Parity effect and single-electron injection for Josephson-junction chains deep in the insulating state

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We have made a systematic investigation of charge transport in 1D chains of Josephson junctions where the characteristic Josephson energy is much less than the single-island Cooper-pair charging energy, $E_J \ll E_{CP}$. Such chains are deep in the insulating state, where superconducting phase coherence across the chain is absent, and a voltage threshold for conduction is observed at the lowest temperatures. We find that Cooper-pair tunneling in such chains is completely suppressed. Instead, charge transport is dominated by tunneling of single electrons, which is very sensitive to the presence of BCS quasiparticles on the superconducting islands of the chain. Consequently we observe a strong parity effect, where the threshold voltage vanishes sharply at a characteristic parity temperature $T^*$, which is significantly lower than the the critical temperature, $T_c$. A measurable and thermally-activated zero-bias conductance appears above $T^*$, with an activation energy equal to the superconducting gap, confirming the role of thermally-excited quasiparticles. Conduction below $T^*$ and above the voltage threshold occurs via injection of single electrons/holes into the Cooper-pair insulator, forming a non-equilibrium steady state with a significantly enhanced effective temperature. Our results explicitly show that single-electron transport dominates deep in the insulating state of Josephson-junction arrays. This conduction process has mostly been ignored in previous studies of both superconducting junction arrays and granular superconducting films below the superconductor-insulator quantum phase transition.

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I. INTRODUCTION

For several decades now, chains and arrays of low-capacitance Josephson junctions have attracted much attention as systems in which quantum phase transitions can be studied\textsuperscript{1,2}, and as many-body platforms that could enable novel quantum phases\textsuperscript{3,4} and topologically protected states\textsuperscript{5,6}. However, there have been many more theoretical proposals along these lines than experimental works, and experiments have mostly been confined to several intriguing avenues of research concerning the superconductor-insulator transition\textsuperscript{7}, the dynamics of quantum-phase slips\textsuperscript{8–10}, metrological current standards\textsuperscript{11,12}, conjectured solitonic phenomena\textsuperscript{13,14}, and the use of Josephson-junction chains as superinductors\textsuperscript{15}.

The insulating state of superconducting junction arrays is located below a superconductor-insulator (SI) quantum phase transition, and is synonymous with the destruction of superconducting phase coherence across the array, and localization of Cooper pairs. The Cooper-pair insulator occurs when the characteristic Cooper-pair charging energy significantly exceeds the Josephson coupling energy, $E_{CP} \gg E_J$. The understanding and engineering of charge transport deep in the insulating state presents difficult problems due to the competition between various modes of transport which include both Cooper-pair and single-electron tunneling processes\textsuperscript{16,17}, both of which occur in the presence of significant disorder.

Nearly all studies of charge transport in insulating arrays, however, start from a point of view where it is assumed that the low energy excitations that play a role in conduction are Cooper pairs. This is based on the assumption that the temperature is sufficiently low to ignore the presence of unpaired electrons. A recent calculation of the dc conductivity for arrays deep in the insulating state, based upon single Cooper-pair excitations and including weak disorder, was put forth by Syzranov et al\textsuperscript{18}. Their model proposes Cooper pairs as the sole charge carriers, and describes transport in terms of variable range hopping between adjacent islands as a result of Josephson tunneling. In another theoretical study by Fistul et al\textsuperscript{19}, a model for Cooper-pair transport in the insulating state of 1 and 2 D arrays of Josephson junctions is applied to the interpretation of experimental data from granular superconducting films. Similar lines of thinking have been taken in the analysis of Josephson-junction array experiments, also based on the assumption that charge transport far below the superconducting transition temperature, $T_c$, is predominantly carried by Cooper pairs\textsuperscript{20,21}. It is important to realize that in most experiments, the contribution of quasiparticles cannot be ignored. This contribution has proven important in studies of qubits\textsuperscript{22} and Cooper-pair transistors\textsuperscript{23}, but has only recently been studied theoretically in JJ chains\textsuperscript{24}.

