Existence of negative differential thermal conductance in one-dimensional diffusive thermal transport

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(Received 14 January 2012; revised manuscript received 30 December 2012; published 3 June 2013)

We show that in a finite one-dimensional (1D) system with diffusive thermal transport described by the Fourier's law, negative differential thermal conductance (NDTC) cannot occur when the temperature at one end is fixed and there are no abrupt junctions. We demonstrate that NDTC in this case requires the presence of junction(s) with temperature-dependent thermal contact resistance (TCR). We derive a necessary and sufficient condition for the existence of NDTC in terms of the properties of the TCR for systems with a single junction. We show that under certain circumstances we even could have infinite (negative or positive) differential thermal conductance in the presence of the TCR. Our predictions provide theoretical basis for constructing NDTC-based devices, such as thermal amplifiers, oscillators, and logic devices.

DOI: 10.1103/PhysRevE.87.062104 PACS number(s): 44.10.+i, 44.05.+e, 05.60.−k

Introduction. In recent years, nonlinear thermal transport, particularly in low-dimensional systems, is of significant interest from both fundamental and practical perspectives [1,2]. For example, thermal rectification has been theoretically and experimentally studied in many nanostructures [3–11] and heterogeneous bulk materials [12–14]. Negative differential thermal conductance (NDTC), an unusual thermal transport phenomenon where the heat current across a thermal conductor decreases when the temperature bias increases, is an essential element for the construction of thermal transistors [15] and thermal logic [16] and is shown to exist in many nonlinear one-dimensional (1D) systems [9,11,15,17–24] and vacuum gaps [25]. Many mechanisms such as nonlinear interactions [26], molecular anharmonicity [11,21,22,27], interplay between the thermal driving force and the thermal (boundary) conductance [17,18,20,23], thermal interfaces [15,17,19], and others [25] have been proposed to explain the existence of NDTC. Interestingly, several numerical studies [18–20,23] have suggested that NDTC may vanish as the system length becomes large (approaching diffusive thermal transport). However, it has not been definitely answered whether NDTC universally vanishes for diffusive thermal transport. Besides, the role played by thermal interfaces in NDTC has not been well studied. Here, we provide a generic and analytic study of these issues in 1D diffusive thermal transport described by the Fourier's law. We prove that NDTC cannot exist when the temperature at one end is fixed and there are no abrupt junctions. However, we show that NDTC in this case is still possible if a junction with temperature-dependent thermal contact resistance (TCR) is introduced. Unlike previous theories and simulations [9,11,15,17–24] that dealt with specific toy models that are often difficult to access experimentally, our predictions provide a generic way toward building NDTC-based devices.

We consider a general 1D system in the diffusive thermal transport regime whose thermal conductivity $\kappa(x,T)$ is a function of the coordinate x and the local temperature $T(x)$. The position dependence of the thermal conductivity $\kappa(x,T)$ is explicitly expressed, since the system we consider can have a spatial dependence of structure or composition (e.g., strain or mass gradient). This phenomenological description is valid as long as the mean free path (MFP) of heat carriers is much smaller than the size of the system, where the microscopic details are unimportant. This approach generates analytic results regarding the existence of NDTC, and it is instructive in system design to pursue the applications of NDTC.

For a finite 1D system that lies in the coordinate range $[x^L,x^R]$ (Fig. 1), the local heat current $q(x)$ can be calculated from the Fourier's law:

$$q(x) = -\kappa(x,T)\frac{dT}{dx}. \tag{1}$$

For thermal transport without heat sources or sinks, the heat current is conserved and the steady-state thermal transport equation reads

$$\frac{d}{dx}\left[\kappa(x,T)\frac{dT}{dx}\right] = 0. \tag{2}$$

Once the temperature at two ends of the system are given [28], i.e.,

$$T(x^{L(R)}) = T^{L(R)}, \tag{3}$$

the temperature profile $T(x)$ is uniquely [29] determined by Eq. (2) and the boundary conditions Eq. (3), and the resulting heat current q (independent of x) flowing in the system can be computed from Eq. (1).

