Effect of the Morin transition upon electron transport in magnesium doped single crystal hematite
by Calvin Lee Ransom

A thesis submitted to the Graduate Faculty in partial fulfillment of the requirements for the degree of
DOCTOR OF PHILOSOPHY in Electrical Engineering
Montana State University
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Abstract:
The effect of the weak ferromagnetic-antiferromagnetic transition on the electron transport properties
has been observed in synthetic, single-crystal, magnesium-doped hematite (nominally 0.5 percent
replacement of iron). Separate measurements of the transverse voltage were taken with current flowing
along the [111] direction and within the (111) plane. The predominant results are: 1) The activation
energy of the transverse voltage is quite different for current flow along the [111] direction and in the
(111) plane. The energies are -1.3 and 1.3 ev, respectively.

2) The transverse voltage produced for current flow along the [111] direction is altered as the crystal
undergoes the weak ferro- magnetic-antiferromagnetic transition at the Morin temperature (TM),
whereas no change is observed at TM for transport in the (111) plane.

3) No normal or anomalous Hall effect is observed for current flow in either the [111] direction or the
(111) plane; however, a second order dependence upon the applied magnetic field is observed for the
transverse voltage.

4) The data also indicates that the activation energy of the transverse voltage is dependent upon the
current density (more properly the applied electric field) within the sample under test.

The observed results are interpreted in terms of a model which considers the effect of the magnetic
state upon the electron transfer process within the crystal. The electron transfer energy is assumed to be
a function of ?, where ? is the angle between the quantization directions (determined by the large
internal magnetic fields) of the iron ions between which the electron is assumed to hop.

5) It was also observed that the transverse voltage of the samples studied was dependent on the
magnetic structure. The effect is believed to be the result of the sample being an array of resistors in
which each resistor is a magnetic domain and within the domain the electrical properties are a function
of its magnetic state,
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IN MAGNESIUM DOPED SINGLE CRYSTAL HEMATITE

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An interesting and as yet little investigated problem is the consideration of charge transport in materials that possess a magnetic state.

This thesis considers only a very limited portion of this problem, but it is hoped that it will, because of the interesting experimental results obtained, provide motivation for more studies in this area.

The work begins with a brief discussion of why the magnetic state of a material may affect its conductivity.

Terms and concepts which may be unfamiliar to the reader are introduced, affording a glimpse of the overall problem. Then, in separate sections, these concepts are more completely described and simple models are used to illustrate them. These simple models are shown to apply to hematite and hence the hypothesis to be tested is formulated.

Chapter Two describes the experimental configurations. Chapter Three presents and discusses the results of these experiments.

The conclusions are summarized in Chapter Four.
ABSTRACT

The effect of the weak ferromagnetic-antiferromagnetic transition on the electron transport properties has been observed in synthetic, single-crystal, magnesium-doped hematite (nominally 0.5 percent replacement of iron). Separate measurements of the transverse voltage were taken with current flowing along the [111] direction and within the (111) plane. The predominant results are:

1) The activation energy of the transverse voltage is quite different for current flow along the [111] direction and in the (111) plane. The energies are 1.3 and 1.3 ev, respectively.

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3) No normal or anomalous Hall effect is observed for current flow in either the [111] direction or the (111) plane; however, a second order dependence upon the applied magnetic field is observed for the transverse voltage.

4) The data also indicates that the activation energy of the transverse voltage is dependent upon the current density (more properly the applied electric field) within the sample under test.

The observed results are interpreted in terms of a model which considers the effect of the magnetic state upon the electron transfer process within the crystal. The electron transfer energy is assumed to be a function of $\theta$, where $\theta$ is the angle between the quantization directions (determined by the large internal magnetic fields) of the iron ions between which the electron is assumed to hop.

5) It was also observed that the transverse voltage of the samples studied was dependent on the magnetic structure. The effect is believed to be the result of the sample being an array of resistors in which each resistor is a magnetic domain and within the domain the electrical properties are a function of its magnetic state.
INTRODUCTION

In considering charge transport in a material that may possess a magnetic state, be it ferromagnetic, ferrimagnetic, or antiferromagnetic, the question arises, "What will be the effect of the internal magnetic fields, associated with the magnetic state, upon the mobile charges?"

If one follows the approach suggested by Weiss in his successful explanation of spontaneous magnetization one would consider the internal magnetic field given by

\[ \vec{H}_I = \vec{H}_A + \gamma \vec{M} \]  

where \( \vec{H}_A \) is the externally applied magnetic field, \( \vec{M} \) is the magnetization of the material under consideration, \( \gamma \) a constant of unknown origin, and \( \vec{H}_I \) is the internal magnetic field.

This suggests that the Lorentz force would be of the form

\[ \vec{F} = q \vec{v} \times (\vec{H}_A + \gamma \vec{M}) \]  

neglecting for the moment any possible electric fields. \( q \) and \( \vec{v} \) are, of course, the charge and velocity, respectively, of the mobile charge.

In ferromagnetic materials \( \gamma \vec{M} \) is of the order \( 10^5 - 10^7 \) oe. It would thus seem that such fields cannot be neglected in considering possible Hall effects. This is indeed found to be the case.

If the class of materials considered is extended to include antiferromagnetics the magnetic fields now involve an additional term to
describe the effect at a two-sublattice antiferromagnet.

The concept of a sublattice is convenient and appropriate when discussing antiferromagnetism and ferrimagnetism. It is used to describe a crystal structure in which layers of atoms all possess a common magnetic orientation. Thus, for example, a two-sublattice antiferromagnetic material may be imagined as a structure composed of alternating layers, each layer consisting of atoms whose magnetic moments are parallel within a layer but in each layer the magnetic moment is antiparallel to the adjacent layers. This is illustrated in Figure 1.

