Review Report on PhD Thesis

Faculty: **Central European Institute of Technology**
Brno University of Technology in Brno

Academic year: **2024/2025**

Student: **Ing. Michal Horký**

Doctoral study program: **Advanced Materials and Nanosciences**

Supervisor: **Ing. Vojtěch Uhlíř, Ph.D.**

Reviewer: **dr. Karel Výborný**

PhD thesis title: Controlling the magnetic phase transition in spatially confined structures

Topicality of doctoral thesis:

transition between ferromagnetic and antiferromagnetic phase of FeRh continues to attract attention until nowadays - as explained in the report, this renders the reported research very topical

Meeting the goals set:

there were no formal goals defined to my knowledge; as explained in the report, thesis reports on successful investigation into metamagnetic properties of FeRh (and related materials) which can be understood as an achieved objective

Problem solving and dissertation results:

see the attached report

Importance for practice or development of the discipline:

this work broadens our knowledge about the effect of strain on phase transition in FeRh which is important to the field of metamagnetic materials

CEITEC
VYSOKÉ UČENÍ TECHNICKÉ V BRNĚ

Formal adjustment of the thesis and language level:

(4) The study is duly completed by a state doctoral examination and the defense of a dissertation, which proves the ability and readiness for independent activity in research or development or for independent theoretical and creative artistic activity. The dissertation must include original and published results or results accepted for publication.)

Questions and comments:

It is difficult to judge which part of the research was performed by the candidate himself as the published articles are collaborative. The candidate tried to comment on this occasionally (for example in the caption of Fig. 5.3) and I expect that large parts of work on sample fabrication and experiments themselves were carried out by him personally. My preference would have been if this were commented on in Chapter 9 alongside the list of publications. See the attached report for questions #1 to #4 and additional comments.

Conclusion:

It is safe to assume, my comment above notwithstanding, that the author of the aforementioned thesis carried out substantial number of experiments and presents them in a self-contained form which is ready for oral defense. The reviewed thesis fulfils in my opinion all requirements posed on theses intended for the award of PhD degree.

Brno, February 05, 2025

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dr. Karel Výborný

Thesis of Michal Horký, hereafter referred to as the candidate, submitted to the CEITEC (under Brno University of Technology) deals primarily with fabrication and magnetotransport measurements on FeRh thin layers and microstructures thereof. This alloy exhibits a transition from low temperature antiferromagnetic (AFM) to high temperature ferromagnetic (FM) state and along with few other metamagnetic systems (such as Mn-based alloys, also touched upon in the thesis), it has been under scrutiny for over 85 years and remains in the focus of research effort until nowadays. The candidate has contributed to this field by detailed measurements of anisotropic magnetoresistance (AMR) and magnetometry across the phase transition supported by magnetic imaging (described in Chapter 6 and partly in Chapter 5) and by advancing the original idea of FeRh/MnRh multilayer structures (or superlattices described in Chapter 7). Chapters 3 and 4 summarise the experimental methods and Chapter 2 introduces the two main materials while paying special attention to the transition between AFM and FM phases. By a safe margin, this thesis contains enough original research to qualify for the award of the PhD. title and during the defense, I would like to discuss interpretations presented therein, in particular, those of AMR in narrow channels oriented along different crystallographic directions. I suspect that the candidate lacks certain specialist knowledge about this effect which may change the interpretation in the end but this is unlikely to have an adverse impact on the defense itself. Moreover, other parts of candidate's work covered by the thesis have already been published (I mean here specifically the article Horký et al. '22, `doi: 10.1021/acsami.1c22460`) and there is thus no doubt that large part of the work has already been 'verified by others'.

Regarding the last mentioned article, I take this as a good opportunity to showcase the disturbingly high density of typos (and awkward formulations too) to be found in the thesis. On page 149, the candidate has not even managed to cite his main work properly! By mistake, the doi's of Horký et al. '22 and Arregi et al. '18 (on the next page) are identical. Regarding Ghahfarokhi et al. (of unknown year) I did not even manage to find the article — it is certainly not my duty to fish for publications of the candidate and even if I do not sense any bad intention here, I very much prefer such mistakes being corrected before the thesis is published in any form.

