Quantitative measurements of the thermopower of Andreev interferometers

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Using a new second derivative technique and thermometers which enable us to determine the local electron temperature in a mesoscopic metallic sample, we have obtained quantitative measurements of the low temperature field and temperature dependent thermopower of Andreev interferometers. As in previous experiments, the thermopower is found to oscillate as a function of magnetic field. The temperature dependence of the thermopower is nonmonotonic, with a minimum at a temperature of \( \approx 0.5 \) K. These results are discussed from the perspective of Andreev reflection at the normal-metal/superconductor interface.

I. INTRODUCTION

In the last few years, investigations of normal metal/superconductor (NS) interface structures have shown a wealth of new and interesting phenomena associated with the penetration of superconducting correlations into the normal metal. These effects can be understood as arising from phase-coherent Andreev reflection of quasiparticles at the NS interface \( \phi \): an electron is retroreflected as a hole at the NS interface, with the concurrent generation of a Cooper pair into the superconductor. The motions of the electron and hole are correlated, and coupled by the microscopic interface \( [1] \): an electron is retroreflected as a hole at the NS interface, with the concurrent generation of a Cooper pair into the superconductor. These effects can be understood as arising from phase-coherent Andreev reflection of quasiparticles at the NS interface \( \phi \).

In addition to the electrical transport properties, the thermal transport properties of proximity coupled NS structures have also been investigated \([1, 5] \). Experiments on the thermopower \( S_A \) of Andreev interferometers \([6, 7] \) have shown that it also oscillates as a function of the magnetic flux \( \Phi \) coupled through the interferometer loop. However, a number of characteristics of the thermopower of Andreev interferometers are not understood. First, the oscillations of \( S_A \) can be either symmetric or antisymmetric with respect to \( \Phi \); the symmetry depends on the topology of the sample, but the reason why this is so is not understood. Second, the temperature dependence of \( S_A \), determined by numerically estimating the temperature gradient in the sample using a simple heat flow model, was found to be non-monotonic, with a maximum in amplitude at a temperature of \( \approx 140 \) mK \([8] \), reminiscent of the reentrant behavior seen in the temperature dependent resistance of proximity coupled normal metals. However, the temperature at which this maximum occurs is much higher than the characteristic temperature scale \( T = E_c/k_B \) which sets the scale for reentrant behavior, as demonstrated by measurements of the temperature dependent resistance of the same samples, which showed no reentrant behavior in the measured temperature range \([9] \).

In this Letter, we present new data on the thermopower of Andreev interferometers, taken using a new second derivative method, and employing a local thermometry technique \([10] \) which permits us to make quantitative measurements of the thermopower without recourse to theoretical modeling. The Andreev interferometers we have measured are shorter in length and have a topology different from those in previous measurements. Their thermopower is purely

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antisymmetric in $\Phi$, as observed before. The temperature dependence of the thermopower is non-monotonic, but the temperature at which the maximum in amplitude is observed is in the range of $T \approx 0.5$ K, indicating that the length of the sample may play a role in the temperature dependence of the thermopower.

II. SAMPLE FABRICATION AND MEASUREMENT

Figure 1(a) shows a schematic of the sample design. A dc current $I$ put through the vertical normal-metal heater wire on the left heats the electrons in the middle of the heater to a temperature $T_h(I)$. The thermal voltage across the two contact pads ($V_{th}$) on the right at the temperature $T_c(I)$ has contributions from the Andreev interferometer and a reference normal metal electrode. The upper voltage probe contains the Andreev interferometer (outlined by a dotted line), and the lower voltage probe contains the reference electrode, which is a pure gold wire in our case. The Andreev interferometer probe and the reference probe join at a single point at the heater, whose temperature $T_h(I)$ can be measured by a proximity effect thermometer (the ‘hot’ thermometer). At the other end, both the Andreev interferometer and the reference wire terminate in a large normal-metal contact whose temperature can be measured by another proximity thermometer (the ‘cold’ thermometer).

![FIG. 1. (a) Schematic of our sample design for measuring the thermopower of an Andreev interferometer. Dark gray is superconductor, lighter area is normal metal. The Andreev interferometer is encircled by a dotted line. $T_h$ and $T_c$ are the local electronic temperatures at the hot and cold sides of interferometer, measured by the hot and cold proximity thermometers. $T_b$ is the base temperature of the refrigerator. The thermal voltage is measured using the contacts labeled $V_{th}$. (b) Scanning electron micrograph of one of our actual devices fabricated by conventional electron-beam lithography techniques. (c), (d) and (e) - schematic of three different types of Andreev interferometers: ‘parallelogram’-, ‘house’- and ‘hook’-like configurations respectively.](image)

