Macroscopic Quantum Tunneling and Related Effects in a One-Dimensional Superconductor

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(Received 2 October 1989)

The results of a study of the superconducting state of very-small-diameter PbIn wires are described. The smallest samples, which had diameters below 200 Å, exhibited significant dissipation at all temperatures below T_c. In addition, their voltage-current characteristics exhibited oscillatory structure, which became more pronounced as the temperature was decreased. These results are consistent with a model in which the phase of the order parameter is treated as a quantum degree of freedom. The observed behavior is then due to quantum tunneling of the order parameter, and the existence of discrete energy levels.

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

Some years ago it was shown[1] that the behavior of a one-dimensional superconductor is analogous to the motion of a particle in a "washboardlike" potential, Fig. 1(a). Previous theoretical[1] and experimental[2] work has shown that for systems with relatively large diameters (here a system may be considered to be one-dimensional if its diameter is less than the coherence length) the motion of this particle may be treated classically. However, recent experiments[3] suggest that if the diameter is made sufficiently small, the particle must be treated quantum mechanically. In particular, small-diameter systems exhibit behavior analogous to quantum tunneling; i.e., the particle in Fig. 1(a) tunnels between adjacent potential minima. This is very similar to the phenomenon of macroscopic quantum tunneling in superconducting tunnel junctions, which has recently been of great interest.[4] In our previous experiments with one-dimensional superconductors[3] the smallest diameters were $\sim$400 Å. In this paper we describe experiments which seem to show that if the diameter of the system is made even smaller, other quantum effects become observable. These effects include coherent motion of wave packets composed of states from many different potential wells, and effects due to the quantized energy levels within a given well.

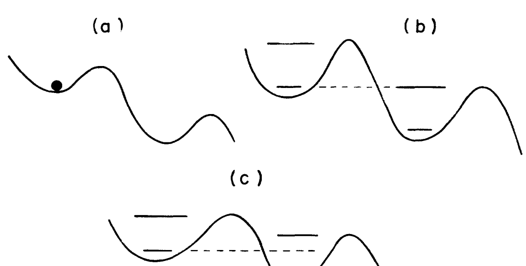

FIG. 1. (a) Schematic of a washboardlike potential; the horizontal axis is the position of the particle. (b) Same as (a), but showing schematically the quantized energy levels within each well. Here the tilt of the washboard is such that the levels in adjacent wells are coincident, leading to the possibility of resonant tunneling. (c) Same as (b) but now the tilt is such that the levels are not coincident.

The superconducting order parameter may be written as $\psi = fe^{i\phi}$. When there is a nonzero current the phase difference across the system is also nonzero, since $\Delta\phi \sim I$. The state of the superconductor, i.e., the "position" coordinate in Fig. 1, can be described by a variable which is closely related to $\Delta\phi$, and the adjacent minima in Fig. 1 correspond to states whose values of $\Delta\phi$ differ by $\pm 2\pi$. For a classical particle, motion from one minima to an adjacent one, also known as phase slip, occurs via thermal activation over the intervening potential barrier (this is the basis for the thermal-activation theory of dissipation in one-dimensional superconductors[1]). However, as the diameter of the system is reduced, the mass of the particle is also reduced, and it should eventually become necessary to treat the particle quantum mechanically.

There are several important parameters which characterize the particle and the potential. First, the average slope of the washboard is proportional to the current according to[1]

$$\Delta F_I = \pm hI/2e \,, \tag{1}$$

where ΔF_I is the difference in (free) energy[1] between two adjacent minima, e is the electronic charge, and I is the current. The height of the barrier between adjacent wells is, in the limit $I \to 0$, given by[1]

$$\Delta F_0 = \sqrt{2} H_c^2 \xi \sigma / 3\pi \,, \tag{2}$$

where H_c is the critical current, ξ is the coherence length, and σ is the cross-sectional area of the system. The behavior can be probed by measuring the voltage; from the Josephson relation one has

$$\Delta V = (\hbar/2e) \partial \Delta\phi / \partial t \,, \tag{3}$$

where ΔV is the voltage across the system. In terms of the motion of the particle, (3) implies that the *faster* the particle moves down the washboard, the *larger* the voltage. The mass of the particle is also crucial to this problem, since the level spacing will depend on the mass together with the curvature of the potential well. In our previous work[3] we suggested that near T_c the Ginzburg-Landau relaxation time, τ_{GL}, would be the characteristic time scale in this problem, and hence that the level spacing would be $\hbar/\tau_{GL}$. This suggestion appears to be consistent with recent theoretical work.[5] However, at temperatures well below T_c, which are of primary interest in the present paper, the relevant time scale may instead be[5,6] $\tau_0 = \sqrt{3}\xi/v_F$, where v_F is the Fermi velocity. At low temperatures τ_{GL} and τ_0 are comparable, and are in the range $(1-4)\times 10^{-13}$ s for our system.

