Rewiew of a habiliation thesis

Title: Modeling of electronic and optical properties of two-dimensional materials

Name of the candidate: Mgr. František Karlický, Ph.D.

Overview of the thesis content

The habilitation thesis under review is based on a coherent body of twelve peer-reviewed publications co-authored by the candidate and published in reputable, high-impact journals in the field of materials science. With a maximum of four authors per publication—most often only two—the candidate's substantial personal contribution to the research is evident.

The thesis addresses a topic of high current relevance and scientific impact: the theoretical modeling of electronic and optical properties of two-dimensional (2D) materials. Given the continuing emergence of new 2D materials and their broad range of potential applications in nanoelectronics, optoelectronics, and energy conversion, the subject is both timely and strategically important. The field is characterized by rapid development, and the need for predictive theoretical approaches is particularly acute, making the candidate's work highly pertinent.

The structure of the thesis is clear and logical, comprising four main chapters, supported by 63 references and supplementary material (including the core articles). Following the introductory chapter, the author discusses the general role of theoretical modeling in the prediction and understanding of electronic and optical properties of 2D materials. This includes a concise overview of current challenges in the field, offering a critical perspective on the limitations of existing approaches and the need for further methodological development.

The core of the thesis presents and briefly comments on results from many-body reference calculations applied to several groups of 2D materials: halogenated and hydrogenated graphenes (References [1, 4]), binary hexagonal materials (References [2, 7]), and transition metal carbides—MXenes (References [8–12]). Complementary insights are provided by DFT-based and semiempirical methods (References [3, 5, 6]), which are used to predict and compare advanced material properties.

From a formal standpoint, the thesis is very well prepared. The text is clearly written, logically organized, and visually well presented. Typographical and grammatical errors are negligible, and the overall readability is very good.

In summary, the thesis represents a significant and timely contribution to the theoretical understanding of 2D materials. It demonstrates the candidate's expertise in advanced computational techniques and their application to a class of materials at the forefront of contemporary condensed matter and materials physics.

Key achievements and scientific relevance

The scientific relevance of the thesis is high, as it addresses core methodological and application challenges in the theoretical study of two-dimensional (2D) materials—an area of intense ongoing research due to its broad technological potential. The research focuses on two complementary research directions pursued by the candidate over the past six years: (1) the development and application of accurate, many-body computational methods for determining reference electronic structures and optical spectra, and (2) the implementation of computationally efficient, approximate methods suitable for large-scale predictions across diverse material families.

This dual approach is both scientifically relevant and practically necessary. While the first line of research emphasizes physical accuracy and deep methodological understanding, it is constrained by the high computational cost of many-body techniques. Nonetheless, these precision-based calculations are critical, as they serve as benchmarks for validating the second line of research, which involves more scalable, approximate techniques. The careful cross-checking of approximate predictions against reference results or available experimental data adds robustness and credibility to the findings.
The materials studied—halogenated and hydrogenated graphenes, binary hexagonal 2D semiconductors, and monolayer transition metal carbides (MXenes)—are all at the forefront of materials research. Each class exhibits distinct electronic and optical characteristics, which are relevant for applications ranging from flexible electronics to catalysis and energy storage.
Furthermore, the entire body of work has been peer-reviewed in respected journals, ensuring the scientific quality and acceptance of the results within the community. The candidate's ability to navigate both high-level theoretical development and application-focused prediction underscores the scientific depth and broader relevance of the thesis.

Final assessment

The habilitation thesis presents a well-structured and scientifically sound body of work centered on the electronic and optical properties of two-dimensional (2D) materials, with a strong emphasis on theoretical modeling. The author has successfully pursued two parallel research directions—high-accuracy reference calculations using many-body methods, and the development and validation of computationally efficient approximations for broader applicability across material classes. The thesis demonstrates a deep understanding of both physical principles and computational techniques, while also offering practical tools for future predictive modeling in this rapidly evolving field.
The importance and timeliness of the research cannot be overstated. The field of 2D materials remains one of the most dynamic areas of contemporary condensed matter physics and materials science. New material families such as MXenes, MBenes, and 2D perovskites continue to emerge, and novel phases—ranging from topological insulators to excitonic insulators—are actively being explored. The diversity of physical phenomena in 2D systems, such as high optical absorption, tunable bandgaps, and magnetic ordering, underpins a wide spectrum of potential applications. Furthermore, the integration of individual 2D layers into van der Waals heterostructures introduces a new level of material design, with properties that can be tailored beyond those of the constituent monolayers.
This field's rapid development and broad technological impact are underscored by the early awarding of the Nobel Prize for graphene research, and the initiation of large-scale programs like the EU's Graphene Flagship. The candidate's contribution aligns well with these global efforts, offering both methodological insight and novel predictive results.
The thesis is formally well-prepared, clear in its structure, and supported by high-quality publications. Taken as a whole, it represents a substantial and original contribution to the field of theoretical materials science.

Recommendation: I fully support the acceptance of this thesis as a habilitation work.

Discussion Points for Consideration:

1. In your application of TD-HSE06 to incommensurate van der Waals heterostructures, you generated large supercells to model prototypical bilayers such as hBN/WSe_2 and MoS_2/WSe_2. How well do your computed optical spectra and interlayer excitonic features reflect available experimental data, and to what extent can your approach be further developed to capture realistically the moiré potentials or exciton minibands observed in twisted heterostructures?

2. An important feature of transition metal dichalcogenides (TMDs) is their valley-selective optical response and interaction with circularly polarized light. To what extent do the methods introduced in your thesis enable the calculation of polarization-resolved spectra? How do these results compare in scope to those following the Maialle-Silva-Sham formalism traditionally employed to describe valley coherence and exciton dynamics?
3. In your treatment of fluorographene, a significant result was the resolution of the apparent gap discrepancy between experiments and many-body GW calculations—primarily attributed to strong excitonic effects. Given the additional roles of defects, inhomogeneity, varying levels of fluorine substitution, and substrate interactions, how do the methods you employed account for these environmental and structural complexities? Could you elaborate on your approach to modeling such real-world conditions and its limitations or potential improvements?
4. Could you please confirm whether you'd like the question to focus on the selection criteria for including specific publications in the habilitation thesis—despite the existence of other works by the candidate in higher-impact journals?

Prague, 15th May 2025 prof. RNDr. Jana Kalbáčová Vejpravová, Ph.D.