BRNO UNIVERSITY OF TECHNOLOGY
FACULTY OF ELECTRICAL ENGINEERING AND COMMUNICATION

Opponent Review of Doctoral Dissertation

Applicant: Zuzana Košelová

Title of Dissertation: Deposition of Functional Oxide Thin Films to Improve the Performance of electron Sources

Opponent: Thiago Albuquerque de Assis

Opponent's Department: Department of Physics (GFI) / Institute of Physics (EGF) – Fluminense Federal University

In accordance with the Study and Examination Rules of BUT, in his/her review the opponent will mainly comment on:
a) the topicality of the dissertation,
b) whether the dissertation achieved its given objective,
c) the problem-solving procedure and the results of the dissertation along with the concrete contribution of the doctoral student,
d) the significance for practical application or the progress in the field,
e) formal and language qualities of the dissertation,
f) whether the dissertation fulfils the conditions of Section 47 (4) of the Act,
g) whether the student proved his/her creative abilities in the given research field and whether the work does or does not comply with the standard requirements placed on the dissertations in the given field. The review is not valid without this conclusion.
It is necessary to add a concise commentary to each of the points below.

Ad a) Topicality of the dissertation

The topic of the dissertation is topical.
Comment: The topic of the dissertation, in my opinion, is timely and relevant to the field of vacuum nanoelectronics and cold field electron emission. The development of coated tungsten emitters and the investigation of dielectric and oxide films as modifiers of emission behavior remain of significant technological and scientific interest, particularly for applications in electron microscopy and high-resolution electron sources. This study addresses a problem of practical relevance, namely the assessment of how controlled oxide-layer formation influences field electron emission stability and efficiency.
From a topical standpoint, the dissertation aligns well with ongoing international efforts to improve emitter longevity and macroscopic current uniformity by introducing surface-engineering strategies. It also reflects the transition from purely metallic emitters to hybrid metal–oxide systems, a direction that has gained importance for the development of advanced field emission sources. Although the physical mechanisms underlying the influence of dielectric coatings on tunneling and emission require careful theoretical

treatment, the overall research theme is appropriate and contributes meaningfully to an active and evolving area of applied surface science and field electron emission physics.

Ad b) Objective of the dissertation

The objective of the dissertation was achieved.
Comment: The dissertation defines a clear objective: to determine how the formation of tungsten oxide layers affects the field electron emission performance of metallic tips. The work follows a coherent experimental sequence that includes oxide growth under controlled conditions, morphological and chemical surface characterization, and emission testing under high vacuum operation. The methodological progression from planar tungsten substrates to sharp emitters provides a consistent basis for linking oxide properties with emission behavior. The study shows that the oxide growth method and the resulting layer composition directly influence the emission stability and the threshold electrostatic field required for tunneling. The correlations established between oxidation conditions, surface chemistry, and emission characteristics confirm that the research objective has been achieved in my view. Electrostatic simulations included in the dissertation are used to interpret the qualitative influence of oxide thickness and surface curvature on the local electrostatic field distribution. These results motivate further theoretical analyses directed toward a more complete description of tunneling through composite metal–oxide–vacuum systems. The data generated in this study provide an empirical basis for such future developments. I believe that, within the scope of its experimental program, the dissertation fulfills its declared objective and establishes a framework that connects controlled oxide growth with the modulation of field electron emission behavior.

Ad c) Problem-solving procedure and the results of the dissertation and the concrete contribution of the doctoral student

The problem-solving procedure and the results of the dissertation are above average.
Comment: This dissertation follows a staged problem solving procedure: (i) preparation of oxide layers under controlled conditions by two routes; (ii) morphological and chemical characterization using complementary techniques; (iii) validation on sharpened emitters; and (iv) evaluation of field electron emission behavior with analysis based on established formalisms. The choice to begin with planar substrates and then transfer optimized conditions to tips creates an interesting path from process parameters to emitter performance, allowing direct attribution of observed behaviors to specific surface modifications. The results are organized to connect each methodological block with measurable outcomes: oxide-growth route and surface state are related to electron emission onset and stability; microscopy and spectroscopy inform the interpretation of emission patterns; and the adoption of orthodoxy testing provides a consistency check for the emission regime and extracted parameters. This structure links coating characteristics to emission behavior under high-vacuum operation. A central contribution is the correlation framework that maps oxide properties to emission response. By comparing coating routes and documenting their effects on surface roughness, composition, and thickness, the dissertation identifies trade-offs between lower onset fields and long-term stability, and discusses how contaminants introduced during processing can influence tunneling and durability. This correlation, built from microscopy, spectroscopy, and emission tests, constitutes a concrete outcome that can guide process selection for different operating constraints.