By applying an infinitesimal variation $\delta T^{L(R)}$ of the boundary temperature at one end, i.e., $T^{L(R)}$ is varied to $T^{L(R)} + \delta T^{L(R)}$, while the temperature $T^{R(L)}$ at the other end is fixed, the resulting temperature profile is varied to $T(x) + \delta T(x)$. This temperature profile variation $\delta T(x)$ can induce a variation δq of the heat current. We define the differential thermal conductance (DTC) as

$$G \equiv \frac{\delta q}{\delta(T^L - T^R)}, \tag{4}$$

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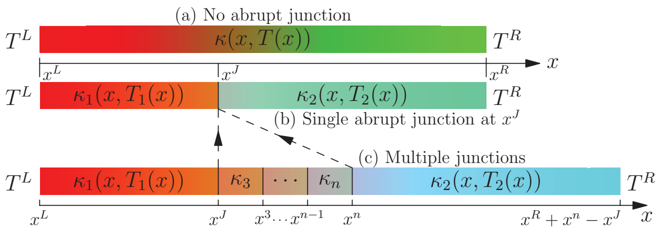

FIG. 1. (Color online) Schematics of 1D systems without any junctions (a) and with a single junction (b) and multiple junctions (c). The junctions are indicated by vertical black lines.

or specifically

$$G = G^L = \frac{\delta q}{\delta T^L}, \qquad \text{when } T^R \text{ is fixed; or}$$
$$G = G^R = -\frac{\delta q}{\delta T^R}, \qquad \text{when } T^L \text{ is fixed.} \tag{5}$$

In the following, we consider the cases with and without the junctions to discuss the existence of NDTC.

Systems without abrupt junctions. As shown in Fig. 1(a), if $T^{R(L)}$ is fixed (without loss of generality, we can assume $T^L > T^R$), we will show that there is no NDTC and the heat current q will increase as the temperature bias increases by increasing (lowering) $T^{L(R)}$ (NDTC could still exist when the temperatures at two ends vary simultaneously [30], however, we limit our study in the cases that the temperature at one end is fixed).

Qualitatively, we first point out that the nonexistence of NDTC here is a direct consequence of the uniqueness of the solution of Eq. (2), as we graphically demonstrate in Fig. 2(a). Take the case that T^R is fixed as an example. If the temperature T^L is increased to T'^L ($> T^L$), the temperature profile $T(x)$ (black line) with $T(x^{L(R)}) = T^{L(R)}$ and heat current q are changed to $T'(x)$ (red line) with $T'(x^L) = T'^L$ and $T'(x^R) = T^R$ and heat current q'. First of all, we must have $q' \neq q$. Otherwise, the first-order differential equation $\frac{dT}{dx} = -\frac{q}{\kappa(x,T)}$ about T with initial condition $T(x^R) = T^R$ would have nonunique solutions [$T(x)$ and $T'(x)$], which is not allowed. Second, q' cannot be smaller than q [proportional to the slope of $T(x)$ at x^R], because otherwise there will be an intersection of $T'(x)$ (represented by the dashed line) and $T(x)$ at some $x^I < x^R$. We then must have $T'(x) = T(x)$ in the coordinate range $[x^I, x^R]$ due to the uniqueness of the solution to Eq. (2), and thus $q' = q$ (contradiction). Therefore, we must have $q' > q$; i.e., the heat current monotonically increases with temperature T^L when T^R is fixed and there is no NDTC. Similar arguments apply to the case when T^L is fixed.

We have derived the analytical expressions for the DTCs as (Appendix 1)

$$G^L = J^{-1}, \quad G^R = F(x^R)J^{-1}, \tag{6}$$

where

$$F(x) \equiv \exp\left\{\int_{x^L}^{x} \frac{1}{\kappa(x', T(x'))} \frac{\partial \kappa}{\partial T} \frac{dT}{dx'} dx'\right\},$$
$$J \equiv \int_{x^L}^{x^R} \frac{F(x')}{\kappa(x', T(x'))} dx'. \tag{7}$$

