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Abstract
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Effect of quantum reflection over the barrier on thermionic refrigeration

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ABSTRACT

We study the effect of quantum reflection over the barrier (ROB) in a thermionic cooling device. We find that the performance of refrigerators can be enhanced by the ROB effect if the bias voltage and the lattice thermal resistance of the semiconductor in the barrier region are both sufficiently high. Furthermore, the figure of merit $ZT$ can be higher due to the ROB effect if the workfunction of the cathode is low and the lattice thermal resistance is high. The overall optimum $ZT$ calculated with and without the ROB effect are 6.5 and 7.1, respectively. The origin of the ROB correction to $ZT$ is that the quantum reflection becomes asymmetric for the carrier transport in opposite directions.

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I. INTRODUCTION

Thermionic refrigerators (power generators) have attracted considerable attention recently due to their potential for reliable, environmentally friendly, scalable and high-efficiency operation.1-13

Thermionic emission is a process where electrons are thermally emitted from the surface of materials. The current density $J_e$ of thermionic emission is determined by the well-known Richardson–Dushman (RD) law,14

$$J_e = AT^2 \exp\left(\frac{-\Phi_0}{k_BT}\right),$$

where $T$ is the absolute temperature in Kelvin, $\Phi_0$ is the workfunction of the material in electronvolts, $k_B$ is the Boltzmann constant, $A = 4\pi e m_e k_B^2/h^3 = 120 A cm^{-2}K^{-2}$ is the Richardson constant, $e$ is the electron charge, $m_e$ is the electron mass, and $h$ is the Planck constant. For convenience, we set electron charge $e > 0$, here and hereafter, and ignore the minus sign as if all carriers in our system are holes rather than electrons.

A typical vacuum-state thermionic refrigerator14 (VSTR) consists of two electrodes separated by a small vacuum gap. In a solid-state thermionic refrigerator15-17 (SSTR), the gap between two electrodes is filled with a solid, usually a semiconductor.

The electrodes can be either metals or semiconductors. Many different semiconductors have been investigated for use in SSTR, including AlGaAs,18 SiGe/Si,19,20 Hg$_{1-x}$Cd$_x$Te,21 and InP.22 In a thermionic cooling process, thermally activated electrons with energy greater than the workfunction are emitted from one electrode and travel over the energy barrier to reach the opposite electrode. The net electrical and heat flux across the barrier can be controlled by an applied bias voltage and a temperature difference between two electrodes. Cooling can be achieved when the applied bias exceeds a threshold value.

The performance of a refrigerator is given by the coefficient of performance (COP), defined as

$$\text{COP} = \frac{Q_1}{W},$$

where $Q_1$ is the heat extracted from the low temperature reservoir and $W$ is the work input by an external source.

The main advantage of SSTRs is that the device can have a much lower workfunction. This is because the effective barrier height in a SSTR is the difference between the cathode’s workfunction and the solid’s electron affinity. Some schemes using negative electron affinity to increase the COP have been proposed.24-26

Another disadvantage of VSTR is the space-charge effect which
increases the effective workfunction and reduces the output. In a SSTR, this effect is much less significant as compared to a VSTR since we can design the barrier width (the width of a solid layer) to be much smaller than the vacuum gap in VSTR.\(^1,^4\) On the other hand, there is a significant heat backflow in a SSTR (from the hot electrode to the cold one) due to the finite thermal conduction (lattice thermal conductivity) in the barrier layer which is a layer of semiconductor in our system. The heat backflow reduces the COP.

In recent years, the thermionic emission and corresponding refrigeration (power generators) have been studied in many different ways such as by using two dimensional materials,\(^2,^3,^6,^27\) photon-enhanced thermionic emission,\(^11,^14,^29\) and van der Waals heterostructures.\(^27,^29\)

In a thermionic process, the electrons traverse the barrier ballistically without loss of energy. In a thermoelectric process, electrons traverse the conductor diffusively. We now linearize the electrical current \(J_{ec}\) and thermal current \(J_{qc}\) with regard to that bias gradient \(\nabla V\) and temperature gradient \(\nabla T\) in a thermionic process,

\[
J_{ec} = L_{11}^0 (\nabla V) + L_{12}^0 (-\nabla T),
\]

\[
J_{qc} = L_{21}^0 (\nabla V) + L_{22}^0 (-\nabla T).
\]

By using this linearization, we can make an analogy between the thermionic and the thermoelectric process, that is, we can define corresponding "thermoelectric" quantities in a thermionic process,\(^1,^15\)

\[
\sigma = L_{11}^0, \quad (5)
\]

\[
S = \frac{L_{12}^0}{L_{11}^0}, \quad (6)
\]

\[
\kappa = \frac{L_{22}^0 - L_{12}^0 L_{12}^0}{L_{11}^0}, \quad (7)
\]

where \(\sigma\) is the electrical conductivity, \(S\) is the Seebeck coefficient, and \(\kappa\) is the thermal conductivity. The performance of a thermoelectric or a thermionic device can be characterized by the figure of merit \(ZT\),\(^4,^12\) given as

\[
ZT = \frac{\sigma S^2 T}{\kappa}, \quad (8)
\]

where \(T\) is the absolute temperature. The higher the \(ZT\) is, the better the device performs.

