Limitations in cooling electrons by normal metal - superconductor tunnel junctions

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We demonstrate both theoretically and experimentally two limiting factors in cooling electrons using biased tunnel junctions to extract heat from a normal metal into a superconductor. Firstly, when the injection rate of electrons exceeds the internal relaxation rate in the metal to be cooled, the electrons do no more obey the Fermi-Dirac distribution, and the concept of temperature cannot be applied as such. Secondly, at low bath temperatures, states within the gap induce anomalous heating and yield a theoretical limit of the achievable minimum temperature.

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Refrigerators are generally characterised by their cooling power, coefficient of performance, and operating temperature under various working conditions. To assign a temperature to a system, one needs to assume that the energy relaxation within the system is faster than any rate associated with the heat flux between the system in concern and its surroundings. If this condition fails, the energy distribution of the particles of which the system is formed is non-thermal, and applying the concept of temperature is strictly speaking inappropriate. Such a limit can be achieved in submicron-size coolers at low temperatures. The structure we study is a symmetric configuration of a NIS refrigerator, formed by a series connection of two Superconductor (S) - Insulator (I) - Normal metal (N) tunnel junctions sharing the N island to be cooled in between them (SINIS). We demonstrate two striking phenomena occurring in these electron microcoolers at low temperatures: evidence of non-thermal energy distribution of the cooled electrons and re-entrant behaviour with anomalous heating at low bias voltages. The cooler performance is typically limited by coupling of the electrons to the underlying lattice (electron-phonon, e-p coupling). This has, however, strong dependence on temperature $T$: relaxation rate $\tau_{e-p}^{-1}$ slows down on lowering $T$ typically as $\tau_{e-p}^{-1} \propto T^3$. Consequently, at low enough temperature, characteristically around 100 mK, the behaviour of the cooler can be described as if the lattice would not exist at all. The interplay of the rates for e-p, electron-electron (e-e) and the injection through the junctions determines the distribution in the normal metal. If e-p or e-e rates are fast, the system assumes Fermi-Dirac energy ($E$) distribution $f_0(E,T_e)$:

$$f_0(E,T_e) = \frac{1}{1 + \exp(E/k_B T_e)}, \quad (1)$$

In the limit of very strong e-p relaxation, $T_e$ equals the temperature of the lattice. We call this equilibrium. On the other hand, if e-p relaxation is slow, and e-e relaxation is fast, $T_e$ is, in general, different from the lattice temperature (quasi-equilibrium). Finally, the slow e-e relaxation as compared to the injection rates implies that the electrons assume a non-equilibrium energy distribution $f(E)$, which cannot be written as Eq. (1), and it is not possible to assign a true temperature to them.

Our discussion is motivated by a puzzling experimental observation in many coolers at temperatures below or around 100 mK. Figure 1 shows in (a) a typical SINIS cooler. The device has been fabricated by standard electron beam lithography. The central part forms the N island of copper (purity nominally 99.9999 %). The injecting tunnel junctions, with normal-state resistances $R_T$ (100 $\Omega$ - 2 k$\Omega$), contact the island symmetrically into the two superconducting aluminium reservoirs at the two ends of the N island. The overlapping extra copper shad-
ows outside the SINIS structure provide better thermalisation of the Al reservoirs. Two additional NIS probe junctions, with normal-state resistances $\gg 1 \text{ k}\Omega$ in the centre are used to measure the temperature of the N island or to probe the distribution through the differential conductance of the nominally symmetric series connection. $R_T \simeq 200 \text{ \Omega}$ and thickness of the copper film $\simeq 30 \text{ nm}$ are the essential parameters of the sample (S1) whose data are presented in this paper. The data in Fig. 1 (b) were taken by applying a constant current $I_0$ through the probe, which, due to the thermal rounding of the current-voltage characteristics, provides, by detecting voltage $V_P$ across it, a measure of the electron temperature on the N island. We calibrated this dependence by varying the bath temperature of the cryostat. Typically one applies $I_0$ such that the voltage remains within the gap region of the superconductor, $V_P < 2\Delta/e$, whereby excess heating or cooling by the probes is avoided. Here $\Delta$ is the energy gap of the superconductor and $e$ is the electron charge ($\Delta/e \simeq 0.2 \text{ mV}$). Figure 1 (b) shows $V_P$ against injection voltage $V_C$; the several curves represent different bath temperatures. The approximate temperature calibration is given on the right vertical axis. At all but the lowest bath temperature of about $50 \text{ mK}$, the curves show the expected refrigeration behaviour symmetrically around $\Gamma$. The lowest-temperature curve demonstrates, however, a feature which appears as heating at low values of $V_C$ and re-entrant cooling again close to $2\Delta/e$. This behaviour is common with many similar samples, and it appears only below $200 \text{ mK}$. Moreover, the measured conductance curves of the probe junctions (Fig. 4) indicate that at low temperatures the actual temperature we assign in a $V_P$ measurement depends on the choice of $I_0$.

