Perspective

Importance of Thermal Transport for the Design of Solid-State Battery Materials

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Battery technologies have evolved rapidly over the past decade, including the advent of solid-state batteries. In this time, it has become apparent that thermal management is paramount for device operation and lifetime. However, the fundamental importance of the thermal properties of materials, such as thermal conductivity, in engineering design and mitigating the risk of catastrophic failure is yet to be fully understood. This Perspective aims to provide motivation for the fields of thermal transport and ionic transport to join forces to understand heat transport for better battery design, especially in light of solid-state batteries. From the basic characterization of thermal conductivity in bulk materials to considering the full complexity of battery composites during electrochemical cycling, there are many potential directions for fundamental and applied investigations. We anticipate that studying heat transport in battery materials has the added benefit of extending the design space to other functional devices.

The difficulty in controlling heat transport in solid-state energy devices, including microelectronics, batteries, and thermoelectrics, is often a limiting factor in improving device performance. Especially in batteries, not only can excessive heat cause degradation that leads to a loss of charge capacity over time, but thermal runaway can occur when the battery overheats to catastrophic failure. Thus, understanding heat evolution and thermal transport in batteries is an important step to improve lifetime and safety. It is from this perspective that we provide the motivation for the importance of bringing together the fields of thermal transport and battery research, particularly to study solid-state batteries, which epitomize the overall complexity of battery systems and require a state-of-the-art understanding of thermal transport mechanisms. Here, we identify the basic and applied scientific directions that may prove fruitful for the next generation of battery thermal management.

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I. INTRODUCTION

The transport of charged particles (electrons and ions) through a material results in the generation of heat due to the various microscopic interactions broadly categorized as resistance. Known as Joule heating, the rate at which electrical energy is converted into thermal energy is proportional to both current and voltage, meaning that "high-rate" and "high-voltage" batteries inherently generate more heat [1]. Additionally, this thermal energy generated inside the battery tends to accumulate over time if the rate of heat removal is slower than the rate of production. The intrinsic ability of battery materials to dissipate heat depends on the magnitude of their effective thermal conductivity, where higher thermal conductivity is desirable for faster heat removal. It is also possible to actively cool the battery pack to draw heat out faster; however, this additional power consumption increases with the thermal resistance of the battery. Altogether, the thermal management of a battery is necessary to control the operational temperature [1–4].

Thermal management in lithium-ion batteries, especially as a multicomponent and composite system, is complex and challenging [5,6]. This is irrespective of whether a liquid or solid electrolyte is used, although the rising development of solid-state batteries is expected to bring unique thermal design challenges, especially since they are proposed to achieve high power densities [7]. The great influence of relatively subtle temperature changes on ion

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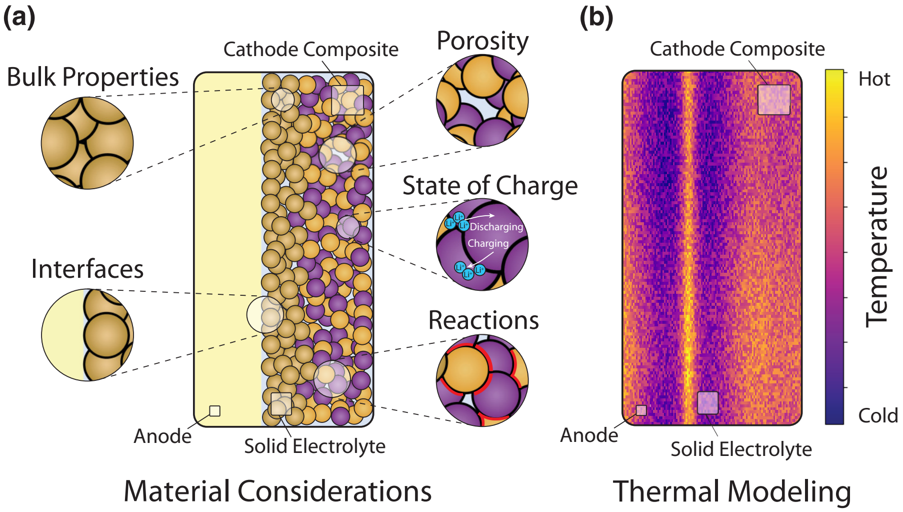

