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On the effective thermoelectric properties of layered heterogeneous medium

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The effective thermoelectric behavior of layered heterogeneous medium is studied, with the distribution of temperature, electric potential, and heat flux solved rigorously from the governing equations, and the effective thermoelectric properties defined through an equivalency principle. It is discovered that the effective thermoelectric figure of merit of a composite medium can be higher than all of its constituents even in the absence of size and interface effects, in contrast to previous studies. This points toward a new route for high figure of merit thermoelectric materials. © *2012 American Institute of Physics*. [doi:10.1063/1.3674279]

I. INTRODUCTION

Thermoelectric materials have attracted significant interests recently for their capability in converting waste heat directly into electricity utilizing the Seebeck effect,[1] wherein a voltage difference proportional to temperature difference is developed when two dissimilar materials are joined together and the junctions are held at different temperatures. The efficiency of thermoelectric conversion is governed by a dimensionless figure of merit ZT, which is intimately related to electric and thermal transport properties of thermoelectric materials,[2]

$$ZT = \frac{\varepsilon^2 \sigma T}{\kappa}, \tag{1}$$

where ε, σ, and κ are Seebeck coefficient, electric conductivity, and thermal conductivity, respectively, and T is the temperature. Clearly, in order to have high ZT and thus high conversion efficiency, the thermoelectric material needs to have not only high Seebeck coefficient, but also high electric conductivity and low thermal conductivity. This turns out to be rather difficult, since all these properties are intimately related to each other, making it hard to control them individually. For example, while high electric conductivity requires high concentration of charge carriers, a modest carrier concentration is optimal for high Seebeck coefficient. Furthermore, high electric conductivity is usually accompanied by high thermal conductivity because of Wiedemann-Franz Law.[3] As a result, it is very challenging to achieve high electric conductivity and Seebeck coefficient and low thermal conductivity simultaneously in a single-phase material.

In the past decade, there are mainly two strategies in developing high performance thermoelectric materials, including (1) searching for bulk materials with intrinsically high figure of merit and conversion efficiency; and (2) engineering hybrid composite materials to enhance and optimize their conversion efficiency. Hybrid materials, especially nanostructured materials, are very attractive for thermoelectric energy conversion. By combining different materials together, it is possible to overcome the intrinsic constraints between electric conductivity, thermal conductivity, and Seebeck coefficient in a single-phase material through microstructure engineering and optimization, and thus achieve high electric conductivity, Seebeck coefficient, and low thermal conductivity simultaneously. For example, a large enhancement in thermoelectric power was observed in nanocomposites consisting of bismuth nanowires embedded in porous alumina and porous silica,[5] and a ZT value as high as 1.6 has been reported in $K_{1-x}Pb_{m+\delta}Sb_{1+\gamma}Te_{m+2}$ system containing nanoinlcusions, which possesses simultaneously low thermal conductivity and high electrical conductivity.[6] Other high thermoelectric figure of merit heterogeneous systems include La-doped n-type PbTe-Ag_2Te nanocomposites with large nanometer-scale precipitates,[7] melt spun $Bi_{0.52}Sb_{1.48}Te_3$ bulk materials with nanocrystals embedded inside the amorphous matrix,[8,9] and nanostructured $Ag_{0.8}Pb_{m+x}SbTe_{m+2}$,[10] among others.

While vast amount of experimental works in thermoelectric materials focus on nanostructured composites,[4–14] there have been only very limited theoretical efforts toward the analysis and understanding of the effective behavior of heterogeneous thermoelectric materials, despite their importance. Instead, most previous theoretical studies focused on size and interfacial effects at nanoscale using molecular dynamics and quantum mechanics.[15–23] This motivates us to examine whether the effective thermoelectric figure of merit of a heterogeneous composite can be higher than all its constituents, excluding the effects of size and interface. If the answer is yes, it will offer us a new route for high figure of merit thermoelectric materials, while the current state of art focuses on nanocomposites that utilize quantum effects at nanoscale and phonon scattering at interfaces,[4–23] which are much more difficult to process and control than regular composites. The question has actually been visited by a number of investigators before. In 1991, it was shown that the effective figure of merit of a composite can never exceed the largest figure of merit in any of its constituents, in the absence of size and interfacial effects.[24,25] This conclusion was drawn from a variational principle similar to Hashin-Strikman bound in elasticity.[26] However, for thermoelectric materials, temperature, which is

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not uniform in general and varies from point to point, enters into constitutive equations as a coefficient, and thus should not be treated as material constants. Furthermore, heat flux was often assumed to be divergence free,[27] and under such assumption, temperature distribution satisfies Laplace equation, as in a regular heat transfer problem uncoupled from electric current. For thermoelectric materials, especially those with high figure of merit, this is also not true.

