Easy axis switching in magnetite

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Abstract. The easy magnetic axis switching in magnetite is investigated. Magnetization data confirmed activation character of the process with activation energy of the same order as that of the Verwey transition, suggesting common origin. On the other hand this activation energy rises with pressure (up to 1.2GPa), unlike $T_V$. The axis switching is clearly reflected in field dependence of resistivity and the direct structural data showed that it is simultaneous with the reorganization of structure. Thus, control of the structure can be possible with the application of magnetic field, as in shape memory materials. Finally, NMR showed that all, possibly decoupled entities: lattice distortion and charge and orbital orderings, change simultaneously while the axis switching occurs.

1. Introduction

The Verwey transition -the first order phase transformation at $T_V=124$K – is mainly known due to the drop of resistance by two orders of magnitude while heating above $T_V$. The Verwey temperature $T_V$ diminishes with cation substitution, nonstoichiometry (usually caused by iron atoms deficiency), and pressure; in case of Ti, Zn and nonstoichiometry the $T_V$ vs. lattice defects relation breaks into two parts, separating first and second order Verwey transition [1].

Following the first papers [2, 3], the transition was usually believed to be related to Fe\textsuperscript{+3} and Fe\textsuperscript{+2} ions on octahedral positions in the spinel lattice, Fig. 1a, lacking its individuality at high temperature (electron is equally spread across all octahedral Fe positions above $T_V$), but maintaining it below $T_V$ (electron is frozen on particular positions at low $T$ forming well specified Fe\textsuperscript{+2} and Fe\textsuperscript{+3} cations). Thus, magnetite undergoes order-disorder electronic transition, as symbolically presented on Fig. 1b.

However, neither the exact iron of +2 and +3 valance, nor the mixed valence above $T_V$ were supported by experiments and this strict view of ionic charge order is not accepted now [4, 5]. Instead, the more relaxed charge ordering, Fe\textsuperscript{−2.4} and Fe\textsuperscript{−2.6}, on octahedral iron positions have been found [6] together with the ordered Fe octahedral orbitals [7, 8, 9]. Moreover, Resonant X Ray Diffraction (RXD) technique allowed to find that both these types of orderings are decoupled [10, 11]. Finally, it
became clear that the transition is due to intertwined electron-electron strong correlations as well as strong electron-lattice confinement [12].

**Figure 1.** a) Tetrahedral and octahedral sites in the structure of magnetite and their formal valances at room temperature, b) Tetrahedra formed by octahedrally coordinated Fe with formal valence of Fe$^{2.5}$ above $T_V$ and with +2 and +3 valences ordering suggested originally by Verwey (and not realized in reality).

The Verwey transition is reflected in virtually all physical characteristics including the abrupt change of lattice symmetry from high $T$ cubic $Fd\overline{3}m$ to low $T$ monoclinic [13, 6], as shown schematically on Fig. 2. There is also the accompanying change of magnetic anisotropy primary axes: while $<111>$, $<110>$ and $<100>$ are easy, intermediate and hard direction above 130K, monoclinic $c$ axis becomes the easy axis below $T_V$ [14, 15], Fig. 3.

**Figure 2.** Schematic view of the changes of magnetite unit cell from cubic (black) to monoclinic $Cc$ (red) once the sample is cooled below $T_V$. 
Since magnetite structure changes at $T_V$ from high temperature cubic $Fd\bar{3}m$ to monoclinic $Cc$, each of the high temperature cubic $<100>$ directions may become the low temperature monoclinic $c$-axis. As a result, the material breaks into several structural domains unless the external magnetic field $B>0.2T$ along particular $<001>$ is applied while cooling through the transition. This particular direction will then become both the unique $c$-axis and also the magnetically easy axis (Fig. 3).

![Figure 3.](image)

**Figure 3.** a) Magnetic easy (green), intermediate (blue) and hard (red) directions of cubic magnetite phase. Once magnetic field is applied along, e.g. [001], while cooling below $T_V$, the cell is doubled in this direction which also becomes the easy magnetic axis in low $T$ phase, b) Zero-field cooling results in structural domains with $c$ doubled along three original $<001>$ directions.

If now the sample is magnetized along other $<100>$ direction below $T_V$ (and higher than 50K), as in Fig. 4 (e.g. at $T=70K$), a reorientation of magnetic moments, i.e. axis switching (AS), may take place and this $<100>$ direction becomes a new easy axis [16, 17]. The phenomenon was first noticed in magnetization measurements by Calhoun and his analysis showed its activation character with the characteristic activation energy of 363K, i.e. of the same order of magnitude as the energy relevant to the Verwey transition.