To understand the observed behaviour, we consider the properties of the kinetic equation describing the dominant processes in our system. We assume that the two reservoirs are identical and the quasiparticles have a thermal distribution of Eq. 1, with $T_C \equiv T_S$ on them. In steady state $f(E)$ on the N island is then determined by

$$\frac{\delta}{e^2 R_T} [n(E_R) [f_0(E_R, T_S) - f(E)] + n(E_L) [f_0(E_L, T_S) - f(E)]] = I_{coll}[f; E]. \quad (2)$$

Here $I_{coll}[f; E]$ is the collision integral discussed below, $\delta$ is the level spacing on the island, $E_{LR} = E \pm eV_C/2$ are energies on the left (L) and right (R) reservoirs, and

$$n(E) = |\text{Re}(\frac{E + i\Gamma}{\sqrt{(E + i\Gamma)^2 - \Delta^2}})| \quad (3)$$

is the broadened BCS density of states (DOS) of the superconductor. $\Gamma$ smears the DOS singularity at $E = \pm \Delta$ and allows for states within the gap, e.g., due to inelastic electron scattering in the superconductor or by inverse proximity effect due to nearby normal metals. A more phenomenological choice is to add a non-zero constant $\Gamma/\Delta$ to the ideal singular DOS ($\Gamma \equiv 0$). This choice would not essentially affect our conclusions.

It is illustrative to investigate first the case where relaxation (both e-e and e-p) is very weak, i.e., when $I_{coll}[f; E] \equiv 0$ in Eq. 2. Then we obtain an explicit expression for $f(E)$ as

$$f(E) = \frac{n(E_R) f_0(E_R, T_S) + n(E_L) f_0(E_L, T_S)}{n(E_R) + n(E_L)}. \quad (4)$$

The solution of Eq. 2 is plotted in Fig. 2 (a) for five different values of $v_C \equiv eV_C/\Delta$, assuming $\eta \equiv \Gamma/\Delta = 1 \cdot 10^{-4}$, and $T_S = 0.1T_C$. The BCS relation $\Delta \simeq 1.764k_B T_C$ has been assumed for the critical temperature $T_C$. This solution exhibits some nontrivial features. At low values of the bias voltage up to $v_C \sim 1$, the distribution first broadens whereafter it starts to get narrower again, and at $v_C = 2.0$, it becomes very narrow, effectively demonstrating strong cooling. In each case, except at $v_C = 0$ (equilibrium), the distribution is not thermal. At $v_C > 2.0$ the distribution would become even more unusual, but we do not consider this regime in detail for reasons to be explained below. Yet, at those voltages the distribution effectively broadens again, suggesting already the non-monotonic behaviour, heating - cooling - heating upon increasing $v_C$. The distribution is tested by measuring the differential conductance $dI/dV_P$ of the probe junctions (Fig. 1 (a)). The relation between