Because of these difficulties, very few works on the homogenization of thermoelectric composites exist in literature, far less than elastic composites, despite their obvious technological importance in energy harvesting and solid state cooling. We seek to address these issues in this paper, by analyzing the effective thermoelectric properties of layered heterogeneous medium. More complicated three-dimensional composites as studied in Ref. 32 is currently under investigations. The paper is organized as follows. The governing equation of thermo-electricity is introduced in Sec. II, and the one-dimensional (1D) analysis of temperature and electric field distribution in layered heterogeneous medium is presented in Sec. III, from which the effective thermoelectric properties can be derived. Numerical results and discussion are then given in Sec. IV, confirming that the effective thermoelectric figure of merit of composites can indeed be higher than both of its constituents.

II. GOVERNING EQUATIONS OF THERMOELECTRICITY

We consider the coupled transports of heat and electrons in a thermoelectric material, with the respective transport equations given by[28,29]

$$-\mathbf{J}_N = \left(\frac{T\sigma}{e^2}\right)\frac{1}{T}\nabla\mu - \left(\frac{T^2\sigma\varepsilon}{e}\right)\nabla\frac{1}{T}, \tag{2}$$

$$\mathbf{J}_Q = -\left(\frac{T^2\sigma\varepsilon}{e}\right)\frac{1}{T}\nabla\mu + (T^3\sigma\varepsilon^2 + T^2\kappa)\nabla\frac{1}{T}, \tag{3}$$

wherein the electron flux $\mathbf{J}_N$ is coupled with the heat flux $\mathbf{J}_Q$ through the Seebeck coefficient ε, with e and μ being the charge and electrochemical potential of the electron, respectively. Assume that the chemical potential is independent of temperature and thus can be ignored, such that

$$\mu = e\phi, \tag{4}$$

where ϕ is electric potential, and notice that the electric current density can be derived from electron flux,

$$\mathbf{J} = e\mathbf{J}_N, \tag{5}$$

the transport equations governing electric current density and heat flux can be derived as follows:

$$-\mathbf{J} = \sigma\nabla\phi + \sigma\varepsilon\nabla T, \tag{6}$$

$$\mathbf{J}_Q = -T\varepsilon\sigma\nabla\phi - (T\varepsilon^2\sigma + \kappa)\nabla T = T\varepsilon\mathbf{J} - \kappa\nabla T. \tag{7}$$

In the absence of thermoelectric effect where $\varepsilon = 0$, the uncoupled transport equations of electricity and heat are recovered. Since energy is transported by both electrons and heat, the energy flux $\mathbf{J}_E$ can also be derived from the current density and heat flux as

$$\mathbf{J}_E = \mathbf{J}_Q + \phi\mathbf{J}. \tag{8}$$

Notice that both temperature gradient ∇T and temperature T enter the thermal transport Eq. (7), making the coupling nonlinear and thus much more difficult to solve than a normal heat transfer problem in uncoupled medium.

We consider a system wherein both charges and energy are conserved, such that both current density and energy flux are divergence-free,

$$\nabla\cdot\mathbf{J} = 0, \tag{9}$$

$$\nabla\cdot\mathbf{J}_E = 0. \tag{10}$$

On the other hand, the heat flux is no longer divergence-free,

$$\nabla\cdot\mathbf{J}_Q = -\nabla\cdot(\phi\mathbf{J}), \tag{11}$$

in contrast to a normal heat transfer problem in uncoupled medium.

III. ONE-DIMENSIONAL ANALYSIS

We consider a layered thermoelectric medium, wherein all the material properties and field variables are only dependent on spatial coordinate x, and independent of y and z. Since the current density is divergence-free, its magnitude J turns out to be a constant in such a

1D configuration,

$$J = |\mathbf{J}| = \text{const}, \tag{12}$$

which simplifies the governing equations of thermoelectricity considerably, making analytic solutions possible. Meanwhile, such 1D configuration is most relevant for practical thermoelectric modules. In the following subsections, we examine the solution for a homogeneous thermoelectric first, and then extend the solution to a layered heterogeneous medium.

