Resistive Transitions in Ultrathin Superconducting Films: Possible Evidence for Quantum Tunneling of Vortices

Y. Liu,[a] D. B. Haviland,[b] L. I. Glazman,[c] and A. M. Goldman

Center for the Science and Application of Superconductivity and School of Physics and Astronomy, University of Minnesota, Minneapolis, Minnesota 55455

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Unusual temperature dependence of electrical resistance has been observed in ultrathin superconducting films of Pb, Al, and Bi. In small magnetic fields, at low temperatures, sheet resistances varied with temperature as $R \approx R_0 \exp(T/T_0)$, where T_0 and R_0 are constants. This result is not expected in conventional models of vortex motion involving thermal activation. On the other hand, it may be explained in a quantum tunneling picture.

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The creep and flow of vortices have been subjects of study because they give rise to nonzero electrical resistance even below the superconducting transition temperature. In the classical model of thermally activated creep of vortices the resistance vanishes exponentially at low temperatures with a dependence given by an Arrhenius form [1]. Non-Arrhenius behavior at low temperatures could result from an alternative to thermal processes involving macroscopic quantum tunneling [2]. This phenomenon itself, taking into account dissipation, was first studied by Caldeira and Leggett [3]. Subsequently there have been theoretical [4] and experimental studies focused on small Josephson junctions [5–10] and one-dimensional wires [11]. Although there has been no firm experimental confirmation of macroscopic quantum tunneling of vortices, it has been argued that anomalous temperature dependences found for the rates of decay of remnant magnetizations provide support for the existence of the phenomenon [12,13], and these experiments have stimulated a number of theoretical treatments [14–16].

In this Letter we report on measurements of electrical resistance of ultrathin films, in relatively small magnetic fields, that exhibit an unusual temperature dependence, $R \approx R_0 \exp(T/T_0)$, which cannot result from thermally activated flux creep. Here T is the temperature, R_0 and T_0 are constants. This particular functional form is consistent with a modification of a model of quantum tunneling of vortices in films [2].

Four-terminal electrical measurements were made on ultrathin films of Pb, Bi, and Al prepared by *in situ* deposition of metal onto glazed alumina substrates previously coated with amorphous Ge. All evaporations were carried out at temperatures lower than 18 K. Small amounts of metal were deposited in successive steps producing a set of films with differences in thickness so small that their microstructures hardly changed from one film to another. Films made this way are very thin (about a monolayer for Pb), uniform on macroscopic scale, and are extreme type-II superconductors. They were not warmed above 14 K so as to avoid irreversible annealing. Quoted thicknesses are nominal in that they are derived from readings of a calibrated quartz crystal oscillator monitor which is sensitive to deposited mass per area. Details of the experimental techniques are described elsewhere [17].

Magnetic fields were applied in the direction perpendicular to the film plane using a split-coil superconducting magnet. The low-temperature apparatus was contained within a metal Dewar, and all electrical leads entering the vacuum chamber were filtered with low-pass electromagnetic interference (EMI) filters (Erie No. 1233-006) which were soldered into a conductive shield. In series with these filters, inside the shield, were 20-nH inductors. The combination of filters and inductors exhibited voltage attenuations of 15 dB at 1 MHz, 60 dB at 10 MHz, and finally 75 dB beyond 100 MHz.

In Fig. 1, values of resistance per square, $R(T)$, for various magnetic fields are plotted for a nominally 3.86-Å-thick Pb film. The normal-state resistance had a small negative temperature coefficient ($dR/dT < 0$). The value of $R(14\ \mathrm{K})$ for this film was equal to 6.0 kΩ. The superconducting transition temperature T_{c0}, defined as the temperature at which the resistance was half of $R(14\ \mathrm{K})$, was 1.62 K. The striking result shown in Fig. 1 is that

