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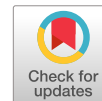
Scattering-tuned metal thermoelectrics, a new paradigm

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Scattering-tuned metal thermoelectrics, a new paradigm

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In contrast to conventional semiconductor materials, this review outlines a new strategy for metals to be high-performance thermoelectric materials. We have proposed “intrinsic energy filtering” as a strategy to overcome the inherently low Seebeck coefficients in metals. This strategy involves engineering electronic structures where a dispersive conduction band and a localized flat band coexist near the Fermi level, creating strong energy dependence in the carrier relaxation time, thereby generating a large Seebeck coefficient S . Effectiveness has manifested in distinct material systems; Ni–Au alloys, where disorder-mediated s – d interband scattering achieves exceptionally high power factors $\sim 34 \text{ mW m}^{-1} \text{ K}^{-2}$. Ni₃Ge exhibits a relatively large $S \sim -80 \text{ } \mu\text{V K}^{-1}$ through interband scattering. The strategy was further extended to the Kagome metal Ni₃In, with topological flat bands, revealing that Zener tunneling can suppress S , and we outline strategies to circumvent this quantum transport phenomenon. These developments point to a new horizon for the development of next-generation thermoelectric materials, unlocked by the precise control of scattering mechanisms. © 2026 The Author(s). Published on behalf of The Japan Society of Applied Physics by IOP Publishing Ltd

1. Introduction

Thermoelectric power generation is a unique technology that can convert waste heat into useful electrical power by solid-state devices without moving parts, and has therefore attracted attention for the expectation of improving energy efficiency and the possible contribution to the development of carbon-neutral society.^{1,2)} Recently, thermoelectric power generation is also expected to be utilized as the power source of IoT sensors.^{3–5)} The thermoelectric device, which is often referred to as a thermoelectric module, consists of arrays of n -type and p -type thermoelectric materials. Actually, the principle is the same as the Peltier module, which can function as a heat pump by passing an electric current through the device.

The conversion efficiency of a power generation device is described as

$$\eta = \frac{T_H - T_C}{T_H} \left[\frac{(1 + zT_{\text{ave}})^{1/2} - 1}{(1 + zT_{\text{ave}})^{1/2} + (T_C/T_H)} \right], \quad (1)$$

where T_H and T_C are the temperature at the hot and the cold sides, respectively. zT represents the dimensionless figure of merit of thermoelectric materials and is defined by $zT = S^2 T / \rho \kappa$, with S , T , ρ , κ being the Seebeck coefficient, absolute temperature, electrical resistivity, and thermal conductivity, respectively. Here, the thermal conductivity can be divided into two terms, $\kappa = \kappa_{\text{latt}} + \kappa_{\text{elec}}$, where κ_{latt} and κ_{elec} are the thermal conductivities due to lattice (phonon) and electrons. zT_{ave} in Eq. (1) indicates the temperature-average of zT . With the increase of zT , the conversion efficiency η monotonically increases and eventually reaches the Carnot efficiency $\eta = (T_H - T_C)/T_H$. Thus, developing thermoelectric materials with high zT is important. However, increasing zT is challenging because of the trade-off between S , ρ , κ .^{6–9)}

A lot of works have been devoted to overcome this issue. As a result, state of the art TE materials record zT of nearly 3.^{10–13)} One of the guiding principles is to utilize semiconductors with low thermal conductivity. This has been achieved by various ways of phonon-engineering, including using materials with complex crystal structures or large unit cells,^{13–15)} with weak-bonding units that can bring anharmonic lattice vibrations,^{16–21)} or by introducing nano structures to reduce the phonon mean free path.^{22–25)}

Now, we consider the electronic requirements, namely, the strategies to achieve high Seebeck coefficients and high power factors (PFs), S^2/ρ . First of all, a semi-classical approach has been adopted to describe electronic transport in materials. From the Mott formula, the Seebeck coefficient of a degenerate system is described as

$$S = \frac{\pi^2 k_B^2 T}{3 |e|} \left| \frac{\partial \ln \sigma(\epsilon)}{\partial \epsilon} \right|_{\epsilon=E_F} \\ \cong \frac{\pi^2 k_B^2 T}{3 |e|} \left| \frac{\partial \ln N(\epsilon)}{\partial \epsilon} + \frac{\partial \ln \tau}{\partial \epsilon} \right|_{\epsilon=E_F}, \quad (2)$$

Here, $\sigma(\epsilon)$, $N(\epsilon)$, and τ represent the spectral conductivity, electronic density of states (DOS), and the carrier scattering time, respectively.^{26,27)} Normally for semiconductors, the carrier scattering time τ can mostly be regarded as a constant within the narrow energy window of $k_B T$. Thus, the second term in Eq. (2) is neglected. Therefore, it is required that the DOS has a sharp energy dependence around E_F to exhibit a large Seebeck coefficient. Carrier-doped semiconductors are therefore suitable, as $N(E)$ shows a sudden change near the band edge. In contrast, normal metals such as Cu and Ag are not good TE materials because the energy dependence of the DOS is very small at E_F . Up to now, semiconductor-based TE materials have been very promising to achieve high zT and high conversion efficiency. On the other hand, the PF, S^2/ρ , is a direct measure of the output power, and has been another fundamental factor to measure the performance of



TE materials. Historically, there have been well known high PF materials in intermetallic alloys and compounds but these systems were not extensively studied for TE application, mostly because of their low zT due to high thermal conductivity. However, when it comes to the power generation from abundant waste heat resources, PF can be a more critical and practical factor than zT , as it is related to the upper limit of the minable power density per area.²⁸⁾ This is especially true when one can use cooling water or efficient cooling tools for the low temperature part. Then, intermetallic compounds and transition metal alloys could become very attractive as candidates for high power-factor TE materials. In addition to the high output power density, these metallic TE materials have other advantages; including mechanical robustness, thermal stability, easy processability and manufacturability, and so on. Furthermore, recently, the possibility of active cooling with Peltier devices^{29,30)} is attracting attention due to the rapidly increasing heat problems in data centers and power devices.³¹⁾ In this case, a high thermal conductivity is also advantageous to transfer the heat out of the hot devices via thermal conduction through the legs, as well as further enhancing heat pumping capabilities via active cooling by the Peltier effect, which is directly proportional to PF. Thus, metallic TE materials with high PF and high thermal conductivity are now of great interest to contribute as possible solutions to the heat problems in society. Metallic thermoelectric materials can be roughly classified into several categories. We will briefly describe them below.

