RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 48, NUMBER 13 1 OCTOBER 1993-I

Nature of hysteresis in glass transitions of the first order

V. B. Geshkenbein
Landau Institute for Theoretical Physics, Moscow, Russia

L. B. Ioffe*
Department of Physics, Rutgers University, Piscataway, New Jersey 08855

A. I. Larkin*
AT&T Bell Laboratories, Murray Hill, New Jersey 07974
(Received 28 May 1993)

The distribution of relaxation times in a glass is very broad. As a result the free energy of a glass slowly approaches its equilibrium value but never reaches it. This slow relaxation explains why, in the hysteresis cycle, the melting temperature is higher than the solidification temperature. We estimate the width of this hysteresis for a general type of transition into the glassy state. We apply these estimates to the special case of the vortex melting transition and find good agreement with recent experiments.

(a) Introduction. Recently Safar *et al.*[1] reported detailed measurements of flux lattice melting in high-T_c superconductors in fields $H_{c1} \ll H \ll H_{c2}$ on a very clean sample. They observed considerable hysteresis as well as discontinuity of resistivity at a certain temperature. It is natural to interpret this temperature as a first-order melting transition in the flux lattice. This interpretation was recently confirmed by the measurements of the angular dependence of the melting temperature.[2]

The "solid" flux phase formed below this transition temperature lacks long-range order: it is destroyed by weak disorder.[3] This phase has glassy properties: it has many metastable states separated by large energy barriers. Since some barriers can be arbitrarily large, this state never reaches the thermal equilibrium.[4–9] In the absence of disorder the melting of the solid flux phase is a first-order transition.[10] The melting transition is affected slightly by a weak disorder; it remains a sharp first-order transition.[11,12] A character of the melting transition may be changed by a strong disorder completely.[11–14] A relative strength of disorder depends on the applied magnetic field, so both regimes may be realized in the same sample in different fields. In the conditions of the experiment[1] the disorder is not sufficiently strong to change the character of the transition.[15] We shall consider only the limit of weak disorder, which is realized in the experiment.[1]

The glassy properties of the solid result in the novel mechanism of hysteresis at the melting transition. The free energy of a glass approaches its equilibrium value very slowly. Melting happens when the free energy of the liquid state equals the free energy of the solid state. Consider the hysteresis cycle through solidification and melting. We start in the liquid state and begin to lower the temperature until solidification occurs. The free energy of the nascent solid is determined by the time scale of the solidification process. After solidification the state continues to approach equilibrium and its free energy decreases. Next, we raise the temperature of the solid back to the former solidification point. But now the free energy of the solid is lower than the free energy of the liquid; hence no transition takes place until higher temperature.

The mechanism for hysteresis is not specific to the flux lattice, but may happen in all glasses displaying first-order phase transitions. The specific property of this hysteresis is that its width depends on the time the system spends in the glass phase (aging time). Such dependence was observed in a recent experiment.[16]

In spin glasses the first-order transition usually happen when the spin glass coexists with a ferromagnet or an antiferromagnet. In this case the order parameter is likely to jump when the external temperature or magnetic field is varied (spin flip transition). The other well known example of a first-order transition in spin glasses with local ferromagnetic ordering is the jump in magnetization that occurs when the direction of the applied field is reversed.[17]

(b) Barrier dynamics in glassy state. In glasses the distribution of barriers between metastable states is very broad. At any particular moment of time the glass is in some metastable state. The free energy of this state contains the entropy of the thermal fluctuations that occur on a time scale shorter than the aging time. There are other metastable states in which the free energy is lower but those states are separated from this state by large barriers. With time the glass surmounts larger and larger barriers and buries itself into deeper states. We consider only the barriers leading to deeper states. Let us denote by $P(U)dUU^{-1}$ the density of these barriers with heights in the interval $(U, U+dU)$. Neglecting very rare processes in which the system returns from a deep state back to a shallow one, we get

$$\frac{dP(U)}{dt} = -\tau_0^{-1} \exp(-U/T) P(U), \tag{1}$$

where τ_0^{-1} is the microscopic attempt rate.