FIG. 1. (a) Solid-state battery configuration with a lithium metal anode (pale yellow); a solid ionic conducting separator (beige circles); and a composite cathode (yellow and orange circles) consisting of ionic conductors, cathode active materials, and potential additives. Variety of interfaces are highlighted at which heat generation may be expected. (b) Schematic of a potential temperature distribution snapshot in the solid-state battery [shown in panel (a)] during operation. Goal of multiscale material modeling is to enable reliable calculation of such heat maps to inform device design.

transport and interfacial stability makes thermal management an indispensable part of battery design. From an engineering perspective, battery packs on the kilowatt-hour scale in electric vehicles may lead to different thermal problems than batteries in mobile devices with energy storage of a few watt hours [8,9]. In any case, accurate thermal property characterization of the constituent materials is an essential first step toward developing design principles at the battery cell level. Quantifying thermal conductivity relevant to battery design is not a small task, as it requires considerations of the numerous different interfaces and interphases [7,10], porosity [11], defect concentrations, and ongoing chemical and electrochemical reactions [12,13], as exemplarily shown for a solid-state battery configuration in Fig. 1(a). In addition, on a fundamental level, understanding how atomic vibrations carry heat in battery materials may ultimately provide insights into the vibrational origins of ion transport [14,15].

Characterizing the thermal properties in these materials is important for modeling temperature distributions in battery devices [Fig. 1(b)], towards the goal of optimizing operational conditions [16,17]. For instance, low temperatures are known to impair the functionality of batteries using liquid electrolytes that can freeze. Higher temperatures increase the kinetics of interfacial reactions, as well as the risk of catastrophic device failure due to thermal runaway in liquid electrolyte cells [18,19]. Solid-state batteries also exhibit temperature-dependent chemical and electrochemical degradation kinetics, leading to additional interphases that affect performance [12,13,20,21], and, just recently, thermal runaway of sulfide-based solid-state batteries was shown at elevated temperatures [22]. Nevertheless, elevated temperatures might be beneficial during charging of a lithium metal anode in solid-state batteries, if the improvements in lithium plating outweigh other detrimental effects [23–25]. For such engineering optimizations, it is important to understand the temporospatial heat distribution during cycling, especially since interfacial resistances can result in significant Joule heating [1].

In this Perspective, we propose the necessity for multidisciplinary research directions to advance both fundamental and applied ionics and solid-state battery development from a thermal transport point of view. By listing potential research directions, we hope to provide guidance to a new field that explores connections between thermal and ionic transport, as well as the magnitude and mechanisms of thermal transport in ionic conductors and solid-state batteries. Given the overwhelming complexity of battery materials and battery systems, we expect this field to push the boundaries of our understanding of atomic vibrations, heat transport, interfaces, and interphases, as well as functional material design. So far, most of these research directions have not yet been thoroughly investigated, and some of their impacts may be speculative. Herein, we summarize some state-of-the-art investigations and identify seemingly logical next steps for (i) the systematic investigation of thermal transport in solid-state battery components, and (ii) building up a vibrational perspective of ion transport.

II. CONTEMPORARY UNDERSTANDING OF THERMAL TRANSPORT

Thermal transport in electronically insulating solids is facilitated by the transfer of vibrational energy between atoms. These vibrations are quantized as phonons, and the theory of phonon thermal transport has been developed for over a century [26–28]. It is now proven that phonons can have different transport character, depending on the complexity of the atomic structure and the bonding interactions between atoms (e.g., anharmonicity) [29–31]. In structurally simple solids, like most crystalline materials with only a few atoms per unit cell, phonons propagate through the material in a gaslike manner, with a velocity, carrying heat an average distance (called the mean free path) before a scattering event occurs (e.g., by point defects, grain boundaries, or other phonons) [32]. These vibrations are associated with a large apparent correlation of atomic motion, and mean free paths range from tens of nanometers to tens of microns in single-crystal silicon [33]. In amorphous solids and structurally complex crystals, the large number of quasi-degenerate vibrational modes, especially overlapping optical modes, gives rise to an atomic scale hoppinglike transport of vibrational energy that conducts in parallel with gaslike phonon transport [34]. These vibrational modes can be called *diffusons* (to give a quasiparticle-like name to phonons with this distinct transport character) and appear to have less correlation in atomic displacements, but are still spatially extended modes capable of thermal transport [35]. The so-called phonon "participation ratio" and "mode shape" can be used to categorize phonon vibrations using relative atomic displacement vectors [36]. The character of phonon modes can also be found by analyzing their contribution to the total phonon thermal conductivity, κ_{ph}, which can be written as the sum of the phonon gaslike (κ_{PGM}) and diffuson (κ_{diff}) channels of thermal transport: $\kappa_{\text{ph}} = \kappa_{\text{PGM}} + \kappa_{\text{diff}}$ [37]. The existence of two-channel transport is rigorously derived from considerations of the phonon heat flux operator, where gaslike transport is represented by diagonal matrix elements and diffusons by off-diagonal elements [30,31,37].