Figure 1(b) shows a scanning electron micrograph of an experimental realization of this design implemented using multi-level electron beam lithography on oxidized silicon substrates. The 65 nm thick Au normal metal regions were patterned and evaporated first, after which the 65 nm thick Al film was evaporated following an O$_2$ plasma etch to ensure good interfaces between the Au and Al films. The overall length of the normal part of the Andreev interferometer and the reference wire is $\approx 2.7$ $\mu$m. From weak localization measurements on long Au wires with similar properties, the electron phase coherence length $L_\phi$ was found to be $L_\phi \sim 3.5$ $\mu$m at $T = 300$ mK, and the diffusion constant in the Au was $D = 1.4 \times 10^{-2}$ m$^2$/s, resulting in a thermal diffusion length $L_T = \sqrt{\hbar D/k_B T} = 0.32$ $\mu$m at $T = 1$ K.

In previous work [9], thermopower measurements on Andreev interferometers of two different topologies were reported. In the first (see Fig. 1(c)), denoted the ‘parallelogram,’ a part of the superconducting part of the Andreev interferometer was placed along the temperature gradient. The second (Fig. 1(d)), denoted the ‘house,’ had no superconducting parts along the temperature gradient. This difference gave rise to characteristically different behavior as a function of magnetic field: while the thermopower oscillations in the house interferometer were symmetric with respect to magnetic field, the thermopower oscillations in the parallelogram interferometer were antisymmetric with respect to field. The topology of the samples in this experiment is similar to the parallelogram geometry, although only one arm of the superconductor is in the path of the temperature gradient (Fig. 1(e)).

The samples were measured in a $^3$He refrigerator with a base temperature of 260 mK. The ‘hot’ and ‘cold’ proximity effect thermometers were first calibrated by measuring their four-terminal resistance with an ac resistance bridge as a function of the temperature $T_b$ of the refrigerator, with no current through the heater. $T_b$ was then kept constant, and the resistance of the thermometers measured as a function of the dc heater current. By cross-correlation of the two measurements, we could obtain the electron temperatures as a function of heater current ($T_h(I)$ and $T_c(I)$). This process was repeated for different values of $T_b$. Figure 2 shows the result of this measurement for the hot thermometer at a few representative temperatures.
The thermal voltage generated between the two voltage contacts (see Fig.1(a)) is given by the equation

\[ V_{th} = \int_{T_e(I)}^{T_h(I)} [S_A - S_N]dT. \]  

(1)

Here \( S_A \) is the thermopower of the Andreev interferometer, and \( S_N \) is the thermopower of the reference. We shall assume that \( S_N \) is small or that it does not vary as a function of external parameters such as the magnetic field, so that it can be neglected in our analysis. (Since the reference wire is made from Au, both conditions are satisfied in our case.) For small values of the dc current (\( \leq 2 \mu A \)), the measured \( T_c(I) \) is essentially independent of temperature and equal to \( T_b [12] \). Taking the derivative of Eq. (1) with respect to current, one then obtains

\[ \frac{dV_{th}}{dI} = S_A \frac{dT_h}{dT}. \]  

(2)

In our previous work [3], the thermopower was determined using this relation. \( dV_{th}/dI \) was measured by superposing a low frequency (\( \sim 10 \) Hz) ac current on top of the dc heater current, and measuring the resulting ac voltage between two large normal metal contacts kept at \( T_h \) at the same frequency. By estimating \( dT_h/dI \) based on a simple heat flow model, we could then estimate \( S_A \). Since \( T_h(I) \) is symmetric in \( I \), \( dT_h/dI = 0 \) at \( I = 0 \), so that one needs to apply a finite dc current in order to obtain a finite ac voltage. The application of a finite dc current heats the electron gas, and complicates the analysis of the temperature dependence. In order to avoid this problem, one can consider the derivative of equation (2) at \( I = 0 \)

\[ \left. \frac{d^2V_{th}}{dI^2} \right|_{I=0} = S_A \left. \frac{d^2T_h}{dI^2} \right|_{I=0} + \left. \frac{dS_A}{dI} \frac{dT_h}{dT} \right|_{I=0} = S_A \left. \frac{d^2T_h}{dI^2} \right|_{I=0} \]  

(3)

since \( dT_h/dI = 0 \) at \( I = 0 \). From the measured value of \( d^2V_{th}/dI^2 \) and a knowledge of \( d^2T_h/dI^2 \) at \( I = 0 \), one can determine \( S_A \). \( d^2V_{th}/dI^2 \) is determined by measuring the ac voltage at a frequency of \( 2f \), where the frequency of the ac current through the heater is \( f \) (with no dc current). \( d^2T_h/dI^2 \) at \( I = 0 \) is determined by taking the numerical derivative of the curve shown in Fig. 2. The resulting values at different \( T_h \) temperatures are shown as an inset to Fig. 2. This procedure gives us a direct quantitative value for \( S_A \), without the need to apply the dc current through the heater and to model the heat flow in the wire in order to estimate the electron temperature.