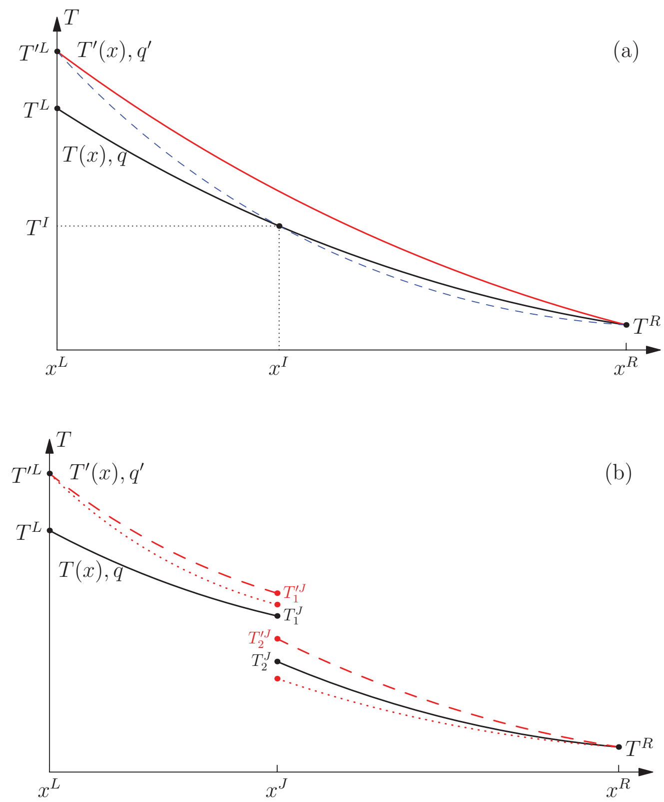

FIG. 2. (Color online) A schematic example of temperature profiles of systems (a) without any junctions and (b) with a single junction at x^J when the temperature at x^R is fixed and the temperature at x^L is increased from T^L to T'^L. The dotted lines in (b) would give rise to NDTC.

Such expressions are useful to calculate the magnitude of DTCs from the temperature profile without the needs to know directly the heat current and its variation [as in Eq. (4)]. They also directly prove the nonexistence of NDTC here: since $F(x)$ and κ are positive, we have $G^L > 0$ and $G^R > 0$.

Systems with a single abrupt junction. As shown in Fig. 1(b), we assume that the abrupt junction is located at $x^J \in (x^L, x^R)$. The system can be considered as two subsystems without any abrupt junctions coupled together at x^J. We suppose that the subsystems in $[x^L, x^J]$ and $[x^J, x^R]$ have thermal conductivity $\kappa_1(x, T_1)$ with temperature profile $T_1(x)$ and $\kappa_2(x, T_2)$ with temperature profile $T_2(x)$, respectively. We denote

$$T_1^J \equiv T_1(x^J),\ T_2^J \equiv T_2(x^J),$$
$$T^L \equiv T_1(x^L),\ T^R \equiv T_2(x^R). \tag{8}$$

At the junction, the two subsystems are coupled through a thermal contact resistance (TCR) R^J, defined such that the heat current q flowing through the system satisfies [31]

$$T_1^J - T_2^J = qR^J(T_1^J, T_2^J). \tag{9}$$

We have generally assumed that the TCR depend on two temperatures, T_1^J and T_2^J. The TCR $R^J(T_1^J, T_2^J)$ provides the complete characterizations of the junction at the phenomenological level.

The possibility of NDTC here can also be interpreted graphically. Again, take the case that T^R is fixed as an example. If the temperature T^L ($> T^R$) is increased to T'^L ($> T^L$), the temperature profile $T(x)$ with $T(x^{L(R)}) = T^{L(R)}$, junction temperatures $T^J_{1,2}$, and heat current q are changed to $T'(x)$ with $T'(x^L) = T'^L$ and $T'(x^R) = T^R$, junction temperatures $T'^J_{1,2}$, and heat current q', as illustrated in Fig. 2(b). Because of the discontinuous jump of temperature at x^J, we could have $q' > q$ if $T'^J_2 > T^J_2$ (red dashed line) or $q' < q$ if $T'^J_2 < T^J_2$ (red dotted line) where the latter situation gives rise to NDTC. We derive the conditions of the existence of NDTC in more detail below.

Analytically, the DTCs are (Appendix 2)

$$G^L = \left(1 - q\frac{\partial R^J}{\partial T^J_1}\right)\frac{F_1(x^L)}{R^D}, \quad G^R = \left(1 + q\frac{\partial R^J}{\partial T^J_2}\right)\frac{F_2(x^R)}{R^D}, \tag{10}$$

with

$$F_i(x) \equiv \exp\left\{\int_{x^J}^{x} \frac{1}{\kappa_i(x',T_i(x'))}\frac{\partial \kappa_i}{\partial T_i}\frac{dT_i}{dx'}dx'\right\}, \quad i = 1,2, \tag{11}$$

defined on $[x^L,x^J]$ and $[x^J,x^R]$, respectively, and

$$R^D \equiv R^J + \left(1 - q\frac{\partial R^J}{\partial T^J_1}\right)J_1 + \left(1 + q\frac{\partial R^J}{\partial T^J_2}\right)J_2, \tag{12}$$

where

$$J_1 \equiv \int_{x^L}^{x^J} \frac{F_1(x')}{\kappa_1(x',T_1(x'))}dx', \quad J_2 \equiv \int_{x^J}^{x^R} \frac{F_2(x')}{\kappa_2(x',T_2(x'))}dx'. \tag{13}$$

If the TCR is independent of the junction temperatures T^J_1 and T^J_2, the partial derivatives of R^J in Eqs. (10) and (12) vanish, and since F_i in Eq. (11) and J_i in Eq. (13) are positive, thus, $G^{L(R)} > 0$ and there is no NDTC. Therefore, a temperature-dependent TCR is necessary for the existence of NDTC. However, as we will see, it is not a sufficient condition.