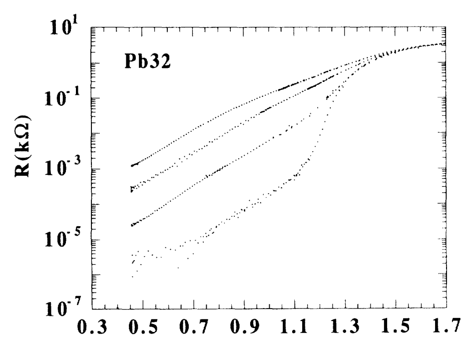

FIG. 1. Sheet resistance R vs temperature T for a Pb film with $T_c = 1.62$ K in various magnetic fields. From top to bottom, $H = 2070$, 945, 98, and 0.76 G. The sheet resistance at a temperature of 14 K is 6.0 kΩ, and the nominal thickness of the film is 3.86 Å. The superconducting transition temperature was chosen as the temperature at which the resistance falls to half of its value at 14 K.

the resistance at low temperatures appears to vary exponentially with temperature in a non-Arrhenius fashion, i.e., $R\sim\exp(T/T_0)$, spanning two to three decades of resistance. The coefficient T_0 is essentially independent of the magnetic field although a 5% to 10% change was found for the highest field (≈ 2100 G). Results very similar to those shown in Fig. 1 were found for various Bi and Al films over ranges of resistance extending from two to five decades.

Values of resistance of the film shown in Fig. 1 are plotted versus magnetic field at $T=0.46$ K in Fig. 2. The resistance is seen to first increase very rapidly for small fields (<100 G) and then increase more gradually as the field is increased further. At higher fields (>300 G), some hysteresis with respect to increasing and decreasing the field is seen, which suggests "freezing in" of vortices. The observed nonlinearity in $R(H)$ at high fields is probably a consequence of vortex unbinding by the applied field which is a strongly nonlinear process [18]. It should be noted that the temperature dependence displayed in Fig. 1 was valid in magnetic fields in both the low- and high-field regimes of Fig. 2.

Films of Pb with various normal sheet resistance in the same deposition sequence were all found to exhibit essentially the same behavior. As the normal sheet resistance was decreased, T_c increased accordingly. However, the characteristic features of the resistive transition as a whole remained unchanged. The approximate temperature associated with the onset of the unusual resistance tail, T_t, was roughly proportional to T_c. The characteristic temperature T_0, on the other hand, was found to be essentially constant over a significant range of normal sheet resistance. This is shown in Fig. 3 where values of T_0 for various Pb films are plotted versus $R(14\text{ K})$. Here the thicknesses were 3.48 and 4.25 Å for the most and least resistive films, respectively. In principle, a similar analysis could have been carried out on the Bi and Al films. For the Bi films, which were the focus of an investigation of the thickness dependence of the conductance, magnetoconductance data were too limited to permit a systematic study of the dependence of T_0 on R. The values of T_0 for the Al films varied in a random manner over the range $0.05\text{ K}\le T_0\le 0.102$ K, and exhibited no systematic variation of T_0 with R. This we believe to be a consequence of inhomogeneities of these films possibly due to their being heated during deposition as will be discussed below.

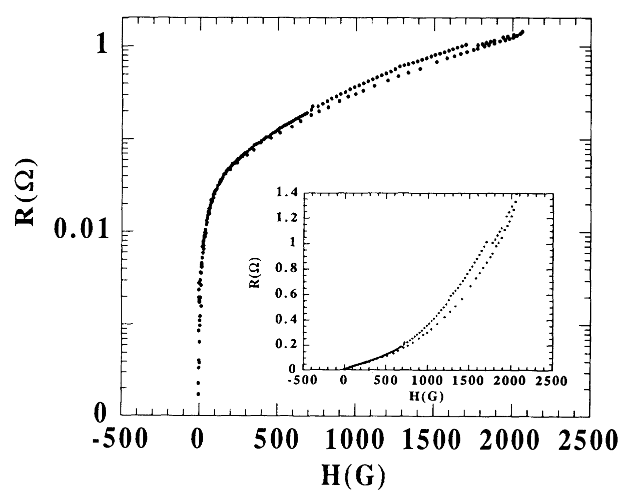

FIG. 2. Logarithm of the sheet resistance vs magnetic field at $T=0.46$ K for the film shown in Fig. 1. Inset: A linear plot.

