High-Efficiency Thin-Film Superlattice Thermoelectric Cooler Modules Enabled by Low Resistivity Contacts

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V-telluride superlattice thin films have shown promising performance for on-chip cooling devices. Recent experimental studies have indicated that device performance is limited by the metal/semiconductor electrical contacts. One challenge in realizing a low resistivity contact is the absence of fundamental knowledge of the physical and chemical properties of interfaces between metal and V-telluride materials. This study presents a combination of experimental and theoretical efforts to understand, design, and harness low resistivity contacts to V-tellurides. Ab initio calculations are used to explore the effects of interfacial structure and chemical compositions on the electrical contacts, and an ab initio based macroscopic model is employed to predict the fundamental limit of contact resistivity as a function of both carrier concentration and temperature. Under the guidance of theoretical studies, an experimental approach is developed to fabricate low resistivity metal contacts to V-telluride thin film superlattices, achieving a 100-fold reduction compared to previous work. Interfacial characterization and analysis using both scanning transmission electron microscopy and energy-dispersive X-ray spectroscopy show unusual interfacial morphology and the potential for further improvement in contact resistivity. Finally, the improved contacts are harnessed to realize an improved high-performance thermoelectric cooling module.

V-telluride materials such as Bi₂Te₃ and Sb₂Te₃ have been of interest for thermoelectric applications[1–5] and recently for their topological insulator properties.[6–9] In particular, thin films of such materials have shown excellent performance for on-chip cooling,[10–13] with even better performance predicted theoretically.[14] To realize the full promise of these materials, it is necessary to eliminate parasitics that can limit the performance. As such, metal contacts play an important role for thin-film thermoelectric (TE) devices, especially in high heat-flux applications (e.g., chip cooling) where low contact resistivity (ρₐ) is critical to device performance.[15–17] Recent work[18] has demonstrated low electrical contact resistivity ρₐ in the range 1–2 × 10⁻⁶ Ω cm² in TE modules based on (Bi,Sb)₂Te₃ superlattices using the evaporation of Cr/Ni/Au to fabricate metal electrodes. For thin-film thermoelectric modules with the TE thickness <2 μm, further reduction of ρₐ to 10⁻⁸ Ω cm² is needed for the contact resistivity to be a small fraction of the resistivity of the thermoelectric element itself.[18]

Creating such low contact resistivities is challenging from a fabrication perspective,[19] but also because little is known about the fundamental properties of metal contacts to these materials. For example, even basic properties such as the atomic and electronic structure of the metal/TE interface are largely unknown. This makes it difficult to optimize the contact resistivity and to establish the fundamental limits[20] that are possible. To address this challenge, we present an integrated theoretical and experimental effort toward understanding the limits of low-ρₐ in realistic metal contacts to advanced TE materials. We present a new multiscale theoretical approach combining ab initio calculations and continuum mesoscopic models to investigate the structural, electronic, and transport properties of electrical contacts to novel TE materials used in thin-film, superlattice V-telluride devices. We show that the nature of these semiconductor materials leads to unusual contact properties, such as strong n-type doping near the interface and interfacial atomic dipoles that completely determine the band bending. We predict that significant improvement over previously reported experimental data is possible, and we present new experimental data that demonstrate a 100-fold reduction in contact resistivity. Detailed atomistic spatially resolved measurements of the new contacts show that additional improvement should be possible. Importantly, we demonstrate that the reduction in contact resistivity can be harnessed to improve the thermoelectric efficiency of cooling modules.

