

Anomalous Little-Parks Oscillations in Mesoscopic Loops

H. Vloeberghs,⁽¹⁾ V. V. Moshchalkov,^{(1),(a)} C. Van Haesendonck,⁽¹⁾ R. Jonckheere,⁽²⁾ and Y. Bruynseraede⁽¹⁾

⁽¹⁾Laboratorium voor Vaste Stof-Fysika en Magnetisme, Katholieke Universiteit Leuven, B-3001 Leuven, Belgium

⁽²⁾Interuniversity Microelectronics Center, Kapeldreef 75, B-3001 Leuven, Belgium

(Received 18 February 1992)

Anomalous resistance oscillations have been observed in mesoscopic superconducting Al loops. As for the classical Little-Parks effect, the oscillation period is governed by the superconducting fluxoid quantization. The oscillation amplitude shows, however, an unusual magnetic-field and current dependence for low flux quanta, which is completely different from the Little-Parks oscillations.

PACS numbers: 74.90.+n, 72.15.Gd, 74.30.-c

It is well established that in small thin-walled superconducting cylinders, the transition temperature T_c is an oscillatory function of the magnetic field H [1]. The T_c oscillations, which are usually referred to as the Little-Parks (LP) oscillations [2], result from the fluxoid quantization, which implies that the sum of the externally applied magnetic flux ϕ and the flux induced by the superconducting screening currents is quantized in units of the flux quantum $\phi_0 = hc/2e$. While the screening currents are absent when $\phi = n\phi_0$, the screening currents reach their maximum amplitude when $\phi = (n + \frac{1}{2})\phi_0$, resulting in a maximum depression of T_c for the latter flux values. Experimentally [2-4] the LP oscillations of T_c can be observed as a periodic variation of the resistance with magnetic field at a fixed temperature $T = T^*$, usually taken near the midpoint of the normal-to-superconducting (N-S) transition. In this case the $R(T = T^*, H)$ dependence is characterized by a regular appearance of resistance minima in magnetic fields H_n corresponding to $\phi/\phi_0 = n$. The earlier experiments [2-4] also indicated that the N-S phase boundary is slightly shifted towards lower temperatures at higher measuring currents I , but the shape and relative amplitude of the resistance peaks are quite insensitive to the variation of I .

In this Letter we report on our experimental observation of unusual magnetoresistance oscillations $R(H)$ in mesoscopic superconducting loops, which are characterized by (i) a peculiar nonmonotonous variation of the $R(H)$ oscillation amplitude with magnetic field, (ii) a strong dependence of the oscillation amplitude on the applied current, and (iii) a field enhancement of the transition temperature T_c for a small number of flux quanta. These observations are directly related to the problem of quantum interference in elementary cells forming superconducting networks [5]. The advantage of using mesoscopic samples is that they are sufficiently small to fulfill the interference condition $\xi(T) \sim r$ (r is the loop radius) and at the same time large enough for studying typical quantum effects up to a μm length scale.

The mesoscopic Al loops are prepared using a Cambridge Instruments EBMF 10.5 electron-beam system operating at 20 kV. Using a bilayer resist, square liftoff

profiles are obtained with size ranging between 1 and 2 μm and a linewidth of 0.15 μm (see inset in Fig. 1). Thermal evaporation of high-purity Al (99.9995%) is used to deposit the 25-nm-thick Al lines in the liftoff profiles. The substrates are Si wafers on which a 50-nm-thick Si_3N_4 layer has been deposited. In order to obtain a smooth film surface without any cracks at the grain boundaries, the evaporation is done in a reduced helium atmosphere ($p \approx 10^{-3}$ Torr). The measurements are performed in a standard helium-3 cryostat allowing us to vary the temperature between 0.4 and 20 K. The applied magnetic field, which is produced by a superconducting coil, never exceeded 200 Oe. The position of zero magnetic field was determined within 1 Oe from the critical field of a bulk Al wire. Using a commercial four-terminal ac resistance bridge (Linear Research 400) voltage variations smaller than 1 nV could be detected.

The sheet resistance R_{\square} of the thin Al films used for the loop structures varied between 1.6 and 1.8 Ω/\square at 4.2 K, indicating the metallic character of our weakly disordered Al structures. The resistance ratio $R(300 \text{ K})/R(4.2 \text{ K})$ ranges between 2.02 and 2.10 and corresponds to a mean free path $l \approx 15 \text{ nm}$; i.e., l is comparable to the film thickness $t = 25 \text{ nm}$. The measured resistance ratio and the sheet resistance as well as our studies of the film topography with an atomic force microscope confirm the homogeneity and the continuity of the Al lines.

