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Negative magnetoresistance in small superconducting loops and wires

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We report an experimental attempt to measure resistively the predicted periodic reentrant behavior of small superconducting loops in external magnetic fields. The superconductivity in the contact probes was deliberately weakened in order to minimize their influence on the loops. As expected, the loops exhibit Little-Parks oscillations near the top of the resistive transition. At lower temperatures, instead of the reentrant behavior, the samples show an unexpected negative magnetoresistance. We interpret this result as being due to a nonequilibrium situation existing between normal and superconducting regions in the sample.

The study of submicrometer-size metallic loops has been an exciting branch of solid-state physics over the past few years. In the case of normal (nonsuperconducting) metals, when the loop perimeter becomes less than or comparable to the electron phase-breaking length, there is a plethora of new phenomena,[1] manifesting the long-range spatial quantum coherence. In the case of a superconducting loop, there has been considerable theoretical work[2-5] addressing the expected critical behavior as its perimeter becomes smaller than $\xi(T)$, the superconducting coherence length. Previous experimental study[6] of loops made of a superconducting material has concentrated on their properties in the normal state, where the perimeter is much larger than $\xi(T)$. Here, we report on experiments designed to explore the properties of small superconducting loops and wires in external magnetic fields at temperatures near and below T_c.

The critical behavior[2-4] of an isolated superconducting loop depends on the ratio $\alpha \equiv R/\xi(T)$, where R is the radius of the loop. If the cross-sectional dimensions of the wire defining the loop are much smaller than $\xi(T)$, the critical field of the wire itself can be quite large. Under these conditions, when $\alpha \gtrsim \frac{1}{2}$ the loop is driven normal only when an external magnetic field is large enough to destroy the superconductivity in the wire. In this regime, one expects the magnetoresistance close to T_c to show oscillations with a period corresponding to a flux quantum in the loop as illustrated by the Little-Parks[7] experiment. In contrast, for $\alpha \lesssim \frac{1}{2}$ the loop is expected to be driven periodically into the normal state at any $T < T_c$ for a range of applied flux inside the loop close to $\Phi = (n + \frac{1}{2})\Phi_0$, where n is an integer and $\Phi_0 \equiv h/2e$ is the flux quantum. The width of this range over which the loop stays normal depends on the value of α; the smaller its value, the larger the range. Qualitatively this behavior is easily explained as follows. The phase gradient along the loop for an applied flux Φ is $\nabla\phi = \Phi/(R\Phi_0)$, with its maximum value at $\Phi = \Phi_0/2$. The phase boundary is defined by $\nabla\phi \simeq 1/\xi(T)$. When $\alpha > \frac{1}{2}$, this condition is never satisfied, whereas for $\alpha < \frac{1}{2}$ there is a range close to $\Phi/\Phi_0 = \frac{1}{2}$ over which the critical boundary exists. de Gennes[2] considered the effect of a dangling arm attached to the loop within the framework of the linearized Ginsburg-Landau (GL) formalism, and expected the diamagnetism to be substantially enhanced. It has also been predicted by Straley and Visscher,[4] using nonlinear GL equations, that while the enhancement is not as great as de Gennes predicted, the range over which a loop stayed normal did decrease as the length L of the arm increased; for $L > \xi(T)$ the loop always stays superconducting. An inductive measurement of an isolated loop would provide a direct means of testing these predictions. The extremely small signal expected in such an experiment, however, led us to an alternative resistive measurement using ion-etched contact probes of weakened superconductivity. If the weakly superconducting or normal probes do not have a significant influence on the loop, we expect to observe the reentrant behavior as abrupt changes in magnetoresistance for values of flux close to $(n + \frac{1}{2})\Phi_0$.

