Electrical Transport Properties in Metal-Insulating Nanocomposite Films

by

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Publications

Giant Hall effect and spin-dependent transport in granular NiFe-SiO$_2$ films

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On the origin of the giant Hall effect in magnetic granular metals


Electric relaxation in conducting metal-insulator nanocomposites

Pakhomov A B, Wong S K, Yan X.

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Low frequency divergence of dielectric constant in metal-insulator nanocomposites with tunneling

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Phenomenological considerations in Percolating Magnetic Nanostructures

S K Wong, B Zhao, T K Ng, X N Jing, X Yan and P M Hui

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Abstract

The DC resistivity, magnetoresistivity and Hall effect were studied in percolating nanostructured composites such as, NiFe-SiO$_2$ films as a function of composition and temperature. A phenomenological model was proposed based on quantum size effect, to explain correlation between magnetoresistivity, Hall resistivity and resistivity. The impedance of granular co-sputtered films NiFe-SiO$_2$, Au-SiO$_2$ and Pt-SiO$_2$ measured in the frequency range of 20Hz-1MHz. The results suggest that the effective dielectric constant is divergent at low frequency. In the proposed explanation, both polarization and tunneling between clusters of metal particles are taken into account.
Chapter 1

Introduction

Composites consisting of nanometer-scale metal particles immersed in an insulating matrix have attracted considerable interest due to their unusual transport, magnetotransport, dielectric and magnetic behavior [1]. It is known that by embedding ferromagnetic metal granules in a non-magnetic immiscible medium, variety of physical properties will be enhanced. In order to make comparison with ferromagnet-insulator composite, non-magnetic insulator composites have also been under investigated.

Resistivity, magnetoresistivity and impedance granular NiFe-SiO₂, Au-SiO₂ and Pt-SiO₂ films, have been investigated at various metallic volume fractions p. Because Ni (Au, Pt) and SiO₂ are almost immiscible, cosputtering creates a structure. At low p, metal particles of several nanometers in diameter are imbedded in an insulating host and at high p, insulator inclusions are dispersed in the metal host. The vanishing size of the intergranular contacts, electron interaction, and localization effects all contribute to variety of resistivity or impedance with different metal fraction. One of the interesting features is that both ordinary and extraordinary Hall effect increase ~4 orders of magnitude, which is larger than the maximum enhancement factor ~20 of simple percolation predicted. This enhancement, so called giant Hall effect (GHE) [2,3], was observed in helium temperature. One of our study is we observed this enhancement also in room temperature. A phenomenological model was proposed to describe this observation.

Although, large amount of DC electrical measurements have been done, there are still problems in understanding mechanisms of conduction and localization in
cerements, particularly in the concentration range close to and above the metal-insulator transition. In an effort to complete the study of electrical transport of ferromagnet-insulator composites, effective conductance and capacitance of NiFe-SiO₂, Pt-SiO₂ and Au-SiO₂ films, with metal volume fractions p exceeding the percolation threshold p_c (~0.53), were measured as functions of frequency in the range 20 Hz - 1M Hz. When p is close to p_c, the percolation-type dispersion is observed at high frequency. The most striking result is the rapid increase of effective capacitance of conducting samples at low frequency, where the variation of resistance is very small. We attribute this behavior to the combination of metallic conduction with tunneling processes.

**Electrical transport properties in Granular Metals**

1.1 *DC electrical conductivity*

By using suitable sputtering condition or annealing, there are three distinct structural regions in granular metals: metallic region, intermediate transition region and insulating region.

In the metallic region, the metal grains touch with each other and form a metallic continuum with dielectric inclusions. Most physical phenomena of bulk ferromagnetism, which depend on transfer of electrons, are retained. However, properties like electrical conductivity and temperature coefficient of resistivity (TCR) are considerably modified due to strong electron scattering from the dielectric inclusions and the grain boundaries.

