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Thermoelectric Transport by Surface States in Bi$_2$Se$_3$-Based Topological Insulator Thin Films

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We develop a tractable theoretical model to investigate the thermoelectric (TE) transport properties of surface states in topological insulator thin films (TITFs) of Bi$_2$Se$_3$ at room temperature. The hybridization between top and bottom surface states in the TITF plays a significant role. With the increasing hybridization-induced surface gap, the electrical conductivity and electron thermal conductivity decrease while the Seebeck coefficient increases. This is due to the metal-semiconductor transition induced by the surface-state hybridization. Based on these TE transport coefficients, the TE figure-of-merit $ZT$ is evaluated. It is shown that $ZT$ can be greatly improved by the surface-state hybridization. Our theoretical results are pertinent to the exploration of the TE transport properties of surface states in TITFs and to the potential application of Bi$_2$Se$_3$-based TITFs as high-performance TE materials and devices.

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Thermoelectric (TE) materials have continuously drawn a great deal of attention to date since they are important energy materials. Searching high-performance TE materials for efficient heat-electricity conversion is one of the greatest challenges in materials science. The energy conversion efficiency of TE devices depends on transport coefficients of the constituent materials through the figure of merit,$^{[1]}$ which is defined as $ZT = T \sigma S^2/(\kappa_e + \kappa_p)$, where $T$ is the temperature, $\sigma$ is the electrical conductivity, $S$ is the Seebeck coefficient, and $\kappa_e$ and $\kappa_p$ are the electronic and phononic contributions to the thermal conductivity. The highest record of $ZT$ for present-day TE materials is on the order of unity.$^{[2]}$ Any small increment in this value will result in many new applications for TE devices. Therefore, it is an important and challenging issue to search for TE materials with large $ZT$.

It is clear that, to increase $ZT$, one has to decrease the thermal conductivity and/or increase the electrical conductivity and Seebeck coefficient. Many different approaches have been proposed and attempted to improve $ZT$, e.g., by suppressing the phonon conduction,$^{[3]}$ by tuning the doping concentration,$^{[4]}$ by tailoring the electronic structure,$^{[5]}$ and by reducing the dimensionality of the material system.$^{[6]}$ Despite these important approaches, searching high-performance TE materials with large $ZT$ is still a long-term goal in materials science and awaits new approaches to come. The recent discovery of topological insulators (TIs) may shed new light on this pursuit. TIs are a new class of quantum materials with an insulating bulk band gap and gapless helical surface states.$^{[7,8]}$ The most exotic property of TIs is that their helical surface states carrying spin-polarized dissipationless currents due to topological protection. As is known, many currently known TIs such as Bi$_2$Te$_3$ and Bi$_2$Se$_3$ are also excellent TE materials. However, the nontrivial surface states in TI materials, which were unknown in the earlier research of TE materials, might be utilized to design high-performance TE devices.

Although there has been a lot of research work on TI surface states, one of the major obstacles in studying their transport properties is an unavoidable contribution of TI bulk states to the transport.$^{[9,10]}$ Therefore, to investigate the TE transport properties of TI surface states, a key problem is how to realize the surface-dominant transport in TIs. In practice, this problem can be solved by tuning the Fermi level of the TI system into its bulk band gap, e.g., via chemical doping and/or gate voltage.$^{[11–13]}$ When the Fermi level is tuned into the bulk band gap, the contribution of bulk states to the TE transport can be neglected. In this work, we develop a tractable theoretical model to investigate the TE transport properties of surface states in TI thin films (TITFs) of Bi$_2$Se$_3$. We chose Bi$_2$Se$_3$-based TITFs as our model systems since they have the following advantages in studying the TE transport properties of surface states.

1. The large bulk band gap of Bi$_2$Se$_3$ (∼0.3 eV)$^{[14,15]}$ allows the topological surface states to dominate the TE transport, even at room temperature, as long as the Fermi level lies inside the bulk band gap. (2) As compared with TI bulks, TITF structures have a large surface-to-volume ratio$^{[16]}$ which can further enhance the surface dominance in the TE transport.