The immediate question then arises, if AS concerns only magnetic axes switching or magnetic axes are moved simultaneously with crystal axes, especially with monoclinic $c$ axis. If this were the case, one might expect that the magnetic field can trigger some electronic processes similar to those that occur at the Verwey transition. We would then have the unique chance to manipulate with Verwey transition-like processes, but at lower temperatures and with magnetic field. The purpose of the present paper is to elaborate on the axis switching phenomenon taking into account that it might be also linked to the mechanism of Verwey transition.

Magnetite is a very special material, where all important interactions (electron- electron, electron-phonon), vital in solid state physics, in particular, in oxides, are represented and the structure is relatively simple (just two kinds of atoms); for these reasons magnetite started to be considered as a test ground for experimental and theoretical techniques. It is, thus, important to realize that although the axis switching is a particular phenomenon, possibly linked to the phase transition in a particular material, the problem is universal for condense matter science, specifically to the physics of oxides. Since very small doping or nonstoichiometry levels greatly affect the Verwey transition it is obvious
that proper sample preparation procedures are crucial for the reliability of the experimental results. All the data presented below were obtained on single crystalline samples grown at Purdue University by the skull melter, crucibleless technique [18]. This technique allows for the control of the oxygen partial pressure during growth, thereby ensuring that the melt remains within the stability range of the material. After preparation the crystals were subjected to subsolidus annealing under CO/CO2 gas mixtures to establish the appropriate metal/oxygen ratio [19].

In the paper, magnetic studies of AS are presented first, also under pressure. In the next section the other global characteristic, electrical resistivity, will be shown to reflect the changing axis. We will then prove by the direct experiment that, indeed, changing of magnetic easy axis is similar to the change of c axis. Finally, the introductory data concerning microscopic observation of AS (by NMR) will be presented.

2. Magnetic observation of AS

As already mentioned above, when magnetite single crystal is field cooled below $T_V$ with a magnetic field along one of cubic $a$ axes (as [001] in Fig. 3), this particular axis becomes the $c$ axis of monoclinic low temperature $Cc$ structure and, simultaneously, magnetic easy axis. This can easily be verified by magnetic measurements in this direction (Fig. 4a, hollow triangles): the initial slope is highest possible and the saturation is reached easily [20]. The measurements along perpendicular direction, an unspecified magnetic axis for low $T$ phase ([100] cubic direction), at $T=4K$ and 55K on Fig. 4a result in $M(B)$ curve that does not show saturation and the slope is much lower. At $T$ close to $T_V$ (e.g. bulk triangles, $T=70K$, on Fig. 4) application of field causes axis switching, easily seen as the sudden change of a slope, after which the signal finally reaches the saturation, while the field lowering results in $M(B)$ curve identical to that in an easy direction.

A sudden change of slope may be used for switching field $B_{sw}$ estimation (in fact, this is the external switching field $B_{sw,ext}$, where demagnetizing field is included; note that in all figures and relations $B_{sw}$ after correcting for demagnetizing field is presented), although the process is time-dependant and $B_{sw}$ estimated this way is not precise (in this type of activation processes external parameter that finally triggers the process to occur is not a well defined parameter at all). Nevertheless, we have measured several samples and at different temperatures, and it was found that with raising temperature the field at which the axis switches gets lower, as shown on Fig.4b, in accord with Calhoun’s description of the process. The probability of the magnetic-field -induced electronic transfer to change the axis reads [16]:

$$P(B_i) = \exp(-U/kT)(1 - \exp(-\mu_B B_i/kT)) \equiv (\mu_B B_i/kT)\exp(-U/kT)$$

(1)

where $U$ is the activation energy of the electron to be able to jump to the other position without magnetic field and $\mu_B B_i$ is the energy excess of unfavorable magnetic axis position in the internal switching field $B_i$ ($\mu_B$ stands for Bohr magneton). To start the cascade of experimentally observable electronic jumps (this parameter is very ill-defined and this is a reason why $B_{sw}$ is difficult to measure and use), the probability (1) has to achieve the critical value $p_c$

$$\mu_B B_i/kT)\exp(-U/kT) = p_c$$

(2)

Thus, the magnetic field required to switch the axis fulfills the relation $B_{sw} = B_i = p_c T_V e^{U/kT}$, shown, in other parameterization on Fig.4b. Note that activation energy for this particular sample is ca 350K; for other samples, we found $U$ in the range of 290-300K.

Apparently, the observation of the axis switching by the magnetization method is very easy and efficient. This was the reason why this method was applied to observe the pressure dependence on AS.

