Cooling Enhancement Using Inhomogeneous Thermoelectric Materials

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Abstract

The maximum cooling temperature of a thermoelectric refrigerator made of uniform bulk material is limited by its dimensionless figure-of-merit ZT. Cascaded stages are typically needed in order to obtain a higher cooling temperature. Multiple stage configurations have disadvantages of device complexity, and reduced efficiency due to the non-ideal heat spreading between different stages. In this paper, we prove that the maximum cooling temperature can be increased by using a single stage made of inhomogeneous material. This optimization is different from conventional graded materials where there is a large temperature gradient and local material properties are optimized in order to achieve the highest ZT at the local temperature under operation. The new optimization is attributed to the redistribution of the Joule heating and Peltier cooling profiles along the current and heat flow directions. The cooling efficiency can also be increased by a moderate amount. Numerical simulations are used to optimize the doping profile for a thermoelectric cooler based on single crystal silicon.

Maximum Cooling of Thermoelectric Materials

It is well known that the maximum cooling temperature of a uniform thermoelectric material is equal to 1/2 ZT^2, where T is the absolute temperature at the cold side, and Z is the material thermoelectric figure-of-merit. The nondimensional figure-of-merit is defined as $ZT = \left(S^2\sigma / K\right)T$, where S is the Seebeck coefficient, and σ and K are the electrical and thermal conductivities, respectively. [1] Finding thermoelectric materials with high ZT values is hard because a large electrical conductivity is usually accompanied with a small Seebeck coefficient and a large thermal conductivity. The trade off between the electrical conductivity and the Seebeck coefficient can be seen in Fig. 1 for bulk Silicon crystals.

Multiple stage thermoelectric coolers can be used to achieve lower temperatures. However, heat spreaders should be applied between adjacent stages to reduce the heat load density of the subsequent stage. This introduces extra thermal resistance and reduces the total cooling temperature.

By engineering the Peltier cooling and Joule heating profiles in a single-stage three-dimensional thermoelectric device, the maximum cooling can be enhanced by ~ 25% according to a finite element simulation. This is due to 3D heat and current spreading. But it could only be achieved with an array of coolers to control the local current. [2] Most generally, it has been proved that the maximum cooling of a single element thermoelectric material cannot be improved by changing its geometry. [3]

In this paper, we optimize the electrical conductivity and the Seebeck coefficient profile in a one-dimensional current and heat flow configuration in order to increase the maximum cooling of a single thermoelectric element. We will show that decreasing material electrical conductivity monotonically from the cold side to the hot side can enhance the maximum cooling greatly. This idea is different from the conventional "functionally graded thermoelectric material (FGM)" in that it is not based on the temperature dependence of thermoelectric properties. The local ZT is not maximized by changing material type or composition as done in FGM. [4-7] Similar to FGM, Mahan studied efficiencies of power generation and refrigeration for an inhomogeneous Bi_2Te_3 material. [8] He showed that a moderate 7% improvement can be obtained. Linearly graded materials with constant volume average ZT were studied for cooling enhancement. [9] Here we focus on enhancing the maximum cooling temperature of a refrigerator above the limit given by its maximum ZT by using optimized inhomogeneous material.

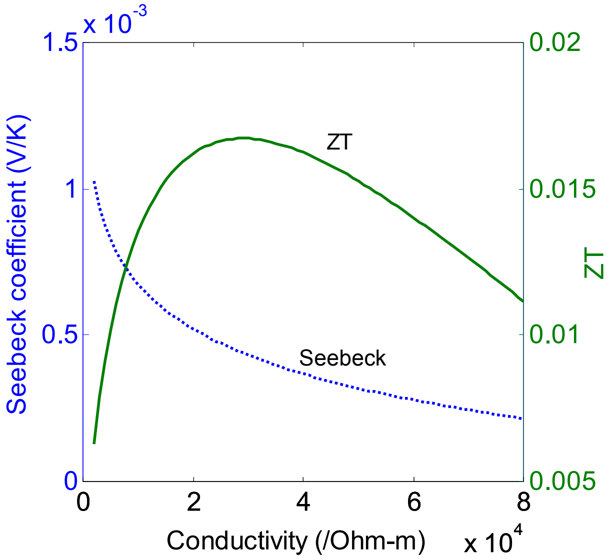

Figure 1: Seebeck coefficient and ZT as a function of the electrical conductivity for bulk crystal Silicon at 373K.

