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Mg_3Sb_2-based thermoelectrics: materials, interfaces, and devices

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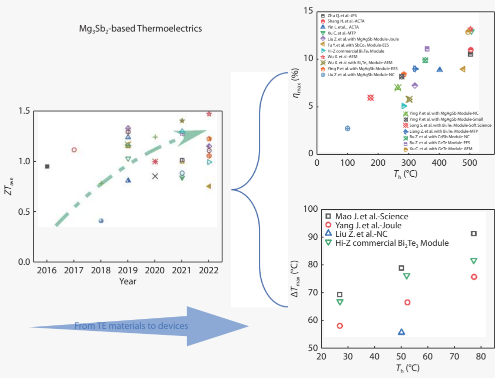
Thermoelectric power generators enable the direct conversion between waste heat and electricity near room temperatures, providing an environmentally friendly solution toward mitigating the ever-increasing global energy issues. Over the past years, we have witnessed significant advances in Mg_3Sb_2-based thermoelectric conversion materials. However, the device-relative efforts lag behind the materials-level works. In this mini-review, we summarize the advances in Mg_3Sb_2-based thermoelectrics from materials to devices. Further, we shine some light on the device-level challenge, including the design of thermoelectric interface materials, the stability issue, and the system-level full-parameter optimization. Finally, we discuss the new application scenarios exploration to inspire confidence in device-level efforts towards practical applications.



Thermoelectric (TE) power generation devices realize the direct conversion between heat and electricity, presenting many indispensable applications in special scenarios[1–3] such as energy supply of space explores [4], massive sensors for the internet of things (IoT) [5,6], and waste heat recovery from power plants, car exhaust, or fossil fuels [7,8]. In addition, the cooling function of TE devices shows great potential because of the precise and fast micro-zone temperature control [9,10], especially for the thermal management of microchips [11,12]. Very recently, high-efficiency TE devices assembled with state-of-the-art TE materials are frequently emerged in the prestigious news, such as PbTe [13], half-Heusler [14], GeTe [15], SnSe [16], Cu_2Se [17], $AgSbTe_2$ 2, Mg_3Sb_2[9], and so on. In particular, eco-friendly, and cost-effective Mg_3Sb_2-based TE devices show extraordinary popularity and desirable superiority at a middle-low temperature range [9,10,18–20], which are expected to replace N-type Bi_2Te_3[21,22] that is the exclusive TE material presently applied in the commercial market. However, the most mature Bi_2Te_3 and its alloys thermoelectrics are still dominant for the widely accepted solid-state or limited power generation application in the commercial market [21,23]. Recently, Mg_3Sb_2-based thermoelectric materials attract exclusive attention in either the middle-low temperature range or near room temperatures application for the rich abundance of Mg as compared with the rare element Te in the Bi_2Te_3 [9,10,18–20]. Compared with quick and significant progress in the Mg_3Sb_2-based TE materials, the Mg_3Sb_2-based devices progress hardly due to the challenges in electrode interface design and device assembling [24,25]. As a result, there is still a yawning gap between theoretical and experimental conversion efficiency (η), and output power density (ω) for the reported Mg_3Sb_2-based TE devices [18–20,26–38].

Before moving to the main content, we would like to clarify the terminology of the key component materials of a TE device based on our previous work [24]. For example, there are anode materials, cathode materials, electrolytes, and separators for the ionic battery [39]. The assembling of a TE device also requires similar key component materials, such as (a) thermoelectric conversion materials (TEcMs) that realize the energy conversion, (b) thermoelectric interface materials (TEiMs) that play an important role to maintain the reliable and efficient transport of heat and electricity between electrode strip and TEcMs, (c) protection material, usually used to prevent the evaporation of the TEcM and/or thermal insulation, (d) housing materials, providing structural support and electrical insulation. The aforementioned four types of materials just follow the four quadrants of a two-dimension space expanded by electrical conductivity (σ) in the vertical axis and thermal conductivity (κ) in the horizontal axis. The classic n and p-type materials fall in the second quadrant, which requires a high σ and low κ, referred to as TEcMs. While the metallization layer materials require a high σ and high κ, setting at the first quad-

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rant, referenced as TEiMs [24].

In this mini-review, we focus on the recent new progresses from the Mg_3Sb_2-based TE materials to devices. Further, we discuss the device-level challenges including the TEiMs design, the stability optimization, and the system-level full-parameter optimization. In the outlook, we also look ahead to the new application scenarios exploration towards practical applications.

