Excellent thermoelectric performance of ZrTe$_2$ thin films and device


1 Department of Applied Physics, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong, China
2 Department of Physics, Hong Kong University of Science and Technology, Hong Kong, China
3 College of Physics, Chongqing University, Chongqing 401331, China
4 School of Physics, Sun Yat-sen University, Guangzhou 510275, China

Abstract

Achieving high thermoelectric power factor in thin film heterostructures is essential for integrated and miniaturized thermoelectric device applications. In this work, we demonstrate a mechanism and device performance of enhanced thermoelectric power factor through coupling the interfacial confined two-dimensional electron gas (2DEG) with thin film conductivity in a transition metal dichalcogenides-SrTiO$_3$ heterostructure. Owing to the formed conductive interface with two-dimensional electron confinement effect and the elevated conductivity, the ZrTe$_2$/SrTiO$_3$ (STO) heterostructure presents enormous thermoelectric power factor as high as $4 \times 10^5 \mu W cm^{-1} K^{-2}$ at 20 K and $4800 \mu W cm^{-1} K^{-2}$ at room temperature. Formation of quasi-two-dimensional electrons gas at the interface is attributed to the giant Seebeck coefficient, and enhanced electrical conductivity is suggested to be originated from charge transfer induced doping in the ZrTe$_2$, which leads to extremely large thermoelectric power factor. By taking the thermal conductivity of STO substrate as a reference, the effective $zT$ value of this heterostructure can reach 1.5 at 300 K. This high thermoelectric figure of merit is demonstrated by a prototype device based on this heterostructure which results in 3K temperature cooling by passing through a current of 100 mA. This superior thermoelectric property makes this heterostructure a promising candidate for future thermoelectric device, and more importantly, paves a new pathway to design promising high-performance thermoelectric systems.

Keywords: Thermoelectric; ZrTe$_2$ thin film; high mobility; interface; transition metal dichalcogenides

$^*$ These authors contribute equally
*corresponding contact:
Jiyan.dai@polyu.edu.hk; xiaoyuan2013@cqu.edu.cn; songhua.cai@polyu.edu.hk; phjwang@ust.hk
Introduction

High-performance thermoelectric thin films have been attracting growing interests as a hot topic in condensed matter physics and materials science recently. The development of micro-cooling devices and miniaturized self-powered devices are technically important for improvement of large-scale integrated circuits, which drives the study of thin film thermoelectric. For example, in microprocessors, the “hot spot” regions with higher power density frequently cause the fatal thermal failure. Therefore, thin film-based thermoelectric device becomes a potential solution for in-demand cooling of circuits. Furthermore, high-performance thermoelectric thin films are also essential to build micro-power generators for self-powered devices in environments where a thermal gradient exists; this is significant for power engineering and management in future electronics.

The performance of thermoelectric material can be evaluated by a dimensionless figure of merit $zT = S^2\sigma T/(\kappa_e + \kappa_l)$, where $S$, $\sigma$, $T$, $\kappa_e$, $\kappa_l$ represent Seebeck coefficient, electrical conductivity, absolute temperature, electronic thermal conductivity and lattice thermal conductivity, respectively. To enhance the figure of merit, numerous efforts have been devoted to improve the power factor (power factor $= S^2\sigma$) or reduce the thermal conductivity. Among those methods to realize large thermoelectric power factor in thin film/substrate, interface engineering has been proven to be an effective approach to enhance Seebeck effect of the heterostructure. Especially, the presence of quasi-two-dimensional electron gas (quasi-2DEGs) at the interface provides a dominant contribution to the enhancement of $|S|^2$. As an ideal substrate with unique structure, physical and chemical properties, SrTiO$_3$ (STO) has been widely applied in thin film growth as well as the exploration of novel low-dimensional physics. For example, the unique electronic phenomena generated from the heterointerface between epitaxial oxide layer and the STO substrate such as the very intriguing LAO/STO system has raised much attention. However, although the LAO/STO interface generally shows enhanced $|S|$, itself has not been found to exhibit an outstanding thermoelectric power factor for meaningful applications mainly due to the limitation on its electrical conductivity. For designing of available high-performance thermoelectric system, it is necessary to couple both high electrical conductivity and the enhanced thermoelectricity on a single heterointerface.

