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Referee's opinion on the PhD thesis of RNDr. Petr Opletal entitled "Peculiarities of magnetism on the verge of ferromagnetic ordering"

The doctoral work of RNDr. Petr Opletal focuses on studying the phase diagrams and magnetic properties of high-quality single crystals of three compounds, UCoGa, URhGa and $UCo_{0.990}Ru_{0.010}Al$. They belong to the hexagonal ZrNiAl group of UTX compounds. Magnetism in all three compounds originates from the 5*f*-electron states of uranium. One of the main goals of this work is to investigate the pressure evolution of ferromagnetic ordering in these 5*f*-electron itinerant ferromagnets and to determine the (possible) critical points in the phase diagrams.

The PhD thesis consists of 6 chapters, presenting the theoretical background, the experimental methods, the obtained results and discussions. These chapters are followed by conclusion, biography, list of figures and tables and list of publications.

In the first chapter (the theory) the helpful information for understanding the magnetic properties of 3*d*-5*d*- and 4*f*-5*f* electron systems, band magnetism, exchange interactions, spin fluctuation, phase transition is presented. In the second chapter the experimental techniques used in the work are described. It is worth to mention here, that beside of the success of handling the complicated single-crystal growth, the author used many research techniques for the sample characterization and measurements (magnetization, specific heat, electrical resistivity). Moreover, **the measurements of magnetization, electrical resistivity and Hall effect were performed not only at ambient pressure, but also in applied hydrostatic pressure up to 9 GPa**. Some basic information related to the investigated systems is presented in chapter 3. Namely, the four different possibilities for the phase diagrams for quantum itinerant ferromagnets accordingly to the Belitz-Kirkpatrick-Vojta (BKV) theory are shown: 1) the discontinuous phase diagram with metamagnetic wings, 2) the suppression of the second-order ferromagnetic transition (and an appearance of the first-order type) as well as the appearance of the metamagnetic transition (the paramagnetic-ferromagnetic phase transition induced by applying magnetic field) at the tricritical point (TCP), 3) the suppression of ferromagnetic phase and appearance of spin-density-wave (SDW) or antiferromagnetic (AFM) phase at the quantum critical point (QCP, or quantum critical wingpoint (QCWP)) and 4) the appearance of a tail-like phase diagram accompanied by a spin glass-like behavior due to a high amount of disorder. The first possibility (1) was proposed for a clean ferromagnet (without disorder or low disorder), while the later three possibilities (2), 3) and 4)) were related to the amount of disorder in a disorder ferromagnet. Some critical values estimated for