FIG. 2: Energy distribution $f(E)$ against $E/\Delta$ in (a), (c) and (e), and differential conductance $G$ against probe voltage $v_P$ in (b), (d) and (f) at three different values of e-e collision parameter $K$, which present very slow, intermediate and very fast e-e relaxation from top to bottom, respectively. Parameter values $\eta = 1 \cdot 10^{-4}$ and $T_S/T_C = 0.1$ have been assumed.
is the dimensionless collision integral, \( G \equiv \frac{dI}{dV_p} \) has been plotted in Fig. 2 (b) for the collisionless distributions of Fig. 2 (a) against \( v_p \equiv eV_p/\Delta \). These curves exhibit some similarities to those of biased diffusive metal wires \(^1\), especially at low values of \( v_C \), where quasi-constant DOS at low energies \( n(E) \approx \Gamma/\Delta \) mimics resistive normal-metal wire. Another important limit is quasi-equilibrium with electrons perfectly decoupled from the phonon bath. The latter condition is well justified at low \( T_S \) by a standard estimate of the minimum temperature \( T_{\text{min}} \) determined by the balance between cooling power and e-p coupling only \(^{12}\). In quasi-equilibrium fast e-e relaxation forces the distribution into a thermal one (Eq. 1\(^*\)). Any temperature satisfies this and we need to determine \( T \) by setting the net heat flux through the island, \( P(T,T_S) \), equal to zero. In our case we can write \(^2\)

\[
P(T,T_S) = \frac{2}{e^2R_T} \int_{-\infty}^{\infty} n(E)[f_0(E_R,T) - f_0(E,T_S)]E_RdE = 0, \tag{6}
\]

where factor 2 on the right hand side takes into account the flux through the two identical junctions L and R. The solutions of Eq. (6) allow us to plot the distributions and differential conductance at different values of \( v_C \) as in Fig. 2 (c) and (f). The re-entrant heating-cooling-heating behaviour survives but less pronounced than in the collisionless case. It transforms into more conventional cooling-heating behaviour above \( T_S = T^* \), given by

\[
(\Delta/k_B T^*)^{3/2} \exp(-\Delta/k_B T^*) \approx \eta/\sqrt{2\pi}. \tag{7}
\]

Thus \( T^* \) depends approximately logarithmically on \( \eta \) and assumes a value \( T^*/T_C \approx 0.125 \) when \( \eta = 1 \cdot 10^{-4} \). Equation (7) is obtained by equating at low bias the quadratic in \( v_C \) heating and cooling terms arising from the states inside and outside the gap, respectively. For an intermediate strength of e-e interaction, the distribution functions were obtained by Eq. (2) with the e-e collision integral given in Ref. \(^4\). In the case of a diffusive normal-metal island whose transverse dimensions are smaller than the coherence length \( \xi_0 = \sqrt{\hbar D/\Delta} \), one finds

\[
I_{\text{coll}} = \kappa \sqrt{\Delta I_{\text{coll}}}
\]

where

\[
I_{\text{coll}} = \int \frac{d\omega d\theta}{\omega^{3/2}} \left[ f(E)f(\epsilon\Delta)(1 - f(E - \omega\Delta))(1 - f((\epsilon + \omega)\Delta)) - f(E - \omega\Delta)f((\epsilon + \omega)\Delta)(1 - f(E))(1 - f(\epsilon\Delta)) \right] \tag{8}
\]

is the dimensionless collision integral, \( \kappa = \sqrt{\pi D\delta} \), \( D \) and \( L \) are the diffusion coefficient and the length of the island. Dividing Eq. (2) by \( \delta/e^2R_T \), we obtain a dimensionless equation where the strength of e-e scattering is governed by \( K \equiv 2\sqrt{2r_R R_C \sqrt{\frac{\Delta}{T_C}} L} \). Here \( R_C \equiv \hbar/e^2 \) and \( E_T \equiv \hbar D/L^2 \) are the resistance quantum and the Thouless energy of the island, respectively.