Here, we demonstrate the realization of giant thermoelectric power factor in ZrTe$_2$/SrTiO$_3$ heterostructure, which mainly attribute to the coupling between interfacial quasi-2DEG generated in STO substrate and the high conductivity of transition metal dichalcogenides (TMDs) film. An interfacial reaction between the pulsed-laser deposition (PLD) deposited layered ZrTe$_2$ thin film and STO substrate leads to the appearance of quasi-2DEG which enhancing the Seebeck coefficient. Meanwhile, the conductivity is also dramatically enhanced which may contribute from the doping effect of TMD film induced by charge transfer. Combining both the high Seebeck coefficient and electrical conductivity, this heterostructure system eventually presents an outstanding thermal power factor as well as thermoelectric figure of merit $zT$ value, which is further demonstrated in a cooling
device. This work provides a promising pathway for designing of high-performance thermoelectric systems and opens new possibilities for exploration and development of intriguing thermoelectric materials, structures as well as thin film-based electronic devices.

**Methods**

In this study, PLD was used to grow ZrTe$_2$ films using an alloy target with Zr:Te=1:5; and insulating (100) STO was selected as the substrate. The substrate-target distance during the deposition was 5 cm, with the base pressure of around $5 \times 10^{-5}$ Pa, and the films were grown at optimized substrate temperature ($T_s$) of 550 °C. To preserve the ZrTe$_2$ film from oxidation in air, an additional AlN capping layer was deposited on the top of ZrTe$_2$ film at 200 °C. The laser source was a KrF excimer laser with a wavelength of 248 nm. Different thicknesses of ZrTe$_2$ films from 5 nm to 10 nm, 35 nm and 60 nm were deposited on STO substrate and the AlN capping layer is about 70 nm with amorphous structure.

X-ray Diffraction (XRD) with Cu K$\alpha_1$ source was used to determine the orientation and crystalline phase of ZrTe$_2$ film. To determine the microstructure and thickness of ZrTe$_2$ film, transmission electron microscope (TEM) was used for cross-sectional characterization. High-resolution TEM (HRTEM) images were acquired on JEOL 2100F TEM, operated at 200 kV. Scanning TEM (STEM) images were acquired on Thermofisher Titan G2 60-300 aberration corrected S/TEM. The accelerating voltage of electron beam is 300kV. The convergence angle of electron probe for STEM-HAADF imaging is 22.5 mrad and the collection angle of annular detector is 79.5-200 mrad. A Gatan dual-EELS system was used for STEM-EELS data acquisition. For EELS line scan, the acquisition time per pixel is 1 s. The acquisition energy range is 420 to 720 eV with 0.1 eV dispersion, including the major or minor edges of oxygen and Ti.

A 9T-PPMS (Physical Properties Measurement System) was utilized for measuring the Seebeck coefficient and electrical conductivity at a temperature range from 2 K to 300 K. The measurement of Seebeck coefficient and electrical conductivity in the range from 300 K to 600 K was carried out on Joule Yacht MRS-3 thin film thermoelectric parameter test system. The thermoelectric cooling experiments were conducted using a home-built setup. To make a good electrical contact, a layer of Au stripes across the sample width were deposited on ZrTe$_2$ thin film directly followed by the AlN capping layer. Then silver epoxy was used to connect gold contacts with current source leads after removing AlN on top of gold stripes while keeping the rest part of sample protected. The total contact resistance of both terminals is about 10 Ω. Two Pt-100 thermometers were used to record temperatures on both sides. The current was applied using a Keithley 6221 AC/DC current source.

