# Thermopower anomaly in multiple barrier structures

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(Received 2 October 2003; published 5 December 2003)

We have predicted that in suitable multiple barrier structures without applied bias, the thermopower may change sign because of quantum transport process. We have performed detailed numerical calculation to show that the predicted thermopower anomaly can be experimentally observed. This thermopower anomaly does not violate thermodynamic laws.

DOI: 10.1103/PhysRevB.68.233302 PACS number(s): 73.40.Gk, 05.60.Gg, 72.20.Pa, 73.63.Hs

When a temperature gradient along a direction, assuming the $x$ axis, $dT(x)/dx$ is maintained in a metal without applied external electric field, a thermoelectric field

$$E=Q\frac{dT(x)}{dx} \tag{1}$$

is generated in the metal, where $Q$ is known as the thermopower. In semiclassical theory where electrons move diffusively, the thermoelectric field directs opposite to the temperature gradient, and so $Q$ is always negative. We will prove in this paper that for quantum transport process, $Q$ can be positive without violating the thermodynamic law. A sign change of $Q$ will induce a reverse of the direction of the thermoelectric field $E$. Consequently, the thermoelectric current density $j$ will also change sign. Since it is more convenient to calculate $j$, in this paper we will demonstrate the thermopower anomaly with the reversal of thermoelectric current. We will perform a numerical study to show that this thermopower anomaly is measurable. Its influence on thermoelectric properties will be discussed.

To demonstrate the physical picture with a simple one-dimensional electron transport, we consider a sample of finite width between 0 and $d$, attaching to two conducting leads at $x=0$ and $x=d$. In the $yz$ plane the potential is constant which we set to zero, and $k_\parallel$ labels the electron wave vector for its motion in the $yz$ plane. Along the $x$ axis the potential $V(x)$ is zero in the two conducting leads $x<0$ and $x>d$, but can be any function of finite value within the sample region $0<x<d$. This potential is schematically illustrated in Fig. 1(a). After we explain the fundamental physics, we will perform a detailed numerical investigation for a double barrier system and a multiple barrier system, the potential profiles of which are plotted in Figs. 1(b) and 1(c), respectively.

Let the conducting lead at the left side ($x<0$) be hot at the temperature $T_h$ with the corresponding chemical potential $\mu_h$, and the conducting lead at the right side ($x>d$) be cold at the temperature $T_c$ with the corresponding chemical potential $\mu_c$. Consider an electron coming in from the left lead with the kinetic energy $\epsilon_\perp$ along the $x$ axis and the parallel wave vector $k_\parallel$ in the $yz$ plane. Let $T(\epsilon_\perp)$ be the transmission probability through the sample, and $f_h(k_\parallel\epsilon_\perp,T_h)$ [or $f_c(k_\parallel\epsilon_\perp,T_c)$] the electron distribution function in the right [or left] lead. Then the thermoelectric current density flowing through the sample from the hot lead to the cold lead is given by

$$j=\frac{e}{\pi\hbar}\sum_{k_\parallel}\int_0^\infty d\epsilon_\perp T(\epsilon_\perp)[f_h(k_\parallel\epsilon_\perp,T_h)-f_c(k_\parallel\epsilon_\perp,T_c)]. \tag{2}$$

Let us assume that in a specific sample the energy dependent transmission probability $T(\epsilon_\perp)$ is zero except for a small energy region $\varepsilon-\Gamma<\epsilon_\perp<\varepsilon+\Gamma$ in which $T(\epsilon_\perp)\simeq 1$. In this case the current density simplifies to

$$\frac{j}{2e\Gamma}\propto T_h\ln(1+e^{(\mu_h-\varepsilon)/kT_h})-T_c\ln(1+e^{(\mu_c-\varepsilon)/kT_c}). \tag{3}$$

The chemical potentials in the leads have the standard temperature-dependent expressions

$$\mu_l=\epsilon_{f,l}\left[1-\frac{1}{3}\left(\frac{\pi k_B T_l}{2\epsilon_{f,l}}\right)^2\right] \tag{4}$$