To understand the electrical properties of contacts to (Bi,Sb)₂Te₃ materials and their realistic low ρₐ limit we carried out a series of large scale ab initio calculations of the Sb₂Te₃–Cr
interface illustrated in Figure 1a. Cr was chosen because it has shown good adhesion properties to V-telluride materials,[21,22] while Sb\textsubscript{2}Te\textsubscript{3} possesses lower electrical resistivity than Bi\textsubscript{2}Te\textsubscript{3}, possibly implying a lower contact resistivity. In addition, we also chose to simulate the p-type doped TE material since experimentally[18] it has higher contact resistivity compared to the n-type material, and thus greater gains might be expected from improvement of that contact. While metal interdiffusion at metal/TE interfaces is known to arise[23] and Cr is a common dopant[24,25] for Sb\textsubscript{2}Te\textsubscript{3}, such effects are beyond the scope of this work, and we therefore focus on the clean interface in our simulations.

We use ab initio calculations to simulate the details of the contacts. Ab initio calculations are necessary because the band alignment at metal/semiconductor interfaces is determined by the atomistic details of the interface. For example, it is well known that Fermi level pinning due to interface states leads to Schottky barriers even when the band alignment should give an Ohmic contact. The importance of ab initio calculations in determining contact resistivity has been demonstrated previously in the case of conventional semiconductors.[26–29] We perform the calculations using the Vienna ab initio simulation package (VASP) code[30,31] to simulate realistic low-strained metal/TE interfaces using large supercells (thousands of atoms), which allows us to incorporate antisite defects that yield p-type free carrier concentrations similar to those expected to maximize cooling performance (see the Supporting Information for additional details of the computation). Figure 1b shows the calculated electronic structure of bulk undoped Sb\textsubscript{2}Te\textsubscript{3}. We find a quasidirect bandgap of 100 meV with both the valence band maximum (VBM) and conduction band minimum (CBM) having sixfold valley degeneracy. From the calculated bandstructure we also calculated the density of modes,[32] from which hole and electrons transport effective masses of ≈ 0.25 m\textsubscript{0}, where m\textsubscript{0} is the free electron mass, were obtained perpendicular to the Sb or Te layers. Using a dense k-point grid we have also calculated the carrier concentration (holes and electrons) as a function of position of Fermi level, as shown in Figure 1c.

To simulate the p-doped semiconductor we created two antisite defects (Te replaced by Sb) in our supercell. At this density of antisite defects the Fermi level is located in the valence band, with no localized defect states in the bandgap. Given that Te has one more valence electron than Sb, each antisite defect leads to one free hole in the degenerate doping regime. We thus realize a total doping concentration expected to lead to optimal TE properties, namely, p = 6.5 × 10\textsuperscript{19} holes cm\textsuperscript{-3}. Based on the carrier concentration results of Figure 1c, the Fermi level lies 75 meV below the VBM for this doping level, as indicated with the dashed line in Figure 1b.

The simplest model for the band alignment at the metal/semiconductor interface is the direct alignment due to the metal work function \(\phi\) and the semiconductor electron affinity \(\chi\). We thus calculated these two quantities from ab initio (see the Supporting Information for details), finding \(\phi = 4.4\) eV and \(\chi = 4.7\) eV, as shown in Figure 1a. This large \(\chi\) implies that a number of metals will naturally prefer to form n-type contacts...
to Sb$_2$Te$_3$; in addition, the large electron affinity will lead to electron transfer from the metal to the semiconductor, which will dope the semiconductor n-type near the interface. This has important consequences for contacts to p-type materials, since we can expect a p–n junction to form near the contact which will increase the contact resistivity.

Of course, the local atomic interactions at the interface can change the position of the Fermi level at the interface expected from direct band alignment. These important effects are revealed by our ab initio simulations of the full interface. For example, we find significant atomic disorder of Sb$_2$Te$_3$ near the interface with Cr, while Cr stays quite ordered (see Figure 1a) (while the Seebeck coefficient in the disordered region may be changed compared to the rest of the TE material, the device Seebeck coefficient will not be strongly affected because the device Seebeck is an average over the thin film thickness, and the disordered region is only a very small fraction of the film thickness).