**Results and discussion**

The fine structure of as-grown samples is directly related to their performances, so the microstructure and composition of all samples were firstly investigated. Figure 1(a) shows the cross-sectional High-resolution TEM (HRTEM) image of the ZrTe$_2$ film and STO substrate. One can see that the layer-structured ZrTe$_2$ occupies the major part of the deposited film, presenting clear (0001) lattice fringes.
parallel to the substrate. This result is consistent to the layered structure of ZrTe$_2$ as shown in the schematic in Figure 1(b). ZrTe$_2$ has a hexagonal close-packed crystal structure, with space group of $P\overline{3}m1$. A stable 1T (trigonal) ZrTe$_2$ phase has an octahedral coordination of metal atoms$^{26}$ that can be illustrated as a CdI$_2$-type structure$^{27}$. A unit cell of ZrTe$_2$ is denoted by a cyan rectangle in Figure 1(b) and can also be identified in the magnified high-resolution TEM (HRTEM) image. The crystal structure of our ZrTe$_2$ film was also characterized by XRD. Figure 1(c) presents the XRD pattern of the sample with 60 nm-thick film, where peaks at $2\theta = 13.3^\circ$, $26.9^\circ$, $40.8^\circ$ and $55.4^\circ$ correspond to the c-axis (0001), (0002), (0003) and (0004) diffraction peaks of hexagonal ZrTe$_2$ ($a = 0.395$ nm, $c = 0.663$ nm, JCPDS #54-560), respectively. These results imply that the ZrTe$_2$ film is preferentially grown along c-axis, which is also consistent to the layer-by-layer characteristic observed in TEM. However, it should be pointed out that, besides the c-axis orientation, other orientation peaks can also be found in the film, suggesting that the ZrTe$_2$ film growth is not epitaxial.

**Figure 1 Crystal structure characterization** (a) High-resolution TEM image of the ZrTe$_2$ film on STO substrate, where an interfacial layer of ZrO$_2$ reaction phase can be identified between the STO substrate and the ZrTe$_2$ film. (b) Magnified image of yellow rectangle region in (a) showing the correspondence of real unit cell with the crystal structure model of ZrTe$_2$. The unit cell is denoted by a rectangle to compared with magnified high-resolution TEM image of the ZrTe$_2$ film. (c) Typical XRD patterns of the ZrTe$_2$ films. The ZrTe$_2$ simulation pattern (JCPDS #54-560) is also illustrated for comparison.

It is worth noting that between the layered ZrTe$_2$ and the STO substrate, there exists a ~10 nm thick interfacial layer with different lattice structure with ZrTe$_2$. A detailed analysis of the lattice spacings
and their angles suggest that this interfacial layer belongs to ZrO$_2$ (See Figure S1). This implies that interfacial reaction occurs between the deposited ZrTe$_2$ and the STO substrate, and the interfacial characteristic has remarkably influenced the physical properties of both ZrTe$_2$ and the STO substrate. This can be revealed by the STEM energy disperse x-ray (EDX) spectroscopy element mapping and the electron energy loss spectroscopy (EELS) analysis of the interfacial region as shown in Figure 2. It is apparent in EDX mapping results shown in Figure 2(a), that the distributions of O and Te are not uniform along the thickness direction, i.e., the signal intensity of Te increases near the interface with a decrease of O signal. It is surprise to see that O even exists in the whole film, suggesting an out diffusion of O from STO which can be proven by EELS line-scan analysis across the interface as shown in Figure 2(b) and (c). By extracting the signals of Ti and O edges from the EELS spectrum, the energy loss near edge structures (ELNES) reveals that the 4 unit cells of STO from interface exhibits a different chemical state with bulk STO. From the 4$^{th}$ layer of STO up to the interfacial layer, the valence state of Ti cations degrades from Ti$^{4+}$ to Ti$^{3+}$ (peak shift and shape variation in Figure 2(b) with a corresponding change of O k-edge fine structure (Figure 2(c)). The interfacial characteristics imply a conductive STO surface, which analogous to the similar situation in the oxide heterostructures such as LAO/STO cases.

![Elemental mapping and interfacial valence states of Ti and O ions](image)

