Evidence for Macroscopic Quantum Tunneling in One-Dimensional Superconductors

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(Received 25 April 1988)

Measurements of the superconducting-to-normal transition in very-small-diameter In wires are presented. The results are discussed in terms of the currently accepted theory, in which dissipation below T_c is attributed to thermally activated motion of the Ginzburg-Landau order parameter over the free-energy barriers which separate metastable states. This theory is consistent with our results for temperatures within about 0.2 K of T_c, but at lower temperatures it fails qualitatively. We suggest that the low-temperature behavior is dominated by quantum-mechanical tunneling *through* the free-energy barrier, and show that our results are consistent with this picture.

PACS numbers: 74.40.+k, 05.40.+j

The current understanding of dissipation in one-dimensional superconductors near T_c is based largely on the work of Little,[1] Langer and Ambegaokar,[2] and McCumber and Halperin.[3] According to this picture, the current carrying state is only metastable, and dissipation occurs when the system passes, via thermal activation, over the associated free-energy barrier to a state of lower free energy. If one considers the case in which the system is biased with a constant voltage, V, then using the Josephson relation one obtains $\partial(\Delta\phi)/\partial t = 2eV/\hbar$. Here ϕ is the phase of the Ginzburg-Landau (GL) order parameter and $\Delta\phi$ is the phase difference across the system. An applied voltage thus causes $\Delta\phi$ to increase with time, which results in an increasing supercurrent, I_s, since $I_s \sim \Delta\phi$. Eventually the critical current is exceeded, as superconductivity (at that current) becomes energetically unstable. However, before the critical current is reached, it is possible to have a thermodynamic fluctuation in which the magnitude of the order parameter goes to zero in a localized region, allowing the system to reduce $\Delta\phi$, and thereby lower I_s. These phase-slip events serve to decrease the current, and result in dissipation. According to Langer and Ambegaokar,[2] the phase slips occur when the order parameter passes, via thermal activation, over the free-energy barrier separating states whose values of $\Delta\phi$ differ by 2π. A detailed theory of this process has been worked out,[2,3] and accounts reasonably well for a number of experiments.[4] In the previous experiments, the samples were typically 0.5-μm-diam whisker crystals; this was sufficiently small so as to be one dimensional as far as superconductivity is concerned (at least near T_c), but sufficiently large that, for low sample currents, dissipation could be observed only very near T_c. In this Letter, results are presented for samples with significantly smaller diameters, for which it is possible to study the dissipation at temperatures much farther below T_c. We find that while the thermal-activation picture is consistent with the results near T_c, it appears to break down as the temperature is decreased. Our results seem to suggest that there is another process by which the phase slippage occurs. We consider the possibility of quantum tunneling[5,6] *through* the free-energy barrier, and show that our results are consistent with this mechanism.

The samples were very narrow In strips (i.e., wires) which were fabricated from evaporated In films with step-edge lithographic techniques.[7] The cross sections were approximately right triangular, and the diameters quoted here correspond to $\sqrt{\sigma}$, where σ is the cross-sectional area. Selected samples were examined with scanning electron microscopy, and were found to be very similar in appearance to AuPd wires which have been described in great detail in Ref. 7. The samples had diameters in the range 400–1000 Å, with normal state resistivities of 4–12 $\mu\Omega$ cm (depending on the wire size, with the thinner wires having the higher resistivities). The wire diameters appeared to be uniform to within typically 100 Å or better, which was near the resolution limit of the scanning electron microscope, and the grain size was in the range 100–200 Å. The measurements were performed with standard dc techniques, with careful shielding of the current and voltage leads to avoid problems with external noise.

