Avalanches and Slow Relaxation: Dynamics of Ultrathin Granular Superconducting Films in a Parallel Magnetic Field

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We report unusual dynamics of ultrathin granular Al films in the hysteretic region near the parallel critical field. Avalanches in film resistance, corresponding to the collapse of macroscopic superconducting regions, are observed. As films approach the normal state, large avalanches give way to slow, stretched-exponential decay interspersed with smaller avalanches. The size distribution of the avalanches exhibits power-law behavior over three decades. We discuss the origins of the avalanches and the slow relaxation in terms of a 2D random array of Josephson junctions.

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Disorder is known to be relevant to a wide variety of complex systems displaying nonequilibrium behavior. One characteristic of these systems is slow, nonexponential time relaxation out of a metastable state, such as seen in magnetic glasses [1]. Another is the tendency to avalanche as an external parameter is varied adiabatically. Avalanches, in particular, have been seen in systems that are strongly hysteretic, such as the athermal martensitic transitions [2], ferromagnetic systems [3], sandpiles [4], and fluid invasion of porous media [5]. However, relaxation due to fluctuations is not observed in these systems. Avalanches occur only when the system is driven through the hysteretic region by tuning a relevant parameter.

In this Letter, we report avalanches and exceptionally slow, nonexponential relaxations in ultrathin granular superconducting films in a parallel magnetic field, $H_\parallel$. Virtually all previous studies of the dynamics of granular superconducting films, as well as their companion model of the two-dimensional (2D) random array of Josephson junctions, have been considered in the context of the vortex state [6]. As we will show, the dynamics of such systems in the virtual absence of vorticity can be very unusual. Not only does parallel field circumvent the flux flow resistance, but, more importantly, $H_\parallel$ can be used to bring the films far out of thermodynamic equilibrium via a first-order transition. This system is unique in that both disorder and relaxation are important. In particular, after it is driven out of equilibrium by $H_\parallel$, the system seems to self-organize spontaneously, and avalanchelike jumps in resistance are observed on all scales.

It has been known for years [7] that superconductivity in ultrathin films in $H_\parallel$ could become limited by a first-order transition if the spin-orbit scattering is weak. This is the spin-paramagnetic limit, $H_{c\parallel}$, at which the Zeeman splitting of the quasiparticles, $-\chi_p H_{c\parallel}^2/2$, equals the superconducting condensation energy, $-N(0)\Delta^2/2$, where $\Delta, \mu_B, N(0)$, and $\chi_p = 2N(0)\mu_B^2$ are the superconducting gap, Bohr magneton, the density of states for one spin direction in the normal state, and the Pauli spin susceptibility, respectively. Earlier work on thin Al films suggested that the transition went from second order to first order near 600 mK [8]. However, no significant hysteresis was ever found to substantiate a first-order transition. Recently, we have extended experiments to lower temperatures [9] and have found a tricritical point, $T_{tri} \sim 270$ mK, separating the low-temperature first-order transition line and the usual second-order transition line. Below T_{tri}, a giant hysteresis develops. Typical resistance data during field sweeps are shown in Fig. 1. In the hysteretic region, avalanches and slow, stretched-exponential relaxation are the signatures of the dynamics of the system.

Ultrathin granular Al films were made by a standard electrochemical anodization process [10]. Scanning force microscopy images showed a typical grain diameter of about 25 nm. We believe that the thickness of the film was near 5 nm. Four-probe resistances were measured using a lock-in amplifier operating at 27 Hz, in a dilution refrigerator with a 90-kG superconducting magnet. The area between the two probe leads was 1×1.5 mm^2. A probe current of 1 nA was used unless otherwise

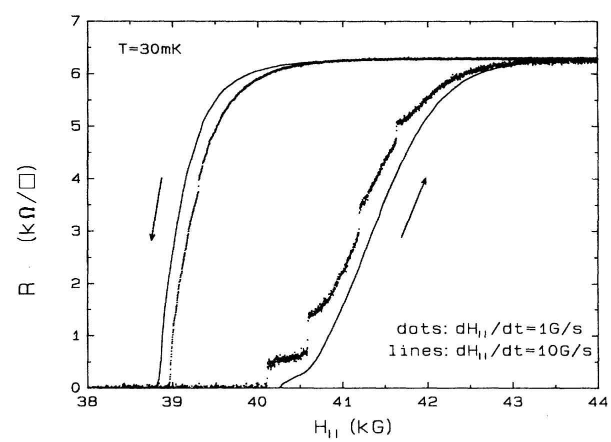

FIG. 1. R versus $H_\parallel$ at 30 mK for a film with $R_N = 6.25$ k$\Omega/\square$, showing two hysteresis loops at different field sweep rates, $dH_\parallel/dt$.

