Little-Parks oscillations in a single ring in the vicinity of the superconductor-insulator transition

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We present results of measurements obtained from a mesoscopic ring of a highly disordered superconductor. Superimposed on a smooth magnetoresistance background we find periodic oscillations with a period that is independent of the strength of the magnetic field. The period of the oscillations is consistent with charge transport by Cooper pairs. The oscillations persist unabated for more than 90 periods, through the transition to the insulating phase, up to our highest field of 12 T.

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I. INTRODUCTION

Transport properties of amorphous superconducting films are strongly influenced by Cooper pairing, Coulomb repulsion, and disorder. The interplay of these effects leads to the very interesting physics of superconductor-insulator transition (SIT), which is now routinely observed in dirty metallic films [1–3]. This quantum phase transition can be driven by variation of disorder [4], thickness [5], magnetic field (B) [6], composition, and carrier concentration [7].

One of the central questions regarding the physics of the SIT is to which extent Cooper pairing is relevant in the insulating phase terminating superconductivity. From the theoretical side, there are two complementary approaches. The so-called Fermionic theory of suppression of superconductivity [8], being quite successful in describing the reduction of the transition temperature T_c via Coulomb interaction, including full suppression of superconductivity, does not take into account the effects of Cooper pairing in the normal, or insulating, state of the film. The alternative approach considers competition of Anderson localization and superconductivity [9–12] and, contrarily, admits activated transport by Cooper pairs in the insulating regime [13].

Experimentally, the importance of Cooper pairing in the insulating state can be probed by both tunneling spectroscopy and transport measurements. In the first approach, one directly measures the superconducting gap in the insulating phase, which indicates the presence of localized Cooper pairs [14–16]. The second approach (which we adopt in this paper) is based on specifically addressing effects, which are related to the crucial property of the Cooper pairs—their ability to maintain coherence at a macroscopic distance. This idea can be traced back to one of the first indications to the importance of the Cooper-pairing principle—the Little-Parks experiment [17]. Since then, a series of experiments was performed following the same logic [18–21].

Recently, following the experiment of the Valles, Jr. group [22], we applied this idea to amorphous indium oxide (a:InO) films [23]. We used a self-arranged array of holes to create a sample composed of a network of rings of a disordered superconductor. Our measurements demonstrated the existence of oscillations with a period consistent with an elementary charge of $2e$ (Cooper pairs) in the insulating regime. However, as in other experiments [18–21], we were able to detect only a few oscillations due to their decay with B. The reason for this decay was not clear and can, in principle, be twofold: (1) the intrinsic effect of the magnetic field, which quickly destroys spatial coherence of the Cooper pairs on the scale of the elementary cell of the array and (2) the effect of fluctuating size of the individual loops of the array, which smears out oscillations at larger fields. In addition, it was not possible to exclude the possibility of Josephson array physics [24].

The aim of the present paper is to extend our earlier study [23] to the case of a single ring in order to clarify both questions. We concentrated on the direct vicinity of the disorder-induced SIT transition in a:InO. We found that oscillations not only exist in a single ring both below and above SIT, but also persist up to the highest fields available (12 T).

II. FABRICATION

To define the structure, we used the ultra-high-resolution electron-beam lithography (EBL). In order to minimize the size of a:InO contacts directly adjoined to the structure, we had to implement the EBL process twice with an overlay precision of less than 20 nm between phases: In the first step we produced the inner Ti/Au contacts, followed by the fabrication of the a:InO ring (using a second EBL step). Each time a thermally oxidized silicon wafer (Si/SiO_2 with a typical value of the surface roughness less than 1 nm; oxide layer 300 nm, and resistivity less than 5 mΩ cm) was spin coated with a bilayer of poly(methyl methacrylate) electron-beam resist of two different molecular weights. The desired structure was exposed in the resist using an EBL-system JEOL JBX-9300FS. Photolithography was used to prepare four- or six-point Ti/Au electrical (outer) contacts. The a:InO film was e-gun evaporated in an ultra-high-vacuum system (2.5×10^{-7} Torr, Thermionics) from high purity (99.999%) In_2O_3 pellets in a residual O_2 pressure of $\sim 1.5 \times 10^{-5}$ Torr.