(a) *Intermetallic compounds with an energy gap or a pseudo gap at the Fermi level*

Intermetallic compounds are mostly metallic, but some of them are semiconductors or semimetals. This includes silicides, such as Mg_2Si , SrSi_2 , CrSi_2 , FeSi_2 , Ru_2Si_3 , MoSi_2 , etc.³²⁾ In addition, FeGa_3 , FeAl_2 , and RuAl_2 are also known to show semimetallic behavior with pseudo gaps, and are thereby studied as candidates of TE materials.^{33–35)} Furthermore, this class involves the full Heusler Fe_2VAl and Fe_2TiSi , and half-Heusler compounds like $(\text{Hf,Zr})\text{NiSn}$, TiCoSb , NbFeSb , and so on.^{36–40)} It is well known that for these Heusler series the valence-electron concentration (VEC) per atom is a good factor to predict the electronic state,⁴¹⁾ and $\text{VEC} = 6$ is the magic number for which the Fermi level of the Heusler compounds is situated within the energy gap or the pseudo-gap. This empirical relation can be further applied to design double- or triple-Heusler systems; like $\text{Ti}_2\text{FeNiSb}_2$ ^{42,43)} or $\text{Mg}_2\text{VNi}_3\text{Sb}_3$,⁴⁴⁾ etc. In this case, increased number of atoms inside the enlarged unit cell causes significant reduction in the lattice thermal conductivity, thereby achieving larger figure of merit.

For these intermetallic TE materials with an energy gap or a pseudo gap, the carrier concentration can be optimized by the elemental substitution. Thus, the basic strategy to enhance the TE performance is almost similar to the case of classical TE semiconductors. In the case of the Fe-based Heusler alloy, however, the dopant element sometimes modifies the band-edge structure. The effects of non-stoichiometry and/or the anti-site defect especially give a significant effect on

the electronic structure near the Fermi level, thereby large improvements in the PF are sometimes achieved due to the resonant level evolution, which were not expected from the rigid-band like VEC dependence.^{45–49)} Thus, electronic structure calculations give good guidelines.^{50–52)}

(b) *Metals or intermetallic compounds with strong electronic correlation*

Another classification of the metallic TE materials is metallic compounds which have simple Fermi surfaces, but possess large electronic DOS near the Fermi level. This quite often is the case for the strongly-correlated electron systems. Intermetallic compounds with Ce, Sm, Eu, and Yb can have intermediate valence state like $\text{Ce}^{3+ \leftrightarrow 4+}$ or $\text{Yb}^{2+ \leftrightarrow 3+}$ through the hybridization of conduction electrons and the $4f$ levels, which gives rise to a steep quasi-particle band called Kondo resonance near E_F . Here, the physical properties are basically described by a single quasi-particle band with significantly enhanced effective mass m^* , ranging from typically tens to several hundreds order of magnitude larger than that of the free electron. Indeed, a large Seebeck coefficients for metallic compounds of $S = -90 \mu\text{V K}^{-1}$ for YbAl_3 , and larger than $100 \mu\text{V K}^{-1}$ for CePd_3 were reported.^{53–55)} Other intermediate valence compounds such as Sm_4Bi_3 and EuCu_2Si_2 also show relatively large Seebeck coefficients for metals.⁵⁵⁾ Furthermore, a good universal scaling is observed between S/T and the electronic specific heat coefficient γ in a wide range of correlated electron systems,⁵⁶⁾ indicating that the above-mentioned Fermi liquid description well holds. On top of these relatively large Seebeck coefficients, the metallic nature with periodic lattice enables very low electrical resistivity. As a result, very large PFs have been observed; exceeding $10 \text{ mW K}^{-2} \text{ m}^{-1}$ even at room temperature.⁵⁷⁾

The origin of the large PF in these intermediate valence compounds can basically be understood in terms of the steep DOS at E_F due to the Kondo resonance. Wei et al. carefully compared the results of magneto thermoelectric effect and other physical quantities for YbAl_3 and suggested that the large Seebeck coefficient of YbAl_3 is understood by a simple narrow-band model.^{27,58)} In case of $4f$ -based intermediate valence compounds, a Lorentzian type quasi-particle band, $N(\epsilon) \propto \frac{\Gamma_f}{\epsilon_f^2 + \Gamma_f^2}$, is assumed to develop, where Γ_f and ϵ_f are the width of the $4f$ band and its distance from the Fermi level. Again, when the energy dependence of τ is not significant, the second term of Eq. (2) can be omitted, then the temperature dependence of Seebeck coefficient is expressed as:⁵⁹⁾

$$S = \frac{\pm \pi^2 k_B}{3 |e|} \frac{T \epsilon_f}{(\pi^2/3) T^2 + \epsilon_f^2 + \Gamma_f^2}. \quad (3)$$

Although phenomenological, this formula agrees well with the Seebeck coefficients in a wide temperature range for many compounds including YbAl_3 ,⁵⁸⁾ CeNi_5Sn , $\text{Ce}_2\text{Ni}_2\text{Ga}$,⁵⁹⁾ Yb_3Si_5 ,⁶⁰⁾ and so on. The fitting with Eq. (3) yields the values of Γ_f and ϵ_f , and the former should be comparable with the energy parameters like Kondo

temperature T_K estimated from other physical measurements. Good agreement between Γ_f and T_K is indeed observed in these compounds, indicating the validity of the universal behavior of Fermi liquid system.

In case of these intermediate valence compounds, optimization by carrier doping is not effective unfortunately, because of the large carrier concentration and large DOS. Instead, controlling the c - f hybridization to tune the parameters Γ_f and ε_f can be effective to modify the TE performance. Typically, this is actually done through substituting elements with smaller or larger ionic radii to vary chemical pressure on the system. In $\text{CePd}_{3-x}\text{Pt}_x$ ⁶¹⁾ and $\text{Yb}(\text{Si}_{1-x}\text{Ge}_x)_{2-\delta}$ or $\text{Yb}_3(\text{Si,Ge})_5$,⁶²⁾ the substitution leads to expansion of the unit cell, which causes a negative chemical pressure, and results in improved TE performance. Hence, the large PF in this class of materials can be basically understood in terms of a single-band (intra band) phenomenon, although the origin of the heavy quasiparticle band derives from a multi-body effect.