The significant disorder at the interface is also accompanied by a strong chemical interaction between the Cr and the Sb$_2$Te$_3$. Figure 1d shows the charge density difference between the Sb$_2$Te$_3$/Cr system and the isolated Sb$_2$Te$_3$ and Cr subsystems (having the same relaxed atomic positions as in the contact case). A large dipole with positive charge on the semiconductor forms within a few Angstroms from the metal interface. This dipole is opposite to the dipole formed due to the difference between the metal workfunction and the semiconductor electron affinity. The large values of the charge density difference reflect the strong interaction between metal and semiconductor atoms with chemical bonds at the interface. Farther away (more than 1 nm) from the metal interface the charge density difference acquires significant components from free carriers, which are responsible for the band bending as we shall see next. We note that the chemical interaction effect discussed here should be general and should be considered when studying the properties of other metal/semiconductor interfaces.

To further understand the influence of the metal on the electronic bandstructure of Sb$_2$Te$_3$, we calculated the spectral function projected on each of the six quintuplets included in the supercell (the term quintuplet refers to the group of five atomic planes that form one Sb$_2$Te$_3$ unit, see Figure 1a). The results are shown in Figure 2a. We find a completely distorted electronic structure in the first quintuplet, with states seen across the energy range and no discernible bandgap. The bulk semiconductor states become more clearly identifiable starting with the second quintuplet. In particular, we can identify the Fermi level positioned at the edge of the conduction band, indicating that even though the semiconductor is p-type, there is strong n-type doping near the interface due to the metal. One can see that the VBM and CBM start bending up as one moves further away from the interface, but with the VBM still below the Fermi level even in the 5th quintuplet. As we shall see, this is an artifact of the limited k-point sampling that we can employ in our full ab initio calculations. A similar picture holds for the

![Figure 2](image-url). Electronic structure and band bending. a) Projected spectral function on each quintuplet layer for p-doped interface of Sb$_2$Te$_3$ and Cr (the leftmost panel corresponds to the 1st quintuplet layer near Cr, the rightmost to the 6th one near vacuum). b) Band bending in Sb$_2$Te$_3$ near the Cr interface calculated from first principles using a k-point sampling $2 \times 1 \times 1$ compared with the ones obtained via macroscopic modeling using the corresponding ab initio charge carrier densities. c) Band bending as well as VBM and CBM band profiles obtained via macroscopic modeling using the converged ab initio charge carrier densities shown in Figure 1c. Also shown is the contribution to the band bending potential from chemical dipoles near the interface.
undoped Sb$_2$Te$_3$ contacted with Cr, except that here the VBM and CBM are generally aligned about 50–100 meV lower in energy. We also note that the 6th quintuplet is next to vacuum in the simulations, hence the appearance of the topological surface states (Dirac cone) in the projected bandstructure.

We obtain the band bending $V(r)$ for the Sb$_2$Te$_3$/Cr system by subtracting the electrostatic potential of the semiconductor and metal subsystems from the one of the full metal/semiconductor system. We do not find an overall tunnel barrier at the interface as short bond-lengths between metal and semiconductor atoms allow for conduction paths near the Fermi level due to free carrier accumulation in the valence/ conduction band. In each case we have anchored the potential with respect to the Fermi level such that at $d = 5$ nm the VBM corresponds to the value seen in the projected bandstructure (here we have assumed a bandgap of 130 meV as deduced from bulk calculations with the same k-point sampling).

