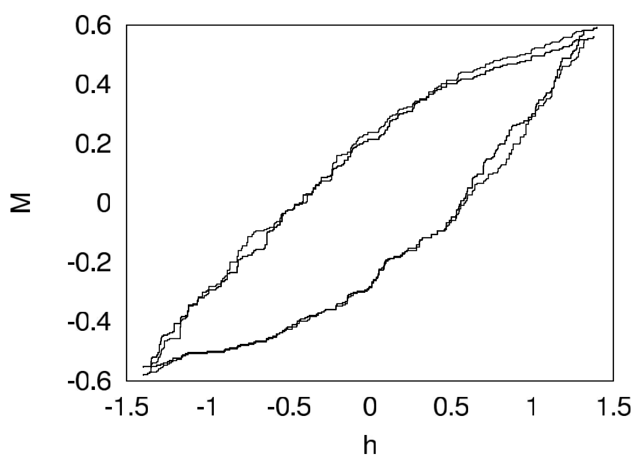

FIG. 1. Steady-state magnetization as a function of magnetic field for a spin glass with 8^3 spins. It can be seen that two cycles of the magnetic field are needed for the magnetization (and in fact the full spin configuration of the system) to return to its initial value.

More generally, m cycles of the magnetic field can be required for the (steady-state) magnetization to return to its initial value. The distribution of m for systems of different sizes at $T = 0$ is shown in Table I. One observes that the larger the system size, the more likely it is to find long cycles.

Although Table I shows an increase in typical cycle size as the system is made larger, this is not entirely caused by the dynamics becoming more complex, but also by the definition of cycle length. For instance if the system were divisible into two independent halves, with a six- and seven-cycle, respectively, the full system would need 42 cycles to return to its original state. To eliminate this, we examine the power spectrum. The magnetization is measured at $H_{\min}$ and $H_{\max}$, the extrema of the cycles of H. The discrete Fourier transform is taken and the power spectrum is calculated. In the example above, the spectrum will show peaks only at multiples of $f = 1/6$ and $f = 1/7$, where $f = 1$ is the frequency of the H field, whereas if the system were truly indivisible, peaks at multiples of $f = 1/42$ would be observed. In addition, the power spectrum is crucial to detecting these effects experimentally, as discussed previously. Although in an experimental situation one would measure the power spectrum for the full time dependent magnetization $M(t)$, the discrete Fourier transform should be a good indicator thereof.

Figure 2 shows the power spectra for $L = 8$ at $T = 0$ and $T = 0.2$, measured in units where the couplings $-1 < J < 1$. This is an average of the spectra for 2048 different realizations of the couplings with a sample length of 512 cycles of the field. Sharp peaks are seen at discrete frequencies, with the dominant peak at $f = 1/2$. At $T = 0.2$, the maximum of the $f = 1/2$ peak is reduced to about half of the zero temperature value. The inset shows a blowup of some of some lower

TABLE I. Probability that m cycles of the magnetic field are required for the magnetization to return to its initial value, for systems of sizes 4^3, 8^3, and 16^3. The number of systems considered for the three different sizes was 100 000, 10 000, and 1000, respectively. Larger m values are more likely for bigger L.

m	$L = 4$	$L = 8$	$L = 16$
1	0.9343	0.68	0.052
2	0.0241	0.2342	0.503
3	0.0359	0.0481	0.044
4	0.0005	0.0179	0.109
5	0.0044	0.0056	0.006
6	0	0.0086	0.206
7	0.0006	0.0021	0
8	0	0.0010	0.007
9	0.0002	0.0011	0.002
10	0	0.0005	0.025
> 10	0	0.0009	0.046