For Yb_3Si_5 , a moderately large PF of 5 to 6 $\text{mW K}^{-2} \text{m}^{-1}$ at $T = 72 \text{ K}$ for sintered samples has been obtained.^{60,63)} On the other hand, a very large PF of 40 $\text{mW K}^{-2} \text{m}^{-1}$ was observed at low temperatures by using a single-crystalline sample.⁶⁴⁾ Similarly, a PF of 34 $\text{mW K}^{-2} \text{m}^{-1}$ was reported for YbAl_3 at 80 K.⁶⁵⁾ There can be much room for further improvement for low temperature thermoelectric applications.

In case of compounds with large DOS near E_F , the electronic state can be affected by strong magnetic interaction, or quite often the compounds are magnetically ordered. Then, in addition to the normal diffusion thermopower, the magnon (spin wave) drag thermopower can sometimes be more dominant, as observed in ferromagnetic Fe, Co, and Ni.^{66,67)} The Heusler compound Fe_2VAl is also known to be close to magnetic ordering, and indeed ferromagnetic state can be induced by slight doping.⁶⁸⁾ A weak ferromagnetism was observed in Cr- and Fe-doped Fe_2VAl alloys, and a clear indication of spin-fluctuation drag thermopower was observed, causing 20% or much higher enhancement in PF.⁶⁹⁾ The merit of spin-fluctuation is that the interaction is available in wide temperature ranges even above the ferromagnetic transition temperature. In addition to these coherent magnetic excitation, Kondo-like local interaction with magnetic moments and carriers can also be found to yield enhanced PF.^{66,70)} Here, the carrier scattering by the other band may also be relevant, which will be introduced in detail below.

(c) *Metallic compounds with significant inter-band scattering*

The last class includes transition metal alloys and compounds and is based on a very new and intriguing concept, which is the main topic of this review article. The distinct difference in this class from the previous ones is that the energy dependence in the scattering time τ is significant, and therefore the second term in Eq. (2) plays the dominant role. This mechanism was shown to be essential in the heavy fermion compound CeCu_2Si_2 , which lies in the vicinity of quantum critical regime.²⁷⁾ Now, it becomes increasingly a promising guideline as a number of high PF materials have been discovered. These include inorganic semimetals, such as MoTe_2 ^{71,72)} and

Ta_2PdSe_6 .^{73,74)} In the former system, a critical scattering due to the polar-non polar structural transition is relevant to the asymmetry in the carrier mobility and the enhanced Seebeck coefficient, which results in a large PF of 30 $\text{mW K}^{-2} \text{m}^{-1}$.^{71,72)} In the latter, an extraordinarily high PF of 240 $\text{mW K}^{-2} \text{m}^{-1}$ has been recorded at low temperature,⁷³⁾ where the multiple bands with significantly different carrier mobilities are considered to be the source of strongly asymmetric scattering time.⁷⁵⁾ Furthermore, even in the well-known TE materials with high PFs, such as the chalcopyrite CuFeS_2 ^{76,77)} and the CoSb_3 -based skutterudites,^{78,79)} the energy dependence of the scattering-time is being considered to be significant.^{80,81)}

Thus, the utilization of the “scattering-time engineering” emerges as the new paradigm for the development of thermoelectric materials, in addition to the “phonon engineering” and the “band engineering.” From the viewpoint of materials design, however, controlling the carrier scattering time is highly difficult. In this review, therefore, recently proposed practical strategies are summarized. Details will be explained later, thereby only a brief introduction is given here. Essentially, the TE compounds in this class should contain multiband structures around E_F which embody different roles. There should be at least one dispersive band, which contributes to the electronic transport with a high mobility. On the other hand, there should be another (other) band(s) with rather non-dispersive DOS. Thus, carrier’s life time becomes very short through the inter band scattering in the narrow energy window where the two bands overlap.⁸²⁾ Outside of the overlapped energy region, on the other hand, τ should have only a weak energy dependence due to the dispersive band. Further, the Fermi level can be tuned so the energy comes close to the non-dispersive band by filling carriers in the dispersive band. This mechanism allows the energy-filtering of low energy carriers, or selective scattering of minority carriers in semimetals. Thus, this mechanism can potentially result in ultra-high PFs.

To illustrate the overall logic of this new paradigm, Fig. 1 summarizes the conceptual pathway from the initial band structure to the resulting thermoelectric performance, alongside the fundamental limitations. As depicted, while the engineered interband scattering creates a steep energy dependence in the relaxation time to boost the PF, this strategy must also navigate constraints imposed by quantum transport. Specifically, phenomena such as Zener tunneling at band crossing points can introduce additional conduction channels σ_{WTE} that smear the transport distribution, thereby suppressing the Seebeck coefficient. Based on this conceptual pathway, proposed candidates for the origins of non-dispersive bands are as follows: (i) heavily doped impurities like in Ni–Au alloy,⁸³⁾ (ii) intrinsically narrow 3 d bands,⁸⁴⁾ which potentially can be finely tuned to the E_F , and (iii) geometrically frustrated orbitals like in Kagome lattice.⁸⁵⁾ Because of the difficulty of calculating the carrier lifetime, this mechanism was not well studied. However, recent development in the microscopic theory of TE phenomena^{52,86)} may enable the precise treatment of this problem, with the help of precise calculation of the electronic structure of materials.