Solving this equation we find the rate at which the

barriers disappear:

$$\frac{dP(U,t)}{dt} = -\tau_0^{-1} e^{-(\exp(-U/T)\, t/\tau_0 + U/T)} P_0(U), \quad (2)$$

where $P_0(U)$ is the initial distribution of barriers. This distribution is broad and smooth in the glassy state. At large time scales $t \gg \tau_0$ the rate (2) has a sharp maximum at a typical barrier height

$$U_t = T \ln(t/\tau_0). \quad (3)$$

The density of barriers can be conveniently described by the "volume" $V(U) = 1/P(U)$ per barrier. We assume that the function $V(U)$ is characterized by one energy scale U_c, by one volume scale V_c, and has a scaling form at large U:

$$V(U) = V_c v(U/U_c), \quad v(x) = x^\nu \text{ at } x \gg 1, \quad v(0) = 1. \quad (4)$$

The function $V(U)$ does not depend weakly on the way in which the glass state was prepared.

At short scales the vortex glass forms a unique ordered state, with no metastability. The transitions between metastable states happen when part of the vortex lattice jumps from one position to another. There is a minimal volume that can jump; this volume plays the role of V_c in (4). The barrier between the states at these scales is U_c. In spin glasses the minimal volume coincides (away from transition temperature) with the unit cell volume; the energy barrier at these scales is simply the exchange interaction constant.

A jump of a larger volume must overcome a larger barrier. In the case of pinning of one-dimensional (1D) structures[18] (single vortex line, dislocation, etc.) the length of the jumping part of the structure plays the role of the volume $V(U)$. The structure of the metastable states in 3D vortex glasses at large scales can be more complicated and such a simple picture may not hold.

In spin glasses the transitions between large metastable states can be visualized as a magnetization flip in a spin cluster. In general this cluster may have a fractal dimension and many clusters may overlap strongly as they do in some spin glass models.[19] In a phenomenological approach to the spin glass dynamics[4,5] one assumes that the properties of these clusters at large scales can be described by a set of scaling exponents. A similar scaling approach was applied to the vortex glass problem by Fisher, Fisher, and Huse[8] at scales larger than the pinning length.

Each transition over the barrier decreases the free energy of the glass by some amount E. The transition over the minimal barrier U_c decreases the energy by roughly the same energy $E \sim U_c$. In the 1D case, this energy is of the order of the barrier energy at larger scales as well. We assume that this decrease always correlates with the energy of the barrier, i.e., that E is a function of U and that this function also has a scaling form:

$$E(U) = U_c e(U/U_c),$$
$$e(x) = e_0 x^\beta \text{ at } x \gg 1, \quad e(0) \sim 1, \quad e_0 \sim 1. \quad (5)$$

The theory of collective pinning assumes[9] that the exponent $\beta = 1$ for the vortex glass. Combining the transition rate (2) with the scaling estimates (4), (6), we get the rate at which the free energy of the glass decreases with time:

$$\frac{dF}{dt} = \int \frac{dP(U)}{dt} E(U) \frac{dU}{U} = -\frac{E(U_t)}{V(U_t) t \ln(t/\tau_0)}, \quad (6)$$

where we performed the integral over U in the saddle point approximation.

To find the change in free energy during glass aging we have to integrate the rate (6) from time scales of solidification t_1 to the aging time t_2. The time scale of solidification is determined by experimental conditions. In most experiments both times scales t_1 and t_2 are macroscopic, whereas τ_0 is the microscopic rate of attempts. In this case it is likely that $\ln(t_2/t_1) \ll \ln(t_1/\tau_0)$ and the change in free energy is

$$\Delta F = -\frac{E(U_t) \ln(t_2/t_1)}{V(U_t) \ln(\sqrt{t_1 t_2}/\tau_0)}. \quad (7)$$

If the aging time is so long that $\ln(t_2/t_1) \sim \ln(t_1/\tau_0)$, we estimate the change in the free energy only in the limiting cases of (i) very large barriers $U_c \gg T \ln(t_2/\tau_0)$ and (ii) very small ones $U_c \ll T \ln(t_1/\tau_0)$. In the first case all energy distributions are determined by a single scale U_c:

$$\text{(i) } \Delta F = -\frac{U_c}{V_c} \ln\left(\frac{\ln(t_2/\tau_0)}{\ln(t_1/\tau_0)}\right). \quad (8)$$

In the second case we use the scaling estimates (6, 4):

$$\text{(ii) } \Delta F = -\frac{U_c e_0}{V_c(\beta-\nu)} \left(\frac{T}{U_c}\right)^{\beta-\nu} [\ln^{\beta-\nu}(t_2/\tau_0) - \ln^{\beta-\nu}(t_1/\tau_0)]. \quad (9)$$

As a result of the energy decrease during aging, melting occurs at a higher temperature than solidification:

$$\Delta T = \frac{\Delta F T}{q}, \quad (10)$$

where q is the latent heat of the melting transition.