As mentioned above, the charge transport is mainly due to over barrier hopping of hot electrons in a thermionic process. In a classical description, it is usually assumed that the transport is ballistic and the transmission over the barrier is 100%. However, if the electrons are regarded as waves, the reflection at boundary is finite and the transmission will be less than 100%. The purpose of this paper is to analyze the effect of the reflection over the barrier (ROB) on a thermionic cooling process. This pure quantum effect is expected to be small and the fact that RD law works well suggests that ROB may have a negligible effect on the emission process. However, for a thermionic device with two electrodes, the effect of ROB on the charge and heat transport along opposite directions is not the same. Thus, while the ROB suppresses the emission, it can either reduce or enhance the overall performance of the device. Therefore, it is important to have a quantitative understanding of the quantum effect.

In this paper, we show quantitatively how the ROB influences the COP and \(ZT\) of a thermionic device. We focus on the difference between two models: with and without the ROB effect. The result of the model without the ROB effect is already known.\(^15,^16\) The ROB effect is analyzed in this work and the results are as follows. Generally, due to the ROB effect, transport probability is less than the classical value of 100%, and the ROB slightly reduces the electric current and the thermal current. However, due to the inequivalent ROB effect in the two electrodes, we find that the COP and \(ZT\) can be higher due to the ROB effect in some regions of workfunction and thermal resistance. The optimum bias (workfunction) of COP (\(ZT\)) is slightly greater (smaller) due to the ROB effect. The overall optimum \(ZT\) is smaller when the ROB effect is included. These findings may be useful in designing and developing high-efficiency thermionic cooling devices.

II. THERMIonic PROPERTIES OF A SINGLE BARRIER STRUCTURE

A. Electric current and thermal current

We consider a single barrier structure shown in Fig. 1, where the cathode and the anode are separated by a semiconductor of thickness \(d\). Here, for simplicity, we assume the same workfunction for the cathode and the anode. A bias voltage is applied to drive the current from the cold to hot electrode. We ignore the space charge...
effect and band bending so that the electric field between two electrodes can be viewed as uniform. This assumption is proper because when semiconductor thickness is small enough, the space charge effect can be ignored and the band bending can be minimized by adjusting the chemical potential of the semiconductor. We choose the position of the cathode/barrier interface as the origin of coordinates and the cathode chemical potential is the zero energy point as shown in Fig. 1. The potential can be written as

\[ V(z) = \begin{cases} 
0, & (z < 0), \\
\Phi - \frac{e}{2}z, & (0 \leq z \leq d), \\
-e_v, & (z > d),
\end{cases} \]

where \( \Phi \) is the workfunction of cathode, \( v \) is the bias voltage, and \( d \) is the barrier thickness. For an electron incident from the left electrode, the wave function can be written as

\[ \psi(z) = \begin{cases} 
\psi_1(\xi(z)) + R_\psi \psi_2(\xi(z)), & (z < 0), \\
A\psi_1(\xi(z)) + B\psi_2(\xi(z)), & (0 < z < d), \\
T_\psi \psi_1(\xi(z)), & (z > d),
\end{cases} \] (9)

where \( k_1 = \sqrt{2m E_\alpha/h} \) and \( k_2 = \sqrt{2m (E_z + e_v)/h} \) are the wavevectors of the wave function in the regions of \( z < 0 \) and \( z > d \), respectively. \( \psi_1(\xi(z)) \) and \( \psi_2(\xi(z)) \) are given as

\[ \psi_1(\xi(z)) = \xi(z)^3 H_1^{(1)} \left( \frac{2}{3} \xi(z)^2 \right) \]

and

\[ \psi_2(\xi(z)) = \xi(z)^3 H_1^{(2)} \left( \frac{2}{3} \xi(z)^2 \right), \]

where \( H_1^{(1)} \) and \( H_1^{(2)} \) stand for the first kind and the second kind of Hankel function, respectively, and \( \xi(z) = \left[ 2mev/\hbar^2 d \right]^{1/2} \left[ z + (E_z - \Phi)d/ev \right] \) is a dimensionless parameter.