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(3) It has been shown experimentally\cite{17} that due to the interface-induced phonon scattering, TTF structures have a much lower phonon thermal conductivity compared with TI bulks. Together with a very small penetration depth of the surface states, the phonon thermal conductivity of the surface transport channel can be neglected in TITFs. (4) More importantly, by decreasing the thickness of the TITF, the surface states can hybridize to open a finite gap.\cite{18,19} We will show that the TE performance of the TITF can be greatly improved by this tunable surface gap, i.e., a large value of $ZT$ can be obtained.

![Fig. 1](image)

**Fig. 1.** (a) Sketch of the TITF with a thickness of $d$. The carrier transport is along the $x$-direction in the surface plane, indicated by the blue and red arrow. The surface and interior regions of the TITF are denoted by the blue and red colors. (b) Schematic band structure of the TITF. The notations BCB, SCB, SVB and BVB represent the bulk conduction-band, surface conduction-band, surface valence-band and bulk valence-band, respectively. The red curves and blue lines denote the bulk and surface states, respectively. The dashed line denotes the position of the Fermi level $E_F$ and the black upward arrow means $E_F$ can be tuned between the surface valence-band and bulk valence-band. Here $2\Delta$ is the surface gap induced by the surface-state hybridization in the TITF.

In the present study, we consider a Bi$_2$Se$_3$-based TITF of thickness $d$, as shown in Fig. 1(a). Such a TITF deposited on a dielectric substrate (e.g., Al$_2$O$_3$) has been successfully fabricated with molecular beam epitaxy.\cite{20} When the thickness of the TITF is thin enough ($d \sim 1$–10 nm), the top and bottom surface states in the TITF can hybridize to open a finite gap at the Dirac point, as shown in Fig. 1(b). The hybridization gap was theoretically predicted\cite{18} and experimentally confirmed.\cite{19} In our previous work,\cite{21} we studied the acoustoelectric properties of TITFs. It was found that the surface states in the TITF can have a strong absorption of the surface acoustic waves propagating along the surface plane of the TITF and the absorption strength can be enhanced by the surface-state hybridization. In this work, we investigate the TE properties of surface states in the TITF and examine the role of the surface-state hybridization.

The surface-state energy spectrum of the TITF can be written as\cite{21}

\[ E_\lambda(k) = \lambda \sqrt{(v_F k)^2 + \Delta^2}, \]  

where $\lambda = 1$ ($-1$) denotes the conduction (valence) surface band, $k = (k_x, k_y)$ is the in-plane wave vector and $k = \sqrt{k_x^2 + k_y^2}$, $v_F$ is the Fermi velocity, and $2\Delta$ is the hybridization-induced surface gap. The zero of energy is chosen at the center of the surface gap. As can be seen, the surface-state energy spectrum of TITFs is no longer gapless and linear but exhibits a gapped and somewhat nonlinear nature, in sharp contrast to that of bulk TI.

We now consider the TE transport properties of surface states in the TITF. The transport direction of surface carriers (electrons or holes) is along the $x$-direction (see Fig. 1(a)). The carrier transport is diffusive and the Boltzmann formalism is applied. By solving the Boltzmann transport equation with the relaxation time approximation, the TE transport coefficients for the surface states in the TITF can be obtained as

\[ \sigma = e^2 L_0, \]  
\[ S = -\frac{1}{eT} \frac{L_1}{L_0}, \]  
\[ \kappa_c = \frac{1}{T} \frac{L_0 L_2 - L_1^2}{L_0}, \]

where $e$ is the electron charge and the integrals $L_j$ ($j = 0, 1, 2$) are given by

\[ L_j = \int_{-\infty}^{+\infty} dE \left( -\frac{\partial f}{\partial E} \Pi(E)(E - E_F)^j \right), \]

