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Reversal of thermopower oscillations in the mesoscopic Andreev interferometer

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We report measurements of phase-periodic thermopower in a diffusive Andreev interferometer. Upon the increase of the dc current applied to the heater electrodes, the amplitude of the thermopower oscillations first increases then goes to zero as one would expect. Surprisingly, the oscillations reappear at yet higher heater currents being inverted compared to low current values. The dependence of the amplitude of the oscillations on temperature strongly correlates with that of the resistance derivative dR/dT.

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In a nonuniformly heated conductor there arises an electric field E proportional to the temperature gradient $E = Q\nabla T$, where Q is known as thermopower. In metals Q is determined[1] by a derivative of the logarithm of conductivity σ with respect to energy ε taken at the Fermi level

$$Q=\frac{\pi^2}{3}\frac{k_B^2 T}{e}\left\{\frac{\partial ln\sigma}{\partial\varepsilon}\right\}_{\varepsilon=\varepsilon_F}, \qquad (1)$$

where k_B is Boltzman constant and e is electron charge. In normal metals with diffusive electron transport the conductivity changes very little with energy and the thermopower has the following order of magnitude:

$$Q=C\frac{k_B}{e}\frac{k_B T}{\varepsilon_F}, \qquad (2)$$

where C is a constant of the order of unity depending on the topology of Fermi surface and the energy dependence of scattering time.

The thermoelectric properties of a normal metal (N) in contact with a superconductor (S) will be strongly modified by the superconducting proximity effect (see Ref. 2 for a review on the proximity effect). The theory predicts that the thermopower in this case can be orders of magnitude larger than predicted by Eq. (2) and that Mott's relation (1) may break down.[3,4] In the geometry of Andreev interferometer (AI), the thermopower has been predicted to oscillate as a function of the magnetic flux Φ through the loop, with a period equal to the flux quantum $\Phi_0 = hc/2e$.[3] Andreev interferometer is a device in which a normal mesoscopic part is connected to a superconducting loop; the conductance of AI oscillates as a function of phase difference between the S banks due to multiple Andreev reflections at the N/S interfaces.[2]

Recently, the oscillating thermopower of mesoscopic (Au/Al) AI has been discovered in a pioneering experiment by Chandrasekhar's group.[5] The value of Q was estimated to be $4\mu V/K$ in agreement with theoretical predictions of few $\mu V/K$.[3,4] Later experiments by the same group with direct measurements of temperature gradients reported thermopower of 100 nV/K which is still orders of magnitude larger than the thermopower of Au at low temperatures.[6] The validity of Mott's relation has not been tested. In previous experiments the thermopower was measured using N probes. Reference 4 has pointed that there can be remarkable difference from the case when the thermopower is measured between N and S probes connected to the N/S interface forming AI.

In this Communication we report measurements of thermopower oscillations vs magnetic field in a (Sb/Al) AI between N and S probes. The comparison of the results with the case of all normal probes is reported elsewhere.[7] As a function of heater current the amplitude of oscillations first increases then goes to zero similar to that in Ref. 5. However, we have discovered another effect: at higher heater currents the oscillations reappear being inverted compared to low current ones. We show that temperature dependence of the amplitude of oscillations correlates strongly with the derivative of resistance by temperature dR/dT so that the amplitude follows Eq. (1). Semimetal Sb was chosen as a normal part because it has a large classical thermopower which can be measured in the same experiment, in order to compare it with thermopower of AI.