Progress in Mg_3Sb_2-based TEcMs

Mg_3Sb_2 has a VB group element (Sb and Bi, respectively). Sb has 3-valence electrons in Mg_3Sb_2, which could be considered an inverse structure of Bi_2Te_3. The crystalline structures of Mg_3Sb_2 and Bi_2Te_3 in trigonal laminar structure, where Te atoms at 3a site (0, 0, 0) are donated Te^1 while Te atoms at 6c site (0, 0, 0.79030(5)) are Te^2. For each atomic layer in both compounds, atoms are stacked in a hexagonal pattern, and these two compounds share the same layer-to-layer stacking sequence, and this is the intrinsic reason for its low lattice thermal conductivity. Furthermore, the high valley degeneracy and relatively low-energy-closed point with the CBM point make Mg_3Sb_2 family the promising TEcM[40].

Mg_3Sb_2-based TEcMs were originally reported that could make the transition between p-type and n-type, being n-type on the side with excess Mg and p-type on the side with excessive Sb in 1956[57,58]. However, it regains our sight since the discovery of the excellent n-type TE properties in 2016[42,59]. Subsequently, Snyder *et al.*[43,50], Pei *et al.*[44,49,60], Liu *et al.*[46], and Ren *et al.*[9] nearly simultaneously reported exciting TE performance near room temperature (below 300 °C) in 2018 and 2019, showing the great potential of Mg_3Sb_2-based TEcMs for the substitution of commercial n-type Bi_2Te_3. In addition, Liu *et al.*[54], Sui *et al.*[56] and Chen *et al.*[45] successfully realize a high *ZT* of over 1.8 at 500 °C, allowing the high average ZT_{ave} Mg_3Sb_2-based TEcMs and thus benefiting the TE devices over a wide temperature range (Fig. 1).

However, the stability of Mg_3Sb_2-based TEcMs remains a great concern in practical applications. Many investigators exposed that the underlying reason causing the degradation was the Mg loss[51,52,59,61]. Fortunately, efforts like the cation or interstitial site doping[52,53,60], hot deformation[10], Mg vapor annealing[43], and depositing protective coating[28,62] could alleviate Mg evaporation and improve the stability of Mg_3Sb_2-based TEcMs. For instance, Liu *et al.* showed that interstitial Mn[46] or Multi-elements transition metals (CrMnFeCoCu)[54] dopant could suppress the Mg-vacancy formation, thereby improving the thermal stability. Snyder *et al.*[53] and Pei *et al.*[60] reported that Yb, Y, and La doping on the cation site could stably control the carrier concertation. Further, Zhao *et al.*[10] exhibited a stable cooling module via a post-hot deformation process to stabilize the Mg vacancies concentration at ambient temperature. Very recently, Ren *et al.*[28] and Liu *et al.*[62] reported that spraying BN and Mg-Mn alloy coating could also suppress the volatilization of Mg and then achieve high thermal stability. Further, electrical and chemical stability are highly concerned. The high current density seems to have little effect on the TE properties[30,62], but the