Perhaps the most convenient quantities to measure in an experiment are the voltage-current characteristics and the resistance. For a very small system, i.e., a very light particle, the quantum tunneling rate will be large, and the particle will never be confined. From (3) this implies that for a nonzero current, there will be a nonzero voltage at all temperatures below T_c. The effective resistance will depend on the tunneling rate. It will become larger as the system is made smaller, since the barrier width should be proportional to ΔF_0, (2), which is proportional to σ. One would also expect level quantization to affect the tunneling rate.[7] If the applied current is such that levels in adjacent wells are coincident, Fig. 1(b), the tunneling rate should be higher than if the levels are not coincident, Fig. 1(c). The two cases would then correspond to regions of large and small values of dV/dI, respectively. A complication which we have not included is the effect of coupling to the environment, e.g., by inelastic processes or through the quasiparticles which may be present. Presumably this coupling will act to broaden the levels and make resonant tunneling effects less pronounced.

The samples were very narrow wires composed of PbIn ($\approx 10\%$ In by weight), which were fabricated using a step-edge method.[8] This alloy was chosen because it is easy to deposit, does not oxidize readily, and has a fairly small grain size. The granularity was reduced by coating the substrate with a thin ($\approx$ 50-Å) layer of Ge just prior to evaporating the PbIn, and also cooling the substrate to 77 K. Wires as small as ≈ 170 Å were made directly with the step-edge method, and they had normal-state resistivities of $\approx 20\ \mu\Omega\,\mathrm{cm}$. The smallest wires were somewhat unstable—if left at room temperature their resistance slowly changed with time, due to either oxidation or (we believe) agglomeration. In some experiments we took advantage of this, and used gentle annealing at temperatures between 77 K and room temperature to reduce the effective diameter further. In this way, samples with effective (i.e., average) diameters as small as ≈ 50 Å were obtained. It seems likely that the sample cross sections were not perfectly uniform, especially in the smallest samples (e.g., with diameters below 100 Å). However, for the samples discussed here annealing was used only sparingly, to produce reductions in the diameter of at most 10%. Problems with inhomogeneities are unavoidable, but one can argue that so long as they are small, they will not affect the basic physics involved, for the following reason. The energy barrier, (2), will be smallest at locations where the diameter is smallest, and while phase slippage will occur preferentially at these places, the effects described above should all still occur.[9] We also note that care was taken in the experiments to heavily shield and filter all leads to the sample, to avoid problems with external noise.[3]

Figure 2 shows some typical results for the resistance as a function of temperature for PbIn wires of several diameters, as indicated in the figure. All exhibit a pronounced drop in resistance near 7.0 K, which we identify as T_c. The value of T_c was essentially the same as that found in coevaporated films, and was slightly below the critical temperature for pure Pb by an amount which dependend on the precise alloy composition (which was slightly different for different evaporations). For the largest wires, with $d \gtrsim 300$ Å, the resistance approached zero rapidly below T_c, and the behavior could be described quantitatively by the thermal-activation model. As the diameter was reduced to the neighborhood of 250 Å, the resistance below T_c vanished much more slowly. This is illustrated by the behavior of the 255-Å sample in Fig. 2; here the resistance exhibited a pronounced "tail" at low temperatures. This can be explained by quantum tunneling through the energy barriers in Fig. 1, which becomes more rapid as the sample diameter, and hence the thickness of the barrier, are reduced. However, the resistance still approaches zero at low temperatures, since the barrier (2) grows as T is reduced, and for this sample size the value of ΔF_0 at low temperatures is large enough to yield a negligibly small tunneling rate as $T \rightarrow 0$. All of the behavior described so far is very simi-