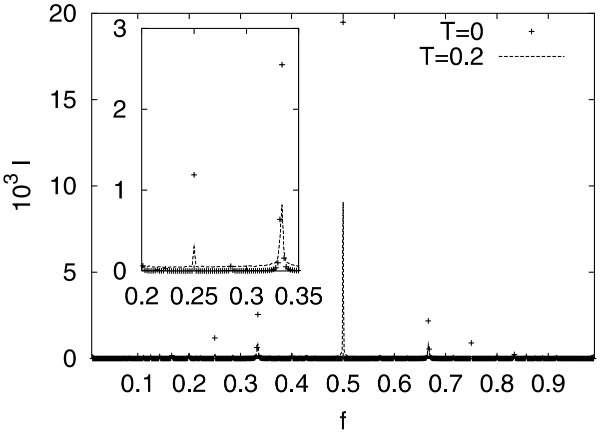

FIG. 2. Average power spectrum for systems of 8^3 spins. The peaks at $f = 1$ and $f = 0$, whose heights are roughly 3×10^4, are suppressed. The power is concentrated at $f = 1/2$, with smaller peaks at other fractions. The data are at temperatures $T = 0$ (+) and $T = 0.2$ (curve). The inset is a blown up portion of the graph with peaks at different subharmonics for both temperatures.

subharmonics, at $f = 1/3$ and $f = 1/4$. Although reduced from their zero temperature value, they are still much higher than the background and should therefore be in principle observable. We have also considered the case when, if there are several spins that are simultaneously unstable, the one which is first flipped is chosen completely randomly. Even in this case a fraction of systems have a steady-state magnetization that have true delta functions at subharmonic frequencies [17], reflecting the robustness of the dynamics to small perturbations.

Figure 3 is an enlarged plot of the power spectrum for $T = 0$ at low frequencies for $L = 4$ and $L = 16$. Although small, the low frequency power is greater for $L = 16$. In addition, the peaks go down to lower frequencies, demonstrating that the dynamics get more complex

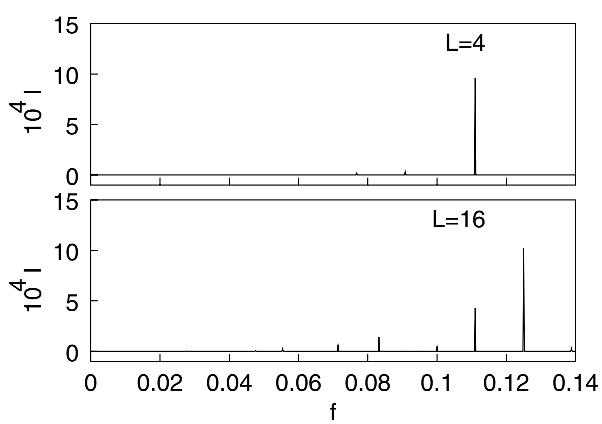

FIG. 3. Low frequency average power spectrum for systems of 4^3 and 16^3 spins. (To avoid confusion with the horizontal axis, the vertical axis has an origin at -1.) There is substantially more power at low frequencies for 16^3 spins, and the spectrum extends down to $1/f \approx 40$, i.e., a 40-cycle that is indivisible. The number of systems is the same as in Table I.

as L is increased in addition to the trivial growth in cycle length discussed in the previous paragraph.

In Fig. 2 the two trivial peaks, at $f = 0$ and 1, are not shown. These are approximately 10^5 times the strength of the peak at $f = 1/2$. As one would expect from Table I, the power spectrum for $L = 4$ has fewer peaks. However, the relative strength of the peaks is larger: the peak at $f = 1/2$ is approximately 10^{-4} times the strength of the peaks at $f = 0$ and 1. More generally, as L is varied, the strength of the trivial peaks will be proportional to the number of spins, i.e., L^3. Although the limited range of L that we have been able to simulate does not allow us to determine the dependence on L of the other peaks, for large L we might expect any system to behave as a collection of reasonably independent regions, and the strength of the low frequency peaks to be proportional to $L^{3/2}$. However, this argument is hazardous, since even a small coupling between different regions of the system might affect subtle details of the dynamics. On the basis of our numerics, we make the more conservative claim, that the strength of the low frequency peaks increases with L, while their relative strength (compared to the peaks at $f = 0$ and 1) decreases with L. In a phase-locked experimental setup, the absolute rather than the relative strength of these peaks is important.