Operatively, the phonon-gas channel of thermal transport always contributes in the $T \rightarrow 0$ K limit, where only long-wavelength acoustic vibrations are thermally excited. The comparatively large group velocities and low degeneracy of the acoustic modes can be used to typify features of the phonon band structure that lead to phonon gas transport (Fig. 2) [38]. Conversely, diffuson transport is expected to be more prominent at high temperatures, especially when the thermal conductivity of solids tends to a constant value [14,29,39]. The phonon band structure indicator for diffusonlike vibrational character is a high degree of energetic overlap between phonon modes (Fig. 2), often with low group velocities (meaning flat energy dispersion) and relatively high anharmonicity. Although both kinds of phonon transport can be present in every material, their relative contributions depend on temperature, complexity of the crystal structure, and bonding.

Electronic carriers, when present, can also contribute significantly to thermal transport through the Wiedemann-Franz relationship, in which a high electronic conductivity leads to a high thermal conductivity, typically found for metals [40,41]. Although ions certainly carry heat as they move through the host lattice, it does not seem, at this point, that they contribute appreciably to the total thermal conductivity [14,42]. Nevertheless, characterizing and understanding thermal transport in solid-state battery materials certainly necessitates a combined perspective of phonon-gas and diffuson transport, as well as electronic contributions, especially when considering so-called "active" materials that participate in the electrochemical reactions.

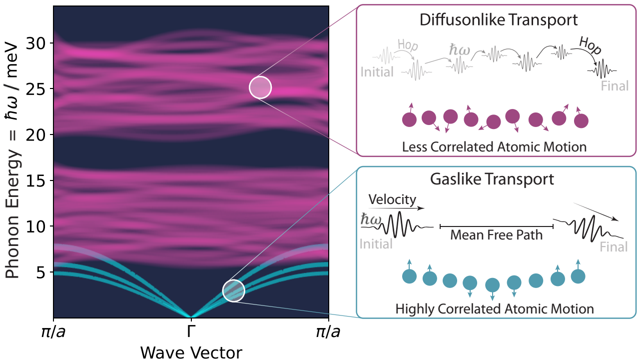

FIG. 2. Phonon character and thermal transport behavior. Schematic phonon dispersion in which acoustic (blue) vibrational modes are highly dispersive and distinct, resulting in gaslike phonon transport of vibrational energy $\hbar\omega$, whereas optical (pink) vibrational modes have little dispersion and a high degree of (quasi-)degeneracy, which gives rise to diffusonlike hopping transport of vibrational energy $\hbar\omega$ as an atomic scale random walk.

III. THERMAL CONDUCTIVITY OF BATTERY MATERIALS

All batteries are multiphase composites with, inevitably, a lot of interfaces. Solid-state batteries, in particular, have distinctive interfaces between the various cell components (solid electrolyte, active material, conductive additive, etc.), as well as with unintended decomposition products (interphases) and pores [12,13,20,21]. Because small particle sizes lead to better composite material packing (facilitating faster charge-discharge rates) and increased contact surface areas, especially between solid electrolyte and active materials, this also contributes to a higher density of interfaces and evolving interphases [11,43]. To develop models for thermal management, a systematic approach to characterizing and understanding heat transport is hence needed [3]. We propose that, first, thermal transport in the "phase pure" individual materials should be investigated, such as the thermal conductivity of single crystals or dense polycrystalline samples. From here, the effects of particle size, porosity, and point defects on thermal conductivity can be systematically studied before advancing to the more challenging work of bringing the individual components together, one by one, to assess thermal conduction in complex composites and across interfaces. Such an approach is expected to provide the requisite information for a parameterized model of effective thermal conductivity across a battery cell.

Here, we review the relatively small body of work on characterizing thermal conductivity in battery materials and discuss some preliminary trends and observations. Given extensive investigations into thermal transport in semiconductor materials for microelectronic and thermoelectric applications, it should be noted that solid-state batteries provide an excellent opportunity to apply and develop concepts in thermal engineering for highly relevant emerging technology.