where $f(E)$ is the Fermi–Dirac function, $E_F$ is the Fermi energy, and $\Pi(E)$, which is the so-called transport distribution function (TDF),\cite{22} is given by

\[ \Pi(E) = \sum_{\lambda,k} v^2_\lambda(k) \tau_\lambda(k) \delta[E - E_\lambda(k)], \]

where $\tau_\lambda(k)$ is the relaxation time, and $v^2_\lambda(k)$ is the group velocity along the transport direction which is given by $v_\lambda(k) = (1/h) \partial E_\lambda(k) / \partial k_x$. As can be seen, the TDF is determined by the material-specific band structures and carrier scattering mechanisms. For the simple Dirac-type band structure given by Eq. (1), the TDF can be expressed as a product of density of states $g_\lambda(E)$, group velocity $v_\lambda(E)$, and relaxation time $\tau_\lambda(E)$, that is,

\[ \Pi(E) = \sum_{\lambda} g_\lambda(E) v^2_\lambda(E) \tau_\lambda(E). \]

From the surface-state energy dispersion of TITFs given by Eq. (1), one can derive the density of states $g_\lambda(E)$ and carrier velocity $v_\lambda(E)$, which are given by

\[ g_\lambda(E) = \frac{|E|}{\pi \hbar v^2_F} \theta(\lambda E - \Delta), \]  
\[ v_\lambda(E) = \frac{(E^2 - \Delta^2)v^2_F}{2E^2} \theta(\lambda E - \Delta), \]
where $\Theta(x)$ is the unit step function. Furthermore, we assume the carrier relaxation time to be a constant, i.e., $\tau(E) = \tau$.

With $g_{\lambda}(E)$ and $v_{\lambda}(E)$ obtained, $L_j$ ($j = 0, 1, 2$) can be expressed as

$$L_j = \frac{(k_B T)^{j+1}}{2\pi \hbar \gamma} \int_0^\infty dx \frac{x^2 - a^2}{x} \times [F_j(x - b) + (-1)^j F_j(x + b)],$$

(7)

where $\gamma = h/\tau$, $a = \Delta/(k_B T)$, $b = E_F/(k_B T)$, $F_j(x \pm b) = (x \pm b)^j f(x \pm b)[1 - f(x \pm b)]$ with $f(x) = 1/(e^{xT} + 1)$, $h$ is the reduced Planck constant, and $k_B$ is the Boltzmann constant.

In this work, the TE transport coefficients $\sigma$, $S$, $\kappa_e$ and TE figure-of-merit $ZT$ are calculated for the surface states in Bi$_2$Se$_3$-based TITFs at room temperature ($T = 300K$). We assume the mean free path $\lambda = 100 \text{nm}$ at $T = 300K$ and the Fermi velocity $v_F = 5 \times 10^5 \text{m/s}$ to be constant and equal for the surface states in gapless and gapped TITF systems. These parameters might have some error bars due to the lack of experimental data for Bi$_2$Se$_3$-based TITFs. The relaxation time is therefore given by $\tau = \lambda/v_F = 2 \times 10^{-13} \text{s}$. As mentioned above, Bi$_2$Se$_3$-based TITFs have many advantages in realizing the surface-dominant TE transport at room temperature such as a gate tunable Fermi level, large bulk band gap, and large surface-to-volume ratio. Here, to further enhance the surface dominance in the TE transport, we consider the Bi$_2$Se$_3$-based TITFs doped with a high density of nonmagnetic impurities. This corresponds to a case with strong disorder, where the bulk-state conduction and phonon heat transport are negligible. However, due to topological protection, the TI surface states are free of being scattered by nonmagnetic impurities and as a result, the surface-state transport is nearly unaffected. Combining all these practical advantages, the surface-state dominant TE transport in Bi$_2$Se$_3$-based TITFs can really be achieved while the bulk-state and phonon contributions to the TE transport can be safely neglected.

