Introduction to thermoelectrics

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The main ideas in the theory of thermoelectrics are discussed. We discuss power generation, thermoelectric cooling, transport theory, the Seebeck coefficient, and phonon drag. © *2016 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution (CC BY) license (http://creativecommons.org/licenses/by/4.0/).* [http://dx.doi.org/10.1063/1.4954055]

Thermoelectric materials are semiconductors that have a strong coupling between the flow of heat and the flow of electricity.[1,2] These materials are important for two types of solid state devices:

- Solid state refrigerators, where the heat inside the refrigerator is removed by running a current through the walls. Usually one wants to cool below room temperature. Thermoelectric materials are needed that operate at or below room temperature.
- Power generation from waste heat: Generating electrical power from heat flow. Usually the heat flow is from a hot source above room temperature. Thermoelectric materials are needed that operate above room temperature.

After World War II, Ioffe[3] suggested the idea of a solid state thermoelectric refrigerator. There followed a world-wide search, led by industrial laboratories in many countries, to identify the best thermoelectric materials. Bismuth telluride (Bi_2Te_3) was found to be the best thermoelectric material at room temperature. Lead telluride (PbTe) was found to be the best material for generating electricity at higher temperatures.

The efficiency of a thermoelectric device, refrigerator or power generator, is a function of *the figure of merit $Z = \sigma S^2/K$*, where

- σ is the electrical conductivity in units of Siemens per meter;
- S is the Seebeck coefficient in units of volts per degree;
- K is the thermal conductivity in units of Watts per degree per meter; and
- the units of Z are inverse temperature, so that ZT is dimensionless, where T is the absolute temperature in units of Kelvin.

For any semiconductor material, impurities or alloys are intentionally added to reduce the thermal conductivity: lowering K increases ZT. For example, bismuth telluride thermoelectric material is usually an alloy such as $Bi_{2-x}Sb_xTe_{3-y}Se_y$. The choice of x and y varies with different manufacturing companies. With alloying, the FOM (figure-of-merit) of this material is about 1.0. That value is too small to make an efficient refrigerator, or an efficient power generator. That is also true of PbTe or other thermoelectric materials.

Research on thermoelectric materials, since Ioffe's time, has focused on discovering new materials with a higher FOM. Those earlier materials, Bi_2Te_3 and PbTe, are both binaries: composed of only two elements. Recent research has focused on growing new materials, not found in nature, with three, four, or more elements. A very few of them turn out to be very good thermoelectrics: the definition of "good" is having a FOM $ZT \geq 1$. Another avenue to improving the FOM has been nanostructuring.

A recent development, in the last decade, has been a very large increase in the manufacture of new thermoelectric devices. They are all either refrigerators or power converters. They nearly all
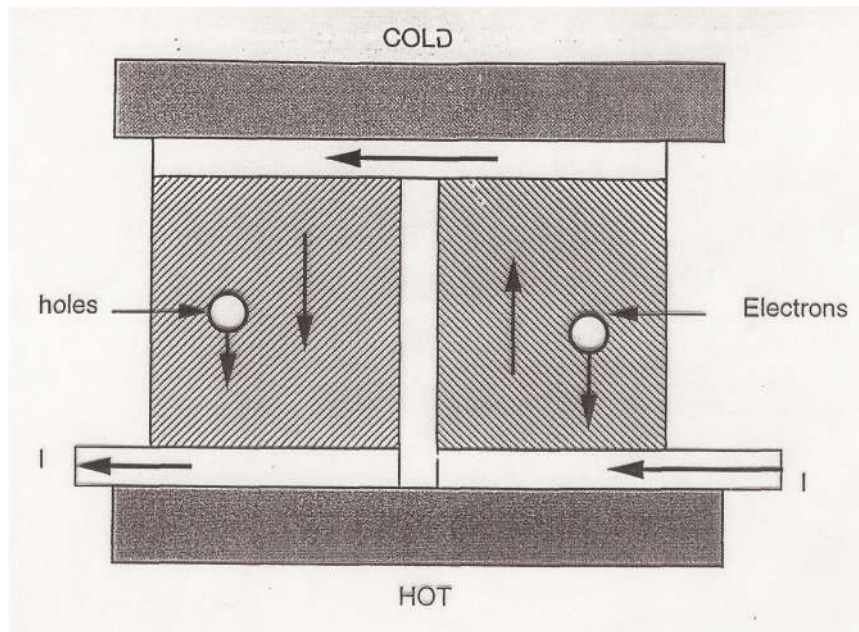
FIG. 1. A *pn*-couple operating in the refrigerator mode. The large arrows denote current flow. On the electron side, electrons flow in the opposite direction of current. On the hole side, they flow in the same direction as the current. Both electrons and holes move heat from cold to hot.

employ bismuth-telluride as the active thermoelectric material. The increased manufacturing is due entirely to better engineering. More and more uses are being found for these devices, where low efficiency is offset by other advantages such as portability or reliability.