In a two-sublattice model

\[
\mathbf{H}_I = \mathbf{H} (\mathbf{H}_A, \mathbf{H}_B)
\]

where \(\mathbf{H}_A\) and \(\mathbf{H}_B\) are the appropriate magnetizations of the two sublattices.\(^{2}\)

At this point the concept of a molecular field begins to cause serious difficulties when describing the motion of a charged particle in the crystal lattice. The fields must vary with position (something not required in the simplest model of a ferromagnet) because there are two sublattices to be considered. In the simplest approach, that in which the magnetic field within a given sublattice is constant, the Lorentz force acting upon a charged particle would depend upon within which sublattice the motion occurs.

As soon as there is to be transport of electrons through the alternating sublattices it becomes apparent that the simple model
Figure 1. Illustration of a Two-Sublattice System Consisting of Layers of Atoms. Within each layer all of the spins of the atoms are aligned.
postulated is much too simple.

While it is still clear that the molecular fields may significantly affect electron transport in antiferromagnetic materials, the models to be used in such a description now become unclear.

A group of materials that may allow these problems to become tenable, although through a more involved approach, are certain of the 3d oxides.

The 3d oxides consist of materials formed of oxides of the elements having incompletely filled 3d electronic shells. Some of these materials are semiconductors, \( \alpha\text{-Fe}_2\text{O}_3 \) and NiO, for example. There is reason to believe (3) the 3d wave functions in these materials do not overlap or only very weakly overlap, and thus a type of conduction occurs in which the mobile electrons spend most of their time localized on an Fe ion and are best described as being localized. Therefore, the system is more appropriately described as possessing discrete energy levels, rather than energy bands, at its Fe or Ni host ion. Then conduction occurs by a transfer of the electron from ion to ion. This type of conduction is frequently referred to as "hopping".

Such a transport mechanism is characterized by an activation energy associated with the movement of these self-trapped carriers. Their mobility will be extremely low because they spend most of their time trapped on the host ion.
However, if indeed the mobile electrons do spend a significant portion of time at a localized site, it is quite likely that an electronic polarization of the surrounding neighborhood will result and the mobile electron will, while localized, be required to satisfy Hund's rules. The mobile charge together with its resultant lattice distortion is known as a polaron.\(^{(4)}\)

If the itinerant electron is subject to Hund's rules, an interesting possibility arises if the material is antiferromagnetic. The stage is then set to satisfy the conditions required of double exchange. Double exchange was first suggested by Zener\(^{(5)}\) and later expanded upon by Anderson and Hasegawa\(^{(6)}\) and de Gennes\(^{(7)}\). Double exchange involves the transfer of an electron from one ion site to another ion site when the two ions have different quantization directions. The energy required to transfer the electron is then angularly dependent upon the relative orientation of the ion sites.

This is exactly the case for $\alpha$-$Fe_2O_3$: Conduction is thought to occur by hopping, the transport of charge occurring among the Fe 3d electrons; it is antiferromagnetic at certain temperatures, thereby magnetically orienting the spins of the Fe ions, but it has the added feature of undergoing a phase transition in which the spins of the Fe ions reorient themselves but no dimensional changes occur within the crystal. During this transition the material goes from an antiferromagnetic to a weakly ferromagnetic state over a temperature of approximately one degree centigrade. This transition, to be discussed in
detail later, is commonly referred to as the Morin transition and will be designated as occurring at $T_M$. This typically occurs in the range of $-10$ to $-40^\circ$C.

Some conductivity data is available for hematite, $\alpha$-Fe$_2$O$_3$, but most of it has included only studies of polycrystalline materials.

Morin's data on sintered hematite doped with titanium "suggests" a change in the Hall voltage as the temperature of the sample passes through $T_M$. He stated in that paper that single-crystal data would be forthcoming but to the best of this author's knowledge it has never been published. In later work, Morin claims no Hall effect should be present. More recent work does predict a Hall effect for polaron motion.

We shall now discuss in more detail the magnetic and electrical conduction properties of hematite and how the conduction process may be influenced by the magnetic state of the material.
A. Magnetic Behavior of Hematite

The crystal structure of a unit cell of hematite is illustrated in Figure 2.

To discuss the magnetic properties of hematite only the orientation of the magnetic moment of the Fe ions need be known. Figure 3 shows the orientation of these ions' magnetic moment with respect to the crystallographic axis, above and below the Morin temperature.

Thus, it becomes apparent that below $T_M$ the sum of the magnetic moments of the Fe ions within the unit cell completely cancel and the material is antiferromagnetic. Not so above $T_M$, where there is a net moment, in the (111) plane, in the amount of

$$M_{\text{weak ferromagnetic}} = 4 |\vec{M}_0| \sin \alpha$$

per unit cell. $\vec{M}_0$ is the magnetic moment of one of the Fe$^{3+}$ ions. This canting phenomena is known as weak ferromagnetism, $\alpha$ being the canting angle.

Dzialoshinski\(^{(16)}\) has shown that a material may exhibit weak ferromagnetism under certain symmetry configuration. Hematite satisfies these requirements.

The energy term Dzialoshinski obtained is of the form

$$E_{\text{weak}} = \vec{D} \cdot \vec{M}_a \times \vec{M}_b$$

where $\vec{D}$ is a constant vector parallel to the trigonal axis and $\vec{M}_a$ and $\vec{M}_b$ are the magnetic moments of the two sublattices, respectively. It should
Figure 2. Crystal Structure of $\alpha$-Fe$_2$O$_3$. The open and shaded circles represent $O^{2-}$ and Fe$^{3+}$, respectively.
Figure 3. Magnetic Structure of $\alpha$-Fe$_2$O$_3$. Arrows represent magnetization of iron ions.
be noted Dzialoshinski stated that this phenomenological theory is possible by symmetry arguments, but he did not specify its origin. Consideration of spin-orbit coupling and the exchange interaction using the perturbation theory of quantum mechanics will indeed produce such a term\(^{17}\).