The structures were made by multilayer electron-beam lithography as shown in the scanning electron micrograph (Fig. 1). The first layer was 40 nm thick Sb (semimetal) followed by second layer of 60 nm thick Al (supercon-

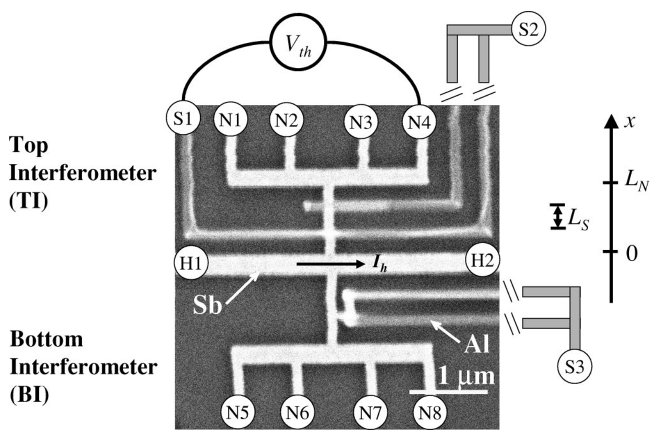

FIG. 1. Scanning electron micrograph of the top (TI) and bottom (BI) interferometers. Electrode notation: $H1$-$H2$ - heater, $N1$-$N4$ TI normal probes, $S1$-$S2$ TI superconducting probes, $N5$-$N8$ BI normal probes, $S3$ - BI superconducting probe. For thermopower measurements current I_h was sent through the heater, while thermoelectric voltage V_{th} was measured between an N and an S electrode.

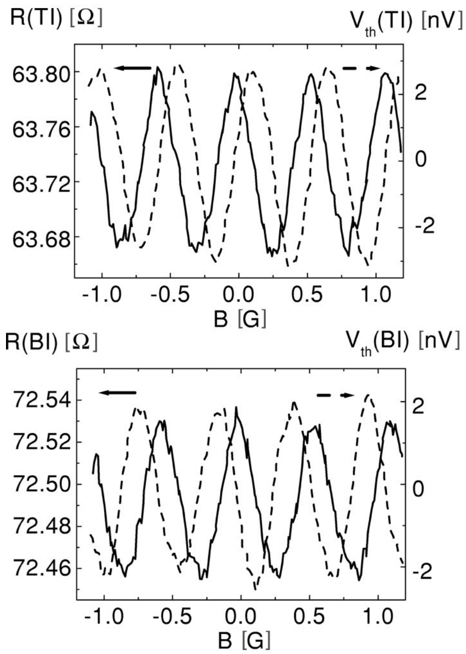

FIG. 2. Magnetoresistance and thermoelectric voltage oscillations at $T=0.28$ K. The period of oscillations corresponds to the flux quantum through the area of the loop. Top panel: TI magnetoresistance measured using current leads $N1$-$H1$ and potential leads $N4$-$H2$ (solid line); TI thermovoltage measured using current $H1$-$H2$ $(1\ \mu A)$, potentials $S2$-$N1$ (broken line). Bottom panel: BI magnetoresistance, current $N5$-$H1$, potentials $N8$-$H2$ (solid); BI thermoelectric voltage, current $H1$-$H2$ $(1\ \mu A)$, potentials $S3$-$N5$ (broken).

ductor). Prior to the deposition of the second layer *in situ* Ar^+ etching was used to clean the interface. Two hybrid loops form two AI which we will call ''top interferometer'' (TI) with interfaces to superconductor situated on the current lines of N-part and ''bottom interferometer'' (BI) with the interfaces being off current lines (see Fig. 1). In this geometry magnetoresistance and thermovoltage on both interferometers can be measured as a function of the same heating current applied between $H1$ and $H2$. This allows us to compare TI and BI and to estimate the temperature gradient across an interferometer, so that the absolute value of thermopower could be determined.

Measurements were performed in a He^3 cryostat in temperatures from 0.28 to 6 K with a magnetic field up to 5T applied perpendicular to the substrate. Resistivity ρ of Sb film was 60 $\mu\Omega$ cm and that of Al film was 1.2 $\mu\Omega$ cm, with diffusion constants D 133 and 223 cm^2/s, respectively. The resistance of interface between the two films in normal state was 8Ω for the interface area $150\times150\ nm^2$.