Pressure affects the Verwey transition in an complicated way, forcing $T_V$, at ca 8GPa down to zero, i.e. exhibiting quantum phase transition [21]. It was also reported that $T_V(p)$ resembles $T_V$ vs. doping relation [22], in particular the transition changes its character from discontinuous to continuous.
Finally, the conjecture was made [23, 24] that the crossover from the normal to inverse spinel takes place in magnetite under pressure; this was, however, refuted based both on Mossbauer [25], and structural [26] studies, and recent XMCD measurements [27]. In any case, the effect of pressure is important and should be measured for the phenomenon like axis switching.

Figure 4. a) In $M(B)$ isotherms along [100] measured after field cooling in $B = 1$T along [001] across $T_c$ (easy axis establishing). As long as the axis switching does not occur, $M(B)$ is reversible and typical for an unspecified magnetic direction ($T_c = 4, 55$K). The arrow shows the onset of axis switching and defines $B_{sw}$; for still higher field and with field lowering, the data reproduce an easy direction (black triangles), proving the permanent character of AS, b) Temperature dependence of switching field, supporting the relation from (2), i.e. Calhoun’s [16] prediction.

We have measured $p$ dependence of axis switching the same way as above, by performing magnetization measurements under pressure below 1.2GPa in the pressure cell described elsewhere [28]. Verwey temperature was measured in magnetic moment vs. temperature ramp under remanence field of electromagnet (the transition is then marked as a sudden change of signal) and is presented on Fig.5 on top of other literature results.

Figure 5. Pressure dependence of the Verwey transition temperature in a low $p$ region studied here.

The representative $M(B_{ef})$ curve (for $p = 0.35$GPa) is shown on Fig.6, together with switching fields, for this pressure value, at different temperatures. Although the determination of $B_{sw}$ is not precise, as
was already mentioned above, we have estimated the $B_{sw}$ for four values of $p$, at different temperatures and the result, for $p=0.37\text{GPa}$, is presented on Fig.6b.

**Figure 6.** a) Results of magnetization measurements of stoichiometric magnetite under hydrostatic pressure of 0.37GPa; the axis switching is seen as a sudden change of $M$ vs. $B_{eff}$ relation, b) typical switching field $B_{sw}$ vs. inverse temperature relation.

Pressure dependence of switching field at 60K and activation energy calculated from linear log($B_{sw}/T$) vs 1000/$T$ relations are stored in Fig.7. We have found that $B_{sw}$ lowers with $p$, while the activation energy increases. It is thus easier to change the axis at higher $p$, but the sample is less susceptible to temperature.

**Figure 7.** Results from the switching field $B_{sw}$ at 60K: a) and activation energy $U$, b) estimation for all studied pressure values of $p=0, 0.37, 0.75$ and $1.2 \text{GPa}$.

Also, the effect of hydrostatic pressure is different than that of chemical pressure, as for nonstoichiometric magnetite [32] (Fig.8): there, both activation energy and the switching fields increase with nonstoichiometry parameter that mimics external pressure.

Since also Zn substitution (that expands the lattice) does exactly the same, we may say that both substitution and doping creates much more complicated disturbance to the lattice than common relation of $T_v$ vs. pressure and disturbance would suggest.
3. Impact of axis switching on electrical resistivity

One of the most spectacular manifestations of the Verwey transition is the 2 orders of magnitude jump of the resistance. Some change of transport properties should thus be observed during the axis switching, if AS were linked to the Verwey transition, as suggested before. To check it, magnetic field dependence of electrical resistance while the AS occurs were measured on PPMS system, equipped with horizontal rotator option, with the experimental layout presented on Fig.9. Two sets of contacts, along [010], and along [100], were attached to the plate-shaped sample. The sample was first field-cooled to the specified temperature below $T_V$ in an external magnetic field $B_{ext}=1T$ set parallel to [100]. The subsequent measurements with $B_{meas}(=0→4T→-4T→0)$ along cooling direction reflect magnetic field dependence of resistivity without axis switching. The axis should be switched in the next experiment, with $B_{meas}$ perpendicular to the cooling field $B_{ext}$. Similar experiments were also performed after field cooling along [010]. In any case, the magnetic domain structure is affected, so the experiment should also show magnetic domain structure impact on resistivity.

Figure 8. Temperature dependence of switching field $B_{sw}$ in Zn-doped, stoichiometric and nonstoichiometric magnetite under ambient and increased pressure. Note that the effect of chemical substitution (or lattice disturbance, as with nonstoichiometry) is much higher, and different, than that caused by the pressure.