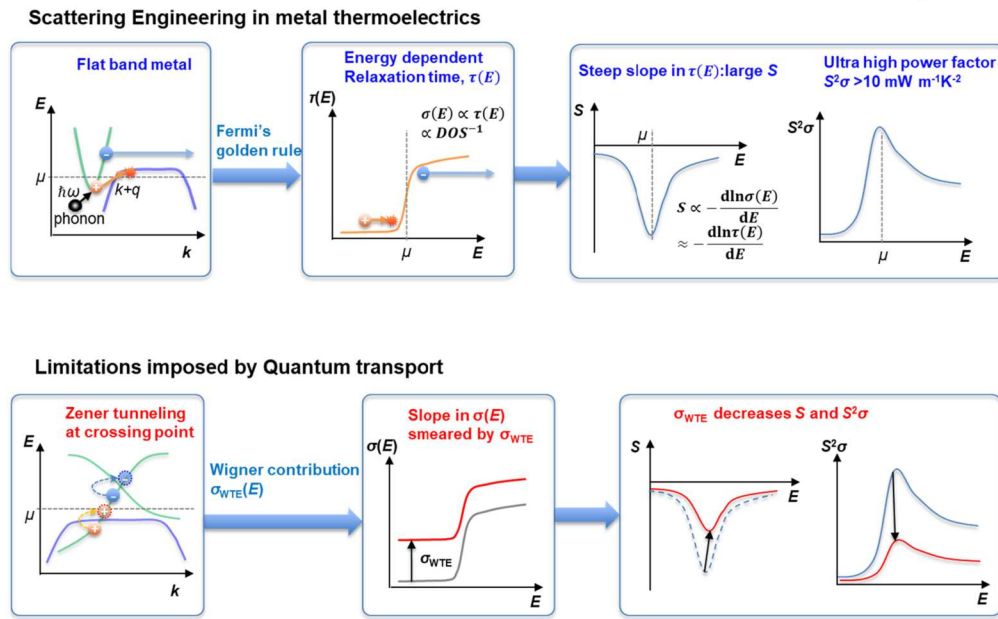


Fig. 1. Conceptual pathway of scattering engineering and the fundamental limitations imposed by quantum transport in metallic thermoelectrics. (Top) The “scattering-time engineering” paradigm. (Bottom) Performance suppression due to Zener tunneling.

In the following section, this category of TE compounds will be introduced. These metallic TE materials are new and have been relatively unstudied until recently, both experimentally and theoretically. However, they actually have exhibited ultra-high PF of the range of $10\text{--}30\text{ mW K}^{-2}\text{ m}^{-1}$ around room temperature. Hence, these materials have great potential of advancing the TE conversion from both large limitless waste heat sources which have some built-in robust cooling mechanism for the cold side, and also the active cooling technology which can be applicable to the explosively increasing number of data centers and power devices.

2. Interband scattering as novel paradigm of boosting TE performance of metals

Distinct from traditional semiconductor-centric approaches, a key research theme in recent years has been how to achieve high thermoelectric performance specifically a large PF in mechanically robust and low-cost metallic materials. Conventionally, common metals like Cu or Al have been considered unsuitable for thermoelectric applications due to their large carrier density and symmetric band structure around the Fermi level, where cancelation of electron and hole contributions results in a low Seebeck coefficient (S). In this section, we review an innovative material design strategy that overturns this conventional wisdom and examine its effectiveness and limitations based on our series of studies.^{83–85)}

The “Intrinsic Energy Filtering” strategy utilized in $\text{Ni}_{1-x}\text{Au}_x$ alloys⁸³⁾ involves designing the electronic structure of metals from the perspective of interband scattering, thereby realizing selective carrier scattering in bulk systems, similar to theoretical predictions for nanostructured materials. Specifically, two ordered materials that demonstrated the success of this strategy are Ni_3Ge with an L1_2 structure⁸⁴⁾ and the Kagome metal Ni_3In , which is predicted to be a topological flat-band (TFB) material.⁸⁵⁾ These studies have not only brought about a breakthrough in the field of metallic

thermoelectrics but have also opened new horizons in understanding charge transport in multiband metals featuring both dispersive and flat bands.

The performance of thermoelectric materials is determined by the S , κ , and electrical conductivity $\sigma = 1/\rho$. In particular, the PF ($S^2\sigma$) is a crucial indicator representing the quality of charge transport. According to the theory proposed by Mahan and Sofo, an ideal thermoelectric material should have a transport distribution function $\sigma(E)$ with a sharp, delta-function-like peak near the Fermi level (E_F).⁸⁷⁾ This implies that if only charge carriers with specific energy contribute to transport, both a large S and high σ can be achieved simultaneously.

The “Energy Filtering” strategy we implemented in recent works^{83–85)} is a concrete design guideline for optimizing the transport distribution in metals. Its core lies in the coexistence of a sharp peak derived from a flat band (FB) and a dispersive band (DB) that supplies conduction carriers near E_F in the electronic DOS $D(E)$ (see Fig. 2). Under this electronic structure, low-energy conduction carriers in the DB are strongly scattered into the FB, where abundant scattering states exist (interband scattering). Consequently, the relaxation time for low-energy carriers becomes extremely short. Conversely, carriers with higher energy are less affected by this scattering and thus possess a longer relaxation time. This strong energy dependence of the relaxation time $\tau(E)$ forms a steep edge in the transport distribution function $\sigma(E)$, generating a large electron–hole asymmetry despite the material being a metal, resulting in the manifestation of a boost of S .

3. Ultrahigh power factors in intermetallic alloys

Thomas Johann Seebeck first discovered the thermoelectric effect in metals more than 200 years ago, but since the mid-20th century, research has predominantly focused on semiconducting materials. Semiconductors inherently display a large Seebeck effect since the transport function $\sigma(E)$

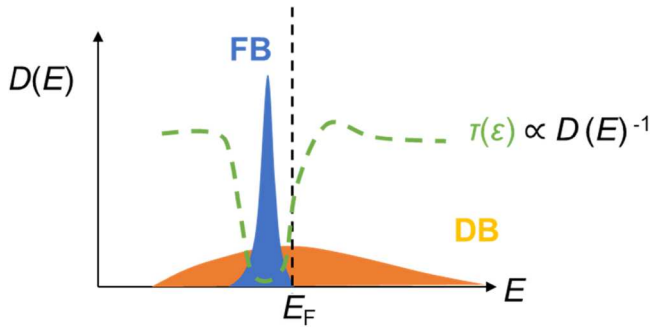


Fig. 2. Schematic diagram illustrating energy filtering. The density of states $D(E)$ shows the coexistence of a flat band (FB) and a dispersive band (DB). Interband scattering suppresses the contribution of low-energy carriers, forming a sharp edge in the relaxation time $\tau(E)$ around E_F .