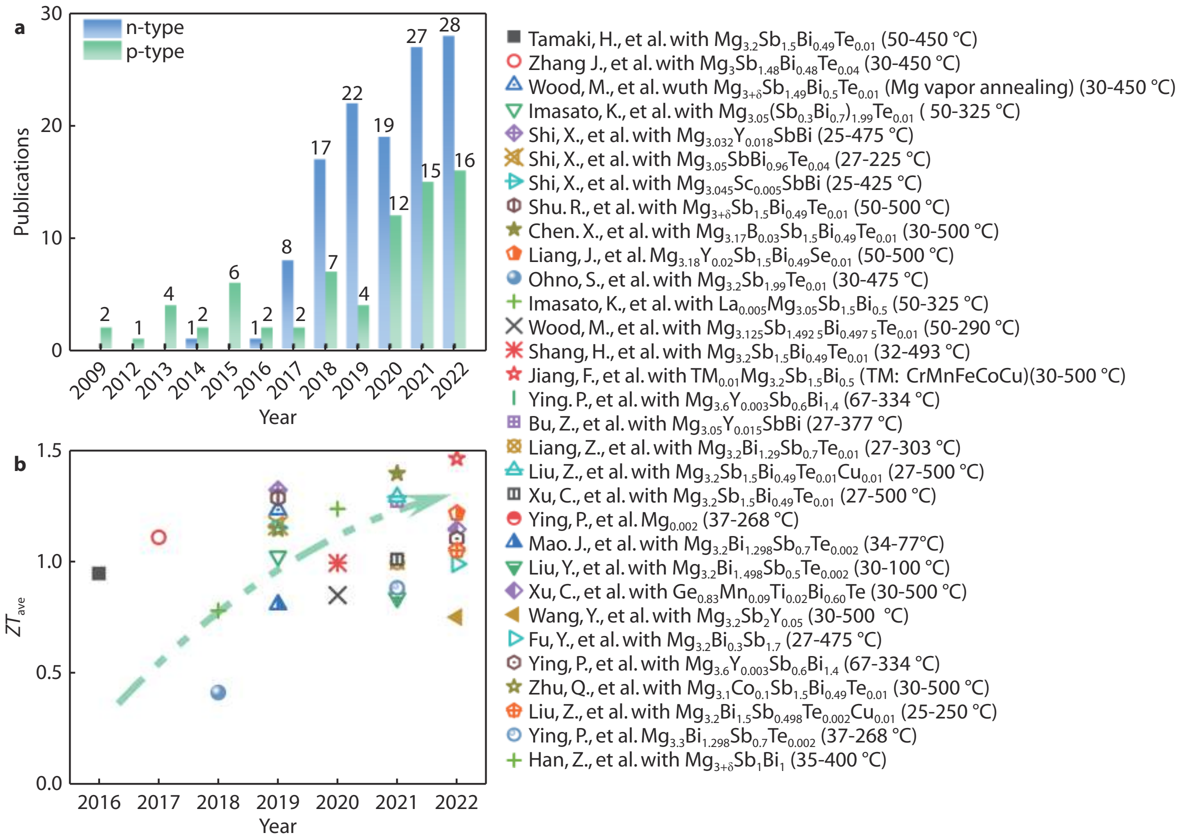

Fig. 1 **a** Publications and **b** ZT_{ave} vs. Year. Literature results[9,18,20,26-28,30-34,36,38,41-56] are included for comparison.

lifetime of Mg_3Sb_2-based TEcMs is more sensitive to moisture other than the considered oxygen[63]. Note that there is no specific parameter to quantify stability at present, and a general criterion to evaluate the stability is worth being defined in the future. Furthermore, the real service environment of a TE device is more complex; thus, we hope that multiple strategies mentioned above working together or creating new solutions to improve stability need to be developed in the future.

Progress in TEiMs for Mg_3Sb_2-based TEcMs

Although the Mg_3Sb_2-based TEcMs are booming and flourishing, the advances in relative devices are slowed and obstructed by the challenges at TEiM/TEcM interfaces[24,25]. The ZT_{ave} relationship between TEcMs and TE devices could bridge by the formula[64], $ZT_D = \frac{L}{L+2\rho_c\sigma}ZT_{ave}$, where ZT_D and are the effective value of TEcM ZT_{ave}. σ, ρ_c, L, and A are the electrical conductivity of TEcMs, specific contact resistivity, length, and cross-sectional area of the TE Leg, respectively. For a commercial TE device (L ~ 2 mm and σ ~10^5 S cm^{-1}), the ρ_c must be < 10 μΩ cm^{-5} and bonding strength (σ_s) should be > 30 MPa, thereby obtaining > 95% output performance and robust mechanical stability[24]. Generally, the fabrication of TE devices is based on soldering or brazing[65] to pursue high σ_s and low ρ_c then allow reliable and high output performance[66]. Unfortunately, most TEcMs exhibit poor weldability owing to their semiconductor behavior[25]; thus the TEiMs, or named metallization layers are inevitably required to realize a reliable bonding between TEcMs and electrodes[67]. Generally, a high σ_s requires necessary diffusion; however, it might also result in an increased ρ_c. It is difficult to balance the high σ_s and low ρ_c for a single-element TEiM; thus, the synergetic achievement in low ρ_c and high σ_s is a great challenge for the whole TE community because of the mutual exclusivity.