We have made a systematic investigation of charge transport for temperatures ranging from 10 mK to 1 K in 1D Josephson arrays that are characterized by large charging energies and high junction resistances. In this regime the arrays are deep in the insulating state. We find that Cooper-pair transport is completely suppressed and charge transport proceeds via single-electron in-
jected into the Cooper-pair insulator. Furthermore, we observe a strong parity effect, with a well-defined cross-over temperature $T^*$, at which the voltage injection threshold decreases sharply. The parity effect has been studied previously in superconducting single-electron transistors, and in Cooper-pair boxes, where it appears as a temperature crossover from 2e to 1e periodicity in normalized gate charge. In contrast, our array measurements show the parity effect has a global effect on charge transport through the whole array. The presence of ~1 thermally-excited BCS quasiparticle per island in the array significantly enhances the tunneling rates of single electrons through the chain, and simultaneously destroys the insulating state of the array, as the voltage threshold for single-electron injection is suppressed to zero.

II. DEVICES AND MEASUREMENTS

We have fabricated 1D chains of Al-AlO$_x$-Al Josephson junctions having a length of $N = 50$ junctions using electron beam lithography, followed by thermal shadow-evaporation and in-situ oxidation of Al films, which have a thickness of 30 nm. We have focused on three arrays with slightly different properties, having in common large junction resistances, $R_J \gg R_Q \equiv (2e)^2/h$ and large Cooper-pair charging energies, $E_{CP} \equiv (2e)^2/2C_J \gg E_1$, where $C_J$ is the junction capacitance, as described in Table 1. The arrays exhibit a Coulomb blockade at low temperatures, both in the superconducting state as well as in the normal state, which is obtained by suppressing the superconducting gap in the films using an external parallel magnetic field.

The samples were bounded to a circuit board, mounted in a microwave tight Cu sample enclosure, and secured to the mixing chamber of a BlueFors LD400 cryogen-free dilution refrigerator with a base temperature of 10 mK. Each DC line was filtered from high frequency radiation using three meters of ThermoCoax, thermally anchored at each stage of the dilution refrigerator, and having a measured low-pass cutoff frequency of ~1 MHz. The lines were additionally filtered using chip LC components on the circuit board, and at room temperature using low-pass LC filters. Several measurements of superconducting single-electron transistors were made using this setup that clearly showed 2e-periodic stability diagrams, indicating negligible quasi-particle poisoning due to the presence of non-equilibrium quasiparticles in the measurement leads.

We have characterized the devices using current-voltage measurements with voltage biases ranging from 5 μV to over 50 mV, and with a DC current resolution as low as ~0.8 fentomampere for currents up to several nanoamperes. In addition, we have used a parallel magnetic field $B_{||}$ to continuously suppress the superconducting gap $\Delta(B_{||})$. From our measurements we have found that the gap depends on the parallel magnetic field through $\Delta(B_{||}) \sim \Delta(0)(1 - B_{||}^2/B_0^2)$, as expected, with $\Delta(0) = 210 \pm 10 \mu eV$, and $B_0 = 0.59 \pm 0.02$ T.

It is important to distinguish between large-scale current-voltage characteristics (or IVCs) and small-scale IVCs. In the presence of a superconducting gap, ‘large-scale’ refers to voltage biases that span the onset of direct quasiparticle tunneling due to pair breaking across every junction in the chain. This occurs for $eV \sim 2N\Delta$. ‘Small-scale’ bias in the superconducting state refers to voltages $V$ that are a substantially small fraction of $2N\Delta/e$, in which case conduction is also referred to as sub-gap transport. As the superconducting gap is suppressed below approximately twice the characteristic single-electron charging energy, $E_C = (2e)^2/2C_J = E_{CP}/4$, the transition from small-scale to large scale occurs at $V \approx E_CN/2e$, above which the Coulomb blockade is lifted across each junction. Our main results are primarily concerned with sub-gap transport; however, the large-scale IVCs measurements are used to experimentally characterize the energy scales and intrinsic disorder of the junction chains.

