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Jury Member Report – Doctor of Philosophy thesis.

Name of Candidate: Tao Fan

PhD Program: Materials Science and Engineering

Title of Thesis: First-principles study of advanced thermoelectric materials: methodology and application

Supervisor: Professor Artem Oganov

Name of the Reviewer: Carlo Gatti

I confirm the absence of any conflict of interest	**Date: 23-08-2022**

The purpose of this report is to obtain an independent review from the members of PhD defense Jury before the thesis defense. The members of PhD defense Jury are asked to submit signed copy of the report at least 30 days prior the thesis defense. The Reviewers are asked to bring a copy of the completed report to the thesis defense and to discuss the contents of each report with each other before the thesis defense.

If the reviewers have any queries about the thesis which they wish to raise in advance, please contact the Chair of the Jury.

Reviewer's Report

The PhD thesis by Tao Fan on "First-Principles Study of Advanced Thermoelectric Materials: methodology and application" is a well-structured report on a quite relevant and successful research project.

The first part of the thesis sets in an appropriate manner the foundations and the scientific/bibliography context of the proposed work, while the following chapters are dedicated to illustrate the core of the work that has been performed, along with the promising results that have been obtained. In particular, Chapter 1 introduces thermoelectricity and the quest for more and more performing thermoelectric materials (TMs). The key properties defining optimal TMs are reviewed, along with the theoretical and computational framework from which these properties may be evaluated. As a natural consequence, the high-throughput screening for thermoelectric materials is introduced and the proposal of a research project aimed at achieving a software code for calculating the transport properties quickly and accurately and at screening materials in a database to find novel compounds with promising properties is outlined. Chapter 2 reviews the existing models for the calculation of the electronic and thermal transport properties and then illustrates those selected for being implemented and significantly generalized/improved in the two developed SW codes, AICON and AICON2, of the present thesis. The technical issues behind this

implementation, the workflow(s) of the codes and their open source distribution under the GNU general public license are outlined. Chapter 2 concludes discussing a test of the proposed and implemented models of the lattice thermal conductivity and of the electrical transport properties on a data set containing 28 well-studied TMs. In chapter 3, the methods introduced in the Chapter 2 are applied to search for promising thermoelectric materials among the structures taken from a database. A first overview of the quality/accuracy of the obtained results is also provided in the chapter. Chapter 4 illustrates in detail the results of the performed TMs screening. The more promising and interesting TMs which have been discovered by the systematic screening are illustrated in terms of their crystal structures, band structures, and transport properties. Chapter 5 concludes. A comprehensive, carefully selected bibliography follows. A final appendix reports the data that could not be covered in Chapter 4 and that, in general, concern TM compounds that have less promising thermoelectrical properties relative to those of the systems illustrated in Chapter 4.

The dissertation is presented in a very ordered manner, it is well structured and carefully written. The exposition is in general concise and clear at the same time. The abstract and the Concluding section have the appropriate length, are crisply clear and adequately informative.

The methods proposed and implemented in the two developed codes are relevant and appropriate and have already been judged favourably by the international reviewers of the two publications listed as number 1 and 2 of the candidate. The results obtained are scientifically very significant, they perfectly comply with the international level and the current state of the art. This judgement is corroborated by their having been published on international journals of great quality and impact. The quality and numbers of publications is perfectly adequate for a PhD thesis. Publications number 1 and 2 deserve a special mention since they offer to the international scientific community two tools (SW codes) of great value for experimental and theoretical research in the quest of novel and ever more efficient TM compounds.

A few minor issues may optionally be considered before the thesis defense:

1. A brief discussion about the scaling problem (chapter 3) may be inserted (why the theoretical results need to be scaled; why they should differently scaled according to the magnitude of the lattice thermal conductivity (LTC); why high LTC are overestimated and low LTC are underestimated (e.g. SnSe); how the scaling problem impacts on the quality/accuracy of the TMs screening process…etc.
2. A two-lines introduction to the features of MPJ vs PBE method as for the band gap evaluation could be inserted at page 68 of the thesis.

During the thesis defense, among other questions, I would like to ask whether the methods and codes proposed in this thesis may be fruitfully combined/embedded into the structural/property optimization codes (say USPEX for instance) and whether it is planned to test in the future the results obtained in this thesis against some experimental, though not yet available, outcomes.

Provisional Recommendation

X☐ *I recommend that the candidate should defend the thesis by means of a formal thesis defense*

☐ *I recommend that the candidate should defend the thesis by means of a formal thesis defense only after appropriate changes would be introduced in candidate's thesis according to the recommendations of the present report*

☐ *The thesis is not acceptable and I recommend that the candidate be exempt from the formal thesis defense*

Milano, 23rd August 2022

Carlo Gatti