From the boundary conditions that \( \psi \) and its derivative being continuous at \( z = 0 \) and \( z = d \), we find

\[ R_\psi = \frac{1 + \frac{1}{k_1} \psi_1(\xi(0)) + \psi_2(\xi(0))}{1 - \frac{1}{k_1} \psi_1(\xi(0)) + \psi_2(\xi(0))}, \] (10)

where

\[ \eta_\psi = \frac{\psi_2(\xi(d)) - ik_2\psi_1(\xi(d))}{\psi_1(\xi(d)) - ik_2\psi_2(\xi(d))}. \]

In above equations, derivatives are with respect to \( z \), not \( \xi(z) \).

The reflection coefficient is given as \( r_\psi = |R_\psi|^2 \). The electric and the thermal currents is given as \( J_\alpha \) and \( J_{Q\alpha} \), where \( \alpha = l(r) \) for the current from left(right) electrode the right(left) electrode,

\[ J_\alpha = \frac{e}{2\pi^2} \int_0^\infty dE_z \int_0^\infty dE_z [1 - r(E_z)] f_{FD}^\alpha \] (11)

and

\[ J_{Q\alpha} = \frac{m}{2\pi^2} \int_0^\infty dE_z \int_0^\infty dE_z E [1 - r(E_z)] f_{FD}^\alpha, \] (12)

where \( f_{FD}^\alpha = f_{FD}(E, \mu_\alpha, T_a) \) is the Fermi Dirac distribution function for the electrons in electrode \( \alpha = (l, r) \), \( E = E_z + E_c \) is the energy for the momentum in the \( z \)-direction necessary to surmount the barrier, \( E_c \) is the energy for the momentum in the plane perpendicular to \( z \)-direction, and \( \mu_\alpha \) is the chemical potentials as shown in Fig. 1.

The total electric current \( J \) and the total thermal current \( J_0 \) are given by

\[ J = J_l - J_r, \] (13)

and

\[ J_0 = J_{Ql} - J_{Qr} - \frac{T_r - T_l}{R_\psi}. \] (14)

The last term in Eq. (9) denotes the heat flow from hot(right) side to cold(left) side, and \( R_\psi \) is the lattice thermal resistance.

### B. Comparison between models with and without the ROB effect

From Eq. (1), the COP with the ROB effect is given as

\[ \text{COP}_1 = \frac{J_0}{v_f}, \] (15)

where \( Q_1 = J_0 \) is the extracted heat per unit area from the cold electrode, \( v \) is the external bias, \( W = v_f \) is the work done per unit area by the external source, \( J \) is the charge current density, and \( J_0 \) are given by Eqs. (8) and (9), respectively.

We now focus on the ROB effect. The result without the ROB effect can be obtained by letting \( r_\psi = 0 \) in Eqs. (6) and (7). We denote \( J_{Ml} = J_\alpha(r_\psi = 0) \), \( J_{M0} = J_\alpha(r_\psi = 0) \), as the electrical and thermal currents in the absence of ROB effect. The corresponding COP is

\[ \text{COP}_0 = \frac{J_{0M}}{v_f M}, \] (16)

where \( J_{0M} = J_{QM} - J_{Q0} - (T_r - T_l)/R_\psi \), and \( J_{0M} = J_{Ml} - J_{M0} \).

Figure 2 shows the bias dependent COP0 and COP1 under certain parameters. Figure 2(a) shows that there exists a critical value of bias when bias is smaller (larger) than the critical value \( \text{COP}_0 > \text{COP}_1 > \text{COP}_0 \). The critical bias is absent in Fig. 2(b). The result indicates that (i) the ROB effect can increase the COP in some cases and (ii) there exist a minimum value of \( R_\psi \) for \( \text{COP}_1 > \text{COP}_0 \) to be possible. The first result is due to the fact that the transmission asymmetry increases with the bias.
This leads to a larger net thermal and electric current from the cold to the hot electrodes. As a result, the COP1 > COP0 at higher bias. The second result on the Rl dependence can be further analyzed. By letting COP1 > COP0, we obtain

\[ R_l > R_{l_{\text{min}}} = \frac{J_M - J}{J_{Q} - J_{Q,M1}} \delta T, \]  

(17)

where \( \delta T = T_c - T_h \) and \( J_{Q,M1} = J_{Q_{01}} - J_{Q_{00}} \). The larger the \( R_{l_{\text{min}}} \) the more difficult to achieve COP1 > COP0.

