UNIVERSITY
OF WARSAW

FACULTY OF
PHYSICS

dr hab. Nevill Gonzalez Szwacki *[Ph.D., postdoctoral degree holder]*

INSTITUTE OF EXPERIMENTAL PHYSICS
tel.: (+4822) 55 32 797
e-mail: gonz@fuw.edu.pl
www: www.tsunano.org

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

The doctoral dissertation of Mr. Rajibul Islam, M.Sc., was written at the International Centre for Interfacing Magnetism and Superconductivity with Topological Matter – MagTop at the Branch ON-6 of the Institute of Physics of the Polish Academy of Sciences, under the supervision of dr hab. Carmine Autieri *[Ph.D., postdoctoral degree holder]*, professor of the Institute of Physics of the Polish Academy of Sciences.

For several years, there has been a huge increase in interest in research on topological (3D) insulators, i.e. materials that are insulators inside with conductive states on the surface, which causes electrons to move only along the surface of the material. Meanwhile, conductive surface states that are protected by symmetry with a non-trivial topological order exhibit spin-blocking behavior, a behavior that is suitable for spintronics. The two-dimensional (2D) counterpart of the topological insulator, i.e. the material with the quantum spin Hall effect, also exhibits symmetry-protected non-trivial topological states with spin order at the edges of the 2D thin layer. In addition to 3D and 2D topological insulators, many topologically protected materials have also been discovered, such as magnetic topological insulators, Dirac and Weyl topological semimetals, and nodal line semimetals. Moreover, several topological superconductors that can accommodate Majorana fermions have also been proposed recently, which can be used to create more stable quantum computers.

As a result, the doctoral dissertation of Mr. Rajibul Islam, M.Sc., on the topological phase of 3D superlattices based on HgTe and 2D MSi_2Z_4 materials (M = Mo, W and Z = N, P, As) perfectly fits into the current research trends devoted to discovering new types of 2D or 3D topological materials, a better understanding of their intriguing properties, and developing potential applications based on topological materials.

The doctoral dissertation is 75 pages long and consists of 4 chapters; it is written in English, summaries in English and Polish, and there is an extensive bibliography (118 items) and copies of 3 scientific articles.

At the beginning of the dissertation, the Ph.D. candidate outlined the subject matter as well as the purpose and scope of the dissertation. There are a lot of materials with a topological phase nowadays, and their concise description and classification is not easy. The author of the doctoral dissertation took up this task in **Chapter 1**. In this chapter, topological materials are divided into two categories depending on whether they remain invariant under the time reversal operation or not. In addition to the characteristics of topological materials, the chapter contains references to specific "suspect" materials and experimentally confirmed that they are topological materials (having two basic features – the existence of an energy gap and the occurrence of surface states placed in the energy gap and having a specific symmetry), as well as materials that have symmetry-protected topological phases, such as topological crystalline insulators, Dirac/Weyl semimetals or nodal semimetals are also discussed. The Author also described new exotic magnetic phases with non-collinear order in 2D and 3D materials, where spin and orbital quantum entanglements are the stabilizing factor.

In this way, the classification of topological phases has been extended to the case in magnetic ordering that coexists with a topological semimetal with Dirac-type relativistic electronic states. This chapter is a concise but complete introduction to the subject of the doctoral dissertation. The only remark that should be made is that the uniform notation in the presented formulas has not been preserved (e.g. the periodic part of the Bloch function is written in several ways), which makes it difficult to link the information contained in the individual subsections.

In Chapter 2, the Ph.D. candidate begins by discussing the calculation methodology based on which the parameters describing the topological properties of the tested materials were determined. According to the description, the exact determination of the Fermi surface begins with low-resolution calculations from the first principles of the electronic structure of the test material using the density functional theory (DFT). Then, the resulting Bloch functions are mapped to the maximally localized Wannier functions, and the matrix elements of the tight binding Hamiltonian are computed. Finally, Wannier functions can be used to interpret eigenfunctions and other quantities in reciprocal space on a much finer lattice compared to the initial first-principles computation. Surface states can be calculated using the supercell method for a modeled thin layer of material. Although this method is simple and takes into account various surface properties, it is quite computationally demanding because it requires a supercell and a large vacuum area. Later in Chapter 2, the Ph.D. candidate discusses the basics of DFT, i.e. the theory that is implemented in the VASP numerical calculation package, used to obtain the electronic structure and wave functions for the tested materials. In general, the presented description is substantively correct and proves the Ph.D. candidate's good knowledge of DFT methods. After the introduction, the first subchapter contains a description of the multi-electron system. Elements of the DFT, such as the Hohenberg-Kohn theorem, the Kohn-Sham method of determining the ground state energy, and the self-consistent calculation of the ground state electron density, and the basic classes of approximations of the exchange and correlation energy functional forms are presented in later sections. Extending the DFT methodology by introducing, for example, the Hubbard correction (DFT+U) or the dispersion correction (vdW) is presented in the subchapter entitled "Beyond GGA". This subchapter is far too short and the abbreviations of calculation methods presented there (e.g. mBJ-GGA) appear without proper introduction. It is not clear why these, and not other approximations to the exchange and correlation functional, were used in the calculations. It is quite a known fact among a small group of specialists that the HSE functional (from the names of the authors: J. Heyd, G.E. Scuseria, M. Ernzerhof) is the most popular hybrid functional for solids, but there is no mention in the paper whether e.g. other hybrid functionals were also tested. The greatest lack of information, however, is in the last subchapter entitled "Wannier based tight-binding model". The Vasp, Wannier90, and WannierTools software were used to describe the topological states related to the geometry of the wave functions. However, these are numerical packages with high functionality and a more detailed description of the calculation scheme for the description of topological phases is missing.