Thermoelectric devices generally have large currents and small voltages. Therefore they are operated in series. The basic device is called a *pn*-couple, as shown in Figure 1, operating as a refrigerator. Current flows up one leg, say from hot to cold, and then down the other leg. One leg must be an *n*-type semiconductor, where the current and heat are carried by electrons. The other leg must be an *p*-type semiconductor, where the current and heat are carried by holes. Practical devices require an efficient thermoelectric of *n*-type semiconductors, coupled with an efficient thermoelectric of *p*-type material. The *pn*-couples are arranged into a module, as shown in Fig. 2.
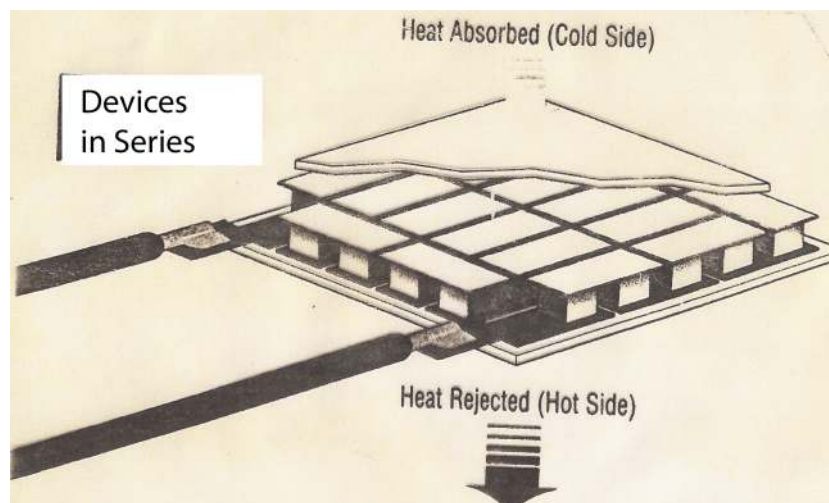

FIG. 2. Many *pn*-couples are arranged in series in a commercial module.

These modules are sold by many manufacturers and provide the cooling or power generation for commercial applications.

The device modeling is actually simple. Given a bar of material, with heat flowing down it, if one runs the electrical current in one direction the device operates as refrigerator. If the electrical current is run in the opposite direction, it operates as a power generator.

The industrial applications of thermoelectric are derived from two equations,[1,2]

$$J = -\sigma \left[\nabla(\bar{\mu}/e) + S\nabla T \right], \tag{1}$$

$$J_Q = TSJ - K\nabla T. \tag{2}$$

The first equation states that the electrical current (J) is produced by either a temperature gradient (∇T) or by the gradient in the electrochemical potential $\bar{\mu} = \mu + eV$. Earlier theories neglected the chemical potential μ in this equation, but it does belong.[4,5] The second equation states that a heat current (J_Q) is generated by an electrical current (TSJ) or by a temperature gradient. The latter term alone is Fourier's Law ($J_Q = -K\nabla T$) of heat conduction. The first term on the right, in Eq. (2), is the connection between heat flow and current flow. Obviously this connection needs a large value for the Seebeck coefficient.

Thermoelectric materials can be used to generate electrical power. The modeling of power generation uses Eq. (1). One has a bar of length L, where a temperature gradient produces a heat flow. One end is hot, at temperature T_h, while the other end is cold at temperature T_c, $\Delta T = T_h - T_c$. The electrical response behaves like a solar cell. There is an open circuit voltage, but as power is drawn from the device, the voltage is lowered. The efficiency of a power generator starts with the Carnot definition: the efficiency is the power P generated divided by the heat J_{Qh} drawn from the hot source.