In the presence of the temperature dependence of TCR, we pick a T_B such that $T_B^{-1} > |R^{-1}\partial R^J/\partial T^J_{1,2}|$, where $R = (T^L - T^R)/q$ is the thermal resistance of the whole system, including the TCR. Inside the regime defined by $|T^L - T^R| < T_B$ in the (T^L,T^R) quarter plane, we have $|q\partial R^J/\partial T^J_{1,2}| < 1$ and subsequently $R^D > 0$ and $G^{L(R)} > 0$: no NDTC is displayed in this low bias regime. Therefore, a temperature bias exceeding T_B is required to observe NDTC, confirming that NDTC is a *nonlinear* thermal transport phenomenon.

As the temperature bias ($|T^L - T^R|$) increases beyond T_B, $G^{L(R)}$ could possibly be negative, leading to NDTC. We denote the dimensionless quantities

$$X \equiv \left(1 - q\frac{\partial R^J}{\partial T^J_1}\right)\frac{J_1}{R^J} = \frac{\partial q}{\partial T^J_1}J_1,$$
$$Y \equiv \left(1 + q\frac{\partial R^J}{\partial T^J_2}\right)\frac{J_2}{R^J} = -\frac{\partial q}{\partial T^J_2}J_2, \tag{14}$$

such that

$$G^L = \frac{X}{1+X+Y}\frac{F_1(x^L)}{J_1}, \quad G^R = \frac{Y}{1+X+Y}\frac{F_2(x^R)}{J_2}. \tag{15}$$

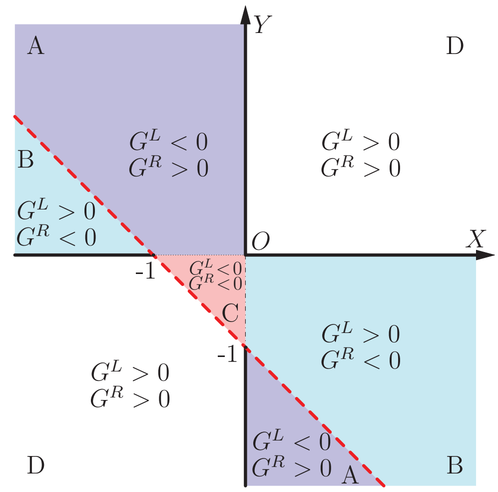

FIG. 3. (Color online) The phase diagram on the X-Y plane that present NDTC.

Now there exists NDTC if and only if at least one of X and Y is negative, which means that at least one of $\partial q/\partial T^J_1$ and $-\partial q/\partial T^J_2$ is negative [32]. We refer to such junctions as those with *intrinsic junction NDTC*, which is now necessary and sufficient for NDTC to occur. Thus, the existence of NDTC in systems with a single abrupt junction is uniquely determined by the properties of the TCRs, regardless of the properties of the system away from the junction.

Furthermore, we can formulate the existence of NDTC on the X-Y plane: find out the points on the plane that correspond to negative G^L or G^R. NDTC exists inside the shaded areas (A, B, and C in Fig. 3), not including the boundaries labeled by the thick solid black and red dashed lines. Note that we have $G^L < 0$ and $G^R = 0$ on the thin dotted line ($-1 < X < 0$, $Y = 0$), while $G^L = 0$ and $G^R < 0$ on the thin dash-dotted line ($X = 0, -1 < Y < 0$). We have $R^D = 0$ and, thus, infinite ($\pm\infty$) DTCs on the thick red dashed line [33]. For the points in the shaded areas and close to the thick red dashed line, we can have very large magnitude of NDTC, useful to design sensitive detectors for temperature fluctuations.