For structural determination we deposit one more test sample along with the experimental one. From the scanning electron microscopy (SEM) we conclude that the internal diameter is 50 nm and the external diameter (d_e) is 150 nm. The external diameter of the disk (unpatterned film, which we used

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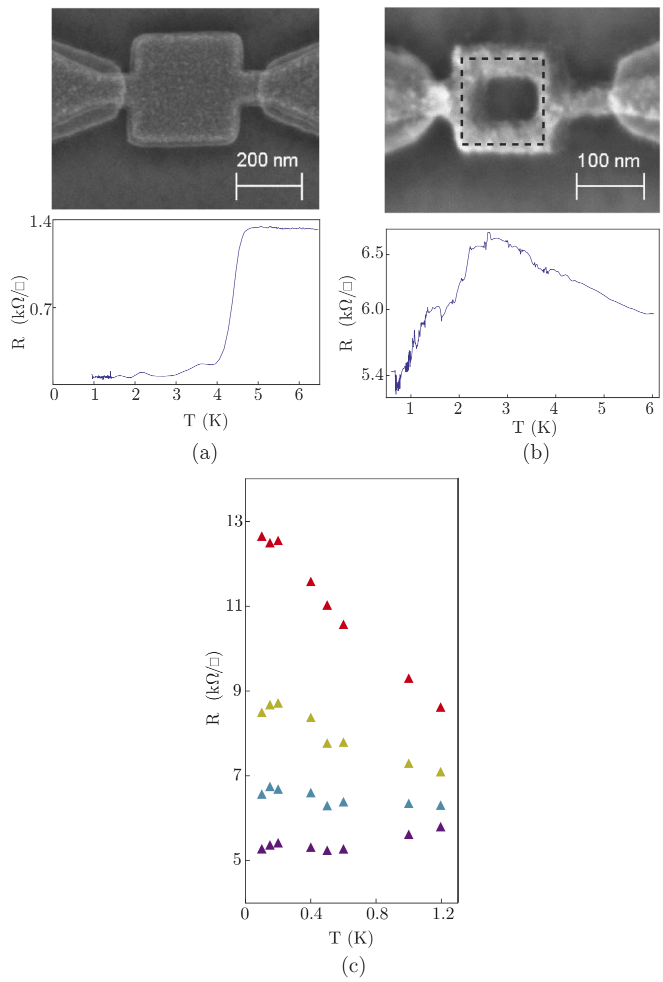

FIG. 1. (Color online) SEM image and $R_\square$ vs T at $B = 0$ T for (a) the disk and (b) the ring. The number of squares for such a ring (five squares) has been estimated from the geometry of the ring. The dashed line in (b) is the trajectory of a particle of a charge $2e$. (c) R vs T for the ring at $B = 0, 0.7, 1.5, 4.0$ T (from bottom to top) showing the shift to insulating behavior.

as a reference) was 320 nm. Accuracy of these measurements was ± 2 nm. The SEM images of the obtained structures are shown in Figs. 1(a) and 1(b) (one of four experimental samples exhibiting oscillations is shown). Atomic force microscopy images showed the thickness variation of about 10%, i.e., the thickness is 30 ± 3 nm. a:InO is known to form relatively uniform films [25], so we expect our structures to be uniform as well.

After gentle lift-off, the sample was mounted on the sample holder, electrically connected with a Au wire. Finally, the sample was immersed into a Kelvinox TLM (Oxford Instruments, Inc.).

We implemented two- and four-probe techniques. In the four-probe measurements resistance of the structure included resistance of small Ti/Au contacts overlapped by a:InO contacts. Such a pair of contacts was less than 1 μm^2 from each side of the structure and was caused by the design limitations. The signal from the sample was amplified by a low-noise homebuilt differential voltage preamplifier and measured using EG&G 7265 lock-in amplifiers at a frequency of 1.8 Hz. In order to minimize heating of the structure, we used a low excitation current of 1 nA.

