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Current-Induced Crossover of Flux Periodicity from *h*/2*e* to *h*/*e* in a Superconducting Nb Nano-Ring

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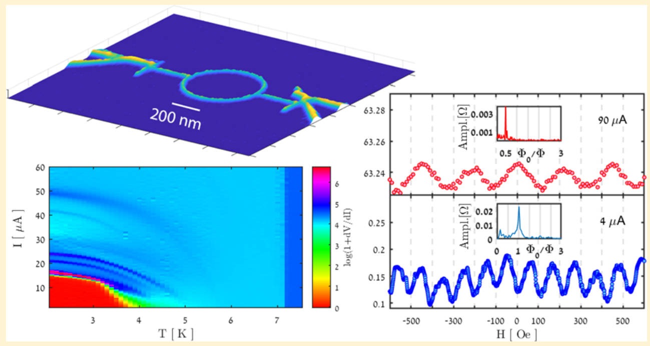
Supporting Information

ABSTRACT: Magnetoresistance measurements in a granular Nb nanoring reveal current-induced crossover between two distinct quantum coherence effects. At low bias currents, Cooper-pair coherence is manifested by Little−Parks oscillations with flux periodicity of $h/2e$. At high bias currents, magnetoresistance oscillations with flux periods of h/e are observed and interpreted as Aharonov−Bohm oscillations, reflecting the phase coherence of individual quasi-particles. The model explaining these data views the ring as a chain of superconducting grains weakly coupled by tunnel junctions. Low bias currents allow coherent tunneling of Cooper pairs between the grains. Increasing the current above the critical current of all the junctions creates a quasi-particles conduction channel along the ring, allowing for quantum interference of quasi-particles.



KEYWORDS: *Superconductivity, nanorings, quantum interference, Little−Parks effect, Aharonov−Bohm effect*

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Quantum phase coherence and interference effects, commonly associated with superconductors,[1] are also observed in mesoscopic normal metals at low temperatures so that the scattering of electrons is almost entirely elastic.[2] Such effects are experimentally manifested in magnetoresistance (MR) measurements, in which the resistance of a superconducting or a metal ring is measured as a function of the magnetic flux threading it. Mesoscopic normal-metal rings with narrow rims exhibit the Aharonov−Bohm MR oscillations with a flux period of h/e,[3] whereas superconducting rings exhibit MR oscillations with a flux period of $h/2e$, $2e$ being the hallmark of the electron pairing. In the present work we show a crossover of the flux periodicity in a granular Nb mesoscopic ring, from $h/2e$ at low bias currents to h/e at higher currents in a range below the depairing current. This crossover is explained by viewing the granular ring as a chain of superconducting islands weakly coupled by tunnel junctions. The $h/2e$ flux periodicity is associated with the Little−Parks effect in a ring consisting of Josephson junctions in the zero voltage state.[4] The magnetoresistance oscillations with a flux period of h/e are interpreted as an Aharonov−Bohm effect arising from phase coherence of quasi-particles flowing in the resistive channel created by the Josephson junctions in the voltage state.

Thin Nb films were grown by magnetron sputtering on SiO_2 substrates. X-rays diffraction (XRD) and reflection (XRR) measurements revealed granularity with grains size between 10 and 12 nm. Nb rings were patterned on these films using e-beam lithography followed by reactive ion etching (RIE). Altogether, seven samples with the same diameter (500 nm) and different rim sizes (20−80 nm) were measured. The effect reported below was observed in three samples with the lowest rim size. Here we report representative results obtained in one of these samples, with a diameter of 500 nm, and a rim's width and thickness of 23 and 20 nm, respectively. Figure 1 shows a scanning electron microscopy (SEM) image of this sample. Transport and magnetoresistance measurements were performed by employing a commercial physical properties measurements system (PPMS, Quantum-Design) for temperatures 3−10 K and bias currents between 1 and 128 μA.