A. Analysis of a homogeneous thermoelectric

We first consider a homogeneous material with uniform distribution of material properties, as shown in Fig. 1(a), and subjected to specified temperatures and electric potentials of (T_0, ϕ_0) and (T_1, ϕ_1) at both ends. All the material properties

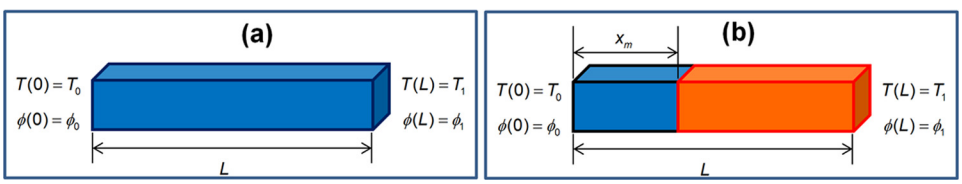

FIG. 1. (Color online) Schematics of layered thermoelectric medium; (a) homogeneous thermoelectric; (b) layered heterogeneous thermoelectric.

TABLE I. Thermoelectric properties of Bi_2Te_3 (Ref. 30) and $Ag(Pb_{1-y}Sn_y)_mSbTe_{2+m}$ (Ref. 31).

Material	ε ($\times 10^{-6}$ V/K)	σ ($\times 10^3$ S/m)	κ (W/m/K)
Bi_2Te_3	200	110	1.6
$Ag(Pb_{1-y}Sn_y)_mSbTe_{2+m}$	270	22	0.77

are assumed to be independent of temperature, and thus Thomson effect is ignored.[29] This allows us to simplify the field equations governing temperature and electric potential distributions as follows:

$$\frac{d^2T}{dx^2} = -\frac{J^2}{\sigma\kappa}, \tag{13}$$

$$\frac{d^2\phi}{dx^2} = \varepsilon\frac{J^2}{\sigma\kappa}. \tag{14}$$

Equation (13) can be solved for temperature distribution as

$$T = -\frac{J^2}{2\sigma\kappa}x^2 + c_1x + c_2, \tag{15}$$

with integration constants determined from boundary conditions as

$$c_1 = (T_1 - T_0) + \frac{J^2}{2\sigma\kappa}, \quad c_2 = T_0, \tag{16}$$

where all the dimensions are normalized with respect to L, the length of the thermoelectric medium.

To solve for the yet to be determined current density J, we notice from Eq. (6) that the distribution of electric potential is govern by

$$\frac{d\phi}{dx} = \frac{\varepsilon J^2}{\sigma\kappa}x - \frac{\varepsilon J^2}{2\sigma\kappa} - \frac{J}{\sigma} - (T_1 - T_0)\varepsilon, \tag{17}$$

so that

$$\phi = \frac{\varepsilon J^2}{2\sigma\kappa}x^2 - \left[\frac{\varepsilon J^2}{2\sigma\kappa} + \frac{J}{\sigma} + (T_1 - T_0)\varepsilon\right]x + c_3, \tag{18}$$

which allows us to solve c_3 and current density from electric boundary conditions as

$$c_3 = \phi_0 \tag{19}$$

and

$$J = \sigma\varepsilon(T_0 - T_1) + \sigma(\phi_0 - \phi_1). \tag{20}$$

The heat flux J_Q can then be calculated using Eq. (7). This completely solves the distribution of electric potential and temperature in a homogeneous thermoelectric.

B. Analysis of a layered thermoelectric

We then consider a layered heterogeneous medium consisting of two homogeneous thermoelectric phases A and B, as shown in Fig. 1(b), and subjected to specified temperatures and electric potentials of (T_0, ϕ_0) and (T_1, ϕ_1) at both ends, identical to those of homogeneous medium considered in the last subsection. Since the layered medium is pieces-wise uniform, Eq. (15) is still applicable to individual segments, with

$$T = \begin{cases} -\dfrac{J^2}{2\sigma_A\kappa_A}x^2 + a_Ax + b_A, & 0 \le x < f, \\ -\dfrac{J^2}{2\sigma_B\kappa_B}x^2 + a_Bx + b_B, & f < x \le 1, \end{cases} \tag{21}$$