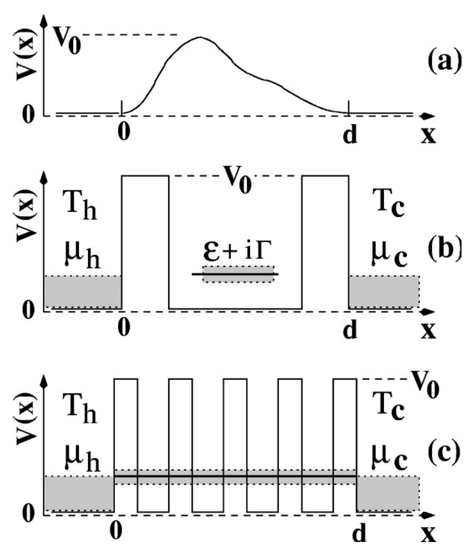


FIG. 1. The potential profile in a sample connected to two conducting leads at $x=0$ and $x=d$; (a) for a general potential, (b) for a double barrier structure, and (c) for a multiple barrier structure.

 

with $l=h$ or $c$, where $\epsilon_{f,h}$ and $\epsilon_{f,c}$ are the corresponding Fermi energies. It is clear that $\mu_c>\mu_h$ since $T_h$ is larger than $T_c$.

We will consider the case that the energy window around $\varepsilon$ for $T(\epsilon_\perp)\simeq 1$ is so located that both conditions $|\mu_h-\varepsilon| \gg k_BT_h$ and $|\mu_c-\varepsilon|\gg k_BT_c$ are satisfied. Then, if $\mu_h<\varepsilon$ and $\mu_c<\varepsilon$, Eq. (3) reduces to

$$\frac{j}{2e\Gamma}\propto T_h e^{(\mu_h-\varepsilon)/k_BT_h}-T_c e^{(\mu_c-\varepsilon_0)/k_BT_c}>0, \tag{5}$$

which is the normal thermoelectric current direction. On the other hand, if $\mu_h>\varepsilon$ and $\mu_c>\varepsilon$, from Eq. (3) we have

$$\frac{j}{2e\Gamma}\propto \mu_h-\mu_c<0, \tag{6}$$

which exhibits a thermopower anomaly, because the reverse of the thermoelectric current direction implies the sign change of the thermopower $Q$.

We have performed a numerical analysis for this simplified energy dependent transmission probability $T(\epsilon_\perp)$, and the result indicates that the thermopower anomaly may occur if $\mu_c>\varepsilon$, $\mu_h>\varepsilon$, and $\Gamma$ is small. We will not go into the details here since later we will present the complete numerical results for both the double barrier and the multiple barrier structure shown in Figs. 1(b) and 1(c) which can be measured. Nevertheless, we must point out that for a sample with a potential profile as shown in Fig. 1(a), the thermopower anomaly does not exist. For such a potential it is clear that $T(\epsilon_\perp)\simeq 0$ for $\epsilon_\perp<V_0$ and $T(\epsilon_\perp)\simeq 1$ for $\epsilon_\perp>V_0$. Since $\mu_c \leq \epsilon_{f,c}$ and $\mu_h\leq\epsilon_{f,h}$ as indicated by Eq. (4), the conditions $\mu_c>\varepsilon$ and $\mu_h>\varepsilon$ imply that $\epsilon_{f,c}>V_0$ and $\epsilon_{f,h}>V_0$. Therefore, in the absence of a temperature gradient, the Fermi energy is already above $V_0$, and the system behaves as a simple metal. It is well known that a simple metal has normal thermoelectric properties.