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specified. Parallel field position was found by adjusting the sample holder mechanically, *in situ,* to maximize both $H_{c\parallel}$ and the width of the hysteresis loop, $\Delta H_{\parallel}$. $\Delta H_{\parallel}$ was found to decrease with misalignment angle θ as $\Delta H_{\parallel} \propto \exp(-\theta/\theta_0)$ with $\theta_0 = 2.4°$. Films that were purposely misaligned up to 1° were also studied and no qualitative difference in the avalanches was observed, but a detailed study has yet to be carried out. Four-probe dc resistances were also measured at base temperature, 20 mK, with all electronics running on dc batteries. We did not find observable deviation from the behavior seen in ac measurements.

The hysteresis, shown in Fig. 1 at 30 mK for a film with normal state sheet resistance $R_N = 6.25$ k$\Omega/\Box$, was found to disappear rapidly near $T_{\rm tri} \sim 270$ mK for all the films studied [9] with $0.1 < R_N < 100$ k$\Omega/\Box$. Avalanches were observed in the entire range of R_N, but the relaxations tended to be slower in films of higher R_N. The superconducting transition temperature T_c was about 1.8 K and varied little with R_N. The width of the hysteresis loops, $\Delta H_{\parallel} \sim 2.2$ kG, also varied little with R_N for $R_N > 1$ k$\Omega/\Box$. In the following, we purposely present data on films with $R_N \sim R_Q[= h/(2e)^2 \approx 6.45$ k$\Omega/\Box]$. Studies on films of various R_N are in progress.

Hysteresis loops were very smooth when $H_{\parallel}$ was swept at a rate $dH_{\parallel}/dt \geq 10$ G/s; see the lines in Fig. 1. However, as the rate was reduced to 1 G/s or lower, they displayed sudden jumps in resistance, as shown by the dots in Fig. 1. Data in the 1 G/s run are noisier since a probe current of 0.1 nA was used, compared to 1 nA in the 10 G/s run. The critical current of the films was of the order of 10 μA for $H_{\parallel} \sim 38$ kG. The avalanche behavior did not change for probe currents ranging from 10 nA to the lowest value of 0.1 nA, indicating that the jumps in resistance were not a critical current effect. We are also certain that the absence of avalanches in the 10 G/s run was not due to eddy current heating. The data in Fig. 1 suggest that only for slow enough sweep rate is the system allowed to organize itself into an ostensibly critical state.

The narrowing of the hysteresis with lowering sweep rate in Fig. 1 is due to relaxation. This, as well as the avalanches, can be seen more vividly by measuring the time dependence of the sheet resistance, R. We performed a simple experiment in which $H_{\parallel}$ was first set to 38 kG, a field slightly below $H_{c\parallel}$ where the film had an unmeasurably small R. We then swept $H_{\parallel}$ up into the hysteretic region at a rate $dH_{\parallel}/dt \sim 3$ G/s while monitoring R. When R reached a desired value R_0, we held $H_{\parallel}$ constant by setting the superconducting magnet into the persistent current mode and started to measure R as a function of time. A larger value of R_0 corresponds to a higher $H_{\parallel}$ at which a field sweep was stopped. Films with $R_N \sim R_Q$ or higher displayed exceptionally slow relaxations. Shown in Fig. 2 are typical runs at 30 mK for $R_0/R_N = 5\%, 10\%$, and 20%,

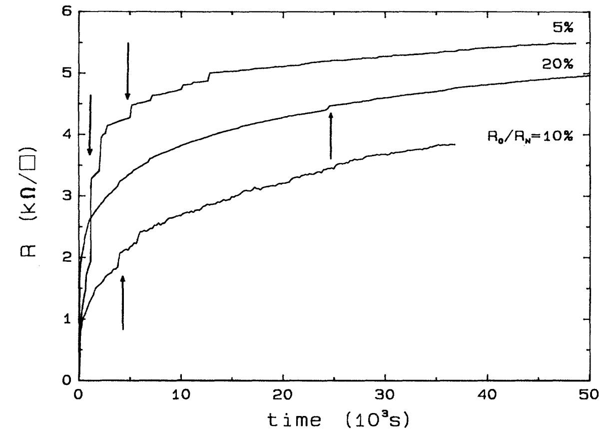

FIG. 2. R versus time after $H_{\parallel}$ was held constant when R_0/R_N reached desired values during field-up sweeps. Arrows indicate some of the avalanches. Note that the $R_0/R_N = 5\%$ curve actually jumped above the $R_0/R_N = 20\%$ curve.