Figure 3 plots \( R_{l_{\text{min}}} \) against bias voltage \( v \). As bias voltage increases, \( R_{l_{\text{min}}} \) decreases. The origin of the \( R_l \) dependence is that the ROB effect makes \( J \) and \( J_{Q} \) smaller than \( J_M \) and \( J_{Q,M1} \). The COP reduction due to the thermal resistance is different between the model without the ROB effect and the model with the ROB effect. For a certain value of \( R_l \), say, \( 5 \times 10^{-7} \) W m\(^{-2}\) K as used in plotting Fig. 2(a), the corresponding critical bias is around 0.033 V, which is slightly greater than the optimum bias of COP0 (around 0.03 V). For bias greater than 0.033 V, COP1 > COP0. If we can further increase \( R_l \) to make the critical value of bias less than 0.03 V and keep other parameters unchanged, COP1 > COP0 can be achieved even for optimum COP0.

\( R_{l_{\text{min}}} \) has a quite different bias dependence under large bias as shown in Fig. 3(b). This is because under a high bias, the thermonic emission is mainly from the carriers with energy close to the workfunction. The number of carriers with higher energy decreases rapidly and their contribution is negligible. Under a high bias, the structure is more asymmetric and the reflection increases, which results in a smaller electrical and thermal current. Furthermore, the \( R_{l_{\text{min}}} \) exhibits a profound quantum oscillation under high bias. Under high bias, the net current is dominated by the current emitted from the left electrode. The reflection coefficient is an oscillatory function of the bias which is manifested in \( R_{l_{\text{min}}} \).

Under a low bias, this quantum oscillation is suppressed by the current emitted from the opposite electrodes.

### C. Figure of merit

The ZT of a thermionic device can be analyzed by making an analogy with a thermoelectric device. For small bias voltage and small temperature difference \( \delta T = T_c - T_h \), where \( T_c = T_i \) and \( T_h = T_j \), and \( J_{Q} \) can be expressed as

\[ J = \mathcal{L}_{11} v + \mathcal{L}_{12} \left( \frac{\delta T}{T} \right) \]

and

\[ J_{Q} = \mathcal{L}_{21} v + \left( \mathcal{L}_{22} + \frac{T}{R_l} \right) \left( \frac{\delta T}{T} \right), \]

where the coefficients \( \mathcal{L}_{ij} \) are given as

\[ \mathcal{L}_{ij} = \frac{e^{i\delta + i\delta}}{2\pi^2 k_B^2} g \int_0^\infty dE \int_\phi^\infty dE_c e^{\frac{E_c}{k_B T} + \frac{E}{k_B T}} (1 - g). \]

(18)

Here,

\[ T = \frac{T_c + T_h}{2}, \]

\[ k_B = \sqrt{\frac{2m(E_c - \Phi)}{R^2}}, \]

\[ c_1 = 2\left( \frac{k_B^2}{k_1} - k_1 \right) \sin (k_B d), \]

\[ c_2 = -2\left( \frac{k_B^2}{k_1} + k_1 \right) \sin (k_B d) - 4i k_B \cos (k_B d), \]

\[ g = \frac{|c_1|^2}{|c_2|^2}. \]
The corresponding thermoelectric quantities are found as \( \sigma = \mathcal{L}_{11} \), \( S = \frac{\mathcal{L}_{12}}{\mathcal{L}_{11} T} \), and \( \kappa = \left[ \frac{\mathcal{L}_{22} \mathcal{L}_{11} - (\mathcal{L}_{12})^2}{\mathcal{L}_{11} T} + \frac{1}{R} \right] d \) \[(19)\] \[(20)\] \[(21)\]

and

\[(ZT) = \frac{\sigma S^2 T}{\kappa} = \frac{(\mathcal{L}_{12})^2}{\mathcal{L}_{11} \mathcal{L}_{22} - (\mathcal{L}_{12})^2 + eT\mathcal{L}_{11}/R} \] \[(22)\]

We can make a comparison between results with and without the ROB effect. The relevant quantities \( \{\sigma_M, S_M, \kappa_M, (ZT)_b\} \) in the absence of ROB have the same form as those in Eqs. (19)–(22) and the coefficients of linear response \( \mathcal{L}^M_{ij} \) is given as

\[ \mathcal{L}^M_{ij} = \frac{e^2}{2\pi^2 \hbar} \int_0^\infty dE \int_0^E dE_z \frac{E^{i(i+j-2)}_z}{k_B T} \] \[(23)\]

Because the ROB correction factor \( 1 - g \) is limited in the range of \( 0 \leq 1 - g \leq 1 \), \( \mathcal{L}^M_{ij} \) is always greater than \( \mathcal{L}_{ij} \), i.e., \( \mathcal{L}^M_{ij} > \mathcal{L}_{ij} \). By varying the bias voltage, we find the maximum COP, given as

\[ \text{COP}_{\text{max}} = \frac{ZT}{(1 + 1/ZT)} \] \[(24)\]

The ROB effect on \( \text{COP}_{\text{max}} \) is contained in the ROB-dependent \( ZT \).