**Figure 2** Elemental mapping and interfacial valence states of Ti and O ions (a) STEM-EDS element mapping across the interface, (b) EELS results of Ti 3d-L edge shift peaks, and (c) O -2p K edge fine structure across the interface.
We have carried out thermoelectric and electron transport properties measurements to different thicknesses of samples at temperatures ranging from 2K to 600K. The ZrTe$_2$/STO heterostructures exhibit negative Seebeck coefficient throughout the whole temperature range, indicating a n-type conducting characteristic; and the general trend is that the thicker films show relatively lower Seebeck coefficient but with high conductivity, while very thin films present highly fluctuated Seebeck coefficient and high resistance. Figure 3(a) shows the temperature-dependent Seebeck coefficients of the ZrTe$_2$/STO heterostructure with the 35 nm and 60 nm ZrTe$_2$ films from 2 K to 600 K. The Seebeck coefficient in the sample with a 35 nm-thick ZrTe$_2$ generally increases with the temperature increases from 2 K and finally reaches the maximum value of about 530 $\mu$V/K at 440 K, above which the Seebeck coefficient begins to decrease. The sample with 60 nm-thick ZrTe$_2$ film shows a similar trend while the maximum Seebeck coefficient appears at 111 K with a value of 368 $\mu$V/K. The temperature-dependent electrical conductivity of both samples are shown in Figure 3(b), which confirms that these heterostructures are highly conductive. The electrical conductivity can reach a remarkable value up to $10^7$ S/cm at low temperature state (2 K). The coexistence of both very large Seebeck coefficient and high electrical conductivity of the ZrTe$_2$/STO heterostructure consequently leads to enormous thermoelectric power factor. As shown in Figure 3(c), the thermoelectric power factors monotonically increase with the decreasing of temperature in both samples and finally achieve a colossal value even exceeds $3 \times 10^5 \mu Wcm^{-1}K^{-2}$ at 20 K.
Figure 3 Thermoelectric and electrical conductive properties

Temperature-dependent thermoelectric properties of the ZrTe$_2$/STO heterostructures with 35 nm and 60 nm-thick ZrTe$_2$ film in the temperature range of 2-600 K. (a) Seebeck coefficient, (b) electrical conductivity, and (c) thermoelectric power factor.

As well known, quasi-2DEG can exist in the conductive STO interface and its presence can be a prime factor for triggering intriguing thermoelectric properties. To clarify the contribution of the possible interfacial effect to the outstanding thermoelectric properties, we further conducted systematic electron transport measurements on the heterostructure. Compared to our previous work\textsuperscript{26}, the sample’s quality has been further improved by optimizing the growth parameters. In these high quality heterostructures, we observed obvious Shubnikov–de Haas (SdH) oscillations superimposed on the large magnetoresistance at low temperatures below 4 K. Figure 4(a) shows the oscillations after subtracting a smooth background from the measured magnetoresistance of a typical 60 nm-thick ZrTe$_2$ film on STO. The periodicity in the reciprocal of magnetic field (1/B) demonstrates the SdH effect is the origin. Fast Fourier Transform (FFT) results further reveal that the oscillating frequency peak is about 29.5 T as shown in Figure 4(b). By analyzing the temperature dependence of the oscillations using the Lifshitz-Kosevitch formula, we obtain an effective mass of 1.3$m_0$ with $m_0$ represents the bare electron
mass (Figure 4(c)). In addition, the Dingle temperature is calculated as 1.4 K based on the measured quantum oscillations. It is interesting to find that the microscopic parameters of the quantum oscillations actually agree well with those acquired in conductive STO surfaces\textsuperscript{28-35}. Thus, we believe that the interfacial effect between the ZrTe\textsubscript{2} thin film and the STO substrate plays a significant role for presenting the very exotic transport properties in our observation\textsuperscript{26}.

Our previous study on the ZrTe\textsubscript{2}/STO heterostructures has revealed a quasi-2D transport characteristic of the hybrid heterostructure, within which the electrons exhibit a high mobility of about $1.8 \times 10^4$ cm$^2$V$^{-1}$s$^{-1}$ at 2 K\textsuperscript{26}. The quasi-2D transport feature is consistent to the layered structure of ZrTe\textsubscript{2} and the property of quasi-2D electrons in the STO interface as well. These characteristics are supposed to be responsible for the observed enormous Seebeck coefficient and power factor. Similar findings also exist in TiO\textsubscript{2}/STO, LAO/STO and FeSe/STO heterostructures\textsuperscript{1,36,37}. In the TiO\textsubscript{2}/STO heterostructure, with the presence of 2DEG, the room temperature (300 K) Seebeck coefficient is as large as 1050 $\mu$V/K; while our ZrTe\textsubscript{2}/STO heterostructures show a value of $|S|_{300K}$ smaller than 500 $\mu$V/K (Figure 3(a)). It is worth noting that the Seebeck coefficient of the heterostructure shows...
obvious dependency with the thickness of the TMD film that a thicker ZrTe$_2$ film will lead to the decrease of $|S|$. Figure S shows comparison of our results to reported Seebeck coefficients from other 2D systems. In fact, a control sample with only the interfacial contribution from ZrO$_2$/STO shows much higher Seebeck coefficient with a value of about 750 $\mu$V/K at 300 K, well consistent with the result in the TiO$_2$/STO heterostructure$^1$ (See Figure S3). Meanwhile, the electric conductivity is also found to be thickness dependent; while this dependence is opposite to that of the Seebeck coefficient. The control sample with only ZrO$_2$/STO shows a much higher resistance, making the Seebeck coefficient measurement fluctuate vigorously and also prevents the system achieving a higher power factor. As a conclusion, the deposition of TMD ZrTe$_2$ thin films on the top of the quasi-2D system induces slight decrease of the Seebeck coefficient of the system due to its relatively lower Seebeck coefficient, but it contributes very high conductivity to the system. With the coupling between the thin ZrTe$_2$ film and the STO-based interface, the largest thermoelectric power factor can be achieved in the ZrTe$_2$/STO heterostructure with an optimized thickness.