Artman, Murphy, and Foner\(^{18}\) considered the temperature dependence of the magnetic dipolar field energy and the fine structure anisotropy energy and were able to show that these energies are very close in magnitude but compete for the directional orientation of the magnetic moment of the Fe ions within the crystal. Using antiferromagnetic resonance to obtain the constants needed in their theory, they were able to predict quite closely the ratio of the Morin temperature to the Neel temperature. The Neel temperature is the temperature at which the material becomes paramagnetic.

There have been a number of studies of the magnetic properties of hematite. Perhaps the most systematic studies were those of Lin\(^{19, 20, 21}\). He studied the magnetization, susceptibility and transition temperature. The last paper\(^{21}\) dealt with synthetic single crystals while the others concerned themselves with natural crystals. Because of the difficulties encountered in growing synthetic crystals, most studies are on natural crystals. Among those studies there is naturally much inconsistency of the data and it is obviously difficult to make studies of the effect of doping in such crystals. Sintered crystals have been used quite extensively. These are polycrystalline and thus have grain
boundaries which have drastic effects on demagnetizing fields in magnetic materials and should be avoided in studying fundamental properties of magnetic materials unless, of course, one is interested in just such effects.

Two types of deviations from the idealized Morin transition do appear in such studies: (1) the Morin transition may possess a large "line width". The transition occurring over as much as a 40°C range; (2) a shift of the Morin temperature to lower values as the materials are doped. These two effects may occur simultaneously. This behavior is illustrated in Figure 4.

The origin of this modified behavior is not understood. One author has claimed to observe that the widened transition is actually composed of many small shifted transitions, and he suggests that the total response is that of many separate regions of the crystal undergoing the normal narrow transitions.

This may be imagined much like the flipping of many domains similar to the Barkhausen jumps observed in ferromagnetic materials.
Figure 4. Possible Morin Transitions.
B. Charge Transport by Hopping

Consider a ring containing iron ions in which one ion has a different charge than the others. This is illustrated in Figure 5. Such a structure has the same energy as one in which the Fe\textsuperscript{2+} ion is at the j\textsuperscript{th} site rather than the i\textsuperscript{th} site. This is true even if the Fe\textsuperscript{2+} causes the adjacent ions to become polarized or shift positions on the ring.

There may exist a barrier which must be overcome to move the electron from the Fe\textsuperscript{2+} site to an adjacent Fe\textsuperscript{3+}, thereby shifting the Fe\textsuperscript{2+} site, but the system is the same regardless of the original position of the Fe\textsuperscript{2+} ion, assuming steady state conditions.

Under proper conditions it would be possible to produce a hole-electron pair (Fe\textsuperscript{2+} - Fe\textsuperscript{4+}) on a ring originally containing only Fe\textsuperscript{3+} ions. Similarly, a hole or an electron could be introduced by the substitution of an ion which has one fewer or one more electron, respectively, than Fe\textsuperscript{3+}. If this hole or electron were somehow moved many sites from the dopant position, the ion being assumed stationary on the chain, then conduction could occur, as outlined above, among the Fe ions only.

This is precisely how hematite is made semi-conducting — either by doping or by the creation of intrinsic hole-electron pairs at high temperatures\textsuperscript{(11-12)}. Pure hematite at room temperature has a resistivity of approximately $10^{10}$ ohm-centimeters. The conduction process is thought to occur by a hopping of 3d electrons from Fe ion to Fe ion.
Figure 5. Cyclic Chain on which Conduction Occurs by Hopping.
The decision of what dopant to use in obtaining a hole or an electron depends upon three factors: (1) the easiest to solve is the choice of sign of the carrier. This involves choosing an atom whose valence is such that when placed in the lattice it produces a locally deficient or excess electron. (2) The interstitial atom must be small enough to fit into the lattice without excessive distortion of the lattice dimensions, an extreme case causing so much distortion of the lattice that the crystal does not "grow". (3) The last factor to be considered is difficult to discuss analytically. The carrier associated with the dopant must not be so tightly bound to that site that it requires excessive energy to "activate" it.

Two elements that have been successfully used to produce conduction by holes and electrons are magnesium and titanium, respectively. Typical dopings are less than one percent atomic substitution for iron. Successful growth of single crystals large enough to be used experimentally require dopings of less than one-half percent Mg and Ti, in this author's experience. For such dopings, typical resistivities are 10-100 ohm-centimeter.
C. Double Exchange

The description of double exchange to be presented here will follow that outlined by Anderson and Hasegawa. Consider the electron illustrated in Figure 6 being transferred from ion $i$ to ion $j$ while subjected to the following constraints:

(C1) The directions of $\mathbf{S}_i$ and $\mathbf{S}_j$ are fixed (for example, by the local internal magnetic field in the crystal).

(C2) The electron to be transferred is either on the $i$th or the $j$th ion and while there is subject to Hund's rules.

To describe the energy associated with the transfer of the electron from site $i$ to site $j$ the matrix theory of quantum mechanics is used. The basis functions are those of the isolated ions. They are to be designated by $|d_i\rangle$ and $|d_j\rangle$, the $d$ referring to the $d$ electrons of iron, for those of the $i$th and $j$th sites, respectively.

When the itinerant electron is localized on the $i$th ion it will be assumed that its spin orientation relative to the ion's spin is described by the Hamiltonian

$$H = -2J \mathbf{S}_i \cdot \mathbf{S}$$  \hspace{1cm} (6)

where $J$ is the Heisenberg intraexchange integral, $\mathbf{S}_i$ is the spin of the $i$th ion, and $\mathbf{S}$ is the spin of the electron. Hund's rules reduce this to two possible states

$$E = \pm J |\mathbf{S}_i| = \pm J S$$  \hspace{1cm} (7)
Itinerant Electron

$i^\text{th}$ ion

$j^\text{th}$ ion

$S = \text{Spin of electron}$

$S_c = \text{Spin of ion}$

Figure 6. Coordinates Defining $\theta_{ij}$. 
corresponding to whether the electron's spin is parallel or antiparallel to \( \vec{S}_i \). While at site \( j \) the electron's energy will similarly be given by

\[
E = \pm J \left| \vec{S}_j \right| = \pm J S \tag{8}
\]

What is the sign of \( J \)? Hund's rules state that if the electronic shell is less than half full, the next electron added to the atom will "go in" with its spin parallel to the others and antiparallel if the electronic shell is more than half full. Hence, the lowest energy when the shell is less than half full would require \( J \) to be positive and if more than half full, \( J \) must be negative. We shall see later that the sign of \( J \) is irrelevant in considering which states will have the lowest energy when the electron is transferred from one site to the other.

When considering the electron to be on one site or the other there is no dependence upon the relative orientation of the ion which does not contain the extra electron. However, if a transfer is made from site \( A \) to site \( B \), then the energy associated with this transfer

\[
E_T = \langle B | H_T | A \rangle \tag{9}
\]

must somehow account for the fact that the state given by \( |B\rangle \) and \( |A\rangle \) may not both refer to the same coordinate system. Compensation for this may be done in two ways:

1. Transform the energies for site \( B \) into a coordinate system parallel to \( A \)'s, or
2. Transform the transfer matrix elements so they refer
correctly to the different directions associated with the sites A and B.

The latter procedure will be used. For this, it is necessary to introduce the rotation operator given by

\[
R_u(\phi) = e^{iJ\phi} \quad (10)
\]

for a rotation by an amount \( \phi \) about an axis \( u \). \( J_u \) is the component of angular momentum along \( u \), taking \( \hbar = 1 \).

The two possible spin states of the electron when referred to the direction of \( \vec{S}_i \) will be designated by \( \alpha \) and \( \beta \). Those corresponding to \( \vec{S}_j \) by \( \alpha' \) and \( \beta' \). Thus the eigenstates for the electron on ion \( i \) are represented by \( |d_i\alpha\rangle \) and \( |d_i\beta\rangle \), and those on \( j \) by \( |d_j\alpha'\rangle \) and \( |d_j\beta'\rangle \).

Inserting the proper discrete values of the allowed angular momentum equation (7) becomes

\[
E(d_i\alpha) = -J S_i = -J S \quad (11)
\]

\[
E(d_i\beta) = -J(S_i+1) = J(S+1) \quad (12)
\]

Similarly, when the electron is at the \( j \)th site

\[
E(d_j\alpha') = J S_j = -J S \quad (13)
\]

\[
E(d_j\beta') = J(S_j+1) = J(S+1) \quad (14)
\]
The transfer matrix elements, if $S_i$ and $S_j$ are parallel, would be

\begin{align*}
\langle d_i \alpha \mid H_T \mid d_j \alpha' \rangle &= t \tag{15} \\
\langle d_i \beta \mid H_T \mid d_j \beta' \rangle &= t \tag{16} \\
\langle d_i \alpha \mid H_T \mid d_j \beta' \rangle &= 0 \tag{17} \\
\langle d_i \beta \mid H_T \mid d_j \alpha' \rangle &= 0 \tag{18}
\end{align*}

The last two equations simply state that the transfer only occurs when the initial and final spin states are the same on the respective ion sites (again, a requirement needed to satisfy Hund's rules).

What does the transformation of the transfer matrix elements involve? Consider the coordinate system shown in Figure 7. The rotation operator takes the form

\begin{equation}
R_u(\theta) = e^{i \theta/2 \sigma_u} \tag{19}
\end{equation}

\begin{equation}
= 1 \cos \theta/2 - i \sigma_u \sin \theta/2 \tag{20}
\end{equation}

where $\sigma_u = 2 S_u$ (i.e., the Pauli spin matrices have been introduced).

For the coordinate system chosen the rotation is about the $y$ axis and hence

\begin{equation}
\sigma_u = \sigma_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \tag{21}
\end{equation}

and
Figure 7. Coordinates Defining the Rotational Transformation of the Spin.
The spin coordinates now transform as

\[ a = \cos \theta/2 \, a' + \sin \theta/2 \, \beta' \]  
\[ \beta = -\sin \theta/2 \, a' + \cos \theta/2 \, \beta' \]  

This resembles a rotation of a coordinate system \( x' - y' \) through an angle \( \theta \) to produce a new system, \( x - y \), except for the factor of \( \theta/2 \). This results because the "thing" being transferred has, being an electron, a spin of one-half. Taking this into account results in an energy matrix as shown below:

\[
\begin{pmatrix}
-d_i a & d_i \beta & d_j a' & d_j \beta' \\
-d_i a & -J S_i & 0 & t \cos \theta/2 & t \sin \theta/2 \\
-d_i \beta & 0 & J(S_i+1) & -t \sin \theta/2 & t \cos \theta/2 \\
-d_j a' & t \cos \theta/2 & -t \sin \theta/2 & -J S_i & 0 \\
-d_j \beta' & t \sin \theta/2 & t \cos \theta/2 & 0 & J(S_i+1)
\end{pmatrix}
= H
\]
The thing to be noted in equation (25) is that the off-diagonal elements, i.e. those associated with the transfer of the electron from one site to another, are now angularly dependent upon the relative angular orientation of the sites between which the electron moves.