respectively, for the film in Fig. 1. Data were taken at 20 s intervals. For large values of R_0/R_N, such as 10% and 20%, the time traces exhibited slow decay with small but measurable avalanches interspersed in the time trace. We have indicated some of them by the arrows in Fig. 2. Notwithstanding the avalanches, these curves can be fitted very well to a stretched-exponential form, $(R_N - R)/(R_N - R_0) = \exp[-(t/\tau)^{\gamma}]$, over four decades in time, as shown in Fig. 3. The time constant τ was of order 3×10^4 s, and γ equaled 0.5 and 0.4 for the 10% and 20% runs, respectively. More extensive studies at various temperatures below $T_{\rm tri}$ will be reported elsewhere [11]. There were no hysteresis, avalanches, and relaxation outside the hysteretic region, right above $T_{\rm tri}$, or in a perpendicular field.

As R_0/R_N was lowered to 5% or smaller, the time traces became dominated by avalanches at early times and were too discontinuous to be fitted reasonably to

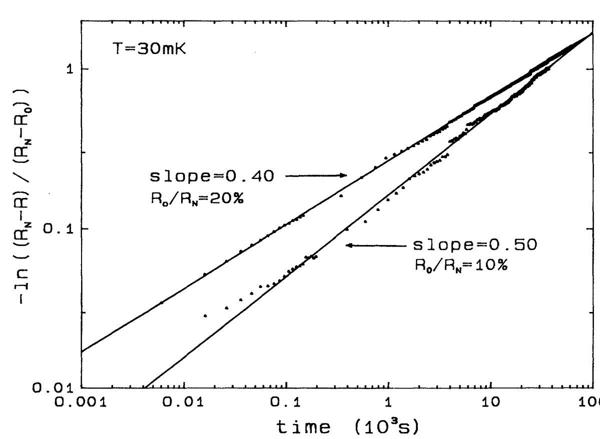

FIG. 3. Log-log plot for the data from Fig. 2. Lines are fits to a stretched-exponential form.

any functional form. Ironically, as can be seen in Fig. 2, relaxations at low values of R_0/R_N (such as 5%) resulted in such large avalanches in the early part of the time traces that the resistance usually jumped above those started at larger values of R_0/R_N (such as 20%). For $R_0/R_N =$ 0.5%, the curves were completely discontinuous, as can be seen in the top portion of Fig. 4. In the inset to Fig. 4, we plot a magnified region at later times to show the saw-toothed behavior resulting from repeated avalanches and subsequent relaxations. This unusual behavior is not understood, but is reminiscent of "fluctuators" seen in the conductance of disordered low-dimensional devices [12]. Similarly, due to the 2D nature of our films, fluctuations at a particular local site could have a measurable effect on the transport properties of the entire film [13].

To study the details of the behavior in the top part of Fig. 4, we plot in the lower portion of Fig. 4 the avalanche size, ΔR, as a function of time where ΔR is the resistance jump for an avalanche. This plot represents an avalanche spectrum. Figure 5 shows the size distribution of the avalanches where we plot the number of events, N, versus ΔR with the size being divided into six bins on a logarithmic scale spanning three decades. The data were collected from three time traces similar to the one shown in Fig. 4. Shown by a solid line in Fig. 5 is a power-law form, $N \sim (\Delta R)^{-1}$, as a guide to the eye. We emphasize that, when we divided time traces such as in Fig. 4 at the middle, the size distributions constructed from each part did not seem to deviate from the power-law behavior seen in the entire data set. The inset to Fig. 5 is a semilogarithmic plot of the number of events, N, versus the time interval between avalanches, Δt, with the solid line being a least-squares fit which gives a characteristic

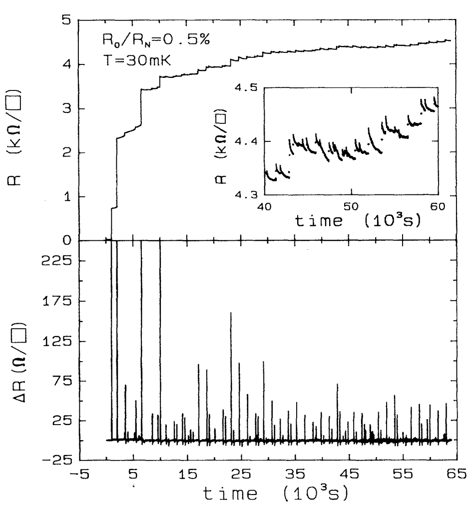

FIG. 4. Top: A typical time trace for $R_0/R_N = 0.5\%$. Inset: A magnified portion showing the saw-toothed behavior. Bottom: Avalanche spectrum from the time trace. Four biggest avalanches are off scale.