A. Thermal transport in solid electrolytes

The flammability, toxicity, and low energy density of organic liquid electrolytes frequently used in state-of-the-art lithium-ion batteries can be mitigated by using solid-state electrolytes [44,45]. Although their thermal stability is generally superior, it is not yet known if solid electrolytes have thermal conductivities that can provide better thermal management in solid-state batteries, relative to liquid electrolyte cells.

A recent study by Cahill and co-workers [46] posited that "good" ion conductors were "bad" thermal conductors, reporting thermal conductivities for several Li^+ and Na^+ solid electrolytes below 1 $\mathrm{W\,m^{-1}\,K^{-1}}$ at room temperature [see Fig. 3(a)]. Such low thermal conductivities are in the typical range for glasses and complex thermoelectric materials and are not much higher than the thermal conductivity of typical liquid electrolytes ($\sim$0.2 $\mathrm{W\,m^{-1}\,K^{-1}}$ at room temperature) [46,52]. However, a study directly comparing ionic and thermal conductivity in fast Ag^+ ion conductors shows no apparent correlation between the magnitude of ionic and thermal conductivity [14]. Nevertheless, the atomic structure (e.g., complex and disordered) and bonding (e.g., soft and anharmonic) conditions associated with fast ion transport [15,53–55] are also associated with diffuson thermal transport. In neither case, however, are rigorous structure-property metrics devised, and it is not known to what extent ionic and thermal transport can be interdependent. Experimentally, the temperature dependence of the thermal conductivity of the studied solid electrolytes is in qualitative agreement with what is theoretically expected for diffuson-dominated transport at high temperature [Fig. 3(a)] [39].

Other recent work focused on characterizing phonons in superionic conductors spectroscopically, showing strong anharmonicities and the breakdown of well-defined phonon modes at the onset of superionic conduction [15,54,55]. However, no drastic change in thermal transport was found to coincide with this transition [54]. Thus, it appears that fast ion transport can affect the appearance of phonons, as inferred from inelastic neutron or x-ray scattering experiments, but not necessarily the appearance of phonons from a thermal transport perspective. This apparent inconsistency further motivates the fundamental study of vibrational character and phonon-ion relationships.

So far, only initial work has investigated the thermal conductivity of solid electrolytes [44,46]. One reason might be the experimental difficulty due to the strong moisture sensitivity of many Li^+ and Na^+ solid electrolyte candidates or that the two fields of thermal transport and ionic transport have yet to link research interest. Computationally, the use of lattice dynamics or molecular dynamics to investigate thermal conductivity in highly defective structures can also be challenging [56,57]. Another experimental hurdle is the inability to fabricate fully dense samples, leading to effectively lower thermal conductivity values [58]. By systematically investigating the effective thermal conductivity as a function of relative density, in conjunction with effective medium theory analysis, an estimate for the thermal conductivity of the fully dense material can be obtained [59]. We suggest that this methodology be standardized so that results between studies can be compared, especially since relative density and pore distribution are likely to be key parameters for device-scale thermal models. We also recognize that measurement errors tend to increase as thermal conductivity decreases, meaning that error estimation should not be forgotten, and statistical methods may need to be employed when comparing low thermal conductivity materials [60].

Overall, there is a possibility that solid electrolytes can provide faster heat dissipation and better battery thermal management than liquid electrolyte cells. However, this depends largely on the relative magnitude of Joule heating that occurs (predominantly at interfaces) compared with the thermal conductivity of the material and device design. Initial results suggest that thermal conductivity in solid electrolytes is at least about 2–3 times higher than liquid electrolytes at room temperature, but it is not clear if this is enough of an improvement to mitigate the need for active cooling [46]. Because the solid electrolyte separator layer is designed to be as thin as possible, it may also be that the thermal properties of the solid electrolyte material are more important in the composite cathode, where heat generation can occur due to both high interfacial density and ongoing electrochemical reactions. Nevertheless, the fundamental study of thermal transport in solid electrolyte materials advances our understanding of phonon-ion relations and helps to shape future solid-state battery designs.

