Glassy Vortex Dynamics Induced by a Random Array of Magnetic Particles above a Superconductor

Young Sun* and M. B. Salamon
Department of Physics and Materials Research Laboratory, University of Illinois at Urbana–Champaign, Urbana, Illinois 61801, USA

K. Garnier and R. S. Averback
Department of Materials Science and Engineering and Materials Research Laboratory, University of Illinois at Urbana–Champaign, Urbana, Illinois 61801, USA
(Received 2 September 2003; published 4 March 2004)

The magnetic relaxation of a Nb film covered with a random array of permalloy particles has been studied using various procedures. When the sample undergoes a field-cooled process, the magnetic relaxation becomes logarithmic in time. The relaxation rate is nearly temperature independent at low temperature and characteristic glassy dynamics—aging and memory effects—are observed. These results are interpreted as the consequence of pinning by the statistical variation of the number of nanoparticles within the area of a vortex core.

DOI: 10.1103/PhysRevLett.92.097002 PACS numbers: 74.25.Qt, 74.78.Db, 75.50.Lk, 75.75.+a

Flux pinning and vortex dynamics in type-II superconductors have been an active research field for many years because of their relevance for applications and for fundamental physics. The response of flux lines to the presence of artificial pinning centers has, consequently, become an attractive topic. Various artificial pinning structures, such as arrays of holes (antidots) [1,2], of nonmagnetic dots [3], and of magnetic dots [4–8], have been studied. When the artificial pinning centers are arranged in a periodic lattice, pronounced commensurability effects take place, as revealed in magnetization [1], resistivity [4], and critical current density [8] measurements. Among these artificial pinning structures, magnetic dots/particles are of peculiar interest because of the additional magnetic nature of the pinning centers. A variety of interesting and unusual effects, such as the asymmetric flux pinning [8], the reconfiguration of the vortex lattice [5], and magnetic-field-induced superconductivity [9], have been observed.

In this work, rather than a regular array of large magnetic dots ($d > 100$ nm) as in most of the previous work, we investigate the effects of a random array of nanoscale magnetic particles ($d < 10$ nm) as pinning centers on a Nb thin film. We focus on the magnetic relaxation behavior because it can provide illuminating information on the nature of the vortex lattice. It is found that the relaxation behavior depends on the cooling history, namely, zero-field cooling (ZFC) or field cooling (FC). Moreover, a nearly temperature-independent relaxation rate and aging and memory effects are observed after FC, but not following ZFC. These results suggest that the magnetization state of the magnetic particles, frozen into alignment in the FC process, plays a crucial role in the flux pinning and that a random array of spin-aligned magnetic particles can induce a "glassy state" in conventional superconductors.

The sample in this study is a thin Nb film covered with a monolayer of permalloy ($Ni_{81}Fe_{19}$) nanoparticles. Nb films with a thickness of 100 nm were prepared by dc magnetron sputtering onto Si(100) substrates that are held at liquid nitrogen temperature during deposition. Some of the as-prepared Nb films were then put into an inert gas condensation system as substrates to receive permalloy particles. A 2 in. magnetron sputtering gun with a permalloy target was used to create a plasma. A high pressure of inert gas causes the sputtered atoms to nucleate and form clusters. The clusters are then extracted by a pressure difference through a series of apertures and collected on the Nb films placed in this path. By controlling the sputtering time we can make the clusters form a monolayer of nanoparticles with the required density. Figure 1(a) shows a TEM image of the magnetic particles. The particles have spherical shape and the size distribution ranges from 3 to 8 nm. These particles are randomly dispersed on the Nb film surface, forming an irregular array of magnetic particles. Magnetization measurements were performed by using a Quantum Design superconducting quantum interference device (SQUID) magnetometer from 1.8 to 300 K. The magnetic field is applied perpendicular to the film surface.

The superconducting transition temperature of the sample as well as a pure Nb film was checked by ac susceptibility measurements with zero-field cooling. Both of them have a T_c of 8.0 ± 0.1 K, which implies that no apparent macroscopic reduction of T_c occurs when the particles are not magnetized. As shown in Fig. 1(b), the $M(H)$ curve of the sample exhibits unusual shape below T_c. When temperature is not far below T_c, the $M(H)$ curve shows several dips, which are absent in a pure Nb film [10]. These dips are not regular in magnetic fields, maybe due to the randomness of the particle array.