Interface contact problems have been extensively studied in classically commercial Bi_2Te_3 devices[68,69], but have not received sufficient attention in Mg_3Sb_2-based TE devices. Recently, researchers have attempted to use Fe [9,18,19,27,28,32], Ni[9,33], and Nb[34] as the TEiM layer for Mg_3Sb_2-based TE power generation devices. For instance, Ren *et al.*[31], Pei *et al.*[30], and Schierning *et al.*[18] respectively reported that the ρ_c of Fe/Mg_3(Sb, Bi)$_2$ interfaces are approximately 2.5, 14.6, and 26.6 μΩ cm^2 (Fig. 2). However, the ρ_c of Fe/Mg_3(Sb, Bi)$_2$ interfaces could increase to 60 μΩ cm^2 after the power generation service[19,29,30], suggesting that interface diffusion or reaction occurs. Similarly, Fu *et al.* used Nb as the TEiM for n-type Mg_3Sb_2 and achieved a low ρ_c of 9.7 μΩ cm^2 together with an increased ρ_c of 26 μΩ cm^2 after the high-temperature service, thereby creating a significant gap between the experimental and theoretical power generation performances[34]. Some alloying TEiMs for n-type Mg_3Sb_2-based TEcMs, including 304 stainless steel (304SS)[29], Mg_2Cu[10], $Fe_{90}Sb_{10}$[70], NiFe, NiCr, NiCrFe[35], $Mg_{3.4}Sb_3Ni$[36], Fe_7Mg_2(Cr, Ti)[37], and FeCrTiMnMg[62] were subsequently reported to show lower ρ_c values (Fig. 2). For example, Ren *et al.* reported that the NiFe/$Mg_3Bi_{1.5}Sb_{0.5}$ contact exhibits a highly stable ρ_c of 13 μΩ cm^2 after aging for over 2100 h[35]. In addition, we have suggested an alloying approach to design the TEiM rather than using available alloys, which has already been verified as an effective technique with some necessary characteristics, including high bonding propensity, coefficient of thermal expansion matching, diffusion passivation, and dopant inactivation[24]. As a result, Fe was selected from 15 elements as the matrix element, considering the criteria of high σ_s and low ρ_c, and further designed two ternary alloys (Fe_7Mg_2Cr and Fe_7Mg_2Ti) with σ_s > 40 MPa and ρ_c < 5 μΩ cm^2 37. Muti-elements TEiM (FeCrTiMnMg) has been further applied for the $Mg_3Sb_{1.5}Bi_{0.5}$ TE devices according to the above alloy screening principles, allowing the excellent interface stability and visually proven by in-situ TEM technique[62]. Of note, in the Mg_3Sb_2-based TEcMs family, the higher the carrier concentration, the lower the ρ_c[71]. Therefore, the TEcMs with different carrier concentrations should adapt to different TEiMs, and more attention ought to be devoted to the contacts investigation.

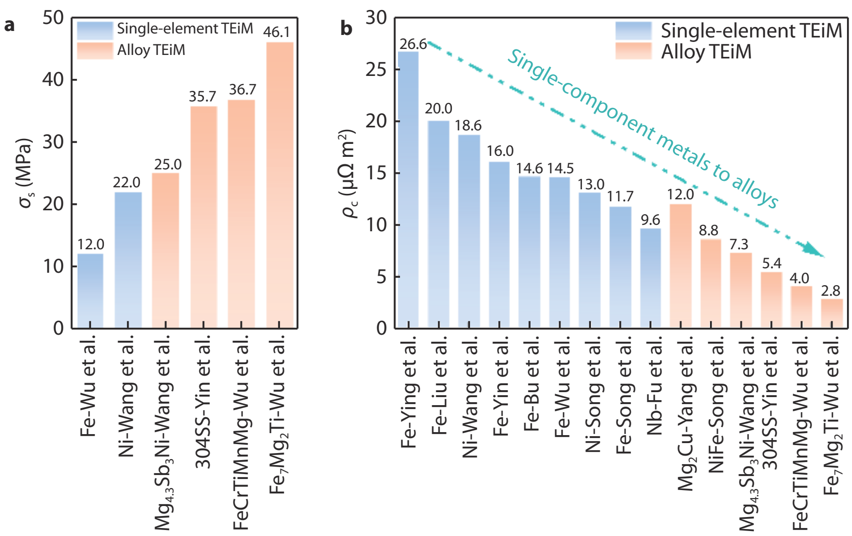

Fig. 2 **a** The initial values of σ_s and **b** ρ_c of the TEiM/TEcM interfaces near room temperatures. Literature results[10, 18, 20, 29, 30, 34-37, 62] are included for comparison.[58] Copyright 2022, Wiley-VCH.