displays large *relative* asymmetry near a band edge. This can be rationalized even from the simple Mott formula, which approximates the Seebeck coefficient by the *relative* asymmetry in $\sigma(E)$ via $S \approx \frac{1}{\sigma(E)} \frac{d\sigma(E)}{dE} \bigg|_{E_F}$. Since $\frac{1}{\sigma(E)}$ is large near a band gap, even a small term $\frac{d\sigma(E)}{dE}$ will result in a large S . On the other hand, the conductivity σ itself will be small, creating a tradeoff. Therefore, to realize a large PF (and ultimately zT) it is crucial to obtain a large S stemming from a large $\frac{d\sigma(E)}{dE}$, sometimes referred to as the “electronic quality factor” or “weighted mobility” in the field.^{88,89} Hence, the *absolute* asymmetry in $\sigma(E)$ must be large. In other words, the properties of the band edge must be such that the Seebeck will remain large even for higher doping and larger $\sigma(E_F)$. However, since TE performance in semiconductors is typically optimized at values of $\sigma(E_F)$ still far from those seen in good metals, heat transport is dominated by phonons $\kappa_{\text{latt}} \gg \kappa_{\text{elec}}$, requiring substantial tuning with respect to their lattice thermal conductivity. In many metals, on the other hand, κ_{elec} is sufficiently larger than κ_{latt} , particularly at high temperatures since κ_{latt} decreases with temperature due to phonon-phonon Umklapp scattering, whereas $\kappa_{\text{elec}} = L\sigma T$ (with L being the Lorenz number). This substantially simplifies the expression for the dimensionless figure of merit in metals to $zT \approx S^2/L$. We extracted thermoelectric property data for several metallic materials from the literature and plotted S versus L as seen in Fig. 3. Indeed, many metals satisfy $zT \approx S^2/L$, reducing the multidimensional optimization problem encountered in semiconductors to the singular challenge of obtaining a large Seebeck coefficient in metals.

From Fig. 3, it becomes clear that, next to intermediate-valence systems such as YbAl_3 and CePd_3 , the highest S in metals is realized in binary alloys containing group 10 and 11 elements. Indeed, it has long been known that binary alloys containing nickel and copper (e.g. constantan) or palladium and silver display an unusually large Seebeck effect, resulting in their application in temperature sensing as thermocouples. The underlying origin of their high Seebeck coefficient is rooted in a combination of almost fully filled d shells and partially filled s states (see Fig. 4). This creates a band structure that looks similar to the one sketched in Fig. 1, where charge carriers can scatter from the s bands into the more localized d states (s - d scattering) below E_F , while no such transitions are allowed above E_F (due to the lack of a substantial d DOS). Interestingly, while the interband

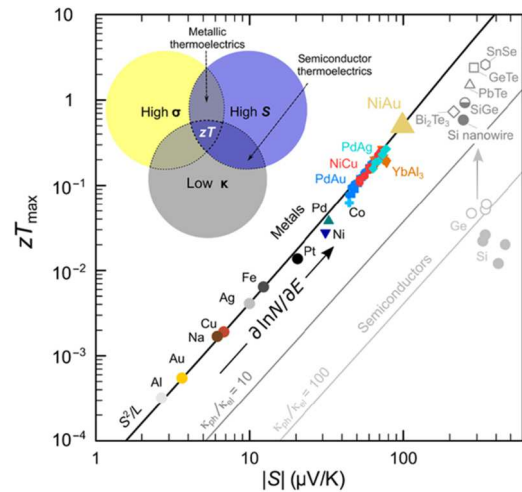


Fig. 3. Wiedemann-Franz scaling of figure of merit with Seebeck coefficient. When electrons dominate the heat transport over phonons, zT simplifies to $zT = S^2/L$, which is experimentally observed across the elemental metals and various binary metallic alloy systems. Semiconductors require tuning of the lattice thermal conductivity to approach the “metallic limit.” (This figure is reproduced from Ref. 83 under the Creative Commons Attribution 4.0 International License.)

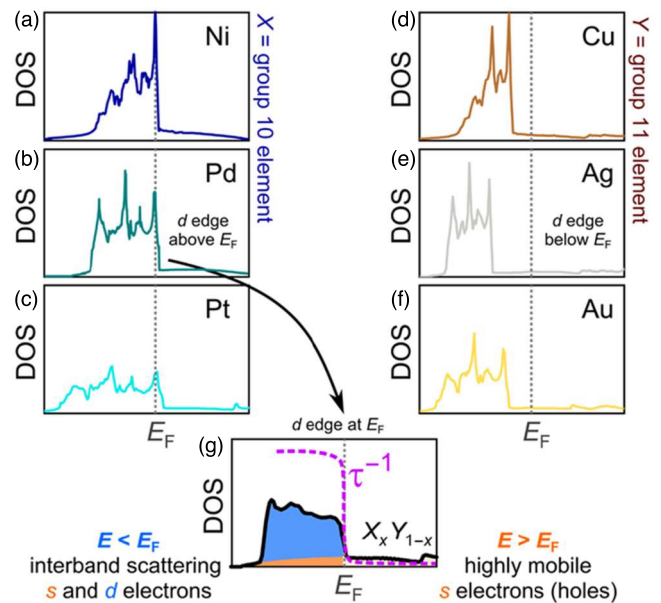


Fig. 4. Density of states of the group 10 and group 11 elements. Transition metals from the Ni group have almost fully filled d shells, creating a steep edge in their DOS just above the Fermi level. On the other hand, the DOS of the Cu group has fully filled d states and is characterized by an almost energy-independent behavior due to the broad s band. In binary alloys, the d edge can be tuned due to the difference in chemical potential between group 10 and 11 elements. (This figure is reproduced from Ref. 83 under the Creative Commons Attribution 4.0 International License.)

transitions are mostly mediated by phonons in pristine, ordered compounds, such as Ni_3Ge and Ni_3In ,^{84,85} the dominant scattering mechanism in highly concentrated $A_x B_{1-x}$ alloys (A = group 10 transition metal and B = group 11 element) is disorder, resulting in the famously temperature-independent resistivity in alloys such as constantan. The fact that interband transitions are mediated by disorder and not phonons implies that the Fermi surface and Fermi wavevector must be large enough such that interband