Magnetoresistance measurements were performed using conventional ac bridge technique. For thermopower measurements a heating current was a sum of dc current I_h and a small ac current I_m. The variation in thermoelectric voltage V_{th} was measured between S and N electrodes (Fig. 1) using lock-in amplifier on the frequency of ac modulation. Thermopower measured between electrodes $S1-N4$ was identical to that measured between $S2-N4$. A superconducting magnet was used to sweep magnetic field. The zero of magnetic field on graphs in fact corresponds to zero current through the magnet as there can be arbitrary magnetic flux trapped in the magnet, resulting in a shift along the B axis. The relative position of thermopower and magnetoresistance oscillations was double checked by repeated measurements to ensure they were measured with the same reference point.

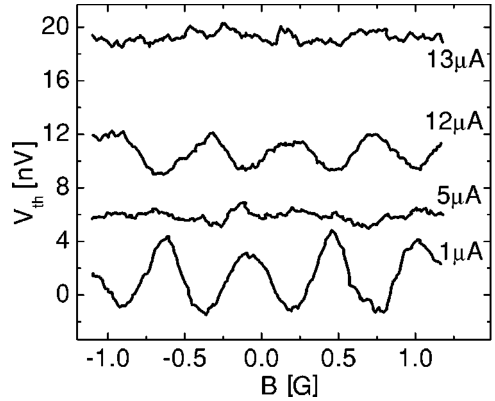

FIG. 3. Thermoelectric voltage of the TI as a function of magnetic field for heater currents $I_h=1$, 5, 12, and 13 μA. $T=0.28$ K.

Oscillations of thermoelectric voltage V_{th} are leading the magnetoresistance ones by $\pi/2$ for TI and are lagging by $\pi/2$ for BI (Fig. 2) for the same polarity of the connection of S and N electrodes to measure V_{th}. We think that there is temperature gradient (due to electron-phonon coupling) along N wire for BI up to N/S interfaces, so that the closest to the N reservoir N/S contact will have higher temperature for BI, contrary to TI. Thus, the temperature gradient will be opposite for BI and TI, resulting in opposite phase of thermopower oscillations.

It is interesting to note, that in our experiment the oscillations of the thermopower for the BI with N/S interfaces off classical current lines between the N reservoirs (corresponding to the house structure of Ref. 5) were $\pi/2$ shifted from magnetoresistance oscillations (as opposed to the two being in phase in Ref. 5). The symmetry of oscillations has been discussed in Refs. 3,6 but there is still no clear understanding of its dependence on sample topology.

Figure 3 shows V_{th} vs magnetic field oscillations of the TI for four different dc currents I_h. With increasing I_h the oscillations first disappear, and then remarkably reappear inverted compared to low I_h measurements. The phase of the thermopower oscillations at each I_h was checked against that of the magnetoresistance, which remained the same for all temperatures and currents. We measured five different samples and all of them showed similar reversal of thermopower oscillations. Magnetic field independent shift of the curves in Fig. 3 with increasing I_h was reproducible on the same sample but was not consistent from sample to sample. The origin of this shift is unclear. Figure 4 shows in detail the dependence of the peak-to-peak amplitude A_{th} of V_{th} oscillations for both our interferometers on the applied heater current. Both interferometers showed remarkable new maximum, which was also observed on other samples.

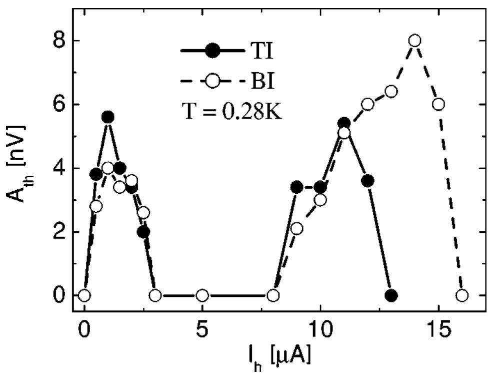

FIG. 4. Amplitude of thermoelectric voltage oscillations as a function of dc heater current. Filled circles: TI; open circles: BI. $T=0.28$ K.