We chose aluminum as the material due to its relatively large $\xi(T)$. The first set of 65-nm-wide samples made[8] from a 34-nm-thick aluminum film is shown in Fig. 1. There are two separate loops of nominal radius $R = 0.38$ μm, and a 1.5-μm-long wire. One of the loops has a 0.21-μm dangling arm attached to it. The lighter rectangles in Fig. 1(a) show regions where the fine lines were

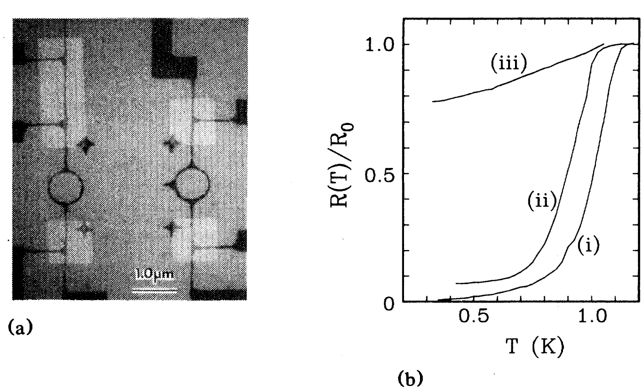

FIG. 1. (a) An electron micrograph of the aluminum loop sample. The lighter regions correspond to the ion-etched part. (b) The resistive transitions of the three samples at 5-nA (rms) bias current: (*i*) the loop with the dangling arm, (*ii*) the bare loop without the dangling arm, and (*iii*) the etched straight wire. The resistances are normalized to their respective values at $T = 1.2$ K for easier comparison.

thinned by argon ion etching. The straight wire was used to characterize the ion-etched portions of the sample. Four-terminal electrical measurements were performed in a sorption-pumped ^{3}He Dewar using a lock-in amplifier at a frequency of 27 Hz. The lowest temperature reached was $T \simeq 0.3$ K.

The resistive transitions of the three samples in Fig. 1(a) at a rms bias current of 5 nA are shown in Fig. 1(b). The loop without the dangling side arm does not go completely superconducting down to the lowest temperature, but reaches $\simeq 6\%$ of the normal-state resistance with the midpoint of the transition (taken to be the mean-field T_c) at $\simeq 0.9$ K. The loop with the side arm does reach the zero-resistance state with $T_c \simeq 1$ K. The damaged straight wire reaches only $\simeq 75\%$ of the normal-state resistance at 0.3 K. These results are obviously different from the behavior expected for typical samples made of Al thin films that show relatively sharp resistive transitions with $T_c \simeq 1.4$ K. We believe that the behavior shown in Fig. 1(b) is essentially the consequence of the proximity of the normal (etched) and superconducting (unetched) parts of the sample.

The typical response of the loop samples in an external magnetic field is shown in Fig. 2. For temperatures close to the top of the resistive transition, both loops showed oscillations in magnetoresistance (MR) corresponding to the Little-Parks oscillations previously seen in superconducting cylinders.[7] Figure 2(a) shows the MR for the loop without the dangling arm at $T = 1$ K. These oscillations confirm that the loops in the sample were continuous. The period of the oscillations is in good agreement with the estimate obtained from electron micrographs. As the temperature is lowered, the Little-Parks oscillations

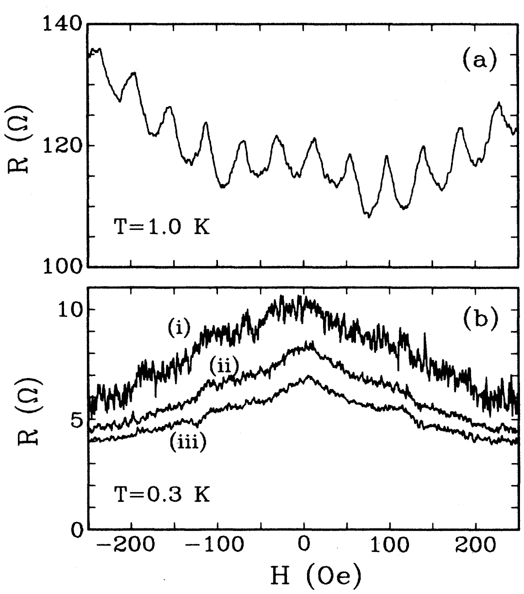

FIG. 2. (a) The magnetoresistance of the bare-loop sample at 1 K. (b) The magnetoresistance of the bare-loop sample at 0.3 K at three different rms bias currents: (*i*), 5 nA; (*ii*), 15 nA; and (*iii*), 25 nA. The magnitude of the negative magnetoresistance decreases as the bias current increases (see the text).

persist down to $\simeq 0.9$ K. Below $\simeq 0.9$ K, a negative MR near the zero field develops. At lower temperatures, as shown in Fig. 2(b), the negative MR is the dominant feature and there is no sign of the periodic oscillations. The noise observed in the traces is mostly random noise superposed on some reproducible structures. The amplitude of the negative MR increases with decreasing bias current to a few percent of the normal-state resistance. Similar behavior was observed for the loop with the dangling arm. The negative MR was also present for the ion-etched straight wire.

