Applied Physics Letters

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Cite as: Appl. Phys. Lett. **121**, 070401 (2022); https://doi.org/10.1063/5.0115322
Submitted: 27 July 2022 • Accepted: 28 July 2022 • Published Online: 18 August 2022

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AIP Publishing

Applied Physics Letters EDITORIAL scitation.org/journal/apl

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Submitted: 27 July 2022 · Accepted: 28 July 2022 ·
Published Online: 18 August 2022

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Note: This paper is part of the APL Special Collection on Thermoelectric Materials Science and Technology Towards Applications.
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https://doi.org/10.1063/5.0115322

INTRODUCTION

This special issue of *Applied Physics Letters* presents pioneering articles in the field of thermoelectric (TE) materials and devices. TE materials are pivotal in delivering one of the solutions to the global energy crisis as they can successfully convert the waste heat into a useful electrical energy. This realization compels researchers to develop high-performance, environmentally friendly, and earth-abundant materials, which would be beneficial for heat to electrical energy conversion in power plants, households, automobiles, space technology, etc. The TE efficiency of any material is governed by a dimensionless figure of merit $zT = S^2\sigma T/\kappa$, where the parameters S (Seebeck coefficient), σ (electrical conductivity), and κ [total thermal conductivity = κ_e (electronic thermal conductivity) + κ_{lat} (lattice thermal conductivity)] are strongly intercorrelated and make a formidable task to realize high value of zT.[1] The only independent parameter in this equation is κ_{lat}, reducing of which would be highly effective in triggering the overall TE performance.[2,3] The zT value around unity used to be a yardstick for the advancement of thermoelectric materials; however, recent developments have made a new paradigm shift with values much above unity. Doping of external elements can substantially optimize the carriers, modulate the electronic structure, or affect the phonon dynamics[4] and, thus, have a significant impact on the TE performance. Intriguingly, flexible TE materials have successfully been integrated into body-worn fabrics, which simply use heat from body to generate electricity. Therefore, thermoelectricity can be anticipated as one of the potential prime candidates in near future energy management.

MODULATION OF ELECTRICAL TRANSPORT

Carrier concentration optimization has always been an efficient route to improve TE performance in solids. For instance, Ag concentration in CaAgP has a significant impact on its carrier concentration where $CaAg_{0.9}P$ is found to be the most effective one with $zT = 0.43$ at 660 K.[5] Ce doping in the SnSe crystal, on the other hand, shifts the carrier types from *p*-type to *n*-type where the electron concentrations increase from 10^{16} to $10^{19}\,cm^{-3}$.[6] The introduction of Pb into the hexagonal Bi_2Te_2Se behaves as an electron acceptor and gradually moves the *n*-type electronic transport toward *p*-type.[7] Conversely, substituting Cu with Ga in $Ba_8Cu_{16}P_{30}$ clathrate reduces the hole concentration, which results in a higher S compared to that of pristine counterpart.[8] Moreover, Ag doping in $Zn_{0.625}Cd_{0.375}Sb$ causes the formation of Zn vacancies, which has a strong influence on the temperature variation trends of the electronic transports.[9]

Electronic structure engineering is an extremely effective tool in designing highly efficient TE materials. Recently, it has been shown that Se impurities in $Ag_2S_{1-x}Se_x$ modify the conduction band minimum, reduce the bandgap, and lower the effective mass of electrons, which ends up with improved carrier mobility.[10] On the other hand, strong asymmetries in the transport between valence and conduction bands can permit remarkable phonon limited electronic conductivity resulting in elevated power factor $S^2\sigma$ in half-Heusler material family.[11] Valence band convergence has a profound effect on S, and thus, highly converged valence bands in $CaCd_2Sb_2$ alloyed $CaMg_2Sb_2$[12] and $Sn_{0.57}Ge_{0.3}Sb_{0.1}Mn_{0.03}Te_{0.95}Se_{0.05}$[13] assist to achieve high Seebeck

coefficient. As a result, a high zT of ∼1.35 at 823 K is realized in $Sn_{0.57}Ge_{0.3}Sb_{0.1}Mn_{0.03}Te_{0.95}Se_{0.05}$. $Yb_{14}MSb_{11}$ (M = Mn, Mg, etc.) has recently attracted broad attention to the thermoelectric community as high-performance *p*-type TE materials. However, the complex crystal structure of these Zintl compounds impedes to comprehend the electronic band structure correctly, which makes a daunting task to engineer the bands. Using molecular orbital (MO) theory, the Zintl chemistry of $Yb_{14}MSb_{11}$ compounds has been delineated. Electronic band structure calculations demonstrate that the conduction band minimum in $Yb_{14}MgSb_{11}$ and $Yb_{14}AlSb_{11}$ is comprised of either antibonding MO, which is created from $(Sb_3)^{7-}$ trimer, or mixture of Yb, M, and Sb atomic orbitals.[14] In addition, the nonbonding p_z orbital of Sb contributes to the singly degenerate valence band. Such chemical comprehension of the electronic structure empowers the strategies to engineer the electronic transports in these materials. Parzer *et al.* have established that with increasing the Al concentration in Fe_2VAl_x, impurity resonant state formation takes place near the valence band edge, which drastically increases the power factor.[15]