The blue and cyan shaded areas A and B in Fig. 3 are not bounded on the X-Y plane. They correspond to one of G^L and G^R is negative and the other is positive, i.e., $G^L G^R < 0$, which is equivalent to

$$XY < 0. \tag{16}$$

Equation (16) includes the situation that $G^{L(R)}$ is infinite (on the thick solid red lines inside the second and the fourth quarters of the X-Y plane). We can write Eq. (16) in a more transparent way by rewriting the temperature dependence of the TCR $R^J(T^J_1,T^J_2) = R^J(\hat{T}^J,\bar{T}^J)$, where $\hat{T}^J \equiv T^J_1 - T^J_2$ and $\bar{T}^J \equiv T^J_1 + T^J_2$:

$$\left|q\frac{\partial R^J}{\partial \hat{T}^J} - 1\right| < \left|q\frac{\partial R^J}{\partial \bar{T}^J}\right|. \tag{17}$$

Equation (17) implies that $|q\frac{\partial R^J}{\partial \bar{T}^J}| > 0$, i.e., the TCR must be dependent on $\bar{T}^J$. This is physically significant, because in thermal transport we have a natural temperature ground of absolute zero temperature. This demonstrates the drastic difference between the thermal and electrical transport, where in the latter case the junction behavior only depends on the voltage difference (not the average voltage) across the junction.

The red shaded area C in Fig. 3 is bounded, and it corresponds to the case that both $1-q\partial R^J/\partial T_1^J$ and $1+q\partial R^J/\partial T_2^J$ are negative and R^D is positive, equivalent to

$$q\frac{\partial R^J}{\partial \hat{T}^J}-1>\left|q\frac{\partial R^J}{\partial \bar{T}^J}\right|,\quad |q|<\frac{R^J+J_1+J_2}{\left|\frac{\partial R^J}{\partial T_1^J}\right|J_1+\left|\frac{\partial R^J}{\partial T_2^J}\right|J_2}. \quad (18)$$

Equations (17) and (18) imply that

$$|q|>\left(\max\left\{\left|\frac{\partial R^J}{\partial T_1^J}\right|,\left|\frac{\partial R^J}{\partial T_2^J}\right|\right\}\right)^{-1} \quad (19)$$

and

$$|q|>\left(\min\left\{\left|\frac{\partial R^J}{\partial T_1^J}\right|,\left|\frac{\partial R^J}{\partial T_2^J}\right|\right\}\right)^{-1}, \quad (20)$$

respectively, suggesting a positive minimum on the heat current for the existence of NDTC. This again indicates that the existence of NDTC is in the *nonlinear* regime, beyond low heat current.

At the onset of NDTC, we have either $G^L=0$ or $G^R=0$ and the magnitude of heat current (versus temperature bias) reaches its local maximum. Taking the case of $G^L=0$ as an example, we have the following variation rates at the vicinity of $G^L=0$ (Appendix 2):

$$\begin{aligned}\frac{\delta(T_1^J-T_2^J)}{\delta T^L}&=F_1(x^L)-G^L(J_1+J_2)\approx F_1(x^L)>0,\\ \frac{\delta(T_2^J-T^R)}{\delta T^L}&=G^LJ_2.\end{aligned} \quad (21)$$

When the system enters the NDTC regime of $G^L\lesssim 0$, the temperature increase δT^L of T^L is distributed over $[x^L,x^R]$ in such a way that the temperature drop over $[x^J,x^R]$ is decreasing with increasing T^L (which is the manifestation of NDTC), while the temperature drop over the junction is increasing with T^L, as shown in Fig. 2.

Systems with multiple abrupt junctions can exhibit both NDTC and infinite DTCs, but the detailed conditions for their occurrence are more complicated. It can be proved that NDTC still requires that at least one of the junctions possess intrinsic junction NDTC (Appendix 3). Nevertheless, these junctions can be grouped into a single effective junction with its properties determined by the way the junctions are organized (e.g., the order and the connection materials) and by the properties of those individual junctions, as shown in Fig. 1(c). After identifying the effective TCR R^J_{eff}, we can treat the system with multiple junctions as one with a single junction. The discussions in the previous section can be readily applied by simply replacing R^J with R^J_{eff}.

This procedure also provides us a routine to engineer the TCR. For example, we can construct a system composed of three segments in $[x^L,x_1^J]$, $[x_1^J,x_2^J]$, and $[x_2^J,x^R]$. Suppose that $[x^L,x_1^J]$ and $[x_2^J,x^R]$ contain the same kind of uniform material and the material in $[x_1^J,x_2^J]$ is also uniform but different. We can have a single effective junction with its TCR $R^J_{\text{eff}}=q/(T_1^J-T_2^J)$, where T_1^J is the temperature at x_1^J at the side of $[x^L,x_1^J]$, T_2^J is the temperature at x_2^J at the side of $[x_2^J,x^R]$, and q is the heat current flowing across the effective junction. In this way, we have a symmetrical effective junction, i.e., $R^J_{\text{eff}}(T_1^J,T_2^J)=R^J_{\text{eff}}(T_2^J,T_1^J)$.