Cooling Enhancement Using Graded Materials

In a uniform thermoelectric cooler, the Peltier cooling is compensated for by the Joule heating so that the maximum cooling temperature is limited. If a constant heat load is applied at the cold side and the hot side is thermally heat sunk, the temperature difference across the uniform material is

$$\Delta T = R_{th}\left(SIT_c - \frac{1}{2}I^2R - Q_c\right), \qquad (1)$$

where I is the current, T_c is the temperature at the cooling side, ΔT is the temperature difference across the material, and R and R_{th} are the electrical and thermal resistances of the material, respectively. The Peltier cooling is localized near the interface with the metal contact. The 1/2 factor in front of the total Joule heating comes from the integral of the distributed heating produced inside the material. In a thermal circuit analysis, as long as we are only concerned with the temperature difference, the distributed Joule heating can be replaced by two heat sources localized at the two ends of the uniform material, each with a value equal to the half of the total Joule heating. When the cooling power is zero, it is easy to find that the maximum cooling temperature is $1/2\ ZT_c^2$ by optimizing ΔT with respect to I. It is logical to wonder if the maximum cooling temperature can be increased by redistributing the Joule heating and Peltier cooling using inhomogeneous materials.

Thermal conductivity of a semiconductor is usually dominated by the lattice contribution. If the doping density changes with position, the electrical conductivity changes much faster than the Seebeck coefficient and it is inversely proportional to it. Thus, the thermoelectric power factor ($S^2\sigma$) and thermal conductivity are almost constant in a finite range of carrier concentration around that of the maximum ZT value. Furthermore, to simplify the analysis for the proof-of-concept, we first tested low ZT materials and small cooling temperatures so that we could neglect the temperature variation in the materials and its effects on the local Peltier cooling power and physical properties of the material.

Let's start with a staircase Seebeck profile. As an example, the three- section element is shown in the inset of Fig. 2. The left end of the first section (x=0) is thermally insulated and the right end of the last section (x=L) is thermally heat sunk. The first section of the staircase material has a Seebeck coefficient S_1, an electrical conductivity σ_1, and a length L_1. The optimum current density to achieve the largest cooling temperature for this section is $J_{opt} = S_1 T \sigma_1 / L_1$. Under this optimum current, in thermal circuit analysis half of the Joule heating located at the cold side cancels half of the Peltier cooling at the cooling surface.

In choosing the Seebeck profile, we want half of the Peltier cooling power at the interfaces of adjacent sections to be cancelled by half of the Joule heating in the adjacent two sections at J_{opt}, the optimal current for the first section. Thus, half of the Peltier cooling at each interface should remain, and the entire Joule heating in the body is compensated for. This requires that the Seebeck coefficient of the n^{th} section is $(2n-1)S_1$, the electrical conductivity is $\sigma_1/(2n-1)^2$ (assuming constant power factor), and the section length is $L_1/(2n-1)^2$. From the thermal circuit analysis, it is easy to conclude that the optimal current for the N-section material is the same as J_{opt} of the first section and the maximum cooling temperature is

$$\Delta T = \frac{1}{2} ZT^2 \sum_{n=1}^{N} \frac{1}{2n-1}, \qquad (2)$$

which is larger than $1/2\ ZT^2$. The cooling enhancement of staircase materials of up to 6 sections is shown in Fig. 2. The cooling temperature in the unit of $1/2\ ZT^2$ jumps whenever the Seebeck coefficient S_L (x=L) increases to a point where one more section can be added. One may wonder if a continuously graded material could achieve better results. For a one-dimensional thermoelectric transport, the heat equation at steady state can be written as

$$\frac{d}{dx}\left(K(x) \frac{dT(x)}{dx} \right) = -\frac{J^2}{\sigma(x)} + JT(x)\frac{dS(x)}{dx}. \qquad (3)$$

Again, to simplify the proof-of-concept and obtain an analytical solution, we assume that the thermal conductivity K and the power factor $S(x)^2\sigma(x) = A$ are constant in a length L. The Seebeck coefficients at the starting and ending positions are represented by S_0 and S_L, respectively. The variation of Peltier cooling power due to small temperature change is always negligible since it is only a small fraction in the total Peltier cooling power $(\Delta T / T << 1)$ at room temperature. It can be shown that the cooling temperature of such a continuously graded material is

$$\Delta T = \frac{1}{K}\left(\int_0^L dx \int_0^x dx' \left(-\frac{J^2 S^2(x')}{A} \right) + \int_0^L JTS(x)dx \right). \qquad (4)$$

Taking the differential with respect to the current, we can obtain the optimal current density as

$$J_{opt} = \frac{T\int_0^L S(x)dx}{\frac{2}{A}\int_0^L dx \int_0^x S^2(x')dx'}. \qquad (5)$$

The resulting maximum cooling temperature is

$$\Delta T_{\max} = \frac{1}{2} ZT^2 \frac{\int_0^L S(x)dx \int_0^x S(x')dx'}{\int_0^L dx \int_0^x S^2(x')dx'} \geq \frac{1}{2} ZT^2. \qquad (6)$$

It can be observed that the maximum cooling temperature is larger than $1/2\ ZT^2$, if the Seebeck coefficient increases monotonically with position. It is interesting to note that the increase of maximum cooling temperature above the $1/2\ ZT^2$ limit is independent of the absolute magnitudes of the Seebeck coefficient, as long as the ratio S_L/S_0 is fixed. It is also independent of the length L and the absolute gradient of the Seebeck profile, as long as its shape is scaled as $S(ax)$ where a is a constant. This means that the grading optimization is not limited to a specific range of material properties or dimensions of the device.