Because the films are extremely thin, we believe that holes in the film are the major cause of pinning. Hence the pinning is strong and there should be no vortex lattice [19]. This suggests that the observed magnetoresistance is due to independent rather than collective motion of vortices. For ultrathin films, it would be natural to consider electrical resistance at low temperatures in a magnetic field to result from the thermally activated hopping of vortices between pinning sites. However, at low temperatures thermal activation may be much less important than quantum tunneling of vortices between pinning sites. Since the tunneling barrier separating vortex configurations at two different sites depends on the magnitude of the order parameter, which is temperature dependent, there can be a temperature-dependent resistance even in the quantum tunneling mode which would ordinarily be temperature independent.

For a quantitative description, we estimate the action integral S for the tunneling between the above-mentioned configurations following a modification of the model discussed in Ref. [2] that assumes overdamped dynamics for vortices. The latter is the relevant mode for films [15]. If a vortex tunnels a distance r_b through a barrier with a height of the order of its core energy, the action can be estimated as $S\sim\gamma(\hbar^2/e^2)r_b^2/\xi^2(T)R_N$. This determines the tunneling probability W_t of a single vortex,

$$\ln W_t = -\gamma[\hbar r_b^2/e^2 R_N \xi^2(T)]\,, \tag{1}$$

where γ is a positive numerical constant, R_N is the nor-

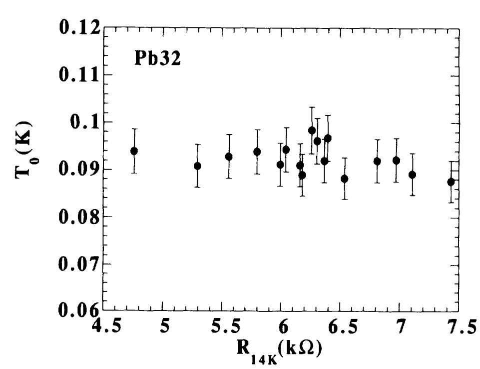

FIG. 3. Plot for seventeen Pb films of the parameter T_0 vs $R(14\text{ K})$ assuming $R\sim\exp(T/T_0)$ as a description of $R(T)$. The error bars are as indicated.

mal sheet resistance, r_b is a tunneling distance, and $\xi(T)$ is the temperature-dependent coherence length. For a superconducting film, r_b is essentially a random variable and percolation theory should be used to obtain the vortex-induced resistance R from (1). In an estimate of $\ln R$ ($\sim \ln W_t$) that neglects numerical constants we can simply replace r_b^2 by a configurational average $\langle r_b^2 \rangle$ (provided that the distribution of values of r_b^2 is not exponentially wide). The mobility of the vortices is proportional to the tunneling rate and the resistance of the film due to this mechanism can be estimated. Using $\xi(0) = 0.85(\xi_0 l)^{1/2}$, $\xi_0 = 0.18\hbar v_F/[1.76\pi k_B T_{c0}]$ (the BCS result) [20], and $R_N = (\hbar/e^2)(2\pi\hbar/m v_F l)$ (the 2D result), where v_F is Fermi velocity, one obtains

$$\ln R = 1.22\gamma(\langle r_b^2 \rangle m k_B/\hbar^2)(T - T_{c0}) . \quad (2)$$

