



# Estimation of electron-phonon coupling via moving least squares averaging: a method for fast-screening potential thermoelectric materials



S. Bang<sup>a</sup>, J. Kim<sup>a</sup>, D. Wee<sup>a,\*</sup>, G. Samsonidze<sup>b</sup>, B. Kozinsky<sup>b,c</sup>

<sup>a</sup> Department of Environmental Science and Engineering, Ewha Womans University, Seoul, 03760, Republic of Korea

<sup>b</sup> Research and Technology Center, Robert Bosch LLC, Cambridge, MA 02139, USA

<sup>c</sup> Harvard John A. Paulson School of Engineering and Applied Sciences, Harvard University, Cambridge, MA 02138, USA

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## ABSTRACT

In this communication, we present a method of predicting the Seebeck coefficient and electrical conductivity of inorganic semiconductors by estimating the electron-phonon (el-ph) coupling from the first principles for fast-screening potential thermoelectric (TE) materials. The method we propose, i.e. the electron-phonon averaged via moving least squares (EPA-MLS) method, combines the EPA method and the MLS averaging strategy. To demonstrate the performance of the EPA-MLS method, the Seebeck coefficient and electrical conductivity of a half-Heusler compound, i.e. HfCoSb, were computed with the EPA-MLS method and compared with the results from the EPA method and comparable experimental data sets. The results show that the EPA-MLS method exhibits several advantages over the original EPA method. The smoother interpolation reduces the risk of spurious numerical behaviors. The EPA-MLS method also requires less human intervention for tuning numerical parameters, since the calculation result of the EPA-MLS method exhibits robustness against the change of associated numerical parameters. The method may even reduce the overall computational cost by allowing the employment of a coarser resolution. All these advantages make the EPA-MLS method a suitable tool for fast-screening potential TE materials. One more example of an archetypical Skutterudite, i.e. CoSb<sub>3</sub>, is also provided for showing that the method can be used for TE materials with more complex structures.

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## 1. Introduction

Thermoelectric (TE) materials provide an attractive alternative option for electricity generation from heat. Without any moving parts, thermoelectric generators (TEGs) made with TE materials can be useful as compact waste heat recovery systems in vehicles or for electricity generation in cases where other technologies are inappropriate [1,2]. TEGs are also considered as a part of hybrid generation systems when combined with solar cells [3]. The materials can also be used for creating TE coolers, which can work as a cooling system for electric devices and solar cells [4,5]. Typically, the efficiency of TE energy conversion is expressed in terms of the temperature of each heat reservoir and the TE figure of merit  $ZT = S^2\sigma T/k$ , where  $S$  is the Seebeck coefficient,  $\sigma$  is the electrical conductivity,

$k$  is the thermal conductivity, and  $T$  is the temperature [6]. As  $ZT$  is one of the most important control parameters for the efficiency of TE energy conversion, significant efforts have been exerted on creating materials with high  $ZT$  values. Approaches to improve the performance of materials include construction of superlattices [7], nanowires [8], nanostructured grains [9], and hierarchical spatial structures [10]. Many different types of materials, including filled Skutterudites [11,12], advanced chalcogenides [13–18], and Heusler alloys [19,20], have also been tested.

Computational design of functional materials using high-throughput fast-screening methods has attracted a large amount of attention [21]. The field of research on TE materials is not an exception, and there have been a number of reports with various computational fast-screening methods applied for the discovery of novel TE materials [22–26]. Such efforts typically require computational capability of estimating the value of  $ZT$  for various different materials quickly. Several approaches were reported to compute the lattice thermal conductivity in TE materials from the first

\* Corresponding author.

E-mail address: [dhwee@ewha.ac.kr](mailto:dhwee@ewha.ac.kr) (D. Wee).

principles [27–29], while the estimation of the electronic transport properties including  $S$  and  $\sigma$  still remains a difficult problem. For electronic transport properties, computational results limited only to simple model systems are available in most cases [30,31].

Electronic transport coefficients in semiconductors, including inorganic TE materials, can be obtained by solving the semiclassical Boltzmann transport equation within the relaxation time approximation [32]. Thus, the simplest approach that can be used for the estimation of the electronic transport properties is the constant relaxation time (CRT) approximation, in which one arbitrarily assumes one single value for the relaxation time  $\tau$  [33]. However, such an approach introduces an arbitrary constant, i.e. the relaxation time, and does not possess any predictive capacity, which makes the approach unsatisfactory for screening TE materials from the first principles. It is, therefore, necessary to find a more predictive method for the relaxation time.

Matthiessen's rule states that the total scattering rate  $\tau^{-1}$  of electrons is the sum of the rates associated with intrinsic (electron-electron, electron-phonon [el-ph]) and extrinsic (impurities, grain boundaries, alloy disorder) scattering mechanisms. To screen potentially promising candidates for TE applications, one must first identify the intrinsic properties of the material, since the extrinsic properties are being tuned during the actual process of synthesis. In automotive TE power generation, the relevant temperature is around 400 °C at the hot side of the device, at which el-ph interaction becomes the dominant scattering mechanism [23,31]. The first-principles estimation of the el-ph interaction has been pursued by several different approaches with various levels of computational complication. At one end of the spectrum, there is the deformation potential (DP) approximation, which is one of the

different band structures from one another. As the result, an appropriate bin size for a screening process cannot be determined a priori. A trial-and-error experiment becomes necessary, increasing human intervention and hindering automation.