 © 2004 The American Physical Society

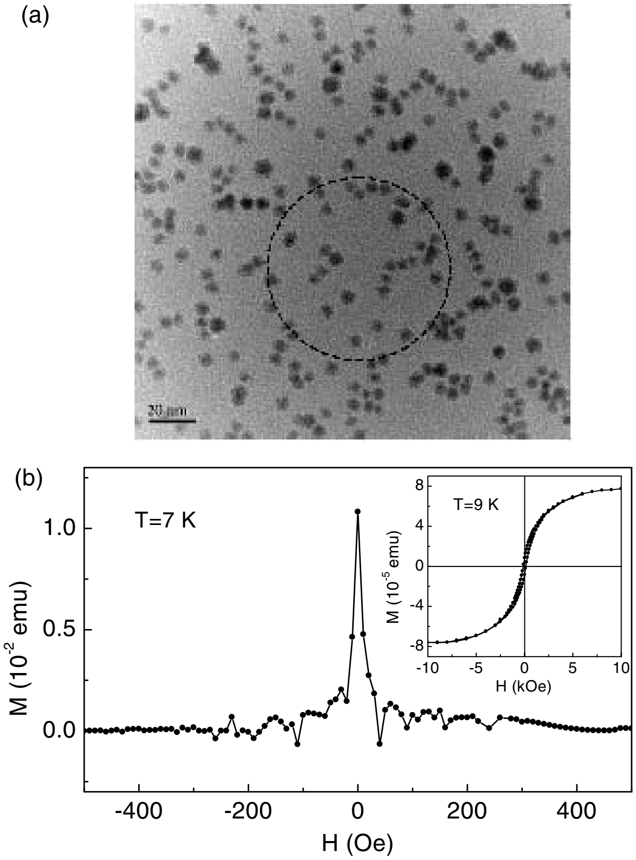

FIG. 1. (a) A TEM image of the magnetic particles, captured on a grid during deposition on Nb. The circle approximates the area of a vortex core. (b) M-H curves of the sample at 7 and 9 K (inset).

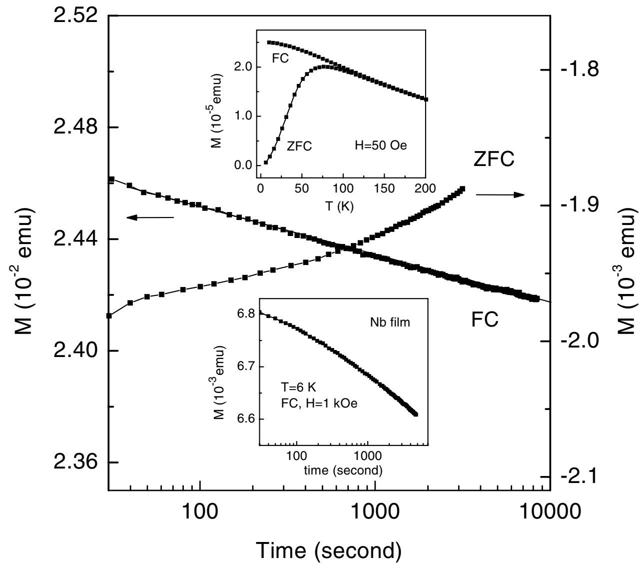

FIG. 2. Magnetic relaxation of the sample at 6 K for both the ZFC and FC processes. The FC relaxation is logarithmic in time while the ZFC relaxation is much faster. The upper inset illustrates the difference between ZFC and FC magnetization for a typical magnetic nanoparticle system. The lower inset shows the magnetic relaxation of a Nb film without magnetic particles.

Above T_c, the sample exhibits typical superparamagnetic behavior of ferromagnetic particles [10].