IVC and differential conductance ($dI/dV$) data are shown for the three devices of Table 1 in Figure 2. The asymptotic normal state conductance at large bias voltage $V$ determines the chain average of the normal state tunnel resistance $R_J$ per junction. From this, one can extract the average Josephson coupling energy using the Abegaokar-Baratoff relation $E_J = \frac{1}{2}\Delta(R_Q/R_J)$. The Cooper-pair charging energy $E_{CP} \equiv (2e)^2/2C_J$ for each device is found experimentally by extrapolat-

| Device | Junction Area ($\mu m^2$) | Island Volume ($\mu m^3$) | $R_J$ | $E_{CP}$ | $E_1$ |
|--------|--------------------------|--------------------------|-------|----------|-------|
| A      | 0.015                    | 0.0029                   | 248 kΩ| 1500 μeV | 2.7 μeV|
| B      | 0.015                    | 0.0029                   | 312 kΩ| 1700 μeV | 2.2 μeV|
| C      | 0.003                    | 0.0016                   | 786 kΩ| 6800 μeV | 0.85 μeV|
FIG. 2. Large-scale current (upper plot) and conductance (lower plot) measurements for devices A, B and C in the superconducting state for magnetic field $B_z = 0$ (solid lines), and for device B in the normal state obtained with $B_z = 0.7$ T (dotted lines). The inset in the upper plot shows the small-scale IVC at $T = 25$ mK for device B at femtoampere current resolution. Conductance $(dI/dV)$ measurements in the lower plot are normalized to $(NR_j)^{-1}$ for each device.

The absence of charge disorder would be aligned when the voltage bias across individual junctions equals $2\Delta/e$. The broadening and concomitant reduction of this BCS peak therefore gives a measure of the relative disorder of the chains due to offset charges and fabrication inhomogeneity.

As clearly seen in Figure 2, the BCS peak for device C is significantly more broadened as compared to devices A and B. In addition, there appears to be a random structure in the $IVC$ data for this device around the onset for direct quasiparticle tunneling. This is expected as device C has a significantly larger $E_{CP}$ compared to device A and B, which makes it much more sensitive to random offset charges. In addition, device C was fabricated in a separate processing run, using different lithographic development parameters, and therefore may have more intrinsic disorder due to reduced film and junction quality.

Small-scale $IVC$ data for device B is shown in the inset of Figure 2. Note that the current scale in the inset of Figure 2 is 5 orders of magnitude lower than in the main plot. There is a clear voltage threshold $V_t \simeq 1.3$ mV for the onset of femtoampere currents. Threshold voltages can be distinguished quite clearly in the differential conductance at low temperatures, as shown in Figure 3. The region $|V| < |V_t|$ shows a current blockade and a zero-bias conductance $G_0 = (dI/dV)_V=0$ that is identically zero, or at most, lower than our measurement resolution, $G_0^{\text{min}} = 10^{-12}$ $\Omega^{-1}$. We experimentally determine the voltage threshold $V_t$ as the absolute value of the voltage bias at which the conductance rises above $10^{-11}$ $\Omega^{-1}$ (see lower plot in Figure 3), which is a factor of 10 greater than the measurement resolution. As seen in Figure 4, there is a characteristic temperature $T^*$ at which $V_t$ drops sharply to zero, and above which a measurable zero-bias conductance is observed.

A subset of the temperature dependent $IVC$ data for device B is shown in the lower plot of Figure 3 for $B_z = 0$ and temperatures 25, 300, 350, 400 and 450 mK. $T^*$ for device B in zero field is found to be 270 mK (see Figure 4). The data at 300 mK and above show a conductance peak around $V = 0$ that grows with temperature, but starts out with a small mini-gap that appears to be a remnant of the blockade region below $T^*$. The conductance peak arises from the overlap of the BCS DOS from island to island across the chain, and only becomes evident when there are thermally excited quasiparticles occupying these states. (For a large Josephson junction such thermally excited quasiparticles give rise to a logarithmic singularity at $V = 0$ in the $IVC$ at finite $T$).