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Progress in Mg_3Sb_2-based TE devices

For the past two or three years, state-of-the-art Mg_3Sb_2-based TE devices are originally emerging, exhibiting outstanding power generation performance in the temperature range from RT to 500 °C (Fig. 3). Early, *Ren et. al.* reported an Mg_3Sb_2-based single-leg TE device with a high η of 10.6% when hot side temperature (T_h) reached 500 °C in 2019[27]. Subsequently, *Zhang et. al.* applied 304SS as TEiM instead of single-element Fe and obtained an excellent single-leg device with a η of 9% at a T_h of 400 °C in 2020[29]. Further development begins in 2021; TE module devices with p-type Bi_2Te_3[9,10,31,35,48,62], MgAgSb[18,20,32,38,72], CdSb[19], GeTe[26,30], and SKD[34] were reported sequentially and yielded great success. During this flourishing period, the key factors considering practice application are gradually concerned, including scalable fabrication[33], electrical stability[30,62], chemical stability[63], and thermal stability[28,34–36,38,62]. We have mentioned above that reliable TE devices are contributed by optimizing TEcMs[10,38], designing TEiMs[29,34–36,62], and applying protective coating[28,62]. Typically, He *et al.* successfully fabricated reliable TE devices by higher temperature sintering and Mg heavy overcompensation, and the robust modules are proved via thermal shock testing over 30,000 times when T_h varied between 50 and 227 °C[38]. Wang *et al.* used Nb as the TEiM for n-type Mg_3Sb_2 and achieve a η of 9% in the Mg_3Sb_2-$CoSb_3$ modules, showing excellent thermal stability at a T_h of 500 °C [34]. The high ω_{max} of about 5.35 W cm^{-2} was achieved in the Mg_3Sb_2-GeTe device owing to the excellent power factor of TEcM and the low ρ_c of the TE devices[26]. Note that the ω_{ma} is unified by using the envelope area of devices. Very recently, Liu *et al.* have reported a multi-element TEiM (FeCrTiMnMg) and functional protective coating (Mg-Mn-based alloy) to de-