### III. RESULTS AND DISCUSSION

We have numerically calculated the \( ZT \) for various temperatures and thermal resistance values. The ROB effect is dependent on the barrier width \( d \). To minimize both the tunneling and scattering, \( d \) is longer than the tunneling length and shorter than the electron mean free path (MFP). The tunneling length is around 5–10 nm for most semiconductors and the MFP is in the order of 100 nm. We used \( d = 50 \text{ nm} \) in our calculations. Figure 4 depicts \( (ZT)_b \) and \( (ZT)_t \) for several sets of parameters. We make the following observations. (i) There is a critical workfunction, below this critical workfunction, \((ZT)_b > (ZT)_t\) as shown in the insets of Fig. 4. In this situation, the COP1 can be optimized to be greater than COP0, (ii) The optimum \( \Phi \) is lower in the presence of ROB. This implies that the ROB effect can reduce the required workfunction where the maximum \( ZT \) occurs. (iii) The optimum \( (ZT)_t \) is always smaller than optimum \( (ZT)_b \). For example, the optimum \( (ZT)_t \) is about 6.5 for parameters with \( R_t = 5 \times 10^{-7} \text{ W}^{-1} \text{ m}^{-2} \text{ K} \) and \( T = 500 \text{ K} \) as shown in Fig. 4(a) with orange color, while the optimum \( (ZT)_b \) is about 7.1 for same parameters, leading to a reduction of about 8%. If we optimize workfunction and bias at same time, the ROB effect will reduce the overall optimum COP.

The condition for \((ZT)_t > (ZT)_b\) is given as

\[ \frac{(\mathcal{L}_{12})^2}{\mathcal{L}_{11} \mathcal{L}_{22} - (\mathcal{L}_{12})^2 + eT\mathcal{L}_{11}/R_t} > \frac{(\mathcal{L}_{12})^2}{\mathcal{L}_{11} \mathcal{L}_{22} - (\mathcal{L}_{12})^2 + eT\mathcal{L}_{11}/R_t} \]

We denote \( R_{\text{min}}^\phi \) as the minimum thermal resistance which satisfies the condition

\[ \frac{eT}{R_{\text{min}}^\phi} \left[ \frac{\mathcal{L}_{11} (\mathcal{L}_{12})^2 - \mathcal{L}_{11} (\mathcal{L}_{12})^2}{\mathcal{L}_{11} \mathcal{L}_{22} - (\mathcal{L}_{12})^2} \right] = \left( \frac{\mathcal{L}_{12}}{\mathcal{L}_{11} \mathcal{L}_{22} - (\mathcal{L}_{12})^2} \right) \]

In Fig. 5(a), we show how \( R_{\text{min}}^\phi \) varies with \( \Phi \) at \( T = 300 \text{ K} \). As \( \Phi \) increases, \( R_t \) increases exponentially and rapidly approaches a very large value. If we choose a typical value of lattice thermal conductivity as \( 1 \text{ W m}^{-1} \text{ K}^{-1} \), then \( R_t = 5 \times 10^{-8} \text{ W}^{-1} \text{ m}^{-2} \text{ K} \). The corresponding \( \Phi \) is around 0.01 V, which is quite small. If we can increase \( R_t \), we can relax the restriction of \( \Phi \). But if the workfunction is too high, \( R_{\text{min}}^\phi \) becomes negative as shown in Fig. 5(b). Therefore, it is impossible for \((ZT)_t > (ZT)_b\) when workfunction is too high. This is shown in Fig. 4, where \((ZT)_b > (ZT)_t\) under a large workfunction \( \Phi \).

In conclusion, the ROB effect in a single barrier thermionic cooling device has been investigated. It is found that the COP in the presence of ROB can be higher than that in the absence of ROB, COP1 > COP0, if the bias voltage \( v \) and the lattice thermal resistance \( R_t \) satisfy relation Eq. (12). \( ZT \) is higher in the presence of the ROB effect, \((ZT)_t > (ZT)_b\), if the workfunction \( \Phi \) and the lattice thermal resistance \( R_t \) satisfy relation Eq. (20). The optimum \( (ZT)_t \) is smaller than optimum \( (ZT)_b \). These results can be useful in designing thermionic devices.
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DATA AVAILABILITY

The data that support the findings of this study are available within the article.

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