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Magnetic susceptibility studies of single-crystalline zinc ferrites under pressure

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Abstract

Magnetic susceptibility measurements of magnetite and zinc ferrite ($Fe_{3-x}Zn_xO_4$, $x = 0.0174$, 0.049) single-crystal samples were performed under pressure up to 1.2 GPa in the temperature range close to the Verwey transition T_V. The application of pressure p decreases T_V linearly. The χ_{AC} signal is not affected above T_V, but rises below T_V with p; this was explained by increasing domain wall movement. Finally, the isotropy point T_{IP} increases with pressure indicating that magnetism does not actively participate in the Verwey transition.

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Keywords: Magnetite; Magnetic susceptibility under pressure; Verwey transition

1. Introduction

One of the most interesting properties of magnetite is the Verwey transition that manifests, on heating above $T_V \approx 124\,K$, in the sharp rise of conductivity by two orders of magnitude and in anomalies in many other properties, e.g. magnetic susceptibility. The temperature of the transition depends on the sample's deviance from the ideal stoichiometry [1] or doping [2], and on pressure, see e.g. [3–5]. Since no systematic studies of pressure dependence of low-doped magnetite have been reported so far and since only one report [4] concerns magnetic properties of magnetite under pressure, we therefore found it worth to study the Verwey transition of zinc ferrite samples under pressure by means of AC magnetic susceptibility.

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2. Experiment

Single-crystalline magnetite and zinc ferrite samples ($Fe_{3-x}Zn_xO_4$, $x = 0$ and 0.0174, 0.049) were grown from the melt by the cold crucible technique, annealed under CO/CO_2 gas mixture and rapidly quenched. AC susceptibility measurements were made under hydrostatic pressures up to 1.2 GPa by using a gas-pressure-operated high-pressure cell made of non-magnetic Cu–Be alloy in following experimental conditions: $77\,K < T < 130\,K$, $H_{AC} = 10^{-3}\,T$, $f = 273\,Hz$, with $\langle 1\,0\,0\rangle$ direction set parallel to the AC magnetic field. The sample was first cooled below T_V under pressure and then measured on heating.

3. Results and discussion

The in-phase component χ'_{AC} of AC susceptibility for stoichiometric magnetite is shown in Fig. 1 as an example; for the sake of clarity, only some measurement runs are presented. These data were used to calculate the Verwey transition temperature T_V (as the temperature of $d\chi_{AC}/dT$ maximum), the step in magnetic susceptibility $\Delta\chi_{AC}$ at T_V and the position T_{IP} of the isotropy point (where the first-order anisotropy constant vanishes resulting in a peak in susceptibility; see the inset of Fig. 3). The pressure dependence of some of those parameters are presented in Figs. 2 and 3.

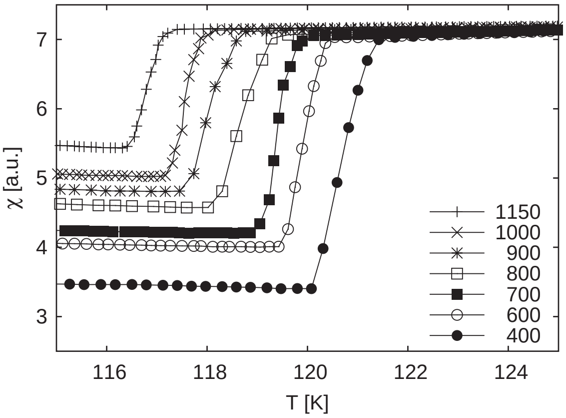

Fig. 1. Temperature dependence of $x = 0$ sample magnetic susceptibility for selected p values (given in MPa in key).

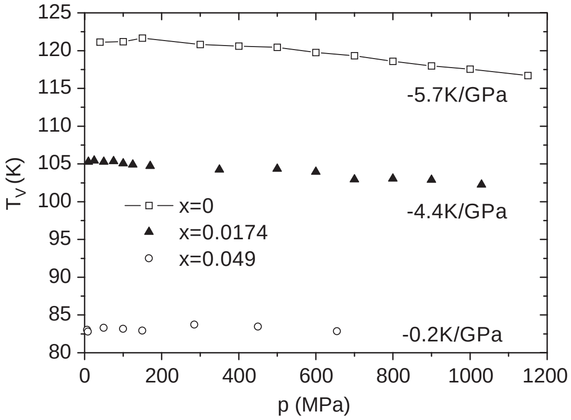

Fig. 2. Pressure dependence of Verwey transition temperature. The slopes of linear T_V vs. p relations are also displayed.

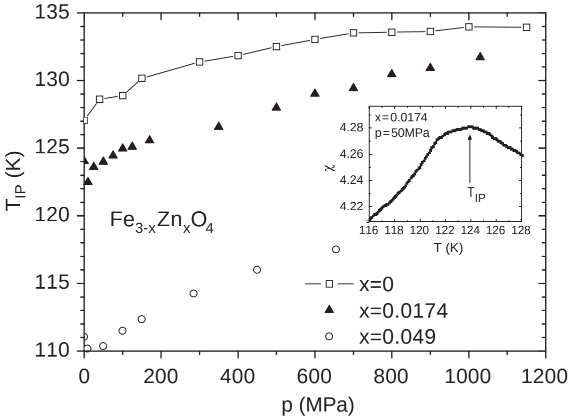

Fig. 3. Pressure dependence of isotropy point for the $x = 0$, 0.0174 and 0.049. The inset shows how T_{IP} was estimated.

The Verwey temperatures decrease with p generally in a linear manner with dT_V/dp equal to $-5.7(\pm1.2)$, -4.4 and $-0.2\,K/GPa$ for $x = 0, 0.0174$ and 0.049, respectively.

The susceptibility value is not affected by pressure above T_V but rises below T_V. As explained in detail in the forthcoming paper [6], the step in susceptibility may be due to the magnetic domain movement changing at the transition. Indeed, when magnetite is cooled below T_V, it breaks into many structural domains. Their small size limits the movement of magnetic domains, since much greater energy is needed to move the structural domain wall than the magnetic one. The application of pressure while cooling magnetite below the transition is one of the methods of eliminating structural domains. Therefore, in magnetite cooled below T_V under pressure, the average size of structural domain may be greater, and the magnetic domain wall can move more freely, resulting in higher magnetic susceptibility.

Fig. 3 shows that T_{IP} moves to higher temperatures with increasing pressure, while the opposite tendency was reported [7] for increasing Zn content in zinc ferrites. Verwey transition can finely be tuned by departure from ideal stoichiometry and/or doping, from first to second order. Rosenberg [8] found similar changeover to second-order transition with pressure as the one caused by Zn and Ti doping, or nonstoichiometry. Thus, since pressure and Zn-doping affect the Verwey transition in a similar manner, but their influence on magnetic characteristics (T_{IP}) is different, it suggests that magnetic degrees of freedom do not actively participate in the mechanism of the transition.

In conclusion, we have reported pressure dependence of the AC susceptibility for magnetite and two zinc ferrite samples. We have shown that T_V lowers with pressure, while the isotropy point T_{IP} rises. This is an indication that magnetic interactions do not drive the transition.

Acknowledgements

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