Figure 9. Experimental layout for checking AS impact on electrical transport. The sample (triangle shaped with two pars of contacts upper right corner) is field cooled (in external field $B_{ext}=1T$, along [100]) below $T_V$ and measured in field $B_{meas}$ aligned in the same direction (no AS). Then, the resistance is measured with field along [010], forcing AS. The sample is then heated, field cooled again, but with $B_{ext}$ pointing along [010] and the whole measuring procedure is repeated. Black arrow shows magnetic easy axis direction.
Fig. 10a presents the results of measurements after the sample was FC along [100] direction and the field was subsequently applied along this line. No AS should occur in this geometry and the curve should reflect magnetoresistance and a possible magnetic domains influence on resistance. Although the resistance rises initially with field, it results from still unset temperature (and due to the fact that resistance of magnetite is extremely sensitive to temperature change below $T_V$). The linear $R$ vs. $B$ decrease will be addressed later on.

The representative result of the experiment when the sample was magnetized in the direction perpendicular to field cooling axis, causing AS to occur, is shown on Fig. 10b. Here, the rapid changes (shown in the expanded form on Fig. 10c) of much higher amplitude than any other effects observed previously, mark the switching field; note that all figures are presented in the same scale. Similar effects, in the form of sharp spikes, were observed in the symmetrical experiment mentioned on Fig. 9 and also after zero field cooling when the field was applied in any of cubic <100> directions. However, neither these spikes are identical for both directions, nor in any other resistivity feature any clear systematics can be observed. For example, the rapid maximum in $R$ along [100] could be found with simultaneous drop of $R$ along [010] (Fig. 10c) and this results could be completely different in subsequent measurements even for the same conditions.

As already mentioned, for all the experiments, the negative magnetoresistance can be observed. To see if this is a real effect, we have estimated the resistance changes in case the changing sample temperature (which was monitored for each field) was taken into consideration. As we have reported separately [33], the effect can entirely be explained by the $T$ flow with the magnetic field: magnetite does not exhibit any substantial magnetoresistance at $T<T_V$, contrary to literature reports [34, 35, 36].

Summarizing electrical resistivity measurements, the axis switching phenomenon can be observed in electrical transport. On the other hand, no clear systematics, whatsoever, as to $R$ dependence on magnetic field along, or perpendicular to the current flow direction, or the magnetic domain structure.
could be found. It means, most probably, that all parameters we control, i.e. c-axis order and magnetic order, are not the most important parameters that affect the electrical resistance.

4. Structural Studies

In all the experiments presented above we have observed the switching of magnetic axis and confirmed its activation character; however, no insight in the mechanism of this phenomenon was proposed. In particular, it was not discerned if AS is solely a switching of magnetic easy axis, without ion displacement, or by changing the axis we actually change the charge arrangement, i.e. redirect monoclinic c axis. Below, we are showing that this last situation occurs: the application of magnetic field changes the ionic order established on cooling the sample below $T_V$, i.e. some new superstructure reflections appear due to the switched monoclinic c axis.

We have mounted stoichiometric magnetite single crystal on the four-circle KM4 diffractometer with graphite-monochromatized Mo Kα radiation, observing (4 4 1/2) and (7/2 0 4) -type reflections (in cubic notation), caused by doubling of cubic lattice constant in monoclinic c direction. When the sample was zero-field cooled below $T_V$ (we used Oxford Cryosystems nitrogen gas cooling), the domains with three c axes were formed, and all six superstructure reflections were observed, as on Fig. 11a. Application of a magnetic field (supplied by specially prepared NdFeB magnet system) along $[001]$ resulted in only one peak for each type (Fig.11b), proving doubling of lattice parameter in this particular direction. Thus, external magnetic field not only changes magnetic axes, but also affects crystal structure by changing the direction of monoclinic c axis.

![Figure 11a](image1.png)

**Figure 11a.** The $\omega$ scan of (4 4 1/2) and (-4 0 7/2) type reflections after ZFC the sample to 100K. Note similar intensity within two groups of reflections, proving similar abundance of structural twins

![Figure 11b](image2.png)

**Figure 11b.** With the application of ca. 0.2T along [001] twins with c along [100] and [010] greatly diminish, leaving only [001] twin identified by (4 4 1/2) and (-4 0 7/2) superstructure peaks.

Qualitatively the same results were obtained when B was applied along [010] (the third direction was not available do to goniometer vs. magnet geometry). We have thus proved by direct method that magnetic and structural monoclinic axes are switched simultaneously. In other words, we could, to some extend, manipulate the crystal structure with a magnetic field.

Similar conclusion that structure can be modified by magnetic field was presented in [37]. However, the structural effects described there originated from the subtle interplay between frustrated FM and AFM exchange interactions, unbalanced by an applied magnetic field. These processes are not present in our situation due to the large difference between $T_N$=858K and $T_V$. 