where $f = x_m/L$ is the volume fraction of phase A. Assume both temperature and electric potential are continuous at interface, $x = f$, and notice that energy flux is a constant, so that the heat flux is also continuous at the interface. Combining these two continuity conditions with two boundary conditions, we derive the following equation that can be solved for the integration constants:

$$\begin{bmatrix} 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 1 \\ f & 1 & -f & -1 \\ J(\varepsilon_A - \varepsilon_B)f - \kappa_A & J(\varepsilon_A - \varepsilon_B) & \kappa_B & 0 \end{bmatrix} \begin{bmatrix} a_A \\ b_A \\ a_B \\ b_B \end{bmatrix} = \begin{bmatrix} T_0 \\ T_1 + \dfrac{J^2}{2\sigma_B\kappa_B} \\ -\dfrac{J^2}{2\sigma_B\kappa_B}f^2 + \dfrac{J^2}{2\sigma_A\kappa_A}f^2 \\ \dfrac{(\varepsilon_A - \varepsilon_B)J^3f^2}{2\sigma_A\kappa_A} + \dfrac{(\sigma_A - \sigma_B)J^2f}{\sigma_A\sigma_B} \end{bmatrix}. \tag{22}$$

In a similar manner, the electric potential can be solved from Eq. (17) as

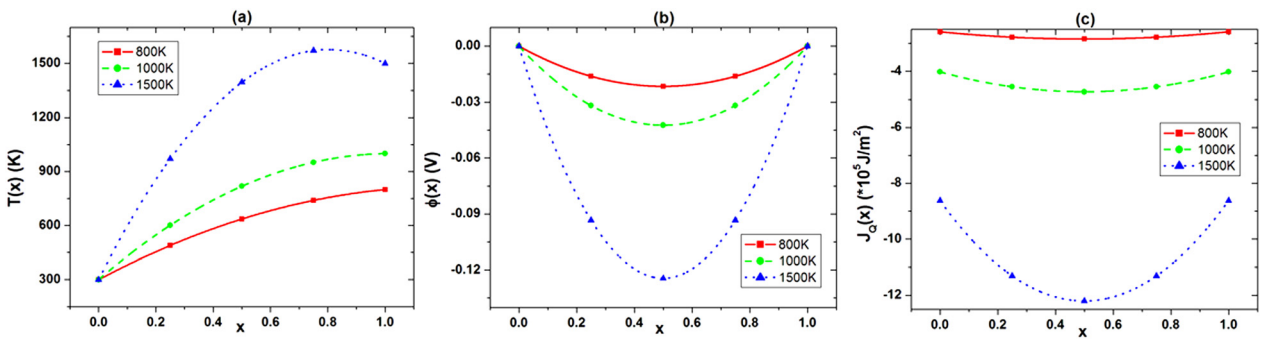

FIG. 2. (Color online) The distributions of temperature, electric potential, and heat flux in a homogeneous thermoelectric under an imposed temperature difference.

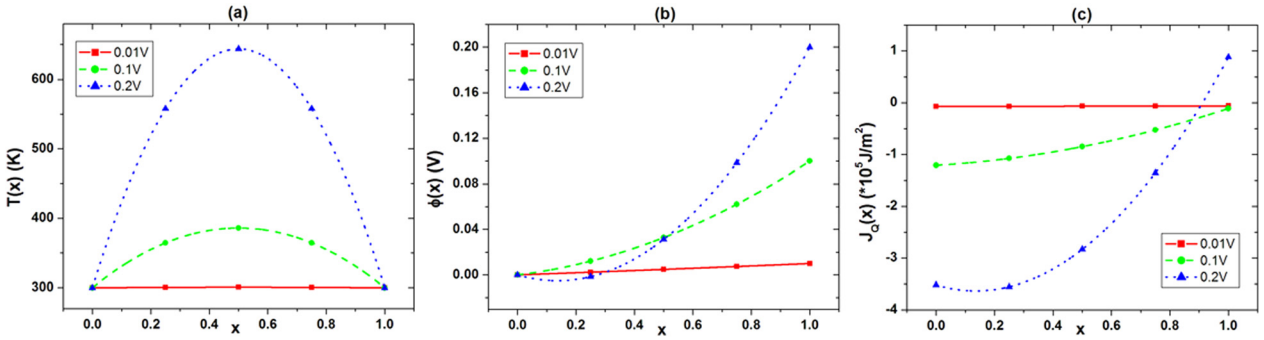

FIG. 3. (Color online) The distributions of temperature, electric potential, and heat flux in a homogeneous thermoelectric under an imposed electric potential difference.