MIMIMIZING THERMAL CONDUCTIVITY

Microstructural engineering, including the introduction of point defects, nano-/micro-structures, and grains boundaries, in the matrix of a material has always been proven to be one of the useful phonon blocking strategies to suppress κ_{lat}, which is beneficial for overall improvement in the TE performance. For example, the incorporation of Cu in $Bi_2Te_{2.7}Se_{0.3}$ drives several point defects formation, which significantly scatter the heat carrying phonons and the zT reached around 0.98 near room temperature.[16] Conversely, the inclusion of disordered nanopores in the thin film of $Ca_3Co_4O_9$ can result in a remarkably reduced κ.[17] Using perturbed molecular dynamics, Fujii *et al.* have demonstrated that grain boundaries with various length scale in two different directions of MgO have different contribution to the scattering of phonons.[18] However, this nano-structuring often inhibits the electronic mobility and, thus, materials with intrinsically low κ_{lat} are highly desirable. In this regard, Wang *et al.* have shown that the strong anharmonic phonon scattering through the Umklapp process and acoustic–optical phonon coupling causes very low κ_{lat} in single crystalline Bi_2O_2Se and the maximum zT reaches ∼0.188 at 390 K.[19] The van der Waals heterostructure of layered Bi_4GeTe_7 also exhibits intrinsically ultra-low $\kappa \sim 0.42\ W\ m^{-1}\ K^{-1}$ at 380 K due to the presence of large anharmonicity in the lattice.[20]

NEW THERMOELECTRIC MATERIALS

The involvement of theoretical studies and predictions in materials have remarkably advanced this field of thermoelectrics. Rao *et al.* have elucidated that the metastable R8 phases of Si can be used as a TE power generator as it exhibits one magnitude lower κ_{lat} compared to that of stable diamond-like cubic Si throughout the temperature range of 300–500 K.[21] Using 4 atoms in the primitive unit cell and formation energy within 0.3 eV of the convex hull, Berland *et al.* have shown for 1093 cubic materials that many of them are narrow bandgap semiconductors, which were wrongly interpreted as metallic previously.[22] Interestingly, Wang *et al.* have combined theoretical comprehension with experimentally observed results to simulate a method to realize the electrical transport in polycrystalline SnSe.[23]

The advancement of thermoelectrics always seeks for new materials and compositions. Semi-metallic $Cr_{0.91}Te$ recently has emerged as a multifunctional material with desired magnetic entropy change and promising zT. The trio effect of enhanced Seebeck coefficient due to the magnon drag effect and suppressed κ_{lat} originating from the phonon–spin coupling leads to such an encouraging TE performance.[24] Interestingly, Kuga *et al.* show that the anisotropies in *S* and σ have different origins of Fermi surface and hybridization between conduction and 4f electron, causing the concurrent increase in thermopower and σ in α-$YbAlB_4$, which can be a potential TE material.[25] From temperature variation of transport properties, Kim *et al.* have investigated the band structure of semi-metallic 2D $PtSe_2$ nanosheet, which can be fruitful to understand the band structure of other 2D materials as well.[26]

DESIGN AND INTEGRATION OF DEVICES

Despite improving the figure of merit above unity for several TE materials, achieving high energy conversion efficiency (η) of the devices for practical applications constructed by those materials is a daunting job. Fujimoto *et al.* have proposed a method to generate TE power via transverse thermo-spin conversions. They have demonstrated that by fastening ferromagnets to a material with a strong spin–orbit coupling, the nonequilibrium spin accumulation owing to the spin current converts into electrical voltage.[27] Tseng *et al.* have investigated the statistics of mechanical properties in silicide-based TE materials, which can be helpful to guide in engineering the reliable TE devices.[28] Baitinger *et al.* have successfully fabricated clathrate-based thermoelectric modules using $Ba_{7.8}Au_{5.33}Ge_{40.67}$ as *p*-type and $Ba_8Ga_{16}Ge_{30}$ as *n*-type legs on basis of an in-depth investigation of the phase diagram, demonstrating the potential of type-I clathrates for waste heat recycling.[29] A η of ∼5.5% is achieved in a two-pair module assembly using $Cu_{26}Cr_2Ge_6S_{32}$ and $Pb_{0.98}Ga_{0.02}Te$-3% GeTe, keeping the temperature difference between hot and cold end $\Delta T = 390$ K.[30] Similarly, Knura *et al.* have offered double-tuned functionally graded TE material and fabricated *n*-type $Pb_{0.75}Sn_{0.25}Te_{1-x}I_x$ leg device, which is shown to exhibit a very high $\eta \sim 12.0\%$ for $\Delta T = 540$ K.[31] The TE power density of $\sim 218.8\ \mu W\ cm^{-2}$ for $\Delta T \sim 41$ K can be obtained in assembled *p*-type $Bi_{0.5}Sb_{1.5}Te_3$ and *n*-type Bi_2Te_3 flexible wearable TE films.[32] Interestingly, a TE power generation device of SiGe combined with a Pd catalytic combustor is observed to exhibit maximum power density as high as $81\ kW\ m^{-3}$, equivalent $0.4\ W\ cm^{-2}$ assuming a 2 mm leg length.[33] Yan *et al.* systematically demonstrated that Peltier heat can increase the η in Bi_2Te_3-based two-segmented module if Seebeck coefficient along the electric current increases.[34] Finally, He *et al.* have demonstrated that CoO-based magnon junctions, i.e., $Y_3Fe_5O_{12}Y_3Fe_5O_{12}/CoO/Y_3Fe_5O_{12}$, can be utilized as a primary developing block to assemble ambient condition magnonic devices and circuits, which can have possible applications in magnon transistor, logic, and memory.[35]