Having obtained the detailed properties of the interface from ab initio calculations, we employ a macroscopic model that captures these details while allowing the modelling of band bending far away from interface together with the prediction of the contact resistivity (although fully ab initio approaches to obtain the contact resistivity have been developed, the size and complexity of this system prohibits the use of such approaches.) Since our novel multiscale approach is general, we expect it to be applicable to a broad range of metal/semiconductor interfaces. As discussed in the Supporting Information, we self-consistently solve for the continuous charge distribution and electrostatic potential for spatially uniform p-type doping. We use a parabolic band approximation with effective mass obtained from the ab initio simulations. We also include the atomistic charges obtained from the ab initio simulations which arise from the strong chemical interaction at the interface. As shown in Figure 2b the combined ab initio/macromolecular approach can be used to predict the band bending in excellent agreement with the fully ab initio one in both the doped and undoped cases. Our approach allows to converge the band bending up to large distances away from the interface, as well as to consider arbitrary doping levels. The converged result for a doping of $6.5 \times 10^{19}$ holes cm$^{-3}$ is shown in Figure 2c. Remarkably, we find that the band bending near the interface is mostly dominated by the chemical dipole charges, i.e., the p-type doping matters little near the interface and in fact the semiconductor is strongly n-type up to 3 nm away from the interface. Moving away from the interface, the chemical dipole charge decreases and quickly becomes smaller than the free carrier charge due to doping, leading to a band bending determined by the doping (as can be deduced from the leveling of the electrostatic potential due to just the chemical dipoles $V_{\text{dipole}}$ as shown in Figure 2c). The semiconductor becomes p-type at 5 nm from the interface. Thus, hole transport takes place through an effective n–p junction inside the semiconductor: holes injected from the contact are thermally excited over a Schottky barrier equal to the semiconductor bandgap (thermionic emission), or tunnel through a barrier of width $W = 2$ nm (thermionic field emission). This situation is illustrated in Figure 3a.

We calculate the contact resistivity using conventional models for tunneling and thermionic emission as detailed in the Supporting Information, with the band bending for different doping obtained from the macroscopic model benchmarked with the ab initio simulations. We find that the barrier width is inversely proportional to the doping level (see the Supporting Information), with negligible dependence on temperature. Other parameters are obtained directly from the ab initio simulations: the tunneling effective mass and barrier height. Figure 3b shows the calculated contact resistivity as a function of p-type doping for several temperatures. Three transport regimes can be identified: at high doping, the tunneling width is small and direct tunneling near the Fermi level dominates transport. At intermediate doping we find that the main contribution comes from thermionic field emission. Finally, at low doping hole transport takes place via thermionic emission,
hence the stronger dependence of $\rho_C$ on temperature. At room temperature and for doping levels similar to those realized experimentally,[18] (namely 3–5 × 10¹⁹ holes cm⁻²) we find values of $\rho_C$ in the range 1–3 × 10⁻⁸ Ω cm², up to two orders of magnitude less than previously reported experimentally.[18] In addition to establishing a lower limit for the contact resistivity of realistic interfaces, our results also establish a benchmark to which future contact design modeling can be compared. Our results also provide the quantitative dependence of the contact resistivity on doping, which will have to be taken into account when optimizing the doping of future TE thin film devices.

Under the guidance of theoretical predictions we have tested experimentally the possibility of low $\rho_C$ by utilizing an improved technique to fabricate electrical contacts to V-telluride materials. The samples to be metallized were a p-type Bi₂Te₃/Sb₂Te₃ thin-film superlattice and an n-type Bi₂Te₃–Se₃ thin-film alloy. Both types of films were grown epitaxially via metal–organic chemical vapor deposition on semi-insulating GaAs substrates.[2] These materials were loaded into a vacuum chamber and pumped down to a background pressure of less than 9 × 10⁻⁷ Torr. They were then back-sputtered using an Ar⁺ plasma (power = 350 W; time = 10 min; pressure = 20 mTorr; Ar flow rate = 50 sccm) in an effort to treat the sample surfaces in situ. Finally, the desired metal stacks were deposited onto the samples via sputtering with an Ar⁺ plasma. Previous work has shown that similar processes resulted in decreased contact resistivity for Ni and Co contacts to Bi₂Te₃.[36] Schematics of the metal stacks can be found in the Supporting Information. This surface treatment and metallization process can also be used to fabricate contacts onto other TE materials, such as PbTe and skutterudites, although the effectiveness of this technique on such materials is unknown.