Figure 2a shows the temperature dependence of the ring resistance for several measuring currents. A sharp drop of the resistance is apparent at $T_c \sim 7.2$ K for all the measuring currents, followed by a current-dependent broad transition. For increasing the measuring current up to 32 μA, superconductivity along the ring is gradually destroyed. For currents between 32 and 128 μA, a remarkable behavior is observed at low temperatures, namely, a decrease of the resistance with increasing temperature. The observed temperature and current dependence of the resistance become clear by viewing the granular ring as a chain of Josephson coupled superconducting

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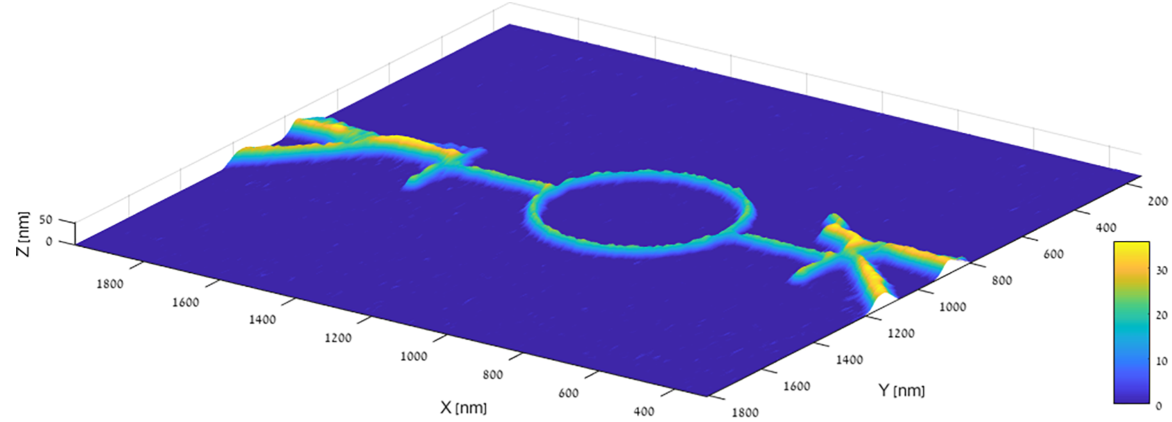

Figure 1. SEM image of the Nb ring with a diameter of 500 nm and rim's width and thickness of 23 and 20 nm, respectively.

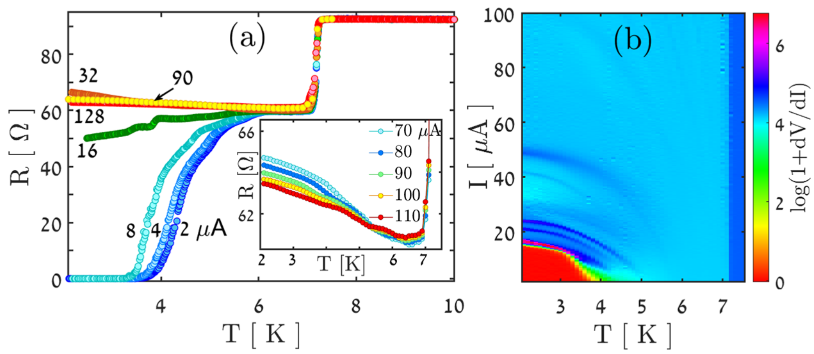

Figure 2. (a) Temperature dependence of the ring resistance for the indicated measuring currents. Inset: $R(T)$ for bias currents between 70 and 110 μA. (b) Color encoded diagram of the differential resistance, dV/dI, in the current–temperature plane. The dark arcs indicate the temperature dependence of the critical current of the different Josephson junctions in the ring.

islands. The initial sharp drop at ~7.2 K to approximately 65% of the normal resistance indicates the onset of superconductivity in the isolated superconducting islands within the ring. The broad transition for low bias currents (2–8 μA) is associated with gradual achievement of Josephson coupling between the superconducting islands, eventually driving the ring into a zero resistance state. In the intermediate current range (represented by the 16 μA in the figure), part of the Josephson junctions in the ring switch into the voltage state, resulting in nonvanishing resistance. This resistance saturates for larger currents, represented by the curves for 32–128 μA in the figure, indicating that *all* the Josephson junctions in the ring are in the voltage state. In this state, a continuous resistive channel associated with the quasi-particle current along the ring is established.[5] The quasi-particles are generated either by thermal excitations or due to breaking up of Cooper pairs by the current. The excited quasi-particles in each junction can tunnel already at voltages smaller than the gap voltage, resulting in a finite resistance, $R_{sg}(T)$, known as the "subgap resistance". The magnitude of $R_{sg}(T)$ is determined by the amount of excited quasi-particles and is given by $R_{sg}(T) = (n_{total}/n(T,I_b))R_N$, where n_{total} is the total density of electrons in the normal state, $n(T,I_b)$ is the density of the excited quasi-particles at temperature T and bias current I_b, and R_N is the normal resistance of the Josephson junction.[5] For a constant bias current, R_{sg} decreases with temperature as $n(T)$ increases. As the bias current increases, R_{sg} drops and its decrease with temperature becomes more moderate, indicating that the generation of the quasi-particles is dominated by the current. These effects are visible in the $R(T)$ data of Figure 2a for high bias currents, and more clearly in the inset to this figure. It is important to note that even for the largest measuring current the grains are still superconducting as the depairing current density, $J_{dp} = \frac{\Phi_0}{2\pi\mu_0\lambda^2\xi} = 1.5 \times 10^8$ A/cm^2, is an order of magnitude larger than the density of the largest bias current in our experiment. This, in fact, is also evident from the large drop of the resistance at 7.1 K, only a small part of which can be attributed to the effect of the superconducting electrodes. Furthermore, this large drop is followed by an almost constant resistance down to low temperatures, excluding an influence of the proximity effect caused by the electrodes. We note that the above value for J_{dp} estimated for bulk Nb, may be an overestimate for Nb films. As reported in ref 6, the depairing current density in Nb films may be as low as ~4×10^7 A/cm^2 at 3.5 K. However, even this low estimate is significantly larger than the current where the crossover to h/e periodicity is observed.