selected compounds were also presented. **This chapter provides a good and deep understanding of physics of the phase diagram by the author, which is very important for the interpretation of the obtained results**. In chapter 4 the structure of the ZrNiAl compounds from UTX family (T - transition metal, X - p-metal) is described. The three investigated compounds presented in the PhD work have the hexagonal ZrNiAl structure type. In chapter 5, the author reviews the experimental results obtained on single crystals of three mentioned compounds. The first part (5.1) concerns to UCoGa. The measurements were performed on **two single crystals of UCoGa** prepared in different conditions and underwent the post-annealing at 800°C and 900°C. In such a way, **the author could examine the influence of single-crystal growth parameters as well as of the thermal treatment on the single-crystal quality and magnetic properties** (e.g. the magnetocrystalline anisotropy, the magnetic domain structure). *But it is not clear what was the difference in the growth parameters. The author wrote (in page 46) "The crucible was set to rotate 2.5 rpm clockwise for both single crystals". But the next sentence was "The first single crystal rotated counter-clockwise with speed 4 rpm and for second single crystal its rotation was turned off"*. The author performed investigations on different pieces taken from those single crystals. Namely, the following samples were used: three pieces of the first single crystal (SC1) without or undergoing the post-annealing (labeled as SC1A, SC1A800 SC1A900), two pieces of the second single crystal (SC2) without post-annealing labeled as SC2A and SC2C ("A" and "C" stands for the part of the ingot closer to the neck and the end). While three samples, SC1A800 SC1A900 and SC2A, show a quite similar behavior and have similar T_C value (48 K), the sample SC2C reveals different behaviors, with lower T_C values (44 K), bigger hysteresis loop, bigger coercive magnetic field, bigger residual resistivity, bigger critical exponent of the pinning constant as well as bigger relative density. It was contributed to the higher concentration of defects (worse quality of SC2C), due to a higher evaporation of gallium related to a longer time of growth in the parts further from the neck of the ingot. The surface topography of UCoGa was investigated by AFM and MFM on a small piece of single crystal cut perpendicular to the easy magnetization axis. While AFM was performed at 50 K (above T_C), MFM was carried out at different temperatures. **Especially by performing the MFM at 20 K in applied magnetic fields, the author could observe the evolution of magnetic domains**. Irrespective of the fact that UCoGa was found not to follow the Bloch domain wall model as for most bulk ferromagnetic materials, **the detail investigations and analysis of MFM data by the author give a good insight of the magnetic domain structure in UCoGa**. *But it was not clear the results were obtained on SC1 or SC2 single crystal.* From the temperature dependence of magnetization measured on the same sample, it has Curie temperature T_C = 45 K. *It means it is closer to the T_C of SC2C-the worse quality sample?* A part of SC2A800 sample was used to measure electric resistivity and magnetization at hydrostatic pressure up to 9 GPa. The low-temperature data was fitted using the relation $\rho = \rho_0 + AT^n$. The author reported a big change at 5-6 GPa, e.g. the step-like change in the derivative (dR/dT) into a broad bump (Fig. 5.11b), an abrupt change of ρ_0, A and n (Fig. 5.13a). *However, the step-like drop could be seen clearly only at ambient pressure and at 3 GPa. The broad bump was already observed at 4.2 GPa and 4.7 GPa.* **The author concluded that the phase diagram of UCoGa follows the expected discontinuous phase diagram for a metallic quantum ferromagnet with the estimated critical (temperature and pressure) value T_{TCP} = 30 K and P_{TCP} = 6 GPa.** From the magnetization curves and Arrots' plots at 2 K in 3 different pressures for UCoGa, **the author could estimate the spin-fluctuation parameters** using the Takahashi theory (given in table 5.2). The second part (5.2) presented the data obtained on a single crystal of $UCo_{0.990}Ru_{0.010}Al$, which has T_C = 16 K and shows the discontinuous phase diagram. The sing-crystal was prepared earlier, since the results of magnetic measurements in an ambient pressure were already a part of author's diploma thesis.

In this work, the author presented the results obtained in applied pressure to suppress ordering temperature of ferromagnetic transition and reach TCP, QPT and QWCP. **The author found the suppression of the ferromagnetic phase transition at T_{TCP} = 8 K and P_{TCP} = 0.2 GPa. The QCP where metamagnetic transition disappears is found at P_{QCP} = 1.6 GPa and H_{QCP} = 5 T**. The third part (5.3) concerns the results obtained on URhGa with T_C = 41 K. **The significant achievement of the author was that he managed to get the first-ever single crystal of URhGa and investigated its physical properties. Especially, the author explained the uncommon behavior of the pressure effect on the ordering temperature in this compound.** Namely, although URhGa and URhAl are isoelectronic and have similar crystal structure parameters and other physical properties, the difference in T_C value and the pressure dependence of the ordering temperature between those two compounds are attributed to the 4*p*-electron states of gallium which implies the change of the 5*f*-electrons states through hybridization and thus some changes in the electronic structure. Besides, **the author showed that the phase diagrams of UCoGa and URhGa revealed a quite different behavior. By applying pressure the ordering temperature increases in URhGa, e.g. T_C = 41 K at ambient pressure which reaches maximal value of 48 K at 6 GPa and unchanges at higher pressures (up to 9 GPa)**. **It was explained as an effect of a low itinerancy (or high localization) of 5*f*-electron states.** In the last part of chapter 5 (5.4) the critical exponents of UCoGa and URhGa is discussed. The discussion of the obtained results is presented in part 6. The author focused first on discussions of the results of UCoGa and URhGa at ambient pressure and then of the results of UCoGa, URhGa and $UCo_{0.990}Ru_{0.010}Al$ at hydrostatic pressures.