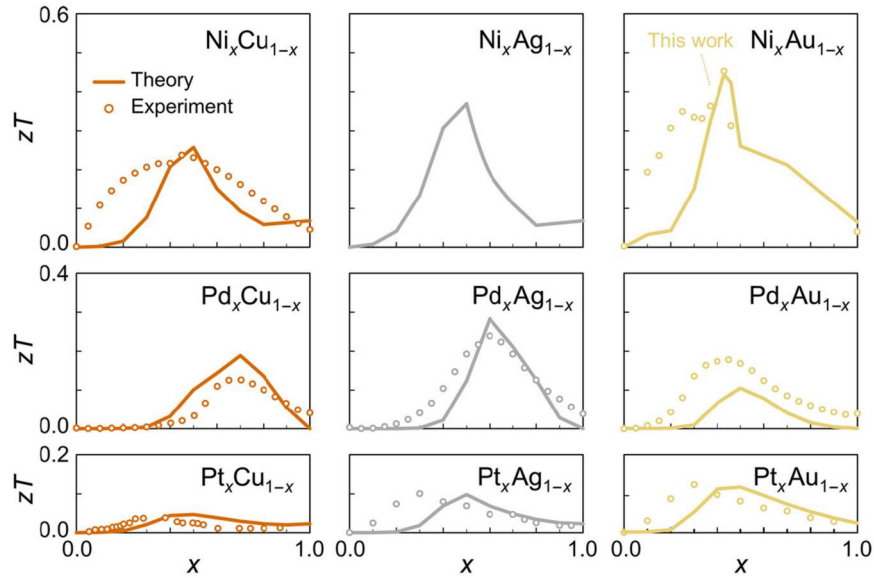


Fig. 5. Composition-dependent figure of merit for alloys of all combinations between the group 10 and 11 elements. Theoretical results were obtained by assuming $zT = S^2/L$ and S calculated from a simple $\tau(E) \propto D^{-1}(E)$ interband scattering model using the alloy-averaged $D(E)$ from density functional theory KKR-CPA calculations. (This figure is reproduced from Ref. 83 under the Creative Commons Attribution 4.0 International License.)

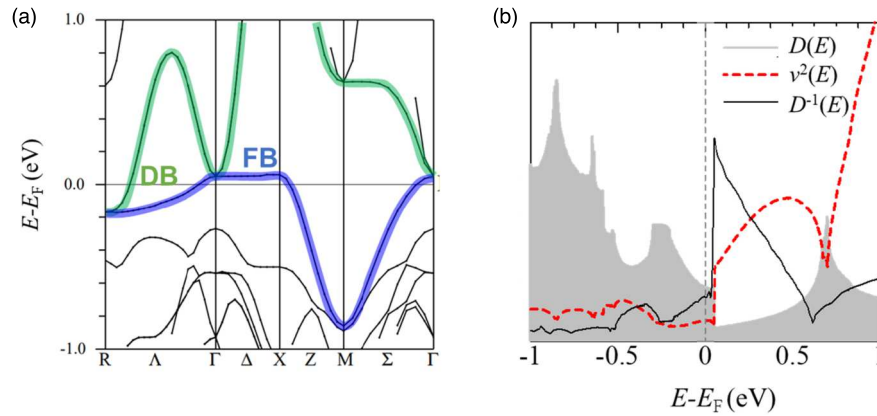


Fig. 6. (a) The band structure of Ni_3Ge features both a flat band (FB, blue line along the Γ -X direction) and a dispersive band (DB, green line) exists at the R point. (b) Density of states $D(E)$, squared group velocity $v^2(E)$, and the relaxation time model assuming $\tau(E) \propto D^{-1}(E)$. $D^{-1}(E)$ exhibits a sharp peak in the energy region just above the Fermi energy E_F .

transitions are allowed even without momentum transfer of phonons. This should be kept in mind when designing new metallic alloy systems for thermoelectrics. In alloys, where phonons are too short-lived and damped to dominate carrier scattering, Fermi surface geometries must be such that static disorder can yield transitions into states that are at most $|k| = k_F$ away in momentum space.

Which combination of group 10 and 11 elements realizes the largest S ? In order to predict the maximum performance and optimal alloy configuration, we systematically investigated the DOS of all possible combinations between Ni, Pd and Pt with Cu, Ag and Au (see Fig. 5) by means of alloy-averaged density functional theory (DFT) calculations in the Kohn–Korringa–Rostoker formalism and coherent potential approximation. Our theoretical predictions revealed that alloys comprising Ni and Au realize the highest S , which was experimentally confirmed. This way, we discovered record-high PFs among bulk materials at $T > 300$ K, with PF reaching up to $34 \text{ mW m}^{-1} \text{ K}^{-2}$ in $\text{Ni}_{0.1}\text{Au}_{0.9}$ at 560 K and ultrahigh average values up to

$30 \text{ mW m}^{-1} \text{ K}^{-2}$ in an extremely broad temperature range 300–1100 K.

4. Materials screening yields flat bands and high thermopower in Ni_3Ge and Ni_3In

In our systematic quest for cheap, abundant candidate materials, our first clean compound to demonstrate the effectiveness of this intrinsic energy filtering strategy was Ni_3Ge . Through large-scale computational materials screening using DFT, we identified the intermetallic compound Ni_3Ge with an L1_2 structure as an ideal candidate for realizing this paradigm. As shown in the band structure of Ni_3Ge in Fig. 6(a), a FB derived from the localized $3d$ orbitals of Ni atoms exists just 20 meV above E_F along the Γ -X direction, overlapping with a dispersive conduction band present at the R point. Figure 6(b) shows the $D(E)$, the squared group velocity $v^2(E)$, and a model of $\tau(E)$ assuming significant interband scattering, where $\tau(E) \propto D^{-1}(E)$. $D^{-1}(E)$ exhibits a steep peak just above E_F , which generates the asymmetric transport distribution function $\sigma(E)$.

