Kraków, 24 April 2923

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POLISH ACADEMY OF SCIENCES

in Kraków

dr hab. Andrzej Ptok
[Ph.D., postdoctoral degree holder],
professor of the Institute of Nuclear Physics of the Polish Academy of Sciences
Department of Materials Research by Computers

Review of the doctoral dissertation of Mr. Rajibul Islam, M.Sc., entitled: "Topological phases of 3D superlattices and 2D materials: theoretical modeling"

The doctoral dissertation submitted for review by Mr. Rajibul Islam, M.Sc., entitled: "Fazy topologiczne 3D supersieci oraz materiałów 2D: modelowanie teoretyczne" (original title in English: "Topological phases of 3D superlattices and 2D materials: theoretical modeling"), was prepared as part of doctoral studies at the Institute of Physics of the Polish Academy of Sciences in Warsaw, under the supervision of the supervisor dr hab. Carmine Autieri *[Ph.D., postdoctoral degree holder]*, professor of the Institute of Physics of the Polish Academy of Sciences.

The dissertation, written in English, is of a theoretical nature and it is a written work that constitutes a collection of published and thematically related scientific articles (in accordance with Art. 187.3 of the Act on the "Law on Higher Education and Science"). The dissertation has a total of 75 pages (including three publications of the Ph.D. candidate), and it is divided into 4 chapters, preceded by: *Abstract* (in Polish and English), *Dedications and acknowledgments*, *Author's declaration*, *List of publications* (included in the dissertation and beyond its scope) and *A list of drawings*. The last unnumbered chapter, i.e. *Bibliography*, contains 118 references. The core part of the dissertation (4 chapters mentioned earlier) is divided into *Introduction*, a description of the *Methodology*, a chapter on *Selected articles that make up the dissertation with a summary* (to which 3 scientific articles are attached), and a *Perspectives* chapter.

Comments on the content of the dissertation.

Chapter One, *Introduction*, describes the issue of the "order parameter" in Landau's theory and the limitation of its applicability in the context of the quantum Hall effect. On this basis, the author justifies the need to introduce non-local order parameters (topological invariants). This chapter includes a subchapter of topological materials (due to time-reversal symmetry-breaking, i.e. *time reversal symmetry*, and the existence of an energy gap – Fig. 1.1) - however, this classification should be discussed in more detail in the main text of the chapter, and the meaning of abbreviations repeated in the description of the drawing. The author then describes the theoretical aspects of topological phases in systems preserving time reversal symmetry (section 1.1). Interestingly, the abbreviation TR (or *time reversal*) is not explained in the text (although it appears in Figure 1.1 or the title

of Section 1.1). In the paragraph preceding Section 1.1, the notation TRS is used interchangeably: … *in TRS Reserved and TRS broken symmetry*. In the second case, *symmetry* is redundant because it is already abbreviated as TRS (i.e. time reversal symmetry). A similar problem relates to the abbreviation TKNN (first sentence of page 2) – although in this case the meaning can be guessed from the context of the sentence that it is an abbreviation of the names *Thouless-Kohmoto-Nightingale-Nijs*.

The part describing systems that break time reversal symmetries appears as subchapter 1.1.2, containing a description of e.g. antiferromagnetic systems – this fragment should be separated in this section. Fig. 1.2 is used to describe the band inversion in the HgTe and CdTe systems (inversion of the Γ_6 and Γ_8 states) – it is a rather non-intuitive example (no simple explanation of the meaning of these states in the context of atomic orbitals). In addition, there are some editing problems, e.g. *ladnau* should be *Landau*, missing spaces in some places (permanently between text and references, *Dirac semimetal(DSM)* or *Weyl semimetal(WSM)* subsection 1.1.2).

The introduction may be difficult for a reader unfamiliar with the topic discussed. There are many concepts here that are poorly explained or not explained at all. Although the dissertation deals with *ab inito* calculations (DFT), theoretically the relations given in the introduction do not explain how given quantities (e.g. topological indices) can be numerically determined (e.g. there are no appendices that would describe this aspect). For didactic reasons, the introduction should be much more extensive and concern generally understood topological systems – mainly experimental results (which were not lacking in the introductions to the Ph.D. candidate's scientific articles) bringing the subject of topological materials in the context of practical implementation.

Chapter 1 ends with the title of section 1.2 *The main theme and content of the thesis*, the content of which has been removed from previous versions of the doctoral dissertation. This means that in the current version, **there is no section ending the chapter and defining the purpose of the doctoral dissertation**. In addition, **this chapter contains 7 figures, but there is no information about their origin** – i.e. literature references.