Although temperature is not a valid concept in non-equilibrium, we can define an effective temperature \( T_{\text{eff}} \). A natural choice is to require that \( T_{\text{eff}} \) satisfies the standard relation between the temperature and the thermal energy density of electrons in Sommerfeld expansion, which yields

\[
k_B T_{\text{eff}} = \frac{\sqrt{6}}{\pi} \int_{-\infty}^{\infty} \frac{\theta(E)}{[f(E)-1+\theta(E)]EdE}. \tag{9}
\]

Here \( \theta(E) \) is the Heaviside step function. This \( T_{\text{eff}} \) coincides with the true temperature in (quasi-)equilibrium, and it is not affected by the strength of e-e relaxation as such. Yet in a biased SINIS, \( T_{\text{eff}} \) depends on the strength of e-e relaxation, because of the heat exchange with reservoirs with non-constant DOS. Figure 3 shows \( T_{\text{eff}} \) as a function of \( v_C \) at \( T_S/T_C = 0.1 \) and \( \eta = 1 \cdot 10^{-4} \) for different rates \( K \). In the collisionless limit, the rise of \( T_{\text{eff}} \) is largest (almost threefold) and the maximum is reached at \( v_C \approx 0.8 \). On increasing the collision rate, the maximum \( T_{\text{eff}} \) gets lower and it is reached at a lower value of \( v_C \), and ultimately in quasi-equilibrium \( (K = \infty) \), the maximum is reached at \( v_C \approx 0.4 \) and its value is about \( 0.12T_C \). The minimum temperature in quasi-equilibrium at \( v_C \approx 2 \) is given approximately by \( T_{\text{min}}/T_C \approx 2.5\eta^{2/3} \) (inset of Fig. 3). Although the experimental working temperatures well below 100 mK imply that \( \eta < 0.01 \), we cannot make a direct comparison to this theoretical result in the present device. For this we would need a thermometer calibration down to the lowest temperatures, the electron system should be forced to quasi-equilibrium (magnetic...
impurities would help), and the S reservoirs should be well thermalised.

To assess the degree of non-equilibrium, we measured $dI/dV_P$ at various values of $V_C$, which allows for a semi-quantitative comparison between the experiment and theory (see Eq. (5)). In the data, especially below about 200 mK, we concentrate on the values of both $V_C$ and $V_P$ below the gap ($< 2\Delta/e$) because of the excessive heating of the reservoirs when current increases abruptly at the gap edge. The data at $T_S = 340$ mK in Fig. 4 (a) exhibit cooling behaviour in quasi-equilibrium: the conductance curves measured get first sharper monotonically on increasing $V_C$ from 0 to 0.35 mV, whereas data at $V_C = 0.45$ mV are more smeared, i.e. hotter than any other curve. All data sets conform in shape without crossing, demonstrating near-to-quasi-equilibrium behaviour. The data in Fig. 4 (b), taken at the base temperature of the cryostat of about 50 mK (best fit to $dI/dV_P$ would yield $T_S \approx 100$ mK), show, on the contrary, that the energy distribution in this case deviates from the thermal one when applying bias $V_C$. Data at $dI/dV_P \leq 0.03$ first indicate that the low-bias conductance becomes larger when increasing $V_C$ from 0 up to 0.1 mV (unlike at 340 mK), where-after the curves start to get sharper, but they heavily cross each other in this regime. At 0.45 mV the data present significant heating again. The mere fact that the data-sets corresponding to different values of $V_C$ criss-cross in the regime below 0.4 mV is a demonstration of non-equilibrium. A few data sets from (b) are magnified in (c), and the corresponding theoretical results, assuming $K = 0.05$, have been shown in (d). The resemblance is obvious, although the theoretical lines show abrupt rise from $dI/dV_P = 0$ at $V_P = V_C$ due to the influence of the gap edge. This feature is smeared in experiment most likely because of noise and finite excitation level (25 $\mu$V p-p) in the measurement.

In summary, we have shown that slow electron-electron relaxation restricts the use of the concept of temperature in electron coolers at low temperatures, and that the non-zero DOS within the gap of the superconducting reservoirs gives rise to anomalous heating and determines the ultimate minimum temperature that can be achieved.

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