Turning to physics, Stoner-Wohlfarth modelling and AMR phenomenology would have deserved more attention. My first question (see the next page) about magnetoresistance in Fig. 6.2 should highlight the need for solid arguments in the interpretation of measured data. A detailed study of the anomalous Hall effect (AHE) would have been worthwhile and Sec. 6.4 is much shorter than what is desirable — leaving the question why only four temperatures were measured aside, a more detailed discussion and comparison to refs. [143] and [145] would be helpful.

Introductory chapters 1 and 2 serve their purpose but they sometimes lack the appropriate depth (Sec. 1.3, 1.4.1 or Sec. 2.2 as detailed on the next page) and sometimes describe unnecessary details (Eqs. 1.11 and 1.12 are never used in subsequent argumentation) or are superfluous completely: the spin Hall effect plays important role in spintronics nowadays but I cannot see any relation to presented measurements on FeRh or MnRh (moreover, I am not sure if the idea behind Fig. 1.8 is correct, a question that can be discussed during the defense if time permits). Effect of magnetic domains (in AFM phase, in analogy to Fig. 1.1) is not discussed at all. On the other hand, the idea of FM layer remaining at low temperatures in otherwise AFM sample should be supported by additional arguments in ideal case. The presented thesis nevertheless certainly fulfils the goal of exploring the influence of strain on the metamagnetic transition and moreover, it brings new experimental data on (Mn,Fe)Rh alloys. Direct applications (such as memory devices) are not yet likely but presented results are valuable from the point of view of fundamental research.

The presented thesis fulfils requirements under Act 111/1998 coll. (Sec. 47) as **it demonstrates the ability of the candidate to carry out scientific research** and his creative approach in this endeavour. I am happy to **recommend the thesis for defense**, questions and some additional detailed comments follow on next two pages.

Praha, Feb5 2025

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General comments

- phenomenology of AMR is richer than what Eq. 1.7 (on p. 17) assumes (see also p. 102) and on the other hand, calling MR data in Fig. 6.3 (on p. 93) anisotropic MR is unfortunate: it is simply MR from which, under some assumptions, AMR can be extracted; crystalline and non-crystalline AMR should be discussed, see for example New J. Phys. 10, 065003 (2008)

- spin flop is the key concept used to manipulate (collinear) AFM order and thus almost ubiquitous in magnetotransport on the LT phase of FeRh; yet, it is poorly explained (it would be appropriate to discuss it on the level of Stoner-Wohlfarth model) and context given on p. 24 is in my opinion insufficient. Further, explications on p. 88 below are somewhat obsolete (compare ref. [13] to Tab. I in Phys. Rev. B 97, 235111) and relation to AMR in other AFM materials could be discussed, on p. 97 the interpretation should be critically compared to literature such as Phys. Rev. Materials 4, 064403 (2020).

- anomalous Hall effect (AHE) in the FM phase is not sufficiently discussed and the possibility of AHE in the AFM phase is not excluded (Eq. 6.2 on p. 116 and in Sec. 1.4.2 is popular but not entirely correct), see the recent commentary of Stefan Blügel, for example (`doi: 10.1038/s41567-024-02750-3`)

- stylistic and formal quality of text is lower than optimal at times; apart from specific minor issues listed at the end of this report, I noticed awkward language at certain parts of the thesis (systems are featured by very low magnetic moments, p. 36 for example) or copy-pasting pieces of text on p. 109 and 110 which is inappropriate.

Particular comments

- p. 13: non-collinear AFMs are completely ignored in the discussion; on the other hand, temperature dependence of magnetisation in Fig. 1.2b deserves a more detailed explanation (critical behaviour close to T_C, low-temperature behaviour etc.)

- p. 15: direction of magnetic field for graph in Fig. 1.4b matters; the dependences were not 'derived from [10]', they were reproduced

- p. 17: anisotropic behaviour described by Eq. 1.7 is not 'ascribed to spin-orbit interaction among *s*-electrons', the spin-orbit interaction requires non-zero orbital momentum (the candidate alludes probably to the case of transition metals where the *d*-electrons feel appreciable spin-orbit interaction

- p. 23: multiple minor issues here — what is the 'so-called transition curve'? (Explain what phase diagram is meant.) What is 'martensitic phase transition'? What are those 'many criteria' to sort phase transitions? Electronic properties and band structure are not the same — so called superconducting gap is an inherently many-body feature.

