Verwey transition on iron oxide nanocubes elucidated by electron energy loss spectroscopy and density functional theory simulations

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Keywords: DFT simulations, EELS, TEM, Verwey transition

In the last decades, the Verwey transition, i.e., the phase transition that takes place in magnetite (Fe3O4) at low temperature, has been an unsettled discussion topic in condensed matter physics[1]. At room temperature (RT) magnetite is a half-metal with a cubic structure (Fd3□m), but at temperatures around 125 K it changes to an insulator with a monoclinic phase. However, the exact crystalline arrangement of the insulating phase is still an open question, where currently the proposed candidate phases are sthe Cc, C2/c and P2/c structures[2-4].

In the present work, the structure of the low temperature magnetite for single 25 nm nanocubes has been assessed combining experimental imagining and spectroscopy approaches in the Transmission Electron Microscope (TEM) and Density Functional Theory (DFT) calculations. The experiment was carried out in a liquid nitrogen cryo-holder allowing to acquire the data both at 100K (low temperature, LT) and at RT. The DFT calculations of the iron electron energy loss spectroscopy (EELS) edge of the RT phase and the potential LT phases were performed using WIEN2k package[5-6].

For HT, both the high resolution TEM and the EELS experimental data are in good agreement with the Fd3□m structure, as expected. However, for the low T phase, the high resolution TEM images do not yield a univocal result. Interestingly, DFT calculations suggest that a difference in the Fe L3 edge energy loss near edge structure (ELNES) can be used to distinguish between the diverse candidate phases. The LT experimental EELS data will be compared with the DFT results obtained from the different monoclinic candidates to elucidate the LT phase of magnetite in these nanocubes.

References

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