

Normal-metal–insulator–superconductor interferometer

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Hybrid normal-metal–insulator–superconductor microstructures were fabricated. The structures consist of a superconducting loop connected to a normal-metal electrode through a tunnel barrier. An optical interferometer with a beam splitter can be considered as a classical analog for this system. All measurements were performed at temperatures well below 1 K. The interference can be observed as periodic oscillations of the tunnel current (voltage) through the junction at fixed bias voltage (current) as a function of a perpendicular magnetic field. The magnitude of the oscillations depends on the bias point. It reaches a maximum at energy eV , which is close to the superconducting gap and decreases with an increase of temperature. Surprisingly, the period of the oscillations in units of magnetic flux $\Delta\Phi$ is equal neither to h/e nor to $h/2e$, but significantly exceeds these values for larger loop circumferences. Possible explanations of the phenomena are discussed.

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I. INTRODUCTION

The simplest optical interferometer consists of a beam splitter, a pair of mirrors, and an opaque screen. A metal loop with two electrodes can be considered as an analog to the described optical interferometer. One node corresponds to the electron-beam splitter and the second node to the screen. The total current through both paths (at a fixed voltage, for example) is then equivalent to the light intensity at the screen. The application of a perpendicular magnetic field alters the “interference pattern.” The conductivity of a normal-metal loop is periodic in units of magnetic-flux quantum $\phi_0^N = h/e$, where e is the electron charge.¹ To preserve the coherence, the size of the loop should be smaller than the phase breaking length ℓ_φ . Micron-sized metal structures at low temperatures ($T < 1$ K) are subject to this limit. The utilization of superconductors should eliminate this size limitation due to macroscopic quantum coherence. The only problem is that in the pure superconducting state, resistive measurements are useless. Other system parameters, such as critical temperature² or magnetization,³ should be measured.

II. EXPERIMENTAL

A number of normal-metal–insulator–superconductor (NIS) microstructures has been fabricated. They can be considered as a solid-state analog of an optical interferometer with a beam splitter. A closed loop of aluminum is overlapped at one point by a copper electrode through a thin oxide barrier (Fig. 1). At sufficiently low temperatures, aluminum becomes superconducting. Due to macroscopic phase coherence, there is no random alternation of the electron phase inside the loop of a superconducting interferometer, while the finite resistance of the whole NIS system enables electric measurements. Structures were fabricated by two-angle metal evaporation through an e -beam patterned double layer P(MMA-MAA)/PMMA mask. The typical aluminum thickness d_{Al} was ~ 35 nm, and the linewidth ~ 120 nm, for small samples. For larger loops ($> 10 \mu\text{m}$) 250-nm lines were used. Before deposition of the top copper electrode

($d_{Cu} \sim 30$ nm) the aluminum surface was oxidized in O_2 atmosphere at a pressure of about 1 mbar for 1–2 min. The nominal overlapping area between copper and aluminum was about $100 \times 100 \text{ nm}^2$. The tunnel resistance R_T at room temperature varied from sample to sample from roughly 1 to 60 k Ω , increasing by $\sim 20\%$, while cooling down to liquid-helium temperatures. The majority of experiments were made in a voltage-biased mode (Fig. 1). The derivative of the variation of the current with respect to the voltage, $dI/dV(V)$, characteristic was measured by a lock-in technique, using a $\sim 1 \mu\text{V}$ ac modulation of the biasing voltage. Current-biased dependencies were also studied. The resistance of the metal parts was roughly a few tens of ohms. Thus, the dominating part of the voltage drop was due to the tunnel barrier. Experiments were made in a ^3He - ^4He dilution refrigerator placed inside an electromagnetically shielded room. Only battery powered front-end amplifiers were kept inside the room. These were connected to the remaining electronics outside, through carefully shielded coaxial cables carrying analog signals. Three stages of filtering were used.

