Possible observation of phase separation near a quantum phase transition in doubly connected ultrathin superconducting cylinders of aluminum

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The kinetic energy of superconducting electrons in an ultrathin, doubly connected superconducting cylinder, determined by the applied flux, increases as the cylinder diameter decreases, leading to a constructive regime around half-flux quanta and a superconductor to normal metal quantum phase transition (QPT). Regular step-like features in resistance vs. temperature curves taken at fixed flux values were observed near the QPT in ultrathin Al cylinders. It is proposed that these features are most likely resulted from a phase separation near the QPT in which normal regions nucleate in a homogeneous superconducting cylinder.

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The existence of a constructive regime near half-flux quanta in ultrathin, doubly connected superconductors provides a rather unique mechanism for destroying superconductivity in samples with restricted geometry. In a doubly connected system, such as a ring, fluxoid quantization demands that the equilibrium-state superconducting coherence length, $\xi(0)$, the kinetic energy of the superconducting electrons near half-flux quanta will exceed the superconducting condensation energy, leading to the suppression of superconductivity by increasing kinetic rather than repulsive interaction energy $\xi(0)$. Experimentally, this prediction was confirmed in a cylindrical geometry because $\xi(0)$ tends to be larger in a cylinder than in a ring when the two have similar diameters.

Cylinders possessing a destructive regime may be considered as an one-dimensional (1D) system. Motivated by an early experiment on possible observation of quantum phase slips, 1D superconductors have been studied extensively experimentally in recent years using singly connected nanowires. Our work on doubly connected 1D superconductors has been focused primarily on destructive-regime physics, in particular, the existence of a novel quantum phase transition (QPT) tuned by kinetic energy, as opposed to wire thickness, and the exotic normal state in the destructive regime. The kinetic energy in cylindrical samples is determined by applied flux, which can be controlled rather precisely, allowing detailed measurements on this QPT. In this Letter we report the observation of a possible phase separation in a homogeneous 1D superconducting state near the destructive regime in ultrathin Al cylinders.

Ultrathin cylinders of Al were fabricated by evaporating Al onto rotating quartz filaments, pulled from quartz melt and attached to a thin glass slide with a notch cut out. The Al film thickness was measured by a quartz crystal thickness monitor during deposition. The diameter of the cylinder was calculated from the period in resistance oscillation, using a natural period of $h/2e$ in magnetic flux, as done previously. A scanning electron microscopy (SEM) study was performed in several cases after all low-temperature measurements had been carried out to measure the diameter directly and check the structural homogeneity of the cylinder. The two methods of determining diameter yielded similar values. Different from the previous work, we have pursued 4-wire measurements by attaching fine Au wires directly to the cylinder spanning the gap of the glass slide using Ag epoxy or paste, as opposed to using films evaporated on the glass slide as the only measurement leads. The length between the voltage leads defined by two Ag epoxy or paste dots is usually more than 100 $\mu$m. Unfortunately, electrical contacts with the ultrathin cylinder do not always survive to low temperatures, in which case two-wire measurements were carried out. Cylinders were manually aligned to be parallel to the magnetic field. Electric transport measurements were carried out in a dilution refrigerator equipped with a superconducting magnet with a base temperature below 20 mK. All leads entering the measurement enclosure were filtered by RF filters working at room temperatures.

Figures 1a and 1c show the resistance as a function of temperature, $R(T)$, at different applied flux values from 0 to $\Phi_0/2$ for Cylinders Al-1 and Al-3, respectively. Parameters for these and other cylinders used in this study are summarized in Table I. Cylinder Al-1 was one of the samples used in the original experiment on the destructive regime. As the system approaches $\Phi_0/2$, regular step-like features, identified alternatively as minima in $dR/dT$, are seen at fixed resistance values. At low fields, these features become less distinct, and disappear when the field is sufficiently small. Even though a single step is seen at zero fields in Al-1, Al-3, and several other samples we measured, it always disappears at slightly higher fields. Although the precise physical origin for this step...
Essentially identical step-like features in comparable magnetic fields suggest that the step-like features are not due to 2-wire measurements because of the following observations. First, the diameter of the cylinder is sufficiently close to that required for possessing a destructive regime. For Al-4, which is only slightly larger than Al-1 and Al-3, the step-like features are present even at 10 nA. More importantly, at high bias currents, some step-like features presented at lower currents actually disappeared, again inconsistent with the PSC picture; Finally, we measured $R(T)$ at fixed magnetic field with a battery rather than a digital current source that may have introduced electrical noises to the sample despite the damping of the RF filters. Since electrical noises are known to generate PSCs, stronger step-like features are expected for digital current source measurements if these features were due to PSCs. Experimentally, the opposite is true.

Figure 2 shows $R(T)$ traces at fixed flux values up to half-flux quantum for cylinders with a diameter slightly larger than that required for destructive regime (Table I). Multiple, but relatively irregular step-like features were observed in Al-4, which is only slightly larger than Al-1 and Al-3. The irregularity in the step-like features appears to be related to the slight variation in diameter (Table I) as revealed by the $R(H)$ measurements. The overall trend of the step-like features are very similar to those found in Al-1 and Al-3. In Al-5, which is larger in diameter but uniform, only a couple of step-like features were found. For Al-6, with the largest diameter (Fig. 2d), no step-like features were observed. This systematic behavior was observed for all cylinders (9 in total) in which the presence of such step-like features was examined closely, suggesting that the step-like features can be induced by magnetic field so long as the diameter of the cylinder is sufficiently close to that required for possessing a destructive regime.

For the two samples shown in Figs. 1a and b, the empirical $\Phi - T$ phase diagrams are constructed (Fig. 3a and b). The phase diagrams suggest that for cylinders with sufficiently small diameters to host a destructive regime, the step-like features were found near the QPT between the superconducting and normal ground states at $T = 0$. If we consider the diameter of the cylinder as the third...
FIG. 2: a-c) $R(T)$ curves at various flux values for cylinders with a larger diameter (Table I). The magnetic field corresponding to $\Phi_0/2$ is 460-533 G for Al-4 which is slightly non-uniform in diameter, 295 G for Al-5, and 190 G for Al-6. The measurement current was 100 nA for all three cylinders; d) An SEM picture for Al-6. The Al grains are seen to be quite uniform with an average size of $69 \pm 16$ nm. The diameter measured in the SEM image is $268 \pm 10$ nm, as compared to $263 \pm 7$ nm obtained from the $R(H)$ measurements.

axis in the parameter space, the above results seem to suggest that the step-like features emerge in a quantum critical regime. It is interesting to note that the appearance of the step-like features shown in Figs. 1 and 2 is accompanied by a broadening of $R(T)$, a feature very similar to that observed near a superconductor-insulator transition (SIT) in 2D [15] homogeneous systems. Even though whether the superconductor-normal metal transition at onset of the destructive regime is a continuous QPT featuring a critical regime is yet to be established, it is likely that whatever drives this QPT is also responsible for the emergence of the regular step-like features.

FIG. 3: Empirical phase diagram for Al-1 and Al-3. The upper curve marks the highest $T$ at which a step was identified at fixed flux while the lower curve shows the onset of finite resistance.

In 2D SIT, the QPT is widely believed to be driven by phase fluctuation due to the suppression of fluctuation in the number of Cooper pairs, $N$. Because of the relation, $\Delta N \Delta \phi > 1$, where $\phi$ is the phase of the superconducting order parameter, the suppression in $\Delta N$ enhances $\Delta \phi$. However, $\Delta N$ is not suppressed near the destructive regime. Therefore it seems that even though the phase fluctuation may be present because of the reduced dimension, it can not dominate the QPT. A different path must be explored in order to understand this QPT and the accompanied step-like features. In this regard, the remarkable regularity of the latter as revealed in Fig. 1 may provide useful hints. To quantify this regularity, in Fig. 4a, we plot $R_{\text{step}}$, at which the step-like features were found, as a function of $n$, which denote the order for the emergence of the step-like feature as $T$ increases (Fig. 1). We see clearly that $\log_{10} R_{\text{step}} \propto n$, namely, $R_{\text{step}}$ grows exponentially.

To explain such exponential growth in $R_{\text{step}}$, and the emergence of the step-like features, we propose the following phenomenological model. As the system approaches the destructive regime, normal regions will nucleate in a homogeneous superconducting state. Each normal region will encircle the entire cylinder, forming a ring, or a band, referred here as a normal band. The first normal band will form near the center of a uniformly superconducting cylinder as $T$ is raised. With increasing $T$, each of the two superconducting sections will break into two sections, followed by breaking the 4 superconducting sections, and so on (Fig. 4b). Such a bifurcation process can lead to an exponential growth in the number of normal bands, $N$, given by $N = 2^n - 1$. If all normal bands have the same length, we have $R_{\text{step}} = NR_1$, where $R_1$ is the resistance associated with a single normal band, leading to evenly spaced steps on a logarithmic scale as
seen experimentally.

Further analysis shows good self consistency in this normal-band model. The slope in Fig. 4a is only slightly smaller than expected log\(_{10} 2\). The value of \( R_1 \) is also consistent with that expected for a single normal band. The length of a normal band, \( L_b \), should be \( 2\xi(0) \) based on energetic considerations (see below). However, \( R_1 \) should correspond to the resistance of a length twice of \( \xi(0) \), the charge imbalance length \([8]\). Typically \( \xi(0) << \Lambda_q \). Therefore, \( R_1 = 2\Lambda_q \cdot \rho_n / A \), where \( A \) is the cross-section area, similar to that for PSCs \([11]\). Based on the experimental value of \( R_1 \), \( \Lambda_q \approx 2\mu m \) for Al-1 and 3, a very reasonable number for Al \([11, 13, 14]\). In principle, \( \Lambda_q \) is a function of the applied field and temperature \([11, 13, 14]\). Therefore \( R_1 \) at different \( \Phi \) values should be different. However, our calculations show that \( \Lambda_q \) varies within 10% for all curves with step-like features in Fig. 1. Such a variation in \( R_1 \) is invisible in a logarithmic plot.

Even though this picture of normal-band bifurcation seems to provide a consistent account of our data, it is surprising that a regular spatial variation of the order parameter should be allowed as this would in general cost energy. On the other hand, as the destructive regime is approached, the free energy of the normal state is only slightly higher than that of the superconducting state because of the large \( v_s \). To minimize the free energy cost, a normal band should only be long enough to support a normal-band formation-2\( \xi(0) \). Once a normal band is formed, two S-N interfaces should bring about an interface energy. The applied field in this case is perpendicular rather than parallel to the interface, different from the typical situation considered in bulk Type I or Type II superconductors. The energy associated with such an interface has not been calculated. Two adjacent superconducting sections may also be coupled by Josephson coupling, likely to lead to a gain (lowering) of free energy. All these factors have to be considered to provide an energetic underpinning for the normal-band formation.

It is interesting to note that steps in \( R(T) \) were reported long ago in Al cylinders with a diameter larger than or equal to 1.4 \( \mu m \) in a narrow temperature range \([10]\). These cylinders were too large to possess a destructive regime. Most importantly, all steps seen at finite fields were also found at zero fields. Therefore the physical origin of these steps, not identified in the original work, cannot be the same as what we have observed in ultrathin superconducting cylinders. It was proposed previously that a heterogeneous mixed state featuring isolated superconducting spots would be formed at the high-temperature part of the superconducting transition due to impurity and strain effects \([17]\). It was further predicted that the presence of these superconducting spots will lead to steps in \( R(T) \). No steps were actually observed in their \( R(T) \) curves taken on cylinders with a diameter larger than or equal to 1.2 \( \mu m \) \([17]\), which were again too large to exhibit a destructive regime.

In summary, we have observed step-like features near a QPT at the onset of the destructive regime in ultrathin, doubly connected superconducting cylinders. A tentative model based on phase separation is proposed to explain the emergence of these step-like features. More theoretical input and further experimental studies will be needed to fully understand the physical origin of these step-like features. For example, reducing the wall thickness of the cylinder will increase the amount of disorder and decrease the orbital effects. Varying the magnetic field angle with respect to the axis of the cylinder, which were found to give rise to Abrikosov vortices in a large cylinder, may shed light on the microscopic origin of the proposed phase separation near the QPT in our ultrathin cylinders.

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