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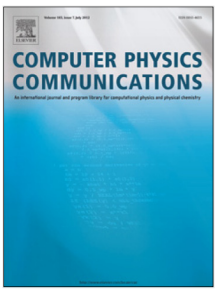

Efficient technique for computational design of thermoelectric materials

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Efficient technique for computational design of thermoelectric materials

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Efficient thermoelectric materials are highly desirable, and the quest for finding them has intensified as they could be promising alternatives to fossil energy sources. Here we present a general first-principles approach to predict, in multicomponent systems, efficient thermoelectric compounds. The method combines a robust evolutionary algorithm, a Pareto multiobjective optimization, density functional theory and a Boltzmann semi-classical calculation of thermoelectric efficiency. To test the performance and reliability of our overall framework, we use the well-known system Bi_2Te_3-Sb_2Te_3.

I. INTRODUCTION

Finding alternatives to fossil energy sources is a priority for many scientific and engineering fields. Thermoelectric energy conversion, that is converting waste heat into electricity, is a particularly attractive method as thermoelectric devices are highly reliable, integrable, stable, and compact[1]. Applications of thermoelectrics include conventional coolers, laser cooling, cryogenic infrared night vision equipment, telecom lasers, electronic cooling and even devices for outer space exploration[2].

Thus, thermoelectric materials have been intensively studied during the last decades, however the energy conversion efficiencies obtained have been quite low, thus limiting the use of thermoelectric devices as promising alternative energy sources (for a review see Ref. [3] and references therein).

The difficulties found in enhancing the thermoelectric efficiency are manifold and depend on the optimization of several, often clashing, parameters. The efficiency or figure of merit, ZT, that characterizes each material is given by the combination of different transport coefficients as follows:

$$ZT = \frac{\sigma S^2 T}{\kappa_e + \kappa_l} \tag{1}$$

where σ is the electrical conductivity, S is the Seebeck coefficient, T is temperature, and κ_e and κ_l are electronic and lattice thermal conductivities, respectively. These quantities have been studied individually, from the experimental and theoretical points of view, and they are frequently tailored according to a particular application. For instance, a large Seebeck coefficient is usually obtained using only one type of carriers (n-type or p-type), however materials with large S tend to have low electrical conductivity σ, thus adjustments have to be made in order to maximize the figure of merit[3]. Also, great efforts have gone into minimizing the thermal conductivity, but this task is far from easy. On one hand, from the first-principles point of view, the description of electrons and holes transporting heat, κ_e, is to some extent tractable within the Boltzmann semi-classical theory and a constant relaxation time[4]. On the other hand, κ_l depends on the structure, rigidity, atomic masses and other characteristics of the lattice[5]. Computations of vibrational properties and lattice thermal transport[6] of individual materials have been done (for a review, see e.g. Ref. [7]), but they are computationally expensive.

In this work, as our main contribution, we demonstrate that an evolutionary algorithm, in combination with density functional theory (DFT)[8,9] and Boltzmann semi-classical calculation of transport properties can be used to find Pareto-optimal solutions in terms of energy and thermoelectric efficiency within finite time, provided a few criteria are met. The ability to predict the most stable crystal structures purely from first-principles using an evolutionary approach such as USPEX[10–12], by providing only the chemical composition, has had a number of successful results widely discussed in the literature. Now we extend this method, to look for structures that are not only the most stable, but at the same time, possess a large figure of merit ZT. This type of optimization task is part of the so-called