AGH UNIVERSITY OF SCIENCE AND TECHNOLOGY

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Referee's opinion on the PhD thesis of M.Sc. Anna Maria Adamska

entitled:

„Variation of actinide magnetism in uranium based hydrides and other selected systems"

The doctoral work of Mrs. Anna Maria Adamska was made in frames of collaboration and agreement between the Faculty of Mathematics and Physics of the Charles University in Prague and the Faculty of Physics and Applied Computer Science of AGH-University of Science and Technology in Kraków. The thesis concerns the research of structural, magnetic and electronic properties of uranium based intermetallics and their hydrides. The main goal of this work was to understand the role of uranium in formation of magnetic properties of three uranium based systems: hydrides of UTGe compounds (U = Fe, Co, Ni, Rh, Pd, Ir, Pt), UFe_{2+x} thin films and selected UPu alloys. The most interesting results are presented for the UTGe hydrides.

The work consists of 3 chapters, presenting theoretical background, description of experimental methods used in this work and the presentation of results. These chapters are followed by conclusions. In the theoretical chapter the information helpful to understand the specific magnetic properties of actinides, band magnetism, exchange interactions and characteristic features of hydrides is presented together with basic information on physical properties of the investigated systems. As concerns the presentation of the Stoner Model I would prefer to speak in terms of electron-electron exchange instead of the empirical molecular field constant. The provided information on Quantum Critical Point seems to be not necessary as it was not used in the interpretation of provided results. However the presentation of this chapter proves good and deep understanding of physics by the author, what is very important for her future scientific development.

In the second chapter the experimental techniques used in the work are described. It is worth noting here, that besides of the complicated sample preparation and hydrogenation technique the author used many research techniques – X-ray diffraction as the main tool for the sample characterization, magnetic measurements (magnetization, ac and dc susceptibility), specific heat measurements, Mössbauer spectroscopy for the hydrides with Fe and the Rutheford Backscattering Spectroscopy (RBS) to characterize properties of UFe thin films. It is worth to mention here, that the measurements were performed in diverse collaborating institutions. Besides of Prague and Kraków parent institutions, experiments were done in the Institute for Transuranium Elements in Karlsruhe, Institute of Nuclear Physics of the University Frankfurt/Main, High-field Laboratory in Dresden, Joint Laboratory for Magnetic Studies in Prague, Technology and Nuclear Institute in Sacavem, Portugal and Mössbauer Spectroscopy Division of the Pedagogical University in Kraków. The possibility to perform measurements in so many laboratories gave Mrs Adamska good opportunity to get experience in diverse measurement methods and to develop her experimental skills.

In the third, the most important chapter the results are reviewed, discussed and interpreted. The big part of analyzed results consists of XRD data and here the skillful use by the author of the fitting tools such as Rietveld method is clearly visible. Mrs. Adamska presents very systematically this big amount of experimental data. For each group of samples the presentation and discussion of obtained structural, magnetic and complementary results is very clear and consistent followed by the summary. With respect to the analyzed results and interpretations I have following remarks:

It is difficult to understand why there is so much discrepancy between the XRD spectra for $UFeGeH_{1.7}$ and $UFeGeH_{2.0}$ which in principle posses the same structure.

In the analysis of all the magnetic data the calculation of diamagnetic contribution to the susceptibility should be performed, as it can be visible in my opinion for the investigated systems.

For $UFeGeH_x$ in the analysis the plots of $1/\chi$ could be useful in the same way as presented for compounds with Co and Ni. It should be also commented why in Mössbauer effect data there is not visible Fe impurity found in magnetic measurements.

For $UCoFeGeH_x$ it is interesting, that appear two β phases with different hydrogen content and their origin in the unstable γ phase is not convincing, as it can be related rather with the microstructure and the process of hydrogen absorption in the powder grains. Very important and original are the results of high pressure measurements.

For $UNiFeGeH_x$ interesting are the results of thermal expansion and magnetic measurements and the correlation between the reduced expansion below 100 K and magnetic properties (e.g. susceptibility for $UNiGeH_1$) should be broader discussed.

As concerns fitting the susceptibility curves with Curie –Weiss Law I have impression, that the data points for fitting were not optimally selected in the temperature range for some cases, because corresponding fitted parameters are non-realistic.

There is a broad discussion concerning the influence of the hydrogen pressure on the formation of α and β hydrides and their hydrogen content, what has important influence on the magnetic properties of hydrides. Here the measurements of PCT curves would be very useful, especially for the case, where at higher pressure lower concentrations of hydrogen are obtained.

Besides of the above remarks the analysis of $UTGeH_x$ series gives many new and valuable results substantially contributing to our knowledge on the magnetism of uranium compounds.

Very interesting are results of XRD and RBS for UFe films, showing their surface structure and internal composition. As the next step the Mössbauer effect investigations should be performed. My only comment here is, that in this case writing the formula UFe_{2+x} is misleading, because x reaches 2 and even 6, what is far away from the starting Laves phase. In addition in the text the percentage notation was used, e.g. $U_{14}Fe_{86}$ – page 107.

For two compositions of U-Pu alloy (U41Pu59 and U59Pu41) the XRD magnetic and specific heat typical magnetic properties were found.

In the final "Conclusions" the author very clearly reviews the main achievements of the work. The most impressive is showing, that: only in the Zr-Be-Si type structure of $UTGeH_x$ the high hydrogen concentration is possible; the dependence of hydride volume on T element; complicated magnetic behaviour of UTGe hydrides and the conclusion that hydrogen may be used to tune the magnetic properties of UTGe systems. Very valuable was the result, that increasing amount of Fe in UFe system can substantially increase the magnetic Curie temperature.

It is worth to mention, that the results described in this PhD work has been already published in 6 scientific papers with Mrs. Adamska as first author and 2 further works has been submitted to the journals. This clearly proves high scientific level presented in her PhD work.

To conclude, I would like to stress that the presented results are new, valuable and scientifically sound. They bring important contribution to our knowledge of magnetic and structural properties of uranium compounds and their hydrides. The information concerning properties of obtained hydrides of UTGe compounds are useful for the research of new materials for application in hydrogen storage. The thesis proves, that the Mrs. Adamska is a very good physicist, especially skillful in experiment and in future can create further valuable scientific works.

The PhD work of Mgr A. M. Adamska represents high scientific level, fulfills all the formal requirements of the Czech and Polish regulations concerning the PhD theses and I recommend it for further proceeding of the doctoral procedure by the Faculty of Mathematics and Physics of the Charles University in Prague.

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