$$\phi = \begin{cases} \dfrac{\varepsilon_A J^2}{2\sigma_A \kappa_A} x^2 - \left(\dfrac{J}{\sigma_A} + \varepsilon_A a_A\right) x + c_A, & 0 \le x < f, \\ \dfrac{\varepsilon_B J^2}{2\sigma_B \kappa_B} x^2 - \left(\dfrac{J}{\sigma_B} + \varepsilon_B a_B\right) x + c_B, & f < x \le 1, \end{cases} \tag{23}$$

with the integration constants c_A and c_B determined from the boundary conditions as

$$c_A = \phi_0, \tag{24}$$

$$c_B = \phi_1 + \frac{J}{\sigma_B} + \varepsilon_B a_B - \frac{\varepsilon_B J^2}{2\sigma_B \kappa_B}. \tag{25}$$

From the continuity of electric potential at the interface, the current density can then be solved as

$$J = -\frac{1}{(\varepsilon_B - \varepsilon_A) f(1-f)(\sigma_A(1-f) + \sigma_B f)} \times (\Psi_1 + \Psi_3 - \sqrt{\Psi_4}), \tag{26}$$

with

$$\Psi_1 = [\sigma_A(1-f) + \sigma_B f][\kappa_A(1-f) + \kappa_B f],$$
$$\Psi_2 = (\varepsilon_B - \varepsilon_A) f(1-f)\sigma_A \sigma_B,$$
$$\Psi_3 = \Psi_2(\phi_1 - \phi_0 + \varepsilon_B T_1 - \varepsilon_A T_0),$$
$$\Psi_4 = -2\Psi_2\{\Psi_1(\phi_1 - \phi_0) + [\sigma_A(1-f) + \sigma_B f][\kappa_A \varepsilon_B (1-f) + \kappa_B \varepsilon_A f](T_1 - T_0)\} + (\Psi_1 + \Psi_3)^2.$$

The heat flux J_Q can then be calculated by using Eq. (7). This completely solves the distribution of electric potential and temperature in a layered heterogeneous thermoelectric.

C. The effective thermoelectric properties

To describe the effective behavior of a heterogeneous thermoelectric, we define its effective thermoelectric properties through the following equivalency principle—given identical boundary conditions of temperature and electric potential, a heterogeneous thermoelectric with a set of effective thermoelectric properties should have identical current density and energy flux as a homogeneous thermoelectric with the same set of properties. With such equivalency, it is clear that the heterogeneous and homogeneous thermoelectrics can be exchanged under the specified boundary conditions. We examine the effective electric conductivity first. Consider a boundary condition of imposed electric potential difference only with $\Delta T = 0$, and compare the current density between homogeneous thermoelectric and layered medium, we conclude that the effective electric conductivity of the layered composite is given by

$$\sigma^*(\Delta\phi, \Delta T = 0) = \frac{J}{\phi_0 - \phi_1} = -\frac{J}{\Delta\phi}, \tag{27}$$

with the current density given by Eq. (26). Expanding current density J into Taylor series of $(\varepsilon_B - \varepsilon_A)$, we derive the effective conductivity of the layered thermoelectric to the first order of $(\varepsilon_B - \varepsilon_A)$,

$$\sigma^*(\Delta\phi) = \frac{\sigma_A \sigma_B}{\sigma_A(1-f) + \sigma_B f} - \frac{f(1-f)\sigma_A^2 \sigma_B^2 \Delta\phi(\varepsilon_B - \varepsilon_A)}{2(\sigma_A(1-f) + \sigma_B f)^2(\kappa_A(1-f) + \kappa_B f)}, \tag{28}$$

which clearly depends on the boundary condition in addition to the material constants of the constituents and the volume fraction, a characteristic distinct from linear medium. On the