According to the thermal-activation model, the resistance below T_c can, in the low-current limit, be written as[2,3,8]

$$R_{\rm TA} = \frac{\Phi_0 \Omega}{I_1} e^{-\Delta F/k_B T}, \tag{1}$$

where $\Phi_0 = hc/2e$, ΔF is the magnitude of the free-energy barrier, Ω is the associated attempt frequency, and $I_1 = k_B T/\Phi_0$. For currents, I, much less than the critical current, ΔF and Ω can be expressed in terms of the critical field, H_c, coherence length, ξ, and the GL relaxation time $\tau_{\rm GL}$,

$$\Delta F = \sqrt{2}\sigma H_c^2 \xi / 3\pi, \tag{2}$$

$$\Omega = \frac{L}{2\pi^2 \xi \tau_{\rm GL}} \left(\frac{3\pi \Delta F}{k_B T} \right)^{1/2}, \tag{3}$$

$$\tau_{\rm GL} = \frac{\pi \hbar}{8 k_B (T_c - T)}, \tag{4}$$

where L is the length of the sample. Near T_c, $H_c \sim \Delta T \equiv (T_c - T)$, and $\xi \sim (\Delta T)^{-1/2}$. Hence, ΔF goes to zero at T_c as $(\Delta T)^{3/2}$, and since this factor enters in the exponent in (1), it dominates the temperature dependence of R_{TA}. It is important to note that the theory (1) is only applicable when $\Delta F \gg k_{\mathrm{B}}T$. Since ΔF vanishes at T_c, this means that (1) will not apply very near T_c.

Figure 1 shows some typical results for the resistance as a function of temperature for three samples. To within the experimental uncertainties, these results are independent of measuring current, for the currents employed here (typically $\lesssim 10^{-8}$ A). We first consider the large (720 Å) sample in Fig. 1. The solid curve shows a fit to (1). The breakdown of (1) is evidenced by the rapid decrease in the solid curve very near T_c. At lower temperatures, (1) describes the results fairly well over several decades of resistance,[9] although we note that it was necessary to adjust ΔF downward from the theoretical prediction (2) by a constant factor[10] of ~ 4. This discrepancy with (2) may be due to inhomogeneities in the samples, i.e., "weak spots," at which the effective cross-sectional area is smaller than the average value. If so, then the phase slippage occurs preferentially at the weak spots, but the nature of the phase slips remains the same; that is, they are still described by the thermal-activation theory [but with a slightly different value of Ω, Eq. (3)]. It does not appear that such behavior would significantly affect any of the arguments given below.[11] In any case, the results for the large sample in Fig. 1 are in good agreement with the form predicted by the thermal-activation model. Let us now consider the smallest (410 Å) sample in Fig. 1. The solid curve again shows a fit to the thermal-activation theory, (1). It was again necessary to adjust ΔF downward from (2) by a factor of ~ 4, to obtain a satisfactory fit for $\Delta T \lesssim 0.2$ K. However, at lower temperatures R deviates significantly from (1). It is clear from (1) and (2) that this behavior *cannot* be explained with the thermal-activation model. The temperature dependence predicted by (1) is much stronger than what is observed at lower temperatures. The behavior for the third (505 Å) sample in Fig. 1 is also seen to show strong deviations from the thermal-activation form at the lowest temperatures.