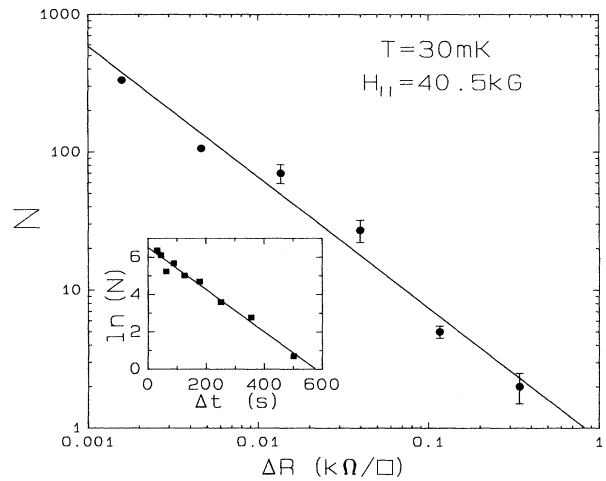

FIG. 5. Number of avalanches, N, versus avalanche size, ΔR. The straight line is a guide to the eye. Inset: Semilogarithmic plot of N versus the time interval between avalanches, Δt, with the straight line being a least-squares fit.

time scale for the avalanche interval of order 100 s.

Our system is very different compared to other systems [2–5] having avalanches. Of particular interest is that, when out of equilibrium, our films seem to self-organize spontaneously into a very fragile state that eventually avalanches even without further tuning of $H_{\|}$, suggestive of self-organized criticality [14]. However, the distribution of the intervals between avalanches, shown in the inset to Fig. 5, does not exhibit a power-law behavior. We also noticed that, in the descending part of the hysteresis during either a field sweep or a time decay, avalanches were smaller in size and happened only occasionally. Avalanches are mostly seen as the system evolves from a low-entropy state, with superconducting order, to the normal state. This asymmetry is also seen in the withdrawal of superfluid helium from a porous medium [5].

In order to make a conjecture as to the probable causes for the observed behavior, we will model our granular films as 2D random arrays of Josephson junctions [9,15]. Approaching $H_{c\|}$, the energy of the normal state for a grain is lowered by an amount $v_i \chi_p H_{\|}^2/2$ which is very close to the superconducting free energy $v_i N(0)\Delta^2/2$, where v_i is the volume of the ith grain. It is interesting to note that the grains are so small that $v_i N(0)\Delta^2/2 \sim 10\Delta$. Hence, there are of the order of 10 Cooper pairs on a grain, and the effect of charge quantization is therefore important as we will discuss later. Intergrain Josephson coupling, which further lowers the energy of the superconducting state by an amount $E_J \sim (R_Q/2R_N)\Delta$, is a significant fraction of the energy of a grain for $R_Q/R_N \sim 1$ and may be essential in determining whether or not a superconducting grain turns normal. If a grain that is Josephson coupled to its neighbors turns normal, then the energy of its neighbors will be raised due to the loss of their Josephson couplings with that grain. Such a scenario could cause a chain reaction, leading to the collapse of superconductivity within a large cluster, thereby producing an avalanche. The size of a big avalanche can a be a quarter of R_N or larger in our data. Assuming that the resistance scales with the area of the normal region, then a large avalanche corresponds to the collapse of a cluster containing 10^9 grains. Although we cannot exclude heating due to the latent heat released at the transition as the origin of the avalanches, we believe that the Josephson coupling is a more probable cause since the latent heat is probably small due to the near balance of the superconducting and normal state energies at the transition. We also found that bigger avalanches are observed in films of lower R_N, pointing to the importance of E_j.