To study the effects of the magnetic particles on the vortex dynamics, we performed magnetic relaxation measurements with both ZFC and FC processes. In the ZFC measurements, the sample is cooled from above T_c to the studied temperatures; after the temperature is stable, a 500 Oe field, which is below the upper critical field of the sample up to 7 K, is applied and the magnetization is recorded as a function of time. In the FC measurements, the sample is cooled in a 1 kOe field from 80 K (far above the blocking temperature of the permalloy particles) to the measuring temperatures; after the temperature is stable the field is cut off and the remanent magnetization is measured with time. In Fig. 2, we show the typical magnetic relaxation curves of the sample below T_c. At each temperature, the relaxation for the FC process is logarithmic with time. In contrast, the relaxation for the ZFC process is not logarithmic with time, but rather much faster than a logarithmic function, with a larger fractional change even though the applied field is only half of that in the FC process. Since the magnitude ($\gg 10^{-4}$ emu) of these relaxations is about 3 orders larger than the magnetic relaxation ($\sim 10^{-7}$–10^{-6} emu) of the particle moments, it reflects only the flux creep in the superconductor. The distinctive relaxation behaviors between the ZFC and FC processes suggest that the magnetization state of the magnetic particles plays a crucial role in the flux pinning. It is well known that the magnetization state of a magnetic particle system depends on the cooling process, as illustrated in the upper inset of Fig. 2. During the FC process the moments of the particles are aligned along the field direction, whereas the moments are frozen in random directions after ZFC. The slower logarithmic relaxation for the FC process compared with the ZFC process implies that the pinning is more effective when the particle moments are aligned. This can be further confirmed by measuring a pure Nb film. As shown in the lower inset of Fig. 2, the magnetic relaxation of a pure Nb film with the same FC process is not logarithmic but faster than it. Only when the temperature is low enough ($T < 3$ K) does it approach logarithmic relaxation, which can be understood within the Anderson-Kim model in the low temperature limit [11].

Because the relaxation is logarithmic with time for the FC process, we can uniquely determine the normalized relaxation rate defined as

$$S = \frac{1}{M_0}\frac{dM}{d\ln t} \quad \text{or} \quad R = \frac{d\ln M}{d\ln t} \tag{1}$$

and study its temperature dependence. The temperature dependences of S and R from 7 K down to 1.8 K are

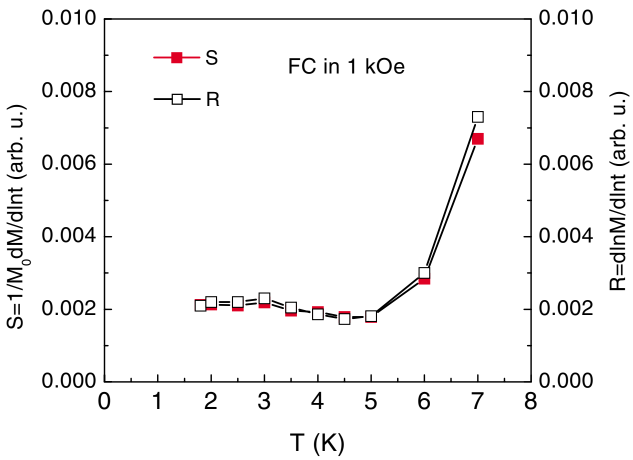

FIG. 3 (color online). Temperature dependence of the normalized magnetic relaxation rate $S = (1/M_0)(dM/dt)$ and $R = d\ln M/d\ln t$ for the FC process. At low temperatures, both S and R are nearly temperature independent.

plotted in Fig. 3. Both give consistent temperature dependence. For a conventional relaxation process by thermal activation over a characteristic energy barrier, one would expect the relaxation rate to decrease toward zero with decreasing temperature. However, a surprising result in our sample is that the relaxation rate is almost temperature independent at low temperature (Fig. 3) and it definitely extrapolates to a nonzero value at vanishing temperature. A nonzero constant relaxation rate has been reported in some superconductors and was usually interpreted as evidence for quantum tunneling of vortices [12,13]. However, quantum tunneling is unlikely to occur at such high temperatures for a conventional superconductor like Nb. There must be other physics underlying these phenomena. In fact, recent theoretical work based on a so-called "restricted occupancy model" has suggested that a nonzero constant relaxation rate can be the natural consequence of the off-equilibrium dynamics in a classical glassy system, giving rise to logarithmically slow relaxation and other glassy phenomena [14]. We discuss our results in light of this model below.

The most convincing evidence for a glassy state is the appearance of the characteristics of glassy dynamics—aging and memory effects—such as have generally been observed in spin glasses and other glassy systems. Experimental demonstration of both aging and memory effects for superconductors is usually achieved by performing the so-called "field-jump" experiments, which were first adopted for high temperature superconductors [15]. In such experiments, the sample is first cooled in a field H_0 to a studied temperature below T_c; the system is then left in its quasiequilibrium state for a certain waiting time t_w before the applied field is increased by ΔH to the measuring field H_1. Just at this time does one start measuring the relaxation curve $M(t)$. The aging and memory effects are characterized by the appearance of a strong dependence of $M(t)$ on the waiting time t_w and an inflection point in the M vs $\ln t$ plot at $t = t_w$.

