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Evaluation of the PhD thesis

"Modification of quantum states in strongly correlated systems by applying uniaxial pressure"

by mgr inz. Mateusz Aleksander Gala

I was asked to provide a report on the PhD thesis by mgr inz. Mateusz Aleksander Gala. I have read his thesis with great interest, and at the start I would like to clearly express my overwhelmingly positive opinion on the content and quality of the PhD thesis. I believe that Mateusz Aleksander Gala's dissertation satisfies all the formal and content requirements.

This thesis deals with a question that is longstanding within the area of strongly correlated materials — to disentangle the coupling between lattice strain, electronic correlations, and phase transitions. A set of highly sophisticated experimental approaches centered around application of uniaxial strain were applied to a chosen strongly correlated material: magnetite. The selected model material is explored in profound detail, using very carefully controlled arsenal of measurements. The main findings of this dissertation are that uniaxial strain changes the global crystal symmetry, and modifies the charge-orbital order — however, without changing the local electronic configuration. The experimental conclusions of this dissertation rest upon remarkably firm experimental grounds, carefully verified on multiple samples.

Dissertation contents

In the following, I will briefly summarize my view of each chapter, and list a few questions following the content. The questions are marked by bullet points.

Overall, the thesis is clearly structured and well organized. It begins with an exceptionally well written introduction into magnetite. This first chapter presents an extensive survey of physics of magnetite relevant to this thesis, describing the state of this field of research through many useful figures. The topics it focuses on are the Verwey transition, and the linked charge, structure and orbital orders. Role of hydrostatic pressure is explored, together with chemical doping and stoichiometry. Specifically, there is significant motivation for studying magnetite under uniaxial strain. This initial chapter spreads over 40 pages and lists 88 literature entries. The text is impressively pedagogical and engaging. This chapter also directly states the new contributions to the field, made by this thesis: to investigate how the uniaxial compression along the cubic axes [100] and [011] influence the Verwey transition, through electronic and magnetic response. The second stated purpose of the thesis was to design, construct, and calibrate a custom-built uniaxial pressure cell.

My questions relating to this part of the thesis would be:
- Are there any alternative materials that the author thinks would make sense to explore in similar depth as he did here for magnetite? Which materials would be worthwhile?
- Where would you expect strong changes under uniaxial pressure, and where could you look for some “big questions” in condensed matter physics? What would those questions be?
- Which experimental probes under uniaxial pressure would you apply?

After this pedagogical review of magnetite, the author presents us with a chapter on experimental details about the uniaxial pressure application and sample preparation. The second chapter is a good overview, which really becomes a set of instructions, or a manual on how to use the developed uniaxial pressure cell. It is also discussed how the magnetite samples are synthesized. Everything that is described is described with a sufficient level of detail. The reader can certainly appreciate the complexity of working with uniaxial pressure. What I found particularly interesting was the discussion on calibrating the force applied to the uniaxial cell. The author clearly states that one has to measure X-ray diffraction in order to know the applied uniaxial stress. The whole chapter is illustrated very nicely, and I have no doubt that it will serve as a reference to those who will use the SOL strain cell in future. Clearly, this chapter shows that the research methodology is high quality, coherent and extremely thorough.

Here, my questions would be:
- Is hydrostatic pressure perhaps less complex than uniaxial pressure?
- In absence of XRD, what is the best way (most suited experiment) to assess the deformation that really takes place under uniaxial compression?

In Chapter 3, the reader can once again appreciate the sheer multitude of experiments, calibrations, and sample preparations that took place. A minor issue here is that the top titles in some places are overlaying on top of page numbers, because they are rather long.
The chapter describes the two main objectives that were pursued here: to determine the crystallographic and elastic response of magnetite to compression along the two previously noted cubic axes, and also to calibrate the uniaxial cell used for all these experiments.

In Chapter 4, the main subject of investigation are defects in magnetite, ones caused by stress or intrinsic ones, and their role in the Verwey transition temperature. Similar to previous chapters, a very pedagogical guide is given, which leads the reader through all the intricacies of working with uniaxial stress. I particularly appreciate thought through, insightful figures such as Fig. 4.4, which relates mosaicity of the crystal to the imaging done in the relevant experiment.