For first-order transitions due to a varying magnetic field, the width of the hysteresis is

$$\Delta H = \frac{\Delta F}{\Delta M}, \quad (11)$$

where ΔM is a jump of magnetization at the transition.

Taken together, formulas (7)–(11) determine the hysteresis width in a first-order transition to a glassy state. The width of the hysteresis increases if $q \to 0$ (or $\Delta M \to 0$). This might happen when the tricritical point is approached. However, in all second-order transitions into the glass states known to us, the width of the distribution of metastable states goes to zero at the transition as a high power of the order parameter. So, the ratio (10),(11) actually tends to zero when the tricritical point is approached.

(c) Estimates of the hysteresis width in the vortex melting transition. Now we apply the general formu-

las derived above to the melting of the vortex lattice as observed in experiment.[1] First, we check the conjecture that the discontinuity of resistivity in this experiment is due to the melting of the vortex lattice. We use Lindemann melting criteria, which relates the thermal mean-square displacement of the vortices at the melting temperature to the distance a between vortices: $\sqrt{\langle u_T^2 \rangle} = c_L a$, where c_L is the Lindemann number. We estimate the thermal mean-square displacement of the vortices by employing the Debye approximation to replace the sum over the whole Brillouin zone by an integral over a sphere, chosen to contain the correct number of phonons: $T_m = 16 C_{66} c_L^2 a^3 \epsilon$,[20,21] where $\epsilon = \lambda_{ab}/\lambda_c$ is a parameter characterizing the anisotropy of the material and C_{66} is the shear modulus of the vortex lattice. To estimate the shear modulus C_{66} we express it through the penetration depth λ: $C_{66} = \Phi_0 B/(8\pi\lambda)^2$.[9] Extracting the Lindemann number from this equation we get $c_L = 0.15$ which is in a very good agreement with the values obtained in other experiments or numerical simulations.[9]

Below we show that in the samples used in this experiment the pinning length R_c is sufficiently long: $R_c \gg a$. However, it is still shorter than penetration depth $\lambda \gg R_c$, which is very large in the high-T_c materials, especially in the vicinity of T_c. In this pinning regime, the pinning length along the direction of the field L_c is related to the pinning length in the transverse direction R_c by $L_c = \epsilon R_c^2/a$. For $YB_2Cu_3O_7$ we have $\epsilon^{-2} \approx 60$.[22] Thus, the minimal volume that can jump between metastable states is $V_c = \epsilon R_c^4/a$. The energy scale of these jumps is determined by the elastic energy of the lattice deformations at these scales: $U_c = C_{66}\xi^2 L_c \sim (T_m/16c_L^2)(\xi^2 R_c^2/a^4)$, where ξ is the coherence length of the superconductor.

To estimate U_c we represent it as a product of two terms $U_c = [\epsilon C_{66}\xi^4(T)/a][(R_c/\xi)^2]$ and estimate them separately:

$$\frac{\epsilon C_{66}\xi^4(T)}{a} = \frac{\epsilon \Phi_0^{5/2} B^{3/2}}{(16\pi^2)^2 \lambda(T)^2 H_{c2}^2(T)} \approx \frac{T_c}{32\pi\sqrt{\pi Gi}} \left(\frac{B}{H_{c2}(0)}\right)^{3/2} \frac{T_c}{T_c - T}, \quad (12)$$

where $Gi = [T_c/H_c^2(0)\epsilon\xi^3(0)]^2/2$ is the Ginzburg number, and $H_c(0)$, $\xi(0)$, $H_{c2}(0)$ are linear extrapolations to $T = 0$. For $YB_2Cu_3O_7$ we estimate (12) $\epsilon C_{66}\xi^4(T)/a \approx 0.2$ K at $T = 80$ K in fields $H = 6$ T applied in the experiment.[1] Lastly, to estimate U_c we need the value of the pinning length R_c. Let us define the critical current j_c as the current which suppresses the pinning completely: at currents $j > j_c$ the resistivity reaches its flux flow value. The pinning length can be determined from the expression for the critical current: $j_c = j_0(\xi/R_c)^2$, where j_0 is the depairing current:

$$j_0 = \frac{cH_{c2}}{6\sqrt{3}\pi\kappa\lambda}. \quad (13)$$

For $YB_2Cu_3O_7$ we estimate $j_0(0) = 10^8$ A/cm^2 and $j_0(T_m) \approx 3\ 10^6$ A/cm^2. Using the value $j_c = 10^3$ A/cm^2 we find that $U_c/T \sim 10$ in the regime of the experiment.[1] This estimate of the pinning energy means that for the experimentally accessible time scales [$\ln(t_2/\tau_0) \sim 20$] the most important are the barriers that are somewhat larger than U_c.

The width of the hysteresis (10) is also determined by the latent heat of melting. The only energy scale involved in melting is set by the melting temperature T_m and the only spatial scale is the lattice spacing, so $q = \zeta T_m/\epsilon a^3$, where ζ is a numerical coefficient. A reliable value of this numerical coefficient is not known. Earlier numerical simulations show that this value is small $\zeta \approx 0.3$–0.5.[23,24] However, simulations performed on larger lattices show that the transition becomes split into two.[25] The melting of the lattice happens at the lower transition. The specific heat does not show any observable anomaly at this temperature, indicating that this coefficient is very small. This smallness is likely to be a consequence of a numerically small difference between the energies of quadratic and triangular lattices. We also performed rough analytic estimates of the entropy associated with this transition and get $\zeta \sim 0.15$. Combining these estimates and assuming that $U_c/T \approx \ln(t_1/\tau_0) \geq \ln(t_2/t_1)$ so that (7) can be applied, we get the width of the hysteresis:

$$\frac{\Delta T}{T} \approx \zeta^{-1} \ln(t_2/t_1) \frac{\epsilon a^3}{V_c} \approx \zeta^{-1} \ln(t_2/t_1) \left(\frac{a}{\xi}\right)^4 \left(\frac{j_c}{j_0}\right)^2, \quad (14)$$

which gives $\Delta T/T \sim 10^{-3}$ in reasonable agreement with the experiment.[1]

If an electrical current flows, large energy barriers disappear: $U \leq U(j)$. Since the dissipation of the current is limited by the transitions over these barriers, the resistivity $R \propto \exp[-U(j)/T]$. In a scaling regime the energy of the largest remaining barrier is related to the current by $U = U_c(j_c/j)^\mu$. The value of the exponent μ extracted from the experimental data is small: $\mu \approx 0.2$.[26] The absence of large barriers means that there are no processes which take very long. All equilibration processes are cut off at the time scale $\tau_0 \exp[U(j)/T]$. When τ_0 becomes less than the solidification time t_1, the vortex glass reaches the steady state during the solidification and hysteresis disappears. Assuming that $\ln(t_1/\tau_0) \approx 2U_c/T$ we estimate that the current sufficient to smear the hysteresis completely should be $j_s \approx 0.03 j_c$. In this estimate we assumed that the scaling formula for $U(j)$ is still valid for $U/U_c \sim 2$. It is possible to avoid this assumption and extract this current from the magnetization experiment. The remanent magnetization of the sample is produced by the current j_M that does not decay on the time scale of t_1: $U(j_M) = T\ln(t_1/\tau_0)$, this current measured at the temperature just below the melting coincide with the current that smears the hysteresis: $j_M \approx j_s$.

The longer the time of the aging (or the smaller is the current) the larger is the hysteresis width: at longer aging times (or smaller currents) the melting of the solid happens at higher temperature. Neither the aging time nor the current affects the solidification process significantly. This prediction is in complete agreement with the experiment[16] in which the drift of the resistivity curve

obtained during the heating part of the hysteresis cycle was observed when a current was changed, whereas the cooling part of the curve stayed still.

