

Pressure-induced enhancement of thermoelectric performance in palladium sulfide

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ABSTRACT

Thermoelectric (TE) materials, which can directly convert waste heat into electric power, have attracted considerable interest because of their reliability and great potential for practical applications, especially in the current time faced with energy shortage. Recent advances in developing TE materials for power generation always optimize at high temperatures with the figure of merit (zT) above 1. However for the cooling or wearable devices, the high-efficiency TE materials with optimized temperature range near room temperature are strongly in demand. If the optimized TE performance can be tuned from high temperature to around room temperature, the extensive commercial application for microdevices could be expected. Here, we choose polycrystalline palladium sulfide as an example to show that pressure can significantly enhance the TE performance. With the measurements of the resistivity, Seebeck coefficient, and thermal conductivity under pressure up to 10 GPa, a times enhancement of the zT value has been obtained around room temperature. The largest value of zT at high pressures near 10 GPa is comparable to the value at ambient pressure near 800 K. The results indicate that pressure as an irreplaceable thermodynamic variable has positively regulated the TE performance around room temperature.

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Currently, the world is in thirst for green energy resources because of the excessive consumption relating to energy supply and environment. High-efficiency thermoelectric (TE) materials are an ideal system for power generation devices because of their ability to convert heat directly into electricity without producing unusable heat [1–3]. The effectiveness of a TE material is criticized by the dimensionless figure of merit (zT), which can be expressed as $zT = S^2 T \rho^{-1} \kappa^{-1}$, where S is the Seebeck coefficient, T is the absolute temperature, ρ is the electrical resistivity, and κ is the thermal conductivity, respectively. For TE devices, a conversion efficiency of at least 15% ($zT \geq 1$) is needed to reach the critical requirement, especially for the application in consumer markets [4]. With the increasing thirst for higher TE efficiency, many approaches have

been developed, such as (i) doping and alloying [4–7], (ii) variation in mesostructure [8], and (iii) band structure or lattice structure approaches [9–11]. However, there is still a long way to go to cross the theoretical zT value of 3 for broad applications because of the complex relationship of the three parameters (S , ρ , κ). In addition, recent advances in TE materials with $zT > 1$ for power generation are usually in mid-temperature (500–900 K) or high-temperature region (>900 K) [2,12]. However, the applications of thermoelectricity near room temperature are still hampered because of the lack of materials with zT above 1. Therefore, the room temperature TE materials with zT above 1 are urgently needed, especially in the applications of the micropower generators and microprocessors [13,14]. Unlike the temperature-turning effect (lattice expansion) on thermoelectricity, pressure-tuning may be a viable approach to change physical properties through lattice shrinking.

Pressure is a fundamental thermodynamic variable that can dramatically drive the modifications of crystal structure, electronic

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structure, and magnetic structure of materials without introducing impurity, resulting in concomitant changes to their physical and chemical properties [15]. For TE materials, pressure tuning can not only change their TE performance but also offer a means to guide the search for higher zT [16]. From literature surveys, electrical resistivity usually has a better behavior at high pressures which is beneficial to the enhancement of zT [17,18]. Recently, measurements of Seebeck coefficient under pressure have been promoted by many experimental efforts, and the behavior of Seebeck coefficient under pressure is diversiform [19–21]. So far, little has yet been done to measure κ for TE materials under pressure. Here we choose a bulk material, palladium sulfide (PdS) as an example to demonstrate how applying pressure can tune and improve the TE performance based on the developed high-pressure techniques of Seebeck coefficient and thermal conductivity measurements.

PdS, which belongs to transition metal sulfides, has a tetragonal structure with the space group of $P4_2/m(84)$ (inset of Fig. 1a) and various interesting properties [22–25]. This binary PdS is a potential base thermoelectrical material with large power factor (PF) of $27 \mu\text{Wcm}^{-1}\text{K}^{-2}$ around 800 K at ambient pressure [26]. The high-pressure TE parameters of this sample have been collected based on the technique development using diamond anvil cells (DACs). The pressure dependent zT of this material around room temperature is