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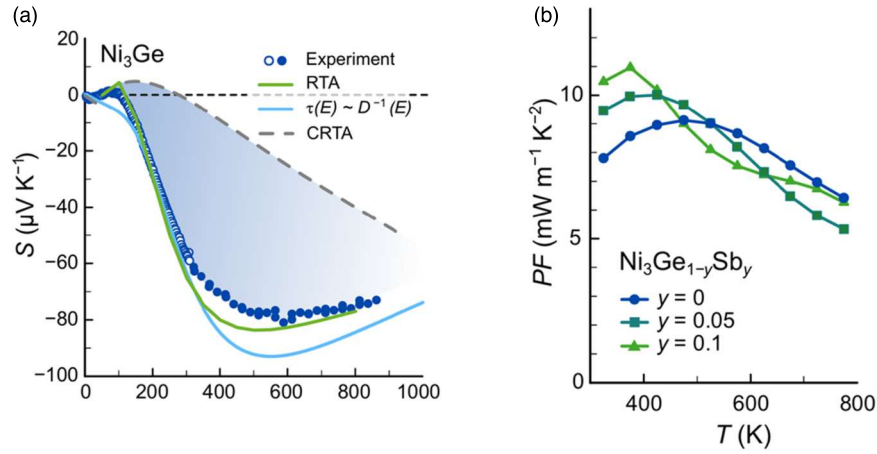


Fig. 7. (a) Experimental Seebeck coefficients (S) of Ni_3Ge compared with theoretical values calculated using different models including relaxation time approximation (RTA), $\tau(E) \propto D^{-1}(E)$ and constant relaxation time (CRTA). (b) Temperature dependence of the power factor (PF) for $\text{Ni}_3\text{Ge}_{1-y}\text{Sb}_y$. (This Figure is reproduced from Ref. 84 under the Creative Commons Attribution 4.0 International License.)

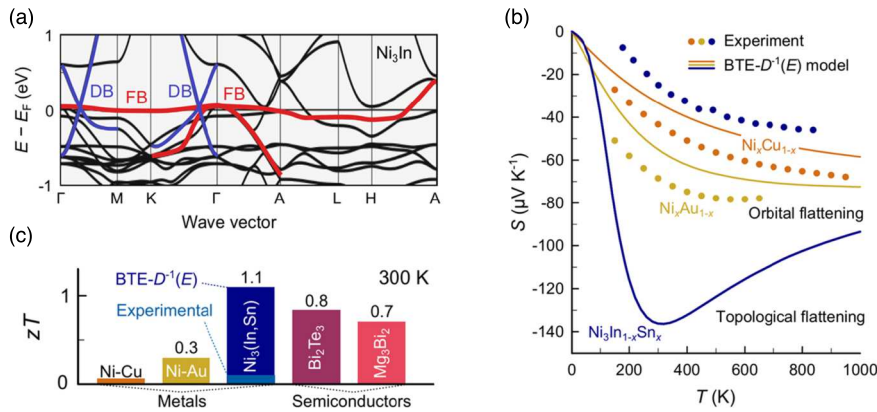


Fig. 8. (a) Band structure of Ni_3In . (b) Experimental Seebeck coefficients (S) of $\text{Ni}_3\text{In}_{1-x}\text{Sn}_x$ and other metallic thermoelectric materials ($\text{Ni}_x\text{Au}_{1-x}$, $\text{Ni}_x\text{Cu}_{1-x}$) compared with calculated values assuming $\tau(E) \propto D^{-1}(E)$. (c) Theoretical values of the dimensionless figure of merit zT . (This Figure is reproduced from Ref. 85 under the Creative Commons Attribution 4.0 International License.)

In Ref. 84 we synthesized stoichiometric and Sb-doped Ni_3Ge samples. As shown in Fig. 7, undoped Ni_3Ge exhibited a very large negative Seebeck coefficient of $S_{\text{max}} = -80 \mu\text{V K}^{-1}$ at 600 K, despite its metallic electrical resistivity of $\rho = 80 \mu\Omega \text{ cm}$. The Sb-doped $\text{Ni}_3\text{Ge}_{0.9}\text{Sb}_{0.1}$, in which E_F was optimized, achieved an exceptionally high PF $11 \text{ mW m}^{-1} \text{K}^{-2}$ in the temperature range from room temperature to 400 K. This value surpasses that of most high-performance thermoelectric materials. Furthermore, the S calculated using the semi-classical Boltzmann transport equation and a simple model assuming $\tau(E) \propto D^{-1}(E)$ reproduced the experimental S well. Indeed, S obtained by calculating the electron-phonon interaction using density functional perturbation theory within the relaxation time approximation (RTA) aligns well with both experiment and the $\tau(E) \propto D^{-1}(E)$ model. This strongly supports the conclusion that the proposed energy filtering mechanism is actually realized in clean bulk compounds through intrinsic phonon-mediated interband scattering. Note that in NiAu alloys and constantan^{83,90–92} interband transitions are predominantly mediated via impurity scattering, but in both cases the electron-hole asymmetry emerges from the phase space of interband transitions.

Aiming to apply the energy filtering strategy demonstrated in Ni_3Ge to a more ideal platform, we conducted a high-

throughput computational materials screening and found that the Kagome metal Ni_3In was suitable.⁸⁵ The “TFB” of Ni_3In is an extremely narrow feature in the DOS arising from destructive phase interference of electron hopping paths caused by the geometric frustration of d -orbitals orbitals. Figure 8 shows (a) the band structure of Ni_3In , (b) the S calculated assuming $\tau(E) \propto D^{-1}(E)$, and (c) the calculated dimensionless figure of merit zT alongside other metallic thermoelectric materials. Since the flat band of Ni_3In has an even narrower bandwidth than the d -orbital derived band of Ni_3Ge , a large S exceeding $-100 \mu\text{V K}^{-1}$ and high thermoelectric performance ($zT > 1$) were theoretically predicted.