In conclusion, we have studied the steady-state 1D thermal transport in the diffusive regime without heat sources or sinks. The Fourier's law is applied to calculate the differential thermal conductance. We find that NDTC (in the case that the temperature at one end is fixed) cannot exist in systems without any abrupt thermal junctions. However, we could have NDTC if and only if a junction with intrinsic junction NDTC is introduced. Our predictions provide a theoretical foundation to experimentally realize NDTC through careful thermal contact engineering, though it remains an open question to realize a junction with intrinsic junction NDTC.

This work is partially supported by the Semiconductor Research Corporation (SRC)—Nanoelectronics Research Initiative (NRI) via Midwest Institute for Nanoelectronics Discovery (MIND) and the Cooling Technologies Research Center (CTRC) at Purdue University. J.H. thanks Xiulin Ruan (Purdue University) and Xingpeng Yan for useful discussions.

APPENDIX

1. Systems without abrupt junctions

To calculate the differential thermal conductance (DTC), we start from the variation of $q=-\kappa(x,T(x))\frac{\mathrm{d}T(x)}{\mathrm{d}x}$:

$$\delta q=-\kappa(x,T)\frac{d}{dx}\delta T-\frac{\partial\kappa}{\partial T}\frac{dT}{dx}\delta T. \quad (A1)$$

We define

$$U^{L(R)}(x)\equiv\frac{\delta T(x)}{\delta T^{L(R)}}, \quad (A2)$$

which according to Eq. (A1) satisfies

$$\frac{d}{dx}U^{L(R)}+\frac{1}{\kappa(x,T)}\frac{\partial\kappa}{\partial T}\frac{dT}{dx}U^{L(R)}+\frac{\eta^{L(R)}G^{L(R)}}{\kappa(x,T)}=0. \quad (A3)$$

where $\eta^L=1$ and $\eta^R=-1$. At the boundaries we have

$$\delta T(x^{L(R)})=\delta T^{L(R)}\quad\text{while}\quad\delta T(x^{R(L)})=0, \quad (A4)$$

since $T(x^{L(R)})=T^{L(R)}$. Thus, from Eq. (A2), the boundary conditions for Eq. (A3) are

$$U^L(x^L)=1,U^L(x^R)=U^R(x^L)=0,U^R(x^R)=1. \quad (A5)$$

Equation (A3) is an inhomogeneous linear ordinary differential equation, and the coefficients $\frac{1}{\kappa(x,T)}\frac{\partial\kappa}{\partial T}\frac{dT}{dx}$ and $\frac{\eta^{L(R)}G^{L(R)}}{\kappa(x,T)}$ are functions of x only, since $T(x)$ is already formally solved from Eq. (2) and boundary conditions Eq. (3). The solution to Eq. (A3) is

$$U^{L(R)}(x)=\frac{U^{L(R)}(x^L)}{F(x)}-\frac{\eta^{L(R)}G^{L(R)}}{F(x)}\int_{x^L}^{x}\frac{F(x')}{\kappa(x',T(x'))}dx', \quad (A6)$$

where

$$F(x)\equiv\exp\left\{\int_{x^L}^{x}\frac{1}{\kappa(x',T(x'))}\frac{\partial\kappa}{\partial T}\frac{dT}{dx'}dx'\right\}. \quad (A7)$$

By evaluating Eq. (A6) at $x=x^R$, we obtain

$$G^{L(R)}=J^{-1}\eta^{L(R)}[U^{L(R)}(x^L)-F(x^R)U^{L(R)}(x^R)], \quad (A8)$$

where

$$J \equiv \int_{x^L}^{x^R} \frac{F(x')}{\kappa(x',T(x'))}dx'. \tag{A9}$$

From Eqs. (A5) and (A8), we have

$$G^L = J^{-1}, \quad G^R = F(x^R)J^{-1}. \tag{A10}$$

2. Systems with a single junction

For a system composed of two segments lying in $[x^L,x^J]$ and $[x^J,x^R]$, we denote the temperature profiles $T_1(x)$ and $T_2(x)$ and the thermal conductivity $\kappa_1(x,T_1(x))$ and $\kappa_2(x,T_2(x))$, receptively. If a heat current is flowing in the system, the temperature profiles satisfy

$$\begin{aligned} q &= -\kappa_1(x,T_1(x))\frac{dT_1}{dx}, \quad x^L < x < x^J, \\ q &= -\kappa_2(x,T_2(x))\frac{dT_2}{dx}, \quad x^J < x < x^R, \end{aligned} \tag{A11}$$

with the boundary conditions

$$T_1^J - T_2^J = qR^J\left(T_1^J,T_2^J\right), T_1(x^L) = T^L, T_2(x^R) = T^R. \tag{A12}$$