Another contribution is the integration of qualitative electrostatic modeling to interpret trends observed experimentally. The finite-element analysis is used to rationalize how coating-induced changes in geometry and dielectric environment affect local fields at the apex, complementing the empirical correlations without attempting to replace them. This pairing of measurement and modeling strengthens the attribution of mechanisms and indicates how surface engineering can be used to modulate emission. The candidate's concrete contributions can be summarized as: (a) establishing a reproducible workflow to grow and assess oxide layers for field emitters; (b) implementing a measurement protocol that couples microscopy, spectroscopy, and emission analysis with orthodoxy checks; (c) producing a comparative assessment of coating routes with process-property-performance links; and (d) articulating an application-oriented view in which the coating method is selected according to stability and voltage requirements. Together, these elements form a transferable methodology for surface-engineered emitters.

Some suggestions for strengthening the contribution: Standardize field-emission notation in line with current literature practice (e.g., using a local-voltage conversion length and a separate, dimensionless field-enhancement factor), so that dimensional analysis is transparent across the report and consistent with the orthodoxy assessment. This increases comparability with future studies and avoids ambiguity in parameter naming.

Overall, the problem-solving procedure is coherent, the results support the stated aims, and the doctoral student's contribution is identifiable in the design of the workflow, the execution of the multi-technique characterization and field electron emission analysis, and the construction of a correlation-based interpretation that can be reused for emitter optimization in related contexts.

Ad d) Significance for practical application or progress in the field

The significance for practical application or progress in the field is above average.
Comment: The experimental findings of this dissertation are directly connected to the optimization of field electron emission sources used in practical devices. By identifying how oxide-layer formation alters emitter stability and operational thresholds, this work provides guidance for adjusting fabrication parameters to meet performance requirements in electron microscopy and related vacuum technologies. The comparative evaluation of oxidation routes offers process-level information that can be directly applied to the preparation of robust emitters without requiring complex nanostructuring. The results also highlight a relevant trade-off between ease of emission initiation and long-term current stability, a consideration that is relevant to the design of field emitters for continuous operation. These observations advance the practical understanding of how thin dielectric layers can be used not only as protective coatings but also as active components that modulate electron tunneling behavior. For clarity in communication and future use of these results, the terminology should distinguish electrostatic field from electric field when describing the static fields responsible for emission. This refinement ensures conceptual precision and aligns the work with established conventions in field emission literature. Overall, this dissertation contributes experimental knowledge that can support incremental progress in the design and reliability of oxide-coated metallic emitters for vacuum electronic applications.

Ad e) Formal and language qualities of the dissertation

Formal and language qualities of the dissertation are above average.
Comment: The dissertation is well organized, and its overall structure follows the expected academic format, progressing logically from the theoretical background to experimental methods, results, and discussion. Figures and tables are placed close to the relevant text, which contributes to clarity. However, the formal presentation would benefit from a more consistent treatment of symbols, physical dimensions, and terminology to ensure alignment with established conventions in field emission physics.
An important point concerns the use of the symbol β. In the text, β is sometimes referred to as the field enhancement factor, which should be dimensionless, but it also appears defined as the inverse of the product KR, giving it a dimensional meaning of inverse length (if K is a dimensionless quantity). This inconsistency may lead to ambiguity when interpreting the results or comparing them with established theoretical models. To avoid this, it is recommended that γ be reserved for the dimensionless field enhancement factor, while ζ represents the local voltage-to-field conversion length, which has dimension of length. If β is maintained, its dimensional role should be explicitly defined. The inclusion of a concise list of symbols and their units at the beginning of the dissertation would resolve these ambiguities and improve technical precision.
The terminology throughout the text is generally appropriate, but a consistent distinction between electrostatic field and electric field is important. Since the study deals exclusively with stationary fields responsible for electron tunneling, electrostatic field is the correct term in this context. The use of electric field may imply time-dependent or electromagnetic effects that are outside the scope of this dissertation, in my view. Ensuring consistency in this terminology would improve conceptual accuracy and readability.
In summary, the dissertation meets the formal and linguistic standards expected for doctoral work. Minor revisions in symbol definition, field terminology, and unit consistency would enhance its precision and facilitate future reference by other researchers in the field.

It would also be advisable to refer to the semiclassical tunneling approximation by its complete form, "JWKB," which acknowledges the contribution of Harold Jeffreys alongside Wentzel, Kramers, and Brillouin. Jeffreys introduced the connection formulas that ensure the continuity of the semiclassical solution across turning points, making his contribution essential to the mathematical foundation of the method widely used in field emission theory. Adopting the term "JWKB" would therefore be both historically accurate and conceptually precise.