We have investigated in detail the N-S phase boundary for four square loops with a size of $1 \times 1 \mu\text{m}^2$ and for two square loops with a size of $2 \times 2 \mu\text{m}^2$. For the same loop size the data are completely reproducible from sample to sample. The unusual LP oscillations also appear to be an equilibrium property of the loop structures: Field cooling, zero-field cooling, and reversal of the magnetic field do not affect the results. In what follows we will focus our attention on the measurements of the $1 \times 1 \mu\text{m}^2$ loops.

Typical $R(T)$ transitions for different currents and applied fields are shown in Fig. 1. Several interesting features characterize the N-S transition. First, the $R(T)$ curve for $I = 0.3 \mu\text{A}$ and $H = 0$ shows a *marked resistance increase* at temperatures just above T_c . This peak in the $R(T)$ dependence is suppressed by applying a weak

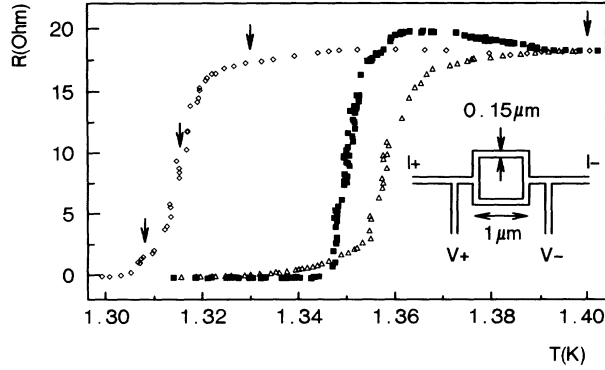


FIG. 1. Temperature dependence of the loop resistance for different currents I and fields H : $I=0.3 \mu\text{A}$, $H=0 \text{ Oe}$ (\blacksquare); $I=0.3 \mu\text{A}$, $H=20 \text{ Oe}$ (\triangle); $I=1 \mu\text{A}$, $H=0 \text{ Oe}$ (\diamond). The arrows mark the fixed temperatures where the $R(H)$ data of Fig. 2 have been obtained. Inset: The Al loop geometry used.

magnetic field or by increasing the measuring current. A similar resistance enhancement just above T_c was reported earlier by Santhanam *et al.* [6] for short Al wires and has been related to coherent phenomena in small superconducting samples and to the presence of an N/S interface [7]. More details about the influence of current and magnetic field upon the anomalous $R(T)$ peak in mesoscopic Al loops will be presented elsewhere [8]. Second, the transition temperature T_c decreases with increasing current, in agreement with the expected shift $\delta T_c(I) \propto I^{2/3}$ [9] and confirming the absence of sample heating for the indicated values of the applied current. Finally, for a constant current the N-S transition is shifted towards *higher temperatures with increasing field*; i.e., there is a distinct field enhancement of the superconducting transition temperature for magnetic fields corresponding to a small number $\phi/\phi_0 < 2$ of flux quanta. This field enhancement of T_c is obviously closely connected to the destruction of the anomalous resistance peak by a magnetic field.

In Fig. 2 we show the resistance variations as a function of the reduced flux ϕ/ϕ_0 for different temperatures (marked by the arrows in Fig. 1). At a relatively high temperature (upper curve in Fig. 2), classical LP oscillations with resistance minima at integer flux quanta are observed. At the lowest temperature (bottom curve in Fig. 2) a zero-resistance ($R=0$) gap opens up and pronounced LP oscillations appear only when the normal state is recovered by increasing the applied field (peaks at $\phi/\phi_0 = \pm 4.5$). At the intermediate temperatures one observes very sharp $R(H)$ peaks with an exotic field dependence of the peak heights at flux values $\phi = \phi_0(n + \frac{1}{2})$ and an anomalous peak, which for the $1 \times 1 \mu\text{m}^2$ loops appears close to $\phi = \pm \phi_0$. Measurements for other loop sizes reveal that the position of the latter feature scales inversely with the area of the Al lines, which form the loop and are penetrated by the magnetic field [8]. The $R(H)$ oscillations in the region $-2 \leq \phi/\phi_0 \leq +2$ are obviously not the