- p. 32: unique properties (of well-defined thin films) should be better explained; I do not think, for example, that 'thermal conductivity' is the very reason why recording media have been developed on the basis of thin films; also I missed the point about 'different density of states' — can this be commented on during defense?

- p. 40: derivation of Bragg's law is textbook (or wikipedia) material but given Fig. 3.5 is shown, I can accept this as a nice introduction of the arguably most important characterisation technique in this thesis; nevertheless, it would then be appropriate to give more details on the Cu thin film (which I suppose was grown by the candidate himself)

- p. 50: the paragraph about RAITH apparatus goes perhaps a little too far to advertise this specific supplier (even if our experience at FZU with their electron lithographs is good)

- p. 88: classifying the antiferromagnetic spintronics as 'prospective industrial branch' is too optimistic in my opinion; the importance of 'increase of electrical resistance or characteristic frequencies' should be explained

- p. 103: could the model (considering 'two AMR values' which possibly correspond to crystalline AMR terms) successfully predict the transverse-AMR, i.e. even part of ρ_{xy})?

Questions

1. change of resistivity ρ (shown for example in Fig. 2.4) could in principle be ascribed to different electronic structure in the FM and AFM phases; however, a quick DFT calculation yields plasma frequencies ω_p which go against the trend of lower ρ in the FM phase (specifically, $\hbar\omega = 6.6$ and 1.7 eV in the FM and AFM phases, respectively. What can be inferred from this information about scattering of carriers in the two phases?

2. magnetoresistance (MR) can be related to magnetic order or to orbital effects (such as the Lorentz force); throughout Chapter 6, it is assumed that the former dominates measurements such as those shown in Fig. 6.8 which is likely correct but supporting arguments would be helpful. Comparison of longitudinal and transversal MR can be used to this end (see for example Fig. 6 in Toth et al. '10, `doi: 10.1140/epjb/e2010-00132-4`) and additional measurements for Fig. 6.2 in the thesis should clarify the situation in FeRh. Does similar situation as in 3d-alloys occur?

3. domains in AFMs are known to have a strong impact on magnetotransport, see for example Phys. Rev. Lett. 130, 036702 (2023); the presence of FM/AFM domains around the phase transition in FeRh has been demonstrated by means of various methods (by MFM in Fig. 2.3 for example) and so even at low temperature, AFM domains with various Néel vector orientations are likely to occur. What impact can possibly the presence of AFM domains have on measurements such as those reported in Fig. 6.6 of the thesis? Discussion of results from ref. [134] on p. 114 assumes a mixture of AFM and residual FM regions, could the small AMR be also plausibly explained by AFM domains only?

4. at the end of Chapter 7, switchable spintronic devices based on MnRh/FeRh superlattices (SLs) are mentioned; I undestand that $M(T)$ can be tuned by the SL design but in such a structure, the tunneling MR, for example, would not simply correspond to such total magnetisation. What are the key properties of individual layers needed for optimal functionality?

Typos and trivial mistakes

- p. 10 below: Chapter 6 focuses... paragraph ends with an apparently incomplete sentence (untypical differences — between what?)

- p. 32 below: [permanent magnets] act *to* electrons (on)

- p. 34 middle: [ref. cite semicore] is a missing reference

- p. 37: computed tomography should be computer tomography; penetration depth is large, not high

- p. 50 (and around): awkward formulations such as 'currents can exceed 20 A even 500 μm should be improved; similarly, on previous page 'no dust particles are accepted to be localized there' (or even confusion about minimum and maximum above Fig. 3.9)

- p. 93: ... attributed to the electron-magnon interaction (not representing)

- p. 102: Eq. 6.1 should contain a symbol for 'angle' (whereas it should be clearly stated what angle it is)

- p. 116: index in Eq. 6.2 should be corrected

- pp. 158, 159, 163: references 72 and 80 are incomplete, ref. 134 can at least be easily found by internet search (but lacks basic bibliographic information such as the journal and page)