Manipulating superconducting fluctuations by the Little–Parks–de Gennes effect in ultrasmall Al loops

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The destruction of superconducting phase coherence by quantum fluctuations and the control of these fluctuations are a problem of long-standing interest, with recent impetus provided by the relevance of these issues to the pursuit of high temperature superconductivity. Building on the work of Little and Parks, de Gennes predicted more than three decades ago that superconductivity could be destroyed near half-integer-flux quanta in ultrasmall loops, resulting in a destructive regime, and restored by adding a superconducting side branch, which does not affect the flux quantization condition. We report the experimental observation of this Little–Parks–de Gennes effect in Al loops prepared by advanced e-beam lithography. We show that the effect can be used to restore the lost phase coherence by employing side branches.

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he problem of superconducting fluctuations, including the existence of a many-body state composed of fluctuating, phase-incoherent Cooper pairs, has been at the forefront of superconductivity research for decades (1). The fluctuating Cooper pair state, which may emerge when the global superconducting phase coherence is destroyed by strong disorder, Coulomb repulsive interaction, or magnetic field, may have been realized in the pseudogap phase of the high Tc cuprate materials. If the fluctuations in such a state could be suppressed and global phase coherence built through engineering means, very high temperature superconductivity could be obtained (2). A similar state of fluctuating Cooper pairs, controllable through geometry and applied magnetic flux, is found in doubly connected ultrasmall superconductors (3–5). Recent theoretical studies have produced some detailed predictions on the nature of the thermal and quantum superconducting fluctuations in this state (6–9). Novel phenomena, such as hc/e as opposed to the hc/2e Little–Parks oscillations as the size of the loop is reduced (10, 11) and the occurrence of superconductivity in the smallest doubly connected samples, the thinnest carbon nanotubes (12), may be expected. New experimental techniques capable of probing these fluctuations were developed (13, 14). Therefore, similar to singly connected mesoscopic superconductors in which some spectacular physical phenomena were found (15–17), ultrasmall doubly connected superconductors are likely to grow into a fertile testing ground for fundamental studies of superconductivity.

The fluctuated quantization in a thin, doubly connected superconductor requires that an applied flux (Φ) threading the superconductor, produce a superfluid velocity \( v_s \sim (1/C)(n - Φ/Φ_B) \), where C is the circumference of the loop, n is an integer that minimizes \( v_s \), and \( Φ_B(= hc/2e) \) is the flux quantum. The periodic modulation of \( v_s \), which reaches a maximum at half-integer-flux quanta, leads to the well-known Little–Parks oscillations in \( T_c \) (18, 19). Based on a Ginzburg–Landau (G–L) theory, de Gennes made a prediction in 1981 (3) that the \( T_c \) oscillation amplitude could become so large for an ultrasmall loop (with negligible wire thickness) that superconductivity itself is destroyed (\( T_c = 0 \)) near half-integer-flux quanta due to the growth of the maximal \( v_s \) and the associated kinetic energy as C shrinks. The resulting nonsuperconducting state is referred to as the destructive regime (3–5).

This destructive regime represents a quantum phase transition (QPT) wherein the ground state is tuned by the applied flux, transitioning from the superconducting to the normal state at a critical flux \( Φ_c \). The applied flux is expected to suppress the superconducting gap to zero at \( Φ_c \), likely leading to a continuous QPT with the onset of the destructive regime a quantum critical point (QCP). Near the QCP, large conductance fluctuations are expected (6, 7). Furthermore, de Gennes predicted that the addition of a “dangling” side branch, which does not change \( v_s \) and should not affect superconductivity in the loop (except possibly via the proximity effect; see below), could stabilize the phase coherence in the entire sample and suppress the destructive regime (3, 4). The presence of the destructive regime and its suppression by the side branch in ultrasmall superconducting loops are referred to here as the Little–Parks–de Gennes (LPdG) effect.