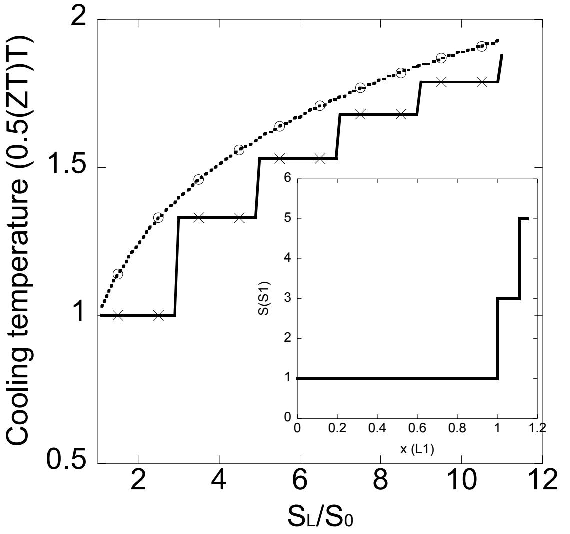

Figure 2: Cooling enhancement of staircase Seebeck profile (x) and the continuously graded material (○). Inset shows the Seebeck profile of the staircase configuration as a function of distance.

The continuously graded version of the above staircase is represented by the function $1/S_0 - 1/S(x) = ax$, whose cooling enhancement is also shown in Fig.2. Its performance is above that of the staircase in the whole range of the Seebeck coefficient ratio, as expected.

Graded Silicon Material

As can be seen in Fig. 1, the thermoelectric figure-of-merit ZT is not constant when the electrical conductivity changes for a practical material such as silicon. There is no obvious solution of equation (3) for optimal cooling current and maximum cooling temperature given realistic material properties. It is expected that an optimal electrical conductivity profile will give a higher cooling temperature that that achieved with the uniform material of the largest ZT. For Silicon, we assume that the thermal conductivity is constant and that the temperature variation along the material can be neglected in the Peltier cooling terms. We then try to find an optimum electrical conductivity profile along the length of the element which can give the highest cooling when the current is optimized. It will be very computation intensive to try all the possible profiles with a high spatial resolution systematically. Rather, we generate random electrical conductivities for N material sections of the same length in a range between σ_1 and σ_N. The Seebeck coefficients are determined according to Fig. 1. The material measures 100μmx100μmx100μm. The current is scanned to find the maximum cooling temperature for a given electrical conductivity profile. Different number of sections, N, and various ranges of the electrical conductivity are tried to see which is much easier to produce larger maximum cooling temperatures. Finally, it is found that 8 sections and electrical conductivity ranging from 4×10^3 /Ohm-m to 8×10^4 /Ohm-m can produce largest coolings. Then we stick to these conditions and find that the electrical conductivity profiles performing the best have a decreasing trend from the cold side to the hot side. The electrical conductivity profile is plotted in Fig. 3 for 52 configurations with highest cooling temperatures, which vary from 4.045K to 4.056K (for a comparison, the maximum cooling temperature for the uniform bulk Si with the largest ZT is 3.117K). The cooling enhancement is about 30% and the variation tolerance of the optimum electrical conductivity profile is fairly large.

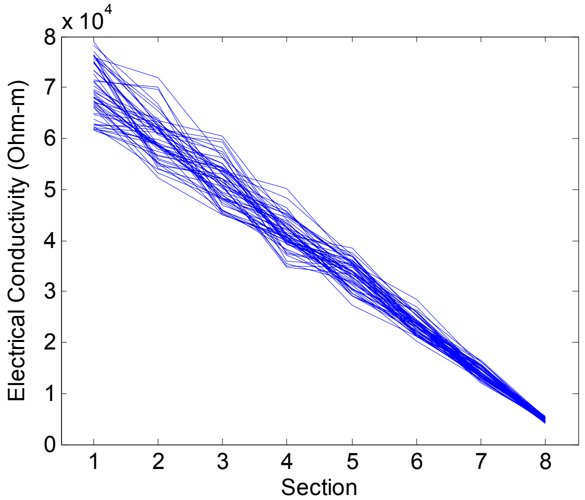

Figure 3: Electrical conductivity profiles of 8-section inhomogeneous Si giving largest cooling temperatures.