B. Thermal transport in electrode materials

Batteries are dynamic systems that operate because an electrochemical potential gradient is sustained internally between the anode and cathode. During operation, the

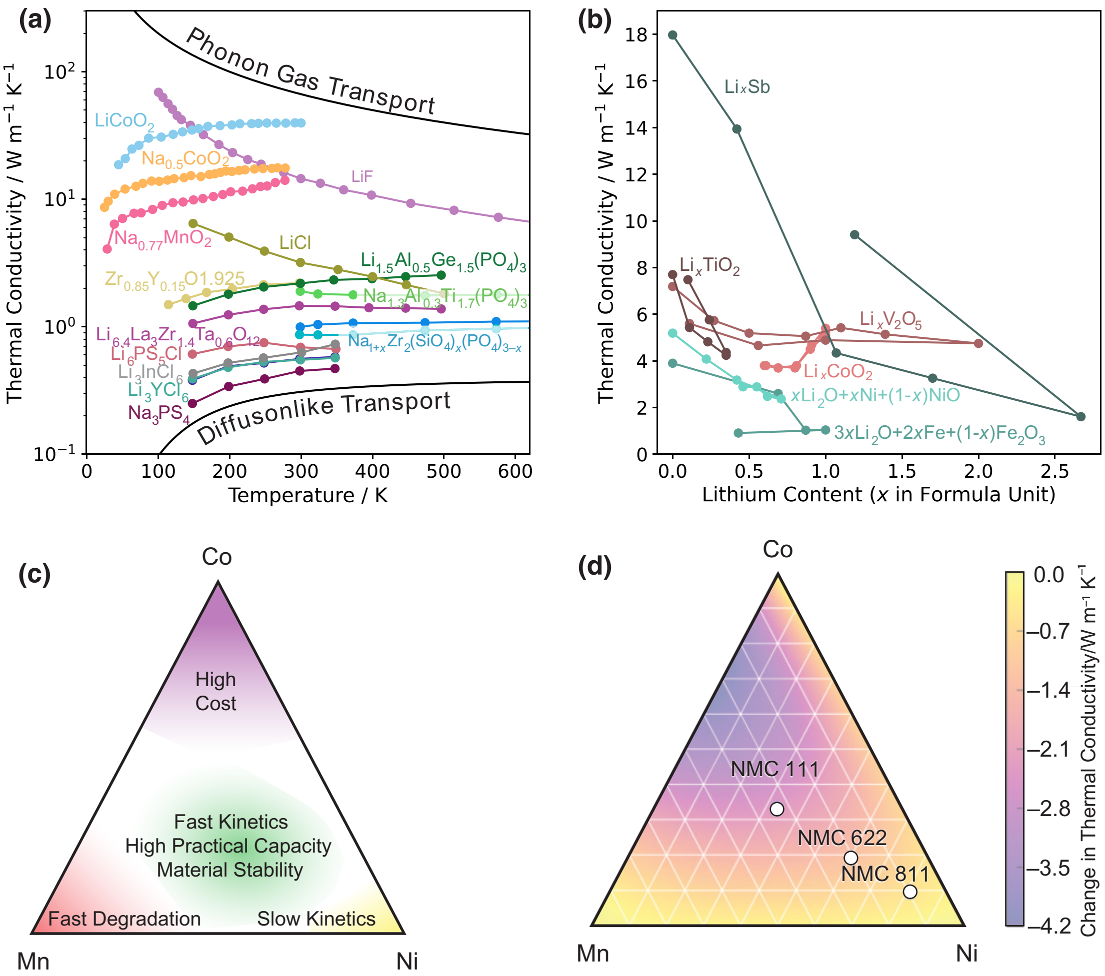

FIG. 3. Characterization of thermal transport. (a) Temperature dependence of thermal conductivity for a variety of active materials [46–48] and solid electrolytes [46,49] reveals that diffusonlike vibrational character may be prevalent. Compared to the thermal conductivity of liquid electrolytes (~0.2 W m^{-1} K^{-1} at 300 K) [46], solid electrolytes may not offer much of an improvement. (b) State-of-charge dependence of thermal conductivity is due to the changing content of mobile ions in the active materials [50,51], structural changes, and any additional microstructural effects, which are a potential cause of the hysteretic behavior. (c) Chemical design space for battery materials, illustrated here considering LiNi$_x$Mn$_y$Co$_z$O$_2$ cathode materials, is currently being explored for electrochemical performance and cost optimization; however, (d) the magnitude of thermal conductivity is also expected to depend on material composition, as demonstrated here in LiNi$_x$Mn$_y$Co$_z$O$_2$ cathode materials (figure after Ref. [48]) with a constant lithium content. Thus, future battery development needs to include thermal management considerations when thermal properties are characterized.

active material at the electrodes can change its thermodynamic state, for instance, lithiation and delithiation in the case of lithium-ion battery electrodes [61]. Simply, this means that the quantity of lithium atoms is changing continuously in the electrode materials during battery cycling, as quantified by the state of charge. Different compositions at different states of charge can have markedly different thermal conductivities [Fig. 3(b)] [50,51,62]. This is expected because not only is the composition of the material changing during the electrochemical reaction, but so are the oxidation state and crystal structure (e.g., local structure and bonding environment) [63,64]. Often, the electrode active materials undergo one or several phase transitions [65–67]. It would be ideal to measure the thermal conductivity of each of these phases independently and in bulk, as suggested for solid electrolytes, to build a foundation for more complex thermal transport studies.