III. PARITY EFFECT

The dependence of the measured threshold voltage $V_t$ on temperature for device A is shown in the upper plot of Figure 4 for several values of the parallel magnetic field, $B_z = 0, 0.25, 0.35$ and $0.45$ T. It is evident that $V_t$
FIG. 3. Differential conductance $dI/dV$ for device B in zero magnetic field. Upper: grayscale image of $dI/dV$ ($\Omega^{-1}$) on a logarithmic scale, showing a temperature dependent threshold that vanishes at a well-defined temperature, identified as the parity temperature $T^*$. Lower: slices of $dI/dV$ for temperatures 25, 300, 350, 400, and 450 mK. For temperatures below $T^*$, the threshold voltage is clearly distinguished as a nearly two orders of magnitude increase in the conductance, as shown by the 25mK data. The voltage threshold $V_t$ is experimentally determined to be the absolute value of the voltage bias at which the conductance rises above $10^{-11} \Omega^{-1}$, which is a factor of 10 greater than the measurement resolution $G_{\text{min}} = 10^{-12} \Omega^{-1}$.

vanishes sharply at a specific temperature that depends on $B_{||}$. We argue that this behavior is a consequence of the parity effect for small superconducting islands. The ground state free energy for an odd number of electrons is higher than that for even numbers by the amount $F = \Delta - k_B T \ln N_{\text{eff}}$, where $N_{\text{eff}}(T) \approx \mathcal{V} \rho(0) \sqrt{2\pi k_B^2 \Delta(T)}$ is the effective number of states at finite $T$ arising from integration over the BCS quasiparticle DOS, $\mathcal{V}$ is the volume of the island and $\rho(0)$ is the density of states for the normal metal at the Fermi energy $\frac{\Delta}{2}$. The free-energy difference $F$ for a single island vanishes at a cross-over temperature $k_B T^* = \Delta / \ln N_{\text{eff}}(T^*)$. Using the island volumes given in Table I, the experimentally determined $\Delta_0$, and taking $\rho(0) = 1.45 \times 10^{47} \text{ m}^{-3} \text{ J}^{-1}$, for the density of states for aluminum, one can compute the theoretically expected parity temperature for isolated islands. For islands such as those in devices A and B, we calculate $T^* = 260 \text{ mK}$, and for the islands of device C, $T^* = 277 \text{ mK}$. One notices that for our device parameters, $k_B T^* \approx \Delta / 9$, which is much less than $T_C \approx 1.3 \text{ K}$ for aluminum.

We experimentally determine the array parity temperature $T^*$ as the temperature at which $V_t$ passes through the voltage threshold observed in the normal state at the lowest temperature, which is indicated in Figure 4 (upper plot) by the horizontal dotted line. For $B_{||} = 0$ we find

FIG. 4. Upper plot: temperature dependence of the voltage threshold $V_t$ for device A for $B_{||} = 0, 0.25, 0.35$ and 0.45 T. Lower plot: magnetic field dependence of the parity temperature $T^*$ for devices A (triangles), B (circles) and C (squares).
the voltage threshold for single-electron injection. In addition, the tunneling rates for single electrons through the chain are significantly enhanced due to the presence of quasiparticles. The precise microscopic mechanism underlying this phenomena is currently being investigated. Some simulation results on parity effects in arrays can be found in Cole et al. Recent experimental results on a hybrid normal-superconducting transistor illustrate enhanced charge tunneling due to a non-equilibrium quasiparticle distribution.

IV. THERMALLY-ACTIVATED
CONDUCTANCE

The zero-bias conductance $G_0 \equiv (dI/dV)_{V=0}$ was measured under applied parallel magnetic fields for temperatures ranging from the parity temperature up to 1K. $G_0$ in all devices is found to follow an Arrhenius law for thermal activation, $G_0(T) = G_\infty \exp(-E_A/k_B T)$, where $E_A$ is the activation energy, as shown in the upper plot of Figure 5. The zero-bias conductance in the normal state at 0.6 T (not shown) continues to decrease above $T^{-1} = 10$, indicating an electronic temperature lower than 100 mK.