Applying the variation $\delta T^{L(R)}$ of the boundary temperature at one end, the resulting temperature profiles T_1 and T_2 are varied to $T_1(x) + \delta T_1(x)$ and $T_2(x) + \delta T_2(x)$, respectively. Of course, we have

$$\delta T_1(x^L) = \delta T^L, \quad \delta T_2(x^R) = \delta T^R. \tag{A13}$$

The heat current q is varied to $q + \delta q$. The junction temperatures T_1^J and T_2^J are varied to $T_1^J + \delta T_1^J$ and $T_2^J + \delta T_2^J$, respectively. We then define the following functions:

$$U_1^{L(R)} \equiv \frac{\delta T_1}{\delta T^{L(R)}} \quad \text{and} \quad U_2^{L(R)} \equiv \frac{\delta T_2}{\delta T^{L(R)}} \tag{A14}$$

on $[x^L,x^J]$ and $[x^J,x^R]$, respectively. They satisfy the following equations:

$$\eta^{L(R)}G^{L(R)} = -\kappa_i\frac{dU_i^{L(R)}}{dx} - \frac{\partial \kappa_i}{\partial T_i}\frac{dT_i}{dx}U_i^{L(R)}, \quad i = 1,2 \tag{A15}$$

and boundary conditions

$$U_1^L(x^L) = 1, U_1^R(x^L) = U_2^L(x^R) = 0, U_2^R(x^R) = 1. \tag{A16}$$

Their solutions are

$$U_i^{L(R)}(x) = \frac{U_i^{L(R)}(x^J)}{F_i(x)} - \frac{\eta^{L(R)}G^{L(R)}}{F_i(x)}\int_{x^J}^{x} \frac{F_i(x')}{\kappa_i(x',T_i(x'))}dx' \tag{A17}$$

where

$$F_i(x) \equiv \exp\left(\int_{x^J}^{x} \frac{1}{\kappa_i(x',T_i(x'))}\frac{\partial \kappa_i}{\partial T_i}\frac{dT_i}{dx'}dx'\right), \quad i = 1,2. \tag{A18}$$

By evaluating Eq. (A17) at x^L for $i = 1$ and at x^R for $i = 2$, we have

$$\begin{aligned} U_1^{L(R)}(x^J) &= F_1(x^L)U_1^{L(R)}(x^L) \\ &\quad + \eta^{L(R)}G^{L(R)}\int_{x^J}^{x^L} \frac{F_1(x')}{\kappa_1(x',T_1(x'))}dx', \\ U_2^{L(R)}(x^J) &= F_2(x^R)U_2^{L(R)}(x^R) \\ &\quad + \eta^{L(R)}G^{L(R)}\int_{x^J}^{x^R} \frac{F_2(x')}{\kappa_2(x',T_2(x'))}dx'. \end{aligned} \tag{A19}$$

The variation of the first equation in Eq. (A12) gives

$$\begin{aligned} &U_1^{L(R)}(x^J) - U_2^{L(R)}(x^J) \\ &\quad = \eta^{L(R)}G^{L(R)}R^J + q\frac{\partial R^J}{\partial T_1^J}U_1^{L(R)}(x^J) + q\frac{\partial R^J}{\partial T_2^J}U_2^{L(R)}(x^J). \end{aligned} \tag{A20}$$

By inserting Eq. (A19) into Eq. (A20), we finally get the DTC

$$\begin{aligned} G^L &= \left(1 - q\frac{\partial R^J}{\partial T_1^J}\right)\frac{F_1(x^L)}{R^D}, \\ G^R &= \left(1 + q\frac{\partial R^J}{\partial T_2^J}\right)\frac{F_2(x^R)}{R^D}, \end{aligned} \tag{A21}$$

where

$$R^D \equiv R^J + \left(1 - q\frac{\partial R^J}{\partial T_1^J}\right)J_1 + \left(1 + q\frac{\partial R^J}{\partial T_2^J}\right)J_2, \tag{A22}$$

with

$$J_1 \equiv \int_{x^L}^{x^J} \frac{F_1(x')}{\kappa_1(x',T_1(x'))}dx', J_2 \equiv \int_{x^J}^{x^R} \frac{F_2(x')}{\kappa_2(x',T_2(x'))}dx'. \tag{A23}$$

From Eq. (A19), we can also readily write down

$$\begin{aligned} \frac{\delta(T_1^J - T_2^J)}{\delta T^L} &= U_1^L(x^J) - U_2^L(x^J) \\ &= F_1(x^L) - G^L(J_1 + J_2), \\ \frac{\delta(T_2^J - T^R)}{\delta T^L} &= U_2^L(x^J) = G^L J_2. \end{aligned} \tag{A24}$$

3. Existence of NDTC for systems with multiple junctions

To prove that the existence of NDTC for systems with multiple junctions requires that at least one of the junctions has intrinsic junction NDTC, we prove the converse by induction, i.e., there is no NDTC if none of the junctions has intrinsic junction NDTC.