Chapter Two, *Methodology*: This chapter presents basic information on the techniques used (e.g. description of the multi-electron system as a justification for the *density functional theory* (DFT), description of the DFT technique itself, exchange-correlation functionals (e.g. LDA, GGA, PAW), and tight binding models in the base of Wannier orbitals). The first paragraph contains information about the software used (i.e. *VASP*, *wannier90,* and *wanniertools*).

While sections 2.1 and 2.2 concern typical calculations, section 2.3 (entitled *Wannier based Wight-binding model*) is particularly important due to the numerical results presented in the author's publications. It seems to me that the description of Scheme 2.1 is generally

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wrong and should be much more extended – it rather presents a strategy for finding tight binding models in the Wannier base of the studied materials, and this is only the first step to "predicting new topological materials" (as the caption on the picture says). The schematically presented approach is generally used, also outside the study of topological systems (it is worth mentioning its successes in numerous theoretical studies of iron-based superconductors).

Section 2.3 is described very sparingly, although it concerns the main technique used in the author's research and presented in the publications included in the dissertation. The phrase "This reduces the size of the matrix" appears – without explaining which matrix is meant (last sentence of the first paragraph of section 2.3). Equation 2.29 is written incorrectly – the index *n* describing the wave functions (Bloch states) cannot simultaneously number the Wannier W_{nR} states. In Equation 2.30, there is an error in the Wannier state indices (similar to Equation 2.31). I expect that in the course of a possible defense, the correct relationships will be presented.

In Section 2.3, a distinction is made between the notation of H taking into account only the position of the primitive cells (Equation 2.32) and the exact positions of the Wannier orbitals (Equation 2.33). This paragraph ends with the sentence: *The formalism in Eq. 2.33 is more accurate in the context of topological materials due its relations with Berry phase and Berry curvature*, however, this context is not given. Please provide it and explain why 2.33 is more appropriate in this context. It is worth mentioning that *wanniertools* uses both expressions (depending on how WANNIER_CENTERS is defined in wanniertools input files). In the next paragraph, relationships on the Berry phase are given. However, they are presented for the 1D system, without taking into account the formalism presented in this section – during a possible defense, please present the exact relationships to determine the Berry phase, using the Hamiltonian in the base of Wannier orbitals.

There is no information in section 2.3 on the method used to calculate the surface states. This is strange because in practice all the results presented in the doctoral dissertation concern mainly research using this method (and implemented in *wanniertools*). This method should be thoroughly presented and discussed.

To sum up, section 2.3 deals with a technique used in virtually every work by the author – however, the description presented is very cursory, and in a didactic context it is practically of no value. For example, there is no detailed description of the strategy used to find maximally localized Wannier states (under *wannier90*) from Bloch states (under VASP/DFT). Doubts arise as to whether the Ph.D. candidate understands the applied methods based on tight binding models obtained from accurate DFT calculations.

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Main content

Chapter Three is a collection of three papers by the author included in the doctoral dissertation. It is divided into three sections for each of the papers. Section **3.2** deals with the search for planar forms in 3D systems based on HgTe/CdTe and HgTe/HgSe superlattices (Phys. Rev. Research 4, 023114 (2022)). Section **3.3** discusses the spin dependence of the electronic structure of the ultrathin 2D MSi_2N_4 layer – exact monolayer and bilayer (Phys. Rev. B 104, L201112 (2021)). Section **3.4** presents the results predicting the existence of the 1T' phase of MSi_2Z_4compounds where M=Mo, W and Z=P, As (Phys. Rev. B 06, 145149 (2022)). Despite the title of the chapter, there is no separate section summarizing the collected results – therefore, combined with the lack of the "aim of the dissertation", it is difficult to say whether the assumed goal has been achieved.

I did not find a comparison of band structures obtained from accurate DFT calculations with band structures obtained from tight binding model in the base of Wannier orbitals in any of the papers. Therefore, there is no certainty that the obtained models correctly reproduce the band structure. During a possible defense, please present such comparisons for the compounds studied in the presented papers.

(Section 3.3) In the paper published in Phys. Rev. B 104, L201112 (2021), Fig. 1 presents the dispersion relations for photons realized in one and two monolayers and in the 2H-$MoSi_2N_4$ crystal – in the case of panel (c), i.e. the result for the crystal, no explanation for flat bands with almost zero frequency. (A similar result for $MoSi_2As_4$ is presented in Fig. 5). Does this mean that the system is at the limit of stability?