The basis of the doctoral dissertation of Mr. Rajibul Islam, M.Sc., consists of 3 publications, including: two in Physical Review B ([2] and [3]) and one in Physical Review Research ([1]). The attached statement shows that Mr. Rajibul Islam, M.Sc., played an important role in the preparation of each publication, starting from the research idea, through research hypotheses, numerical calculations, results analysis and data visualization, and on the preparation of manuscripts (80%, 90%, and 80% share for publications [1], [2] and [3], respectively) to publication. All papers have been published in very good, specialized scientific journals from the *Journal Citation Reports* (JCR) list, so they have been thoroughly analyzed and evaluated in the review process, so I will briefly discuss them, focusing on the most important results.

In Chapter 3, each publication is discussed according to a similar scheme: at the beginning, the author presents detailed goals and formulates research hypotheses, and then presents

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the most important results. At the beginning of the doctoral dissertation, the Author placed a list of 12 other works (8 already published and 4 in the process of publication), where the Ph.D. candidate is a co-author and which are thematically related to the reviewed doctoral dissertation. It is worth emphasizing that Mr. Rajibul Islam, M.Sc., is the first author in 3 publications from this additional list, and the 8 published papers have been published in such journals as Physical Review B, Nanoscale, and 2D Materials.

In Chapter 4, the Author summarized the most important research results and presented his ambitious plans for the future.

In publication [1], **Topological states in superlattices of HgTe class of materials for engineering three-dimensional flat bands**, zinc blende superlattices consisting of HgTe, HgSe, and CdTe were considered. Unlike the trivial insulator CdTe, the bulk semiconductors HgTe and HgSe have a band inversion that can be adjusted by changing the layer thickness, strain, and temperature. In systems built of these materials, topological phases may appear, such as the topological insulator phase, the Weyl semimetal phase, and the phase of the quantum spin Hall effect. In this paper, using calculations from the first principles, the Ph.D. candidate showed that the HgTe/CdTe and HgTe/HgSe superlattices arranged along the (001) direction demonstrate a wide range of topological phases, including nodal line phases for unstressed structures. The band structure of HgTe/CdTe has a band gap of several meV along the high symmetry directions with a "camelback" line along the $\Gamma \rightarrow Z$ direction. Many band crossing points far from the high symmetry direction were observed. By using the tight binding model, the Ph.D. candidate found multiple nodal points at the Fermi level forming a ring. The topological surface state that connects the nodes was interpreted as a state belonging to nodal line semimetal. Strain and hydrostatic pressure lead to the formation of a normal insulating phase in HgTe/CdTe. In addition, the work revealed that the GHgTe/HgSe superlattice is a perfect Weyl semimetal. The Weyl phase in HgTe/HgSe is resistant to hydrostatic pressure, but at critical pressure, it starts to form a Dirac semimetal phase, i.e. HgTe/HgSe is a Weyls semimetal with a topological Lifshitz transition. However, the Weyl phase is affected by uniaxial compression or strain causing a phase transition from the nodal phase to the topological insulator phase.

In publication [2], **Tunable spin polarization and electronic structure of bottom-up synthesized $MoSi_2N_4$ materials**, the spin-dependent electronic structure of ultra-thin 2D MSi_2Z_4 materials (M = Mo or W and Z = N or As) in the 2H phase was analyzed. For this purpose, the Ph.D. candidate performed first-principles calculations, which began with examining the structural stability of the tested 2D compounds and examined properties using the valley degree of freedom for MSi_2Z_4, i.e. the so-called "valleytronics", in these materials. MSi_2N_4monolayers are diagonal-gap semiconductors with large spin-spin splitting at K and K'. The monolayer lacks inversion symmetry, so it shows 100% spin polarization at K and K', and the bands with the opposite spin orientation are locked at K and K', which results from the presence of reversal symmetry in time. Unlike the K points, the bands at Γ and M and M are doubly degenerate. When we consider a bilayer, the inversion symmetry is preserved and consequently the bands are doubly degenerate and the resultant spin polarization of the materials is zero. The applied electric field allows spin polarization to be tuned; the presence of an electric field creates a charge imbalance that breaks the local inversion symmetry, pushing the bands of the second layer downwards if the electric field is positive and upwards if the electric field is negative. In publication [2], the Ph.D. candidate also shows that the bulk material 2H-$MoSi_2N_4$ is thermodynamically stable. The Author suggests that it is possible to synthesize the three-dimensional equivalent of the 2H-$MoSi_2Z_4$ compound using bottom-up synthesis methods. The energy gap of this material decreases with the increase in the number of layers and reaches the value for the bulk material after 8 monolayers, but the nature of the gap remains the same (i.e. the gap is diagonal). Similarly, the spin polarization starts