Assume that the system lying in $[x^J,x^R]$ contains an arbitrary number of junctions and the DTCs for this system are nonnegative. We now add a new segment (with no junctions within the segment) lying in $[x^L,x^J]$ to the existing system. We denote $T^{L(R)}$ as the temperature at $x^{L(R)}$. We assume that the new junction at x^J with TCR $R^J(T_1^J,T_2^J)$ has no intrinsic junction NDTC. Here, T_1^J and T_2^J are the temperatures at x^J at the side of $[x^L,x^J]$ and $[x^J,x^R]$, respectively. Suppose we raise the temperature T^L infinitesimally to $T^L + \delta T^L$ ($\delta T^L > 0$)

while keeping T^R fixed. The temperature $T^J_{1(2)}$ is then varied to $T^J_{1(2)} + \delta T^J_{1(2)}$ and the heat current is changed from q to $q + \delta q$. At the junction, we have $\delta q = G^J_1 \delta T^J_1 - G^J_2 \delta T^J_2$, where $G^J_{1(2)} \geqslant 0$ are the junction DTCs. On the other hand, the system in $[x^J, x^R]$ has nonnegative DTCs; i.e., $\delta q = G^B \delta T^J_2$, where $G^B \geqslant 0$. We thus have $G^J_1 \delta T^J_1 = (G^J_2 + G^B)\delta T^J_2$. Of course, δT^J_1 cannot be negative. If either G^J_2 or G^B is positive, we will have $\delta T^J_2 = G^J_1 \delta T^J_1/(G^J_2 + G^B) \geqslant 0$ and, thus, $\delta q = G^B \delta T^J_2 \geqslant 0$ and there is no NDTC. If both G^J_2 and G^B are zero, we will have $\delta q = G^J_1 \delta T^J_1 \geqslant 0$ and there is no NDTC.

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[28] To control the temperatures at two ends of the system, both ends may be directly in contact to two ideal heat baths so that the thermal contact resistances between the heat baths and the system may exist. The temperatures at the two ends of the system can be controlled by varying the temperatures of the heat baths. It is not necessary to require that the temperatures of the heat baths are identical to the temperatures at the two ends of the system. In practice, with careful design, the contact resistances between the heat baths and the systems can be negligibly smaller than the thermal resistance of each segment (no junctions inside each segment) of the system and the thermal contact resistances between those segments. In this case the temperatures of the heat baths are approximately equal to the temperatures at two ends of the system.

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[30] If the temperatures at both ends vary simultaneously and depend on a parameter u [i.e., $T^{L(R)}(u)$], the DTC is $G = \frac{G^L dT^L/du - G^R dT^R/du}{dT^L/du - dT^R/du}$. It is possible to have the numerator of G to be negative while its denominator is positive, if the form of $T^{L(R)}(u)$ is designed carefully.

[31] Usually, $R^J(T^J_1, T^J_2)$ is reduced to $R^J(T^J)$ with $T^J = (T^J_1 + T^J_2)/2$ when $|T^J_1 - T^J_2| \ll T^J$, as being studied in many experiments [E. T. Swartz and R. O. Pohl, Rev. Mod. Phys. **61**, 605 (1989)]. However, that form of R^J is not applicable to the cases involving large currents that we are particularly interested in here.

[32] To have NDTC (negative G^L or G^R), it is necessary that at least one of X and Y is negative. Conversely, if at least one of X and Y is negative, since both X and Y (assumed to be continuous with temperature bias) would be positive in the limit of vanishing temperature bias, there exists a critical temperature bias below which both X and Y are positive and slightly above which at least one of X and Y is negative and $|X| \ll 1$ and $|Y| \ll 1$; thus, one of G^L and G^R must be negative ($1 + X + Y$ is positive) and there exists NDTC.

[33] The zeros of G^L and G^R exist at $q = [\partial R^J/\partial T^J_1]^{-1}$ and $q = -[\partial R^J/\partial T^J_2]^{-1}$, respectively. The zeros may accidentally cancel the singularity at $(X,Y) = (0,-1)$ or $(X,Y) = (-1,0)$, but this cancellation rarely happens and can be avoided by slightly perturbing the boundary temperatures.