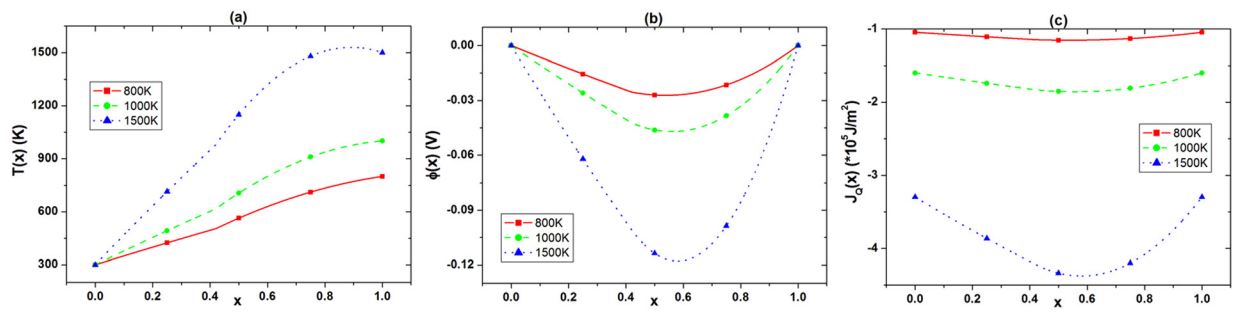

FIG. 4. (Color online) The distributions of temperature, electric potential, and heat flux in a layered thermoelectric under an imposed temperature difference.

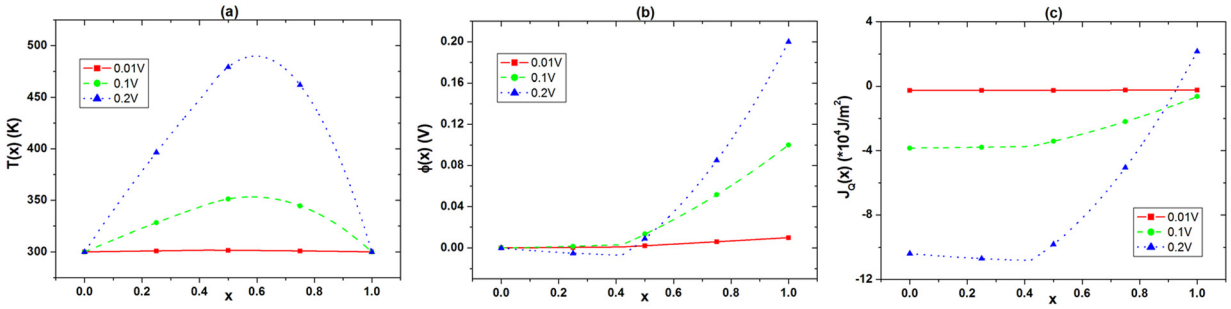

FIG. 5. (Color online) The distributions of temperature, electric potential, and heat flux in a layered thermoelectric under an imposed electric potential difference.

other hand, if we impose open-circuit boundary condition such that $J=0$, then the effective Seebeck coefficient can be derived as

$$\varepsilon^* = \frac{\kappa_A(1-f)\varepsilon_B + \kappa_B f \varepsilon_A}{\kappa_A(1-f)+\kappa_B f}, \tag{29}$$

while the effective thermal conductivity can be derived as

$$\kappa^* = -\frac{J_Q}{T_1 - T_0} = \frac{\kappa_A \kappa_B}{\kappa_A(1-f)+\kappa_B f}. \tag{30}$$

Although they do not appear to be dependent on the boundary condition explicitly, open- circuit boundary condition is implied implicitly. From the effective thermoelectric properties, the effective thermoelectric figure of merit is then defined as

$$Z^* = \frac{\sigma^* \varepsilon^{*2}}{\kappa^*}, \tag{31}$$

and we are interested in whether Z^* of the heterogeneous thermoelectric can be higher than both its constituents.

IV. RESULTS AND DISCUSSION

A. Homogeneous thermoelectric

We first consider a homogeneous thermoelectric Bi_2Te_3 of $L=0.01$ m, with its thermoelectric properties listed in Table I. Consider first that only a temperature difference is imposed, with $T_0=300$ K at cold end and three different temperatures of $T_1=800$, 1000, 1500 K at hot end. Notice that the hot end temperatures could be higher than the melting point of material, and these are used solely for mathematical argument. The distributions of temperature, electric potential, and heat flux are shown in Fig. 2, where it is observed that the nonlinearity in temperature distribution is small for small and modest temperature differences, but becomes significant under relatively large temperature difference. For $T_1=1500$ K, the maximum temperature occurs inside the medium, not at the end, due to Joule heating. Associated with such nonlinear distribution of temperature, substantial variation in heat flux is also observed under large temperature difference, though such variation decreases