THERMOELECTRIC THIN FILMS

Thin films always stay at the vanguard of research in thermoelectrics for the low-power applications. Hoang *et al.* have shown that the inclination of microstructure in $CuCr_{0.85}Mg_{0.15}O_2$ is dependent on the thickness of films that tailors the thermopower.[36] By using solution process, textured $SnSe_2$ thin films with preferred crystallographic orientation are successfully fabricated, and a power factor of $3.69\ \mu W\ cm^{-1}\ K^{-2}$ is further realized.[37] Improvements in the power factor are also observed in Mg_3Bi_2 films grown on sapphire,[38] B hyper-doped Si nanocrystalline thin films,[39] and P doped SiGe-based thin films.[40] Lima *et al.* have

revealed that Ga doping in the Mg_2Sn thin epitaxial film serves both the roles of phonon scattering centers and carrier concentration enhancer.[41] On the other hand, Al, Ga, and In have different impact on the thermoelectric properties of a ZnO thin film due to the different extent of distortion in the hexagonal wurtzite crystal structure of ZnO originating from the ionic size mismatch between Zn and the dopants.[42] Also, an outstandingly high S of 1.5 mV K^{-1} is observed in oxidized thin films of Ru_2Si_3.[43] Khodzitsky *et al.* have shown that the voltage responsivity of THz photothermoelectric detector built on the $Bi_{88}Sb_{12}$ frequency-selective surface is three times higher compared to the detector based on a continuous film.[44] These achievements provide effective approaches to optimize the thermal and electronic transport properties of thin films.

FLEXIBLE THERMOELECTRICS

Flexible thermoelectric materials, which can be efficaciously integrated into body-worn fabrics, have recently drawn huge interest to the scientific community because of its capability to generate electricity utilizing the body heat. Theoretical prediction on boron–nitrogen dimer doped graphphenyl shows a promising result where the thermoelectric properties can be tuned by rotating the angle of the phenyl ring.[45] Lu *et al.* have shown the great possibility of adjusting the thermoelectric parameters of poly(3,4-ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS) films for biomedical purposes.[46] Bi_2Te_3 blended PEDOT:PSS exhibits very low thermal conductivity compared to that of pristine Bi_2Te_3 due to the phonon scattering at the interface. This composite shows a remarkably high zT of ∼1.19 at 405 K.[47] Similarly, the device fabricated by spraying the starch-based biopolymer/graphene nanoplatelets ink onto the cellulose paper followed by PEDOT:PSS spraying demonstrates synchronized elevation in S and σ.[48] Ag_2X (X = S, Se, and Te)-based materials also are potential candidates for flexible thermoelectrics. While I doped deformable and ductile Ag_2S displays a promising performance owing to the improved carrier concentration,[49] the bacterial cellulose/Ag_2Se nanocomposite yields to the power factor of ∼3 μW cm^{-1} K^{-2} due to the changes in the carrier concentration and carrier mobility.[50] The degree of crystallization in $Ag_2S_xTe_{1-x}$, on the other hand, differs according to the S:Te ratio where $Ag_2S_{0.4}Te_{0.6}$ is comprised of two S and Te-rich $Ag_2(S,Te)$ glass states. The Ag_2S-based glass acts like a Newtonian fluid leading to the outstanding ductility, which shows an efficient route to design flexible inorganic materials.[51]

CONCLUSION

We are delighted to publish this special topic on thermoelectric materials science and technology toward applications in *Applied Physics Letters*. These set of articles offer a remarkable advancement of the state of the art of this field and provide the readers an effective path to acquire uptodate perceptions into this rapidly advancing field of materials science. We, the guest editors, thank all the authors and reviewers for making it happen.

DATA AVAILABILITY

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

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