Let us now illustrate how these off-diagonal elements may vary with the direction of the electron motion in a crystal, assuming that this motion occurs by a hopping from one site to the next so that the two-ion model may be used. Consider the two-dimensional lattice shown in Figure 8. The arrows indicate the quantization direction of the ions. Suppose, for simplicity, the corresponding wave functions are the same along and symmetrical about the x and y axes as illustrated by the fourleaf appearance of the wave functions. If the electron were to go from one ion to the next along the \([10]\) direction then \(\theta = 0\) and (25) becomes

\[
\begin{bmatrix}
-JS & 0 & t \cos \theta/2 & t \sin \theta/2 \\
0 & J(S+1) & -t \sin \theta/2 & t \cos \theta/2 \\
t \cos \theta/2 & -t \sin \theta/2 & -J.S & 0 \\
t \sin \theta/2 & t \cos \theta/2 & 0 & J(S+1)
\end{bmatrix}
\]
Figure 8. Symmetric Wave Functions in a Two Dimensional Crystal. The arrows represent the magnetic moments of the atoms.
However, if the electron transfer is along the [01] direction then \( \Theta = \pi \)
and (25) becomes

\[
\begin{bmatrix}
-JS & 0 & t \cos \pi/2 & t \sin \pi/2 \\
0 & J(S+1) & -t \sin \pi/2 & t \cos \pi/2 \\
t \cos \pi/2 & -t \sin \pi/2 & -JS & 0 \\
t \sin \pi/2 & t \cos \pi/2 & 0 & J(S+1)
\end{bmatrix}
= \begin{bmatrix}
-JS & 0 & 0 & t \\
0 & J(S+1) & -t & 0 \\
0 & -t & -JS & 0 \\
t & 0 & 0 & J(S+1)
\end{bmatrix}
\]  \( (27) \)

It will be shown in the next section that the expression for charge transport involves the use of off-diagonal elements, which do appear to be different for this simple two-dimensional model.

When the unit cells of hematite are joined to form a crystal the structure generated is quite similar to that of Figure 8. The Fe ions form a two-sublattice structure which is illustrated in Figure 9. Within each layer of the sublattice the magnetic moments of the ions are parallel to each other.

For charge transport within the (111) plane, \( \Theta = 0 \) both above and below the Morin transition, whereas current flow along the [111] direction has \( \Theta = \pi \) for \( T < T_M \) but \( \Theta = \pi - 2\alpha \) for \( T_M < T < T_{\text{Neel}} \). \( \alpha \) is typically of
Figure 9. Sublattice Magnetization Directions of Hematite.
the order of 5°. Thus, if double exchange is to be a significant factor in discussing the energy of mobile electrons in hematite, then as the temperature is varied through $T_M$, the value of $\theta$ is unchanged for transport within the (111) plane and hence the matrix elements describing the system are those shown in equation (26). However, for electron transport along the [111] direction with $T < T_M$, $\theta = \Pi$; but for $T > T_M$, $\theta = \Pi - 2\alpha$. Thus, below $T_M$ the energy matrix is that of equation (27) but above $T_M$ it becomes

$$
\begin{bmatrix}
-JS & 0 & t \sin \alpha & t \cos \alpha \\
0 & J(S+1) & -t \cos \alpha & t \sin \alpha \\
t \sin \alpha & -t \cos \alpha & -JS & 0 \\
t \cos \alpha & t \sin \alpha & 0 & J(S+1)
\end{bmatrix}
$$

Thus, in going from $T < T_M$ to $T > T_M$, the energy matrix, for current flow in the [111] direction, undergoes a change in the two by two submatrices off the main diagonal. If the expression for the charge transport in hematite is dependent upon these off-diagonal elements it appears there may be a change of the transport properties along the [111] direction at $T_M$ (because the off-diagonal elements change at $T_M$) whereas for transport along the (111) plane no change would occur because there is no change in the corresponding matrix elements at $T_M$. 

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D. A More Detailed Look at the Hopping Mechanism

A classical model of hopping, considered on a microscopic basis, would describe a current as a succession of electron movements (transfers) over a discrete distance (from ion site to ion site). Thus, in writing an expression for the current as

$$\bar{J} = q n \bar{v} \quad (29)$$

it would seem appropriate to replace the velocity, $\bar{v}$, by a term,

$$\bar{v} = \langle \omega \rangle_{a \rightarrow B} \bar{d}_{a \rightarrow B} \quad (30)$$

where $\langle \omega \rangle_{a \rightarrow B}$ is the probability per unit time for transfer from site $a$ to site $B$, which may be degenerate; i.e., there may be equivalent sites in the crystal, and $\bar{d}_{a \rightarrow B}$ is the vector distance traversed in the transfer.

The net current would be the average of all such hops in the crystal; i.e.,

$$\bar{J} = \langle \sum q n_a \langle \omega \rangle_{a \rightarrow B} \bar{d}_{a \rightarrow B} \rangle \quad (31)$$

where $n_a$ is the number of particles per unit volume at sites equivalent to $a$.

Using first order time-dependent perturbation theory the transition probability is given by (26)
where $V_{ba}^a$ is the matrix element connecting states $|a\rangle$ and $|b\rangle$; i.e., $V_{ba} = \langle b | V | a \rangle$, and $\rho_b$ is the density of final states at $b$.

Thus it becomes apparent how the transfer matrix elements influence the current in a hopping process. See comments after (25).

The above description of current flow is by no means unique. It should be just as valid to use Equation (33) where the quantum mechanical equivalent of $\vec{v}$ is used. This may be done in two ways:

1. $\vec{v} = (\frac{1}{i\hbar}) [ \vec{F}, H ]$ (33)

where $\vec{v}$ is the time rate of change of the position vector, $\vec{F}$, and hence is obtained by commuting $\vec{v}$ with the Hamiltonian.