We must manipulate Eq. (1). These devices are operated in a steady-state mode. Current flows down a bar, so the motion is one-dimensional. From the equation of continuity $\partial J_x/\partial x + \partial\rho/\partial t = 0$, one has $\dot{\rho} = 0$ for steady-state operation. Then J_x is a constant. Multiply Eq. (1) by $\rho = 1/\sigma$ and integrate over the length of the bar,

$$\rho J L = -\frac{1}{e}\Delta\bar{\mu} - S\Delta T. \tag{3}$$

The power developed is $P = -J\Delta\bar{\mu}/e$,

$$\eta = \frac{P}{J_{Qh}}, \tag{4}$$

$$P = J[\rho J L + S\Delta T], \tag{5}$$

$$J_{Qh} = ST_h J - \frac{K\Delta T}{L} + \frac{\rho J^2 L}{2}. \tag{6}$$

The last term in the heat flow comes from the Joule heating of the bar. Both P and J_{Qh} depend on the electrical current J. One finds the maximum efficiency, called the *optimum efficiency*, by maximizing: $0 = \delta\eta/\delta J$.

$$\text{OE} = \frac{\Delta T(\gamma - 1)}{\gamma T_h + T_c} \tag{7}$$

$$\gamma = \sqrt{1 + Z\bar{T}}, \quad \bar{T} = \frac{1}{2}(T_h + T_c) \tag{8}$$

As an example, operating over the temperature range $T_h = 400$ K, $T_c = 300$ K, with $Z\bar{T} = 1$, then OE = 5.0%, which is a very small efficiency for converting heat flow to electrical power. The ideal Carnot engine has $\eta = \Delta T/T_h$ which gives OE = 25% for the same operating temperatures. For comparison, steam turbine generators have an efficiency of about 20%.

An important use for thermoelectric power generation has been in space probes, by both Russia and the USA. Satellites that revolve around the Earth can generate electricity using solar cells pointed at our Sun. However, probes to distant bodies, such as Jupiter or Pluto, get too far from the Sun and solar panels are not adequate. *Radioisotope Thermoelectric Generators* (RTGs) are

used for power generation. The device has cylindrical geometry. The core of the cylinder is a material that generates heat. Typically it is a low density radioactive material such as an oxide of plutonium-238. Spontaneous nuclear fission generates heat that keeps the core area at a temperature over 1000 K. Thermoelectric elements surround this core. As the heat diffuses outward, it passes through the thermoelectric modules, which produce electrical power. Since plutonium-238 has a half-life of 88 years, the power is generated for decades. Russia also used RTG's to power remote stations in the arctic region. These often used strontium-90 as the radioactive material.

Many automobile companies are developing thermoelectric power modules for cars and trucks. The waste heat from the (i) exhaust and (ii) engine block are used to generate the electrical power needed to operate the vehicle. The major technical problem is the weight of these devices: both Bi-Te and PbTe are very dense. Newer, lighter weight materials, are needed for these applications to produce electricity in vehicles. The exhaust heat is around 700 K.

All of these applications are generating electricity with a source that operates over a wide temperature range. At the Jet Propulsion Laboratory (JPL) in the USA, RTG's often are designed to operate with $T_h = 1273$, $T_c = 473$ K. Every thermoelectric material has a high value of the FOM only over a narrow range of temperature, say ± 100 K from its peak value. Actual devices are segmented. The bar of length L is composed of several different materials bonded together in series along its length. The hot end has one material, while the colder end has another material. Perhaps a third material is the middle segment. Each material is chosen because it has a high FOM over the temperatures in its section of the bar. This is true for both the *n*-type and the *p*-type segments. So many different thermoelectric materials are need for the actual device. On the JPL website, they report that present technology creates devices with an efficiency of 12%, and a power density of 9 Watts per kilogram.

A very large possible use of thermoelectric power generators is to utilize waste heat from power plants, and factories, to produce additional electricity. These devices would use Bi-Te, the only efficient material near room temperature. A very large problem is that tellurium is rather scarce in the Earth's crust: there is not very much of it. There is not enough for such large scale operations. A zintl compound was recently reported to have a high FOM ($ZT \sim 1$) near room temperature,[6] but the material is unstable due to fast diffusion of alkali ions.[7] Other new materials are needed.

A thermoelectric device can presently cool to about $\Delta T = 100$ K below room temperature. The problem with going lower in temperature is that there are no materials with a high FOM at lower temperatures. Most cooling applications are for achieving a $\Delta T \sim 30$ K below room temperature. The efficiency is again given by the Carnot formula of the ratio of the heat removed from the cold side (J_c) divided by the power expended,

$$\eta(J) = \frac{J_c}{P} = \frac{ST_h J - \frac{K\Delta T}{L} - \frac{\rho J^2 L}{2}}{J(\rho J L + S\Delta T)}. \tag{9}$$

Again the current J is varied to obtain the maximum efficiency, which is called the *coefficient of performance*,

$$\text{COP} = \frac{\gamma T_c - T_h}{\Delta T(\gamma + 1)}, \tag{10}$$

since the best thermoelectric materials for refrigeration have $ZT \sim 1$ then $\gamma \sim \sqrt{2}$ and device efficiency is low. The ideal Carnot efficiency is obtained in the limit that $ZT \to \infty (\gamma \to \infty)$.

