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Jury Member Report – Doctor of Philosophy thesis.

Name of Candidate: Iosif Leibin

PhD Program: Materials Science and Engineering

Title of Thesis: Spectra and mobility of open-shell atoms in rare gas crystals: effects of interaction anisotropy

Supervisor: Professor Alexei Buchachenko
Assistant Professor Dmitry Aksenov

Name of the Reviewer: Amar Vutha

I confirm the absence of any conflict of interest YES	**Date: 2024-08-04**

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

Reviewers report should contain the following items:

- Brief evaluation of the thesis quality and overall structure of the dissertation.
- The relevance of the topic of dissertation work to its actual content
- The relevance of the methods used in the dissertation
- The scientific significance of the results obtained and their compliance with the international level and current state of the art
- The relevance of the obtained results to applications (if applicable)
- The quality of publications

The summary of issues to be addressed before/during the thesis defense

Quality of the thesis and overall structure:

The quality of the thesis is good. It is written in a standard format and structure, and has appropriate details and references. The thesis addresses an extremely complicated physical problem using a set of

clearly described methods. Overall I consider this dissertation to be both original and a significant contribution to the body of knowledge in this field.

Relevance of the topic to the content:

The topic and the content of the dissertation are well-matched. The results obtained in the paper elucidate a number of experimental aspects of the spectra of matrix-isolated atoms.

Relevance of the methods used:

As an experimentalist, I am not a practicing expert on the methods used for numerical calculations. But to the best of my knowledge, the methods chosen for this investigation are sound and appropriate. They are described with a good amount of detail in Chapter 3. I particularly found the methods used for Sm in RG to be novel and interesting.

Scientific significance of the results obtained and comparison with international level:

The results described in this thesis are highly significant, being some of the most accurate and detailed investigations of the physics of atoms trapped in rare-gas matrices. These results create new means to gain insight into the complicated physics of trapping in rare-gas matrices.

Relevance of obtained results to applications:

The results described in this thesis will be keenly followed by experimentalists working on matrix-isolated atoms and molecules, a field which has been resurgent recently. The numerical methods and techniques used in this thesis will likely be adopted by other groups interested in studying not just rare-gas matrices but also impurity atom behaviour inside large clusters and crystals. Applications of these methods could lead to improved atomic clocks and magnetometers.

Quality of publications:

The publications listed on page 3 of the dissertation are all in reputed international journals and are of high quality.

Issues to be addressed before/during the defense:

Minor points:

m1. The use of hyperlinks within the document (using the LaTeX package hyperref, for example) would greatly help readers to navigate back and forth between different sections of the thesis and/or the references. See comments under M2.a-c below.

m2. There are a number of minor typos in the document. While they do not affect any of the scientific content, they should be easy to fix using a spellchecker. I recommend that the document should be carefully proofread before final submission.

Major points:

M1. <u>Atoms vs molecules.</u> The last paragraph of Section 4.3.3 mentions that the physics of matrix-isolated P-state atoms is likely to be similar to that of diatomic molecules in matrices. It should be clarified whether this comment refers to homonuclear diatomic molecules (e.g., Mn_2), where the electron density does resemble that of P states in atoms, or whether it is also applicable to polar

diatomic molecules (e.g., BaF) where the electron density is quite asymmetrically distributed.

M2. References and details. I recommend the addition of references in a few places, and slightly clearer explanations of symbols. For example:

a. Paragraph following equation (1) on page 44: it is unclear which of these is related to alpha, and which one to beta, and how. A reference to further details, or a more detailed explanation, would be useful.

b. Section 2.6.1: for a non-expert like me, it was not easy to understand terms such as «GGA functionals». An explanation of such new terms, wherever they are introduced in the text, would make the thesis useful to readers who are new to the field. Internal references would also help. For example, the discussion in Section 4.1.1 where the «TT model» is referred to could use an internal reference back to Equation (34), where this model is described in more detail.

c. There are some discussions where the details are hard to follow. For example, on page 119 in last paragraph of Section 4.5.4, we have the sentence «reaffirming the original authors' interpretation». Original authors of which paper? What was their interpretation? To make the thesis easier to follow and understand, internal references to sections of the thesis where these ideas were first introduced — or external references to the original papers — to should be included for such paragraphs. Again, hyperlinks in the document would be very useful in such situations.

M3. «Convex hull». On page 44 paragraph 1, it is indicated that the convex hull method would be described in greater detail in Section 3. (Should that be Chapter 3? Section 3.1? Again, an internal hyperlink would be helpful here). However the paragraph after Equation (43) in Section 3.1 has just one single line regarding the stability diagram and the convex hull.

It is possible that the term «convex hull» is used with a particular meaning in theoretical quantum chemistry. But to someone who is not an expert in this field — but quite familiar with the concept of a convex hull from geometry — the use of this term in the thesis is a bit confusing. As this notion of convex hull is repeatedly mentioned in the thesis, it will be useful to readers if the following basic ideas are clearly laid out:

a. Exactly *what* distribution of points in a 2D plane is this convex hull being calculated for?

b. Why is the convex hull a useful concept? Instead of just using the plot of Delta E(N_del) versus N_del, why bother with the convex hull?

Provisional Recommendation

☒ *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*