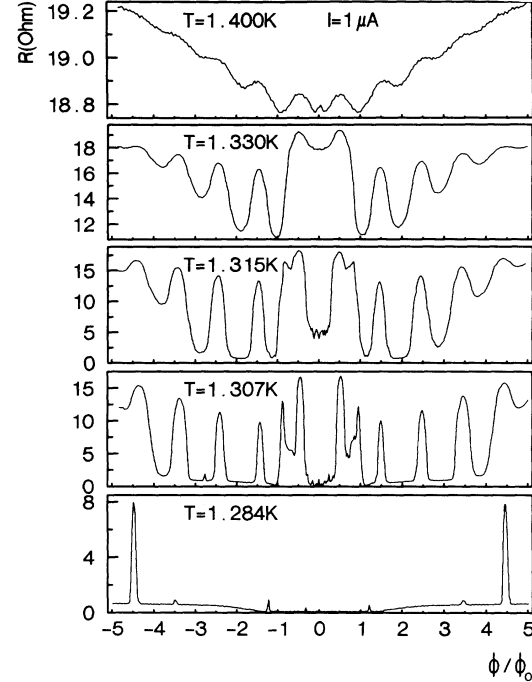


FIG. 2. Influence of temperature on the magnetic-flux (in units of the flux quantum ϕ_0) dependence of the resistance measured for a constant current $I=1 \mu\text{A}$. The different temperatures correspond to the arrows in Fig. 1.

ordinary LP oscillations.

As illustrated in Fig. 3 the $R(H)$ oscillations in the mesoscopic Al loops also have a peculiar current dependence for the same normalized temperature. By fixing $T/T_c(I)$ we take into account the shift $\delta T_c \propto I^{2/3}$ [9] caused by the current increase. Increasing the current also causes a gradual evolution of the $R(H)$ oscillations from an ordinary LP behavior towards an anomalous regime ($I=1 \mu\text{A}$) characterized by additional peaks at $\phi = \pm \phi_0$ and a nonmonotonous field dependence of the peak heights. A further current increase ($I=3 \mu\text{A}$) is accompanied by a strong narrowing of the $\phi/\phi_0 = \pm \frac{1}{2}$ peaks.

By measuring the shift of the midpoint of the superconducting transition in different fields (see Fig. 1) we can reconstruct the $T_c(H)$ phase boundary. The dotted line in Fig. 4 corresponds to the calculated phase boundary for $I \rightarrow 0$ [1]. The finite linewidth causes the weak parabolic $T_c(H)$ background on which the LP oscillations are superimposed. At low measuring currents ($I < 0.03 \mu\text{A}$) the experimental phase boundary coincides with the theoretical model. For larger measuring currents important deviations from the classical LP effect occur for $-2 < \phi/\phi_0 < 2$. For a higher magnetic flux the $T_c(H)$ variation again corresponds to the classical LP oscillations.

The inset of Fig. 4 shows the anomalous low-field part of the N-S boundary, which is obtained from the R vs H

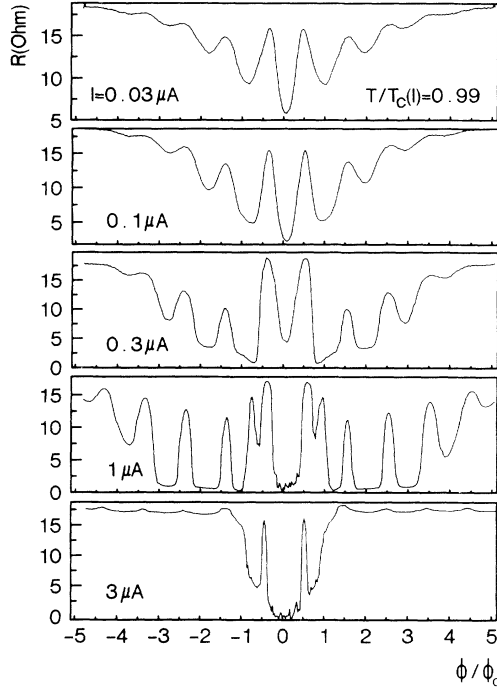


FIG. 3. Influence of the measuring current on the magnetic flux (in units of the flux quantum) dependence of the loop resistance measured at the same normalized reduced temperature $T/T_c(I) = 0.99$.

measurements (see Figs. 2 and 3). The displayed boundary corresponds to a threshold value $R = 2 \Omega$. Comparing the results shown in Fig. 4 and in the inset of Fig. 4 we may conclude that the details of the phase boundary (including the field enhancement of T_c) are insensitive to the choice of the threshold value for the superconducting order parameter, which is used to reconstruct the boundary. As illustrated by the inset of Fig. 4, the field enhancement of T_c also appears symmetrical about zero field and is observed up to $\phi/\phi_0 = 2$, corresponding to a magnetic field $H \approx 40$ Oe.