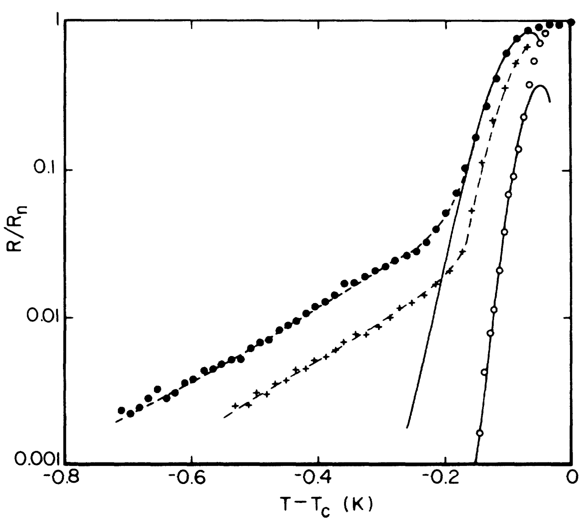

FIG. 1. Resistance, normalized by the normal stae value, as a function of temperature for three In wires; the sample diameters were 410 Å (●), 505 Å (+), and 720 Å (○). The solid curves are fits to the thermal activation theory (1), while the dashed curves are fits to (7). Where the solid and dashed curves overlap, only the former is shown for clarity. The samples had lengths of 80, 150, and 150 μm, and normal-state resistances of 5.7, 7.1, and 1.2 kΩ, where we list the values in order of sample size, with the value for the smallest sample first.

As the temperature decreases, the size of the free-energy barrier (2) relative to $k_{\mathrm{B}}T$ increases, leading to a rapid reduction in the thermal-activation rate. One might expect that as this rate becomes negligible, the rate of some other process by which the phase can relax, might become dominant. It has been suggested[12] that tunneling *through* the energy barrier could become significant under these conditions. We will now show that our results are consistent with this mechanism for phase slippage. However, we want to emphasize that this explanation of our results is certainly not unique, and it is quite conceivable that some other, as yet unidentified, mechanism could well be dominant.

To the best of our knowledge, there is no quantitative theory of macroscopic quantum tunneling (MQT) for a one-dimensional superconductor. However, we can work from analogy with the well-developed theory of MQT in other systems to estimate the form that such a theory would take.[13] In general, the tunneling rate, Γ_{MQT}, is given by an expression of the form[5,6,14]

$$\Gamma_{\mathrm{MQT}} = A e^{-B/\hbar}, \tag{5}$$

and the parameters A and B depend on whether the motion of the "particle" which tunnels is strongly or weakly damped. The thermal-activation theory is based on time-dependent GL theory,[15] which predicts a purely diffusive time dependence for the order parameter. Accordingly, (1) is based on the theory of thermal activation in the strongly damped limit.[16] We will therefore assume that MQT in our case is also in the strongly damped limit.[17] In this regime, the theory predicts[6] $B = 2\pi\eta(\delta\phi)^2/9$, where $\delta\phi$ is the distance under the barrier, and η is the effective "viscosity." From thermal-activation theory,[2,3] we expect $\delta\phi \sim 1$, and examination of the time-dependent GL equation yields[3,15] $\eta = \tau_{\mathrm{GL}}\sigma H_c^2 \xi/4\pi$. The estimation of the prefactor A in (5) is not as straightforward. In the limit of large damping the theory[6,18] predicts $A = 8\sqrt{6}\omega_0 \alpha^{7/2}(V_b/\hbar\omega_0)^{1/2}$, where ω_0 is the natural frequency of oscillation of the particle which tunnels, V_b is the barrier height (which for our case is ΔF), $\alpha = \eta/2m\omega_0$, and m is the "mass" of the particle. The problem now is to estimate m, a parameter which appears to be beyond the scope of previous treatments of time-dependent GL theory.[15] However, there are a number of cases in which propagating modes in superconductors have been derived, indicating that mass terms can exist. In particular, Mooij and Schön[19] have predicted the existence of a propagating plasma mode in very small diameter, one-dimensional superconductors, and shown that this mode leads to phase fluctuations closely associated with those considered here. Since we must know at least the qualitative form of m in order to estimate A, we will assume that the damping factor α is a constant, which we will take to be α_0. Note that the exponential factor in (5) dominates the temperature dependence, so the qualitative behavior found below is largely independent of any temperature dependence of α.

