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Reviewer's report on doctoral thesis

It is my privilege to provide this review report on the doctoral thesis of Mgr. Oleksandra Koloskova, entitled " *U-based thin films: electronic structure and physical properties*", which I found interesting to read.

The thesis presents results from the investigation of electronic and structural properties of the pure and hydrated U and U-T (T: Zr and Mo) thin films prepared by the sputtering technique and their correlation with the magnetic and electric properties. The motivation for studying these thin films was the properties they may demonstrate (anomalous superconductivity and exotic types of magnetism) and certain advantages before studying bulk materials, such as quick preparation, the possibility of precise control of their composition, and small amounts of used materials. The thesis consists of a short introduction followed by three main chapters (Theory, Experimental Details, and Results and Discussion), and it is finished with the Conclusions and Future Outlooks.

In the Introduction and Theory parts, the author underlines the insufficient investigation of actinide materials in general because of the rarity and radioactivity, explains that the magnetic properties of such materials relate to the occupancy and localization of 5f orbital, and informs that in the case of U-based compounds, these properties can be tuned through the interaction with different elements. It also gives an informative overview of differences between materials with the itinerant and localized electronic systems and types of magnetism originating from them, presents theoretical basics of the electronic structure, electric and magnetic properties of actinides in general, and a detailed description of the structure and properties of U compounds and their hydrides. Finally, the theoretical description of the preparation and analytical methods used (reactive DC sputtering, RGA, XPS, UPS, BIS, XRD, XRR) and the knowledge needed to process the obtained experimental data are provided here. In general, this chapter looks complete and makes a good impression on the reader.

The Experimental Details section contains a description of the laboratory equipment used. It presents detailed information about the UHV system developed at JRC Karlsruhe and used for the samples' preparation and in-situ XPS and UPS analyses and provides a description of the techniques used for the structural analysis (XRD, XRR, and TEM), and magnetization and electrical measurements. The chapter also provides some deposition parameters used during the preparation of the thin films (type of supports and its temperature during the growth, target current, H_2 pressure, time of deposition, etc.). However, the important information about the deposition rates is missing here. It is said that in some previous experiments, the deposition rate was about 1 Å/sec, but I think it is very dependent on the deposition parameters used. Providing only the deposition time without the deposition rates is just numbers that say nothing about the thickness of the films, which is essential information.

The third and most crucial chapter contains the experimental results obtained in the thesis. A detailed study of the electronic structure of the differently hydrated U layers is shown at the beginning. After that, the results obtained from studying the electronic and structural properties of the non-hydrated and hydrated U-T films are presented. The significant part of the Results and Discussion section describes the electric and magnetic properties of the layers and tries to correlate them with the electronic and structural properties. Even though this section contains a large amount of experimental data, sometimes there is a feeling of the absence of a systematic approach in the layers' characterization. Indeed, according to the motivation, one would expect always to have the characterization of the electronic structure and structural properties for each sample, then correlate it with the electric and magnetic properties. However, in the case of U-Zr(-H) layers, their electronic structure is studied in detail, while structural studies are absent. Oppositely, for U-Mo(-H) samples, a very detailed XRD and XRR characterization is performed, and no studies on the changes in the electronic structure of the layers with adding Mo are present. Also, having microscopic studies (SEM or AFM, for example) would be nice. There is only one TEM image of UH_2 film in the whole work. However, TEM studies are often crucial for supporting the diffraction results. Despite these shortcomings, the work brings new and original results of the highly unexplored electric and magnetic properties of U and U-T hydrides and might help to understand the physics of high-temperature superconductors.

A list of questions that I'd like the applicant to address during his upcoming defense:

1) Have you checked the composition of the layers deposit for 3000 or 4000s (those prepared for the ex-situ experiments)? Was it the same as in the case of 600s depositions? As you mentioned in the experimental part, the sputtering process is usually unstable, and the film stoichiometry may change during the deposition process.
2) Also, I did not get the idea of the different substrates used. If I understood correctly, the thickness of the prepared layers was above 50 nm in the case of pure UH_x films and above 100 nm for the U-T(-H) ones. Did you really expect to see some difference in the structure of such thick films prepared by sputtering at low temperatures? The roughness of the substrates might play a role. Did you check it?

3) If U-Zr alloys formed in your sputtered samples, I would expect peak shifts in the U and Zr XPS spectra. It seems that the peak positions in Zr 3p and U 4f did not change much with the increase in the dopant concentration. Can you comment on it?

4) The primary purpose of preparing the alloyed hydride films was to stabilize single-phase films, but this was not fulfilled (probably due to the high complexity and variety of processes during the reactive sputtering from two targets). What about preparing the alloyed films by sputtering and then hydrating them at high H_2 pressure? It may bring a bit more control on the film preparation.

General comment: The thesis presents original research results supported by at least 4 original publications in peer-reviewed journals. It is written well, with occasional minor grammatical mistakes and typos. There is no doubt that Mgr. Oleksandra Koloskova demonstrated that she can solve challenging scientific tasks and achieve valuable results. I consider that this work meets all requirements for a Ph.D. thesis and recommend that the candidate be awarded the doctoral degree after a successful defense.

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