III. EXPERIMENT

We first measured the dependence of the resistance (R) of the a:InO disk on temperature (T) at $B = 0$ T. The result is shown in Fig. 1(a). As T is lowered below 4 K the resistance drops abruptly from 1.4 kΩ to 50 Ω. In Fig. 1(b) we present R vs T at $B = 0$ T for the ring. Unlike the disk, it does not show an abrupt change in R, but a drop of 20% at ~3 K is most likely due to the (not fully developed) superconducting transition. We note that despite the sharp drop R, it saturates at a measurable value and remains finite down to $T = 50$ mK. In this regime, the sample demonstrates quadratic positive magnetoresistance at a low field turning into a negative magnetoresistance at $B > 2$ T.

Next, we measured R of the ring as a function of T. Contrary to the disk and films, it does not show any sudden change in the resistance down to the lowest temperature. However, it demonstrates nonmonotonous magnetoresistance, similar to that of the disk. We show R vs T traces at different values of the magnetic field in Fig. 1(c).

On a large scale of B, the disk and the ring demonstrate similar behavior, albeit, in comparison with the disk, the R vs T dependence at $B = 0$ T of the ring is much weaker. They exhibit the high-B phenomenology that we are accustomed to in our previous studies of a:InO films (see Ref. [26]), although, in this case, it is less developed. In Fig. 2, we plot R isotherms over our entire B range. The crossing point of the isotherms at $B_c = 0.8$ T identifies the "critical" B of the magnetic field tuned SIT, followed by the prominent magnetoresistance peak at $B = 8$ T. In this experiment we were not able to determine the crossing point in Fig. 2 better than specifying that it is in the range of 0.8,0.9 T. We believe that relative smallness (compared to measurements on macroscopic films) of the resistance variation with B and T is due to the mesoscopic nature of our sample. Another effect of the finite size, related to the loop geometry, is clearly seen in Fig. 2: Small, about ~1% by magnitude, oscillations of resistance as a function of magnetic field appear, which will be the focus of the remainder of this paper.

We start our analysis of these oscillations with the region of low B. On the plot of R vs B (Fig. 3), more than ten

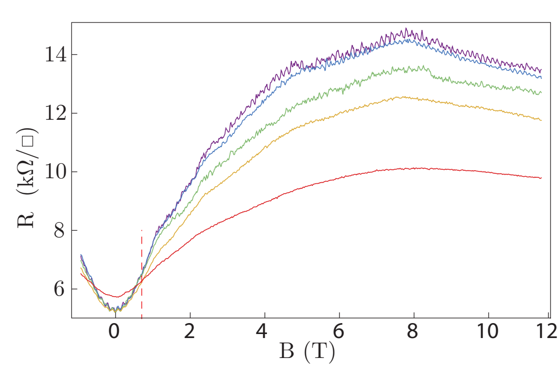

FIG. 2. (Color online) R vs B for temperatures $T = 0.1, 0.2, 0.4, 0.6, 1.2$ K (from top to bottom at $B = 8$ T), and the crossing point $B_c \sim 0.8$ T is shown by the red dashed line.

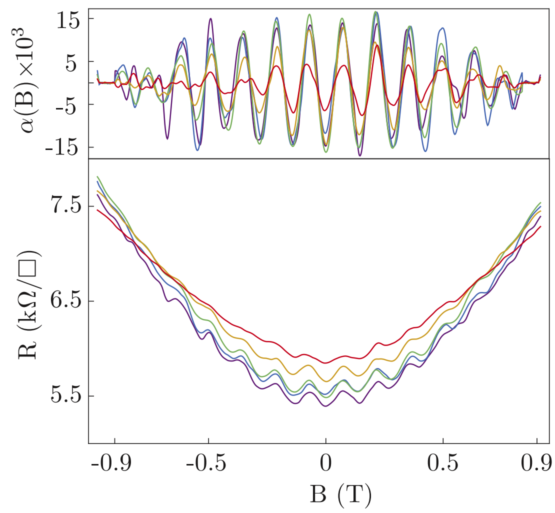

FIG. 3. (Color online) R vs B for the temperatures $T = 0.15, 0.2, 0.5, 0.8$ K (from bottom to top at $B = 0$ T); the inset: oscillating part $\alpha(B)$ (see the text).