As a function of applied $B$-field, the activation energy for devices A and B is linear with $B^2$, as shown in Figure 5, and appears to be equal to $\Delta(B_{||}) = \Delta_0 (1 - B^2_{||}/B_{c2}^2)$, with $\Delta_0 = 214 \pm 3 \mu eV$, and $B_{c2} = 0.59 \pm 0.02$ T. We find that the experimentally determined value of $E_A$ in zero applied field agrees with an independent estimate of $\Delta_0$ gained from the large scale $IVC$ data, $\Delta_{Vq}/2N = 210 \pm 10 \mu eV$, where $V_{q}$ is the voltage that marks the onset of direct quasiparticle tunneling that occurring for $eV_q/N \approx 2\Delta$. An activation energy that equals the superconducting gap can be easily understood because an energy of $2\Delta$ is required to break a Cooper-pair. Since two independent excitations are created, the exponent for thermal activation is $\Delta$ rather than $2\Delta$.

For device C, $E_A$ varies randomly and somewhat irreproducibly with $B_{||}$, taking values between 250 and 350 $\mu eV$. We attribute this to significantly larger disorder present in device C, as inferred from the large scale IV data, and which is also consistent with the much larger $E_{CP}$ for device C. In contrast to this, however, the activation exponent for the conductance evaluated at $V = 1.5 mV$ for device C shows nearly identical behavior to that of $G_0$ for devices A and B. This voltage bias is just outside the observed Coulomb blockade region of device C in the normal state, which is also relevant for unpaired charge carriers when the islands of the chain are superconducting. We conclude that transport above the parity temperature is set by thermally-activated quasiparticles in chains where strong charge disorder does not dominate.

Thermally-activated transport in 1D SQUID-arrays was reported recently by Zimmer et al. The use of
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tunneling, and a flux-independent term that remains
dependent part, as would be expected for Cooper-pair
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mention as an alternative explanation, thermally gener-
pairs undergoing variable-range hopping, although they
measurements, Zimmer
a perpendicular magnetic field. To account for their
plots: activation energy

0
0.1
0.2
$B^2$ (T$^2$)

$E_A$ (µeV)

FIG. 5. Upper plot: log $G_0$ vs. $1/T$ for device B for $B_{||}$=0, 0.3 and 0.4 T. The solid lines are fits to an Arrhenius law. Lower plot: activation energy $E_A$ vs. $B_{||}^2$ for devices A (triangles) and B (circles). The dot-dashed line is a combined fit for both devices to $\Delta(B_{||}) = \Delta_0(1 - B_{||}^2/B_{0,||}^2)$ yielding zero-field gap $\Delta_0 = 214 \pm 3 \mu$eV and $B_{0,||} = 0.59 \pm 0.02$ T.

a SQUID geometry permitted tuning $E_J$ in situ using a perpendicular magnetic field. To account for their measurements, Zimmer et al. assume a zero-bias conductance that is the sum of two contributions: a flux-dependent part, as would be expected for Cooper-pair tunneling, and a flux-independent term that remains when $E_J$ (Cooper-pair tunneling) becomes very small. With $E_J$ suppressed to nearly zero, Zimmer et al. observe an activation exponent of the order of the superconducting gap. These authors interpret their measured $E_A$ as a characteristic charging energy for localized Cooper pairs undergoing variable-range hopping, although they mention as an alternative explanation, thermally gener-
ated quasiparticles.