In the intermediate transition region, the electrical conductivity in this region is due to both percolation along the metallic maze and electron tunneling between
isolated metal grains. By further decreasing $p$, the TCR would becomes negative. It is due to the thermally activated tunneling becomes comparable to the contribution of percolation. The critical transition happened at the percolation threshold composition $p_c$, below which only the tunneling process contributes to the conductivity of the granular metal.

In the insulating region, the metal is in the form of isolated grains with distribution of size dispersed in a dielectric continuum. The electric transport is realized by intergranular tunneling or temperature activated hopping. It was found that the low-field electrical conductivity, $\sigma_L$, of granular metals in the insulating region has a characteristic temperature dependence, $\ln \sigma_L \propto T^2$. At low temperature and high-field electrical conductivity, $\sigma_H$, has an electric field dependence, $\ln \sigma_H \propto -\frac{1}{E}$, where $E$ is the electric field.

Also, the relationships between resistivity, magnetoresistivity, and both extraordinary and ordinary Hall resistivity were observed by varying the metal volume fraction in room temperature. Extraordinary Hall resistivity $\rho_{xy}$, ordinary Hall resitivity $\rho_{xx}$ and magnetoresistivity $|\Delta \rho|$ follow power law dependencies on resistivity $\rho$. Namely, $-\rho_{xy} \propto \rho^{\alpha}$, $-\rho_{xx} \propto \rho^{\alpha'}$, and $-\Delta \rho \propto \rho^{b}$, with $\alpha = 0.69 \pm 0.07$, $\alpha' = 0.7 \pm 0.1$, and $b = 1.2 \pm 0.2$, with resistivity spanning from $10^{-5} \Omega\text{cm}$ to $1\Omega\text{cm}$. 

1.2 AC electrical conductivity

The frequency dependence of the impedance is a sensitive tool for probing the dispersion and relaxation processes due to polarization and charge transfer, which is
determined by the microstructure of complex systems. A few examples of such studies for both nanometer-scale and micrometer-scale metal-insulator composites have been reported in [5-12]. Most of the studies were conducted on composites where the metal volume fraction $p$ was below the metal-insulator or percolation transition. In view of the new discoveries in the field, it is an important task to study in detail the ac properties of conducting metal-insulator nanocomposites. Impedance for NiFe, Au or Pt with insulating matrix, SiO$_2$ was measured in the range between 0.5 and 0.7. With increasing $p$, the dominating conduction mechanisms change from a thermally activated tunneling with exponential temperature dependence of resistivity typical of insulators, via a transitional region, to quasi-metallic at room temperature. Several pronouncing features in the impedance versus frequency can be attributed to different relaxation processes on different spatial scales or of different origins. The results of composites material, which is close to the metal-insulator transition, are often compared to the predictions of percolation scaling theories.

The most striking result is the divergence of the effective capacitance at low frequency. In our simple model, the low frequency real capacitance is associated with the tunneling correlation length $\xi_t$, which can be much greater than the classical percolation correlation length $\xi_p$. We attribute this behavior to a divergent tunneling correlation length, which can be much greater than the classical percolation correlation length for conductor-insulator networks. For a time-dependent process such as tunneling, the importance of the tunneling correlation length increases at low frequency ($<10^1$ Hz).
Chapter 2

Theoretical Background

2.1 Percolation theory

There are a lot of available theories to interpret the transport properties of granular composite material. For example, the effective-medium approximation (EMA) is a method of determining the effective properties of a disordered medium. It can be used for predicting the AC conductivity of disordered systems that are far from their percolation. Since we are only interested in the scaling behavior of transport properties near $p_c$, in this chapter, we discuss only the percolation, which is widely used in many granular composite materials.

2.1.1 Basic quantities in percolation

Percolation processes were first developed by Flory (1941) and Stockmayer (1943) to describe reaction in small branching molecules react and formation of large macromolecules. In the mathematical literature, the study of percolation was arose by S.R. Broadbent and J. M. Hammerley (1957). They were first dealing with the fluid spreading randomly through a medium. It is found that percolation is a very simple type of mechanism for a critical point where physical properties have singularities.