With this assumption it can be shown that $R_{\rm MQT}=\Phi_0\Gamma_{MQT}/I_1$, with[20]

$$\Gamma_{\rm MQT}=\frac{16\sqrt{3}L\alpha_0^4}{\xi}\left(\frac{\Delta F}{\hbar\tau_{\rm GL}}\right)^{1/2}\exp\left(-\frac{\beta\sigma H_c^2\xi\tau_{\rm GL}}{18\hbar}\right), \quad (6)$$

where β is an unknown constant of order unity which arises from uncertainties in the distance under the barrier and the factor of L/ξ enters as the number of independent locations at which a tunneling event can occur. The total probability for escape will be the sum of the thermal activation and MQT rates, so we have

$$R_{\rm TOTAL}=R_{\rm TA}+R_{\rm MQT}. \quad (7)$$

The dashed curves in Fig. 1 show fits of (7) to the results for the 410- and 505-Å samples. We see that (7) accounts extremely well for the behavior of R over the entire temperature range studied.[21] The fit to (7) involves three free parameters; β, α_0, and an overall factor which adjusts the magnitude of the free-energy barrier. If our ideas concerning MQT are correct, we would expect β to be of order unity, and $\alpha_0 \gtrsim 1$. The fits shown in Fig. 1 yielded $\beta\approx 0.3$ and $\alpha_0\approx 1$, which therefore supports our interpretation. The abrupt change in the temperature dependence of R at $\Delta T\approx 0.2$ K is due to a crossover from thermal activation to MQT. The reason for this is easy to see. The thermal activation and MQT rates, (1) and (6), depend in nearly the same manner on ΔF; the major difference involves the factors which normalize ΔF in the exponents. For thermal activation this factor is k_BT, while for MQT it is (apart from a constant factor) $\tau_{\rm GL}^{-1}$. Since $\tau_{\rm GL}^{-1}\sim\Delta T$, it eventually becomes larger than $k_BT/\hbar$, which causes MQT to dominate. Behavior similar to that seen in Fig. 1 has been observed in a number of other samples, with similar results for the factors β and α_0. We can also understand why MQT is not observed in the 720-Å sample considered in Fig. 1, or in previous experiments. The temperature at which the behavior changes over from thermal activation to MQT depends on the variation of $\tau_{\rm GL}$, as just discussed. The temperature at which the crossover occurs, T^*, is easily seen to be independent of the cross-sectional area of the sample, σ. However, as σ increases, the rates of both thermal activation and MQT at T^* decrease, and hence for the larger samples, the resistance is negligible at T^*, which prevents the study of the MQT regime in this way.[22] The results for the 505-Å sample in Fig. 1 support these arguments. The crossover from thermal activation to MQT is seen to occur at approximately the same temperature as for the 410-Å sample, but the resistance at which this occurs is reduced because of the larger value of σ, and hence ΔF.

In summary, we have analyzed the behavior of very thin In wires near the superconducting transition. We have shown that the results below T_c can be consistently interpreted in terms of a combination of thermal activation and MQT. We emphasize that our results do *not* rule out the possibility that some hitherto unidentified mechanism, other than MQT, is dominant at low temperatures. Nevertheless, our results do suggest a number of avenues for further work. First, a proper theory of MQT in this system, especially consideration of the mass term in the equation of motion, would be of great interest. It should be possible to determine all of the parameters which would enter such a theory, and thus enable a detailed test of the theory of MQT. Second, while MQT has been observed previously in other systems, the ability to study it in one-dimensional superconductors may offer interesting new possibilities. The energy scales in one-dimensional superconductors are such that MQT can dominate at temperatures well above 1 K, in contrast to other systems, in which much lower temperatures are generally required. This makes shielding and isolation problems much less severe, and could make this an interesting system in which to look for other related effects, such as macroscopic quantum coherence.[23]

I thank P. Muzikar for many stimulating and critical discussions, T. R. Lemberger for very helpful comments, and E. Sweetland for invaluable assistance with the fabrication. This work was supported by the National Science Foundation through Grant No. DMR-8614862.

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[8]In general, the theory (see Refs. 2 and 3) predicts a current-dependent resistance, but for the low currents employed here (typically $\lesssim 10^{-8}$ A), the resistance is approximately independent of I, and is given by (1).

[9]The range of applicability of (1) found here is very similar to that seen in previous work (Ref. 4).