III. EXPERIMENTAL RESULTS

Figure 3(a) shows \( dV_{th}/dI \) and \( d^2V_{th}/dI^2 \) as a function of the heater current \( I_{dc} \) for the Andreev interferometer sample shown in Fig. 1(a), measured with an ac heater current of rms amplitude \( 2 \mu A \). \( dV_{th}/dI \) is antisymmetric and \( d^2V_{th}/dI^2 \) is symmetric with respect to \( I \), as should be expected for the thermal response, since \( T_h(I) \) is symmetric in \( I \). Due to the small signal to noise ratio in the \( d^2V_{th}/dI^2 \) measurement, the curve has a small offset. To take into account this offset, we match the value of \( d^2V_{th}/dI^2 \) at \( I = 0 \) with the numerical derivative of the \( V_{th}/dI \) vs \( I \) curve taken at the lowest measurement temperature.

As has been reported before [13], the thermopower of these samples oscillates as a function of magnetic field, with a fundamental period corresponding to one superconducting flux quantum \( \Phi_0 = \hbar/2e \) through the area of the Andreev interferometer loop. Figure 3(b) shows \( dV_{th}/dI \) at \( I_{dc} = 5 \mu A \) as a function of the magnetic field perpendicular to the plane of the Andreev interferometer (in terms of magnetic field, \( \Phi_0 \) corresponds to 22.8 Gauss for this Andreev interferometer). The thermopower is antisymmetric with respect to magnetic field. This is similar to the thermoelectric response of the parallelogram sample of Ref. [9]. For comparison, we also show in the same figure the oscillations of the resistance of the Andreev interferometer, which are symmetric with respect to magnetic field.

In order to obtain the temperature dependence of the thermopower \( S_A(T) \), we set the magnetic field at \( H = 5.7 \) G which is equal to \( \Phi_0/4 \), corresponding to the maximum value of \( V_{th}/dI \), and measure the second derivative \( d^2V_{th}/dI^2 \) as a function of temperature with zero dc current through the heater. The filled circles in Fig. 4(a) show the result of this measurement, with an ac current of rms amplitude \( 2 \mu A \) through the heater. To obtain the thermopower, we must divide \( d^2V_{th}/dI^2 \) by \( d^2T_h/dI^2 \) at \( I = 0 \) at the appropriate temperature. The circles in the inset to Fig. 2 show the measured temperature dependence of this quantity. To obtain values at intermediate temperatures, we fit the points to a power law in \( T \), as shown by the dotted line in the inset. Finally, the thermopower \( S_A \) is obtained by dividing the measured \( d^2V_{th}/dI^2 \) by these interpolated values. The open circles in Fig. 4(a) show the result of this calculation. Due to the small value of \( d^2T_h/dI^2 \) at higher temperatures, the data are very noisy in this regime. Nevertheless, the temperature dependence is clearly non-monotonic, with a minimum at \( T \approx 0.5 \) K. As was
observed in previous experiments, this temperature does not appear to be related to the correlation energy $E_c$, which corresponds to a temperature $T = E_c/k_B \approx 0.015$ K. Evidence for this can be seen in the temperature dependence of the resistance of the interferometer, which is shown in Fig. 4(b). The resistance decreases monotonically below the transition temperature of the superconductor, showing no hint of reentrance, with an overall change of about of 3.8%.

FIG. 2. Temperature of hot thermometer as a function of dc current through the heater at different base temperatures $T_b$. Insert shows temperature dependence of the calculated value of $d^2T/dI^2$ at $I = 0$. The dashed line in the inset is a fit to the points by the equation $d^2T/dI^2 = 1.1 \times 10^{-4} \times T^{-3.2}$.

FIG. 3. (a) $dV_{th}/dI$ and $d^2V_{th}/dI^2$ as a function of $I$ for the Andreev interferometer of Fig.1(a). Magnetic field is 5.7 G, corresponding to a flux $\Phi_0/4$ through the area of the interferometer loop. (b) Magnetic field dependence of the $dV_{th}/dI$ at $I_{dc} = 5\mu$A (solid line) and resistance of the Andreev interferometer (dashed line). Dependences were measured at $T_b = 0.296$ K.

FIG. 4. (a) Filled circles represent the measured $d^2V_{th}/dT^2$ as a function of $T$ at a magnetic field of 5.7 G. Open circles represent the calculated value of $S_A$ as discussed in the text. Solid lines are guides to the eye. (b) Resistance of the Andreev interferometer as a function of temperature.
The most surprising aspect of the thermopower of Andreev interferometers is the fact that it can be antisymmetric with respect to $\Phi$, even though the resistance is always symmetric. An antisymmetric magnetic field dependence implies that the thermopower must change sign as a function of $\Phi$. If one associates a negative thermopower with electron-like quasiparticles, and a positive thermopower with hole-like quasiparticles, this implies that quantum mechanical interference modulated by the magnetic field results in a change of the nature of the excitations which carry the thermal current.