2. $\vec{v} = (\frac{1}{m})[\vec{P} + q(\vec{A}/c)] = (\frac{1}{m})[(\hbar/i)V + q(\vec{A}/c)]$ (34)

where $\vec{V}$ is the gradient operator and $\vec{A}$ is the magnetic vector potential as "seen" by the moving charge. If the exact Hamiltonian can be described and the resulting set of eigenfunctions found, then all three descriptions of the current should be valid. The exact solutions cannot, however, be obtained and must be approximated. This implies that varying degrees of approximation must be accepted and thus the values of $J$ obtained by the three approaches outlined above will, in
general, yield different results. The first approach, considering the probabilities for an electron transfer, directly incorporates the concept of hopping on a microscopic basis and has thus been chosen for the discussion of how current "flows" in such materials.

Some of the multiple paths for current flow along the (111) direction or possible sites for hopping are illustrated in Figures 10 and 11.

For the case $T < T_M$, there are two allowed hopping paths.--- (remember we required

$$
\langle d_1 \beta | H_T | d_2 \beta \rangle = \langle d_1 \beta | H_T | d_2 \beta \rangle = 0
$$

as shown by the dotted lines representing the connecting matrix elements $V_1$ and $V_2$, respectively.

For $T > T_M$ there are four possible paths. The only hops considered are to the nearest neighbor sites in the direction of current flow. A question arises with respect to matrix elements such as $V_2$, $V_2'$, and $V_2''$. Are they equal? In the absence of external fields it appears that they are.

Consider for the moment the angular and time dependence of the basis functions used to describe the 3d electrons.

This is of the form

$$
\psi = \psi(\theta, \phi, t) = \Theta(\theta) \phi(\phi) e^{i E_0 t/\hbar}
$$

(35)
Figure 10. Illustration of Allowed "Hops" for $T < T_H$.
The spins of the ions are along the $[111]$ direction, $\odot$ being antiparallel to $0$. 
Figure 11. Illustration of the Allowed "Hops" When $T > T_M$.

The spins of the ions lie in the (111) plane and are canted as shown. Transitions such as $V_2^*$, etc., have been omitted to keep the sketch uncluttered.
but

\[ \Phi(\varphi) = e^{i m_1 \varphi} \quad (36) \]

Putting \( E = \hbar \omega_0 \), we obtain

\[ \psi = \Theta(\omega) e^{i(\omega t + m_1 \theta)} \quad (37) \]

which represents a wave-like rotation of the wave function about the z-axis with angular frequency \( \omega \sqrt{m_1} \). Sketches of such functions are shown for the various 3d states in Figure 12. The itinerant electron spends most of its time trapped and only occasionally jumps. Thus, it gets a rather "fuzzy picture" of its neighbor as shown in Figure 13 which illustrates the possible \( V_2, V_2', \) and \( V_2'' \) for a hop from a 3d\(^4\) to a 3d\(^5\) state. Similar sketches can be drawn for the other transitions in the crystal. From the symmetry exhibited one expects all hops to equivalent sites to involve identical matrix elements because of the time average symmetry of the wave functions.

What is the difference between the two-sublattice hopping put forth earlier and this seemingly more complicated model? Strictly speaking, the two-sublattice model forbids hopping in the [111] direction since the matrix elements of

\[ V \text{ connecting antiparallel spin} = 0 \quad (38) \]

for temperatures above the Morin temperature. The more detailed model still adheres to this requirement of the transfer matrix elements but
Figure 12. "Spinning" 3d Wave Functions.
Figure 13. "Pictorial Hopping" from a $3d^4$ to a $3d^5$ State.
considers a transfer which has a lateral component. In the absence of external fields the average of such transfers must yield a net transverse component equal to zero. Thus the charge is transferred from a position on the \([\text{III}]\) axis of one unit cell to that of an adjacent unit cell, rather than along the \([\text{III}]\) direction at each unit cell. Thus, a net current flow along the \([\text{III}]\) direction is allowed.
THE EXPERIMENT.

Single crystals of hematite doped with magnesium were grown by the methods outlined by Besser (27). The doping was one-half percent (nominal) atomic substitution for iron.

Two samples were cut from the same single crystal being separated by only 12 milli-inches due to kerf loss of the saw blade and approximately two milli-inches were removed when polishing the surfaces of the crystals. Both samples are regular parallepipeds with dimensions of 0.050" x 0.050" x 0.150". The orientation of crystal with respect to the rectangular sides of the sample is illustrated in Figure 14.

Silver contacts were applied by evaporation of silver in a vacuum followed by firing at 300°C for 20 minutes. The contacts on the square ends were 0.050" in diameter. Those on the rectangular sides were 0.010" x 0.050". The 0.010" width was chosen to minimize the shorting caused by this contact. The contact orientation is illustrated in Figure 15.

The induced voltage was measured while passing a constant current through the sample in the longest direction and measuring the voltage difference produced between the smaller rectangular contacts.

The samples were mounted so they could be rotated through 360° while subjected to an external magnetic field and a known temperature.

Because there are many possible orientations of current flow, voltage measured, and magnetic field directions with respect to
Figure 14. Sample Crystallographic Orientation.
Figure 15. Basic Experimental Configuration.
crystallographic directions these orientations will be shown on each figure where the results are presented and will thereafter be referred to by the respective figure number.

The values of magnetization were obtained using a Foner vibrating-sample magnetometer\(^{(28)}\). Magnetic fields were obtained from a Varian 3600 magnet with field dial control of the field magnitude.

The data were taken directly using as x-y recorder having such inputs as a voltage proportional to; the angle of the sample with respect to the applied magnetic field, the temperature of the sample, the magnetization of the sample, and the transverse voltage \(\Delta V\).