An energy window with $T(\epsilon_\perp)\simeq 1$ can be realized in systems shown in Figs. 1(b) and 1(c). We will first perform a simpler calculation for the double barrier resonant tunneling structures, which have been extensively studied in connection to modern electronic devices.[1] To present clearly the physical process, we assume the sample has only one resonant level with energy $\varepsilon$ and half-width $\Gamma$. Then, $T(\epsilon_\perp)$ is zero except for a small energy region $\varepsilon-\Gamma<\epsilon_\perp<\varepsilon+\Gamma$ in which $T(\epsilon_\perp)\simeq 1$. The values of $\varepsilon$ and $\Gamma$ can be tuned by adjusting the well width, the barrier widths, and the barrier height $V_0$. The Fermi energies $\epsilon_{f,h}$ and $\epsilon_{f,c}$ can also be tailor made with different impurity concentration. This is the ideal system to exhibit thermopower anomaly.

A double barrier structure is specified with many structure parameters and materials parameters. To present our numerical results, we will reduce the number of parameters in such a way that the essential physics of thermopower anomaly can be clearly demonstrated. On the other hand, we have to make sure that the sample can be fabricated and the predicted thermopower anomaly can be measured. We will consider a symmetric GaAs/Al$_x$Ga$_{1-x}$As double barrier structure with 50 Å well width and the same barrier width 55 Å for both barriers.

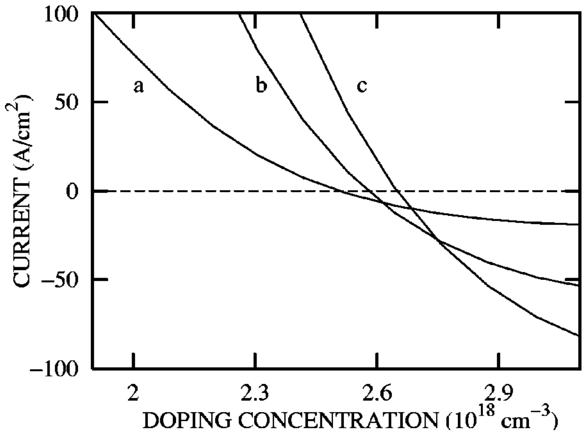


FIG. 2. Thermoelectric current density $j$ in a double barrier structure with varying doping concentration $N_d$ for $T_h=85$ K (curve $a$), 100 K (curve $b$), and 115 K (curve $c$).

We set $x=0.27$ for which the barrier height, as indicated in Fig. 1(b), is $V_0=230$ meV. There is only one resonant level in this structure, and the transmission probability $T(\epsilon_\perp)$ is obtained numerically for the range of $\epsilon_\perp$ from zero to high above the potential barrier. This will guarantee an accurate current calculated from Eq. (2).

The two leads at the left and the right are equally doped with donors concentration $N_d$ cm$^{-3}$, and so the two Fermi energies $\epsilon_{f,h}$ and $\epsilon_{f,c}$ have the same value $\epsilon_f$ which can be easily calculated for given value of $N_d$. We will keep $T_c$ at the liquid nitrogen temperature $T_c=77$ K and vary the temperature $T_h$. Then, we have only two free parameters $N_d$ and $T_h$. In this way not only the numerical results can be presented clearly, but also the corresponding experiments can be carried out.

We will use the well established effective mass approximation, and let $m^*$ be the electron effective mass in GaAs. The total electron energy $\epsilon_{tot}=\epsilon_\perp+\epsilon_\parallel$ in the leads consists of two parts $\epsilon_\parallel=\hbar^2k_\parallel^2/2m^*$ and $\epsilon_\perp=\hbar^2k_\perp^2/2m^*$, where $k_\perp$ is the wave-vector component along the sample growth direction $x$. The current given by Eq. (2) can then be expressed as

$$j=\frac{em^*k_B}{2\pi^2\hbar^3}\int_0^\infty d\epsilon_\perp T(\epsilon_\perp)[T_h\ln(1+e^{(\mu_h-\epsilon_\perp)/kT_h}) - T_c\ln(1+e^{(\mu_c-\epsilon_\perp)/kT_c})]. \tag{7}$$

In our definition, the above current is due to electrons flowing through the double barrier structure. In a real experiment the measured current flows through an external circuit. When we present our numerical results, we have used the convention that a positive $j$ means normal thermopower. When the thermoelectric current reverses its direction, the thermopower changes sign. Hence, in our convention a negative $j$ indicates the thermopower anomaly.