The measured values of specific contact resistivity for the sputtered metallizations were obtained via the circular transfer length method (C-CTLM).[17] C-CTLM calls for the measurement of electrical resistance values of a metallized thin film material across a series of annular gap spacings as a function of gap width (see the Supporting Information for details of the geometry). As the gap width approaches zero, the measured electrical resistance approaches the value of the contact resistance. Representative resistance versus gap spacing data for both p-type and n-type V-telluride materials are given in Figure 4. The slope of the resistance versus gap spacing line is the sheet resistance ($R_s$) multiplied by a geometric constant. The intercept is $-2 \times 10^{-8}$ Ω cm² for n-type contacts and $-7 \times 10^{-9}$ Ω cm² for p-type contacts.

The specific contact resistivity values obtained for a number of sputtered metallizations, on both p-type and n-type V-telluride materials, are given in Table 1. Missing data in Table 1 means the calculated value resulted in a negative contact resistivity or transfer length, which is an unphysical result and therefore not reported. The traditional interpretation of this unphysical result is that the particular contact resistivity (and therefore transfer length) being measured is small compared to experimental variance, implying a very small value of the contact resistivity—likely below $10^{-8}$ Ω cm². This data indicate that the Ar⁺ backsputter/sputtered metallization process can yield lower $\rho_C$ values than the standard evaporation or plating metallization processes which are currently used in state-of-the-art thin-film V-telluride devices.[18] Indeed, we measured the properties of similar contact prepared without Ar sputtering and consistently find contact resistivities around $10^{-5}$ Ω cm² for p-type contacts and $10^{-3}$ Ω cm² for n-type contacts, as shown in the Supporting Information.

To shed further light on the impact of Ar⁺ backsputtering on the contact properties, we performed a scanning transmission electron microscopy (STEM) analysis of the Cr/p-type (Bi,Sb)₂Te₃ material. TEM specimens were prepared by focused ion-beam liftout techniques and were analyzed using a Titan G2 80–200 instrument (FEI, Hillsboro OR, USA) operated at 200 keV and equipped with a SuperX window-less X-ray detector array for energy dispersive x-ray spectroscopic (EDS) analysis. EDS spectrum images were postprocessed using multivariate statistical analysis methods using the Sandia-developed Automated eXpert Spectrum Image Analyzer (AXSIA) software.[19]

Figure 5 shows STEM images and an EDS spectrum image of a sputtered Cr metal contact on a p-type (Bi,Sb)₂Te₃ superlattice.[12] The Cr layer here is nominally 40 nm thick and is topped with an overlay of Au. The grains in the Cr layer exhibit a columnar morphology and, as can be seen in the corresponding EDS spectrum image, some Au appears to have

![Figure 4](image-url)

**Figure 4.** Resistance versus gap spacing data for a) p-type and b) n-type V-telluride material that has been metallized with Ti via sputtering after an in situ Ar⁺ ion backsputter treatment. The black markers represent measured electrical resistance values. The red and blue markers represent $+2\sigma$ and $-2\sigma$ values, respectively. The lines represent least-squares fits to the data.
Table 1. Transfer length and specific contact resistivity values for a series of metallizations for both p- and n-type V-telluride materials. An error of ±2σ has been applied to the data and is also given in the table.