A further support to our view of the granular Nb ring as a chain of Josephson junctions with distributed critical currents is obtained from the I–V curves shown in Figure S-1 in the Supporting Information. The I–V curves are characterized by several voltage steps at different current values. These are clearly seen especially at low temperatures. A voltage step occurs when the current reaches the value of the critical current of a single or a group of Josephson junctions in the chain. To provide an even clearer picture, we show here, in Figure 2b, a color encoded diagram of dV/dI in the current–temperature plane. Voltage steps in this diagram are visible as a

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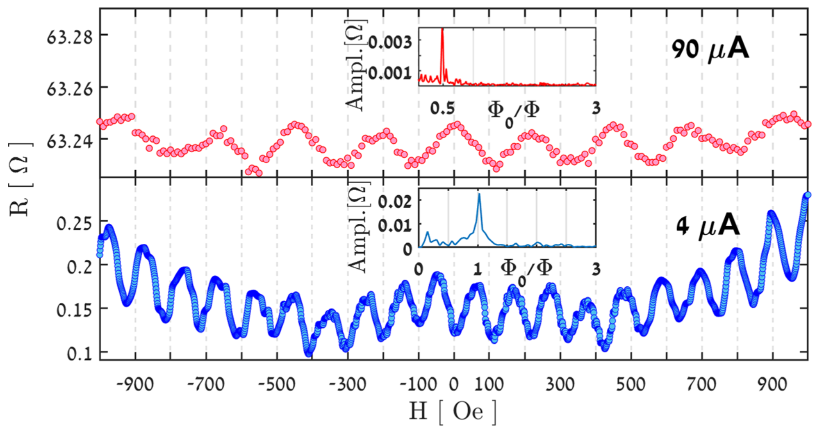

Figure 3. Magnetoresistance oscillations in the Nb ring at 3.5 K, for bias currents of 4 and 90 μA, lower and upper panels, respectively. Note the doubling of the flux periodicity from 105 to 210 Oe, corresponding to a crossover of the flux periodicity from $h/2e$ to h/e, respectively. Insets show the Fourier transform of the data.

set of dark arcs that reflect the temperature dependence of the critical currents of the involved Josephson junctions.

Low current magnetoresistance measurements (1−12 μA) at temperatures between 3.5 and 3.9 K, exhibit oscillations with a field periodicity of ~105 G, corresponding to flux periodicity of $h/2e$ for our ring of area ~2 × 10^5 nm^2. Typical magnetoresistance data, at 3.5 K and a current of 4 μA, are shown in Figure 3 (lower panel). The inset to this figure shows the Fourier transform of the data, demonstrating domination of the $h/2e$ periodicity.

A dramatic change in the flux periodicity to h/e is observed as the measuring current is increased to a range of 75−110 μA. Figure 3 (upper panel) demonstrates the h/e periodicity obtained at 3.5 K with current of 90 μA. The Fourier transform in the inset demonstrates domination of the h/e periodicity. Figure 4 shows the amplitude of the h/e oscillations as a function of the bias current. Interestingly, the amplitude is a nonmonotonic function of I_b, exhibiting a peak around I_b = 90 μA.