The calculated current density as a function of the doping concentration $N_d$ is plotted in Fig. 2 for $T_h=85$ K (curve $a$), 100 K (curve $b$), and 115 K (curve $c$). The changing sign of $j$ with increasing $N_d$ marks the appearance of thermopower

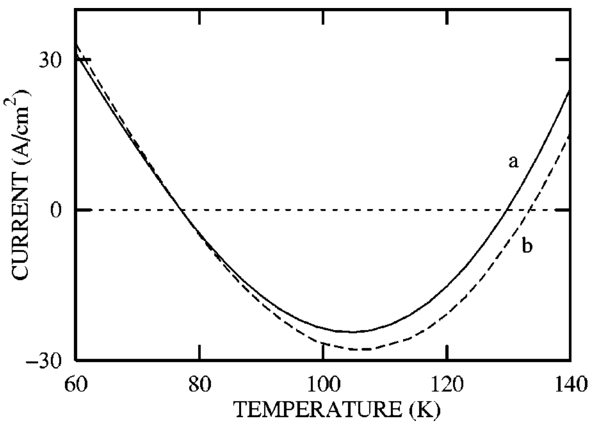


FIG. 3. Thermoelectric current density $j$ in a double barrier structure (solid curve) as a function of $T_h$ for the impurity concentration $2.72\times10^{18}$ cm$^{-3}$. Note that in the temperature region less than 77 K, $T_h<T_c$. The dashed curve is for four barrier structures and eight barrier structures.

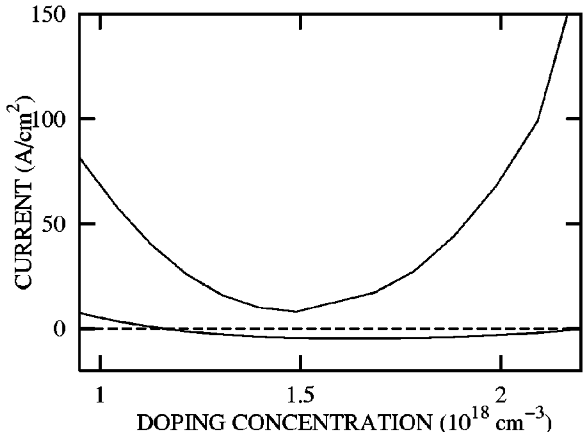


FIG. 4. The behavior of thermoelectric current in a double barrier structure with a wider well which contains two resonant levels. The lower curve is for $T_h=85$ K, and the higher curve for $T_h=135$ K.

anomaly. When this occurs, the anomalous thermoelectric current is sufficiently large to be measured without any difficulty.

Once a sample is made, the impurity concentration $N_d$ cannot be changed. It will be more convenient to detect thermopower anomaly in a given sample by changing the temperature $T_h$. From Fig. 2 we see that such an experiment can be carried out for $N_d=2.72\times10^{18}$ cm$^{-3}$. For this sample structure, the calculated current density is shown by the solid curve in Fig. 3 as a function of $T_h$ in the range from $T_h<T_c$=77 K to $T_h>T_c$. At $T_h=T_c$ the thermoelectric current vanishes, as expected in a symmetric sample. As soon as $T_h$ is raised to above $T_c$, the thermopower anomaly occurs because $j$ becomes negative. When $T_h$ becomes lower than $T_c$, the temperature gradient in Eq. (1) changes sign. In this case, the positive thermoelectric current in Fig. 3 in the temperature region $T_h<T_c=77$ K also marks the thermopower anomaly. This is the correct phenomenon because our sample is symmetric.