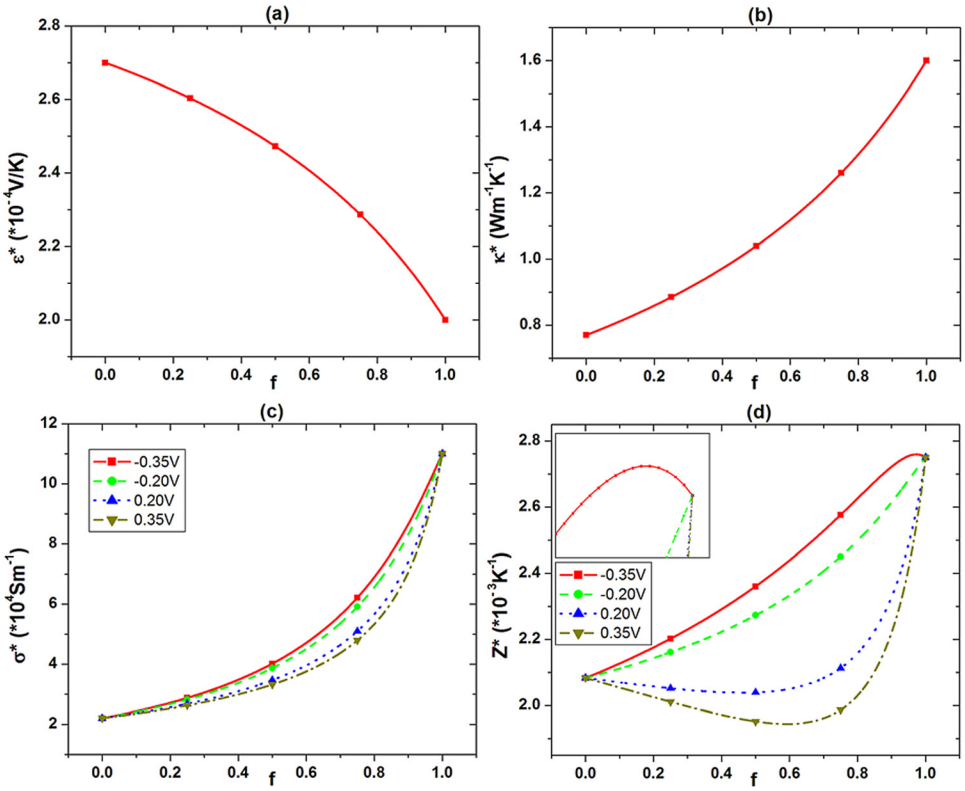

FIG. 6. (Color online) The effective Seebeck coefficient, thermal conductivity, electric conductivity, and figure of merit as functions of volume fraction.

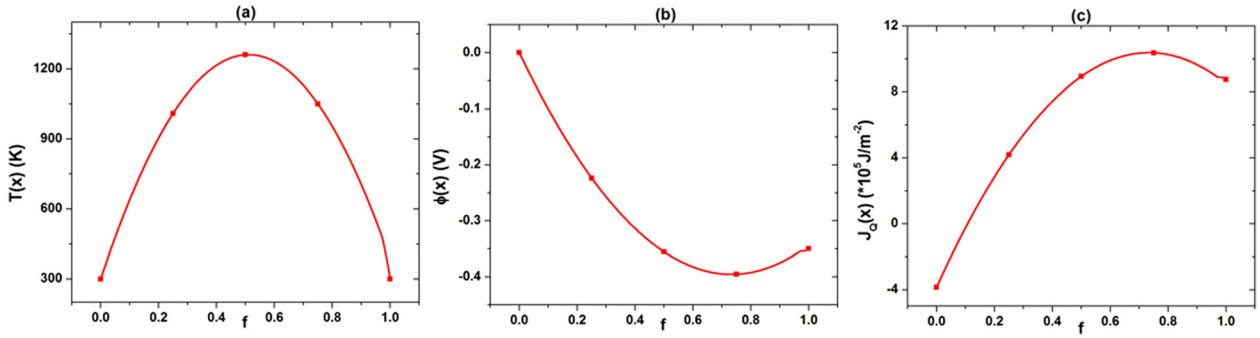

FIG. 7. (Color online) The distributions of temperature, electric potential, and heat flux at the maximum Z^* with $f=0.973$.