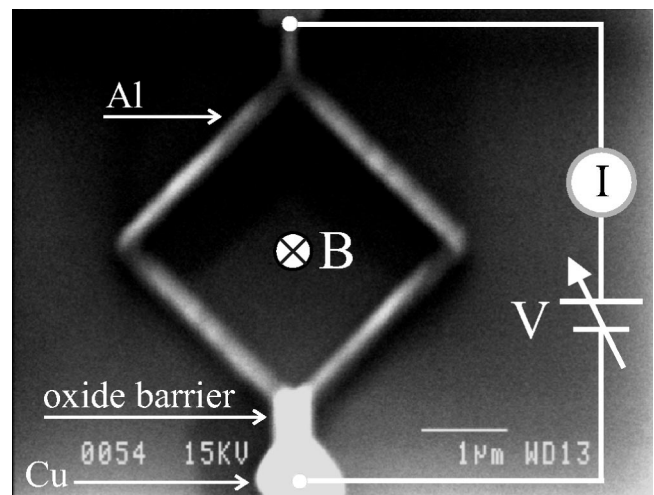


FIG. 1. Scanning electron microscope image of a structure with a $3 \times 3 \mu\text{m}^2$ loop. Schematics of voltage-biased measurements.

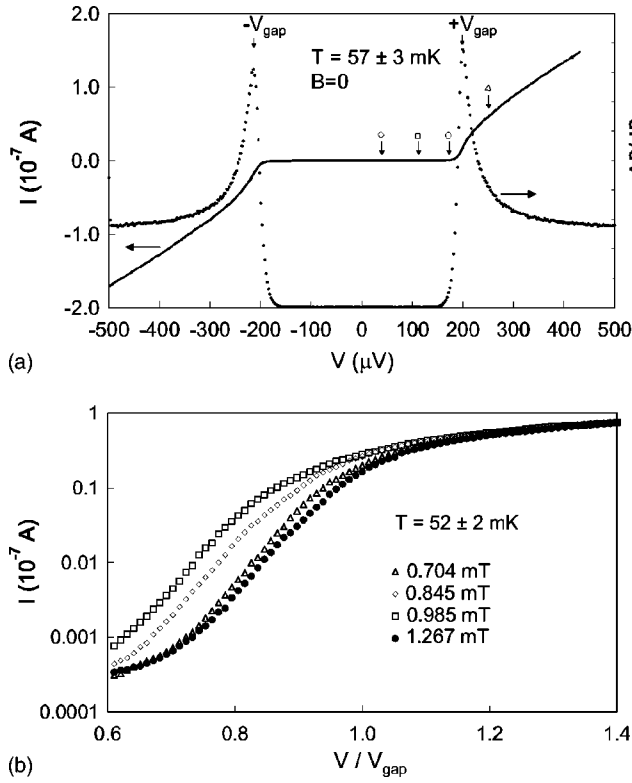


FIG. 2. Sample with a $5 \times 5 \mu\text{m}^2$ loop. (a) Typical $I(V)$ (left axis) and $dI/dV(V)$ (right axis) characteristics at zero magnetic field B . Symbols with arrows show the bias voltages, where the magnetic-field sweeps in Fig. 4 were taken. (b) Zooms of the same $I(V)$ characteristic at various perpendicular magnetic fields.

First, Spectrum Control 51-390-305 filters with a -80 dB cutoff at 300 kHz were placed on top of the cryostat at room temperature. A second stage was located at a ~ 1 K point consisting of capacitive and inductive C - L - C elements (220 nF, 2.2 mH, 220 nF) forming a π filter. The last stage was made of ~ 30 cm Thermocoax Philips cable,⁴ which was thermally anchored to the sample stage. The utilization of filters at room temperature resulted in a marginal improvement of the signal-to-noise ratio, while the performance of the π filter at 1 K appeared to be crucial, since no reasonable signal-to-noise ratio could be obtained without it. Magnetic fields of up to 40 mT were generated by various two- or four-layer superconducting coils wound directly on the outside of the refrigerator's vacuum canister. The coil inputs leading to the current source were also filtered. Magnetic-field sweeps (step by step) were made rather slowly (~ 2 – 5 s/point). In some cases, a car battery was connected through a decade resistor block and was used as a current source. The Earth's magnetic field was not screened. The latter may explain a small offset of the magnetic field (~ 0.05 mT), which appears in the data presented below.

III. RESULTS

At zero magnetic field the current-voltage characteristics, $I(V, B=0)$, show typical behavior for NIS junctions [Fig. 2(a)]. Hereafter, the superconducting gap energy Δ is defined