In this note, we propose to combine the EPA method with moving least squares (MLS) averaging strategy [36] to address the problems mentioned above. It shall be demonstrated that the electron-phonon averaged via moving least squares (EPA-MLS) method exhibits relatively little dependency on associated numerical parameters, requiring virtually no tuning. Moreover, it can be used with coarser calculation results without severe degradation in accuracy. As a result, the EPA-MLS method can become a practical tool for fast-screening TE materials in a more automated environment. We first describe the approach in Section 2 and then validate it by comparing the results to the original EPA method with the bin-based averaging strategy in Section 3. A brief summary follows in Section 4.

## 2. Theory

### 2.1. The EPA method

We first review the features of the EPA method. The description here largely follows the study by Samsonidze and Kozinsky [26], to which the reader may refer for more detailed discussion. The main task of predicting the electronic transport coefficients for electrons within the relaxation time approximation is the evaluation of the inverse of the electron energy relaxation time induced by the el-ph interaction, which is given as follows [37,38]:

$$\tau_{nk}^{-1}(\mu, T) = \frac{\Omega}{(2\pi)^2 \hbar} \sum_{m\nu} \int_{BZ} d\mathbf{q} |g_{m\nu}^{SE}(\mathbf{k}, \mathbf{q})|^2 \times \left\{ \left[ n(\omega_{\nu\mathbf{q}}, T) + f(\varepsilon_{m\mathbf{k}+\mathbf{q}}, \mu, T) \right] \delta(\varepsilon_{n\mathbf{k}} + \omega_{\nu\mathbf{q}} - \varepsilon_{m\mathbf{k}+\mathbf{q}}) \right. \\ \left. + \left[ n(\omega_{\nu\mathbf{q}}, T) + 1 - f(\varepsilon_{m\mathbf{k}+\mathbf{q}}, \mu, T) \right] \delta(\varepsilon_{n\mathbf{k}} - \omega_{\nu\mathbf{q}} - \varepsilon_{m\mathbf{k}+\mathbf{q}}) \right\}, \quad (1)$$

simplest methods [34]. However, such simplification is not always applicable for generic TE materials. At the other end of the spectrum is the electron-phonon Wannier (EPW) method [35], which fully describes the el-ph scattering. However, the EPW method is not completely appropriate for fast-screening due to its multistep procedure with high computational cost.

More recently, a new approach, i.e. the electron-phonon averaged (EPA) method, which combines simplicity and speed with a fully first-principles treatment of the el-ph interaction, has been introduced [26]. By changing the complex momentum-space integration into an integration over energies and simultaneously replacing several terms with their averages within bins over an energy range, the EPA method allows for automated rapid calculations for optimization of electronic transport quantities, while being more predictive than the CRT and DP approximations. The method has been successfully used for screening potential TE materials from a group of half-Heusler (HH) compounds [26]. However, the EPA method with the bin-based averaging strategy first requires determination of the bin size. Excessively large bins may result in the loss of accuracy, while excessively small bins may end up with too few samples within a bin, rendering the averaging process physically unsatisfactory. In addition, even within the same class of materials, the candidate compounds can exhibit very

where  $\Omega$  is the volume of the primitive cell,  $m$  and  $n$  are the electron band indices,  $\nu$  is the phonon mode index,  $\mathbf{k}$  is the electron wavevector,  $\mathbf{q}$  is the phonon wavevector,  $\varepsilon_{n\mathbf{k}}$  is the electron energy,  $\omega_{\nu\mathbf{q}}$  is the phonon energy,  $g_{m\nu}^{SE}(\mathbf{k}, \mathbf{q})$  is the el-ph coupling matrix element appearing in the self-energy expressions [37],  $n(\omega, T)$  is the Bose-Einstein distribution function,  $f(\varepsilon, \mu, T)$  is the Fermi-Dirac distribution function, and  $\delta$  is the Dirac delta function.  $\mu$  is the chemical potential of electrons,  $k_B$  is the Boltzmann constant, and  $\hbar$  is the reduced Planck constant.

The main element of the EPA approximation is to replace the momentum-dependent quantities in Eq. (1) with their energy-dependent averages. The el-ph coupling matrix elements are averaged over the directions of  $\mathbf{k}$  and  $\mathbf{k} + \mathbf{q}$  wavevectors:

$$|g_{m\nu}^{SE}(\mathbf{k}, \mathbf{q})|^2 \mapsto g_{\nu}^2(\varepsilon_{n\mathbf{k}}, \varepsilon_{m\mathbf{k}+\mathbf{q}}). \quad (2)$$

As a result,  $g_{\nu}^2$  becomes a function of two energies, namely,  $\varepsilon_1$  and  $\varepsilon_2$ , which represent the energy of the incoming electron state and that of the outgoing electron state, respectively. Additionally,  $\omega_{\nu\mathbf{q}}$  is also replaced with its average:

$$\omega_{\nu\mathbf{q}} \mapsto \bar{\omega}_{\nu}. \quad (3)$$

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