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O. Koloskova, PhD. thesis, opinion of supervisor

The PhD thesis of Oleksandra Koloskova, "**U-based thin films: electronic structure and physical properties**", represents part of research results of the candidate obtained in the period 2017-2023. A complete account of her research effort, which includes studies of bulk alloyed U hydrides and several U-based ternary single crystalline materials, would be rather extended. Hence numerous results and activities remained beyond the borderline, imposed with the aim so as to have the thesis thematically focused, with its size not exceeding a usual practice.

The work presented is located at a crossroad of two fields, namely the field of solid-state properties of Uranium systems and of thin film synthesis and studies, the latter bringing a new dimension to conventional actinide science by introducing materials far from thermodynamic equilibrium. Unlike traditional bulk (poly- and single-crystalline samples), sputter deposited thin films allow much broader variations of crystal structure, affected by the composition of the working gas, deposition rate, and type and temperature of a substrate. The importance of the work accomplished is in the fact that uranium as a strongly electropositive element is very reactive, requiring extreme vacuum conditions and purification of gases used. Hence uranium thin film studies are performed only in few laboratories around the world. Such difficulties are amplified in case of working with hydrogen, which, if introduced in a UHV chamber, sticks to walls and desorbs adsorbants, which have remained even after baking. This are the reasons why no other group succeeded to prepare oxygen-free U hydrides, even if working with the same equipment, located at JRC Karlsruhe.

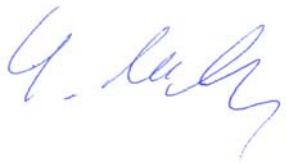
The results contribute to important issues related to interactions of various nuclear fuels with hydrogen, but the main target was understanding of U hydrides as systems with strong electron-electron correlations within the 5*f* states, the global character of which is determined by the polar bonding, tuning the 5*f*-6*d* hybridization by the involvement of the 6*d*'s in the bonding with H-1*s* states. Testing predictions of ab-initio calculations working with diverse particular assumptions requires photoelectron spectroscopy data. As general experience shows, the clean hydride surface needs in situ hydride synthesis. Cleaning of ex situ prepared bulk hydrides (very often pyrophoric) in all so far attempted cases gave severely oxidized material. From this point of view the spectroscopy results are culmination of decades of research all over the world. The spectroscopy involves not only the conventional XPS, but also more surface sensitive UPS with higher energy resolution, and even BIS, the Bremsstrahlung Isochromat Spectroscopy, which probes empty electronic states above the Fermi level.

The achievements are naturally based on many years of instrumental development at ITU Karlsruhe. O. Koloskova was able to master all important instrumental procedures needed for sputter deposition, i.e. substrate manipulations, cleaning, managing the complicated vacuum and gas delivery systems, as well as for subsequent in situ spectroscopy studies. The following ex-situ stage of research, performed in the Prague labs, included XRD characterization and studies of magnetic and transport properties. The candidate was a key person in these studies. This is visible also from the insightful text, describing the instrumentation used and individual techniques. The

language side of the thesis is very good, without ambiguous statements or excessive amount of misprints.

The main achievements comprise the finding, that the metastable *fcc* UH_2 phase occurs systematically for various dopants (Mo, Zr), and its formation, concurrent to the stable UH_3 phase is determined by complicated interplay of sputtering conditions. Seen by the spectroscopic results, both phase are very similar, reflecting the polarity of the U-H bonding, while details carried by the 5*f* states reveal fingerprints of e-e correlations, manifesting in the form of atomic multiplets arising from essentially itinerant states. This carries an important message, that the valence-band spectra should NOT be generally compared by conventional DFT of DFT+*U* calculations (even if those account for cohesion properties, phonons etc.), but one needs to utilize the more advanced Dynamical Mean Field type of theory, including the intra-atomic excitations. In another words U hydrides are not simple 5*f* band systems, brought into the ferromagnetic state by H-induced volume expansion (as believed for decades). The variation of Curie temperature between UH_2 and UH_3 can be explained by the higher depletion of the U-6*d* states in the latter case. On a more general level, the work demonstrates how powerful the in situ synthesis of films and their spectroscopy can be.

As a supervisor, I evaluate the PhD. study of O. Koloskova as very successful. The perhaps too long time it took was, besides the COVID episode closing the Karlsruhe lab for several years, an expression of the effort to conclude main topics she had opened. This manifests in number of high-quality papers in premium journals as in numbers of highly evaluated conference presentations in the field of actinides, which received several junior research prizes. She also proved herself to be able to work independently in a complex environment of a large nuclear research facility with plenty of strange rules and restrictions. Eventually her video was used to recruit new young users into the nuclear research by the Joint Research Centre of European Commission. Most importantly at this point, I am convinced that the thesis, which proves a broad scope and capabilities of independent research of the candidate, is a solid basis for the PhD degree.



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