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5. NMR observation
After we have shown that the magnetic axis switching is simultaneous with the change of monoclinic c axis, it is interesting to check if this change of c axis is linked to the switching of charge and orbital ordering; as mentioned above, three Verwey transitions are present in magnetite, with lattice partially decoupled from orbital and charge orderings. The Resonant X ray scattering studies of this effect, allowing the observation of each type of ordering separately, will be published elsewhere; here, we will focus on microscopic observation of the process by Nuclear Magnetic Resonance.

The axis switching was not studied by NMR method before; the former NMR measurements in external magnetic field were performed at temperatures too low for this phenomenon to occur. On the other hand, NMR spectroscopy can easily see all different iron positions (both tetrahedral and octahedral) in monoclinic \( Cc \) structure [38, 39], allowing, potentially, a precise observation of all different ordering entities. The results of extended studies of AS by this technique are described in the separate paper [40]; here, only the most important results are summarized.

The sample was the sphere of stoichiometric magnetite oriented with [001] axis vertical, parallel to the RF coil axis. External magnetic field, up to 1T, could be set horizontally and the sample could be rotated to allow all the directions in [001] zone to be checked (see Fig.12). Only tetrahedral lines were checked what amounts to A iron nucleus being a probe for its nearest neighbors (B atoms) arrangement.

We have first field cooled the sample to 20K, which sets the c axis in the field direction (say [100]), establishing some atomic arrangement. Then the field of 0.3T was applied along [100] and the corresponding NMR signal for all A lines is shown on Fig. 13a. The field was then rotated such that it points in other cubic direction (Fig.13b); since the field was small enough not to force AS, the NMR pattern reflects the different hyperfine field but with the same atomic arrangement as before. Axis switching was subsequently forced (with the field B=1.3T) after heating the sample to \( T=80K \) just below \( T_V \) and the sample was measured again in B=0.3T and at T=20K, revealing the NMR pattern (Fig. 13c) almost identical to the original one. It thus looks as if all octahedral surroundings of the A atom we observe are, in relation to the field B direction, the same as before. This suggests that not only lattice distortion, but also orbital and charge orders, were changed simultaneously after the application of external magnetic field.

Finally, when the magnetic field axis was reverted to its initial direction and the whole procedure was repeated, the pattern almost came back to the one found before (Fig 13d-f).

Cooling the sample in 1.3 T through the Verwey transition down to 57 K (the field was along [100] axis) and applying 0.5T field along [010] direction enabled the observation of axis switching dynamics (only lines \( A_3-A_7 \) were measured in order to obtain reasonable signal/noise ratio). It is apparent (see Fig. 14) that the electronic system evolved from that on Fig. 13b (the external field was slightly higher) to the one on Fig. 13c within approximately 20 hours.
Figure 13. Selected tetrahedral sites spectra revealing axis switching. Figures 13a, 13c and 13e correspond to external magnetic field (0.3T, T=20K) set along the easy axis (however c and d show the pattern after axis switching) while in 13b, 13d and 13f the external field is perpendicular to the easy axis. Between b and c, and d and e, the sample was heated up to 80K and the field of 1.3T was applied to switch the easy axis. Scheme on right side describes the steps of the experiment.

Figure 14. Axis switching dynamics observed by NMR (T=57K, B=0.5T). See text for details.

We hope that similar procedure, when analyzed together with the spectra for different field direction in (001) plane may result in finding those octahedral positions that had to change most to reorganize charge, orbital and lattice distortion arrangement, as observed experimentally.

6. Conclusions
We have studied the axis switching process with various techniques, both bulk and microscopic. By the magnetization measurements, we have confirmed the activation character of this process. Since the activation energy was similar to Verwey temperature itself, it appeared that this phenomenon may be closely linked to the electronic processes active also in the Verwey transition. However, both magnetization studies in nonstoichiometric and Zn doped magnetite, as well as magnetization measurements under hydrostatic pressure have shown that activation energy increases rather than lowers with $T_V$, rendering the simple proof of a close link of AS with the transition questionable. On the other hand, AS is very well reflected by electric transport, where the strong oscillations are found at the magnetic field that switches the axis. Direct measurements of the superstructure reflections due to lattice constant doubling below $T_V$ proved that AS is simultaneous with the partial lattice reorganization, especially the change of c monoclinic lattice axis. Finally, NMR proved that this reorganization is complete, i.e. tetrahedral iron surroundings with magnetic field along easy axis is identical to that after the axis has been switched.
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