The values of the magnetization were obtained using a Foner vibrating-sample magnetometer\(^{(28)}\). Magnetic fields were obtained from a Varian 3600 magnet with field dial control of the field magnitude. A more detailed discussion of the experimental configuration and how the experiment was conducted is given in Appendix A.
RESULTS

The experiment shown diagrammatically in Figure 15 determines the transverse resistivity, temperature, magnetic field and electric field dependence. That is, the parameters being studied are described by the resistance tensor \( \rho \) given by

\[
\begin{bmatrix}
V_x \\
V_y \\
V_z
\end{bmatrix} = \begin{bmatrix}
\rho_{xx} & \rho_{xy} & \rho_{xz} \\
\rho_{yx} & \rho_{yy} & \rho_{yz} \\
\rho_{zx} & \rho_{zy} & \rho_{zz}
\end{bmatrix}
\begin{bmatrix}
I_x \\
I_y \\
I_z
\end{bmatrix}
\]

in which each \( \rho_{ij} \) is a function of temperature, applied magnetic field, magnetic state of the sample, and applied electric field. If the experiment is characterized by its terminal behavior, then

\[
V_y = \rho_{yx} I_x + \rho_{yy} I_y + \rho_{yz} I_z
\]

\[
= \rho_{yz} I_z \tag{38}
\]

assuming the differential voltmeter has an infinite input impedance. It is, then, \( \rho_{\perp} \) that this experiment characterizes.

Figures 16 and 17 give \( \Delta V \) as a function of temperature for current flow along the [111] direction and in the (111) plane, respectively.

Each of the curves may be used to determine the activation energy of \( \rho_{\perp} \) when the current is along the [111] direction and in the (111)
Figure 16. Temperature Dependence of the Transverse Voltage for Current Flow in the [111] Direction.
Figure 17. Temperature Dependence of the Transverse Voltage for Current Flow in the (111) Plane.
These energies are

\[
E_A^{(111)} = -1.3 \text{ eV} \quad \text{and} \quad E_A^{(111)} = 1.3 \text{ eV},
\]

respectively. Note that these are determined for a specific current density.

There is a rapid change in \( \Delta V \) at the Morin temperature in Figure 16 but not in Figure 17. This is exactly as expected if a double exchange were a significant factor in describing the conduction process.

Figure 18 is a plot of \( \Delta V \) for current flow along the [111] direction and an external magnetic field applied in the (111) plane. \( \theta \) is the angle between the magnetic field direction and the contacts used to measure \( \Delta V \). The temperature was fixed at 25°C; i.e., \( T > T_M \).

The results are not as expected in a normal Hall experiment. The resulting voltage has a \( 2 \theta \) dependence rather than just \( \theta \); i.e., the resulting transverse voltage is not linear in applied magnetic field but is of second order. The origin of this behavior is not known.

Figure 19 shows the effect of rotating the sample with respect to the magnetic field when the magnetic field is in the plane formed by the direction of current flow along the [111] direction and the direction defined by \( \Delta V \). Again, \( T = 25^\circ C > T_M \).

Here the influence of the large anisotropy confining the magnetization of hematite to the basal plane is evident.
Figure 18. $\Delta V$ as a Function of the Basal Plane Magnetic Field Direction.
Figure 19. Effect of the Magnetic Field Strength upon $\Delta V$. 
The anisotropy energy within the basal plane is very weak, being equivalent to tens of oersteds. The magnetization is, however, confined to this plane except for very large fields, on the order of $10^5$ oe. Under such conditions an $M-H$ loop in which $H$ is applied in a plane containing the [111] direction would be similar to that shown in Figure 20(a). But the data of Figure 18 indicates we should be considering the second order field and hence more appropriate is the $|M| - H$ loop shown in Figure 20(b).

Remembering that the sample consists of many magnetic domains, the results are obvious. When $H$ is along the [111] direction the magnetization is still confined to the (111) plane but $\Theta$ is randomly oriented in the plane resulting in no net magnetization.

As soon as $H$ is off axis enough, all the domains are aligned. Thus, what one observes is the net resistance of many small resistors whose value depends upon its magnetic state. Once all of the resistors have the same value (the same magnetic state and orientation), the sample behaves as a single resistor (domain). This is exactly what is shown in Figure 19. The family of curves produced as the magnetic field strength is varied also supports this interpretation of the results. At low fields the sample is essentially demagnetized and hence no variation in $\Delta V$ is observed; as the field strength is increased the effect is to align a greater percentage of the domains and hence a greater "sharpness" of change in $\Delta V$ when the magnetization of the sample switches at $H_c$. 
$\theta_0 = \sin^{-1} \frac{H_c}{H_{\text{Applied}}}$

Figure 20. Hematite $M-H$ Loops.
It has been found experimentally that the temperature at which the Morin transition occurs is a function of the applied magnetic field. Increasing the magnetic field lowers $T_M$ in a nearly linear relationship. This is shown in Figure 21. It is thus expected that the change in $\Delta V$ exhibited in Figure 16 should occur at a different temperature as the external magnetic field is changed. This is observed in Figure 22.

The temperature dependence of $\Delta V$ for three different current densities is shown in Figure 23. Two interesting results appear: (1) apparently the Morin temperature may be changed by varying the current density; (2) the activation energy (or temperature coefficient) is observed to change sign at a certain current density.

The first result may be a result of the experimental procedure. The sample has a resistance of about one kilohm. A change of current from 2 to 2.5 milliamps would result in an additional 2.25 milliwatts being dissipated in the sample. This additional power dissipation could raise the temperature of the sample a few degrees thereby causing an apparent shift in the Morin temperature with increasing current.

It is difficult to imagine any factor attributable to the experimental techniques used which would produce a change in the slope of the curves, (i.e., a change in the activation energy), as the current through the sample is varied.