Bond and site percolation problems are the main concerned problems in classical percolation. For example, sites of the network are occupied with probability $p$ and vacant with probability $1-p$. Two nearest-neighbor sites are called if they are both occupied. A set of connected sites bounded by vacant sites is called a cluster. To determine the transition in the topological structure of the random network from a
disconnected structure to a connected one, there is a important quantities called percolation threshold, $p_c$. This is defined as the largest fraction of occupied bonds (sites) below which there is no sample-spanning cluster. In other words, this is equivalent to the smallest fraction of an infinite cluster existing in the network. In addition to the percolation thresholds, the topological properties of percolation networks are characterized by several important quantities.

Accessible fraction $\lambda^A(p)$, this is the fraction of occupied bonds belonging to the infinite cluster. Backbone fraction $\lambda^B(p)$, inside the accessible fraction, there are bonds characterize the current flow, since some of the bonds in the cluster are dead-end and do not carry any flow. Besides, correlation length $\xi(p)$ is also very important, this is the typical radius of clusters for $p < p_c$. These percolation quantities obey scaling laws that are insensitive to the network structure and its microscopic details.

### 2.1.2 DC conductivity of metal-insulator films

Since then, the concept of percolation has been widely used in metal-insulator composites materials. It was found that some physical quantities such as the effective bulk DC electrical conductivity $\sigma(\omega=0)$ and the DC dielectric constant $\varepsilon(\omega=0)$ of a percolating system should exhibit a power-law dependence on the correlation length $\xi$.

According to percolation theory, there is a percolation threshold concentration $p_c$, and there exists a characteristic correlation length $\xi$ being proportional to $|p - p_c|^\beta$ near $p_c$. This correlation length diverges from either side of the metal-insulator transition.
In fact, these critical behaviors can be predicted by the theoretical analysis on a random resistor network (RNN), which is included in appendix. In network models, the link-nodes-blobs (LNB) picture of Stanley and Coniglio enables a good quantitative estimates on the critical behavior. In this picture, the backbone act as an irregular supernetwork of nodes and links. (A node is any site of the backbone that is connected to the boundary by at least three independent paths and link is the set of backbone bonds between two adjacent nodes.) The average distance between adjacent nodes is the percolation correlation length and it is proportional to $\Delta p = |p - p_c|$ in different dimensions $d$ as:

$$\xi \sim \Delta p^{-\nu},$$

$$\nu = \begin{cases} 
\frac{4}{3} & d = 2 \\
0.89 & d = 3 
\end{cases} \quad (2.1)$$

The average conductance of a link is $g_\xi \sim \Delta p^\zeta$. The average number $N_{\text{SCB}}$ of singly connected bonds (SCB) in a link satisfies $N_{\text{SCB}} \sim l/\Delta p$. Since the number of SCBs has a lower limit on the resistance of a link, $\zeta \geq 1$. Then the LNB can be used to get the conductivity of the network (d is the dimensionality):

$$\sigma_e \equiv g_\xi / \xi^{3-2} \sim \Delta p^{\zeta+2(d-2)\nu} \quad (2.2)$$

The critical exponent $t$ is therefore:

$$t = \zeta + (d-2)\nu \geq 1 + (d-2)\nu.$$  

For a two-component random composite mixture of a good conductor and a bad conductor ($\sigma_m \gg \sigma_i$), the effective conductivity would be:

$$\sigma_e \sim \sigma_m (p_m - p_c)^t$$ for $0 < p_m - p_c \ll 1$, \quad (2.3)

$$\sigma_e \sim \sigma_i (p_c - p_m)^{-s}$$ for $0 < p_c - p_m \ll 1$. 

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The values of \( t \) and \( s \) from calculations on percolating networks are:

\[
\begin{align*}
    t &= \begin{cases} 
        1.30, & d = 2 \\
        2.0 & d = 3 \\
        3 \quad (\text{exact}), & d \geq 6 
    \end{cases} \\
    s &= \begin{cases} 
        1.30, & d = 2 \\
        0.76 & d = 3 \\
        0 \quad (\text{exact}), & d \geq 6 
    \end{cases}
\end{align*}
\]

These power laws are supposed to be universal. The critical exponents depend primarily on the dimensionality of the percolating systems and do not depend on the details of the geometric structures.