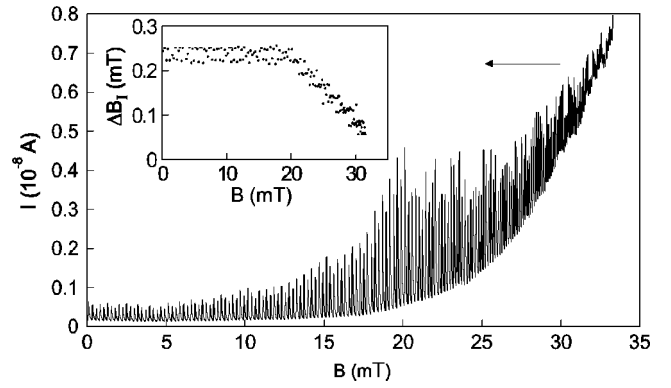


FIG. 3. Sample with a $25 \times 25 \mu\text{m}^2$ loop. $I(V/V_{\text{gap}}=0.31, B)$ dependence at $T=218 \pm 3$ mK. The arrow indicates the direction of the field sweep. Inset: corresponding magnetic-field dependence of the oscillation period ΔB_I .

as $\Delta = eV_{\text{gap}}$, where V_{gap} corresponds to the maximum of the $dI/dV(V)$ dependencies. This assumption is quite justified at temperatures well below the critical temperature T_c of the superconducting electrode (about 1.35 K for our structures).

The application of a perpendicular magnetic field modifies the $I(V, B=\text{const})$ dependencies in a nonmonotonous way [Fig. 2(b)]. The $I(V=\text{const}, B)$ characteristics are quasi-periodic with respect to the magnetic field (Fig. 3). The period of oscillations ΔB_I is not constant, but drops by a few factors at high fields (Fig. 3, inset). As the last phenomenon is not well understood, hereafter only the low-field data ($|B| \leq 2$ mT) is considered, where the magnitude and the period of oscillations are field independent. The $I(V=\text{const}, B)$ dependencies are essentially hysteretic (Fig. 4). Phenomenologically, one may state that the $I(V=\text{const}, B)$ characteristics form a set of “parabolas,” where allowed current states “jump” from neighboring branches of parabolas, depending on the direction of the magnetic-field sweep. Within the range of fields corresponding to a single “parabola,” dependencies are not hysteretic. The $I(V=\text{const}, B)$ characteristics are well reproducible and become noisy at biases V noticeably higher than the gap voltage V_{gap} .

Typical dependencies of the normalized magnitudes of current oscillations $\Delta I/I_{\text{max}}$ as a function of the normalized bias voltage V/V_{gap} for several samples with various loop size are plotted in Fig. 5. There are at least two common features. First, the function $\Delta I/I_{\text{max}}(V/V_{\text{gap}})$ has a maximum slightly below the gap voltage $V/V_{\text{gap}} \sim 0.7$ – 0.8 . Second, this maximum is pronounced at lower temperatures and becomes smeared at temperatures higher than ~ 500 mK. Surprisingly, the normalized magnitude of oscillations $\Delta I/I_{\text{max}}$ reaches nearly 100% at sufficiently low temperatures. As a comparison, the Aharonov-Bohm effect in micron-sized normal-metal rings¹ has the magnitude $\Delta R/R < 10^{-31}$, while in a micron-sized superconducting aluminum ring the magnitude of the critical temperature oscillations^{5,6} (Little-Parks effect) is $\Delta T_c/T_c < 10^{-25}$. The maximum magnitude of the current oscillations decreases slightly with an increase of the loop diameter, but the effect is still well pronounced even for a loop size as high as $25 \mu\text{m}$. For small size loops ($< 3 \mu\text{m}$) no correlation between the maximum magnitude of current