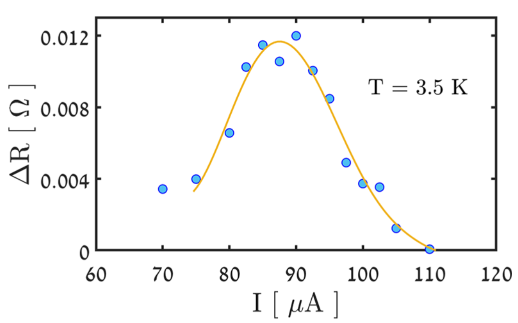

Figure 4. Current dependence of the h/e amplitude near zero field. The error in the amplitude is estimated to be of order 10%. The solid line is a guide to the eye.

The current-induced flux periodicity crossover is explained based on our view of the granular ring as a chain of Josephson coupled superconducting islands. As evident from the $R(T)$ data of Figure 2, the $h/2e$ magnetoresistance oscillations of Figure 3 (lower panel) were measured in the superconducting state where all the Josephson junctions comprising the ring are in a zero voltage state. It is, therefore, natural to associate these oscillations with the Little−Parks effect in the Nb ring, indicating coherence of electron pairs along the whole ring.[4,7]

The existence of Josephson junctions in the ring is expected to modify the waveform of the oscillations to sinusoidal instead of parabolic.[8] Indeed, the observed waveform is closer to sinusoidal rather than to parabolic, as apparent from the Fourier analysis shown in the inset to the lower panel of Figure 3.

The h/e oscillations of Figure 3 (upper panel) appear in the temperature and current range (see Figure 2) where each Josephson junction in the ring is in the voltage state; i.e., the bias current is larger than the critical current of each of the junctions. In this state, a resistive, conduction channel of quasi-particles is established along the ring. Thus, it is plausible to associate the h/e oscillations with the Aharonov−Bohm effect arising from interference of phase coherent quasi-particles flowing along the two branches of the ring consisting of Josephson junctions. Phenomena related to the Aharonov−Bohm effect in condensed matter were observed in the past in various metallic[2] and nonmetallic systems (see, e.g., refs 9−16). The present work demonstrates the effect for the first time in a ring consisting of Josephson junctions.

In view of a lack of a theory for Aharonov−Bohm oscillations in such a unique granular ring, it is instructive to compare the amplitude of the h/e oscillations obtained in this ring (see upper panel of Figure 3) to the amplitude expected for a normal Nb ring of the same size. This will provide an upper bound for the Aharonov−Bohm amplitude expected in our Nb granular ring. For a metal in zero voltage, the phase coherence length can be calculated from[17] $L_\phi(T) = \sqrt{D\tau_\phi}$, where D is the diffusion coefficient and $\tau_\phi = \frac{\tau_{e-e}\tau_{e-ph}}{\tau_{e-e} + \tau_{e-ph}}$ is the time between inelastic collisions. Here τ_{e-e} and τ_{e-ph} are the times between electron−electron and electron−phonon collisions, respectively. For Nb at 3.5 K, $D = 3.5 \times 10^{-3}$ m^2/s, the inelastic electron−electron collision rate estimated from ref 18 is $\frac{1}{\tau_{e-e}} = 2 \times 10^8$ s^{-1}, and the electron−phonon collisions rate estimated from ref 19 is $\frac{1}{\tau_{e-ph}} = 1.7 \times 10^8$ s^{-1}. The average time between inelastic collisions is thus τ_ϕ = 2.6 ns. These yield L_ϕ = 3 μm, which is larger than the size of our Nb ring. Inserting this value in the relation[20] $\Delta G_{h/e} = \frac{e^2}{h}\frac{L_T}{\pi r}\sqrt{\frac{L_\phi}{\pi r}}\,e^{-\pi r/L_\phi}$, using a thermal length $L_T = \sqrt{\frac{D\hbar}{k_B T}}$ = 87 nm, we find $\Delta R \approx$