[10]In the calculations of ΔF, etc., we used $H_c(0)=286$ Oe [see R. W. Shaw, D. E. Mapother, and D. C. Hopkins, Phys. Rev. **120**, 88 (1960)]. The coherence length was estimated from the standard relations [see, for example, M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York, 1975), p. 113], with $\xi(0)=0.86\sqrt{\lambda\xi_0}$, where for In, $\xi_0=2.6\times10^{-5}$ cm [see, A. M. Toxen, Phys. Rev. **123**, 442 (1961)], and λ is the mean free path. λ was estimated from the resistivity and the measured $\rho\lambda$ product [$\rho\lambda=1.1\times10^{-11}$ Ω cm^2, K. R. Lyall and J. F. Cochran, Phys. Rev. **139**, 517 (1967)]. The mean free path was 240 Å for the 720-Å sample in Fig. 1, and 95 Å for the other samples in Fig. 1. We should also note that the magnitude of the critical current was consistent with this reduced value of ΔF inferred from the fits to (1). In addition, the critical current varied approximately as $(\Delta T)^{3/2}$, as expected from Ginzburg-Landau theory. The value of the normal-state resistance obtained from measurements below T_c with currents larger than the critical current was the same as that found above T_c.

[11]If the phase slips occur preferentially at certain locations where the free-energy barrier is lowest, the picture would then be that the tunneling events occur at the same locations. This would not affect our basic discussion of quantum tunneling.

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[17]It is interesting to note that if we were to assume instead that the system is weakly damped, the temperature dependence of B, and hence that of MQT, would be the same as (6), for the following reason. In the weakly damped limit, the factor B is proportional to $\Delta F/\hbar\omega_0$. For our problem, the natural time scale is $\tau_{\rm GL}$, which leads one to expect $\omega_0\sim\tau_{\rm GL}^{-1}$. The resulting value of B is the same as in (6), to within a constant factor. A key point here is that for either strong or weak damping, the MQT rate has a much weaker temperature dependence then the thermal rate, which is precisely what is needed to explain our results.

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[20]The key step in eliminating ω_0 and m from the prefactor in (6) is to use the fact that $\omega_0\sim(V_b/m)^{1/2}$, where the barrier height $V_b=\Delta F$. Since $\alpha=\eta/2m\omega_0$ and $\eta=\tau_{\rm GL}\sigma H_c^2\xi/4\pi\sim\tau_{\rm GL}\Delta F$, this yields $\omega_0\sim2\alpha/\tau_{\rm GL}$, which then leads to (6).

[21]The result for the free-energy barrier (2) based on the GL results for H_c and ξ (see Ref. 10) are only valid near T_c; deviations from the GL form occur at low temperatures. However, even at the lowest temperatures studied here, the deviations are small [see, for example, J. Romijn, T. M. Klapwijk, M. J. Renne, and J. E. Mooij, Phys. Rev. B **26**, 3648 (1982)], $\approx 20\%$ or less, and do not significantly affect the results for $\Gamma_{\rm MQT}$.

[22]It is, however, possible to study the phase-slip rate at low temperatures with measurements at finite currents [see e.g., T. A. Fulton and L. N. Dunkelberger, Phys. Rev. B **9**, 4760 (1974)]. We have performed such measurements, and preliminary results indicate that the rates at low temperatures in the larger wires are much greater than predicted by the thermal-activation model. The expression for the MQT rate (6), with parameters, β and α_0 obtained from the fits to the low-current behavior of the two smallest wires in Fig. 1, gives a good account of these results for the larger wires. This indicates that in *all* samples, both large and small, the phase-slip rate at low temperatures is greatly enhanced over the thermal rate. While these measurements of the phase-slip rate make it possible to study the behavior at temperatures far below T_c, it would also be very useful to extend the measurements in Fig. 1 to lower temperatures. Unfortunately this is difficult, since the resistance becomes very (exponentially) small at low temperatures.

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