The FOM $Z = \sigma S^2/K$. It is increased by (i) reducing the thermal conductivity K, (ii) increasing the electrical conductivity σ, and (iii) increasing the Seebeck coefficient S. This process is limited by a law of nature. The thermal conductivity has a contribution from the heat flow by phonons K_p and by electrons $K_e, K = K_p + K_e$. The Wiedemann-Franz Law relates K_e to the electrical conductivity,

$$K_e = \frac{\pi^2}{3}\sigma T \left(\frac{k_B}{e}\right)^2, \tag{11}$$

where k_B is Boltzmann's constant, and e is again the electrical charge. Thus it is not possible to reduce K_e while increasing σ. The numerical prefactor of $\pi^2/3$ is for metals. In lightly doped semiconductors, the prefactor is two. Most thermoelectric materials are semiconductors, but they are heavily doped so that the formula for metals applies.

One can reduce the phonon part of the thermal conductivity. One method is by including nano-sized inclusions in the semiconductor that scatter the phonons more than electrons.[8–26]

All thermoelectric materials for applications are either doped with impurities or alloyed. This lowers the phonon part of the thermal conductivity, while increasing the electrical conductivity. Mott introduced the concept of critical doping density n_c which varies from semiconductor to semiconductor.[27] If a_D is the effective Bohr radius of an electron bound to a donor in that semiconductor, then the critical density is $n_c^{1/3} a_D = 0.25$. Pearson and Bardeen[28] first located this value for n-type silicon to be $n_c = 2 \times 10^{18}$ cm^{-3}.

- If the density of donors is $n_e < n_c$, the semiconductor is an insulator at low temperature.
- If the density of donors is $n_e > n_c$, the semiconductors has nonzero resistivity at low temperature.
- All materials used in thermoelectric application have $n_e > n_c$. They behave as low density metals.

Most thermoelectric devices operate at temperatures near room temperature or higher. In this regime of temperature, the Seebeck coefficient can be separated into two terms,

$$S = S_e + S_{pd}, \tag{12}$$

where S_e is the contribution of electrons (or holes) during their motion in the conduction (or valence) band. The phonon-drag term S_{pd} is discussed below. Three of the transport coefficients can be evaluated by solving the Boltzmann equation for the electron lifetime $\tau_e(\mathbf{k})$ and then integrating over the band electrons,

$$\sigma = e^2 \int dE \Sigma(E) \left(-\frac{df}{dE} \right), \tag{13}$$

$$\sigma S_e = \frac{e}{T} \int dE \Sigma(E)(E - \mu) \left(-\frac{df}{dE} \right), \tag{14}$$

$$K_e = \frac{1}{T} \int dE \Sigma(E)(E - \mu)^2 \left(-\frac{df}{dE} \right), \tag{15}$$

$$f = \frac{1}{e^{(E-\mu)/k_B T} + 1}, \tag{16}$$

$$\Sigma(E) = \frac{2}{3} \int \frac{d^3 k}{(2\pi)^3} v_k^2 \tau_e(\mathbf{k}) \delta \left(E - E_c - \frac{\hbar^2 k^2}{2m_e} \right), \tag{17}$$

where m_e is the effective mass of the electron in the conduction band, and v_k is the electron velocity in the conduction band. An identical formula is used for hole motion in valence bands. E_c is the minimum energy of the conduction band, or the maximum energy of the valence band. Note that S is the ratio of two integrals. Mahan and Sofo[29] showed that the maximum FOM occurred when $\Sigma(E) = \Sigma_0 \delta(E - \mu - b)$, where $b = \pm 2.2 k_B T$, and Σ_0 is any constant. The delta function density of states can be achieved by having narrow bands, such as in rare-earth materials, or by Fano resonances in impurity states.

The Seebeck coefficient has two terms: (i) S_e due to electrons in conduction or valence bands, and (ii) S_{pd} due to phonon-drag. The phonon-drag term in metals and insulators is quite different. In metals, the majority of the heat current is carried by electrons. As the electrons drift through the crystal, they drag along the phonons. In insulators, the majority of the heat is carried by phonons. As the phonons drift through the crystal, they drag along the few electrons. Both cases are due to the interaction between the electrons and the phonons.