There are two limiting cases of the system; the case in which \( \sigma_M \) is finite and \( \sigma_i = 0 \) (conductor-insulator mixtures), and the case in which \( \sigma_M = \infty \) and \( \sigma_i \) is finite (superconductor-conductor mixtures). In fact, there is a crossover in the behavior described by (2.3) breaks down, if \( \sigma_i/\sigma_M \) is not strictly zero, and \( p_M - p_c \) tends to zero. Since \( \sigma_i \) can never truly vanish or become infinite. Instead, the critical behavior can be described by the scaling expression:

\[
h = \frac{\sigma_i}{\sigma_M}
\]

\[
\sigma_c \equiv \sigma_M (\Delta p)^{\beta} f(z),
\]

\[
z = \frac{\sigma_i / \sigma_M}{\Delta p^{\nu + \tau}}
\]

(2.4a)

\[
\Delta p = |p_M - p_c|,
\]

where the scaling function \( f(z) \) has the following asymptotic behavior:

\[
f(z) \equiv \begin{cases} 
    A + Bz + ... & z << 1, \quad p_M > p_c \\
    A'z + B'z^2 + ... & z << 1, \quad p_M < p_c \\
    A''z^{(r+s)} + ... & z >> 1, \quad p_M \approx p_c
\end{cases}
\]

(2.4b)
The scaling behavior is obtained explicitly in the effective-medium approximation and in the mean-field theory of percolation and has been verified in some numerical simulations. One can also easily notice that the effective conductivity is invariant under the interchange of \( \sigma_M \) and \( \sigma_l \), and therefore we can have

\[
\sigma_e (p, \sigma_M, \sigma_l) = \sigma_e (1 - p, \sigma_l, \sigma_M) \quad (2.5a)
\]

\[
f(p, h) = hf (1 - p, 1 / h) . \quad (2.5b)
\]

### 2.1.3 Application of Percolation Theory on Hall Effect

Hall effect is also an important subject in random composite material. By choosing \( H \parallel z \), the conductivity tensor has the form

\[
\sigma = \begin{pmatrix}
\sigma_0 & 0 & 0 \\
0 & \sigma_0 & 0 \\
0 & 0 & \sigma_0
\end{pmatrix} + \begin{pmatrix}
0 & \lambda & 0 \\
-\lambda & 0 & 0 \\
0 & 0 & 0
\end{pmatrix} \begin{pmatrix}
\delta \sigma_\perp & 0 & 0 \\
0 & \delta \sigma_\perp & 0 \\
0 & 0 & \delta \sigma_\parallel
\end{pmatrix} \quad (2.6)
\]

where \( \sigma_0 \) is the conductivity at \( H = 0 \), \( \lambda \) is the Hall conductivity, and \( \delta \sigma_\perp, \delta \sigma_\parallel \) are the transverse and longitudinal magnetoconductivities, respectively. The relationship between the volume averages of \( \bar{E} \) and \( \bar{J} \) can again serve to define the bulk effective conductivity tensor \( \sigma_e \)

\[
\langle J \rangle \equiv \sigma_e \langle E \rangle , \quad (2.7)
\]

where \( \sigma_e \) now depends on four parameters as above. In low value of \( H \), the local electric field allows to use the perturbation method (the three terms of sum in (2.6) are proportional to successively higher powers of \( H \)) and lead to:

\[
\lambda_e = \frac{1}{V} \int dV \lambda(r) \frac{(\bar{E}^{(s)} \times \bar{E}^{(v)})_z}{E_o^2} \quad (2.8)
\]

and (2.8) can be transformed into,
\[
\frac{\lambda_e - \lambda_i}{\lambda_M - \lambda_i} = \frac{1}{V} \int_{V_M} dV \left( \frac{E^{(x)} \times E^{(y)}}{E^2} \right)_x \quad (2.9)
\]