To explore possible aging and memory effects, we applied this experimental procedure to our sample. Figure 4(a) shows the results obtained at 6 K. The sample is first cooled in $H_0 = 200$ Oe to 6 K; after the temperature is stable, we wait for a time $t_w = 1$ or 2 h; then the field is changed to $H_1 = 400$ Oe and the magnetization is recorded with time from this moment. From Fig. 4, it is apparent that the relaxation with $t_w = 1$ h differs from that with $t_w = 2$ h, which reflects the fact that aging occurs during the waiting time. In each measurement, after an initial transient response ($t < 2$ min) M relaxes logarithmically with time. However, just around a measuring time t equivalent to the waiting time t_w (indicated by arrows in Fig. 4), the relaxation starts to deviate from $\ln t$. Such an echolike response is called a "memory effect" [15]. We also performed similar measurements at 1.8 K with higher magnetic fields ($H_0 = 2$ kOe and $H_1 = 3$ kOe). The results are shown in Fig. 4(b). Clearly, the relaxation depends on the waiting time t_w and an inflection point appears at $t = t_w$. Therefore, these results successfully demonstrate the aging and memory effects in the studied sample. For comparison, we also performed the same measurements on a pure Nb film. However, no appreciable memory effects like those in Fig. 4 occur in a

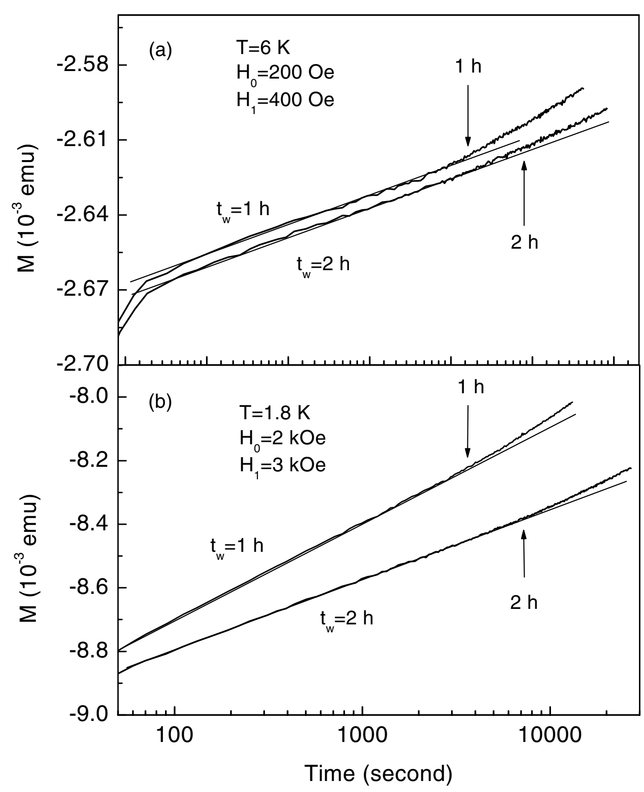

FIG. 4. Aging and memory effects at (a) $T = 6$ K, and (b) $T = 1.8$ K. The arrows indicate when the measuring time equals the waiting time. The solid lines are linear fits.

Nb film without magnetic particles, confirming that the glassy phenomena are introduced by the magnetic particles.

Pinning of vortices by mesoscopic magnetic dots is well documented [4,5]. Rather than dots, large on the scale of the vortex core, we have here a random distribution of nanoscale particles with an areal density ρ and magnetic moment μ_p per particle. Within each vortex core we expect, on average, $N = \pi\xi^2\rho$ particles at low temperatures, with a statistical variation $\sqrt{N}$ (see Fig. 1). It is the spatial variation of magnetic moment $\mu_p\sqrt{N}$ that provides the pinning energy [5]

$$E_p = \frac{\Phi_0 \mu_p \sqrt{N}}{2\pi\lambda^2} K_0\left(\frac{\xi}{\lambda}\right). \tag{2}$$

Here $\lambda = 39$ nm is the penetration depth and $\xi = 38$ nm the coherence length of Nb; Φ_0 is the flux quantum. We have fit magnetization data for similar particles on Si and find a moment of $6000\mu_B$, consistent with 6 nm particles and 25% coverage. If all particles were aligned, the maximum pinning energy would be $E_p = 3.2 \times 10^{-19}$ J. This is to be compared with the vortex-vortex energy [16] between rigid vortex lines of length $\ell = 100$ nm and spacing $L = \sqrt{2\Phi_0/B\sqrt{3}}$ at $B = 0.1$ T,

$$F_{vv} = \frac{\Phi_o^2 \ell}{2\pi\mu_o\lambda^2} K_0\left(\frac{L}{\lambda}\right) = 4.2 \times 10^{-19} \text{ J}. \tag{3}$$

The case of nearly equal pinning and vortex-vortex energy scales was treated by Nicodemi and Jensen [14]. The frozen magnetic moment measured at 0.1 T in our sample is less than one-third the saturation value while the model assumes an average pinning energy of half the maximum. Nonetheless, we expect to find temperature-independent logarithmic relaxation. The key is that the vortex system finds itself far from equilibrium over the observation time and therefore explores collectively a relatively flat energy landscape with typical glassy behavior as the consequence.

