Little-Parks Oscillations in an Insulator

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We present the results of a magnetoresistance study of the disorder-induced superconductor-insulator transition in an amorphous indium-oxide thin film patterned by a nanoscale periodic array of holes. We observed Little-Parks-like oscillations over our entire range of disorder spanning the transition. The period of oscillations was unchanged and corresponded to the superconducting flux quantum in the superconducting as well as in the insulating phases. Our results provide direct evidence for electron pairing in the insulator bordering with superconductivity.

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A key ingredient in the Bardeen, Cooper, and Schrieffer theory of superconductivity is the pairing of electrons in the superconducting state [1]. One of the earliest [2] experimental supports for this prediction was obtained by Little and Parks (LP) [3], who measured the resistance (R) of a thin superconducting cylinder threaded by an external magnetic field (B). They observed magnetoresistance oscillations periodic in the superconducting flux quanta, $\Phi_0 = h/2e$, where h is Planck's constant and e is the charge of the electron, corresponding to a Cooper-pair charge of $2e$. LP-like oscillations were later observed in other, multiply connected, systems such as arrays of superconducting wires [4], arrays of Josephson junctions [5], and patterned thin superconducting films [6].

The transport properties of a superconducting material are strongly dependent on the interplay between superconductivity, electron-electron interaction, and disorder. In highly disordered, thin-film, superconductors this interplay can lead to a direct transition between the superconducting and insulating phases, for the review see Ref. [7]. This superconductor-insulator transition (SIT) can be induced by a variety of means such as disorder, film thickness, external magnetic field, external electric field, or chemical substitution [8–10].

Most (but not all, see Refs. [11,12]) of the theoretical attempts to account for the physics of the SIT [13–18] suggest that, surprisingly, Cooper-pairing plays a central role in determining the transport properties of the insulator bordering with superconductivity. Some of the authors [19] even claim that the insulating phase owes its very existence to Copper-pairing. Although this notion is supported by circumstantial evidence, it still defies a direct verification.

In order to test for the role of electron pairing in the insulating phase, Stewart and his collaborators [20–23] conducted transport measurements of periodically patterned, amorphous, Bi thin films. The effect of the patterning of their Bi films was twofold. First, they observed LP-like oscillations in the insulating (and the superconducting) regime, with a period that was consistent with Φ_0 threading a unit cell of their pattern. But as importantly, the patterning of their films resulted in a profound change to the phenomenology of their insulating phase, in the form of the appearance of the high-B insulating peak [7,24] that was absent in all their unpatterned samples [22].

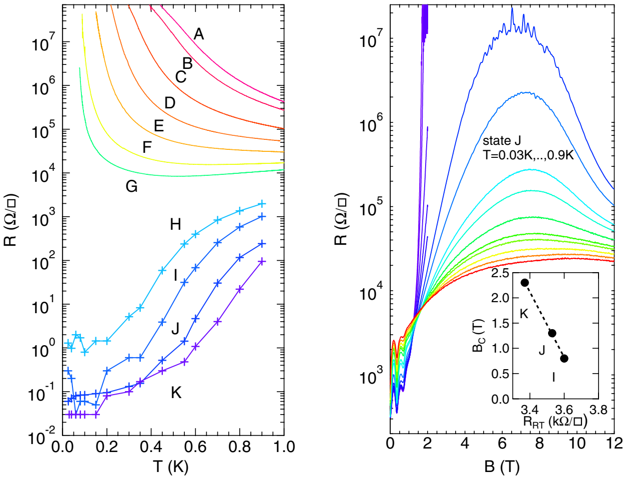
In the Stewart *et al.* experiment, the SIT was driven by varying the film thickness. This resulted in variations in the geometry of the physical samples, as an unavoidable consequence of the topography of the substrate [23]. As a result of this, they claim, the insulator bordering superconductivity consisted of a network of superconducting islands connected by weak links of nonsuperconducting material [25]. They suggested that this artificially introduced inhomogeneity resulted in a transition that was dominated by phase, rather than amplitude [21], fluctuations in the order parameter, in turn leading to the formation of a bosonic, or Cooper-pair insulating state, rather than the fermionic insulator they observed in the unpatterned films. This change in the microscopic physics of the transition, they argued, was responsible for the change in the phenomenology in the insulating regime and the appearance of the insulating peak.

In this Letter we present the results of transport measurements of a nanopatterned, amorphous indium-oxide (a:InO) thin film. By means of a simple annealing procedure (see below) we were able to affect a SIT driven by the amount of disorder in the film [26]. We observed magnetoresistance oscillations with a period corresponding to Φ_0, which remained unchanged as we went from the insulator to the superconducting phases. In contrast with the work on Bi thin films where the magnetoresistance peak was only observed in patterned films, our procedure enabled the SIT in a single physical sample without any change in geometry and without inducing a change in the phenomenology associated with the bosonic insulating state that exists in the absence of any patterning [see

Fig. 1(b)] [24,27]. Unpatterned and patterned a: InO films follow similar high-B magnetoresistance behavior. The signature of patterning appears in the form of oscillations appearing before the large magnetoresistance peak.