What are the possible origins of this asymmetry? In numerical calculations, Claughton and Lambert showed that the thermopower in mesoscopic NS interferometers could be symmetric or antisymmetric with respect to field, depending on the topology of the sample. However, their arguments depended on the devices having an axis of perfect mirror symmetry; for samples such as ours without this feature, the thermopower was predicted to be neither purely symmetric nor antisymmetric. A second explanation can perhaps be found in the phase dependent oscillations of thermoelectric quantities that have been observed in early experiments on so-called ‘thermal SQUIDs,’ which are doubly connected loops in which one arm is fabricated from one superconductor and the second from another superconductor with a different gap $\Delta$. A temperature gradient applied across the loop will result in a different quasiparticle current in each arm. This quasiparticle current is balanced by a counterflowing supercurrent in each arm of the loop. Since the quasiparticle currents in each superconductor are different, the counterflowing supercurrents are also different, resulting in a net circulating supercurrent in the loop. The circulating supercurrent results in a phase gradient which is proportional to the temperature gradient, and which can be detected by measuring the critical current of the loop as a function of magnetic field, for example. With this in mind, one might consider the Andreev interferometer to be a ‘thermal SQUID,’ with one superconductor being the proximity coupled normal metal arm. A temperature differential $\Delta T$ would then result in a phase difference between the two NS interfaces, and a shift in the thermopower oscillations as a function of magnetic field.

A number of experimental observations argue against this interpretation. First, the presence of a supercurrent would be expected to change the phase of the resistance oscillations as well as the thermopower oscillations. As we have observed, the thermopower oscillations are shifted by $\pi/2$, while the resistance oscillations show no shift with respect to magnetic field. Second, the phase shift of the oscillations would depend on the temperature gradient $\Delta T$, which can be controlled by varying the dc current through the heater. However, the phase shift of the thermopower oscillations for this geometry is always exactly $\pi/2$; no change in the phase of either the thermopower or the resistance oscillations is observed as a function of dc heater current. Consequently, we do not believe a temperature induced supercurrent is responsible for the asymmetry. To our knowledge, the dependence of the symmetry of the thermopower on sample topology has not been satisfactorily explained.

The temperature dependence of the thermopower is similar in form to what has been observed before– it is non-monotonic, being zero at higher temperatures, having a maximum in magnitude at some intermediate temperature, and then going to zero again at lower temperatures. Although the electrical transport properties of a variety of NS structures have been calculated in detail, reliable theoretical predictions for the thermopower are not yet available. This is because the usual formulation of the quasiclassical theory of superconductivity which is used as a starting point for the calculations assumes particle-hole symmetry, and hence ignores thermoelectric effects from the beginning. Extensions of the quasiclassical theory to include thermoelectric effects have proved difficult. Nevertheless, one can make some useful observations on the differences between the temperature dependence measured here and that reported in Ref. First, the temperature $T_{\text{min}} \simeq 0.5$ K at which the minimum in thermopower is observed in this experiment is greater than the temperature $T_{\text{min}} \simeq 0.14$ K observed in Ref. The normal line of Andreev interferometer in that experiment had a length of $\simeq 7\mu$m; this sample has a length of 2.7 $\mu$m. For the resistance, $T_{\text{min}}$ is expected to scale with $E_c \sim 1/L^2$, i.e., it is larger for smaller $L$, which is the trend we observe in the thermopower. Second, the overall magnitude of the thermopower is much smaller than that measured in Ref., with a maximum of about 100 nV/K compared to 4 $\mu$V/K observed in Ref. This difference may be understood as arising again from the topology of the samples. The Andreev interferometers in this experiment have a superconductor in the path of the temperature gradient. Experiments on the thermal conductance of the Andreev interferometers similar in topology to this sample show that the thermal conductance of the Andreev interferometer is limited by the thermal conductance of the small superconducting part. Hence, most of the temperature gradient across the interferometer is dropped across the superconducting part, and very little across the proximity-couple normal metal, resulting in a small resultant thermoelectric voltage. This is in contrast to the ‘house’ interferometer whose temperature dependence was discussed in Ref., which had no superconductor along the path of the temperature gradient.

In conclusion, we have made detailed quantitative measurements of the thermopower of Andreev interferometers using a new second derivative technique. The measurements confirm the two main qualitative observations from previous experiments: the unusual symmetry of the thermopower with respect to magnetic field, and the non-monotonic...
temperature dependence of the thermopower. Further theoretical and experimental work is required to understand the origin of these effects.

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