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Magnetoresistance in magnetite: Switching of the magnetic easy axis

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ABSTRACT

The influence of the external magnetic field B ($B \leq 4$ T) on resistivity in magnetite single crystal was studied at few temperatures both below and above the Verwey transition temperature T_V, and in two ⟨1 0 0⟩ type cubic directions. We have succeeded to confirm our predictions that the magnetic axis switching affects electronic transport. It was also found that the transverse resistivity ($\boldsymbol{B} \perp$ current direction) exceeds longitudinal one, but only for $T > T_V$, with no obvious systematics below the transition temperature. This may indicate that atomic disorder, inherent and poorly defined in a material with structural domains, is the primary factor that governs transport properties below the transition temperature. Finally, and concomitant with the last statement, only very small magnetic field dependence in magnetite below T_V may be inferred from our data.

1. Introduction

At the temperature T_V of Verwey transition in magnetite, the crystal structure changes from high temperature cubic to monoclinic [1]. Since each cubic [0 0 1] axis can become the monoclinic c axis (doubled, in comparison to cubic), the sample breaks into several structural domains when freely cooled across the transition [2,3]. In contrast, the application of magnetic field along cubic [0 0 1] (easy magnetic direction both above and below T_V) on cooling prevents most of domains to form and this particular axis becomes c axis below T_V. Also, when external magnetic field is applied below T_V (down to ca. 55 K) along other cubic ⟨0 0 1⟩ directions, the easy axis and c monoclinic direction may switch to this direction. That this magnetic easy axis switching (AS), the effect known already since Calhoun paper in 1953 [2], is accompanied also by the switching of c monoclinic axis, was the outcome of our previous paper [3]. Here we show the preliminary measurements aimed to study how the axis switching is seen in resistivity. Although several magnetoresistance studies in magnetite were described (see, e.g. [4–7]), none of them directly addressed the problem of axis switching. The primary result of our studies is that the electronic transport at low temperatures is affected by the structural disorder rather than magnetic, what is reflected in highly irreproducible resistivity results in a magnetically oriented sample. In contrast, the transverse resistivity (i.e., when B is perpendicular to the current direction) is lower than the longitudinal one above the transition temperature. As a by-product of our studies, we suggest that already very small shift of sample temperature, uncontrolled, but, unfortunately, common in automated measuring systems, may create false results of magnetoresistance of magnetite at low T.

2. Experimental

AC electrical resistance measurements under external magnetic field were performed on PPMS apparatus on magnetite single crystal (triangle-like slice with exposed (1 0 0) type plane; T_V = 123.7 K), skull melter grown from 99.99% pure Fe_2O_3 and annealed for stoichiometry [9].

The experimental arrangement for resistivity studies, performed on PPMS horizontal rotator option, with Cernox thermometer placed ca. 2 mm from the sample is shown schematically on the left inset of Fig. 1. The electrical resistance was measured successively, after magnetic field stabilization ($B < 4$ T), with two set of contacts (i.e., current direction) set parallel to [1 0 0] (R[1 0 0]) and [0 1 0] (R[0 1 0]) directions. The field cooling procedure is shown in Fig. 1: first, the sample was field cooled (1 T along [1 0 0], FC[1 0 0]) from 150 K down to the specified temperatures T_0 (T_0 = 145, 140, 125, 115, 105, 100, 90, 80 K), establishing both magnetic easy axis as well as c monoclinic axis in this particular direction. Then, after 2000 s temperature stabilization, magnetic field was set (according to the scheme: from 0 T → 4 T → − 4 T → 4 T → 0 T) and the resistance was measured along the field direction (i.e. along an easy axis, B[1 0 0]) and, subsequently, along the direction perpendicular to that, B[0 1 0]. In case T_0 is not very low, this final measurement should result in a magnetic and c axis switching [3]. The sample was then heated to 150 K and the whole procedure was repeated starting from field cooling with B along [0 1 0] down to T_0. Owing to sample's platelet-like shape, we consider external magnetic field B very close to the internal field.