In metals, the electrons involved in the transport of heat or electricity are at the Fermi surface. Only electrons of thermal energy $k_B T$ within the Fermi surface are involved. The standard theory

TABLE I. Si samples from Geballe and Hull. Sample dimensions are width (w), height (h), and length (ℓ). Distances in cm, and donor and acceptor densities in cm^{-3}.

No.	w	h	ℓ	N_D	N_A
537	0.470	0.47	3.8	$3.70 \cdot 10^{14}$	$0.9 \cdot 10^{14}$
131	0.086	0.16	1.1	$2.75 \cdot 10^{14}$	$1.0 \cdot 10^{14}$

for phonon-drag was worked out by Bailyn.[30] However, metals are poor thermoelectric materials since they have a small Seebeck coefficient, so this theory is of little interest in thermoelectrics.

In insulators, the heat is carried by phonons. One method of measuring the Seebeck coefficient is using a bar of length L. A small temperature gradient is put along the length of the bar. The ends are electrically insulated, so no current can flow. Then the voltage ΔV is measured and is found to be proportional to the difference in temperature ΔT of the two ends of the bar. At most temperatures, the Seebeck coefficient is the ratio $S = -\Delta V/\Delta T$. However, at very low temperatures, there is an important correction to the Seebeck coefficient due to space-charge effects.[31] Since the ends of the bar are electrically insulating, electrical current cannot flow. The force the phonon current exerts on the electrons must be cancelled by an additional voltage to prevent the electrons from moving. That is the source of phonon-drag.

Early experiments by Geballe and Hull[32] on lightly doped crystals of Si and Ge showed that S_{pd} depended upon the size of the sample. Two samples are described in Table I, and the data are shown in Figure 3. The phonon mean-free-path λ is an important ingredient in the theory of phonon-drag. If the phonons do not scatter from other phonons or isotope fluctuations, they can go entirely across the sample and scatter from the walls of the sample. The larger the sample, the
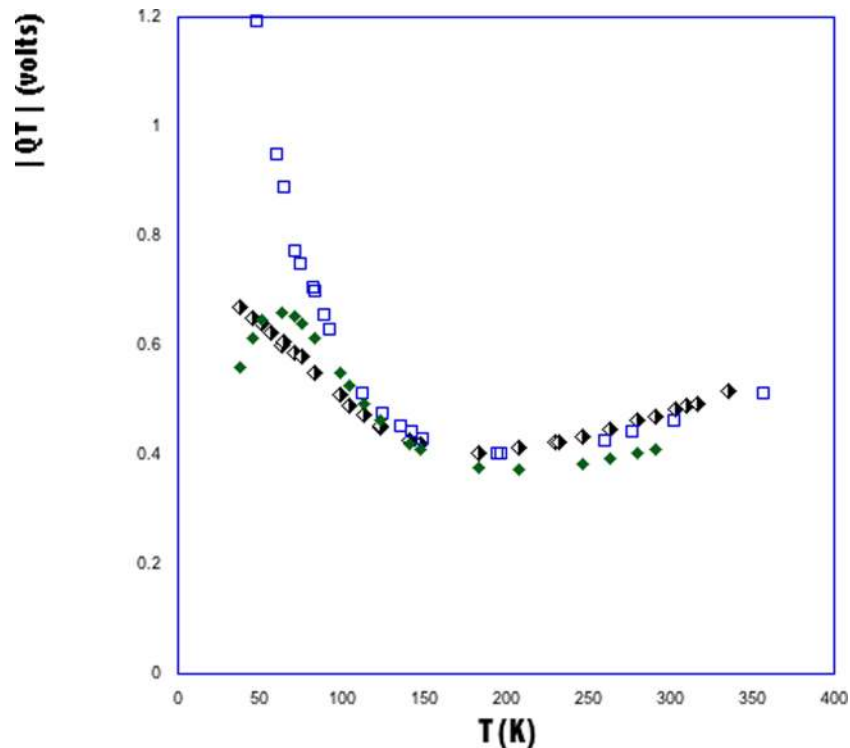

FIG. 3. Seebeck coefficient of silicon. Squares are sample number 537 of Geballe and Hull,[32] and half-black diamonds are sample no. 131. Black diamonds are theory of Mahan *et al.*[33] for sample no. 131. Q is Seebeck coefficient, and theory and experiment are plotted as $|QT|$ which has the units of volts.

