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Department of Experimental Physics

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Assessment of the doctoral dissertation authored by Mateusz Aleksander Gala for the PhD program in the field of Natural Sciences, discipline Physical Sciences at Akademia Górniczo-Hutnicza, Krakow, Poland and Technische Universität Wien, Vienna, Austria

The dissertation manuscript submitted by Mateusz Aleksander Gala in view of the defence of his doctoral work is titled "Modification of quantum states in strongly correlated systems by applying uniaxial pressure". The candidate's work has been carried out in the framework of a joint doctoral program at Akademia Górniczo-Hutnicza (AGH) in Krakow, Poland and Technische Universität Wien in Vienna, Austria, under the joint supervision of Professor Wojciech Tabis and Professor Neven Barisic.

The thesis consists of six chapters, spanning over 166 pages, followed by a substantial appendix section devoted to the discussion of details of the experimental implementation and of the theoretical analysis carried out during the doctoral work. The dissertation addresses highly relevant topics concerning the study of the influence of uniaxial stresses on the electronic and magnetic properties of magnetite (Fe_3O_4). While this material, and particularly the Verwey transition, has been investigated for long time, surprisingly there is a lack of experimental results on the effects of uniaxial strain on the electronic transport and magnetic properties of the material in the proximity of the Verwey transition temperature. Applying uniaxial stresses along well-defined crystallographic directions is expected to induce a strong variation of the Verwey transition temperature and this

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investigation can potentially provide an important insight in the coupling between lattice distortions, charge ordering and magnetic properties of this material.

The thesis manuscript is opened by an introductory chapter, in which the candidate discusses the properties of correlated systems, with an emphasis on magnetite. After describing the crystallographic structure of this material, the discussion focuses particularly on the Verwey transition, which is initially described in terms of structural phase transition. Later in the chapter, Mr. Gala presents experimental and theoretical evidence which frame this process in a much more complex mechanism, in which electronic, magnetic, structural and vibrational degrees of freedom are deeply intertwined. A notable example cited in the dissertation is represented by the positive correlation between the variation of the Verwey transition temperature and Néel temperature in samples with varying stoichiometry. The candidate moves on to the discussion of the influence of hydrostatic pressure on the Verwey transition temperature, showing also preliminary results obtained in the framework of his doctoral studies. After pointing out that in the literature there are very limited examples of reports on the effects of uniaxial stresses on the electronic properties of magnetite, Mr. Gala reviews the available experimental results. When discussing the effect of anisotropic stresses, applied both via a uniaxial pressure cell or on epitaxial film, the candidate mentions that some experimental results are inconsistent with expectations. He identifies the reason for this in the complex strain field present in epitaxial films, possibly in conjunction with interactions with the magnetic substrates. In this context, a question that the candidate should address if whether it is possible to provide a more quantitative estimation of these effects, based for example on the Poisson's ratio of magnetite.

Another interesting way mentioned to apply in-plane stresses is to attach magnetite samples on top of piezo-electric actuators, which can impart stresses upon the application of bias voltage. In this context, can the candidate clarify if the imparted stresses are purely biaxial, or (due for example to asymmetry in the geometry of the PMN-PT film), the stresses are characterized by a uniaxial component?

The chapter concludes with the description of the objective of the thesis and with the outline of the remaining part of the manuscript.

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The second chapter of the dissertation describes the experimental techniques the candidate has used during his experimental work. After a brief overview of the different experimental approaches available to apply uniaxial stresses, the candidate devotes a significant attention to the design and the implementation of the uniaxial strain cell developed in the framework of the doctoral work. Particularly interesting are the sections devoted to the analytical and numerical estimation of the applied stress, based on the geometry of the strain cell. These approaches have then been validated (and proved to be not fully reliable) by an experimental procedure based on the measurement of X-ray diffraction. The reference for the calibration was a sample mounted in a commercial mechanical press, while the home-made strain cells to calibrate were operated manually, recording the lattice parameters and (for relevant crystalline directions) the angle as a function of the title angle of the strain cell screw. Surprisingly, the calibration factors that resulted from this procedure were found to differ along two crystalline directions. While I understand that, as specified in the dissertation, the way the sample is mounted might lead to a certain difference in the calibration factor, can the candidate clarify from where the significant difference of the calibration factors reported for the two considered crystalline directions come?

The chapter further discusses two methods used to grow bulk magnetite crystals and concludes with an introduction to the main characterization techniques used by the candidate over the course of his doctoral studies.

