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Annex No. 10 to the MU Directive on Habilitation Procedures and Professor Appointment Procedures

# HABILITATION THESIS REVIEWER'S REPORT

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| **Masaryk University** | |
| **Applicant** | Mgr. Jiří Chaloupka, Ph.D. |
| **Habilitation thesis** | Exotic Magnetism in Relativistic Transition Metal Compounds |
| **Reviewer** | Frésard Raymond, Professor |
| **Reviewer's home unit, institution** | Laboratoire CRISMAT-ENSICAEN, Caen (F) |

Dr. Jiři Chaloupka's habilitation thesis consists of seven chapters.

After a short introduction to transition metal compounds the steps to the construction of effective low-energy models - easier to handle by means of fully numerical approaches than the ones entailing all electronic degrees of freedom - are outlined in Chapter I, which paves the way to the Heisenberg, Kitaev, and soft spin models. This very pedagogical introduction is clearly a good starting point for an advanced lecture on modern condensed matter theory.

In chapter II, the various contributions to realistic multiband effective Hamiltonians for transition metal oxides and related compounds are presented in quite some details. One is first getting familiarized with local one-body terms (including the crystal field in a symmetrical environment, the spin-orbit coupling and small lattice distortions) in order to establish the most suitable orbital basis. It is then shown how the multiplet structure arises when taking the two-body terms into account. The magnetic moment is appropriately addressed, too. Furthermore, the intersite coupling of the orbitals within the tight-binding approximation is shown, and key examples are presented in depth. One of them is made use of to describe low energy excitations as entailed in the dynamical spin susceptibility of the simplest Mott insulator, aiming at showing that they can be captured by a simpler effective low energy Hamiltonian - here the Heisenberg model. With this as a motivation a general perturbative scheme to construct effective low energy models is explained and worked out in the one-orbital case. Besides, very importantly, difficulties arising from the smallness of the charge transfer gap to the hopping amplitude are pointed out. Unfortunately no particular example is addressed. While most arguments put forward in this chapter are well established, Dr. Chaloupka manifestly succeeded in synthetically gathering them and pedagogically present them in his thesis.

From chapter III on, selected works by Dr. Chaloupka - making use of the above – are presented. It starts with Kitaev materials, and effective models breaking the spin rotational symmetry are derived. The first one is an extended Kitaev model, the JKΓ model. It is further made more realistic by including the trigonal splitting, that itself generates further spin rotational symmetry breaking terms (the Γ' term). It results in the extended Kitaev-Heisenberg model. Even when setting Γ' to zero, the phase diagram of the JKΓ extended Kitaev-Heisenberg model is too involved to be presented here, and Dr. Chaloupka focuses on a subset of parameters residing on a unit sphere in the parameter space, along [85]. In this

remarkable publication the comparison of the Luttinger-Tisza (classical) method to the exact ground state obtained for a 24-site cluster sheds light on the influence of the parameters on the ground state. Such a comparison is especially useful as both methods have their limitations and it took certainly a large amount of work to perform it. Both methods show in particular a Kitaev spin liquid regime, that is however strongly disfavored by J and Γ.

Among the essential results presented in this thesis, the emergence of zigzag phases has been put forward on a rather firm ground, as it is common to both methods, yet in somewhat different regions of the parameter space. Furthermore, the exact ground state has been analyzed using the spin coherent method introduced by Dr. Chaloupka [81]. He goes on by presenting his publication [92] that focuses on the regime of dominant Kitaev interactions with emphasis on the zigzag phases. The computation of the RIXS response reveals a highly interesting gapped spin excitation spectrum, and it is in particular stressed that two energy scales dominate the spectra.

Soft spin systems is a topics to which Dr. Chaloupka brought important contributions, and chapter IV is devoted to them. The emphasis is put on $t_{2g}^4$ ionic configurations, aiming at understanding the properties of materials arising from $Ru^{4+}$ ions. In such 4d ions, the spin-orbit coupling may not be negligible and causes the splitting of the J = 0 ground state to the J = 1 triplet states. With this at hand, Dr. Chaloupka builds an effective singlet-triplet model, which is of the Kugel-Khomskii type, augmented by tetragonal splitting. Its impact on the in-plane and out-of-plane g-factor is calculated. With no tetragonal splitting Δ, both g-factors markedly differ from the spin only value (g = 2), especially $g_c$ that happens to be 1/2. The presented calculation shows that it takes values of Δ considerably larger than the spin-orbit coupling parameter to quench the angular momentum so as to recover the spin only value for both g-factors. In this limit, the pseudo-spin anisotropy is strongly suppressed, too. Another essential result presented in this thesis is provided by the long-ranged order entailed in the model: Triplon condensation at a quantum critical point is found! Regarding excitations, a gapless magnon mode coexists with a Higgs-like gapful mode arising from amplitude fluctuations of the triplon condensate.

At then end of this chapter the main results of his publication [101] are presented. They cover a comparison of calculated dynamical magnetic susceptibility and Raman scattering to experimental data for $Ca_2RuO_4$. The main intriguing features of the experimental spectra are shown to find natural explanations within the investigated model on the present level of calculation, and were related to the calculated magnons as well as to the Bose-Einstein condensation of the triplons.

Chapter V shortly summarizes Dr. Chaloupka's key publications, which are reproduced in Chapter VI. The thesis is summarized in Chapter VII, together with a number of propositions for future work.

Summarizing, with this habilitation thesis Dr. Jiři Chaloupka demonstrated his marvelous capability to explain the works he has undertaken, despite of their complexity. It is furthermore evident that it reiterately succeeded in bridging well established concepts to the most modern ones, leading to important and highly recognized articles published in the best journals. Reading, in this nearly flawless document, about RIXS response, spin liquid, triplon condensation, quantum critical point, Higgs modes and massive numerical work justifies stating that Dr. Jiři Chaloupka is at the forefront of condensed matter theory. For all these reasons I recommend the habilitation committee of Masaryk University to accept the thesis as a part of a successful habilitation procedure wholeheartedly.

**Reviewer's questions for the habilitation thesis defence**

i) What is exactly meant by "Dynamical Gutzwiller treatment"?

ii) Is there a symmetry between $t_{2g}^2$ and $t_{2g}^4$ configurations?

iii) How large is the Bose-Einstein condensation temperature of the triplons?

**Conclusion**

The habilitation thesis entitled *Exotic Magnetism in Relativistic Transition Metal Compounds* by Chaloupka Jiři **fulfils** requirements expected of a habilitation thesis in the field of Condensed Matter Physics.

Date: April 11th, 2021 Signature: