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 destructive regime around half-flux quanta and a superconductor to normal metal quantum phase transition (QPT). Regular steplike features in resistance versus 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 resulting 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 destructive regime near half-flux quanta in ultrathin, doubly connected superconductors [1] 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 superfluid velocity v_s satisfy the relation $v_s = (2\hbar/m^* d)(p - \Phi/\Phi_0)$, where $\hbar$ is Planck constant, m^* is the effective Cooper pair mass, d is the ring diameter, Φ is the applied flux, Φ_0 $(=h/2e)$ is the flux quantum, and p is an integer minimizing v_s [2]. As d decreases, v_s increases. When d becomes smaller than the zero-temperature 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 [1]. 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 [3].

Cylinders possessing a destructive regime may be considered as a one-dimensional (1D) system. Motivated by an early experiment on possible observation of quantum phase slips [4], 1D superconductors have been studied extensively experimentally in recent years using singly connected nanowires [5–9]. 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 [6], and the exotic normal state in the destructive regime [10]. The kinetic energy in cylindrical samples is determined by the 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 [2]. 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 previous work, we have pursued 4-wire measurements by attaching fine Au wires to the cylinder using Ag epoxy or paste. The length between the voltage leads defined by two Ag epoxy or paste dots is usually more than 100 μm. Unfortunately, electrical contacts with the ultrathin cylinder do not always survive to low temperatures, in which case 2-wire measurements were carried out. Cylinders were manually aligned to be parallel to the magnetic field. Electrical 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 1(a) and 1(c) 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 [3]. As the system approaches $\Phi_0/2$, regular steplike 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 is not known, we believe that it is a sample-specific feature

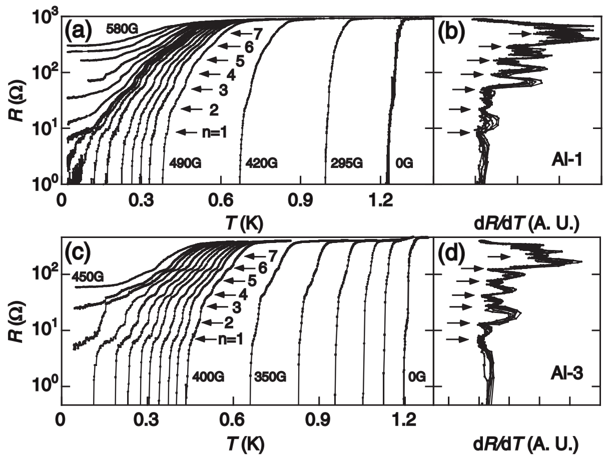

FIG. 1. (a) Resistance as a function of temperature, $R(T)$, at different applied flux values for cylinder Al-1; steps in $R(T)$ are marked by arrows. n denotes the order for the step to appear as the temperature is increased. Between 490 and 580 G, the field values are 500, 505, 510, 515, 520, 524, 526, 527, 528, 530, 535, 540, 550, and 560 G. $\Phi_0/2$ corresponds to 585.5 G; (b) dR/dT for data shown in (a). Steps in $R(T)$ shown in (a) correspond to minima in dR/dT; (c) $R(T)$ curves for Al-3. Between 400 and 450 G, the field values are 405, 410, 415, 420, 425, 430, 435, 440, 441.4, and 445 G. $\Phi_0/2$ corresponds to 580 G; (d) dR/dT for data shown in (c). The bias currents were 25 nA for Al-1 and 100 nA for Al-3.

unrelated to those seen in higher fields. Clearly, the regular steplike features near the half-flux quantum are induced by magnetic field.

Even though both samples shown in Fig. 1 were measured in a 2-wire configuration, the steplike features are not due to 2-wire measurements because of the following reasons. First, these features were seen in both 2- and 4-wire samples (see below). Furthermore, in a control experiment, 2- and 4-wire measurements were carried out on the same cylinder with multiple leads ($d \approx 0.2$ μm). Essentially identical steplike features in $R(T)$ were found in both cases. Sample inhomogeneities featuring variation in local T_c can also be excluded from being the cause of the regular steplike features. To begin with, SEM studies have shown that our cylinders are quite uniform. For those on which sharp SEM images were obtained, grains of Al with rather uniform size were seen. In addition, if the sample were inhomogeneous with separate regions of different T_c values, the superconducting transition in zero fields would not be as sharp as what was observed experimentally. The regular steplike features would also have persisted down to low and zero fields, inconsistent with experimental result.