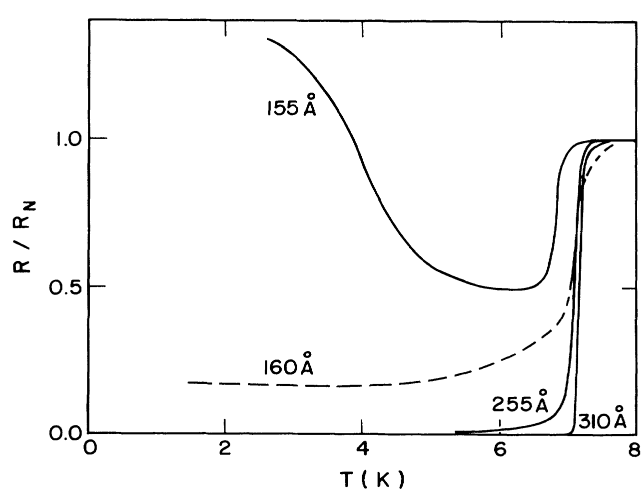

FIG. 2. Resistance as a function of temperature for several samples. The sample diameters are indicated in the figure. Except for the smallest sample, the resistance was independent of the applied current for $I \lesssim 10^{-8}$ A.

lar to that found previously in studies of small In wires.[3]

As the wire diameter was reduced below about 200 Å, new behavior was observed. We first consider the 160-Å sample in Fig. 2. In this case the resistance does not vanish below T_c, even as $T \rightarrow 0$. This can be easily understood from (2). As T is reduced below T_c, the barrier height, ΔF_0 (and hence also the thickness of the barrier), increases rapidly at first, yielding a corresponding decrease in the tunneling rate, which in turn reduces the resistance. However, ΔF_0 and also the tunneling rate eventually become independent of T far below T_c. Since $\Delta F_0 \sim \sigma$, this constant tunneling rate will become larger as the system is made smaller, and it appears that for diameters below about 200 Å the tunneling rate is appreciable even in the low-temperature limit. It should also be noted that this behavior cannot possibly be explained in terms of thermal activation, since all thermal processes must vanish as $T \rightarrow 0$.

Reducing the sample diameter still further[10] leads to qualitatively different behavior. As can be seen from Fig. 2, the resistance of the 155-Å sample *increases* as the temperature is reduced below about 4 K. This effect can be quite large; in some cases the effective resistance at low temperatures is *larger* than the resistance in the normal state. An example of this behavior is shown in more detail in Fig. 3 which shows the effective resistance ($\equiv V/I$) as a function of temperature for a single sample at different measuring currents. The resistance below T_c is seen to be strongly current dependent, which indicates that the system is not Ohmic. A surprising aspect of these results is that at low temperatures the effective resistance becomes larger as the current is *reduced.* We note that the currents employed in Fig. 3 are all much smaller than the critical current for switching to the normal state, which is greater than 10^{-5} A for this sample at low temperatures.

It is instructive to consider the V-I characteristics directly, and some typical results are shown in Fig. 4. The V-I curves are seen to have an enhanced slope near $I=0$. Again, in some cases, the corresponding differential resistance was *greater* than the normal-state resistance. In terms of the analogy with the motion of a particle in Fig. 1, this means that the particle is moving *faster* down the washboard than it would down a smooth inclined plane of the same average slope. It is very hard to see how this could be possible for either a "classical" particle, or for a particle which can simply tunnel between adjacent wells. This result seems to imply that the particle is moving *coherently* among many wells. The qualitative picture we have in mind is similar to the formation of Bloch states in a periodic system. In that case, the states are plane-wave-like combinations of the states in individual wells. A particle in such an extended state would be able to move very easily down a slightly tilted washboard, leading to the large differential resistance as $I \rightarrow 0$ seen in Figs. 2-4. This picture is similar to that developed in recent theoretical discussions of the behavior of very small tunnel junctions.[11]

Close examination of Fig. 4 reveals another interesting effect. The V-I curves display oscillatory structure below T_c. This structure is more evident in the inset to Fig. 4, which shows dV/dI at the lowest temperature. A possible explanation of this behavior is shown in Figs. 1(b) and 1(c). When the quantized levels in adjacent wells are coincident, the tunneling rate should increase, leading to an increase in dV/dI, with the reverse occurring when the levels are far from overlapping. From the period of the oscillations in Fig. 4 together with (1) the

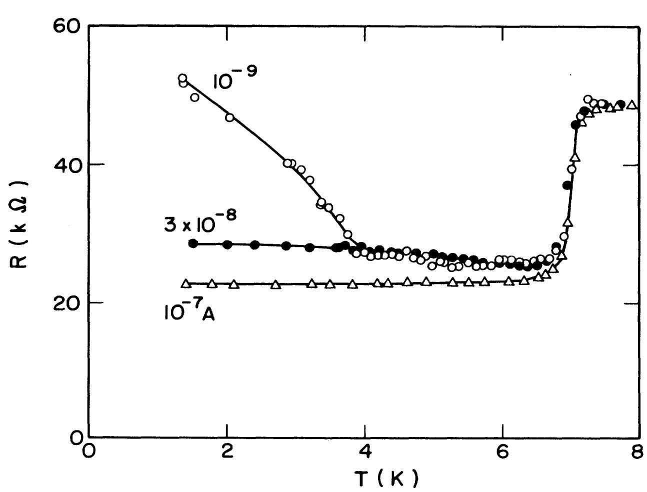

FIG. 3. Effective resistance ($\equiv V/I$) as a function of temperature for a 155-Å sample (different from the one in Fig. 2), for several different applied currents, as indicated.