The linear $\ln R$ vs T dependence is a salient feature of the model borne out by the data. This also enables us to identify T_0^{-1} with the coefficient of $T - T_{c0}$ in Eq. (2). The temperature dependence [21] of the coherence length $\xi^2(T) = \xi^2(0) T_{c0}/(T_{c0} - T)$ is responsible for the unusual temperature dependence. The latter is observed in a regime in which the vortex transfer rate determined by tunneling would be temperature independent if the parameters characterizing the superconducting state were. Given the fact that the thicknesses of various films shown in Fig. 2 vary only by about 0.5 Å, $\langle r_b^2 \rangle$ should not change much for these films. This implies that $T_0 = [1.22\gamma \times \langle r_b^2 \rangle m k_B/\hbar^2]^{-1}$ should be constant, independent of the sheet resistance, as was found for Pb films. We believe the irregular variation of T_0 with R(14 K) for Al films follows from the fact that these films are deposited from a much hotter vapor source than was needed for either Bi or Pb. Thus, the structures of Al films will change during growth whereas those of Bi and Pb will not. Also, the onset of measurable conductances in the case of Al films was found at much greater thicknesses than for the Pb or Bi films, a result which is due to the formation of Al clusters as a consequence of deposition from a hot source.

An important consideration is the competition between quantum tunneling and thermal activation. The probability of the latter, $W_a \sim \exp(-U/T)$, can be easily estimated if the barrier height is of the order of the core energy of a vortex, $U = \varepsilon\Phi_0^2 d/16\pi^2\lambda^2$. Here d is the film thickness, λ is the penetration depth, and ε is a constant [20]. Using the dirty-limit expression for λ and proceeding in a manner well known, e.g., from the theory of Kosterlitz-Thouless transition [18], we obtain

$$\ln W_a = -10.7\varepsilon(\hbar/e^2 R_N)(T - T_{c0})/T . \quad (3)$$

This would lead to an Arrhenius form for the resistance $R \sim \exp(-T_0/T)$. At low temperatures this form clearly does not describe the data. For higher temperatures, it is unclear if it would fit the data. The accessible temperature range, which is limited at the high end by T_{c0}, and at the low end by the onset of the linear dependence of $\ln R$ on T, is too small to carry out a conclusive analysis. Comparison of Eqs. (1) and (3) leads to the following estimate of the temperature T_t below which vortex motion is controlled by tunneling:

$$T_t = 10.7\varepsilon \frac{\hbar}{e^2 R_N} T_0 = \frac{8.77\varepsilon}{\gamma} \frac{\hbar}{e^2 R_N} \frac{\hbar^2}{k_B m \langle r_b^2 \rangle} . \quad (4)$$

This characteristic temperature must be smaller than T_{c0} for the model to be self-consistent. The proportionality of T_t to $1/R_N$ at a constant $\langle r_b^2 \rangle$ in Eq. (4) is in qualitative agreement with our observation of a shift in the onset temperature for the unusual resistive tail towards lower temperatures as R_N increases.

Vortex tunneling can explain the linear temperature dependence of $\ln R$ for $T < T_t$. Full quantitative comparison of experiment and theory is not possible given that the numerical factors ε and γ depend on unknown details of the models underlying Eqs. (1)-(4). It is possible to estimate or set bounds on these factors from the data. Equation (4) can be used to estimate ε. The resultant values for representative films are given in Table I. One can set bounds on γ by first noting that for the quantum tunneling model to be applicable the inequality $\xi^2(T_t) < \langle r_b^2 \rangle < L^2$ should be satisfied at T_t, the beginning of the quantum regime, where L is a characteristic linear dimension in the film. This corresponds to the tunneling distance being smaller than the sample size, but larger than the maximum size of the vortex core in the quantum regime. The upper and lower bounds on γ then follow from the expression $T_0 = [1.22\gamma \langle r_b^2 \rangle m k_B/\hbar^2]^{-1}$ using the inequalities. The lower bound on γ is essentially zero because L is a macroscopic length. See also Table I for a tabulation of the upper bounds.