Our film was prepared on an anodized aluminum-oxide (AAO) substrate. The growth condition of the oxide layer was chosen to give as periodic hexagonal hole pattern [28]. Analyzing scanning electron microscope images we found a hole diameter of 42 ± 5 nm, and a distance between two adjacent holes of 80 ± 5 nm, resulting in a unit-cell area of the hexagonal lattice of 5600 ± 600 nm^2.

The a: InO film was e-beam evaporated on the AAO substrate from stoichiometric 99.999% $\mathrm{In_2O_3}$ pellets [26] (Cerac Inc.) in residual $\mathrm{O_2}$ pressure of 10^{-5} Torr. Since the evaporated film thickness was much less than the AAO hole depth, the hole pattern transferred to the a: InO film. The film was prepared in a Hall-bar geometry by using a shadow mask 3 mm long and 1.5 mm wide. Immediately after evaporation the sample was mounted on the sample holder and electrically connected by Au wire using silver paint. The sample was immersed into a Kelvinox TLM (Oxford Instruments Inc.) dilution refrigerator for the transport measurements. For the insulating samples, a two-probe technique was implemented with electrical currents in the range $0.1 < I(pA) < 70$ amplified by Femto DDPCA-300 preamplifier. Superconducting samples were measured using a four-probe configuration with currents in the range of $1 < I(nA) < 20$. In this case, the signal from the sample was amplified by a homemade, low-noise, differential preamplifier and measured using an EG&G 7265 lock-in amplifier at a low frequency of 3.5 Hz. After completion of the low-T measurements, the sample was annealed at 43 ± 3 °C while maintaining a vacuum of $\sim 3 \times 10^{-2}$ Torr, and then measured again at low-T.



(a) (b)

FIG. 1 (color online). (a) R vs T at $B = 0$ T in annealing states A through K, spanning the range of disorder in nanopatterned a: InO thin-film sample. (b) R vs B in superconducting state J at $T = 0.03$, 0.04, 0.06, 0.08, 0.1, 0.15, 0.2, 0.3, 0.35, 0.45, 0.55, 0.6, 0.7, 0.8, 0.9 K . Inset: Critical B vs R at room T for some of the superconducting states. The dashed line is a linear fit.

In Fig. 1(a) we show R vs T taken at $B = 0$ T for several annealing states spanning our range of disorder. Our a: InO film was initially in an insulating state, attested to by its diverging R as $T \to 0$. The annealing (we label each annealing state in alphabetical order) resulted in weaker insulators and, subsequently, superconductivity commenced with state H. We found that the $R(T)$ traces in the insulating phase followed an Arrhenius law $R(T) \propto \exp(T_0/T)$ where T_0 is the activation T [24]. With each annealing state, T_0 was found to decrease. Unfortunately, in this experiment, we have taken a large annealing step on the approach to superconductivity so we were not able to determine the critical normal-state R (at $T = 0.9$ K) of the disorder-driven SIT to better than specifying that it is in the range of $2 < R(\mathrm{k}\Omega/\square) < 8$. In the superconducting regime, the critical T increased from 0.4 K in state H to 0.9 K in state K. A much more detailed account of this annealing-driven SIT is forthcoming. In superconducting samples, the resistance was strongly dependent on the magnetic field, especially around $B = 0$ T. Meaning that applied magnetic fields of a few mT caused resistance to increase by many orders of magnitude. Therefore, the $R(T)$ dependence for the superconducting samples was reconstructed from $R(B)$ data measured at fixed T's. The value of $R(B = 0)$ was identified as the minimum value observed in a range $-0.1 < B(T) < 0.1$.

We begin the presentation of our B-dependent data by plotting, in Fig. 1(b), the R isotherms obtained from one of our superconducting states, state J, over our entire B range. Before addressing the signature of the patterning in our data evident at $B < 1.5$ T, we note that our patterned sample exhibits the familiar [24] high-B phenomenology that we are accustomed to in our previously studied a: InO films. Typical electron concentration in such films is on the order of 10^{20} cm^{-3} with an upper critical field of about $H_{C2} = 14$ T. This includes the crossing point of the isotherms at $B = 1.5$ T identified with the critical B, B_C, of the B-tuned SIT, followed by the prominent magnetoresistance peak at $B = 7$ T. In the inset of Fig. 1(b), we plot B_C in the superconducting phase at various annealing states versus R at room temperature, R_{RT}, which we take as a rough measure of the level of disorder in our sample. A clear trend was detected in which B_C is decreasing with R_{RT} and the extrapolated vanishing of B_C is expected at $R_{\mathrm{RT}} = 3.7$ kΩ. The clear evidence for the applied nanopattern, was the magnetoresistance oscillations, observed in the superconducting regime below $B = 1.5$ T.