significantly when temperature difference is reduced. Regardless of the temperature difference, the distribution of electric potential is highly nonlinear, despite that both ends are imposed with same potential, resulting in an current density of $J=-1.1,\ -1.54,\ -2.64\times10^6\,\mathrm{A\ m^{-2}}$, respectively. On the other hand, if only an electric potential difference is imposed with $\phi_0=0$ and $\phi_1=0.01,\ 0.1,\ 0.2\,\mathrm{V}$, the corresponding results are shown in Fig. 3. It is observed that the nonlinearity in the distribution of electric potential becomes substantial under a relatively large potential difference, while the temperature distribution is highly nonlinear regardless of potential difference. Large variation in heat flux is also observed when the potential difference is large, and substantial temperature increase is observed inside the medium. These results suggest that neither temperature nor electric potential of thermoelectric materials satisfies Laplace equation, and the deviation is particularly large under large temperature or potential difference.

B. Layered thermoelectric

We then consider a layered thermoelectric consisting of Bi_2Te_3 as phase A and $Ag(Pb_{1-y}Sn_y)_mSbTe_{2+m}$ as phase B, with their thermoelectric properties listed in Table I. Notice that both phases have excellent Seebeck coefficient that are comparable to each other, yet Bi_2Te_3 has relatively high thermal conductivity, while $Ag(Pb_{1-y}Sn_y)_mSbTe_{2+m}$ has relatively low electric conductivity, not desirable for high thermoelectric conversion efficiency. We consider the distribution of temperature, electric potential, and heat flux in the layered thermoelectric first, with either temperature difference or electric potential difference imposed, identical to those considered in the last subsection. The volume fraction of Bi_2Te_3 is taken to be $f=0.42$, and the length remains to be 0.01 m. The corresponding results are shown in Figs. 4 and 5, respectively. The qualitative trends are similar to those observed in homogeneous Bi_2Te_3 under imposed temperature difference. In particular, it is noted that although the thermal conductivity of Bi_2Te_3 is more than 100% higher than that of $Ag(Pb_{1-y}Sn_y)_mSbTe_{2+m}$, temperature drops in these two materials appear to be similar. On the other hand, quite different trends are observed under imposed electric potential difference, where majority of electric potential drop occurs in $Ag(Pb_{1-y}Sn_y)_mSbTe_{2+m}$, which has much smaller electric conductivity.

The effective electric conductivity, Seebeck coefficient, thermal conductivity, and figure of merit of Bi_2Te_3-Ag $(Pb_{1-y}Sn_y)_mSbTe_{2+m}$ layered medium are shown in Fig. 6, with the following boundary conditions imposed:

$$T_0=T_1=300\mathrm{K},\quad \phi_0=0\mathrm{V},$$
$$\phi_1=\{-0.35,-0.2,0.2,0.35\}\mathrm{V}. \tag{32}$$

While the effective Seebeck coefficient and thermal conductivity only show slight deviation from the rule of mixture, as observed in Figs. 6(a) and 6(b), the nonlinear dependence of electric conductivity on volume fraction is significant, and it is sensitive to the voltage difference imposed on the boundaries, as shown in Fig. 6(c). This results in large difference in the effective thermoelectric figure of merit shown in Fig. 6(d). What is most interesting is that there is a peak, albeit small, in the effective thermoelectric figure of merit that exceed both constituents, at $f=0.973$, when the imposed potential difference is $\phi_1=-0.35$. This is significant, since it demonstrates that the effective thermoelectric figure of merit of a heterogeneous medium can be higher than both of its constituents, in contrast to previous studies. The distributions of temperature, electric potential, and heat flux for this particular layered structure under imposed boundary condition is given in Fig. 7, where substantial temperature increase, though still in the acceptable range, is observed. The distributions of temperature, potential, and heat flux are all highly nonlinear, which is essential for enhanced thermoelectric figure of merit. With higher potential difference imposed at boundary, higher enhancement in thermoelectric figure of merit will be obtained, though it may lead to unrealistic high temperature inside the medium.

V. CONCLUDING REMARKS

In conclusion, we have developed rigorous 1D analysis of layered thermoelectric medium, from which the effective thermoelectric properties have been established using an equivalent principle. It is found that the thermoelectric figure of merit of layered medium can be higher than both its constituents, and the key is to utilize the nonlinear distribution of temperature and electric potential inherent in thermoelectric transport.

ACKNOWLEDGMENTS

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