We now turn to experimental considerations. Nanoparticles of spin-glass material, such as $Fe_{0.5}Mn_{0.5}TiO_3$, with $O(16^3)$ separate spins would be a few nanometers in diameter. It should be possible to reduce the grain size somewhat, although, as discussed in the previous paragraph, the subharmonic content is reduced. In order to eliminate possible problems from intergrain interaction, it would advisable to coat or embed the grains in a neutral material to isolate them from each other. With N_g independent grains, the strength of the signal would be proportional to $\sqrt{N_g}$.

As discussed earlier, the field should be varied over a range of order half the saturation field, which would be $\sim$10 T. (If a material with a lower T_c is used, at correspondingly lower temperatures, the magnetic field could be reduced.) The time dependence of the magnetic field should be slow enough to evolve the spins adiabatically and to be experimentally achievable. From the latter constraint, we estimate a frequency of $\sim$0.1 Hz.

In our numerics we did not allow spin flips that were energetically unfavorable. Experimentally, the temperature has to be sufficiently low for the probability of such a spin flip to be small in one cycle of the magnetic field. Assuming that the rate for favorable spin flips is $\sim 10^9$ Hz, we obtain $\Delta E/k_B T \gg \ln(10^{10})$. Estimating $\Delta E \approx 0.25 k_B T_c$, with the critical temperature of $Fe_{0.5}Mn_{0.5}TiO_3$ $T_c \sim 20$ K, experimental temperatures should be less than $\sim$0.2 K.

It should be possible to satisfy all these conditions experimentally. For instance, recent work has found super spin-glass behavior in nanoparticle multilayer composites [18] and in frozen ferrofluids [19], where the effective individual spins are large and T_c is correspondingly high. While this would render the phenomena we wish to observe accessible at higher temperatures, one should be careful because the saturation field increases with T_c.

From a theoretical perspective, it is of interest to ask whether the results we have found here are generic for systems with hysteresis. As mentioned earlier in this Letter, there are strong constraints on many ferromagnetic models that force them to have return point memory, precluding the possibility of multicycle hysteresis loops. With the spin-glass model that we have considered in this Letter, multicycles are seen in two dimensions but not in one dimension.

From a dynamical systems approach, it would be natural to expect a system with many interacting degrees of freedom to exhibit complex dynamical behavior, possibly even chaos. In order to verify whether this is the source of the results reported here, we have carried out similar numerical simulations on the Sherrington-Kirkpatrick (SK) model [20] for spin glasses, as an example of a coupled nonlinear system with several degrees of freedom (the spins). We find that one needs a minimum of five spins to find multicycle steady-state magnetization. In this case, one finds only three cycles and no others aside from the trivial closed loop case. The fraction of systems showing three cycles for five spins is low, approximately 3×10^{-4}. However this fraction rises steeply with the number of spins. One has to be careful in applying general results from dynamical systems, since the dynamics we consider here are of a special kind, being driven by energy minimization. For instance, with a constant magnetic field and any initial state, the system must evolve towards a fixed point. Thus there may be features, e.g., frustration, essential to see the behavior reported in this Letter. Further work is needed to clarify this issue.

Our results for the SK model suggest that systems which are described by a Landau theory with five or more components to the order parameter might, in the correct parameter regime, show multicycle hysteresis behavior. This would have the advantage that the effect would show up in intensive quantities. As discussed earlier in this Letter, for the spin-glass system we have considered here the nonreturn point effects for the average magnetization per spin should vanish in the large size limit [6].

In conclusion, we have shown in this Letter that hysteresis loops have much richer behavior than commonly assumed. For Ising spin-glass models with zero temperature dynamics, a slowly varying cyclical magnetic field produces a subharmonic response in the magnetization. Our estimates indicate that this effect should be observable in low temperature experiments. Broader implications for other systems have been discussed.

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