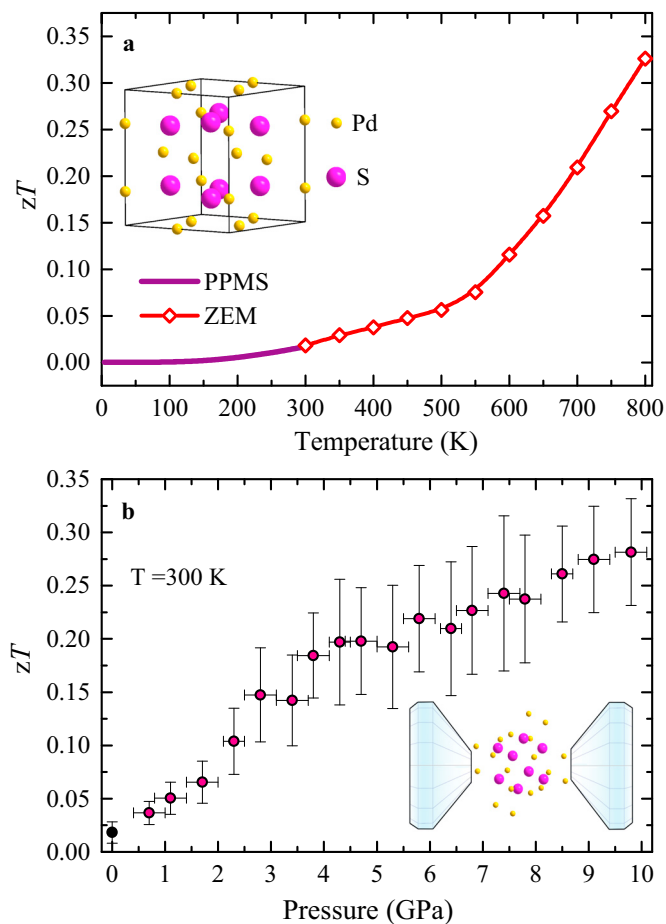


Fig. 1. (a) Temperature dependence of the dimensionless figure of merit zT of PdS at ambient pressure. The inset illustrates the crystal structure of the sample. (b) Pressure dependent zT of PdS around room temperature. Owing to the uncertainty in the measurements of the resistivity, Seebeck coefficient, and thermal conductivity under pressure, the error bars are very large for zT . PPMS, Physical Properties Measurement System, ZEM, Seebeck Coefficient / Electric Resistance Measurement System.

illustrated in Fig. 1b. The TE performance has a giant improvement at high pressures up to 4.5 GPa, and then slightly increases after passing a platform over a narrow pressure region. Obviously, the pressure-tuning effect on the TE performance of PdS has a nearly equivalent influence as the temperature-generated effect (Fig. 1a) [26]. In other words, the well-behaved TE performance of PdS at high temperature has been turned to room temperature by pressure effect. These results indicate that the route of applying pressure has successfully improved the TE performance of PdS around room temperature. Because some TE materials hold zT values around 0.5 at room temperature [1–3], one can expect the realization of zT to be above 3 if the similar pressure effects will be applied.

The detailed pressure dependence of the electrical conductivity, Seebeck coefficient, and thermal conductivity of PdS around room temperature is shown in Fig. 2. In all the experiments, pressure was obtained using DAC and was calibrated using the ruby fluorescence shift at room temperature [27]. All the results obtained under pressure are compared with the values measured at ambient pressure by Physical Properties Measurement System (PPMS) from Quantum Design. For the electrical conductivity (Fig. 2a), the detailed measurements can be seen elsewhere [25]. To give a more credible result, three separate temperature dependence of resistivity runs were carried out at various pressures. Some representative temperature dependent resistivity at various pressures (2.5, 5.6, and 9.5 GPa) are shown in Fig. 3. It can be seen that the resistivity of PdS is gradually suppressed with increasing pressure. The room temperature resistivity at different pressures is summarized in Fig. 2a. Obviously, the resistivity of PdS has a continuous decrease with increasing pressure (about two times). Resistivity for most semiconducting samples always decreases with increasing pressure due to the closing of the band gap [15]. A polynomial fitting is applied to the pressure-dependent resistivity. Then, the fitted results of resistivity are used to calculate zT values.

Based on the definition of the Seebeck coefficient $S = \Delta V / \Delta T$, we developed a high-pressure technique to measure S . The detailed setup diagram used for Seebeck coefficient measurement under pressure is shown in the inset of Fig. 2b. The temperature gradient (ΔT) from the heater to the cooler is measured by two separate W-Ta thin-film thermocouples connected with two digital temperature controllers [28]. The TE voltage (ΔV) along the temperature gradient is read out by a digital multimeter (218-A-5900, Keithley). The selected ΔV dependent of ΔT at the pressure of 0.7, 3.8, and 9.1 GPa is shown in Fig. 4. The Seebeck coefficient values are given by fitting the slopes of $\Delta V / \Delta T$, which are 384 ± 15 , 473 ± 14 , $414 \pm 10 \mu\text{V/K}$, respectively. All the relative errors of Seebeck coefficient are less than 10%. The obtained Seebeck coefficient as a function of pressure around room temperature is summarized in Fig. 2b. The S of PdS increases monotonously with increase pressure and reaches a maximum value of $473.0 \pm 30.5 \mu\text{V/K}$ at 3.8 GPa. After that, it has a continuous reduction with increasing pressure up to 10 GPa. Structure transition has not been observed up to 19 GPa [25]. Likely, the Seebeck coefficient hump around 3.8 GPa is associated with pressure-induced modulation of the band structure or electronic topological transition. The interrelationship between carrier concentration and Seebeck coefficient can be expressed as:

$$S = \frac{8\pi^2 k_B^2}{3eh^2} m^* T \left(\frac{\pi}{3n} \right)^{2/3},$$

where n is the carrier concentration and m^* is the effective mass of the carrier [2]. Therefore, one can conclude that the evolution of Seebeck coefficient at higher pressures is mainly caused by the increased carrier concentration. Similar behavior is not unusual in other samples [19–21]. Here, pressure is limited within 10 GPa for Seebeck coefficient measurements because of the unacceptable error of temperature at higher pressures.

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