It is worth to mention that **the results obtained by the author have been published in 9 scientific papers in which he was as the first author of 4 publications. It clearly indicates the high scientific level presented in his PhD work**.

As a referee, I should also gives some comments and remarks related to the thesis-construction or chapter and section division of the PhD thesis:

1/ The main chapter with the obtained results of the PhD thesis (chapter 5) consisted of 30 pages (from page 46 to 75) divided into 4 sections. In a comparison with it, then the chapter 1 (theory) is too long with 20.5 pages (from page 5 to 25). Besides, chapter 1 is with too many subsections (the number of the main sections = 6, and the total numbers of subsections = 14). Several subsections consisted of less than 1 page (1.1.2 , 1.1.3). The section 1.5 has one page and only one subsection 1.5.1 (one section cannot consist of only one subsection). In my opinion, chapter 1 can be shorten, especially it consists of standard information in many textbooks of physics of condensed matters. Besides, sections 1.4 – 1.6 (resistance, Hall effect, heat capacity) should not belong to the "theory". Some information about resistivity, Hall effect, specific heat can be included into chapter 2, as a short theoretical-background related to the experimental methods. Chapter 2 consists of only 10 pages (from page 26 to page 35) but it is divided into too many sections (6 sections). The main section 2.3, 2.4, each consists of one paragraph with the length of 0.2-0.5 page. It should be as subtitle-lines in a big letter, but not the subsection. Chapter 3 consists of about 7 pages (from page 36 to page 43) and chapter 4 is with only 2 pages (page 44-45). There is only one section (without any subsection) in each of these chapters. Chapter 4 is too short to be as a separated chapter. Besides, the information in chapter 3 and 4 was taken from the literature, such the so-called "state of art". Thus, they should be placed ahead of the chapter of experimental methods, not after it.

2/ The main outcome of the PhD work was presented in chapter 5, i.e. as only one among 6 chapters. As one chapter, it is too long consisted of 30 pages. Besides, there is a big confusion in this chapter (at least for the referee). Why the author put URhGa seperately from UCoGa,

despite of the fact both UCoGa and URhGa are both isoelectronic to URhAl? Especially the author also said that URhGa can be easily compared to UCoGa as it has the same crystal structure and similar T_C-value. In other words, why the author placed $UCo_{0.990}Ru_{0.010}Al$ in between the two Ga-based compounds?
3/ In fact, chapter 5 can be divided into 3 different chapters, each chapter relates to one compound. Other choice is to divide the chapter 5 into 2 chapters, one chapter presented the results at ambient pressure, another one for under hydrostatic pressure. If so, it would be in a good agreement with the content in chapter 6 (i.e. Conclusion, in which the author discussed first the results of UCoGa and URhGa at ambient pressure and then of the results of UCoGa, URhGa and $UCo_{0.990}Ru_{0.010}Al$ at hydrostatic pressures).
4/ There are some errors:
Page 46, second paragraph: precursors of UGa2 (not UGe2)
Page 54 (Fig. 5.11) the values are given in R(μΩ) and dR/dT. It is the resistance. But the author discussed about the electric resistivity.
Page 55, Fig. 5.12b: UCoGa (not URhGa?)
Page 55 (Fig. 5.13), page 60 (fig. 5.19a): the resistance (R) are shown (in mΩ), but the author discussed about the electric resistivity.

As a summary, I would like to stress that the presented results are new and valuable. The significance of this PhD work is that all the results were obtained on single crystals. They bring important contribution to our knowledge of magnetic and structural properties of uranium compounds and in particular the pressure evolution of ferromagnetic ordering in these 5*f*-electron itinerant ferromagnets of the UTX compounds. They are useful for the research of new materials. The thesis proves the ability of RNDr. Petr Opletal for creative scientific work, especially in single-crystal growth and in experiments in ambient pressure as well as in applied high pressures. I would like to state here that my comments and remarks mentioned above are mostly related only to the editorial aspect of the thesis.

The PhD work of RNDr. Petr Opletal represents the high scientific level and fulfills the formal requirements of the Czech regulations concerning the PhD theses. 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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