

Referee report :

Anna Maria Adamska: "Variations of actinide magnetism in uranium-based hydrides and other selected systems"

The presented thesis of Anna Maria Adamska deals with the results of experimental studies of structural and magnetic properties of UTGe hydrides as a function of hydrogen concentration, sputter-deposited UFe_{2+x} films and magnetic properties of one Pu-U alloy.

Thesis consists of four chapters, the merits of thesis is chapter three, where the experimental results are described. Discussion is presented at the end of every sub-chapter and general conclusions are at the end of the thesis. Thesis has altogether of 132 pages including Appendix, more than 120 Figures and 97 well arranged references.

The studied problems are characterized in brief Introduction where also list of studied type of materials is mentioned together with their short characterization.

The models used to describe the behavior of light actinide systems with mostly itinerant magnetic moments are described in the first part of the Chapter one. The description of basic characteristics of band model, Stoner model and model of Pauli paramagnetism is rather detailed and well arranged. The remarks describing the Hill limit, 5f ligand hybridization, orbital moments, exchange interactions, magnetic anisotropy and ferromagnetic superconductors are briefly described. Second part of Chapter contains description of thermodynamics of hydride formation and structural, electronic and magnetic properties U-H and UTX-H. Last part of Chapter 1 is related to the physical properties of U-based compounds, U-Fe and Pu-U systems that are the subject of studies.

Chapter two contains the description of sample preparation, their hydrogenation, structural characterization and experimental technique used. I have just one remark related to the Table 2.1 (page 34) where the UFe_{2+x} amorphous samples are listed together with their characteristics. It would be better to placed this table to the part Results (page 100 and the following) or at least to put there reference. I had similar problem with samples no.1 and no.2 of UCoGe (pages 33 and 63). The number of different experimental methods used by applicant is rather high and the applicant showed his ability to perform complex study of the selected samples. In the part 2.5 Magnetic measurements you could describe how the correction to magnetic impurities had been made. To determine H positions by X-ray is not

possible. Are you planning to perform neutron diffraction studies on deuterides of selected samples? On page 40 you wrote “an alternative method had to be chosen for the determination of hydrogen positions.” What did you have in mind?

In Chapter three the experimental results are described. I highly appreciate the number of new hydrides that were prepared from the six “mother compounds” and the systematic study of their physical properties that were performed. Nevertheless I have some comments and questions.

On pages 56-58 you described the properties of UFeSi and UFeSiH_{0.3} but I did not find any remarks about this sample in the previous chapters.

As I already mentioned the Thesis contains about 120 figures, so number of figures characterizing the hydride properties is maybe too high – e.g. UCoGe hydrides. To concentrate on the basic characteristics of the newly prepared hydrides would be in some cases more useful. The reason of different way of preparation of samples no. 2 and no.1 and differences in their results could be discussed in more detail in the discussion.

Why you choose the UCoGe for high pressure measurements? To arrange sample, ruby and silicon oil into the 200 micron hole is not an easy task. How many trials did you make? I am not sure if the silicon oil is really hydrostatic till 6 GPa, my feeling is that it can be up to 2.5 GPa. You did not have the problems with the ruby X-ray diffraction as it was placed at the same place as the sample? I have one remark to Fig. 3.23. Using different scales to visualize the pressure dependence of different lattice parameters is a little confusing – e.g. the rather small pressure induced increase of lattice parameter *b* looks more important than it is. Do you suppose that this pressure induced increase is a real effect or it can be a consequence of limited pressure data preciseness and rather huge pressure decrease of *a* parameter?

I have one more general question related to the continuation of these studies. The hydrogen concentration is in the most cases remarkably lower than the theoretical concentration of two hydrogen atoms per formula unit. Are you planning to prepare some deuterides and to check their positions in the lattice – e.g. for UNiGeH₁?

It is known that the magnetic ordering temperature increases with decreasing volume in the RFe₂ compounds. It is also true for the UFe_{2+x} crystalline samples at ambient pressure. Having in mind also the sensitivity of Fe magnetism on short range order (number of nearest neighbors) it is a little surprising that a consequence of increase of Fe concentration (and

increase of the number of Fe-Fe nearest neighbors) is the increase of ordering temperature. Can you comment this?

I have a question related to the estimation of γ values for U-Pu alloys. Having in mind remarkable differences for γ values of different Pu phases I would like to know if you can estimate the reliability of using γ values of α -U to estimate of γ of U-Pu alloys with different crystallographic structure?

List of publications contains 12 already published papers in international journals, in 6 of them M.A. Adamska is a first author. Articles have mostly several coauthors that present the ability of the applicant to participate in effective collaboration.

As an evaluation I can state that the applicant succeeded to prepare six new hydrides a several RFe₂ based amorphous alloys and two U-Pu alloys, characterized them by different physical method and the obtained data analyzed using the contemporary methods.

Remarks and suggestion mentioned in this report do not decline the good quality of the thesis, the level and methods of processing are on the high level. I can state that the candidate definitely presented the ability to conduct the research in independent way and the dissertation can be accepted.

Prague, October 25, 2011

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