If the reentrant behavior were present in the loops, large, abrupt periodic oscillations in MR are expected in a temperature range below T_c. The lower end of this range is determined by the temperature at which $\alpha < \frac{1}{2}$. Using the measured resistance ratio of 1.8, we estimate[9] $\xi(0) \simeq 0.1$ μm. This provides only a lower estimate of $\xi(T)$ since the measured resistance includes a substantial section of the ion-etched wire. We find that while the reentrant behavior is expected at $T \gtrsim 0.88$, we see no clear evidence for it in either of the loops.

The negative MR that we do see, however, is quite unusual and unexpected. In superconducting samples, one expects positive MR near the resistive transition due to the suppression of superconducting fluctuations. In disordered thin films, the MR can be negative due to weak localization,[9] but the magnitude is typically a few parts in ten thousand or less. Another well-known mechanism for negative MR is the Kondo effect due to magnetic impurities[10] in metals. We believe that this is not relevant in our samples because most common magnetic impurities do not possess moments in Al, and also the characteristic field scale for this mechanism is of the order of a few kOe. Alternatively, we consider a nonequilibrium model based on the existence of normal-superconducting (N-S) interfaces between the etched and unetched portions of our samples. Previously, such a model has been used by Kadin, Skocpol, and Tinkham[11] to explain the magnetic field dependence of the resistance of phase-slip centers in microbridges. We find that this model can reasonably explain the negative MR.

At an N-S interface[12] the magnitude of the energy gap $\Delta(T)$ in the superconductor recovers to its full value over a length scale of $\xi(T)$. In the presence of an external current, for $(\Delta/k_BT) < 1$, almost all of the excitations from N propagate into S resulting in a quasiparticle current in the superconductor. The conversion of the quasiparticle current to the supercurrent occurs over a length scale of the charge-imbalance relaxation length, λ_{Q^*}. The consequence is an N-S boundary resistance corresponding to a length λ_{Q^*} for each such boundary. As Δ/k_BT increases, the spatial extent of the nonequilibrium region decreases and more of the total current is converted to supercurrent at the interface due to Andreev reflection. For $\Delta/k_BT > 1$, there still remains a boundary resistance corresponding to a length $\xi(T)$ in the superconductor, which is typically much smaller than λ_{Q^*}. In this situation, we expect positive MR just due to the suppression of superconductivity. Thus we are led to the $(\Delta/k_BT) < 1$, regime. We note that $\lambda_{Q^*} = (D\tau_{Q^*})^{1/2}$, τ_{Q^*} being the charge-imbalance relaxation time and D, the diffusion constant. τ_{Q^*} depends[11–13] on Δ/k_BT, the inelastic scattering time τ_i, and the pair breaking time τ_s. The current through the N-S boundary I, and the applied magnetic field H strongly affect the magnitude of τ_s. The final expression for λ_{Q^*} is

$$\frac{1}{\lambda_{Q^*}} \simeq \left[\frac{\pi\Delta(T)}{4k_BT}\right]^{1/2} \frac{1}{l_i(T)} \left\{1+\frac{l_i^2(T)}{\xi^2(T)}\left[\frac{1}{3}\left[\frac{I}{I_c(T)}\right]^2+\left[\frac{H}{H_{c2}(T)}\right]^2\right]\right\}^{1/4}. \quad (1)$$

$\Delta(T)$ is the superconducting gap, $I_c(T)$ is the critical current in zero magnetic field, and $H_{c2}(T)$ is upper critical field of the superconductor. $l_i = (D\tau_i)^{1/2}$ is the inelastic diffusion length. It is clear from Eq. (1) that λ_{Q^*} (and hence the corresponding resistance) decreases as the magnetic field increases. Further, the overall magnitude of the negative MR decreases as the bias current increases. This prediction is in good agreement with the experimental data shown in Fig. 2(b).