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$R^2\Delta G = 0.026\ \Omega$. Obviously, the value of L_ϕ calculated above, using the bulk values for diffusivity and scattering times, is much larger than that expected for our granular ring. Thus, one would expect a much lower value of ΔR for our ring. Surprisingly, the experimentally measured ΔR in our granular sample is comparable to the calculated one for a bulk Nb. This points to an enhanced Aharonov–Bohm effect in our granular ring, which consists of decoupled superconducting grains. The enhancement effect can also be deduced from the fact that the Aharonov–Bohm effect in our experiment is observed at relatively high voltages, whereas the calculations above were made for zero voltage. Clearly, as the voltage increases, L_ϕ, and consequently ΔR, should decrease because the rate of inelastic collisions increases with the electron energy.[21] We thus may conclude that our measured oscillations amplitude is, in fact, much larger than expected for a metallic Nb ring of the same size under the same voltage. This enhancement may be attributed to the reduced density of quasi-particles in the ring consisting of Josephson junctions as compared to the density of electrons in a metallic ring. A lower value of the quasi-particle density reduces the rate of inelastic collisions and thus increases L_ϕ. Note that enhancement of the Aharonov–Bohm effect, however of different origin, was reported in metallic rings with superconducting "mirrors".[22,23]

The amplitude, ΔR, of the h/e oscillations shown in Figure 4, exhibits a nonmonotonic behavior as a function of the bias current. This can be explained as resulting from two competing processes. As the current increases, the number of quasi-particles increases, leading to a larger amplitude. However, a larger bias current is associated with a larger voltage and a larger energy of the quasi-particles, leading to a larger rate of electron–electron collisions and consequently to a reduced length of phase coherence.

As mentioned above, the crossover to the Aharonov–Bohm h/e flux periodicity was observed in three samples with the lowest rim size. This is expected in light of the work of Webb et al.[24] who showed that a clear Aharonov–Bohm h/e periodicity may be observed in rings having an area much larger than the area covered by the rims. When this condition is not fulfilled, an Aharonov–Bohm $h/2e$ periodicity appears that can hardly be distinguished from the Little–Parks $h/2e$ periodicity. Current dependence of the magnetoresistance oscillations obtained in the three samples can hardly be compared as they differ widely, probably due to different granular structure of these samples.

A further support for our interpretation of the h/e periodicity as the Aharonov–Bohm effect is found in measurement of the magnetoresistance as a function of the bias current at a constant magnetic field. As noted by Webb et al.,[25] if the voltage developed across the sample is changed, the interference properties are also affected. This leads to voltage-dependent fluctuations in the conductance that are similar to effects seen when the vector potential is changed. Such an effect is demonstrated in Figure S-2 in the Supporting Information. Oscillations of the differential resistance are clearly observed as a function of the bias current in the same region for which the Aharonov–Bohm oscillations are observed.

We close our discussion by noting that in the intermediate current range, between 15 and 45 μA, magnetoresistance oscillations are not observed. In a current range between 50 and 75 μA there is an indication for a flux periodicity close to $3\Phi_0$, a phenomenon that requires further investigation. Magnetoresistance data in these current ranges are shown in Figures S-3b,c in the Supporting Information. The Supporting Information also includes magnetoresistance data for the whole range in which the h/e periodicity is observed (Figure S-3d,e). We note that no change in the phase of the h/e oscillations is observed in this current range. The phase shift reported in ref 26 was obtained in normal metals in a range of relatively low currents (0–6.8 μA). The absence of an observable phase shift in our experiment could well be related to the unique structure of our sample and to the relatively small variations of the current in a much higher range.

We also note that the existence of Cooper pairs does not necessarily imply an $h/2e$ periodicity. For example, an h/e flux periodicity was predicted for s-wave nanorings with a size *smaller* than the coherence length, ξ_0.[27] Experimental efforts to detect the h/e periodicity in nanorings were partially successful.[28,29] Clearly, the physical origin of the h/e periodicity observed in our Nb ring is different, as the size of the ring is much larger than the coherence length ($\xi_0 \approx 40$ nm).

In conclusion, this work demonstrates two distinct quantum coherence effects in a single Nb ring. A Little–Parks effect, which manifests coherence of Cooper pairs, gives rise to a flux periodicity of $h/2e$ at low bias currents. The Aharonov–Bohm effect resulting from phase coherence of quasi-particles gives rise to a flux periodicity of h/e at high bias current. To the best of our knowledge, this is the first demonstration of the Aharonov–Bohm effect in a ring of Josephson junctions, resulting from phase coherence of quasi-particles tunneling between superconducting islands. The data indicate an enhancement of this effect as compared to the effect in metallic rings. This is attributed to the reduced density of quasi-particles in the superconducting islands which gives rise to a larger phase coherence length.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.nanolett.8b03617.

> $I-V$ curves for the Nb ring in the temperature range 2.5–7.5 K, differential resistance as a function of the bias current at zero magnetic field, and magnetoresistance oscillations in different current regimes (2–150 μA) (PDF)

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Notes

The authors declare no competing financial interest.

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