In conclusion, a Nb film covered with a random array of magnetic particles shows interesting relaxation behaviors. The magnetization state of the particles plays a crucial role in the flux pinning. When the sample is cooled in a magnetic field, logarithmic relaxation, temperature-independent relaxation rate, as well as aging and memory effects appear. All of these results suggest that a random array of aligned small magnetic particles induces a "glassy region" in conventional low-T_C superconductors. It will be worthwhile in future work to explore whether a random array of big magnetic dots (comparable to the size of a vortex core) would lead to similar glassy dynamics. Although these experimental results are currently interpreted as the consequence of pinning by the statistical variation of the number of nanoparticles within the area of a vortex core, it should be noted that there could be other contributions to the observed glassy dynamics. Since the pinning strength depends on the magnetization of particles, the relaxation of particle magnetization itself may cause appreciable change in flux dynamics. This may be true especially when temperature is not low enough. However, the glassy dynamics in our sample persists to 1.8 K where the relaxation of particle magnetization is essentially too weak to be relevant during the measuring time. This indicates that the contribution from relaxation of particle magnetization, if existing, should not be a dominant origin. Future work will examine the I-V characteristics of these samples to further explore vortex pinning and dynamics.

This work was supported by the National Science Foundation under Grant No. EIA-0121568 and in part by the U.S. Department of Energy, Basic Energy Sciences under Grant No. DEFG02-91-ER45439. Some of the experiments were carried out in the Center for Microanalysis of Materials, University of Illinois, which is partially supported by the U.S. Department of Energy under Grant No. DEFG02-91-ER45439.

*Present address: Department of Physics and Astronomy, Rice University, Houston, TX 77005, USA.

[1] M. Baert, V.V. Metlushko, R. Jonckheere, V.V. Moshchalkov, and Y. Bruynseraede, Phys. Rev. Lett. **74**, 3269 (1995).
[2] A.V. Silhanek, S. Raedts, M. Lange, and V.V. Moshchalkov, Phys. Rev. B **67**, 064502 (2003).
[3] A. Hoffmann, P. Prieto, and Ivan K. Schuller, Phys. Rev. B **61**, 6958 (2000).
[4] J. I. Martín, M. Vélez, J. Nogués, and Ivan K. Schuller, Phys. Rev. Lett. **79**, 1929 (1997).
[5] J. I. Martín, M. Vélez, A. Hoffmann, Ivan K. Schuller, and J. L. Vicent, Phys. Rev. Lett. **83**, 1022 (1999).
[6] J. I. Martín, M. Vélez, A. Hoffmann, Ivan K. Schuller, and J. L. Vicent, Phys. Rev. B **62**, 9110 (2000).
[7] O. M. Stoll, M. I. Montero, J. Guimpel, Johan J. Akerman, and Ivan K. Schuller, Phys. Rev. B **65**, 104518 (2002).
[8] David J. Morgan and J. B. Ketterson, Phys. Rev. Lett. **80**, 3614 (1998).
[9] M. Lange, M. J. Van Bael, Y. Bruynseraede, and V.V. Moshchalkov, Phys. Rev. Lett. **90**, 197006 (2003).
[10] Y. Sun, M. B. Salamon, K. Garnier, and R. S. Averback (unpublished).
[11] P.W. Anderson and Y. B. Kim, Rev. Mod. Phys. **36**, 39 (1964).
[12] A. Hamzic, L. Fruchter, and I. A. Campbell, Nature (London) **345**, 515 (1990).
[13] Y. Yeshurun, A. P. Malozemoff, and A. Shaulov, Rev. Mod. Phys. **68**, 911 (1996).
[14] Mario Nicodemi and Henrik Jeldtloft Jensen, Phys. Rev. Lett. **86**, 4378 (2001).
[15] C. Rossel, Y. Maeno, and I. Morgenstern, Phys. Rev. Lett. **62**, 681 (1989).
[16] M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York, 1975), p. 149.