Empirically, the exotic variation of the $R(H)$ oscillation amplitude (see Fig. 2) can be directly related to an anomalous field dependence of the distance ΔT between the field sweep line at $T = T^*$ and the experimental $T_c(H)$ phase boundary, which are shown in Fig. 4. In the same way the modification of the $R(H)$ oscillation pattern with current (see Fig. 3) can be connected to an anomalous current dependence of the distance ΔT .

Theoretically [1], the phase boundary of mesoscopic superconducting loops is expected to depend strongly upon the ratio between the loop size and the Ginzburg-Landau coherence length $\xi(T)$:

$$\xi(T) = \xi(0) \left[\frac{T}{T - T_c} \right]^{1/2}, \quad (1)$$

where $\xi(0) = 0.85(\xi_0 l)^{1/2}$ for our Al lines, which are in

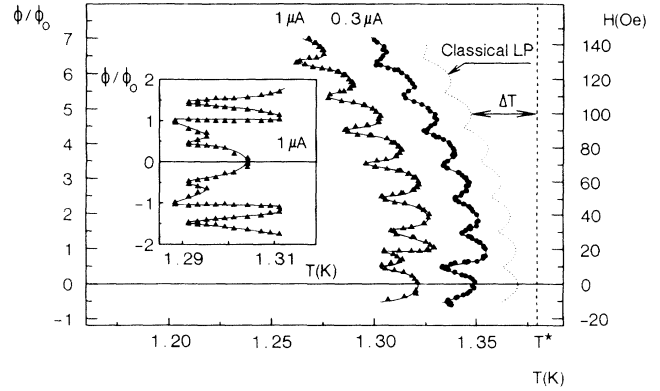


FIG. 4. The N-S phase boundary, which is obtained from the midpoint of the superconducting transition ($R = 9 \Omega$ in Fig. 1) for two different currents. The vertical dashed line represents a field sweep isotherm at $T = T^*$. The dotted line corresponds to the calculated classical Little-Parks phase boundary. Inset: A detailed view of the anomalous field enhancement of T_c for low flux quanta $\phi/\phi_0 < 2$. The data points for the inset have been obtained from the $R(H)$ magnetoresistance curves ($R = 2 \Omega$ in Fig. 2).

the dirty limit, i.e., $l \approx 15 \text{ nm} \ll \xi_0 \approx 1.6 \mu\text{m}$. The resulting coherence length $\xi(0) \approx 130 \text{ nm}$ is in good agreement with the value $\xi(0) \approx 120 \text{ nm}$ determined from the quadratic background of the $T_c(H)$ dependence in our samples (see dotted line in Fig. 4).

The phase boundary of a mesoscopic loop with vanishingly small linewidth has been calculated by Fink, López, and Maynard [10]. For a circular loop with radius r and symmetrical current leads the N-S transition is given by

$$\sin \left[\pi \frac{\phi}{\phi_0} \right] = \pm \left\{ 1 - \frac{1}{4} [AJ\xi^3(T)]^2 \right\}^{1/2} \sin \left[\pi \frac{r}{\xi(T)} \right], \quad (2)$$

where the constant A is determined by the normal-state transport properties. When the current density J is fixed, the solution of Eq. (2) for different ϕ/ϕ_0 values is determined by the ratio $r/\xi(T)$, with the temperature dependence of the Ginzburg-Landau coherence length $\xi(T)$ given by Eq. (1). For our low measuring currents the limit $J \rightarrow 0$ should be appropriate. The corresponding phase diagram, which consists of several chainlike normal areas (white areas) at values of $r/\xi(T) = 1, 2, \dots$, is shown in Fig. 5. The oscillating boundary between T_c , i.e., $r/\xi(T) = 0$, and $r/\xi(T) = \frac{1}{2}$ corresponds to the classical LP oscillations (boldfaced curve in Fig. 5) [1].