In Fig. 2, we plot R vs B for our most superconducting state, state K, measured at $T = 0.08$ K, in the range of $-1 < B(T) < 1$. Nearly four oscillations are seen, superimposed on a rising background. The oscillations period, 0.35 ± 0.005 T, corresponds to Φ_0 threading an

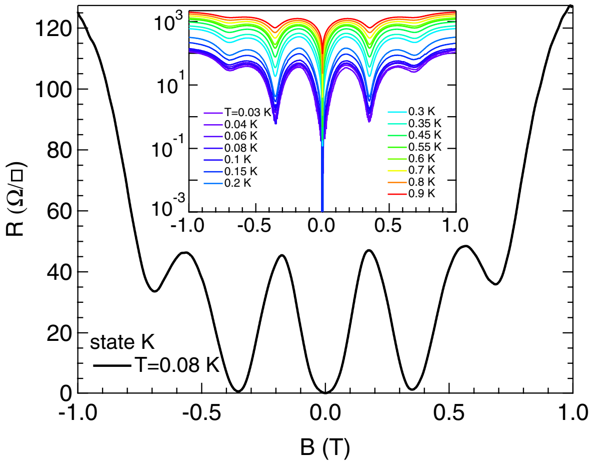
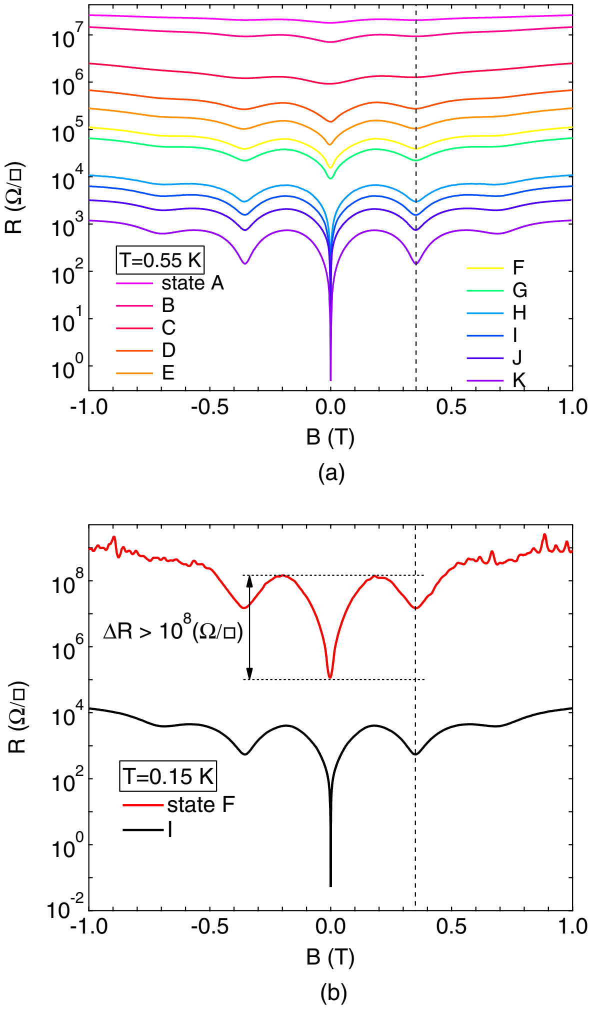


FIG. 2 (color online). R vs B at $T = 0.08$ K for superconducting state K. The period of LP oscillations was 0.35 ± 0.005 T. Inset: R isotherms taken at various T's in state K.

area of 5500 nm^2, that is, within error, the unit-cell area of our AAO substrate. In the inset of Fig. 2 we present $R(B)$ isotherms, obtained from the same state, in the range of $0.03 < T(K) < 0.9$ on a logarithmic scale. The oscillations period is independent of T, indicating that it is determined by the geometry of the pattern rather than by some other physical length scale, which is expected to be T dependent. Following Refs. [4,6] we interpret these results as the LP oscillations.