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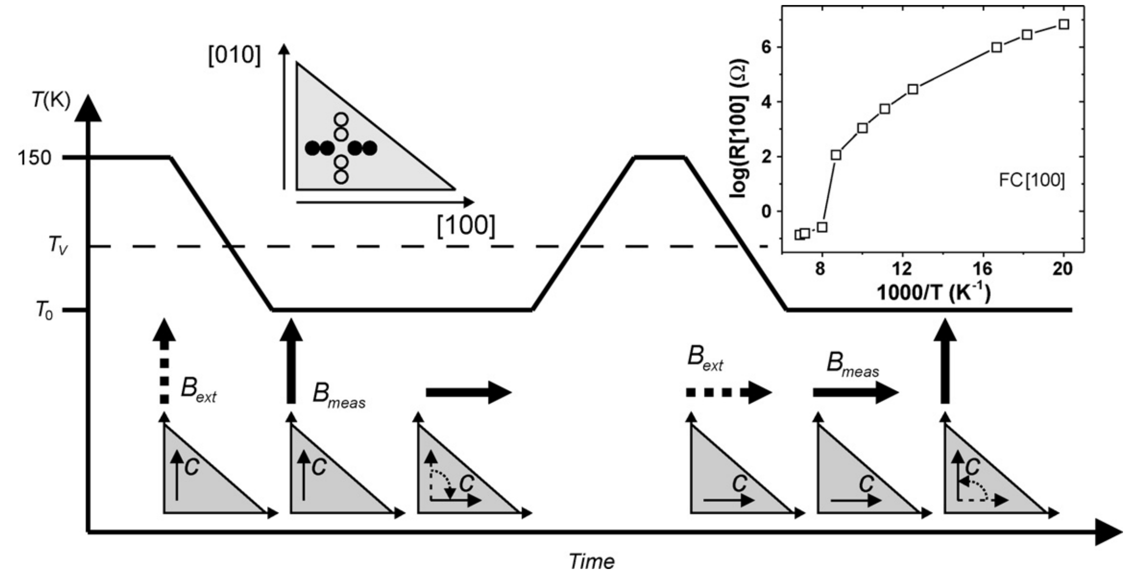

Fig. 1. Experiment sequence for each T_0. The shape of the sample as well as the contact geometry is shown. B_{ext} is the cooling field, B_{meas} is the measuring field. In the inset, rapid, 100 times R increase on cooling for our sample is presented.

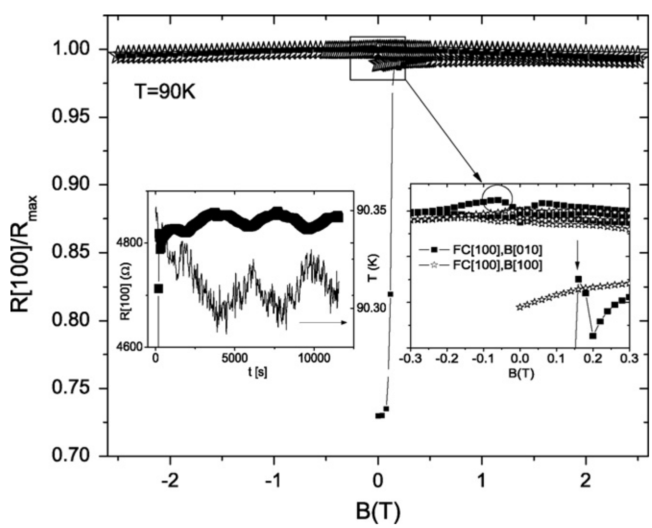

Fig. 2. Normalized resistance R/R_{max} vs. magnetic field B loop for $T = 90$ K after FC[1 0 0] and subsequent B[1 0 0] (stars) and B[0 1 0] (bold squares) measurements: the drastic difference between these curves is caused by AS. The marked region is enlarged in the right inset: here the arrow points to the value of the switching field and the circle shows the resistance influence on magnetic field. The left inset correlates the R[1 0 0] (bold symbols) with the sample's temperature.