Can the steplike features be due to phase slip centers (PSCs) formed at sections of the sample with a small local critical current (I_c) [11]? We believe that they are not, based on the following observations. First, the detailed evolution of the steplike features as a function of magnetic field suggests that the steplike features were not caused by PSCs. Second, we measured a cylinder (Al-3) with different bias currents. It appears that the steplike features are present even at 10 nA. More importantly, at high bias currents, some steplike features present 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 steplike 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, steplike features were observed in Al-4, which is only slightly larger than Al-1 and Al-3. The irregularity in the steplike 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 steplike features is 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 steplike features were found. For Al-6, with the largest diameter [Fig. 2(d)], no steplike features were observed. This systematic behavior was observed for all cylinders (9 in total) in which the presence of such steplike features was examined closely, suggesting that the steplike 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. 1(a) and 1(b), the empirical Φ-T phase diagrams are constructed [Figs. 3(a) and 3(b)]. The phase diagrams suggest that, for cylinders with sufficiently small diameters to host a destructive regime, the steplike 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 axis in the parameter space, the above results seem to

TABLE I. Parameters of cylinders reported in this work. $\xi(0)$ is estimated from the parallel upper critical field $H_{c2}^{\parallel}$ [$\xi(0) = \sqrt{3}\Phi_0/\pi t H_{c2}^{\parallel}$, where t is the film thickness]; the length of the cylinder L is defined by the distance between the edges of the adjacent Ag epoxy or paste dots; the mean-free path l_{el} is estimated from the normal-state resistivity ρ_N using a free-electron gas model. *m.c.* denotes measurement configurations (2- vs 4-wire).

	m.c.	d (nm)	t (nm)	L (mm)	$\xi(0)$ (nm)	l_{el} (nm)
Al-1	2	150	30	0.43	161	13
Al-3	2	151	33	0.29	190	16
Al-4	2	157–169	31.5	0.48	≈ 97	6.1
Al-5	4	212	30	0.11	146	7.9
Al-6	4	263	30	0.11	150	13

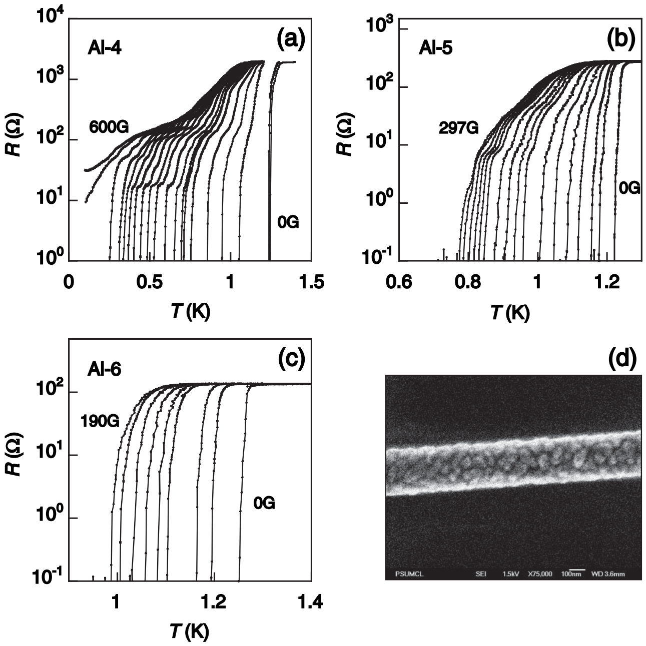

FIG. 2. (a)–(c) $R(T)$ curves at various flux values for cylinders with a larger diameter (Table I). The magnetic field corresponding to $\Phi_0/2$ is 460–533 G for Al-4, which is slightly nonuniform in diameter, 295 G for Al-5, and 190 G for Al-6. The measurement current was 100 nA for all three cylinders; (d) an SEM picture for Al-6. The Al grains are seen to be quite uniform with an average size of 69 ± 16 nm. The diameter measured in the SEM image is 268 ± 10 nm, as compared to 263 ± 7 nm obtained from the $R(H)$ measurements.

suggest that the steplike features emerge in a quantum critical regime. It is interesting to note that the appearance of the steplike 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 [12] 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 steplike features.