Our central finding presented in Fig. 3(a), where we plot $R(B)$ traces of our sample spanning our entire range of disorder. Clear oscillations are observed throughout the range and, most importantly, the period of oscillations remains fixed, corresponding to Φ_0 through a unit cell of the array, deep into the insulating phase. While in this figure we show data at $T = 0.55$ K because, at lower Ts, the insulating states have prohibitory high Rs, lower-T ($= 0.15$ K) data are plotted, for a superconducting state I and an insulating state F in Fig. 3(a) [the corresponding $B = 0$ R vs T can be seen in Fig. 1(a)]. Strikingly, the amplitude of the oscillations in the insulating state can be very large, see Fig. 3(b). At $T = 0.15$ K, for state F it was 10^8 $\Omega/\Box$ and grows even larger at lower Ts. The observation of these oscillations demonstrate phase coherence between Cooper pairs in the insulating regime on a scale larger than the interhole distance. So far, we discussed the B dependence of the resistance near zero V bias. However, a similar oscillatory trend was observed under application of a finite V, where the electronic transport in the insulating states is characterized by high nonlinearity [29]. This is demonstrated in Fig. 4, in which $I - V$ characteristics of insulating state A at $B = 0$ T are presented. Similarly to the nonpatterned films case, at low T these I-V curves exhibit an abrupt jump in I of several orders of magnitude at a threshold dependent on the sweep direction. On increasing V, the system switches from a high-resistance to a



(a)

(b)

FIG. 3 (color online). (a) R vs B from different annealing states at $T = 0.55$ K spanning our entire range of disorder. The states A through G were insulating, while states H through K were superconducting. (b) R, plotted on a logarithmic scale, vs B for insulating state F and superconducting state I. The dashed line corresponds to Φ_0 penetrating an area of one unit cell.

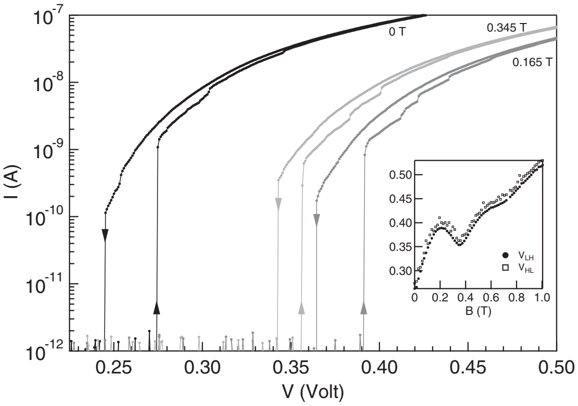


FIG. 4. Nonlinear $I - V$ in insulating state A at $T = 0.025$ K at three different B's. Arrows indicate direction of voltage sweep. Inset: threshold voltages V_{LH} and V_{HL} vs B.

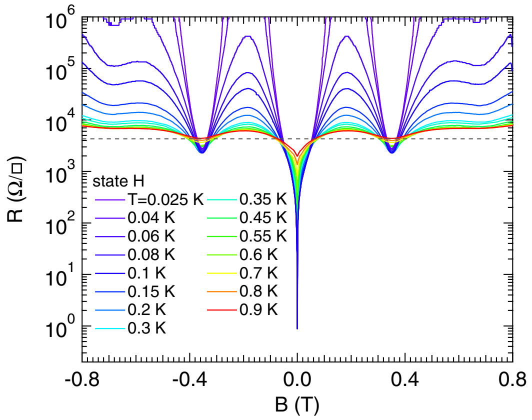


FIG. 5 (color online). Superconducting state H exhibits multiple SIT's driven by B. Transition points indicated by the dashed line at $R = 4.2\ \mathrm{k}\Omega/\square$.

low-resistance state at a point referred to as V_{HL}. A hysteresis is observed when the sweep direction is reversed, until a second switching voltage, V_{LH}, is reached. Tracing these thresholds as a function of B, we observe (inset in Fig. 4) oscillations with period Φ_0 superimposed on a monotonically increasing background. These oscillations appear to reflect the periodic oscillations observed in the Ohmic measurements. In some of the superconducting states multiple crossing points [21] of the isotherms were observed, see Fig. 5. Rs at the crossing points were found to be close to $R = 4.2\ \mathrm{k}\Omega/\square$, similar to other superconducting states [9,24]. For state H at $B > 0$ T there were three such points at $B^* = 0.06$ T, $B^* = 0.31$ T, and $B^* = 0.39$ T. As B increased from zero, the superconducting sample became insulating at $B^* = 0.06$ T. At half filling $B = 0.18$ T the R was larger then $10^7\ \Omega/\square$. The sample returned to the superconducting state at $B^* = 0.39$ T. However, its minimum R at $B = 0.35$ T was larger than at $B = 0$ T. Above $B = 0.39$ T, the insulating state reappeared. With the increased annealing time, the sample became more superconducting and the critical B^* shifted to the higher values.

In summary, we observed LP-like oscillations in the insulating and superconducting phases of a single physical sample of nanopatterned a:InO thin-film. The T-independent oscillations had a constant period throughout the disorder-tuned SIT. For our sample, the patterning induced no significant change in the high-B magnetoresistance. The flux periodicity corresponded to Φ_0 through the unit cell of the patterned array, demonstrating the participation of Cooper pairs in the transport in the insulating phase.

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