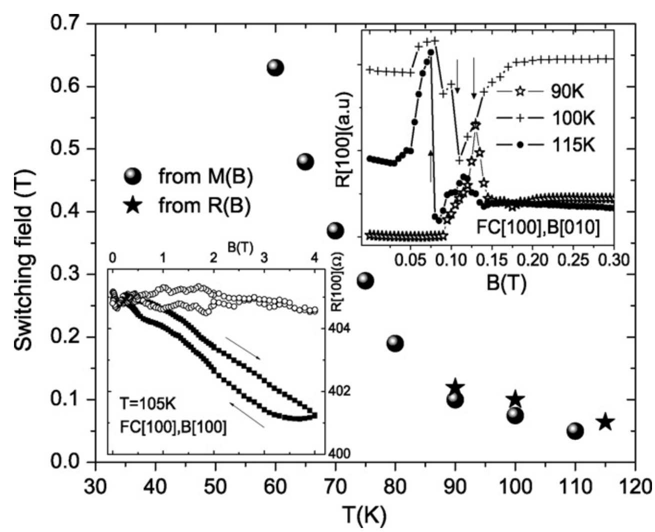

Fig. 3. Switching field values as drawn from magnetic measurements [3] (spheres) and the present studies (bold stars; the relevant R[1 0 0] curves after FC[1 0 0], B[0 1 0] for low magnetic fields are presented on the right inset). In the left inset, experimental R vs. B curve (bold squares) and the curve corrected for sample temperature flow (open circles) are shown.

3. Discussion and conclusions

The general aim of the present studies was to judge which of the two orderings: structural or magnetic, has the higher influence on electric transport properties in magnetite. Above a certain field (close to the magnetic saturation field) and at $T > T_V$, the longitudinal resistance (i.e. when the magnetic field is parallel to the current) always exceeds the transverse one (e.g. R[1 0 0] B[0 1 0] < R[1 0 0] B[1 0 0]); no systematics for transverse and longitudinal resistances was found at $T < T_V$. Since in both temperature regimes the highest field $B = 4$ T saturates the sample magnetically, the observed behavior suggests that the electronic transport at higher T is linked to magnetic moments direction, unlike at lower T, where structural disorder's influence on transport dominates over magnetic moment structure. Similar conclusions were drawn in [5], and, to some extend, in [7].

Dominant role of structural rather than magnetic influence on transport at $T < T_V$ suggests that the changeover of structure caused by axis switching should be reflected in transport properties. Actually, this can be inferred from Fig. 2, where the normalized resistance R[1 0 0]/R_{max} (R_{max} is a highest resistance value for each run) vs. magnetic field B loop for $T = 90$ K ($<T_V$) is presented; here, the sample was first field cooled FC[1 0 0]. Two loops are shown: the one with B[1 0 0] and the other with B[0 1 0], the last one expected to cause AS. While the $R(B)$ loop for B[1 0 0] does not show any abrupt behavior, this is so for B[0 1 0]; here, at certain B value, the resistance increases rapidly (or presents sudden changes, as shown in the right inset of Fig. 3 for slightly different experiment), the behavior apparently caused by AS. The enlargement of the region where

AS occurs is shown in the right inset. The temperature dependence of switching fields, as obtained from the position of the most pronounced change in resistance (shown by the arrows in the right inset of Fig. 3), presented in Fig. 3, is in satisfactory agreement with data drawn from magnetic measurements [3].

Although the structural influence on magnetite resistance dominates over the magnetic (at $T < T_V$), the last one is still visible under low magnetic fields where the sample is demagnetized. The result of this phenomenon: the small bump in R vs. B, is circled on the right inset of Fig. 2 and is currently under investigation.

In the left inset of Fig. 2, the time dependence of resistance, throughout the whole R vs. B loop, is correlated with sample thermometer output: apparently, any change in sample temperature (almost unavoidable in this kind of automated systems) greatly influences magnetite resistance rendering very subtle conclusions difficult. This is additionally shown in the left inset of Fig. 3, where the positive field R vs. B loop at 105 K is presented (bold squares) and compared to $R(B)$ corrected for the temperature resistance changes (see inset of Fig. 1). Apparently, all "magnetoresistance" at this temperature is an artifact effect caused by sample temperature flow. Although at lower temperatures, some negative magnetoresistance exists, the sample temperature flow is still very substantial and should be taken into account.

The results of our measurements have confirmed our prediction that axis switching phenomenon, observed in magnetic and structural experiments should influence also the electronic transport. In fact, this is what might have been expected bearing in mind the strong electron–phonon interactions [8] driving the transition: if the crystal lattice is changed then the electronic structure is influenced. Since it occurs quite close to the transition and since the transition involves reorganization of electronic level population (as $Fe^{2+} \rightarrow Fe^{2.5+}$ in original Verwey model), we might expect influence of magnetic field on the transition. This idea made us to study the axis switching. The problem, however, requires further studies that are currently in progress.

In conclusion, we have shown that the axis switching phenomenon is observed in transport characteristics at $T < T_V$. This fact, together with the lack of clear influence of R on the particular FC and subsequent in field (high, certainly above the saturation) measurements procedure suggests that electronic levels disorder caused by structural disorder is a leading parameter affecting low T electrical transport in magnetite.

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