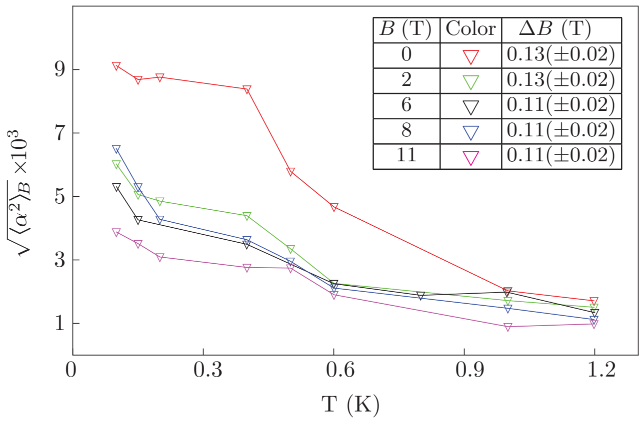

FIG. 5. (Color online) Amplitude of the oscillations vs temperature for different $B = 0, 8, 2, 6, 11$ T. The inset: the period of the oscillations as determined by counting the peaks in the interval of 1 T in the several ranges of B.

oscillations are seen, superimposed on a parabolically rising background. The oscillations period $\Delta B \approx 0.15 \pm 0.02$ T can be easily read from this figure. It is independent of T, indicating that it is determined by the geometry of the ring. The trajectory of a particle of a charge $2e$ encompassing the superconducting flux quantum $\Phi_0 = h/2e$ in a field of 0.15 T is shown in Fig. 1(b) and is consistent with the flux periodicity in integer units of Φ_0. For better characterization of oscillations, it is convenient to define the normalized oscillating part $\alpha(B) = [R(B) - R_s(B)]/R_s(B)$, where $R_s(B)$ is a smooth part of the $R(B)$ dependence (averaged over several oscillations).

Our central result is related to the behavior of $\alpha(B)$ at high B. It is presented in Fig. 4 where we plot $\alpha(B)$ of our ring for the entire range of B at $T = 150$ mK. Oscillations are clearly visible throughout the range, up to our highest B. This result is quantified in a table, shown in the inset to Fig. 5 where we show the period of the oscillations as determined by counting the peaks in the interval of 1 T on several ranges of B. Different rows correspond to different ranges of B: The first row, for example, is for the range of $-0.5, 0.5$ T. It is clear that the oscillations have similar periodicity at different values of B.

Finally, we characterize the T dependence of the amplitude of the oscillations. As a quantity characterizing the amplitude of the oscillations, we choose $\sqrt{\langle \alpha^2 \rangle_B}$ where averaging over the entire range of B is implied (the results are qualitatively the same if averaging over other ranges is performed). The T dependence of this quantity is shown in Fig. 5. It is consistent with our intuition: With increasing T, coherence length of the Cooper pairs decreases, and oscillations disappear at $T \sim 1.2$ K.

In order to further quantify this result, we performed Fourier transform (FFT) of the oscillating contribution. The result is demonstrated in Fig. 6. The main conclusion of this analysis is that the period of the oscillations is the same at different T's and B's. In Fig. 6(a) we show the spectra as a function of T. It is clear that the period of oscillations remains Φ_0 for different temperatures. At the same time, the amplitude of the dominant peak is strongly T dependent. In order to quantify how the oscillatory properties change with increasing B, we perform a series of FFTs at different subregions of the field [see Fig. 6(b)] where several curves correspond to the FFT of signals in different ranges of B: The lowest curve, for instance, shows the data in the range of $-0.5, 0.5$ T. This plot demonstrates that oscillations have similar periodicity in different ranges of the magnetic field (within an error of $\sim$0.02 T).