Preliminary investigations show that a hysteresis may be prevalent when measuring thermal conductivity as a function of state of charge [Fig. 3(b)] [50,51]. This may be the result of irreversible reactions or significant changes in microstructure or interphases that result from electrochemical cycling. It should be noted that electronic conductivity may also contribute significantly to thermal conductivity, with its own state-of-charge dependence [68,69]. The interplay of a multiplicity of different microstructures in electrode materials, including anisotropy, and, perhaps to a greater extent, the coatings on active materials that aim to prevent strong decomposition reactions add further levels of complexity [10,70]. Thus, characterizing thermal transport in electrode active materials may be more tedious than in solid electrolytes.

The chemical composition of materials, as well as microstructural defects, are often used to tune thermal conductivity in energy materials like thermoelectrics [71,72]. Thus, it is interesting to speculate that it may be possible to use the stoichiometry of transition metal oxides (cathode active materials) to simultaneously tune the electrochemical and design requirements of the battery [Fig. 3(c)], including considerations of the thermal conductivity [Fig. 3(d)]. For example, in the $\mathrm{Li(Ni, Co, Mn)O_2}$ system, Ni-rich and Mn-rich compounds are expected to have the highest thermal conductivity, which further motivates the utilization of cobalt-free compositions [48].

Calculating, measuring, and understanding thermal transport as a function of the state of charge is a rather new research direction and only limited studies are available so far [51]. Developments in experimental and theoretical methodologies are clearly needed to investigate the complexities of electrode active materials. However, this is an exciting research topic, as it not only leads to a better database for thermal transport modeling, but also because it is possible to truly understand how small changes in a material's composition and chemical state can affect thermal transport in functional materials.

C. Thermal transport in composites

Just as the thermal transport of anode and cathode active materials is state-of-charge dependent, so is the heat conduction of solid-state battery cells, which are effectively composites of mixed solid particles [see Fig. 1(a)]. During battery operation, heat generation due to internal resistance (Joule heating), as well as from electrochemical reactions (such as intercalation or phase changes) and chemical reactions (e.g., thermodynamic degradation at interfaces), needs to be dissipated [12,20,21,73]. Thus, understanding thermal transport pathways in complex battery composites is the ultimate engineering goal.

Characterizing thermal transport in battery composites is no small feat. Already, measuring temperature distributions in battery cells is a highly involved process [16,17]. Both *ex situ* (as a function of state of charge) and *operando* (during electrochemical cycling) measurements of thermal properties have significant practical challenges. Even more so, solid-state batteries often operate under an external pressure, exacerbating the challenge of measuring *operando* thermal conductivities. Furthermore, phase transformations (including phase transitions and electrochemical side reactions) during cycling take place, which can have a significant influence on the measurement of thermal diffusivity and heat capacity [74]. Altogether, great care must be taken to estimate thermal conductivity in the presence of interphase formations and the effect of local heating during electrochemical reactions.

The effective thermal conductivity of composites is understood by considering the bulk thermal conductivity of the constituent materials, weighted by their respective volume fractions, and accounting for the microstructure (particle shape). This approach is broadly categorized as effective medium theory, and there are various effective medium models [75–77]. One prevalent model, by Bruggeman, is already widely used to characterize ionic conductivity in solid-state battery composites [58,75]. Interestingly, recent work [59] suggested that ion transport and thermal transport might not follow the same Bruggeman scaling relation, which was unexpected from the current understanding of effective medium theories [58]. Given that effective transport coefficients for both ionic and thermal transport are necessary for thermoelectrochemical modeling of batteries, there is significant motivation to understand the limitations of effective medium theories. This can be accomplished through the systematic investigation of transport in composites composed of pores and functional materials with a variety of particle shapes and size distributions.