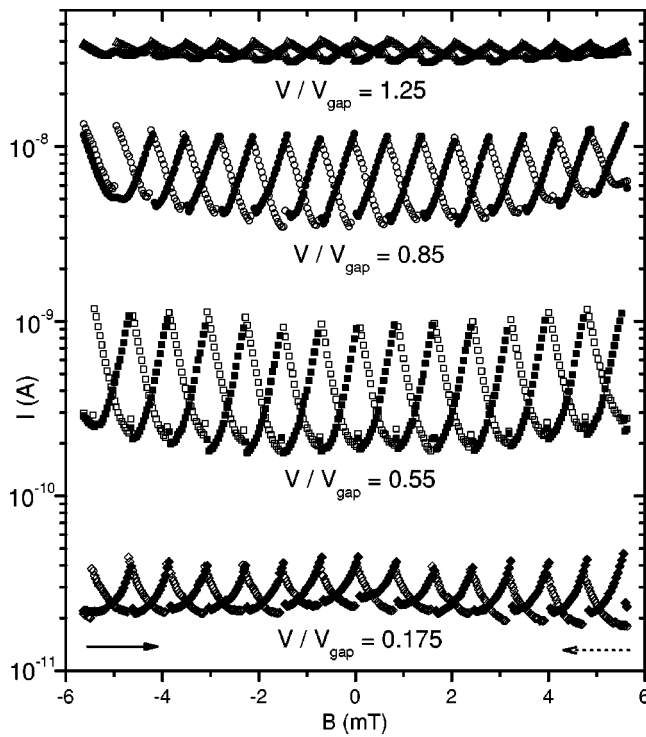


FIG. 4. Sample with a $5 \times 5 \mu\text{m}^2$ loop. $I(V=\text{const}, B)$ dependencies taken at various biases V/V_{gap} , $T = 165 \pm 5$ mK. Symbols correspond to notations in Fig. 2. The solid-line arrow shows direction of the field sweeps for the solid symbols; the dashed-line arrow, for the open symbols.

oscillations and the tunnel resistance in the range from 3 to 60 kΩ has been found. For loops with the side $L > 5 \mu\text{m}$, no effect has been detected for structures with tunnel resistance $R_T > 8$ kΩ. As only a finite number of structures (~ 20) has been studied, the last statement might not have a universal validity.

For a given size of loop, the period of current oscillations ΔB_I in low fields depends slightly on the bias voltage and increases by $\sim 15\%$ below the gap (Fig. 6). The effect is more pronounced at low temperatures (Fig. 6). Probably, the most surprising feature is the absolute value of the period of current oscillations $\Delta\Phi_I$ in units of superconducting magnetic-flux quantum $\phi_0^S = h/2e$. The period increases with an increase of the loop size and reaches a value $\Delta\Phi_I/(h/2e) \sim 60$ for the largest $25 \times 25 \mu\text{m}^2$ structure (Fig. 7). The uncertainty in the definition of the effective loop area, due to the finite linewidth, cannot account for such a high value of discrepancy. Although the majority of experiments were made in the voltage-biased mode $I(V=\text{const}, B)$, the current-biased dependencies $V(I=\text{const}, B)$ were also measured (Fig. 8). Qualitatively, the same oscillating behavior with hysteresis in the magnetic field was observed. However, there are several important differences. First, the shapes of the voltage and the current oscillations differ: the $V(I=\text{const}, B)$ “parabolas” are “up-side down” [Fig. 8(a)]. Second, the normalized magnitude of the voltage oscillations is much smaller than the corresponding current variation taken at the same point of the $I-V$ characteristic [Fig. 8(b)]. Third, the period of the voltage oscillations ΔB_V is smaller than the

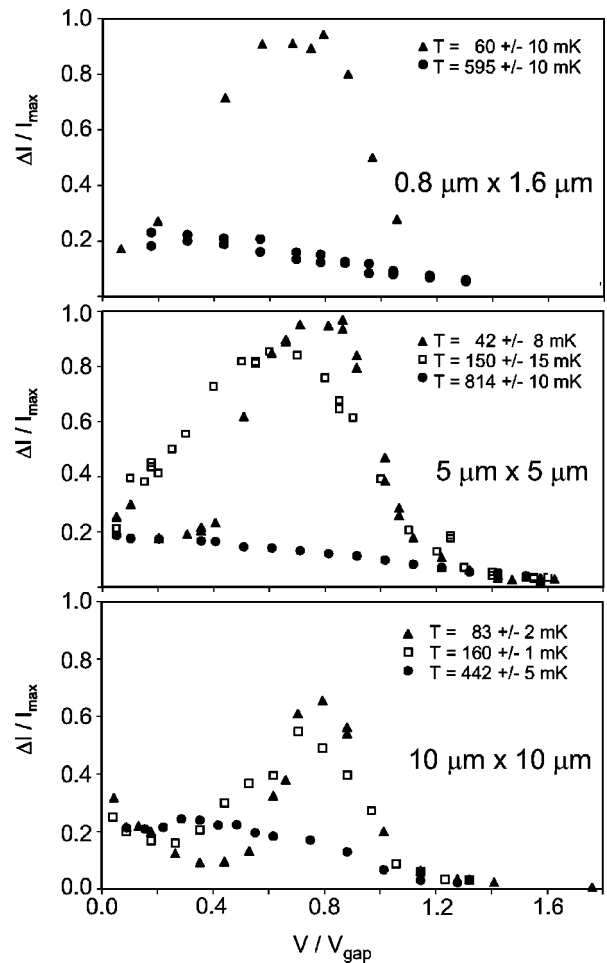


FIG. 5. Normalized magnitudes of the low-field ($|B| < 2$ mT) current oscillations $\Delta I/I_{\text{max}}$ as a function of the normalized bias V/V_{gap} for structures with different loop sizes at various temperatures.