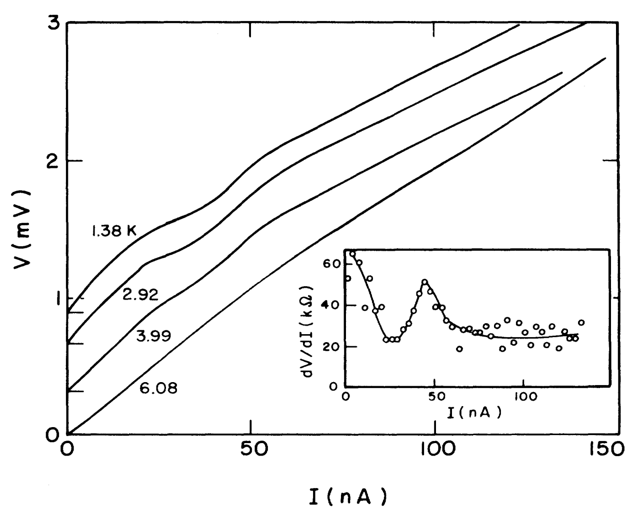

FIG. 4. V-I characteristics for the 155-Å sample considered in Fig. 3, at several temperatures. For clarity, the results for different temperatures have been offset. Inset: dV/dI at 1.38 K.

energy-level spacing can be estimated. If we assume that this spacing is given by $\hbar/\tau$, we find $\tau \approx 1\times10^{-12}$ s at 1.4 K. This is about a factor of 3 larger than either τ_{GL} or τ_0, but given the qualitative nature of our arguments this level of agreement is encouraging. The importance of level quantization is analogous to behavior recently observed in small Josephson junctions,[7] which can also be described by a washboard potential model.

Our interpretation of the results in Figs. 2–4 seems to be qualitatively consistent with the particle-on-a-washboard picture. While a theoretical discussion of quantum tunneling effects in a one-dimensional superconductor has recently appeared,[5] a comprehensive treatment of other aspects of the behavior, including the effects of level quantization, is not available at present. It would therefore be premature to rule out other possible explanations of our results. However, the nonzero dissipation seen in the smallest samples as $T \rightarrow 0$, Fig. 2, would certainly seem to rule out theories based solely on classical thermal activation. In addition, the fact that the differential resistance at low temperatures can be larger than the normal-state resistance appears to imply some type of coherent tunneling process which involves many potential wells, in contrast to simple tunneling between two adjacent wells in which coherence is lost after each tunneling event. These and other features of our results are very reminiscent of the phenomena of macroscopic quantum coherence.[12]

We thank P. Muzikar for many stimulating and critical discussions, and S. Saito for helpful correspondence. This work was supported by the National Science Foundation through Grant No. DMR-8614862.

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[9]It is conceivable that inhomogeneities could lead to effects similar to those we have observed. In particular, one might imagine that there could be "weak spots" which were actually tunnel junctions, etc., which could then behave as discussed in Ref. 4. However, the overall behavior of the resistance between room temperature and low temperatures was not consistent with the presence of such weak links. While this does not conclusively rule out an explanation based on inhomogeneities, etc., the available evidence suggests that they are not a major problem. This point will be discussed further elsewhere.

[10]While the two smallest samples in Fig. 2 had similar diameters, their behavior was quite different. We believe this to be due to the inhomogeneities in the diameter mentioned above. That is, for the sample with a 155 Å average diameter in Fig. 2, there were fluctuations in the size, with the smallest diameter being somewhat less than the smallest diameter of the 160-Å sample. Since the phase slippage will occur primarily at this "weakest" point, the smallest diameter rather than the average diameter will determine behavior. Our results on a number of different samples suggest that the smallest diameter scales roughly with the average diameter. Hence, samples with average diameters greater than 200 Å never exhibited behavior like that of the two smallest samples in Fig. 2.

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