However, it should be noted that if at a much thinner thickness, the interface is not well conductive and EELS results show that the valence state of Ti cations at the interfacial region is still Ti$^{4+}$ instead of Ti$^{3+}$ (see Error! Reference source not found.). Therefore, we come to the conclusion that the interfacial state for forming a confined 2D conductive interface is essential for achieving the outstanding thermoelectric properties in this system. On the other hand, the extremely high conductivity is another crucial factor for realization of the giant thermopower factor. We also prepare a ZrTe$_2$ thin film on the insulating Al$_2$O$_3$ (Figure S) to clarify the origin. It can be found that the ZrTe$_2$ thin film is indeed metallic with a high conductivity but still lower than the value achieved in the ZrTe$_2$/STO system, this also suggests a modulation effect to the ZrTe$_2$ film from the interface. As a reference, the FeSe/STO is a well-explored metal/STO heterostructure system due to the interface-enhanced high temperature superconductivity$^{38,39}$. Strong evidences have shown that the interfacial electron-phonon coupling as well as the electron doping on the FeSe layer should be responsible for the high temperature superconductivity of the FeSe/STO system$^{40-44}$. In particular, the electron doping to the film through the thermal annealing generally decreases the resistance of the system and thus enhances the conductivity$^{45}$. We suggest that similar mechanism may also exist in the metallic ZrTe$_2$/STO heterostructure that the electron doping from the substrate to the TMD film determines the very high conductivity of the system. More efforts are needed to further demonstrate the underlying physical origins.

It is noticeable that there exists a small peak in the Seebeck coefficient at about 20 K (Figure 3(a)), and this can be explained by the phonon-drag effect$^{46-48}$ originated from the STO (100) substrate. One study that related to the thermal transport of STO$^{49}$ exhibits a thermal conductivity peak close to 20 K. When a thermal gradient is applied to the thin film and substrate, nonequilibrium phonons will be generated and transfer their momentum to the carriers; as a result, an additional electric field is formed.
in the film, thus enhance the Seebeck coefficient. The characteristic of phonon-drag effect is that, it is more effective at temperatures where the substrate attains its maximum thermal conductivity; this is the reason why the peak of Seebeck coefficient, as shown in Figure 3(a), is near the temperature where the substrate exhibits its peak thermal conductivity.

For thin films, the substrate thermal conductivity dominated the thermal transport, so we estimate the effective thermoelectric figure of merit \( zT \) is mainly based on the value of thermoelectric power factors of the film and the reported thermal conductivity values of STO single crystal\(^5\). For the 35 nm ZrTe\(_2\)/STO sample, the \( zT \) value at 300 K is about 15, which is extremely higher compared to all reported works according to our knowledge. Even for the 60 nm thick ZrTe\(_2\) film sample, \( zT \) is about 1.5 at 300 K and increases to 13 at 18 K.