(Section 3.4) In the paper published in Phys. Rev. B 106, 245149 (2022) the photon dispersion relation for $MoSi_2P_4$ is also presented – according to Section II of this work, PHONOPY and DFPT (i.e. T=0) were used. So what was the purpose of using MD (result presented in Fig. 1(e)) in the context of photons? This information is not given in the text. Apart from the oscillation of the mean "free energy" around a certain value, this result does not guarantee the absence of soft modes and the stability of the system at T=300 K for a given symmetry but only shows that no structural transition occurred during the (relatively short) simulation. However, it is worth noting that the result of the MD calculation can be used to determine the dispersion relation at T=300 K – so why wasn't it done?

Contribution of a Ph.D. candidate to the publication.

In the sections on scientific publications, there is no information on the Ph.D. candidate's contribution – this contribution is "described" in the statements of the authors and the Ph.D. candidate himself (separate attachments to the doctoral dissertation). The dissertation is based on three papers published in Phys. Rev. Research and Phys Rev. B (two papers). In these works, the Ph.D. candidate is the first author, and these papers have from 7 to 10 authors. As I mentioned earlier

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- the contribution of the Ph.D. candidate and co-authors are presented in separate attachments. However, they are prepared in a very superficial way and enigmatically state:
"The most of the work of the above papers are done by me." – in the Ph.D. candidate's statement, "I declare that my contribution […] has been related to discussion on the scientific project and results as well as the writing part of the paper." – for all other statements by co-authors.
Therefore, the question arises whether such a large group of co-authors was necessary, since most of the "work" was done by the Ph.D. candidate, and none of the co-authors can point to a specific section that they "did".

The quality of the dissertation.

In my opinion, the doctoral dissertation was hastily and inaccurately prepared (as evidenced by the editing errors described above, deficiencies in descriptions, etc.). In the future, however, I recommend greater care in the preparation of texts.

Lack of spaces is common – especially between text and references. Some of the drawings are much too small, which reduces their legibility. These drawings mostly present a band structure or a surface Green's function, i.e. structures that are usually very complex – the details of which are very important – the size of drawings in APS publications is not limited, so they could be made in a two-column form.

The bibliography in the dissertation (page 107) takes the form of a rather unusual reference to references in the form of: *Author, Title, Journal, Volume:Pages, Year*. The adopted form is not uniform for all records, sometimes the references provided have the entry *Page* from-to, others only the number of the first page – it would be preferable to adopt a uniform form. There are also omissions regarding the spelling of *Journal* names and chemical formulas. In this case, the author did not exercise due diligence in checking the references. Sometimes he uses the notation *Review of Modern Physics* (in ref. 21), sometimes *Review of modern physics* (in ref. 25) – with the first notation being preferred; similarly in the case of *Physical review letters* (in ref. 5, 6, 7, 8, 9, etc.) – it should be *Physical Review Letters*. Titles of references were also not properly edited: e.g. topological index Z_2 (in the title ref. 9 and ref. 28), surnames with a lowercase letter (e.g. *dirac* in ref. 13 or ref. 16, *hall* in ref. 19 or *van deer waals* in ref. 20), and chemical formulas (e.g. *snte* in ref. 15, *mnbi3te4* in ref. 19 and in ref. 20, *insulator-bi4br4* in ref. 31, *bi2se3, bi2te3 and sb2te3* in ref. 32 and others).

The purpose of the work is practically not defined – there is no section 1.2 ending chapter 1. There is also no clear summary, although it is mentioned in the title of Chapter 3.

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In conclusion, the doctoral dissertation of Mr. Rajibul Islam, M.Sc., is in the form of a collection of scientific articles (in accordance with Art. 187.3 of the Act on the *Law on Higher Education and Science*). **However, I regret to say that the quality of the presented doctoral dissertation is relatively low – in my opinion, it does not meet the statutory requirements for scientific dissertations** (Art. 187.1 and Art. 187.2 of the Act on the *Law on Higher Education and Science*). **The quality and form of the prepared dissertation** (discussed in numerous comments described above) **raise my serious doubts about the ability to conduct independent research work** – enigmatic statements regarding the authors' contributions do not facilitate the assessment of the Ph.D. candidate's skills; the deficiencies in chapter 2.3 of the dissertation suggest a lack of knowledge of the details of the techniques used. **The scientific problem being solved in the doctoral dissertation was not given** – the lack of a defined goal of the work (missing chapter 1.2) and a clear summary (despite chapter 3) of whether this goal was achieved; it is hard to say whether the Ph.D. candidate is aware of the purpose of the work since he did not define it in the doctoral dissertation. **As a result of the above, I motion that Mr. Rajibul Islam, M.Sc., not be admitted to public defense.**

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