The average of these 52 8-section profiles is plotted in Fig. 4, which is almost linear with position. In Fig. 5, we plot the cooling curves of the uniform Silicon with the largest ZT, and the average of 8-section profiles and the continuously graded material according to its linear fitting. The maximum cooling temperature is 4.059K for the average of 8-section profiles, and is 4.214K for the continuously graded material. The cooling enhancement compared to the uniform material is 30% and 35% respectively.

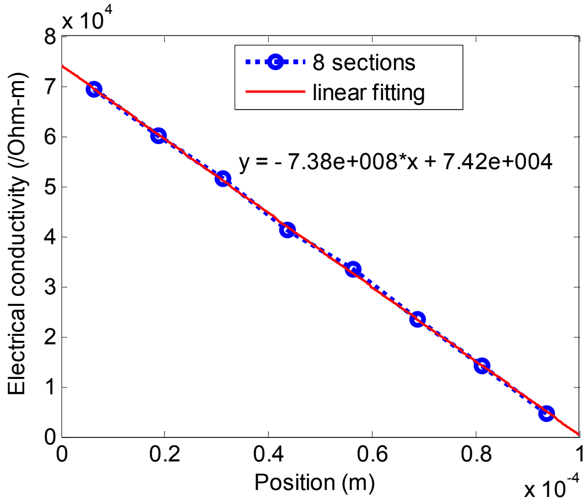

Figure 4: The average of 52 electrical conductivity profiles (8-sections) giving the largest cooling temperatures, and its linear fitting.

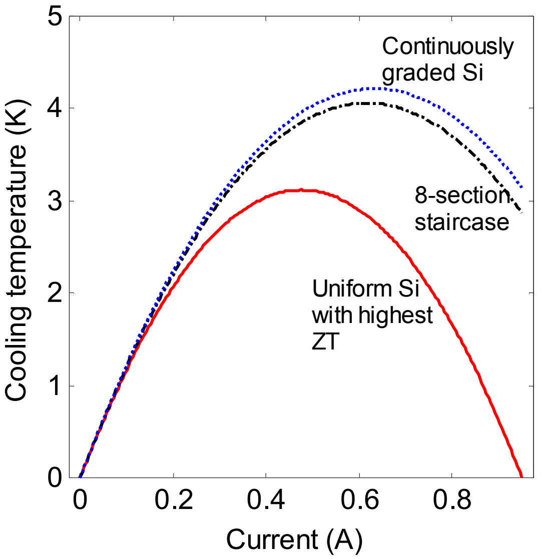

Figure 5: The cooling curves of the uniform Silicon with the largest ZT, and the average of 8-section profiles and the continuously graded material according to its linear fitting.

The Seebeck coefficient and ZT of the linear electrical conductivity profile are plotted in Fig. 6. It can be seen that Seebeck coefficient increases monotonically and its change becomes abrupt at the hot side. In this optimum doping profile, the ZT value can be as small as 25% of its maximum at the hot side. This really proves that the profile of ZT is as important as its maximum value to the maximum cooling temperature.

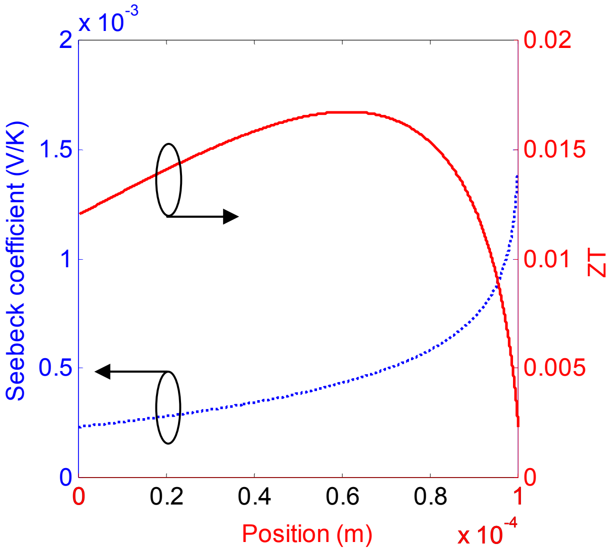

Figure 6: The Seebeck coefficient and ZT as a function of distance for the linear electrical conductivity profile that gives the largest cooling.

Conclusions

It has been shown that the maximum cooling temperature can be greatly increased by using inhomogeneous thermoelectric materials thanks to the redistribution of the Peltier cooling and the Joule heating. A cooling enhancement of 35% can be achieved for graded Silicon crystals according to a numerical simulation where the practical relation of the electrical conductivity and the Seebeck coefficient is taken into account. The temperature dependences of the material properties need to be included into the calculation self-consistently for high ZT materials and one can expect similar cooling enhancement.

Acknowledgments

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