Disorder and granularity may play subtle roles near $H_{c\|}$. Consider a Hamiltonian for a granular film near $H_{c\|}$ with grain charging effects [9,16] being neglected,

$$H = -\sum_{ij} E_J^{ij} S_i S_j - \sum_i \frac{v_i\left[N(0)\Delta^2 - \chi_P H_\|^2\right]}{2} S_i - \sum_i \frac{v_i \chi_P H_\|^2}{2} . \qquad (1)$$

In Eq. (1), S_i equals 1 or 0, in correspondence with the ith grain being in the superconducting state or the normal state, respectively. The first term sums over all nearest neighbors with E_J^{ij} the Josephson coupling energy between the ith and jth grains which reflects quenched disorder. The meaning of the next two terms is clear: They combine to give the superconducting free energy if $S_i = 1$ or the normal state energy if $S_i = 0$.

Equation (1) is an Ising model with both random bonds and random fields. Recently, avalanche dynamics at first-order transitions has been studied at $T = 0$ in the random-field Ising model [17] as well as in the random-bond Ising model [18,19]. The 2D random-bond model [19], in particular, predicts that the avalanche size distribution follows a power-law form, $D(s) \sim s^{-\tau}$ with $\tau \sim 1.45$. There might be a connection between our system and this model. Our system is unique in that the relative importance of the random-field disorder can be tuned. Near $H_{c\|}$, one can tune $H_\|$ such that the random-field term is almost zero, making the random-bond term the dominant term in Eq. (1). However, the model [19] is limited to field sweeps at $T = 0$, while our data were taken during relaxation at finite T.

The stretched-exponential relaxation in Fig. 3 could also be related to the granularity of the system. Near $H_{c\|}$, a grain can be either superconducting or normal depending on its Josephson coupling energies with its neighbors. However, the grains cannot make their choice freely. This is because a superconducting grain favors to have an even number of electrons to avoid the energy penalty of Δ for an unpaired electron [20], while normal grains prefer to be neutral. There must be a charge redistribution on the grains near the transition, a process that has to overcome a grain charging barrier [16] which is of order of 0.1 to 0.2 K for $R_N \sim R_Q$ [9]. We found that relaxations are slower for films of higher R_N, pointing to the importance of E_c which increases with increasing R_N. This charging barrier should give rise to frustration near $H_{c\|}$. Random coupling and frustration could result in the observed slow, stretched-exponential relaxations [1], suggesting a new glassy state for an ultrathin granular superconducting film near $H_{c\|}$, namely a Josephson glass.

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[1] K. Binder and A.P. Young, Rev. Mod. Phys. **58**, 801 (1986).
[2] E. Vives *et al.*, Phys. Rev. Lett. **72**, 1694 (1994).
[3] K.L. Babcock and R.M. Westervelt, Phys. Rev. Lett. **64**, 2168 (1990); P.J. Cote and L.V. Meisel, Phys. Rev. Lett. **67**, 1334 (1991).
[4] H.M. Jaeger, C.-h. Liu, and S.R. Nagel, Phys. Rev. Lett. **62**, 40 (1989).
[5] M.P. Lilly, P.T. Finley, and R.B. Hallock, Phys. Rev. Lett. **71**, 4186 (1993).
[6] D. Dominguez *et al.*, Phys. Rev. Lett. **67**, 2367 (1991).
[7] P. Fulde, Adv. Phys. **22**, 667 (1973).
[8] P.M. Tedrow and R. Meservey, Phys. Rev. B **8**, 5098 (1973).
[9] W. Wu and P.W. Adams, Phys. Rev. Lett. **73**, 1412 (1994).
[10] W. Wu and P.W. Adams, Appl. Phys. Lett. **65**, 1180 (1994).
[11] P.W. Adams and W. Wu (to be published).
[12] T.L. Meisenheimer and N. Giordano, Phys. Rev. B **39**, 9929 (1989).
[13] P.A. Lee and T.V. Ramakrishnan, Rev. Mod. Phys. **57**, 287 (1985).
[14] P. Bak, C. Tang, and K. Wiesenfeld, Phys. Rev. Lett. **59**, 381 (1987).
[15] H.M. Jaeger *et al.*, Phys. Rev. B **40**, 182 (1989).
[16] B. Abeles, Phys. Rev. B **15**, 2828 (1977).
[17] J.P. Sethna *et al.*, Phys. Rev. Lett. **70**, 3347 (1993).
[18] P. Shukla and T.K. Sinha, Phys. Rev. E **49**, R4811 (1994).
[19] E. Vives and A. Planes, Phys. Rev. B **50**, 3839 (1994).
[20] M.T. Tuominen *et al.*, Phys. Rev. Lett. **69**, 1997 (1992).