As a concluding remark about this chapter, it would be interesting to understand what were the motivations that pushed Mr Gala to design from scratch a strain cell, instead of (for example) using prototypes already available at his two home institutions or using commercially available devices.

The third chapter is devoted to the discussion of X-ray diffraction experiments performed by the candidate on single crystal magnetite under compressive uniaxial strain at the European Synchrotron Radiation Facility in Grenoble, France. After summarizing the results of these measurements in terms of dependence of the lattice parameters and lattice angle for uniaxial compression along the ⟨100⟩ and ⟨011⟩ directions, Mr Gala gives additional details concerning the calibration procedure of the home-built strain cells. By measuring

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the lattice parameters of samples oriented in the same manner in a calibrated mechanical press and comparing the dependence of these lattice parameters and the lattice angle on the screw angle, the candidate successfully performed a calibration procedure of the home-build strain cells. To obtain the calibration factor, the candidate performed a linear fit, in which the intercept was effectively the unstrained lattice parameter in the relevant direction. Has this parameter been constrained to some values known from the literature? Importantly, the stress tensor components obtained experimentally have been analysed both in the laboratory reference frame and in the sample reference frame. This allowed the candidate to realize that in the case of stresses applied along the $\langle 011 \rangle$ direction, the crystal is subjected to non-negligible shear stress, with a reduction of the symmetry from cubic to monoclinic. Additionally, the author identified in several instances ~150 MPa as the limit for a fully elastic regime. Would it be possible to attribute the non-linear strain-stress dependence noticed at this stress values also to extrinsic phenomena (for example, imperfect transfer of strain or slippage)?

Chapter 4 focuses on another structural characterization technique, referred to as dark field X-ray microscopy, which is particularly sensitive to the presence of defects or disorder in solids. Importantly, the candidate used two beam sizes in his investigation: An extended beam profile, having a real-space size comparable to that of the studied crystals (box beam in the dissertation), and a line beam, with a much smaller cross-section. The latter enables to probe thin slices of the studied crystals, potentially leading to spatially resolved structural information. The aim of this chapter is to correlate the crystalline quality (including presence of extended defects and intrinsic strain fields) of the samples grown with different techniques with the Verwey transition temperature, evaluated by susceptibility measurements. After a detailed description of the experimental technique, the manuscript shows how one can obtain a quantitative estimation of the axial strain as well as a spatial map of the sample mosaicity. Mr Gala applied this imaging techniques to two crystals of nominally stoichiometric magnetite grown with two different approaches. His results demonstrate that the moisaicity and level of strain of the two samples are different. He tried to correlate this structural information with the measured Verwey temperature, invoking different strain levels or lack of full control of the stoichiometry for

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one of the growth techniques. Additionally, dark field X-ray microscopy has been applied to crystals which have been subjected to large or very large (*i.e.*, beyond the fracture point) stresses, to understand whether these stresses leave traces in the defect field even after decompression. The data acquisition was performed in two steps. In the first one, a "box"-beam (with a spot size approximately as large as the sample) was used to obtain a global overview of the strain and mosaicity on the three examined samples. In a second experimental run, a narrower beam was used, with the goal of extracting spatially resolved data. While the former approach revealed a nice correlation between the sample "stress history" and the mosaicity maps, the latter method did not enable to extract a noticeable difference in the defect fields measured on the three investigated samples. This has been explained by noting that a large beam size makes it possible to obtain signal related to structural inhomogeneities distributed across the full crystal. Conversely, the line beam enhances the spatial resolution, at the cost of a smaller sensitivity to spatially extended defect fields. Another interesting finding concerns the axial strain extracted with the two techniques. The level of strain probed with the line beam (higher spatial resolution) appears larger than what is extracted by using the box beam with a lower spatial resolution. Should one expect that mosaicity and axial strain follow similar trend as a function of the beam size? How correlated are these quantities? Finally, the chapter concludes by demonstrating that extreme uniaxial stresses beyond the fracture points introduce clear structural defects and dislocations but, surprisingly, do not appear to influence the temperature of the Verwey transition, which is instead known to be influenced by point-like defects. Here, a natural question that can be asked is whether the candidate has some hypothesis to explain this inconsistency between the influence on the Verwey transition of defects of different types.