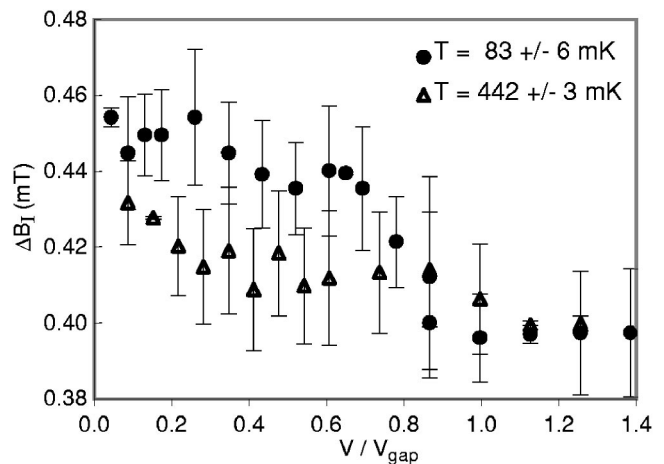


FIG. 6. Sample with a $10 \times 10 \mu\text{m}^2$ loop. Dependencies of the period of the current oscillations ΔB_I measured at low magnetic fields ($|B| < 2$ mT) on the normalized bias V/V_{gap} for two temperatures.

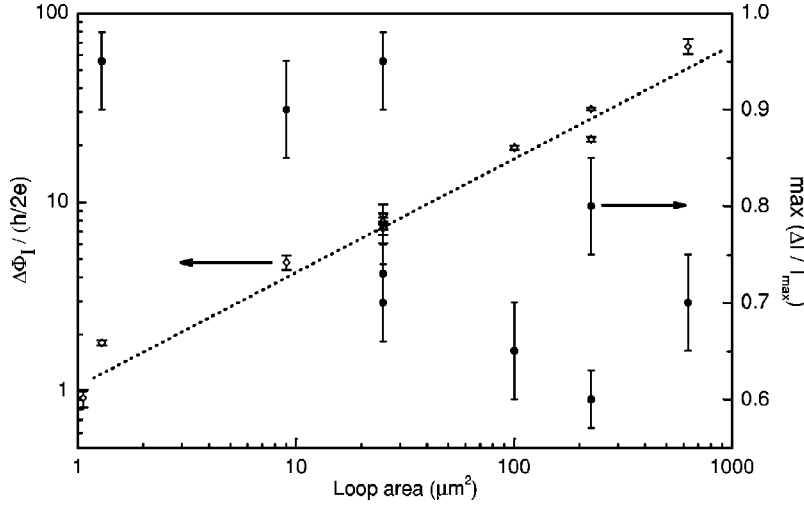


FIG. 7. The period of the current oscillations $\Delta\Phi_I$ in units of the magnetic-flux quantum $\phi_0^S = h/2e$ measured at low magnetic fields ($|B| < 2$ mT) (left axis, diamonds; the dotted line is a guide for the eye); and the maximum magnitude of the normalized current amplitude $\Delta I / I_{max}$ (right axis, circles) at temperatures $T < 100$ mK as functions of the loop area.

corresponding period of the current oscillations ΔB_I [Fig. 8(c)].

IV. DISCUSSION

The authors have no solid explanation for the mentioned phenomena. The most confusing feature is the deviation of the period of oscillations $\Delta\Phi$ from the expected value $h/2e$. The allowed states of a superconducting ring differ by the phase change accumulated over the circumference of the loop. The energy of the n th state is given by

$$\mathcal{E}_n = \frac{2\pi^2 \hbar^2 n_s \sigma}{m^* S} \left(\frac{\Phi}{\phi_0^S} + n \right)^2, \quad (1)$$

where n_s is the density of superconducting electrons, m^* is the effective electron mass, σ is the cross-section area of the wire, forming the loop, S is the loop's circumference, Φ is the magnetic flux through the area of the loop, and $\phi_0^S = h/2e$ is the superconducting-flux quantum. The persistent current is proportional to the derivative of the energy, $I \sim d\mathcal{E}_n/d\Phi$, and shows the characteristic sawtooth behavior with a period $\Delta\Phi_I = \phi_0^S$. Here it is assumed that the system changes its quantum state n , always relaxing to a ground state. The corresponding value of the persistent current at which the system switches to a new state is $j_{sw}^0 \sim 2\pi/S$. What is measured in the experiment is the transport current through the whole NIS structure (Fig. 1), and not the persistent current. However, any explanation based on these conventional "superconducting" properties of the loop section should result in a $h/2e$ periodicity independent of the size of the system (Fig. 7), the range of the magnetic fields (Fig. 2), and the measuring mode (voltage or current bias) (Fig. 8).