It is important to emphasize that the thermopower anomaly occurs with a very small temperature difference $|T_h-T_c|$, provided the sample is properly designed. Experimentally, $|T_h-T_c|$ as small as 100 mK can be well controlled.[2,3] It is well known that in heterostructures the interfaces scattering of phonons reduces the thermal conductivity by about one order of magnitude, and the effect is enhanced with increasing number of interfaces.[4] A multiple barrier structure, as shown in Fig. 1(c), will not only effectively reduce the thermoconductivity, but also decrease the temperature gradient. These are important factors for performing experiments. We have expanded the double barrier structure in Fig. 1(b) to a four-barrier structure and a eight-barrier structure. The resonant level develops into a very narrow miniband of bandwidth about $2\Gamma$. We then performed similar calculations. The results are plotted in Fig. 3 as two indistinguishable dashed curves, because the miniband is already fully developed when the number of barriers increases to four.

To be able to observe the thermopower anomaly, one should avoid to have more than one resonant level or miniband in the sample. Let us illustrate the reason using the double barrier structure as an example. If a second resonant level exists at energy $\varepsilon^*$, this energy will be so high that in the energy range around $\varepsilon^*$ we have $f_h(k_\parallel\epsilon_\perp,T_h)>f_c(k_\parallel\epsilon_\perp,T_c)$. Within this window around $\varepsilon^*$ with large transmission probability, a net electron flow through the sample will be directed from the hot lead to the cold lead as a normal thermoelectric current. The thermopower anomaly will then be suppressed. If we increase the well width of our sample from 50 Å to 85 Å but keep the same barrier structure, a second resonant level will appear in the well. The calculated thermoelectric current density is shown in Fig. 4, where the thermopower becomes entirely normal.

In our above theoretical analysis and numerical calculation, we have not included the temperature effect on the total amount of charge accumulation in the well. Since the difference $\mu_c-\mu_h$ is due to the thermal effect and is small, this temperature effect in the well should have negligible influence on our final numerical results. We have used a self-consistent approach[5] to check this point, and found it indeed correct.

We should mention that the thermopower anomaly does not violate the thermodynamic law with respect to the heat current. The heat current in the system is always directed from the hot to the cold lead. It can be calculated from Eq. (2) with the electron charge $e$ replaced by $\epsilon_{tot}-\mu_{h,c}$, where $\epsilon_{tot}=\epsilon_\perp+\epsilon_\parallel$ is the total electron energy. When the electric current changes sign, both the factor $f_h(\epsilon_{tot},T_h)-f_c(\epsilon_{tot},T_c)$ and the factor $\epsilon_{tot}-\mu_{h,c}$ change sign, which implies the correct phenomenon that the heat current always flows from the hot to the cold lead. It is easy to show that the energy flow is connected to the Joule heating power[6] $jV_{th}$, where $V_{th}$ is the thermoelectric voltage. All these have been confirmed in our numerical calculations.

We have predicted theoretically an observable thermopower anomaly in multiple barrier structures without externally applied bias. Thermoelectric current reversal was discussed very recently for a one-dimensional system without transverse degrees of freedom.[6] Multiple barrier structures have continuum of states in the transverse plane, and are widely used device structures controlled by the applied bias. Since the bias changes the difference $\mu_c - \mu_h$ of chemical potentials just as the effect of temperature, it is interesting to investigate the combined influence of the temperature and the bias on thermopower anomaly, although the analysis and the numerical calculation will be more complicated. We must point out a major difference in sample structure between our problem and the conventional double barrier resonant tunneling diodes. In our case the resonant level $\varepsilon$ should lie below the Fermi energy $\epsilon_f$. In the conventional diode which operates in the region of negative differential resistance, the resonant level $\varepsilon$ is much higher than the Fermi energy $\epsilon_f$. Consequently, our predicted thermopower anomaly can hardly be detected in conventional double barrier resonant tunneling diodes.

Although semiconductor superlattices have been much studied as a potential material for thermoelectric technology, thermopower anomaly has not been observed in such systems. There are two reasons. First, the width of the energy window for $T(\epsilon_\perp) \simeq 1$ is the subband width, which is too wide to yield the thermopower anomaly. Second, the relevant issue in multilayer structures is focused on thermionic process,[7,8] which is carrier transport over the barrier.

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