As an alternative to effective medium theory, equivalent circuit models for thermal transport were developed to explain microstructural effects in thermoelectric materials [72,78]. Here, fundamental investigations of interfacial resistances are necessary [72,79–83]. Solid electrolyte degradation and interphase formation and growth alter thermal conductivity across the interface or interphase of the solid electrolyte and electrode material. Importantly, interfacial resistance and constriction resistances are not explicitly considered in effective medium theory [84]. Using more-detailed physical models can provide immense insights into the role of interfaces in thermal and ionic transport, although it is unclear if such models are practical to implement at the scale of a full battery cell. Thus, we propose the development of multiscale material models to incorporate thermal transport behavior from the atomic level to the microstructural level and up to the system level [85–87].

IV. VIBRATIONAL PERSPECTIVE OF ION TRANSPORT

Having established the significant motivation for thermal transport characterization for battery engineering purposes, it is also important to point out the coinciding opportunity to study ion transport from a vibrational perspective. To date, there is no "phonon theory" of ion transport as there is for heat transport. While the so-called jump or diffusion pathway for ions is relatively easy to determine [Fig. 4(a)], the energetic mechanisms that facilitate transport are not. Thus, the characterization and understanding

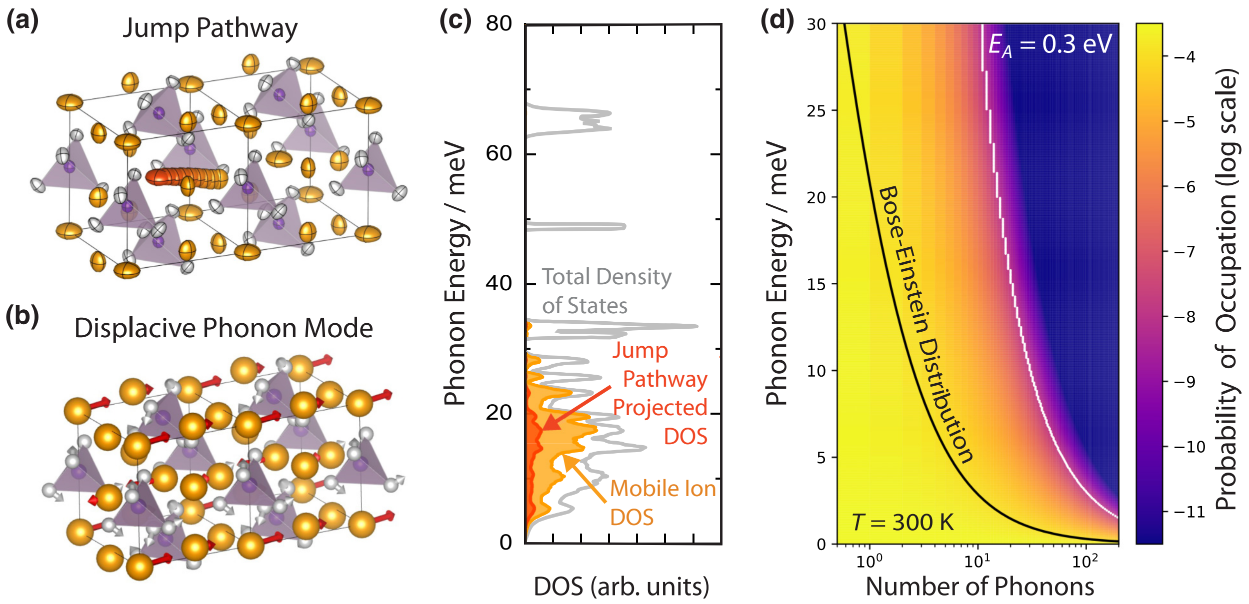

FIG. 4. Vibrational understanding of ionic transport. (a) Schematic of the lowest energy pathway for a mobile ion to move between crystallographic sites. (b) Single low-frequency phonon mode, where phonon eigenvectors indicate a displacive motion of mobile ions along the jump pathway shown in panel (a). (c) By analyzing the phonon eigenvectors for all of the mobile ion vibrational modes, it is possible to see the spectral density of states of vibrations that contribute to the net motion of the mobile ion along the jump pathway. (d) Bose-Einstein distribution gives the average number of thermal phonons expected to be populated at a given temperature and vibrational mode energy (frequency), where lower frequency modes have higher average phonon occupations; however, the instantaneous occupation of phonons in a vibrational mode is a fluctuating quantity and the probability of a given number of phonons follows a Boltzmann distribution (see the color map). Also shown is the number of phonons that would be required in a given phonon mode to achieve a thermal energy of 0.3 eV, which is a typical activation barrier energy for ion transport in fast ion conductors.

of atomic vibrations may be equally important to grasp the fundamental physics of ion transport.