Ad f) The dissertation fulfils the conditions of Section 47 (4) of the Act

The dissertation fulfils the conditions of Section 47 (4)*) Act No. 111/1998 Sb. Higher Education Act: YES

((4) Studies are duly finished with a doctoral state exam and dissertation defence, which prove the ability and readiness to work independently in the field of research or development, or in theoretical and creative arts. The dissertation must comprise original and published results or results accepted for publication.*

Ad g) Creative abilities of the student in the given research field. Compliance with the standard requirements placed on the dissertations in the given field.

The doctoral student did prove his/her creative abilities in the given research field and the work does comply with the standard requirements placed on the dissertations in the given field.

Comment: The candidate demonstrates, in my view, the ability to conduct independent research through the complete cycle of experimental design, data acquisition, analysis, and interpretation. This work presents a coherent investigation into the preparation and characterization of tungsten emitters with controlled oxide layers, integrating a sequence of methods that the author developed and applied independently. This dissertation contains original results that were presented at scientific conferences and published in international peer-reviewed journals. These publications confirm both the authenticity and the dissemination of the research outcomes. The presentation of the problem, formulation of hypotheses, and execution of experiments show readiness to continue independent scientific activity within the field of vacuum nanoelectronics and surface physics.

The structure, methodology, and documented results therefore satisfy, in my opinion, the formal and substantive requirements of a doctoral dissertation. This work evidences the author's capability to perform research of a professional standard and to contribute original results to the advancement of the discipline.

Overall evaluation: This dissertation represents a consistent and technically competent study of tungsten oxide layers for cold field emission applications. It demonstrates the candidate's ability to plan and execute experiments, interpret results, and relate surface properties to emission behaviour. The main strength of this work lies in its systematic experimental program and its contribution to understanding how surface oxidation can be controlled to optimize field electron emission stability. There are, however, conceptual and terminological aspects that deserve further attention. In particular, the physical definition of certain quantities, such as the field enhancement factor and surface roughness, should be refined to conform to established conventions in field emission and surface science. For example, surface roughness is commonly defined as the standard deviation of surface heights, and represents the statistical amplitude of morphological fluctuations. In this dissertation, roughness is sometimes treated as an average height difference, which does not correspond precisely to the statistical definition. Clarifying this distinction would improve the quantitative interpretation of the AFM results and allow more direct comparison with surface evolution studies. This work could also benefit from a clearer separation between geometrical effects—such as curvature and aspect ratio—and physical effects associated with oxide composition and stoichiometry. Disentangling these contributions would strengthen the interpretation of the results and provide a pathway for predictive modelling in future work. The overall impression is that the dissertation achieves its objectives and contributes meaningful experimental knowledge to the field. The observations are robust and provide a foundation for subsequent theoretical and applied developments in the design of metal–oxide field emitters. The candidate's performance reflects readiness for independent research work.

Opponent's questions:

The following questions are intended to encourage critical discussion and to highlight aspects that could further strengthen the scientific depth and broader relevance of the dissertation:

The first question concerns the definition and quantification of surface roughness. How would the interpretation of the AFM results change if the roughness were evaluated using the statistical definition based on the standard deviation of surface heights, rather than an average height difference? Would this alter the observed correlation between surface morphology and field electron emission behavior?

A second question relates to the distinction between geometric and material influences. How can one experimentally distinguish whether variations in emission threshold and stability arise primarily from geometric changes—such as curvature and surface irregularities—or from modifications in oxide composition and electronic structure?

A third question addresses the notation and dimensional consistency of the field-related parameters. Could the author clarify the precise physical meaning of β, γ, and ζ, specify their units (or indicate if they are dimensionless), and indicate how each should be used consistently in the analysis of emission data?

A fourth question focuses on the evaluation of emission stability. Would it be possible to define a quantitative measure of stability, for example as the relative standard deviation of the emission current over a fixed time interval, and apply it systematically across samples to establish a unified comparison of performance?

A fifth question is: could the author provide additional methodological details about the electrostatic simulations performed with COMSOL, such as how the domain size was chosen to minimize artificial boundary effects, and how the boundary conditions were defined for an axisymmetric geometry? It would be important to explain how the mesh was refined near the apex and across the oxide layer, where the electrostatic fields are strongest, and how convergence and independence with respect to mesh density were verified. Since the problem is treated as axisymmetric, the two-dimensional boundaries revolve into rings in three dimensions, and the resulting electrostatic coupling effectively occurs between concentric rings rather than isolated points. A short discussion clarifying how this geometric representation influences the local field distribution and whether it may lead to overestimation or underestimation of certain effects would strengthen the interpretation of the numerical results.

Finally, the experimental findings point toward the potential of multi-layer or composite systems. How could the present approach be extended to investigate more complex oxide configurations, such as W–WOx–Au or WOx–TiO2 combinations, and what challenges in reproducibility and interface control might arise in such systems?

I ☒ recommend ☐ do not recommend the dissertation for the defence.

Date: 05.11.2025

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