To estimate temperature gradient in normal wire we have used proximity effect in the TI as a thermometer.[8] Figure 5 shows the amplitude of the magnetoresistance oscillations of the TI as a function of temperature and dc current I_h. Relating the amplitude on left and right panels of Fig. 5 we obtain the temperature T_m in the middle of the normal part of the TI vs I_h (Fig. 6, low panel inset). The reliable estimation of T_m using the above method is only possible at small currents when T_m is far away from the critical temperature of the superconducting transition. This is because close to the superconducting transition the temperature dependence of the proximity effect is governed by the temperature dependence of the gap rather than actual electron temperature.[9] Also the reservoir temperature measured using $N5-N8$ electrodes departs from T_0 at high I_h.

At our measurements V_{th} relates to the thermopower of AI, Q_A, according to the following formula (see also Refs. 6 and 8):

$$V_{\rm th}=Q_A\left\{\frac{\partial T_m}{\partial I}\right\}_{I=I_h} I_m . \qquad (3)$$

We calculate thermopower for current $I_h=1\ \mu$A at which the reservoir temperature did not deviate from T_0, so that

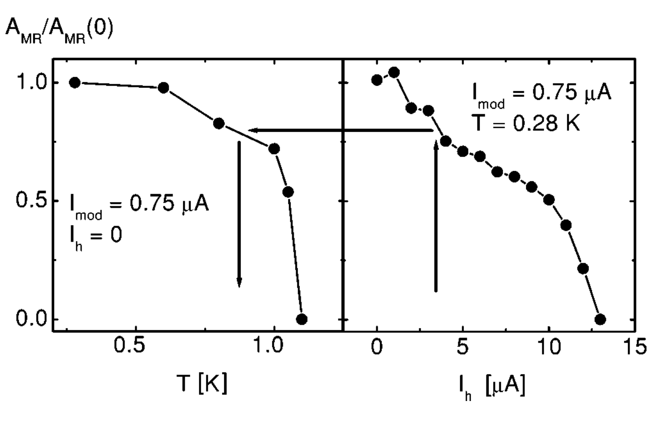

FIG. 5. Reduced amplitude of magnetoresistance oscillations measured using current leads $N1$-$S1$ and potential leads $N4$-$S2$ as a function of temperature (left) and heater current ($H1$-$H2$) (right). Modulation ac current was $I_{\rm mod}=0.75\ \mu$A.

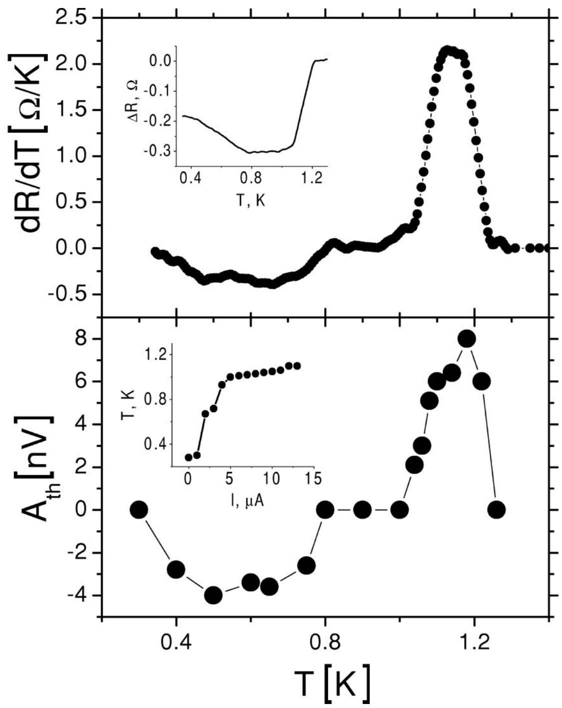

FIG. 6. Top panel: temperature dependence of the resistance (inset) and its derivative on temperature for BI. Bottom panel: amplitude of $V_{\rm th}$ oscillations for BI transformed from Fig. 4 using temperature-current correspondence (inset) extracted from Fig. 5. The sign of $A_{\rm th}$ reflects reversal of oscillations.