Strong discussions exist about vibrational contributions to ion transport, such as the concerted motions of structural motifs facilitating better ionic transport [88–91]. There is some agreement that phonon entropy limits our ability to tune ionic conductivity, but the methodology to calculate this entropy is not settled [14,92,93]. Even relatively simple questions like which attempt frequency should be used in the transition state theory of ion transport, have not really been answered. In this particular case, some believe that one (or a select few) vibrational mode is primarily responsible for ion transport [15,55,94], while others consider the possibility that many vibrational modes contribute to transport [14,54]. The former perspective seems to be based on the presence of low-frequency displacive phonon modes [Fig. 4(b)], analogous to those observed in displacive phase transitions [55,95]. The latter perspective recognizes that many phonon modes contribute to the net ion displacement in the jump direction between adjacent sites [Fig. 4(c)] [15,55]. In any case, it is possible to consider how many phonons would be needed for a specific vibrational mode to have enough energy to overcome typical ion transition barriers [Fig. 4(d)]. Relative to typical phonon occupation at room temperature [see the Bose-Einstein distribution in Fig. 4(d)], it is clear that a large phonon fluctuation would be needed for any vibrational mode to be responsible for an ion jump leading to net displacement and ionic transport [the white line in Fig. 4(d) shows how many phonons are needed to overcome an ion transition barrier of 0.3 eV]. Determining which vibrational modes are important to ion transport and how they are occupied is hypothesized to be the key to unlocking new design strategies for ionic conductors [92,94].

Overall, it is unclear if ion and thermal transport affect each other, if they are separate phenomena, or if phonons are the underlying cause for ionic conduction. Nevertheless, the experimental, analytical, and computational methods developed for understanding thermal transport provide a foundation for spectral investigations of atomic vibrations pertaining to ion transport. For example, methods of investigating changes in atomic dynamics due to changes in temperature, pressure, or through phase transitions, may also be applied to study the fundamentals of ion transport in battery materials. While *ab initio* molecular dynamics simulations prove to be useful for validating experimental work, it is tedious to extract spectral information relating vibrations to ion jumps. Lattice dynamics provides spectral information, but does not yet model ion transport. Future work seems to be needed to truly understand these phenomena and to determine if heat transport or targeted phonon excitation can be used to push ions through a solid [94].

V. SUMMARY AND OUTLOOK

Modeling thermal load and distribution within batteries themselves, and packs on a larger scale, enables engineers to identify and prevent instances of thermal runaway and thermal deterioration of battery materials. Charging times and other factors for device performance and lifetime can depend on the temperature of the battery. In general, efficient charging procedures may be developed from a thermal perspective for both liquid electrolyte and solid electrolyte battery cells. This is especially needed when considering the current push from liquid-electrolyte-based to solid-state batteries.

For instance, considering the low thermal conductivities measured for many solid electrolytes, significant local heating effects may also be appreciable in solid-state batteries. While these can speed up decomposition reactions, kinetically hindered reactions, such as metal plating, may benefit from local heating effects. Ultimately, and speculatively, it may be possible to use these heating effects by finding the sweet spot between beneficial and detrimental influences, for instance, by mitigating the need to heat a battery in colder climates during operation. Along this line of thinking, it may be beneficial for the thermal and ionic transport communities to identify a material-level figure of merit (perhaps a dimensionless ratio consisting of the ionic and thermal conductivities or effective conductivities) that aids in the identification of promising materials, microstructures, and composites for battery cell development. Tying thermal and ionic transport together, in the context of capacity retention, charging-discharging rates, and cell power, is a promising research direction, given the complex optimization that is centered around thermal, ionic, and electronic transport in batteries.

Optimization of battery performance requires accurate system-level modeling based on input parameters such as the effective thermal and ionic properties of materials. These effective properties may be estimated with sufficient accuracy using effective medium approximations. However, identifying the correct effective medium theory depends on a microscopic understanding of transport processes. Thus, there is tremendous importance for further thermal characterization of active materials and solid ion conductors from the fundamental level towards a multiscale thermal transport understanding that can be used in conjunction with cell-level ionic transport modeling [56,96–100]. By studying battery materials, our understanding of thermal transport across interfaces, in composites, and during phase transformations, will be advanced. The lessons learned here may be applied to other functional devices, such as thermal rectifiers and switches, thermoelectrics, and fuel cells. Finally, investigating the role of vibrational modes in ion transport is fundamental to building an understanding of ion transport physics. Future technologies based on ion transport may only be inspired by this understanding.

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