In 2D SIT, the QPT is 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 ϕ is the phase of the superconducting order parameter, the suppression in ΔN enhances $\Delta \phi$. However, ΔN 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 cannot dominate the QPT. A different path must be explored to understand this QPT and the accompanied steplike features. In this regard, the remarkable regularity of the latter as revealed in Fig. 1 may provide useful hints. To quantify this regularity, we plot in Fig. 4(a) the step resistance R_{step} as a function of n, which denotes the order for the steplike feature to emerge as T increases (Fig. 1). We see clearly that R_{step} grows exponentially with n.

To explain such exponential growth in R_{step}, and the emergence of the steplike features, we propose the following phenomenological model. As the system approaches the destructive regime, normal regions will nucleate. 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. 4(b)]. 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. 4(a) 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.

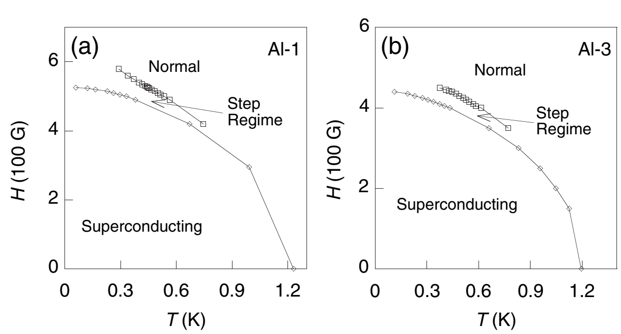

FIG. 3. Empirical phase diagram for Al-1 and Al-3. The upper curve marks the highest T at which a step was identified at fixed flux, while the lower curve shows the onset of finite resistance.

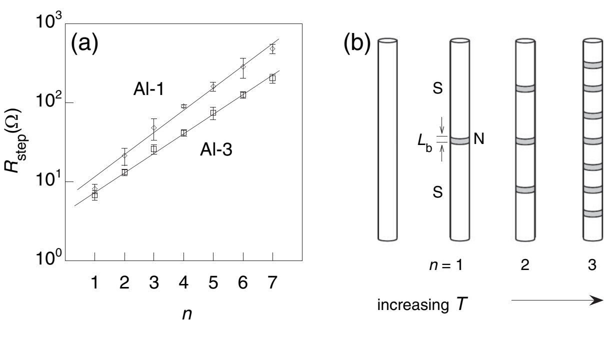

FIG. 4. (a) Step resistance as a function of the index n (Fig. 1) for Al-1 and Al-3. The lines indicate exponential dependence; (b) proposed bifurcation process for the normal-band formation in ultrathin cylinders.

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 Λ_Q, the charge imbalance length [13]. Typically, $\xi(0) \ll \Lambda_Q$. Therefore, $R_1 = 2\Lambda_Q \rho_N / A$, where A is the cross-section area, similar to that for PSCs [11]. Based on the experimental value of R_1, $\Lambda_Q \approx 2$ μm for Al-1 and 3, a very reasonable number for Al [14,15].

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 be only long enough to support two superconducting-normal (S-N) interfaces. As a result, $L_b = 2\xi(0)$. The 2 S-N interfaces should bring about an interface energy, which is difficult to estimate as the applied field in this case is *perpendicular* to the interface. 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 determine the energy of a normal band.

It is interesting to note that steps in $R(T)$ were reported for Al cylinders with a diameter larger than or equal to 1.4 μm [16], too large to possess a destructive regime. The physical origin of those steps, not identified in the original work, cannot be the same as what we have observed in ultrathin superconducting cylinders, as all steps seen there at finite fields were also found at zero fields. 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 predicted that 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 μm [17], again too large to possess a destructive regime.

In summary, we have observed steplike 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 steplike features. More theoretical input and further experimental studies will be needed to fully understand the physical origin of these steplike 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 was 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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