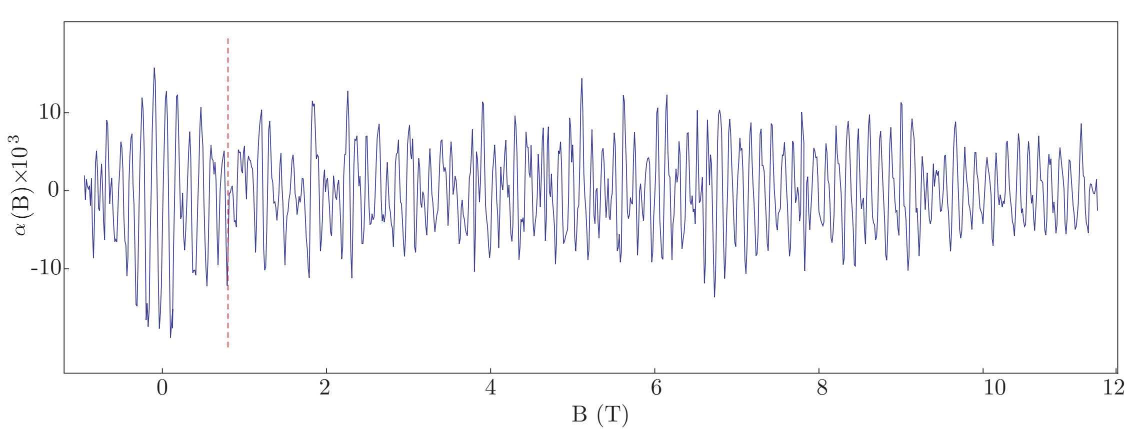

FIG. 4. (Color online) Oscillating part $\alpha(B)$ for $T = 0.15$ K, and the red dashed line shows $B \sim 0.8$ T.

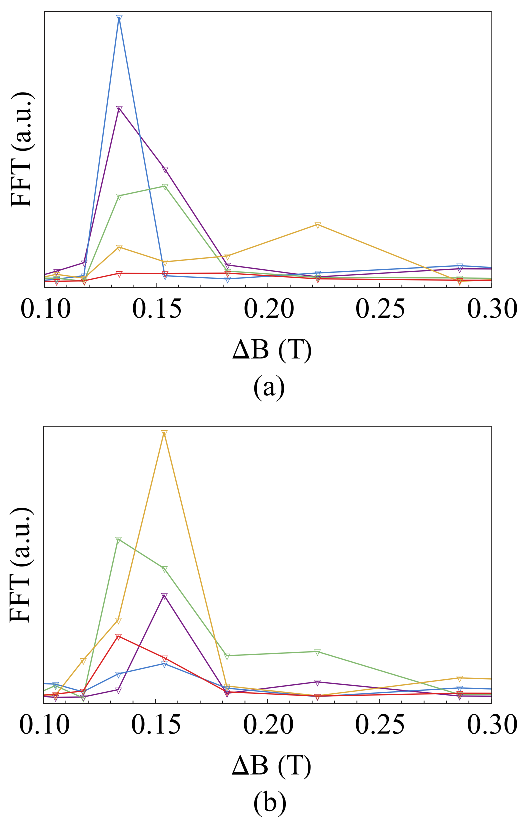

FIG. 6. (Color online) (a) FFT for temperatures of $T = 0.15,0.1,0.2,0.5$ K (from top to bottom at $\Delta B \sim 0.15$ T). (b) FFT for $B = 8,6,0,10,2$ T from top to bottom at $\Delta B \sim 0.13$ T and $T = 0.1$ K. The field window is 1 T, i.e., the purple line is for the range of $-0.5, +0.5$ T.

IV. DISCUSSION

Our main observations are magnetoresistance oscillations of a constant period throughout the available interval of T and B, consistent with a flux periodicity, corresponding to elementary charge $2e$. We argue below that this magnetoresistance is most likely due to electron-electron interaction in the Cooper channel, that is, undeveloped Cooper pairing.