In Fig. 2, we show the TE transport coefficients including the electrical conductivity $\sigma$, the absolute value of the Seebeck coefficient $|S|$, the electron thermal conductivity $\kappa_e$, and the TE figure-of-merit $ZT$ for the surface states in the Bi$_2$Se$_3$-based TITF. The results are shown at room temperature $T=300K$ for different surface gaps $\Delta$ as indicated. Considering that the bulk band gap of Bi$_2$Se$_3$ is about 300meV, we restrict the Fermi level in the range $[-150, 150]$meV for the realization of surface-dominant TE transport. As can be seen, $\sigma$, $|S|$, $\kappa_e$ and $ZT$ are symmetric with respect to the Fermi level $E_F$ due to particle-hole symmetry, reflected by the isotropic surface-state energy spectrum (see Eq. (1)). With the increase of $\Delta$, $\sigma$ and $\kappa_e$ decrease while $|S|$ increases. This is due to the metal-semiconductor transition induced by the surface-state hybridization (it is known that metals have larger $\sigma$ and $\kappa_e$ but smaller $|S|$ than semiconductors). For the fixed $\Delta$, $\sigma$ increases with $|E_F|$ since $\Delta$ is proportional to the surface carrier (electron or hole) density. When $E_F > 0$ ($E_F < 0$), the conducting carriers are electrons (holes) so that $S$ has a negative (positive) value. Compared with $\sigma$ as a function of $E_F$, the situation for $\kappa_e$ is more complex, which first decreases then increases with increasing $|E_F|$ and has a local maximum at $E_F=0$. Therefore, the relation between $\kappa_e$ and $\sigma$ violates the Wiedemann–Franz law, which states that $\kappa_e$ is proportional to $\sigma$, i.e., $\kappa_e = L_0 T \sigma$, with $L_0 = \pi^2 k_B^2/(3e^2)$ being the Lorentz number. According to Ref. [24], this violation makes the TE transport more reversible, which is desirable for the efficient TE energy conversion.

As mentioned above, we have neglected the phonon thermal conductivity $\kappa_p$ in Bi$_2$Se$_3$-based TITFs doped with a high density of nonmagnetic impurities. Hence, the TE figure of merit can be written as $ZT = T \sigma |S|^2/\kappa_e$. According to this expression, it seems to
be difficult to determine whether $Z_T$ decreases or increases with an increase in $\Delta$, since $\sigma$ and $\kappa_e$ decrease while $|S|$ increases with $\Delta$. However, our numerical calculation shows that the TE power factor $P_F = \sigma S^2$ increases with $\Delta$ (see Fig. 3). Together with the reduction of $\kappa_e$, one can see that $Z_T$ increases with $\Delta$. However, our numerical calculation shows that the TE power factor $P_F = \sigma S^2$ increases with $\Delta$ (see Fig.3). Together with the reduction of $\kappa_e$, one can see that $Z_T$ increases with $\Delta$. By tuning the Fermi level properly, $Z_T$ can be optimized and its maximum value can reach about 1.6 for $\Delta = 80$ meV, which is much larger than that for $\Delta = 0$ meV (about 0.25). Therefore, the TE figure-of-merit $Z_T$ of TITFs can be greatly improved by the surface-state hybridization.

In summary, we have theoretically investigated the TE transport properties of surface states in Bi$_2$Se$_3$-based TITFs. The surface-state hybridization in TITFs plays a significant role. With the increasing hybridization-induced surface gap, we find that the electrical conductivity and electron thermal conductivity decrease while the Seebeck coefficient increases. Based on these TE transport coefficients, we have calculated the TE figure-of-merit $Z_T$ and have shown that $Z_T$ can be greatly improved by the surface-state hybridization. A large value of $Z_T$ has been achieved by increasing surface gap. Considering that Bi$_2$Se$_3$ material for the TE industry is as important as silicon material for the electronic industry, our theoretical results indicate that Bi$_2$Se$_3$-based TITFs are promising for TE science and technology.

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