In chapter 5, Mr Gala summarizes his measurements of electrical resistivity, magnetic susceptibility and X-ray absorption spectroscopy performed on stoichiometric magnetite single crystal samples while applying uniaxial stresses along the [100] and [011] directions. The dependence of the electrical resistivity on the applied stress of samples compressed along the [100] directions clearly shows two regimes with two strongly different slopes. This was attributed to the onset of development of microcracks. After extracting the

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dependence of the Verwey transition temperature on the applied stress, the candidate extracted a gauge factor from the slope of this dependence by assuming it is linear. However, this assumption appears in contrast with the experimental data and inconsistent with the explanation provided in the dissertation, so it would be interesting to know whether it is really justified to presume a linear dependence of the Verwey transition temperature over the whole investigated range of pressures, considering the possible onset of microfractures on the samples. The resistivity measurements with uniaxial stresses applied along the [011] direction are much cleaner, due to a better mechanical resilience of the studied material to stresses applied along this direction. A clear linear dependence of the Verwey transition versus applied uniaxial stress is reported, with a much larger gauge factor as compared to the corresponding measurements performed with stresses applied along the [001] direction. The explanation suggested for this observation is related to the possible modification of the band structure induced by the transition to a monoclinic symmetry, which could modify the band width of the relevant electronic bands. Has this hypothesis been backed by theoretical calculations available in the literature?

In a subsequent section, magnetic susceptibility measurements were used to probe the effect of uniaxial strain on the Verwey transition temperature. Consistent with the results of resistivity measurements, also magnetic susceptibility showed that applying stresses along the [011] directions yields a stronger pressure dependence of the Verwey transition temperature. Unlike resistivity measurements, though, the Verwey transition observed with magnetic susceptibility measurements occurred over an increasingly broader temperature range with increasing uniaxial stress. To explain this observation, Mr Gala suggests that in the case of magnetic susceptibility measurements there might be an interplay between applied stress and the dynamics of the magnetic domain walls. What kind of measurements would the candidate suggest to verify this hypothesis?

The last piece of experimental work reported in the manuscript is concerned with the possible influence of uniaxial strain on the local symmetry of Fe ions. This was probed by employing X-ray absorption near edge structure measurements, centred at the K-edge of Fe. The basic idea was to study the pre-edge Fe peak, whose presence is indicative of a lack of centrosymmetricity. In the presence of uniaxial stresses, the Fe ions sitting at the centre of the octahedral unit are expected to see a more asymmetric environment, which should

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lead to an increase of the pre-edge signal. This signal is present in pristine samples due to the non-centrosymmetricity of the Fe ion included in the tetrahedral units. Within experimental resolution and the applied uniaxial stresses, the candidate could not observe any noticeable difference of the Fe pre-edge signal in the presence of in the absence of uniaxial stress. Mr Gala concludes that the (relatively) small distortion introduced by the applied uniaxial stress is unable to modify the centrosymmetric arrangement of the Fe ion in the octahedral unit. In this respect, a natural question would be whether it would be possible to observe the reduced centrosymmetricity by using other perturbations (for example, by modifying the steric hindrance in the sublattices around the octahedra, doping, etc.), possibly in combination with uniaxial pressure.

Globally, the doctoral dissertation is well-structured and written in a very clear manner. The rich amount of details makes its content accessible also to non-specialists. The extensive introduction suggests that the candidate devoted a considerable amount of time to bibliographic research. The design, development, optimization and calibration of a home-made strain cell represent highly remarkable technical achievements. The candidate has taken part in multiple measurement campaigns at large research infrastructures, which typically implies the capability of managing and carrying out highly challenging experiments in a stressful environment and robust data analysis skills. By applying uniaxial stresses, he controllably modified and investigated the delicate interplay between structural, electronic and magnetic degrees of freedom of magnetite. The doctoral work addresses a gap in the literature, in that the majority of high-pressure studies carried out on magnetite are conducted in a hydrostatic pressure environment, which is not expected to modify the symmetry of the crystal structure. In this work, instead, Mr Gala purposefully chooses to apply uniaxial stresses to break the crystal symmetry and introduce structural distortions to the magnetite crystal. The candidate is co-author of multiple scientific articles published in peer-review international journals. On the negative side, however, it should be noted that he is not the leading author in any publication.

In conclusion, in my opinion the dissertation satisfies the statutory requirements needed for the award of the PhD title and my recommendation to the Discipline Council for Physical

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Science of the AGH University and of the Technical University in Vienna is to admit the candidate to the PhD defence.

Wroclaw, 10 June 2026

Alessandro Surrente

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