To confirm that the negative MR is indeed due to an N-S interface, we prepared[14] another sample [see Fig. 3(a) inset] in a different geometry. It consisted of a 35-nm-thick wire of two widths: a narrow region (3.35 μm long, 41 nm wide) with a wider region (2.6 μm long, 0.48 μm wide) at either end. N-S interfaces can be created between the narrow and wide regions by driving the narrow part normal with sufficient bias current below T_c. Figure 3(a) shows the $R(T)$ behavior of this sample measured with a small bias current of 10 nA. T_c is $\simeq 1.51$ K, typical of thin-film aluminum behavior. The sample shows a relatively sharp transition in zero field. As a magnetic field is applied at $T = 1.29$ K, the sample shows two transitions [curve (*i*), Fig. 3(b)]: one at the lower field corresponding to the wider portion of the wire and another at the higher field corresponding to the narrower portion of the sample.[9] Even though there are normal and superconducting boundaries between these two transitions, there is no obvious negative MR. As the temperature is increased, qualitatively the behavior remains the same, but the two critical fields (corresponding to the narrow and wide parts) become lower as expected. In order to have a N-S interface near zero magnetic field, we used a sufficiently large bias current to drive the narrow region normal. At a temperature $T \simeq 1.48$ K, with a 2-μA bias, the MR is shown as curve (*ii*) in Fig. 3(b). A negative MR is clearly present for $H < 20$ Oe. We wish to attribute this negative MR to the magnetic field dependence of λ_{Q^*} as given in Eq. (1). The resistance due to this charge-imbalance region is[15]

$$R_{Q^*} = \frac{2R_\square}{W}\left[1-\frac{\pi\Delta}{4k_BT}\right]\lambda_{Q^*}, \quad (2)$$

where $R_\square$ is the sheet resistance of the film and W is the width of the wire. Figure 4 shows the predicted change in resistance based on Eq. (2). We infer from the resistance at $H = 0$ that the ends of the narrow wire remain superconducting along with the wider parts in the situation rep-

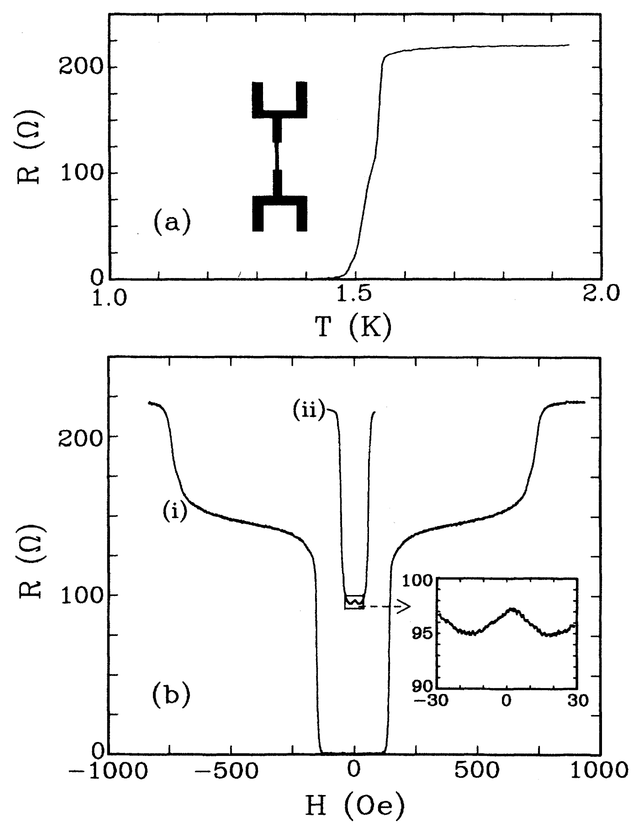

FIG. 3. (a) The resistive transition of a wire of two widths at 10-nA rms bias current. The inset shows the schematic of the sample. The dimensions are given in the text. (b) Curve (*i*) is the MR of the wire at $T = 1.29$ K at a 10-nA rms bias. Curve (*ii*) is the MR of the wire at $T = 1.48$ K at 2-μA rms bias. The inset provides a magnified view of the small field regime of curve (*ii*).