The superior thermoelectric performance of our ZrTe\(_2\)/SrTiO\(_3\) heterostructure is further demonstrated by thermoelectric cooling experiments as shown in Figure 5 at room temperature of \( T_{\text{hot-side}} = 300 \) K and ambient atmosphere. The Peltier effect allows the heat to be pumped from the cold end towards the hot-side (heat sink) by applying positive currents due the n-type nature of our device. As shown in Figure 5(a), clear asymmetric temperature difference \( \Delta T = T_{\text{cold-side}} - T_{\text{hot-side}} \) produced by Peltier effect is seen by reversing current directions although Joule heating dominants at currents above 20 mA. This Joule heating is inevitable as the total resistance of the device is about 20 \( \Omega \) including \(~10 \) \( \Omega \) of sample resistance and \(~10 \) \( \Omega \) of contact resistance. Nevertheless, the Joule heating is overcome at positive current of 20 mA and a net temperature difference of \(-0.15 \pm 0.05 \) K is achieved. To visualize the intrinsic Peltier cooling, the Joule effect is subtracted out in Fig.5 (b) as \( \Delta T_{\text{Peltier}} = 1/2 (\Delta T_{+} - \Delta T_{-}) \) assuming a symmetric response of Joule heating. Noticeable cooling about 3 K is achieved even using a moderate current \( I=100 \) mA for a ZrTe\(_2\)(60 nm)/SrTiO\(_3\) standalone device. Measurements using current in the Amperes region were not successful due to extremely high current density load to the nanometer scale thin film. Nevertheless, the cooling effect is in comparable to that of commercial Bi\(_2\)Te\(_3\) devices in the small current and linear response region as presented by inset in Fig.5(b). Note that such a Kelvin level temperature difference is produced only by a nanometer scale thermoelectric working unit loaded with millimeter-sized substrate, if incorporating with a \( p \)-type leg, it could reduce the heat load significantly, and likely push the temperature difference to a much higher level.
Figure 5 Thermoelectric cooling performances. (a) Net temperature difference $\Delta T = T_{\text{cold-side}} - T_{\text{hot-side}}$ produced by a ZrTe$_2$(60 nm)/SrTiO$_3$ device using varies currents. (b) Peltier cooling $\Delta T_{\text{Peltier}} = 1/2(\Delta T^+_i - \Delta T^-_i)$ evaluated after subtracting the Joule heating by symmetrizing the direct temperature differences at opposite currents shown in (a). The inset in (a) shows a schematic setup used for thermoelectric cooling test. The inset in (b) compares the Peltier cooling of our device to that of a Bi$_2$Te$_3$ based commercial device.

Conclusion
In summary, we demonstrate that the ZrTe$_2$/STO heterostructure possesses colossal thermoelectric power factor at both low temperature and room temperature which has been further demonstrated in a cooling device. The excellent thermoelectric properties can be attributed to the formation of two-dimensional electrical transport property as well as the enhanced conductivity due to charge transfer occurs in the interface. This study reveals the promising thermoelectric properties of ZrTe$_2$/STO heterostructure and provides a new pathway for development of thermoelectric materials and structures. Furthermore, the mechanism exploration is of fundamental importance for both thermoelectric physics and possible application on thin film-based electronic devices.

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Supporting Information for

Enormous thermoelectric power factor of ZrTe$_2$/SrTiO$_3$ heterostructure

Chun Hung Suen$^1$, S.H. Cai$^1$, H. Li$^2$, Long Zhang$^3$, Kunya Yang$^3$, Huichao Wang$^4$*, M.Q. He$^3$, Y.S. Chai$^3$, K. Zhou$^3$, Chi Man Wong$^1$, J.N. Wang$^2$*, X. Y. Zhou$^3$* and Ji-Yan Dai$^{1,*}$

1 Department of Applied Physics, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong, China
2 Department of Physics, Hong Kong University of Science and Technology, Hong Kong, China
3 College of Physics, Chongqing University, Chongqing 401331, China
4 School of Physics, Sun Yat-sen University, Guangzhou 510275, China
Figure S1. (a) HRTEM and (b) STEM image of the interfacial structure. ZrO$_2$ becomes the dominant component near the interface area.

Figure S2. Comparison of temperature dependent Seebeck coefficient among 2D systems.
**Figure S3.** (a) HRTEM image of the thinnest control sample (5nm ZrO$_2$/STO), and (b) the Seebeck coefficient showing that the control sample has much a higher value but unstable due to its high resistance.

**Figure S4** Elemental mapping and interfacial valence states of Ti and O ions  
(a) STEM-EDS element mapping across the interface, (b) EELS results of Ti 3d-L edge shift peaks, and (c) O -2p K edge fine structure across the interface.
Figure S5. Conductivity-temperature relationship in the ZrTe$_2$ thin films on Al$_2$O$_3$ substrate.
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