Results and Discussion

In order to observe the LPdG effect the circumference, C, of the superconducting loop must be smaller than \( πΦ/0 \), where \( ξ(0) \) is the zero temperature superconducting coherence length with a typical value around 100 nm for microstructures of elemental superconductors. Preparation of such an ultrasmall loop, while maintaining long coherence lengths, is a significant challenge. Previous studies of superconducting loop structures almost all featured characteristic circumferences of greater than 15 \( ξ(0) \) (20–23), far too large to observe the LPdG effect. While the use of doubly connected ultrathin cylinders (5, 24–26) led to the successful demonstration of the destructive regime, the full LPdG effect, especially the role played by the side branch, has not been explored experimentally. To achieve the necessary feature sizes and long coherence lengths we fabricated our devices using the procedure detailed in Materials and Methods and measured them using a d.c. electrical transport technique in an RF filtered dilution refrigerator with a base temperature of 20 mK. A typical device, shown in Fig. 1A, features a low level of disorder, with a zero-field transition temperature \( T_{co} = 1.28 \) K and \( ξ(0) = 119 \) nm.

Due to our electrical transport measurement technique our loops cannot be electrically isolated from their environment as done in de Gennes’ original theory. However, at sufficiently high applied magnetic field the bulk electrodes in our loop structures are driven fully normal, providing a normal metal boundary for the narrow leads, which remain superconducting due to parallel critical field enhancement. Therefore the narrow leads conveniently function as the “side branches” of effective length, L, in de Gennes’ theory, even though they are terminated by a normal metal rather than an insulator. This deviation from de Gennes’ original design should not alter the physical picture advanced by de Gennes in which the branches stabilize phase coherence in the loop by adding condensation energy into the system offsetting the kinetic energy associated with \( v_s \). On the other hand, the LPdG

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effect can also be understood in the context of the proximity effect. Essentially the destruction of superconductivity within the loop must also suppress superconductivity in the side branch up to a length of \( \xi(T) \), increasing the energy cost for the destructive regime and thus stabilizing superconductivity in the whole structure or, alternatively, the superconducting side branch could return the “normal” loop in the destructive regime into a superconducting state via proximity effect. Within this alternative picture, the normal metal rather than open end of the side branch could have interesting consequences on details of the LPdG effect.

A destructive regime was observed in our ultrasmall loop structures. Shown in Fig. 1B is a resistance vs. magnetic flux trace taken at 50 mK for a loop with an average circumference, \( C \), of 409 nm and side branch length \( L \) of 146 nm. At zero field the entire device is superconducting, and with increasing field the wide (200 nm) parts of the leads are driven normal first, at a field of 350 G, enhanced over the film critical field of 180 G as determined from resistive measurements. Note that the higher critical field value of the wide parts of the leads results from the critical field enhancement in narrow superconductors (27). As the applied field was increased further (as the corresponding flux approaches \( \Phi = \Phi_0/2 \)) the loop is driven normal, marking the emergence of a superconductor-normal-normal QPT. This sample had full normal state resistance between 0.45 \( \Phi_0 \) and 0.7 \( \Phi_0 \) followed by re-emergence of superconducting behavior, with a resistance minimum at 1,990 G (corresponding to \( \Phi_0 \) for this loop) demonstrating the presence of a destructive regime and QPT.

The existence of QPTs in these ultrasmall doubly connected superconductors depends on the size of the loop. Shown in Fig. 2 is data obtained from two samples of different \( C \) values but a similar lead length, \( L \approx 370 \text{ nm} \). In Fig. 2A, a \( C = 384 \text{ nm} \) sample was found to show a distinctive resistance peak near \( \Phi = \Phi_0/2 \), indicating the destruction of superconductivity. For the sample with \( C = 492 \text{ nm} \), however, superconductivity remains robust to low temperatures at \( \Phi = \Phi_0/2 \) as shown in Fig. 2B.

These experimental results on the occurrence of QPTs can be compared with theoretical predictions. In de Gennes’ original work (3), where the QPT would correspond to the onset of the destructive regime, the width of superconducting loop was assumed to be infinitely small. In our experiment, however, the width is finite. The finite line width of the loop leads to additional kinetic energy because \( v_\text{F} \) in this case will be position dependent, changing the condition for the destructive regime, especially at higher winding number. In order to obtain an estimate of the critical flux value for the onset of the destructive regime for an ultrathin superconducting ring with a finite wire thickness, \( w \), we make use of the prediction on the \( T_c \) modulation in the LP oscillations obtained in the G-L theory without a side branch (28)

\[
\frac{\Delta T_c}{T_{c0}} = \frac{4\pi^2 \xi^2(0)}{C^2} \left[ \frac{(\Phi/\Phi_0 - n)^2}{C} + \frac{\Phi_0}{\Phi_0} \left( \frac{\pi w^2}{C} \right)^2 \right] + \frac{n^2(\pi w^2)^2}{C^2} \left( \frac{1}{3} + \frac{1}{5} \left( \frac{\pi w^2}{C} \right)^2 \right)
\]

from which the critical flux for \( \Delta T_c = 0 \) corresponds to the onset of the destructive regime. For an isolated loop of the same loop size as that of Sample B, Eq. 1 predicts a moderately reduced \( T_c \) of 200 mK with no destructive regime, opposite from the experimental observation. This is unexpected because the inclusion of the measurement leads not considered in ref. 28 should have made the destructive regime less likely as pointed out above. On the other hand, the superconducting fluctuations were not considered in deriving Eq. 1 (28), suggesting that the fluctuations may be important.

In the pair breaking theory developed recently, fluctuations were considered in deriving the conditions for the destructive regime (6–9). Within the pair breaking theory, for loop structures at a given magnetic flux, the energies for a pair of time-reversal single particle states differ by \( \alpha \) (7, 28). A reduced \( T_c \) is obtained by solving the equation

Fig. 1. (A) Scanning electron microscope (SEM) image of a representative sample with the side branch length denoted. (B) Resistance (R) vs. \( \Phi/\Phi_0 \) at 50 mK for the same sample with the average loop circumference \( C \) and lead length \( L \) indicated. The superconducting (S), destructive regime (D), and normal (N) states are shown. Zero resistance is not expected at \( \Phi_0 \) (corresponding to a field of approximately 2,000 G for these samples) because the field is close to the critical field and the wide parts of the leads are normal.

Fig. 2. Resistance (R) vs. magnetic flux (\( \Phi/\Phi_0 \)) and temperature (T) for a sample with a circumference \( C = 384 \text{ nm} \) and lead length \( L = 370 \text{ nm} \) (A) showing the destructive regime near \( \Phi = \Phi_0/2 \) and a sample of \( C = 492 \text{ nm} \) and \( L = 375 \text{ nm} \) (B) showing a destructive regime at \( \Phi = (3/2)\Phi_0 \) but not near \( \Phi = \Phi_0/2 \). These two devices are on the same chip and adjacent to one another and should have similar coherence lengths. Each plot was constructed from R vs. \( \Phi/\Phi_0 \) traces taken at 100 mK intervals from 50 mK to 1.4 K.
\[
\ln \left( \frac{T_c(\alpha)}{T_{c0}} \right) = \psi \left( \frac{1}{2} \right) - \psi \left( \frac{1}{2} + \frac{\alpha}{2\pi k_B T_c(\alpha)} \right) \tag{2}
\]

where \( \psi \) is the digamma function (6–9). Including second order effects related to a nonzero wire width (9), we have

\[
\alpha = \frac{\hbar D}{2R^2} \left( \Phi_0 - n \right)^2 + \frac{\mu^2}{4R^2} \left( \frac{\Phi_0}{\Phi} \right)^2 + n^2 \left( \frac{1}{3} + \frac{\mu^2}{20R^2} \right) \tag{3}
\]

where \( D = \frac{4k_BT_c(\alpha)}{\pi} \) is the diffusion coefficient for disordered superconductors, and \( w \) is the line width. Here the onset of the destructive regime is given by \( T_c = 0 \) of the ring, yielding \( \alpha_c = 0.889k_BT_{c0} \). For Sample B, this theory correctly predicts a QPT, with critical flux values of 0.4569\( \Phi_0 \) and 0.5924\( \Phi_0 \), compared to our experimental observation of a significant deviation from the resistance background from 0.4 to 0.75\( \Phi_0 \). This suggests that the pairbreaking theory more accurately describes our system, even though it seems to also underestimate the width of the destructive regime. A further comparison between the G–L and depairing predictions and our experimental results is shown in Table 1 for Sample C. For this device both theories predict the correct trend, especially the significant difference between \( \Phi = (3/2)\Phi_0 \) and \( \Phi = \Phi_0/2 \) in Sample C. But neither theory predicts the experimentally observed destructive regime at \( \Phi = (3/2)\Phi_0 \) in this sample. This apparent failure of the theory at \( \Phi = (3/2)\Phi_0 \) may be simply due to an underestimation of \( \xi(0) \) in this device or due to the more complicated geometry of our devices compared with those considered in the theoretical models.

The attainment of the destructive regime made it possible to examine recent predictions regarding quantum fluctuations in the destructive regime based on pair breaking theories (6–9), which provided a framework for understanding fluctuation enhanced electrical conductivities (6, 7) or the magnetic responses from the fluctuating persistent currents (8, 9) in the destructive regime. Similar to the classical superconducting fluctuations observed above the critical temperature, diagramatic calculations revealed three dominant corrections to the conductivity, the Aslamasov–Larkin, density of states, and Maki–Thompson corrections (6, 7). The Aslamasov–Larkin correction, corresponding to current carried by the fluctuating Cooper pairs, always enhances conductivity. Meanwhile these fluctuating Cooper pairs remove electrons from the Fermi surface and reduce the conductivity, leading to negative density of states correction. The Maki–Thompson term, reflecting Andreev scattering off fluctuating pairs, is more complicated and can be a positive or negative correction (7). Furthermore, three regimes, the classical, intermediate, and quantum regimes, were identified (6, 7). In the classical regime, at high temperatures and low pair breaking energies the Aslamazov–Larkin term generally dominates, leading to the well-known fluctuation enhanced conductivity of the form (27)

\[
\delta\sigma(0, T) = \frac{\pi e^2}{16\hbar A} \frac{\Phi(0)}{T} \tag{4}
\]

where \( A \) is the cross sectional area. Interestingly, at zero temperature and in the quantum regime, the Maki–Thompson and density of states correction are expected to both suppress conductivity and overpower the Aslamasov–Larkin term resulting in a suppressed conductance. As the temperature increases to reach the intermediate regime, the density of states correction becomes important, leading to an expected fluctuation conductivity of (7)

\[
\delta\sigma(\alpha, T) = \frac{\pi D e^2}{12\sqrt{2}} \frac{T^2}{(\alpha - \alpha_c(T))^{3/2}} \tag{5}
\]

Different from the previous cylinder work where residual resistance observed in the destructive regime was attributed to sample inhomogeneity rather than superconducting fluctuations (25, 26, 29), the reduced sample resistance in the destructive regime clearly comes from superconducting fluctuations because of the ultrasmall sample size. The thermal superconducting fluctuations above the mean-field transition, \( T_{c0} \), expected to be dominated by the Aslamasov–Larkin mechanism in the classical regime, were explicitly demonstrated in our measurements (Fig. 3/4). However, no reduced sample conductance expected in the quantum regime was observed. Given the small \( C \) values of our samples, which lead to large values of \( \alpha \) quantum fluctuations should be at least substantial in our ultrasmall loops at 50 mK, which corresponds to \( T/T_{c0} = 0.039 \). This could result from the normal-superconductor boundary within our loop structure where Andreev reflections should take place, leading to an additional positive conductance correction. Also the side branches themselves may be a large perturbation on either the effective pair breaking energy or the Maki–Thompson correction. Nevertheless, as shown in Fig. 3B, our devices show good agreement with the predictions for the intermediate regime.

A central feature of the LPdG effect, that the destructive regime is strongly affected by the presence of a side branch on the loop (3, 4), was readily tested in the current experiment. In Fig. 4 we show results obtained from two loop structures with similar \( C \) but different \( L \) values. While the sample with short side branches showed a robust destructive regime with full normal state resistance over a wide range of flux, the other, with long side branches, was found to be fully superconducting near \( \Phi = \Phi_0/2 \) at the lowest temperatures. Therefore a sufficiently long side branch does suppress the destructive regime, as predicted (3, 4). Evidently the side branch functions as a reservoir of condensation energy, which balances out the kinetic energy rise in the loop and helps restore global phase coherence by converting fluctuating pairs into a superconducting condensate.

**Table 1. Comparison between predictions of the pair breaking theory and the mean-field Ginzburg–Landau theory to the experimental results for Sample C**

| Flux | \( \alpha/k_BT_{c0} \) | \( T_c(\alpha) \) (mK) | \( T_e \) (GL) (mK) | \( T_e(\text{Meas}) \) (mK) |
|------|----------------|----------------|----------------|----------------|
| \( \Phi = \Phi_0/2 \) | 0.53 | 700 | 750 | 320 |
| \( \Phi = (3/2)\Phi_0 \) | 0.87 | 100 | 400 | <50 |

**Fig. 3.** (A) Change in conductance vs. \( 1/T \) in zero field for Sample B. The dashed line indicates fit to the linear behavior expected from Aslamasov–Larkin theory for fluctuation enhanced conductance in one dimension (27), yielding \( T_{c0} = 1.25 \text{K} \) consistent with that obtained directly from R(T) measurements. (B) Change in conductance vs.\( \alpha/k_BT_c \) at four different temperatures, plotted such that linear behavior corresponds to the expected fluctuation enhanced conductivities within the intermediate regime (7).
quantitatively there exist discrepancies. In particular we observe a resistive state for a lower than expected pair breaking parameter as well as a destructive regime in at $\Phi = (3/2)\Phi_0$ in Sample C. These discrepancies could arise from errors in estimating the superconducting coherence length or from our measurement geometry. Specifically the nonzero line width of our ring and side branches implies that an applied magnetic field will induce screening currents in them, which may add kinetic energy to the system. Moreover, the the finite magnetic field will also suppress the amplitude of the order parameter, $|\psi| (30)$. For Sample C, this effect will suppress the order parameter to $0.97|\psi_0|$ at $\Phi = \Phi_0/2$ and $0.72|\psi_0|$ at $\Phi = (3/2)\Phi_0$, helping to explain the full destructive regime seen in this device at $\Phi = (3/2)\Phi_0$. In spite of these issues the destructive regime in this system provides a useful system to test the predictions for superconducting fluctuations near a QPT.

The QPT seen in our ultrasmall Al loops is different from the two-dimensional superconductor-insulator transitions studied previously (31). Here the sample is of a finite size and therefore long-range phase coherence is not a defining feature of the state on either side of the QPT. Such a QPT has been encountered previously in finite-size systems (32). Ultrasmall superconducting structures may then provide us with a simple model system in which QPT and quantum fluctuations may be accurately controlled by device design and applied flux. Manipulating phase coherence among fluctuating pairs using a side branch to the ultrasmall loop demonstrated in this work should motivate further exploration of macroscopic quantum coherence engineering, a possible pathway to very high temperature superconductivity.

Materials and Methods

All devices used in this experiment were defined through a lift-off procedure and metalized using rapid thermal deposition of 31 nm 99.9999% (6 N) pure Al. The smallest feature sizes of the devices were written using a Leica EBPG-150 etching tool and lithography tool using a single layer PMMA e-beam resist, yielding a 40-nm line width for our smallest features. All devices presented in this work were fabricated at the same time using an identical procedure, which should yield similar levels of disorder in all devices. The values of $\xi(0)$ were estimated using the measured geometry of each sample and from the upper critical field of the devices, which yielded $\xi(0)$ values ranging from 97 to 126 nm for the devices used in this work. In addition, $\xi(0)$ was estimated from the measured resistivity and yielded similar results.

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