However, the experimental results significantly fell short of these theoretical predictions. We synthesized $\text{Ni}_3\text{In}_{1-x}\text{Sn}_x$ by alloying Ni_3In with Ni_3Sn to tune the position of the TFB relative to E_F and measured the thermoelectric properties. The measured S for all samples was smaller than that of Ni_3Ge , with an absolute value of at most $50 \mu\text{V K}^{-1}$. Furthermore, neither the semi-classical transport model of $\tau(E) \propto D^{-1}(E)$, which was successful for Ni_3Ge , nor the RTA calculating electron-phonon interactions from first principles calculation (BTE-RTA) could explain the experimental results at all.

To unravel this significant discrepancy between theory and experiment, we introduced calculations using the Wigner transport formalism-based RTA (WTE-RTA), a more

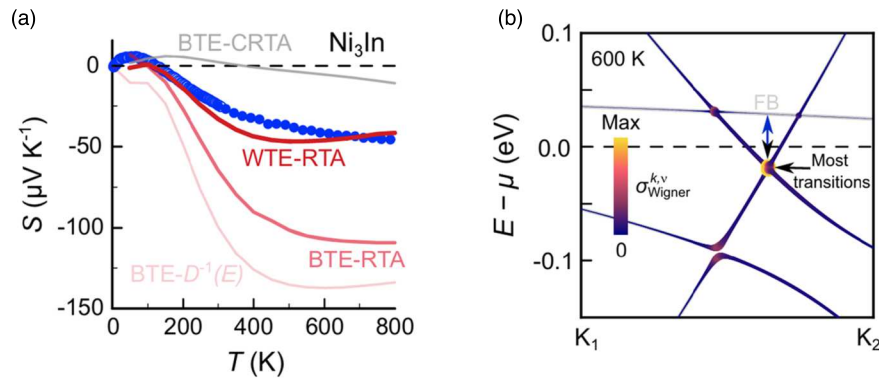


Fig. 9. (a) Experimental Seebeck coefficients (S) of Ni_3In compared with calculations using various relaxation time models based on Boltzmann transport theory (BTE), and quantum transport calculations based on Wigner transport theory (WTE). (b) Mapping of the Zener tunneling contribution derived from Wigner transport onto the band structure of Ni_3In . (This Figure is reproduced from Ref. 85 under the Creative Commons Attribution 4.0 International License.)

advanced theoretical framework that extends beyond semi-classical Boltzmann transport theory (Fig. 9). The results revealed the existence of a new quantum transport phenomenon previously unconsidered in thermoelectric transport: “Zener Tunneling.”

The electronic structure of Ni_3In contains Dirac cones with linear band dispersion in addition to the TFB. As shown in Fig. 9(b), calculations using the WTE identified that an extra conduction channel exists where electrons tunnel directly between these Dirac bands. This is the primary cause for the weakening of the electron–hole asymmetry created by energy filtering, thereby suppressing the S . This demonstrated that the performance of thermoelectric materials can be governed by pure quantum effects that cannot be captured by the conventional semi-classical picture. To suppress this troublesome Zener tunneling and realize a large S in Ni_3In , methods such as separating the Dirac cone and the TFB by applying strain to the material or through elemental substitution are conceivable. Moreover, the availability of Ni_3In single crystals provides the unique opportunity to investigate anisotropy effects of interband scattering—and possibly also direction-dependence of the Wigner contribution to transport. Having said that, low-frequency spectroscopic studies of charge transport, e.g. by optical reflectivity in the infrared and THz ranges, will be highly informative to elucidate the intriguing mechanisms at play.

5. High performance metallic thermoelectrics summary

The series of studies described above has brought two major advancements to the field of metallic thermoelectric materials. First, by proposing the clear design strategy of “Intrinsic Energy Filtering” and experimentally proving its effectiveness in Ni_3Ge , we demonstrated that metallic materials can indeed become high-performance thermoelectric materials. Second, in the process of expanding this strategy to TFB materials, we elucidated the importance of the quantum transport phenomenon “Zener Tunneling,” which cannot be explained by conventional semi-classical transport theory, thereby indicating the necessity of a new theoretical perspective in the design of metallic thermoelectric materials. Through these studies, we are further advancing the search for materials with strong energy dependence of relaxation times, and computationally we have identified several candidate materials that

exhibit large S values exceeding $100 \mu\text{V K}^{-1}$, which will be subject to future experimental endeavours.

6. Outlook

As described in this review, the emergence of metallic thermoelectric materials as viable thermoelectric materials is exciting for various important reasons.

First of all, there is the huge increase of exploratory phase space to search for new attractive materials, which was previously confined largely to semiconducting materials.

The other fundamentally interesting aspect is the novel and powerful thermoelectric enhancement mechanism demonstrated, namely, how utilizing different character energy bands (e.g. flat and sharp near the Fermi level) with different scattering time dependences, could result in intrinsic carrier filtering effects to enhance the Seebeck coefficient, without relying on construction of composite materials with suitably functioning interfaces which can be complex and sometimes challenging for reproducibility.

Furthermore, two attractive directions for applicability of metallic thermoelectric materials are also shown.

Firstly, for the Ni_3Ge -type materials, very large PFs of $\sim 10 \text{ mW m}^{-1} \text{ K}^{-2}$ and not too high thermal conductivity can yield competitive zT values, to try to further enhance for conventional thermoelectric applications. It further should be mentioned that for the metallic materials, the high thermoelectric performance tends to manifest at relatively low temperature including room temperature.

Secondly, for the materials of Ni–Au-type, the huge PFs $> 30 \text{ mW m}^{-1} \text{ K}^{-2}$ and high thermal conductivity make them interesting candidates for the active cooling application, which is becoming increasingly important as the heating problems related to semiconductors, data centers, etc., become critical. Further tuning of the PF and thermal conductivity should be possible, and can lead to attractive materials for this relatively new application.

We think that this review on the new frontiers shown in metallic thermoelectrics will inspire further intense research explorations and discovery of even higher performance materials to come. Altogether, the recent discovery of high-performance metallic thermoelectrics provides an invaluable stimulus for the vast fields of thermoelectricity and condensed-matter research in general. We expect exciting and unexpected results both from experimental and theoretical investigations.

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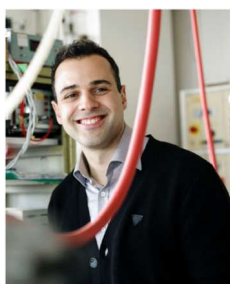
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