- I also found it quite intriguing that extended defect fields do not shift or broaden the Verwey transition. The provided explanation mentions decoupling of lattice disorder from the electronic order parameter. Is this something that the author would have expected before the actual experiments, or was this a surprise? If yes, why so?

A small point of confusion are the color-coded parts of text, I am not sure if they are intentional.

The experiments and instructions of Chapters 2,3 and 4 set the stage for the “real” experiments described in Chapter 5. Overall, the text until now is well-structured and clear. It is obviously intended for an experimentalist reader who plans to undertake uniaxial pressure experiments. This is currently a growing field of research in condensed matter physics. The text is full of detailed instructions and very informative. For a non-expert, it also tells a rounded story, although this story may be somewhat dense or more difficult to follow than for a more informed reader.

Chapter 5 focuses on electronic and magnetic response of magnetite to uniaxial compression. This chapter explores charge correlations, magnetic response, and local Fe coordination are influenced by uniaxial stress near the Verwey transition. Here the main result seems to be that the performed compression of magnetite does not break the local inversion symmetry of the iron-oxygen octahedra. This means that while the uniaxial pressure modifies the global crystal symmetry, it does not influence the local Fe-O geometry. Again, like in previous chapters, an impressive number of samples were studied, and we have very detailed descriptions of experimental procedures. The reader can see very clearly that the experiments were not easy. Samples often crack, or develop microcracks under strain, and the effects on the Verwey transition are not dramatic. Complications arise from inhomogeneous stress distribution within the crystal. I suppose the main lesson would be — beyond the magnetite physics — that it is indeed necessary to study multiple probes (here transport and magnetic susceptibility) in order to address a full picture of the material.

- While I found this chapter very interesting, I kept wondering whether the author had initially intended to perform additional, maybe different experiments?
- Was there an idea how to perhaps change the local Fe-O environment?
- Were there maybe some alternative hypotheses that got dismissed by the results?

Fundamental requirements and how they are satisfied

I believe that the dissertation shows a good general theoretical knowledge of the candidate. Particularly the introductory chapters seem to show a solid and coherent understanding of the strongly correlated materials field. A very thorough field review, presented at the beginning, gives an impression that the candidate not only understands the underlying physics, but is also able to convey it meaningfully and effectively.

Concerning the candidate's ability to conduct independent research, if the experiments presented in this thesis were mostly performed by the candidate, then there is no doubt of his independence. From my reading of the dissertation, he is able to interpret the research using appropriate methods.

Finally, the research presented here is original. The problem studied in this work is difficult, with many experimental complications and setbacks. Samples require preparation, they often crack, the environment is not always straightforward. The experimental work obviously required a great deal of patience. The contributed knowledge, in my understanding, lies mostly in detailed knowledge on how to perform similar experiments on magnetite, or possibly some new materials.

I understand that the presented results will be published. I did not find any reference to any first-author papers by the thesis author. In my judgment, some of these results, particularly in Chapter 5, appear significant and worthy of publishing.

Final remarks

Throughout this work and in particular in the first four chapters, the figures and illustrations are impressively well done and tell a compelling story. Overall, a remarkable level of attention is given to details. This thesis provides a carefully executed and interesting story of challenging experiments on a classical material. I believe it brings important new knowledge on how to perform carefully controlled uniaxial pressure experiments.

In my opinion the dissertation demonstrates the candidate's general theoretical knowledge in the discipline of Physical Sciences, as required under Article 187(1) of the Act of 20 July 2018 – Polish Law on Higher Education and Science and contains an original solution to the problem in the domain of physics as required under Article 187(2).

With all this in mind, I strongly recommend that the PhD thesis be accepted. I believe that the dissertation meets the statutory requirements for awarding the doctoral degree. The candidate's dissertation shows that he is able to design, perform, and understand original research in condensed matter physics. With these contributions described in this thesis, I have no hesitation to positively evaluate the presented thesis work.

Prof. Ana Akrap