We are aware of two physically distinct mechanisms that can lead to such oscillatory magnetoresistance: the electron-electron interaction in the Cooper channel and weak (anti)localization (WL). The first effect is expected to be most prominent close to the superconducting transition as is measured in the Little-Parks scheme [17]. In the vicinity of T_c the resistance of the ring is determined by thermal phase slips, which are influenced by magnetic flux penetrating the ring. Detailed theoretical analysis of this effect was performed in Ref. [20] where it was demonstrated that periodic flux dependence of the activation energy of the phase slips in such a ring allows explaining the magnitude of experimentally observed oscillations in the vicinity of the superconducting transition of the LaSrCuO rings [27]. For the metallic regime outside the transition region in the vicinity of T_c Kulik and Mal'chuzhenko [28] predicted, based on the Ginzburg-Landau approach, that this effect should also be noticeable. It appears due to the presence of fluctuating Cooper pairs, which can be rather long lived, $\tau_{GL} = \frac{\pi}{8T \ln T/T_c}$ and in this respect is due to paraconductivity [29]. Later, Larkin demonstrated [30] that further away from the transition the Maki-Thompson correction would be dominant in magnetoresistance and, hence, will give a dominant contribution in the Little-Parks type of oscillations.

Interestingly, in the experiment of Shablo *et al.* [31] where oscillations of the resistance in the normal state were first observed, they were attributed to paraconductivity, and only later it became clear that it is more realistic that they are actually related to weak localization, which was not well understood. It gives another possible contribution to the observed oscillations [32], see also the experimental study in Ref. [33]. This effect is not associated with electron-electron interaction, but its phenomenology is similar to that of the interaction-induced one. Interestingly, this effect was also predicted to exist in the hopping conductivity regime [34], but the low-field magnetoresistance has a negative sign—opposite to that observed in our measurements.

As was stressed already in the seminal work of Ref. [32], the amplitude of the oscillations in metals is usually determined by a factor $\gamma - \beta(T)$, where γ is coming from the WL part and depends on the symmetry class of the system ($\gamma = 1$ for weak spin-orbit impurity scattering, $\gamma = -1/2$ for the opposite case) and $\beta(T)$ is an effective constant of Cooper interaction [30]. These two effects, although having the same periodicity, have rather different spatial scales: single-particle coherence length L_ϕ and coherence length of the Cooper pair L_ξ. Since we do not have any reliable estimate for L_ξ in our sample, we will concentrate on ruling out the possibility of WL origin of the effect. First we consider the ability of single electrons to maintain coherence on the size of the sample. At the temperatures of our experiment, the main source of dephasing is expected to be electron-electron interaction. According to Ref. [35], coherence length $L_\phi = \sqrt{D\tau_\varphi}$ can be estimated from the dephasing rate $\frac{1}{\tau_\varphi} = \frac{T}{2\pi g}\ln(2\pi g)$. Estimating the diffusion coefficient as $D \approx 1$ cm^2/s and $g = \hbar\sigma/e^2 \approx 0.7$ we find for $T = 0.2$ K the values of $\tau_\varphi \approx 100$ ps and $L_\phi \approx 90$ nm, which are much smaller than the circumference of our ring. Additionally, the magnetic field not only imposes a phase on the interfering electrons, but also induces mass into the Cooperon. This effect becomes more pronounced with the growth of width w of the ring. As shown in Ref. [36], the effective dephasing length for the WL-induced oscillations $\bar{L}_\phi$ is determined by $1/\bar{L}_\phi^2 = 1/L_\phi^2 + \frac{1}{3}(\frac{weH}{\hbar c})^2$. This effect imposes additional restrictions on the number of oscillations, which can be seen in the experiment as follows: $N_{\rm osc} \sim d_e/w$, in our case $N_{\rm osc} \sim 3$ while we resolve over 90 oscillations.

V. CONCLUSIONS

These arguments allow excluding WL for the description of the observed effect. We emphasize that, although we defer an attempt for a theoretical explanation of our observation to a future paper, we nevertheless stress that in all likelihood it is rooted in the stability of Cooper pairing deep in the insulating regime. This is especially interesting because our ring does not show a strong superconducting trend. A more detailed study of these oscillations can shed more light on the role of the Cooper interaction in mesoscopic a:InO rings.

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