longer the average value of λ. Thus, S_{pd} is not an intrinsic number of the material, but depends on the dimensions of the sample. Perhaps this is not surprising, since it has been known for a long time that the thermal conductivity of these samples also depends upon sample dimensions for the same reason. Figure 3 shows the experimental results of two samples of Geballe and Hull on lightly doped silicon; their measurements are squares for sample no. 537 and half-black diamonds for sample no. 131. The solid diamonds are the theory of Mahan et al.[33] for sample no. 131, which is in good agreement with the measurements. Since silicon is so well understood, the theory has no adjustable parameters, yet the fit to the data is very good. The data of Geballe and Hull show that two samples with the nearly same density of impurities, but different sample thicknesses, had very different values for the Seebeck coefficient. The main difference is in their dimensions. The larger sample no. (537) has the higher values of the phonon-drag contribution to the Seebeck coefficient. Experimentalist finds it frustrating that the Seebeck coefficient depends upon the size of the sample and is not an intrinsic parameter.

A very active area of current research is to improve the FOM by either (i) having pores in the sample or (ii) filling the pores with another material.[9–26] Long ago Worlock[8] showed that this lowered the thermal conductivity due to phonons. What does it do to the other transport coefficients? Most of the recent work has been in thin films, where the transport is mostly two dimensional. Many experiments, and many theoretical calculations, have been done for a film with a periodic arrangement of cylindrical holes.[34–40] These have largely focused on the thermal conductivity, which is reduced by the presence of the holes. One paper measured the other transport coefficients.[35] These theories have mostly been large-scale computer simulations involving many atoms.

Most calculation use molecular dynamics.[41–45] They construct a spring and mass model of the vibrations of the solid and follow the motions of the atoms using Newton's Laws. At each time step, they calculate the heat current $\vec{J}_Q(t)$ and then correlate it with itself. The thermal conductivity is

$$K = \frac{1}{3\Omega k_B T^2} \int_0^\infty dt \langle \vec{J}_Q(t) \cdot \vec{J}_Q(0) \rangle, \tag{18}$$

$$\vec{J} = \sum_i \mathbf{v}_i \varepsilon_i + \frac{1}{2} \sum_{i \neq j} \mathbf{r}_{ij} (\mathbf{v}_i \cdot \vec{F}_{ij}) \tag{19}$$

$$+ \frac{1}{6} \sum_{ijk} (\mathbf{r}_{ij} + \mathbf{r}_{ik}) \times (\vec{F}_{ijk} \cdot \mathbf{v}_i). \tag{20}$$

Some workers omit the last term (3-body forces). These calculations take large amounts of computer time.

In 1892, Lord Rayleigh[46] published a paper on the electrical and thermal conductivity of porous materials. His model of porosity was a periodic arrangement of pores: cylinders in two dimensions, and spheres in three dimensions. He provided an analytic theory of conduction in these systems. A solid with a periodic arrangement of pores is now called an inverse opal. Mahan et al.[47] applied Rayleigh's theory to the thermoelectric properties of inverse opals. Rayleigh defined a factor $p(f)$ which gave the reduction in thermal conductivity with increased porosity f. Considering Eq. (1) and setting $\nabla T = 0$, the electrical conductivity of the inverse opal $\sigma_{io} = p(f)\sigma$. Again, in Eq. (1) setting $\nabla(\bar{\mu}) = 0$ gives that $\sigma_{io} S_{io} = p(f)\sigma S$. Dividing these two results gives $S_{io} = S$: the Seebeck coefficient is unchanged for a solid film compared to a film with periodic holes. This result was also found in the numerical calculations of Ref. 35. Since the thermal conductivity $K_{io} = p(f)K$, then the thermal conductivity and the electrical conductivity scale with the same factor $p(f)$. This was also found in the calculations of Ref. 35. Therefore the FOM is the same in the film with holes, as in the original film with no holes. There is no thermoelectric advantage to punching holes in the film. This conclusion also applies to punching periodic spherical holes in a three dimensional materials. Here the Rayleigh theory has not been tested.

The Rayleigh theory comes with caveats. The surfaces of the holes should be perfect: there can be no reconstruction of the bonding nor any passivation. This may be hard to achieve in practice. Furthermore, Rayleigh's theory is classical and is based upon diffusion. When the holes have the dimensions of nanometers, the conduction excitations, electrons or phonons, will have important

diffraction effects, which negates Rayleigh's theory. It will be interesting to see how this changes the classical prediction.

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