Eq. (3) remains valid. For $I_h=1\ \mu$A we have $T_m\approx0.36\pm0.02$ K. Substituting the corresponding value of $\partial T_m/\partial I=0.1\pm0.04$ K/μA at $I=I_h$ obtained from the inset of Fig. Fig. 6(b) and $I_m=0.75\ \mu$A into Eq. (3) yields $Q_A\approx73$ nV/K. We estimate relative inaccuracy of this thermopower value to be about 50%. The theory predicts values of a few μV/K.

We have also measured classical thermopower of Sb $Q_{\rm cl}$ in samples of similar geometry with all normal electrodes. For the same conditions of experiment no $V_{\rm th}$ was observed in this case down to the level of 0.1 nV. This means that $Q_{cl}<1.3$ nV/K at $I_h=1\ \mu$A. Therefore, the experimentally observed ratio $Q_A/Q_{cl}>55$. Note that Eq. (2) predicts $Q_A/Q_{\rm cl}\approx\varepsilon_F/k_BT\approx8000$.

The estimation $Q_{cl}<1.3$ nV/K is an order of magnitude smaller than expected value of $Q_{\rm cl}=36$ nV/K as extrapolated from the table value of $Q_{\rm cl}=36\ \mu$V/K for Sb at $T=273$ K.[10] Here we neglect the phonon drag effect in Sb because it has maximum at 10 K and drops as T^3 at low temperatures, so that it should be negligible at the temperatures of our experiment.[11] We find that experimental values for both Q_A and $Q_{\rm cl}$ are about two orders of magnitude smaller than that predicted by theory, but Q_A is indeed giant compared to $Q_{\rm cl}$ being at least 55 times larger.

Let us now discuss the origin of the second maximum in $A_{\rm th}$ seen at higher I_h in Fig. 4. Figure 6 shows the temperature dependence of the resistance (top panel inset) and its derivative (top panel) for BI. The bottom panel shows amplitude of $V_{\rm th}$ oscillations for BI transformed from Fig.

4 using temperature-current correspondence (inset) extracted from Fig. 5. The sign of $A_{\rm th}$ reflects reversal of oscillations. Strong correlation between dR/dT and $A_{\rm th}$ suggests that $A_{\rm th}$ follows Eq. (1). For $T=0.36$ K we get $dR/dT\approx 0.07\Omega/K$. Substituting $(1/R)(dR/dT)$ for $\partial(\ln\sigma/\partial\varepsilon)$ into Eq. (1), with $R=72.5\Omega$ we get thermopower of approximately 100 nV/K close to our estimation of measured thermopower. At higher temperatures $A_{\rm th}$ can go down because of reduced phase-breaking length. The use of dR/dT in Eq. (1) instead of $\{\partial\sigma/\partial\varepsilon\}_{\varepsilon=\varepsilon_F}$ can be justified by the fact that at low applied voltages the temperature sets the energy window available for quasiparticles. The same correlation as in Fig. 6 was detected on other samples where the second maximum in $A_{\rm th}$ has been observed. Thus, we have established that the new second maximum occurs when the system is driven through the superconducting transition by the heating current. That is probably why it was not observed in Ref. 5.

In conclusion, we have observed the reappearance and reversal of the phase-dependent thermopower in diffusive (Sb/Al) Andreev interferometer at heating current increase. This behavior has been explained on the basis of Mott's relation (1). The absolute value of both Andreev and classical thermopower was found orders of magnitude smaller than theoretical predictions, with Andreev thermopower being giant compared to classical one. This work invites discussion about the role of Mott's relation for the thermopower in mesoscopic N/S structures.

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