| Type | Sputtered metallization | Transfer length, \( L_T \) [μm] | Specific contact resistivity, \( \rho_c \) [Ω cm²] |
|------|-------------------------|----------------------------------|-----------------------------------------------|
|      |                         | +2σ \( L_T \) | \( L_T \) | −2σ \( L_T \) | +2σ \( \rho_c \) | \( \rho_c \) | −2σ \( \rho_c \) |
| P    | Au                      | 2.06 | 0.46 | −1.14 | 1.6 × 10⁻⁷ | 2.6 × 10⁻⁸ | – |
|      | Cr/Au                   | 0.84 | 0.53 | 0.21  | 4.9 × 10⁻⁸ | 2.7 × 10⁻⁸ | 4.4 × 10⁻⁹ |
|      | Cr/Ti/W/Au              | 1.08 | 0.26 | −0.57 | 5.2 × 10⁻⁸ | 9.3 × 10⁻⁸ | – |
|      | Ti/Cu                   | 1.02 | 0.59 | 0.15  | 7.9 × 10⁻⁸ | 3.9 × 10⁻⁸ | – |
|      | Ti/TiW/Cu               | 1.19 | 0.02 | −1.15 | 1.0 × 10⁻⁸ | 1.3 × 10⁻¹⁰ | – |
| n    | Au                      | −0.10 | −0.54 | −0.98 | – | – | – |
|      | Cr/Au                   | 0.16 | −0.09 | −0.34 | – | – | – |
|      | Cr/Ti/W/Au              | 0.33 | 0.20 | 0.06  | 1.2 × 10⁻⁸ | 6.1 × 10⁻⁹ | 2.4 × 10⁻¹⁰ |
|      | Ti/Cu                   | 0.55 | 0.27 | −0.002 | 7.7 × 10⁻⁹ | 3.2 × 10⁻⁹ | – |
|      | Ti/TiW/Cu               | −0.52 | −1.06 | −1.60 | – | – | – |

Figure 5. a) High angle annular dark field (HAADF)-STEM image showing microstructure of the sputtered Cr metallization on a (Bi,Sb)₂Te₃ superlattice. b) HAADF-STEM image showing atomic resolution of the interfacial region near the tip of one of the vertical (Bi,Sb)₂Te₃ spikes. Here, the Cr metal layer is in direct contact with the (Bi,Sb)₂Te₃. The measured 0.20 nm lattice fringe spacing in the metal layer is consistent with the spacing of the Cr (011)-type planes. c) EDS spectrum image collected and analyzed using the multivariate curve resolution (MCR) technique from the region indicated by the yellow box in (a). Five distinct spectral components were identified as follows: red and green correspond to Bi-rich and Bi-deficient layers in the (Bi,Sb)₂Te₃ superlattice; blue corresponds to the Cr metallization. Yellow represents Au-rich regions within the Cr layer. Magenta is an oxygen-rich interlayer between the Cr and (Bi,Sb)₂Te₃. d) Plot of the EDS spectral components using the same color scheme as in (c). For clarity, the component associated with Au is not shown. The inset shows the low energy region of the spectra. The interfacial phase (magenta) is associated with an increase in oxygen signal.
infiltrated along the grain boundaries in this layer. However, while metal diffusion into the TE is a well-known phenomenon, here we find little evidence of metal diffusion into the TE material. Particularly striking is the rough morphology of the interface between the (Bi,Sb)2Te3 superlattice and the Cr coating. We believe that this rough morphology is likely a result of the presputtering of the thermoelectric surface prior to deposition of the Cr. Analysis of the EDS spectrum images in conjunction with atomic resolution high angle annular dark field (HAADF)-STEM imaging (Figure 5b) shows vertical spikes of (Bi,Sb)2Te3 in contact with Cr, at least at their tips. We also identify an additional signal in the troughs (shaded as magenta in Figure 5c) which exhibits signatures of enriched oxygen content (Figure 5d). The observations also suggest some disorder in crystal structure of the (Bi,Sb)2Te3 material near the tip of the vertical spikes, with bending of the lattice planes and a disruption of the normal 5-plane wide quintuple layer structure apparent in the high resolution STEM observations (Figure 5b).

We also observe a similar morphology at the interface between the (Bi,Sb)2Te3 and the Ti metallization (see the Supporting Information). Overall, these observations suggest that controlling the interface morphology may be beneficial to the contact resistivity, and also point to possible further reduction of the contact resistivity via increasing the direct contact area by further removing the oxygen at the interface, which could allow the resistivity to approach the fundamental theoretical limit.