In our experiments three different coils were used, each being calibrated at room temperature and at 4.2 K. The data was found to be quantitatively consistent. Nevertheless, a control test was made. A pure aluminum $5 \times 5 \mu\text{m}^2$ loop, which contained no other materials and no tunnel junctions, was fabricated. Oscillations of the sample's conductivity, while in a resistive state (Little-Parks effect²), were measured using the same experimental setup. The period of oscillations was equal to the expected value $\Delta\Phi = \phi_0^S = h/2e$

within a reasonable accuracy $< 5\%$. The data gives the confidence that the observed periodicity in the NIS systems is not a product of the measurement hardware artifacts. Unusual periods of oscillations in NIS quasi-one-dimensional structures can by no means be explained by the formation of a superconducting sheath at high magnetic fields observed in relatively "bulk" systems.⁷

There are a few recent publications where the magnetization of small superconducting loops has been studied at temperatures well below the critical one.^{8,9} Experimental dependencies $M(B)$ do show periods $\Delta\Phi/(h/2e) > 1$. The proposed explanation⁹ considers the formation of metastable states with screening currents in superconducting loops exceeding the "conventional" value j_{sw}^0 . The ultimate limit of possible persistent currents in a ring is the critical current of the wire forming the loop $j_c \sim 2\pi/\xi \gg j_{sw}^0 \sim 2\pi/S$. The exact solution for the ratio of these currents for a ring of radius R is given in⁹

$$j_c/j_{sw}^0 = \frac{1}{\sqrt{3}} \frac{R}{\xi} \sqrt{1 + \frac{1}{2} \left(\frac{\xi}{R} \right)^2}, \quad (2)$$

The model⁹ uses the Ginzburg-Landau formalism. Strictly speaking, this approach is valid only for a gapless superconductor $\Delta(T) \rightarrow 0$. Applicability of Ref. 9 in the low-temperature limit requires further justification.

Even postulating the formation of the mentioned metastable states resulting in periodicity $\Delta\Phi/(h/2e) \sim j_c/j_{sw}^0 \gg 1$, it is not clear how the screening currents in the loop-shaped superconducting electrode affect the total tunnel current measured in the present work. The tunnel current of a NIS junction as function of the voltage bias V is given by¹⁰

$$I_{NIS} = \frac{1}{eR_T} \int_{-\infty}^{+\infty} \frac{N_S(E)}{N_N(E)} [f(E) - f(E + eV)] dE, \quad (3)$$

where $f(E)$ is Fermi distribution function, and the ratio of the density of states (DOS) in normal and superconducting states is given by the conventional BCS expression,

$$\frac{N_S(E)}{N_N(E)} = \frac{E}{(E^2 - \Delta^2)^{1/2}}. \quad (4)$$

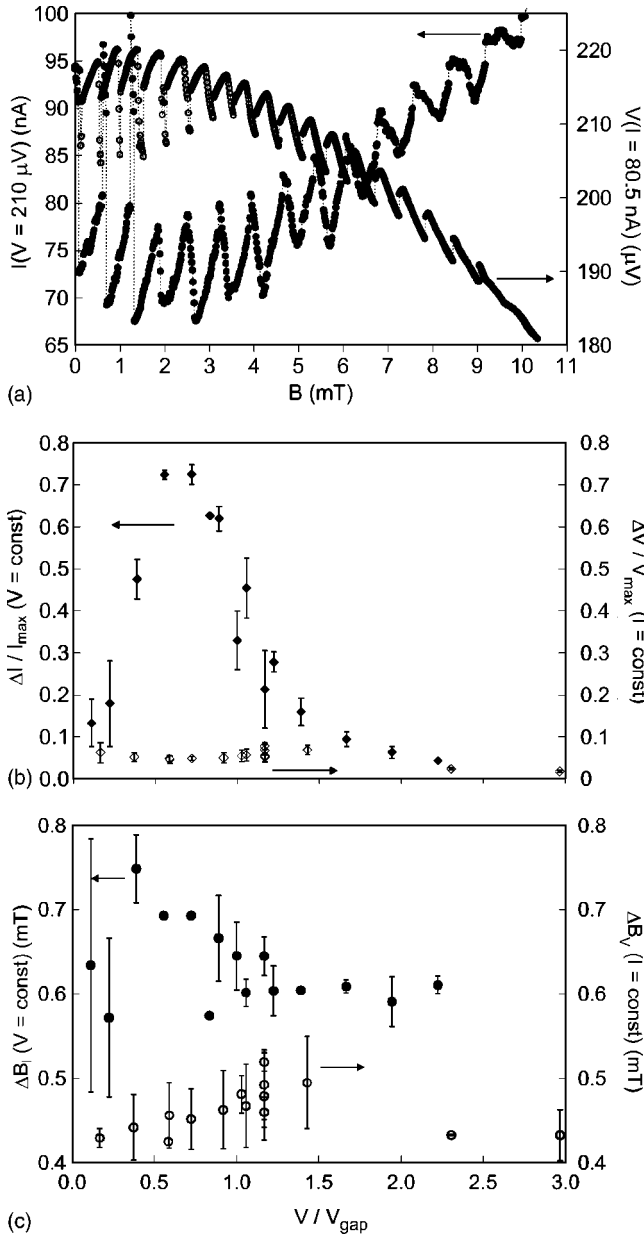


FIG. 8. Sample with a $5 \times 5 \mu\text{m}^2$ loop. (a) Current and voltage oscillations, corresponding to the same point of the I - V characteristic at zero magnetic field: $V=210 \mu\text{V}$, $I=80.5 \text{ nA}$, $V/V_{gap}=1.17$, $T=127 \pm 1 \text{ mK}$. The dotted lines are guides for the eye. (b) Normalized magnitudes of the current and voltage oscillations as functions of the normalized bias voltage V/V_{gap} . (c) Periods of the oscillations as functions of the same argument as in (b). For the voltage-biased data the solid symbols and the left axis are used; for the current-biased data, the open symbols and the right axis are used. For (b) and (c) only, data at low magnetic field ($|B| < 2 \text{ mT}$) was considered.

The very straightforward contribution of an external magnetic flux to the tunnel current oscillations $I_{NIS}(\Phi)$ is the corresponding periodic modulation of the energy gap $\Delta(\Phi)$, the Little-Parks effect,²

$$\frac{\delta\Delta(\Phi)}{\Delta(0)} = \frac{\delta T_c(\Phi)}{T_c(0)} \approx \left(\frac{\xi}{R}\right)^2 \left(n - \frac{\Phi}{\phi_0^S}\right)^2. \quad (5)$$

Note that Eq. (5) is also derived in the Ginzburg-Landau approximation,¹⁰ and, hence, its extrapolation to low-temperature limit requires further justification. Simple calculations based on Eqs. (3)–(5) give the magnitude of the current oscillations $\Delta I/I_{max}$ much smaller than the observed experimental dependencies (Fig. 5). It has been proposed¹¹ that the additional oscillating behavior of the tunnel current might originate from the deviation of the DOS from the BCS expression (4) at high values of the screening currents inside the superconducting loop-shaped electrode. A noticeable impact is only expected at high currents comparable to the critical ones:¹² $\max(j_{screen}) = j_{sw} \sim j_c$. In real samples the actual critical current is reduced in comparison with its theoretical value $j_c \sim 2\pi/\xi$, due to inevitable imperfections. Thus, if the metastable states with high screening currents $j_{screen} \gg j_{sw}^0$ are formed⁹ the DOS can be modulated with period $\Delta\Phi/\phi_0^S \sim j_{screen}/j_{sw}^0 \gg 1$. One might expect that the observed periodicity of oscillations should be a “finger print” of each structure, due to the particular arrangement of imperfections and the corresponding reduction of the critical current value. However, a comparison with our experiment (Fig. 7) tells us that if the critical current is reduced, it is not reduced randomly: structures with the same size of the loop show close values of periodicity in magnetic field scaled with the size of the loop [Eq. (2)].