It is important to note that the result (2) has been obtained from the linearized Ginzburg-Landau equation. At lower temperatures the nonlinear term will become important and may destroy the additional normal chains, which appear for $r/\xi(T) > \frac{1}{2}$. Nevertheless, the existence of areas with a different superconducting order parameter (different critical current density) is still plausible [11]. The chain at $r/\xi(T) = 1$, which is closest to

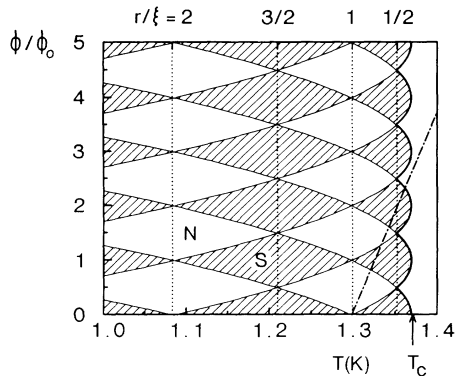


FIG. 5. The H - T phase diagram corresponding to Eq. (2). The white areas correspond to the normal (N) regions, while the hatched areas correspond to the superconducting (S) state. The dash-dotted line represents a field-sweep path. This tilted path takes into account the shift of the superconducting regions towards lower temperatures with increasing field strength, due to the finite linewidth of the mesoscopic loop structures.

T_c , should induce the most pronounced effect. The existence of this chain makes it possible to assume that the observed $R(H)$ oscillations are caused by successive crossing of the N-S boundaries by a tilted field-sweep line (dash-dotted line in Fig. 5). The tilt of the field-sweep takes into account the expected shift towards lower temperatures of the superconducting areas with increasing ϕ/ϕ_0 , due to the finite linewidth of the loop structure. Increasing the current density J in Eq. (2) leads to a shrinking of the superconducting areas in Fig. 5 and may cause the change of the $R(H)$ oscillations with current (see results shown in Fig. 3). A complete theoretical understanding of the N-S boundary will, however, also require an explanation for the anomalous resistance peak above T_c (see Fig. 1), which strongly depends on the magnetic field and the measuring current [6,7].

In conclusion, we have observed anomalous Little-Parks oscillations in mesoscopic superconducting loops. The oscillation amplitude shows a pronounced magnetic-

field and current dependence for small values of the normalized flux ϕ/ϕ_0 . These features are probably closely related to the anomalous field enhancement of T_c as well as to the anomalous resistance bump, which appears above T_c .

The authors are much indebted to G. Neuttiens for his help with the phase boundary measurements. This research has been supported by the Belgian Inter-University Institute for Nuclear Sciences (IIKW), the Inter-University Attraction Poles (IUAP), and the Concerted Action (GOA) programs. V.V.M. has obtained financial support from the Research Council of the Katholieke Universiteit Leuven, while H.V. and C.V.H. have benefited from the financial support of the Belgian National Fund for Scientific Research (NFWO).

- (a)Also at the Laboratory of High- T_c Superconductivity, Physics Department, Moscow State University, Russia.
- [1] M. Tinkham, Phys. Rev. **129**, 2413 (1963); P. G. de Gennes, *Superconductivity of Metals and Alloys* (Addison-Wesley, New York, 1989).
 - [2] W. A. Little and R. D. Parks, Phys. Rev. Lett. **9**, 9 (1962).
 - [3] R. P. Groff and R. D. Parks, Phys. Rev. **176**, 567 (1968).
 - [4] L. Meyers and R. Meservey, Phys. Rev. B **4**, 824 (1971).
 - [5] For a recent review, see *Coherence in Superconducting Networks*, edited by J. E. Mooij and G. Schön [Physica (Amsterdam) **152B**, 1988].
 - [6] P. Santhanam, C. C. Chi, S. J. Wind, M. J. Brody, and J. J. Bucchignano, Phys. Rev. Lett. **66**, 2254 (1991).
 - [7] Y. K. Kwong, K. Lin, P. J. Hakonen, M. S. Isaacson, and J. M. Parpia, Phys. Rev. B **44**, 462 (1991).
 - [8] V. V. Moshchalkov, H. Vloeberghs, C. Van Haesendonck, and Y. Bruynseraede (to be published).
 - [9] B. Pannetier, in *Quantum Coherence in Mesoscopic Systems*, edited by B. Kramer (Plenum, New York, 1991), p. 457.
 - [10] H. J. Fink, A. López, and R. Maynard, Phys. Rev. B **26**, 5237 (1982).
 - [11] H. J. Fink, V. Grünfeld, and A. López, Phys. Rev. B **35**, 35 (1987).