Finally, we discuss briefly the importance of other mechanisms of hysteresis for vortex glass melting. One such mechanism is the nucleation process. The time scale of this process is set by the energy of the nuclei which is determined by the surface energy of the solid-liquid boundary. It is likely that this energy is small in the vortex problem where the energies of all phases are close to each other.[9] Indeed, in the experiment[1] many small steps in the resistivity curve were observed indicating that nucleation is not the limiting process in this experiment. The other such mechanism may be the pinning of the boundary between the glass and the liquid phase. The strength of this pinning is determined also by the properties of the boundary between the glass and the liquid state. For the broad boundary that we expect in this transition, the pinning energy should be small and the effect of the pinning weak. Unfortunately, the theoretical investigation of these effects is plagued by a lack of understanding of the melting transition of the vortex lattice.

We are grateful to D. Bishop, G. Crabtree, P. Gammel, D. Huse, M. Feigel'man, S. Fleisher, W. Kwok, H. Safar, and V. Vinokur for useful discussions and to D. Lidsky for the critical reading of the manuscript.

---

* Also at Landau Institute of Theoretical Physics, Moscow, Russia.

[1] H. Safar *et al.*, Phys. Rev. Lett. **69**, 824 (1992).
[2] W. K. Kwok *et al.*, Phys. Rev. Lett. **69**, 3370 (1992).
[3] A. I. Larkin, Zh. Eksp. Teor. Fiz. **58**, 1466 (1970) [Sov. Phys. JETP **31**, 784 (1970)].
[4] D. S. Fisher and D. A. Huse, Phys. Rev. B **38**, 373 (1988); **38**, 386 (1988); Phys. Rev. Lett. **57**, 2203 (1986).
[5] M. Ocio, J. Hammann, and E. Vincent, J. Magn. Magn. Mater. **90**, 329 (1990); G. J. M. Koper and H. J. Hilhorst, J Phys. (Paris) **49**, 429 (1988).
[6] M. P. A. Fisher, Phys. Rev. Lett. **62**, 1415 (1989).
[7] M. V. Feigel'man *et al.*, Phys. Rev. Lett. **63**, 2303 (1989).
[8] D. S. Fisher, M. P. A. Fisher, and D. A. Huse, Phys. Rev. B **43**, 130 (1991).
[9] For the review of the theory of the vortex states see G. Blatter *et al.*, Rev. Mod. Phys. (to be published).
[10] E. Brezin *et al.*, Phys. Rev. B **31**, 7124 (1985).
[11] Y. Imry and M. Wortis, Phys. Rev. B **19**, 3580 (1979).
[12] V. Pentegov and M. V. Feigel'man, Zh. Eksp. Teor. Fiz. **94**, 345 (1988) [Sov. Phys. JETP **67**, 2154 (1988)].
[13] T. K. Worthington *et al.*, Phys. Rev. B **46**, 11 854 (1992).
[14] A. Bishop *et al.*, Science **255**, 165 (1992).
[15] H. Safar *et al.* (unpublished).
[16] M. Charalambous, Ph.D. thesis, CRTBT, CRNS, Grenoble, 1992; M. Charalambous *et al.*, Phys. Rev. Lett. **71**, 436 (1993); M. Charalambous (private communication).
[17] K. Binder and A. P. Young, Rev. Mod. Phys. **58**, 801 (1986).
[18] L. B. Ioffe and V. M. Vinokur, J. Phys. C **20**, 6149 (1987).
[19] L. B. Ioffe and M. V. Feigel'man, Zh. Eksp. Teor. Fiz. **89**, 654 (1985); J. Phys. (Paris) Lett. **45**, L475 (1984); for a review see V. S. Dotsenko, M. V. Feigel'man, and L. B. Ioffe, Sov. Sci. Rev. A **15**, 1 (1990).
[20] A. Houghton *et al.*, Phys. Rev. B **40**, 6763 (1989).
[21] G. Blatter *et al.*, Phys. Rev. Lett. **68**, 875 (1992).
[22] D. E. Farrell *et al.*, Phys. Rev. Lett. **67**, 1165 (1991).
[23] R. E. Hetzel *et al.*, Phys. Rev. Lett. **69**, 518 (1992).
[24] Y.-H. Li and S. Teitel, Phys. Rev. Lett. **66**, 3301 (1991); Phys. Rev. B **45**, 5718 (1992).
[25] Y.-H. Li and S. Teitel, Phys. Rev. B **47**, 359 (1993).
[26] C. Dekker *et al.*, Phys. Rev. Lett. **68**, 3347 (1992).