Fe_3O_4 thin films: controlling and manipulating an elusive quantum material

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Fe3O4 (magnetite) is one of the most studied quantum materials. Numerous studies have been devoted to describe and to understand its enigmatic Verwey transition, also the first example of a metal-insulator transition in oxides. Yet, the underlying mechanism remains elusive. Nevertheless, the theoretically expected half-metallic behavior generates high expectations that magnetite in the thin film form can be used in spintronic devices such as spin valves, magnetic tunnel junctions, and so on. A tremendous amount of work has been devoted to preparing thin films with high crystalline quality. Using a variety of deposition methods, epitaxial growth on a number of substrates has been achieved. Yet, the physical properties of the thin films are not that well defined as those of the bulk material. In particular, the first order metal-insulator transition, known as Verwey transition, is in thin films very broad as compared to that in the bulk single crystal. The Verwey transition temperature T_V in thin films is also much lower, with reported values ranging from 100 to 120 K, while the stoichiometric bulk has T_V of 124–125 K. It is not clear why the Verwey transition in thin films is so diffuse.

In this work, we investigate systematically the effect of oxygen stoichiometry, thickness, strain, and microstructure on the Verwey transition in epitaxial Fe_3O_4 thin films on a variety of substrates. We use molecular beam epitaxy (MBE) technique under ultrahigh vacuum conditions combined with *in-situ* electron diffraction and x-ray spectroscopic characterization as well as *ex-situ* x-ray diffraction and electrical conductivity measurements. We have been able to determine the factors that affect negatively the Verwey transition in thin films [1], we have investigated in detail the growth mechanism of the interfaces and surfaces which are polar but not-metallic [2], and very recently, we have succeeded in growing magnetite thin films which not only have the Verwey transition as sharp as in the bulk, but also show transition temperatures that are substantially higher than the bulk [3]. One of the break throughs hereby is that we were able to find a class of substrate materials that is particular for magnetite. Using these tailor-made substrates and the strain exerted by these substrates, our record of the Verwey transition temperature so far, which is the world record as well, is 136.5 K, about 12 K higher than that of the bulk single crystal and about 25 K higher than that of epitaxial thin films so far published. Obviously, the occurrence of the Verwey transition in the highly anisotropic strained films has raised also a new question to the intricacies of the interplay between the charge and orbital degrees of freedom of the Fe ions in magnetite.

[1] X. H. Liu, A. D. Rata, C. F. Chang, A. C. Komarek, and L. H. Tjeng,
Verwey transition in Fe_3O_4 thin films: Influence of oxygen stoichiometry and substrate-induced microstructure,
Physical Review B 90, 125142 (2014).
[2] C. F. Chang, Z. Hu, S. Klein, X. H. Liu, R. Sutarto, A. Tanaka, J. C. Cezar, N. B. Brookes, H.-J. Lin, H. H. Hsieh, C. T. Chen, A. D. Rata, and L. H. Tjeng,
Dynamic atomic reconstruction: How Fe_3O_4 thin films evade polar catastrophe for epitaxy,
Physical Review X 6, 041011 (2016)
[3] X. H. Liu, C. F. Chang, A. D. Rata, A. C. Komarek and L. H. Tjeng,
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NPJ Quantum Materials 1, 16027 (2016)