It should be pointed out that we have assumed in the above discussion that the energy levels accessible to vortices on pinning sites are distributed over a range smaller than the thermal energy $k_B T$; this is compatible with our assumption that holes in the film cause pinning. The opposite limit, that of variable range hopping of vortices as discussed in Ref. [14], seems not be consistent with the observations, as the predicted variable-range-hopping result $R \sim \exp(a/T^a)$ is not found.

The above analysis has not considered the possible effect of noise. Although the electrical leads entering the experimental environment were all well shielded, it is still

TABLE I. Parameters as defined in the text for representative films. $\xi(0)$ is determined from the slope of $H_{c2}(T)$ near T_{c0}. The use of $\xi(0) = [0.817(\hbar^3/m k_B e^2)(1/R_N T_{c0})]^{1/2}$ gives values of $\xi(0)$ a factor of 3 larger, and upper bounds on γ an order of magnitude smaller than those listed.

Film	d (Å)	R_N (kΩ)	T_{c0} (K)	T_0 (K)	T_t (K)	$\xi(0)$ (Å)	$\xi(T_t)$ (Å)	ε	γ
Pb	3.86	6.0	1.62	0.10	1.1	41	73	1.5	< 136
Al	28.65	8.25	2.27	0.058	1.9	37	91	6.15	< 151
Bi	16.9	0.719	4.04	0.10	3.95	21	145	0.65	< 34.7

possible that high-frequency Johnson noise from the sections of the electrical leads at high temperatures inside the shielded environment, which corresponds to a very low level of extrinsic noise, could be affecting the measurements. In principle, noise can establish an effective temperature which determines the properties of the electron system, T_{eff}, which is higher than the measured temperature of the thermal bath. This could lead to a flattening of the tail of the resistive transition and should be distinguishable from the temperature dependence presented above. Indeed, flattened tails have been seen in some of our measurements. In the case of an Al film, a flattening of $R(T)$ was found after five decades of variation consistent with Eq. (2) and may represent a "noise floor" in those measurements. A noise floor would result if the barrier for tunneling or activation ceased to change for $T < T_{eff}$. Even if the noise were to affect the activation processes in a more complicated manner, not accounted for by a simple substitution of T by T_{eff}, it is likely that the corresponding $\ln W_a$ would depend on the normal sheet resistance, as in Eq. (3), which would be inconsistent with Fig. 3 where it is not. For example, if the effects of noise were to lead to $\ln W_a \sim U(T)/T_{eff}$, this would result in $T_0 \sim R_N T_{eff}$, a result inconsistent with our data. These considerations would appear to rule out extrinsic or high-temperature noise as being responsible for the observed effects.

It should be noted that a linear dependence of $\ln R$ on T, similar to the one reported here, was observed in granular Al films years ago, without an identification of the relevant physics [22]. It has also been observed more recently [11] in small-diameter In wires in zero applied magnetic field, where it was interpreted in the context of a model of the dissipative tunneling at phase slip centers, in which $\ln R$ seems to be a linear function of $(T_c - T)^{1/2}$ rather than $T_c - T$ [see Eq. (6) in Ref. [11]]. The precise connection of this work to the present study requires further investigation.

Although the resistance measurements shown above were carried out in the linear regime, the *I-V* characteristics were found to exhibit small nonlinearities in the high current limit. Such features would arise within the quantum tunneling picture if the self-field due to the current were taken into account [2]. Further measurements over a wider current range, together with a more detailed theoretical analysis, are needed to elaborate this issue.

In summary, an unusual temperature dependence of the resistance of ultrathin films of Pb, Al, and Bi of the form $R \approx R_0 \exp(T/T_0)$ has been observed in small magnetic fields. It is argued that this dependence is due to quantum tunneling of vortices between pinning sites.

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[a]Present address: Department of Physics, University of Colorado, Campus Box 390, Boulder, CO 80309.

[b]Present address: Department of Physics, Chalmers Technology University, S-412 96, Göteborg, Sweden.

[c]Also at the Theoretical Physics Institute, University of Minnesota, Minneapolis, MN 55455.

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