Priv.-Doz. Mgr. David Holec, PhD
Department of Materials Science
Franz-Josef-Straße 18
A-8700 Leoben
Austria
david.holec@unileoben.ac.at
http://cms.unileoben.ac.at

Montanuniversität Leoben

Materials Science

To: prof. Mgr. Miroslav Černý, Ph.D.
Brno University of Technology

Leoben, September 10, 2025

Report on a doctoral thesis of Ing. Pavel Papež

Dear prof. Černý, dear Mirek,

In this report, I assess the the dissertation titled "*Theoretical study of magnetic states in alloys with different levels of ordering*" by **Pavel Papež** submitted to the Brno University of Technology for the doctoral degree.

The thesis has 102 pages, including a preface and references, and is divided into eight chapters. These eight chapters cover an introductory part (Chapter 1), an overview of the used theory and techniques (Chapters 2 and 3), results (Chapters 4 and 5), outlook and conclusion (Chapters 6 and 7), and a summary of published papers and other scientific activities (Chapter 8).

The thesis is written in a very **personalised style**, which is something I want to highlight: it reflects the personality of the author and without a doubt gives an **authentic feeling**. It is mostly written in easy-to-read and high standard English, with only a minimum number of typos. The former research as well as reproduced figures are properly cited (in total 122 citations, with more than 1/4 being from the last five years). Its **formal and graphical style** is rather professional, although some images could benefit from higher resolution (Fig. 2.2) or more readable fonts (the Python outputs in results, Chapters 4 and 5). Furthermore, "−" should be consistently used for minus sign instead of "-"; "Kohn Sham theory" should read "Kohn–Sham theory"; incorrect brackets in Eq. 3.7 to denote functional of the charge density; or consistent unit system should be used (cf. kinetic energy terms in Eqs. 3.12 and 3.13). These are only minor issues, which do not disturb the flow of reading.

More serious critiques concern the **unbalanced content** of individual chapters. Chapter 1 (Introduction) finishes without any conclusion and without stating the goals of the thesis. Chapter 2 (Ordering in alloys) is very brief, without providing a clear connection to the thesis. Parts related to describing methods (Bader's charge dissection, Gadget, UMLIPs) could have been included in Chapter 3 (Ab initio calculations), which anyway contains content beyond only ab initio methods (e.g. thermodynamics, ordering in alloys, or molecular dynamics). Some formulations are rather vague and non-scientific (e.g. "These potentials are trained on some databases. . . "–which ones? No reference provided!).

Even more obvious imbalance appears between Chapter 4 (CoCrNi alloy, 32 pages, 30 figures) and Chapter 5 (YMn_2, 10 pages, 7 figures).

The provided **analyses are carefully performed** and very detailed. Some results are not yet fully understood, which is acceptable considering the advanced level of the performed calculations. I also very much appreciate that the author did not present only "success stories", but honestly reported also on difficulties or even unsolved discrepancies with respect to reported experiments. Nevertheless, it is a pity that the thesis does not contain more results and insights that Pavel has obtained during his PhD research (e.g. results from his contributed papers or some conference contributions do not seem to be included). Instead, some preliminary results obtained with the GRACE UMLIP (chapter 4.3.5.) are presented, which make a rather incoherent impression with the rest of the provided results.

It is rather unusual that "Goal" would be presented as the second-last chapter. This is because these are not goals of the thesis, but rather a proposal, or outlook, for the follow-up work. The reader is, however, left to guess this himself, since this chapter starts without any introduction.

Some other unclarities include:

1. What is the physical meaning of comparing CHGNet and DFT total energies in Fig. 4.18 and Table 4.4? Since there is no guarantee that the calculation setups of the here performed DFT calculations and those used to produce the training data are compatible, wouldn't it make more sense to focus on formation energies (energy differences) instead?

2. On page 58, it is written: " Looking at Figure 4.24, it can be seen that the majority of errors arise from the Ni ordered structures, as well as the hcp Cr ordered structure. This hints at a possibility of the model to somehow correctly predict the disordered structure out of the box. The ordered structure, which will probably not be included that often in the training dataset, represents quite a struggle." This is a rather vague formulation. Can the author please clarify what is meant by this statement?

3. Figure 4.21, Comparison of forces. I don't understand this figure. Do the CHGNet calculations correspond to the DFT-relaxed cells (forces ~ 0) without any further relaxation? In such case, however, the structure hpc_3 is not properly relaxed (DFT forces are significantly non-zero). This figure is very confusing.

4. Why GRACE can be used only for molecular dynamics and cannot be used for molecular statics evaluation, similarly to CHGNet? In my experience, this is possible.

5. What is the structure of the AFM7 configuration of YMn_2? It is not shown in Fig. 5.2.

6. Table 5.2 presents energies per atom (in eV/at.) with two significant numbers, same as bulk modulus (in GPa). Can the author prove that this is a meaningful accuracy of bulk modulus, provided the accuracy of the energy (or the shown E–V curves in Fig. 5.5, where, e.g., the green fit for AFM1 does not go through all data points)?

7. The author proposes as the future direction to upfit the GRACE UMLIP with own system specific data. How will you make sure that your DFT data are compatible (accuracy, methods, . . .) with those used for training?

Despite the above raised criticism, I read the thesis with great interest. It provides **scientifically interesting and relevant results** using **advanced and up-to-date methods**. The author has clearly demonstrated mastering them, and being an expert in the field of computational atomistic materials science. Therefore, I strongly recommend this dissertation to be **positively evaluated and to be admitted for a public defence**. Considering my above report, I propose it be graded with 2 (on the grading scale 1=best, 5=failed).

Sincerely yours,

David Holec