We harnessed the low contact resistivity of sputtered contacts to V-telluride materials by realizing high-performance thermoelectric coolers. A sputtered Ti/Cu metallization with an Ar+ backsputter was used for the sink-side contact in a thin-film TE test structure, as shown schematically in Figure 6. The p-type material is a Bi0.5Se0.5Te1−xSbx superlattice (thickness 15.3 μm) while the n-type material is Bi2Te3-xSex (thickness 14.2 μm). Details of the material properties can be found in the Supporting Information.

The hot side/cold side temperature difference (ΔT) of the test device was measured as a function of input current. The top and bottom TE module temperatures, Tc and Th, respectively, were read using 25 μm diameter K-type thermocouples positioned on the device. The measurements were taken under vacuum (P < 10⁻⁴ Torr) to minimize convective parasitics. The TE device was sunk to a water-cooled heat sink which was maintained at 23 °C. During the measurements, the temperature of the hot side increased slightly, reaching 24.5 °C at maximum cooling.

The ΔT versus I curve for the Ti/Cu-contacted device is plotted in Figure 6 (additional data can be found in the Supporting Information). Data from a control sample using standard contacts on both the source and sink sides is included for comparison. Both curves follow the expected expression[2]

$$\Delta T = \frac{S T_c}{K} I - \frac{1}{2K} (R_c + R_s) I^2$$  \hspace{1cm} (1)

where S is the Seebeck coefficient, Tc is the cold side temperature, I is the current, K is the thermal conductance, R is the resistance of the thermoelectric material, and Rc the contact resistance. (In our measurements, we stopped the data acquisition soon after ΔT started to decrease after reaching the maximum value. Best fits to Equation (1) are shown to help guide interpretation of the curves.) Equation (1) shows that for fixed material parameters, a decrease in contact resistance leads to a decrease of the Joule heating term. This reduction allows the device to be operated at a higher current, and therefore to achieve a larger ΔTmax.

The increased maximum ΔT value from the device built with sputtered Ti/Cu contacts (ΔTmax = 41.2 K) as compared to that from the device with standard contacts (ΔTmax = 35.9 K) suggests that the decreased contact resistance seen from the sputtered Ti/Cu contacts can be readily translated into improved device performance. The reduction in contact resistance is readily seen from the voltage versus I data shown in the Supporting Information, which gives total resistances of 18 mOhm for the reference module, and 10 mOhm for the module with sputtered contacts. Thus, adaptation of this new metallization process has the potential to enable record-setting performance of V-telluride materials.
devices, specifically those with thermoelectric applications. From the best fits and the above total resistances, we extract device Seebeck coefficients of 343 µV K⁻¹ for the reference device, and 295 µV K⁻¹ for the device with sputtered contacts. These values are in good agreement with the materials Seebeck coefficients reported in the Supporting Information. In addition, this further shows the value of the improved contacts, since at the same device Seebeck coefficient the maximum cooling with the new contacts would be even higher.

In summary, we employed ab initio simulations to understand the fundamental properties of metal/telluride interfaces and to predict their contact resistivity. We find an unusual situation where the strong disorder at the metal/Tev interface leads to chemical dipoles that dominate the band bending near the interface. This effect, accompanied by the high electron affinity of the TE material invariably leads to a contact resistivity that is determined mainly by thermionic field emission. We harness this knowledge to fabricate improved contacts to TE materials, and to demonstrate a significant improvement in the cooling performance of thin film TE devices. We expect that detailed knowledge of the contact properties could lead to further reduction of the contact resistivity by designing new contact geometries. For example, the knowledge that tunneling plays a critical role at the contact suggests that metal protrusions could provide local field concentration that would reduce the band bending width and increase the tunneling probability.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

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