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to decrease exponentially as an odd number of layers increases; for an even number of layers, it is zero because it restores the inversion symmetry. The Ph.D. candidate also established that the nature of the gap changes from straight to diagonal by converting N to AS into Z.

In publication [3], **Switchable large-gap quantum spin Hall state in two dimensional MSi_2Z_4**, the Ph.D. candidate predicted 1T-MSi_2Z_4, a new thermodynamically stable phase for $MSiZ_4$ materials (M = Mo, W and Z = Z, As). In contrast to the trigonal prism of the 2H phase, the M atoms in the 1T-MSi_2Z_4 materials form an octahedron with six Z atoms. The lack of negative phonon frequencies in the dispersion for phonons indicates that it is a thermodynamically stable phase, moreover, molecular dynamics calculations at 300 K show an insignificant change in free energy, which also confirms the lack of spontaneous breaking of bonds in the system. One of the important features of the QSH insulator is the topologically protected helical edge states that connect the valence and conduction bands. Using the Green's function method (which was not described in **Chapter 2**) implemented in the WannierTools code, an edge state was obtained with a symmetry-protected Dirac cone at the point Γ. It was also shown how the electric field affects the topological phase transition: the energy gap decreases with the increase of the electric field, becomes zero at the critical electric field, and increases with further increase of the electric field. This is a key aspect of the quantum spin Hall transistor.

The research results presented in papers [1], [2], and [3] are very important from the point of view of potential applications of the tested topological materials in nanoelectronics.

During the editing of the doctoral dissertation, the Author failed to avoid minor errors and inaccuracies, which I list in the table below as a reviewer's duty.

page	it is	it should be	page	it is	it should be
1	and the of	and the value of	24		
1	don't breaks	don't break	25	four year	four years
1	QHE phase poses	QHE phase possess	25	isoenergic	isoenergetic
2	they shows	they show	25	semi metal	semimetal
2	can be evolve	can evolve	26	create nodal	creating a nodal
2	It also shown	It is also shown	26	isoenergic	isoenergetic
2	for field of	for the field of	26	weyl	Weyl
4	either either	either	46	MSi2N4	MSi_2N_4
21	MBJGGA	mBJ-GGA	53	bandstructure	band structure
24	$u_{nk}(\vec{r}) = nk\left(e^{-ik.\vec{r}}\right)$	$u_{nk}(\vec{r}) = \Psi_{nk}\left(e^{-i\vec{k}.\vec{r}}\right)$	53	start decreases	decreases

These minor shortcomings do not significantly affect the reception of the doctoral dissertation, which presents a high substantive level.

The doctoral dissertation presented for review has been written in English in an understandable way, it is logically composed and quite clearly edited. It contains several original and valuable results published in very good, peer-reviewed scientific journals. In his research, the Ph.D. candidate used advanced theoretical methods and performed many time-consuming numerical calculations. The author is very well versed in the issues related to the topic of the doctoral dissertation concerning the topological phases of matter: he quotes the latest items abundantly and accurately selects them for the issues discussed which proves that he regularly follows the literature.

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It is hard to resist the impression that the doctoral dissertation of the Ph.D. candidate actually consists of two unrelated parts. The first one concerns HgTe/CdTe and HgTe/HgSe superlattices and the second one concerns 2D materials of the MSi_2Z_4 type (M = Mo, W and Z = N, P, As). Both parts could have separate introductions, calculation methodology, and a summary, which would make it easier to understand the issues presented in the doctoral dissertation. It is also not entirely clear why this class of materials was chosen among all those that the Ph.D. candidate dealt with during his doctorate. The dissertation lacked a concise discussion of other achievements of the Ph.D. candidate, which are reflected in the Author's publications from the list of publications related to the doctoral dissertation. By far, the greatest insufficiency is felt when reading **Chapter 2** on the methodology of calculations: it extensively discusses the basics of the density functional theory and lacks a more detailed discussion of the method of obtaining topological parameters characterizing the topological states of matter.

In conclusion, I state that the doctoral dissertation meets all the conditions set out in Art. 13 Sec. 1 of the Act of 14 March 2003 on academic degrees and titles and degrees and titles in the field of art (Journal of Laws of 2003 No. 65, item 595, as amended) and I motion to admit Mr. Rajibul Islam, M.Sc. to the next stages of the doctoral procedure.

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