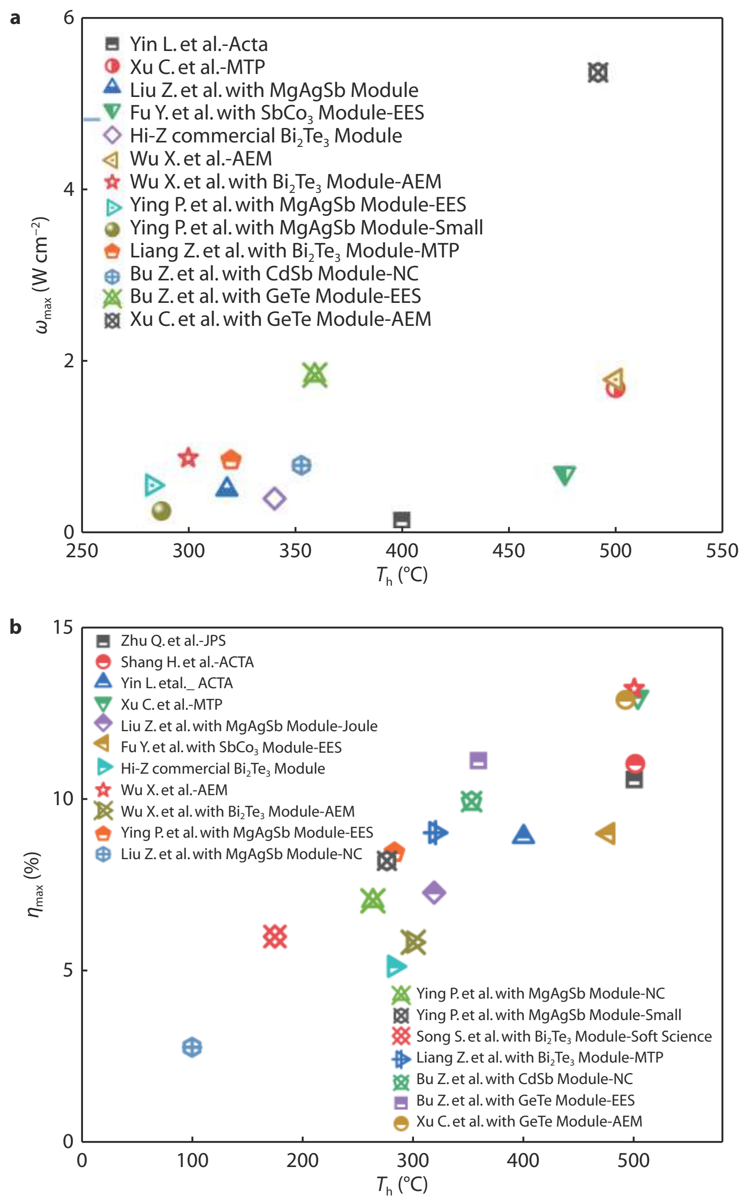

Fig. 3 T_h-dependent **a** ω_{max} and **b** η_{max}. Literature results are included for comparison[18-20,26-38,48,62,72-74].

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crease the chemical potential gradient, reduce the saturated vapor pressure, and increase the diffusion activation energy barrier, thereby realizing a high ω_{max} and η_{max} of 1.7 W cm^{-2} and 13% at a T_h of 500 °C; moreover, the optimized device exhibited excellent stability within 100 h and T_h cycled between 200-400 °C over 200 times with a TE performance degradation < 10 %[62]. In addition, employing device structure design and segmenting TE material with different Sb content are also progressed, thereby achieving outstanding device performance[18,26,31,72]. Further, new application models such as TE cooling[9,10,20,38] (Fig. 4) and flexible power generation devices[48] are pushed forward, which is likely to bring Mg_3Sb_2-based TE devices investigation into the era of explosion.

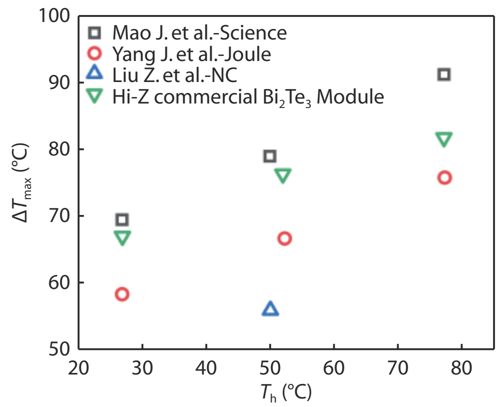

Fig. 4 T_h-dependent ΔT_{max}. Literature results are included for comparison[9,10,20,73].

Summary and Outlook

The TE community has put great efforts into Mg_3Sb_2-based thermoelectrics, however, the challenges at the device level remain critical and must be overcome before industrial implementation. Seeking high *ZT* is always a primary orientation, but the stability of TEcMs especially at elevated temperatures should not be ignored. In addition to the synthesis process and composition optimization, new ways to improve stability without sacrificing TE properties deserves our consideration. The mechanical properties are vitally important, especially for micro-TE devices. Li *et al.* optimized the mechanical properties through the nano-SiC-dispersed[75] and proposed the microelectromechanical technology fabricating micro-TE devices[76], broadening the exclusive dominance for Bi_2Te_3-based thermoelectrics. Therefore, optimizing the mechanical properties of Mg_3Sb_2-based thermoelectrics is absolutely an attractive aspect to broaden the new application scenario.

Designing TEiMs is a key to fully realizing a high device-level performance. Alloying appears to be the trend for rapidly and effectively screening TEiMs; however, more advanced TEiMs still require more scholars to devote. For real TE application systems, collaborative full-size optimization and device life prediction are all indispensable domains, which still need to be well established in Mg_3Sb_2-based thermoelectrics.

Exploring new application scenarios is also a keyway to promoting the development of Mg_3Sb_2-based thermoelectrics, including but not limited to power generation, cooling, sensing, and thermal management. Energy supply for massive IoT sensors[6] and wearable devices[48,77], human temperature perception system of electronic skin [78], micro cold storage for Covid-19 vaccine[79,80], temperature control for laser diodes, infrared detectors, charge-coupled devices, and computer chips[81], thermal cycling for DNA synthesizer[12], etc. have been initially developed. More importantly, exploring innovative applications requires our cross-collaboration in different research fields, thereby creating a green and beautiful lifestyle using thermoelectrics.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

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AUTHOR CONTRIBUTIONS

Xinzhi Wu: writing–original draft. Jian Huang, Zihan Zhou, Zhijia Han, Huan Li, Yupeng Wang, and Zhu Kang: revision. Weishu Liu: revision, supervision.

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