Periodic modulation of the DOS by metastable screening currents can nicely explain unusually high periods of oscillations and gives order-of-magnitude agreement with the $\Delta I/I_{max}$ dependencies at $V \geq V_{gap}$ (Fig. 5). However, theoretical simulations at smaller bias voltages V contradict the experimental ones: calculated dependencies $\Delta I/I_{max}$ are almost constant at $V < V_{gap}$, except in the very vicinity of $V=0$. This discrepancy is probably related to the noticeable subgap current observed in our relatively “high transparent” (low R_T) tunnel structures: for a given bias V , experimental current I (Fig. 2) is systematically higher than the corresponding calculated value [Eq. (3)]. Very probably, the resulting model should incorporate this subgap current contribution.

A possible alternative explanation of the phenomena is a multiple vortex penetration within the superconducting “walls” of the structures while in a mixed state. However, the effective core size of a single vortex is about the dirty-limit coherence length $\xi \sim 150 \text{ nm}$, and is not much smaller than the linewidth of the studied structures. Thus, there is not enough room for vortices to fit within the “walls” of the superconducting loop (contrary to the “wide sample” from⁸ Fig. 5). The test structure, which consisted of a solid $5 \times 5 \mu\text{m}^2$ square overlapped through a tunnel barrier by a copper electrode, was studied. The variation of the current in a magnetic field $I(V=\text{const}, B)$ showed complicated nonmonotonous behavior with no signs of periodicity. This behavior agreed with the expectations that the penetration of a magnetic vortex inside a type-II superconductor requires the overcoming of a temperature-dependent potential barrier. The latter results in nonmonotonous, strongly hysteretic, random magnetic-field patterns. Thus, the origin of the oscillations due to the multiple vortex penetration should be ruled out.

It is probable that related oscillating behavior has been reported for a single-electron transistor composed of a super-

conducting central island in the form of a loop.¹³ The period of oscillations was not constant in a magnetic field and was different for voltage- and current-biased modes. Unfortunately, no solid explanation applicable to our geometry has been proposed.¹³

Experiments involving NIS tunnel junctions allow one to pump nonequilibrium quasiparticles from a normal electrode into a superconductor. Formation of a Cooper pair from injected quasiparticles is governed by at least three processes.¹⁴ The corresponding lifetimes are: quasiparticle scattering (τ_S), branch imbalance (τ_Q), and recombination (τ_R). If there are no other inelastic mechanisms involved, the quasiparticles should preserve their phase at shorter time scales: $t < \tau_\phi^0 = \min(\tau_S, \tau_R, \tau_Q)$. For aluminum charge imbalance, relaxation time can easily exceed ~ 10 ns.¹⁴ Additionally, all the mentioned lifetimes diverge at excitations $eV = \Delta$ and decrease rapidly at higher energies.¹⁵ Thus, one can associate the observed oscillations with the interference of nonequilibrium quasiparticles. However, there are several serious objections against such a proposal. First, the oscillations do not decay rapidly at excitations below the energy gap (Fig. 5), where the concentration of the injected quasiparticles is low. Second, the $5 \times 5 \mu\text{m}^2$ SIS structure (all aluminum, no normal-metal injector) showed similar oscillating behavior. The shape of the $I(V=\text{const}, B)$ dependencies appeared to be different from the Cu-AIO-Al system, but the period was close to the NIS case. Finally, the last problem is the absolute value of the oscillation period. Assuming that the periodicity $\Delta\Phi = h/q$ originates from the interference of quasiparticles, one should require that these nonequilibrium excitations have fractional charge $q < e$. Additionally, it is not clear why for the wide range of injection energies eV the charge becomes “more fractional” as the size of the loop increases. The origin of the difference of periods in current- and voltage-biased modes (Fig. 8) is by no means clear.

V. CONCLUSION

We have observed an unusual interference phenomenon in structures consisting of a superconducting loop (Al) connected to a normal metal electrode (Cu) through a tunnel barrier (Al oxide). All measurements were performed at temperatures well below 1 K. The interference can be observed as periodic oscillations of the tunnel current (voltage) through the junction at fixed bias voltage (current) as a function of a perpendicular magnetic field. The magnitude of the oscillations depends on the bias point. It reaches a maximum at energy eV , which is close to the superconducting gap, and decreases with an increase of temperature. Surprisingly, the period of the oscillations in units of magnetic flux $\Delta\Phi$ is equal neither to h/e nor to $h/2e$, but significantly exceeds these values for larger loop circumferences. The origin of the phenomena is not completely clear. Possible explanations might deal with the formation of metastable high screening currents inside the superconducting loop and the corresponding periodic modulation of the DOS. Probably, effects related to the injection of nonequilibrium quasiparticles should be additionally taken into consideration. Further study is required.

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