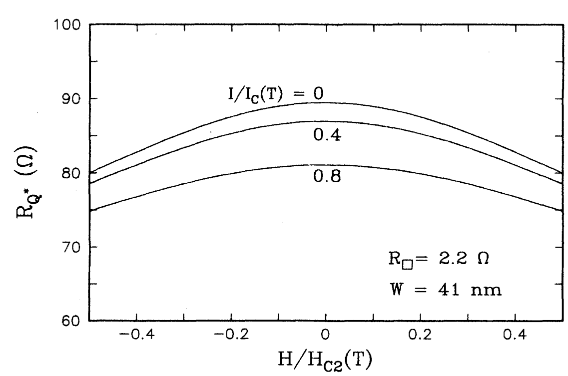

FIG. 4. Estimates of the variation of R_{Q^*} with external magnetic field in the charge-imbalance relaxation model for three bias currents, using Eq. (2). The behavior is similar to those shown in Fig. 2(b) and the inset of Fig. 3(b).

resented by curve (*ii*) of Fig. 3(b). Hence it is reasonable to take the relevant superconducting material parameters in Fig. 4 to be close to those of the narrow wire. We estimate $\Delta/k_B T = 0.44$ and $\xi(T) = 0.5$ μm based on the BCS theory. We need $l_i = 0.75$ μm in order to reproduce such reasonable curves for this sample. This is $\simeq 2$ times smaller than an estimate based on weak-localization experiments[9] and may be explained as due to additional charge-imbalance relaxation mechanisms such as gap anisotropy[16] in the presence of a large bias current. The qualitative agreement between Fig. 4 and the experimental behavior seen in Figs. 2(b) and 3(b) is quite good. A quantitative comparison is difficult due to the uncertainty in determining the background of the experimental curve. Also, Eq. (2) assumes that the sample length is much larger than λ_{Q^*}, which is only satisfied marginally in our sample.

We found that the range in temperature and bias current over which we could see the negative MR of the sample with two widths was quite narrow. In contrast, we could not avoid the negative MR for the samples with ion-etched portions. The origin of this difference may be that an N-S interface exists intrinsically for the ion-etched samples down to 0.3 K, whereas we had to create such an interface using a higher bias current for the sample with two widths. For the etched samples the condition $\Delta/k_B T < 1$ is satisfied at any temperature due to the spatial gradient of Δ, which is suggested by the broad resistive transitions. For the sample with two widths, $\Delta/k_B T < 1$ can be satisfied only in a narrow range because of its intrinsically sharper transition.

Another interesting aspect of the MR curves of the sample with two widths is shown in Fig. 3(b). Based on the geometrical measurements, we expect the resistance of the wide part to be only about 12% of the total resistance. As seen in curve (*i*), the resistance rise at the critical field of the wide part corresponds to $\simeq 65\%$ of the total resistance, indicating that a good fraction of the narrow wire is driven normal even though the critical field of the narrow part is about seven times larger. This is another manifestation of the N-S interface occurring within the narrow part of the wire.

In conclusion, we did not observe the reentrant superconducting behavior predicted for an isolated superconducting smaller than the superconducting coherence length. Instead, a large unexpected negative magnetoresistance was observed in samples designed to have both normal and superconducting regions. A simple model based on nonequilibrium superconductivity of a normal-superconducting interface can qualitatively explain many features of this negative magnetoresistance seen in these samples. Our results point out the complications to be anticipated in experiments[17] that require normal-superconducting boundaries.

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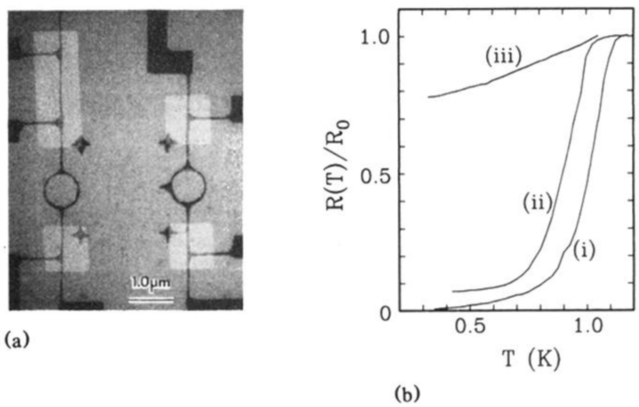

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