It is possible that the change in the slope is a result of the electric field having an effect on the transition probabilities of the hopping electrons. For example, referring to Figure 11, at a given
Figure 21. Shift of the Morin Transition as a Function of the Magnetic Field.
Figure 22. Shift of the Transverse Voltage as a Function of the Applied Magnetic Field.
Figure 23. Transverse Voltage as a Function of the Current Through the Sample.
value of electric field conduction by the transition indicated by $V_4$ may be occurring but the electric field strength may not be sufficient to produce a significant contribution from $V_2$. Upon increasing the electric field the contribution from $V_2$ may become appreciable. The result is an increase in the number of possible hopping sites available for the conduction process.

Holstein and Friedman\(^{(5)}\) mention that the Hall mobility, $\mu_H$, is given by

$$\mu_H = \frac{c}{H} \quad \sigma_{xy} = \frac{c}{H} \quad \mu_{xy} \quad \mu_{xx} \quad (39)$$

where the conductivity tensor $\sigma$ is the inverse of the resistivity tensor. They further state that mobility along the primary direction of current flow, $\mu_{xx}$, is predominantly of the activation form $e^{-E/kT}$ while the transverse mobility, $\mu_{xy}$, may vary as $1/T$ and hence the Hall mobility may exhibit a negative activation energy.

Equation 39 would allow both $\mu_{xy}$ and $\mu_{xx}$ to vary as $e^{-E/kT}$ and under proper conditions the ratio would produce a negative temperature coefficient.

The above discussion is not intended to be rigorous but only to give a plausible explanation of the factors that would contribute to an electric field dependent temperature coefficient for a material in which conduction occurs by hopping.
CONCLUSIONS

This work was intended to be an experimental study to see if materials possessing a magnetic state have electrical conductivity properties which are influenced by the magnetic properties of the material. Hematite was chosen for the study because of its precisely defined transition (Morin transition) in which it goes from a weak ferromagnetic to an antiferromagnetic state. This is believed to be the first study of this kind undertaken using single crystals.

The results indicate that the electrical properties are altered at the Morin transition for current flow in the [111] direction but do not change when the current flows within the (111) plane. These results are interpreted as being a consequence of double exchange.

The data also indicates that the conduction process, which occurs by hopping, is not linearly dependent upon the electric field applied to the sample. A plausible explanation is put forth to explain this behavior using concepts related to conduction by hopping.

It also appears that even though the sample is a single crystal its conductivity is affected by the domain structure of the sample.

Further studies certainly seem appropriate. Perhaps most interesting would be a theoretical study in which an attempt is made to determine why there is no linear Hall effect but only a second order dependence upon the applied magnetic field.
Also of interest would be calculations for the temperature dependence of the electrical conduction in the various directions within the crystal.

There is more need for experimental studies both to more completely characterize the conductivity tensor and to observe the effects of other dopants.

A study correlating the resistance and domain structure could also prove useful.
APPENDIX A

DESCRIPTION OF THE EXPERIMENT

The experimental configuration used to measure the transverse voltage of the sample is shown in more detail in Figure 24. The temperature of the sample is determined by the use of a thermistor. The thermistor is a YSI-Components precision thermistor # 44005 having a 25°C value of 3000 ohms. The temperature is determined by passing 100 microamps through it and measuring the voltage produced by the use of a differential amplifier. The output of this amplifier supplies the x-axis input for a Mosley model 135A x-y recorder. This is the source of the temperature scale for Figures 16, 17, and 23. Knowing the current through the thermistor, the amplifier gain and having a calibration of the resistance vs temperature of the thermistor enables one to determine the temperature of the thermistor.

To obtain a voltage proportional to the angle of the sample holder with respect to the applied magnetic field direction, a potentiometer was connected to the sample rod. This voltage supplies the input to the x-y recorder to provide the θ values in Figures 18 and 19.

The transverse voltage is obtained by the use of a differential voltmeter which supplies the input to the y-axis of the x-y recorder. This provides the y-axis readings on Figures 16, 17, 18, 19, 22, and 23.

The differential voltmeter consists of an integrated circuit operational amplifier produced by Fairchild (µA 709). It is used in a single ended non-inverting mode as shown below:
Leads for current sources and differential voltmeter

Sample Holder

Dewar-filled with acetone to which dry ice is added to lower the temperature of the sample

Magnet pole piece

Enlarged view—approximately 3 times actual size

Thermistor

Sample holder

Tygon tubing

Soldering iron

Printed circuit board on which sample is mounted and leads for sample attached

Hematite sample

Figure 24. The Experimental Configuration.
In this mode and for the values of the gain obtained (.5 and 10) the amplifier has a worst case input impedance of approximately fifty megohms.

A typical run in which the temperature of the sample is varied is carried out as follows: The value of the current through the sample is chosen as are the magnetic field strength and direction. Dry ice is then added to the acetone in the dewar until the lowest temperature desired is obtained. The system is then allowed to "warm-up" by itself and it is during this time that the data is obtained. A typical warm-up rate is approximately one degree centigrade every three minutes.

It was found, during the initial experiments, that because of the small diameter of the dewar in the region containing the hematite sample that the temperature would fluctuate quite rapidly. Adding the Tygon tubing and a soldering iron operating at a low temperature, as illustrated in Figure 24, caused a small stream of bubbles to pass upward through the dewar stem and had the effect of keeping the liquid stirred and at a more uniform temperature.
LITERATURE CITED


LITERATURE CITED (continued)

22. Personally obtained data.
29. The phrase "double exchange" was originally coined by Zener to refer to the transfer of an electron from one Na ion to the adjacent Na ion as the transfer of an electron, say, from the left Na atom to the central Cl ion simultaneously with the transfer of an electron from the central Cl ion to the right Na ion, i.e. the system before the transfer consists of Na Cl Na and after the transfer consists of Na Cl Na. Anderson and Hasegawa later relaxed the definition to include a simple transfer of an electron between two ions, for example, before the transfer Mn Mn and after the transfer Mn Mn.
Effect of the Morin transition upon electron transport in magnesium doped single crystal hematite.