As far as we are aware, Zimmer et al. is the only reported measurement of thermally-activated zero-bias conduction in 1D arrays in the superconducting state. Thermally-activated zero-bias conductance in 2D junction arrays has been reported by two groups some time ago. These authors interpreted their results in terms of a so-called “core energy”, $E_{\text{core}}$, which is the energy required to create an electron-hole pair on adjoining sites (e.g. by moving a single electron by one site), together with the induced polarization charge on neighboring islands. This model is known as the soliton model. In the superconducting state, one finds for a 2D system, $E_{\text{core}} = 2\Delta + E_C/2$, where the first term comes from breaking a Cooper pair, and the second is the electrostatic energy for placing a single electron and hole on adjoining sites. Since two independent excitations are created, $E_A = E_{\text{core}}/2 = \Delta + E_C/4$. While Tighe et al. found quantitative agreement with the core-energy model of localized dipoles, more detailed measurements by Delsing et al. showed substantial deviations from this picture. For large $\Delta/E_{CP}$, Delsing et al. interpreted their results as evidence for Cooper-pair/hole solitons, even though their measured activation energies showed a strong dependence on superconducting gap. Delsing et al. also report thermally-activated conduction in a 1D chain, but only in the normal state.

For a 1D chain with localized dipole excitations, one expects $E_{\text{core}} = 2\Delta + E_C$ using the soliton model of Tighe et al. and Delsing et al., and therefore $E_A = E_{\text{core}}/2 = \Delta + E_C/2 = \Delta + E_{CP}/8$. Our results for 1D chains show that $E_A$ agrees more closely with $\Delta$, with no additional term needed to account for the charging energy of electron-hole pairs on adjacent islands. The localized dipole model ignores tunneling processes that effectively lower the core energy. As noted previously, above $T^*$, a voltage threshold for conduction is no longer found. In summary, conductance above $T^*$ is consistent with the lack of an electrostatic threshold for both charge injection and activated transport.

V. CONDUCTANCE BELOW THE PARITY TEMPERATURE

Finally, we have measured conductance at 20 mK and above the threshold voltage $V_t$ as a function of the magnetic field. Data for device A is shown in Figure 6 taken at a bias voltage, $V = 4$ mV. Here we have used the fit from Figure 5 to express $B_{||}$ in terms of $\Delta$. As the superconducting gap is suppressed by the magnetic field, the conduction $G_I$, in what we will call the “injection regime”, is clearly exponentially enhanced by the factor $\exp(-\Delta/k_B T_{\text{eff}})$. In contrast to the zero-bias conductance, we find the effective temperature $T_{\text{eff}} = 340$ mK, which is considerably larger than the zero-field parity temperature for this device, $T^* = 260$ mK. This shows that charge transport below $T^*$ and above the voltage

$\Delta = 6$
threshold occurs by injection of single electrons/holes into a non-equilibrium steady state, which shows a significantly elevated effective temperature. Future experiments are needed to address the detailed nature of this steady state and its relation to the voltage threshold for conduction, $V_i$, observed at low temperatures.

VI. CONCLUSION

In conclusion, we find that for 1D Josephson-junction chains deep in the insulating regime, where $E_J \ll E_{CP}$, there is a characteristic parity temperature $T^*$, above which the insulating state is destroyed by thermally-excited BCS quasiparticles. Above $T^*$, an observable zero-bias conductance appears and is thermally-activated with an activation energy equal to the superconducting gap. This can be understood most simply if charge carriers are single electrons and holes rather than Cooper pairs. The situation is somewhat analogous to donor ionization in a doped semiconductor, although here the donors are the background of localized Cooper pairs, and the effective ionization energy is strongly renormalized due to the singular BCS density of states. Conduction in temperatures below $T^*$ occurring above the threshold voltage appears to be thermally-activated, with an exponent equal to the ratio of the superconducting gap to an effective thermal energy, $k_B T_{\text{eff}}$. The effective temperature $T_{\text{eff}}$ is found to be significantly higher than the electronic temperature that would otherwise exist in the array. This indicates that a non-equilibrium steady state of unpaired charge carriers becomes established, enabling above-threshold charge transport below the parity temperature in the Cooper-pair insulator. Our results are also relevant to studies of disordered superconducting films, which are often modeled using a picture of weakly coupled superconducting islands.

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