Since the right-hand side of the equation depends only on ohmic properties of the composite, including the ratio of ohmic conductivities \( \sigma_i/\sigma_M \) (it is natural to assume that near \( p_c \), it will depend on the microgeometry through the same scaling variable that appearing in (2.4a). Because the integration in (2.9) is confined to the subvolume \( \sigma_M \), the right-hand side will be \( O(1) \) when \( p_M > p_c \), but \( O(\sigma_i^2/\sigma_M^2) \) when \( p_M < p_c \) (whereas inside the poor conductor \( \sigma_i \) the local field \( \tilde{E}(\tilde{r}) \) is always of order \( E_o \), inside the good conductor \( \sigma_M \), \( \tilde{E}(\tilde{r}) \sim E_o \) only on the percolating backbone.)

Using the relation \( \rho_H = \mu/\sigma^2 \), we can find the weak-field Hall resistivity \( \rho_{He} \):

\[
\rho_{He} \sim \begin{cases}
\rho_{HM} \Delta p^{-k} + \rho_{Hi} (\sigma_i/\sigma_M)^2 \Delta p^{-2t} \\
\rho_{HM} \Delta p^{-k} + \rho_{Hi} \Delta p^{2s} \\
\rho_{HM} (\sigma_i/\sigma_M)^{-k/(t+s)} + \rho_{Hi} (\sigma_i/\sigma_M)^{2s/(t+s)}
\end{cases} \quad (2.10)
\]

where constants of order 1 have been omitted from all terms.

2.1.4 Finite systems and finite-size scaling

We have discussed percolation in infinitely large systems so far. Indeed, the effect due to the finite size of the sample is also important.

For any length scale \( L \gg \xi \), a percolating system is macroscopically homogeneous. But for \( L \ll \xi \), the system is not homogeneous and the macroscopic properties of the system depend on \( L \). In this regime, the sample-spanning cluster is on average self-similar, i.e., it looks the same at all length scales up to \( \xi \), and its mass \( M \) (the total number of bonds or sites) scales with \( \xi \) as
\[ M \sim \xi^{D_c}, \quad (2.11) \]

where \( D_c \) is called the fractal dimension of the cluster. However, \( D_c \) is not a totally new quantity and is given by

\[ D_c = d - \frac{\beta_p}{\nu} \quad (2.12) \]

where \( \beta_p \) is the critical exponent of \( X^p(p) \). For \( L \gg \xi, D_c = d \). Similarly, for \( L \ll \xi \), the backbone is a fractal object and its fractal dimension \( D_{bb} \) is given by

\[ D_{bb} = d - \frac{\beta_b}{\nu} \quad (2.13) \]

where \( \beta_b \) is the critical exponent of \( X^b(p) \). Note that if \( L < \xi \), one should replace \( \xi \) in (2.11) by \( L \). Note also that \( \xi \) is divergent at \( p = p_c \), so that the sample-spanning cluster is a fractal object for any \( L \).

Another important concept in percolation theory is finite-size scaling. Fisher (1971) investigated the effect of finite size of a thermal system on its critical properties near the critical temperature, and developed this scaling. If a quantity \( X \) is predicted to scale as \(|p - p_c|^z \) for sizes \( L \gg \xi \), then we expect it to obey the general scaling law

\[ X(L, \xi) = \xi^{x/u} x_1(L/\xi) \propto \begin{cases} \xi^{x/u} & \text{if } L \gg \xi \\ L^{t/u} & \text{if } L \ll \xi \end{cases} \quad (2.14a) \]

or \[ X(L, p) = (p - p_c)^{x_2} (p - p_c)^{L^{t/u}} \quad (2.14b) \]

The intuitive content of the finite-size scaling law is that the infinitely sharp percolation singularity that affects the conductivity of an infinite sample of the conductor-insulator mixture is rounded off over a small range of values of \( p \) around \( p_c \), of the order of the larger of the two estimates. 

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\[ \delta p_1 \sim h^{1/(\sigma + r)}, \quad \delta p_2 \sim L^{-1/v} \quad (2.15) \]

\(\delta p_1\), already given in (2.4), is a measure of the cross-over due to the finiteness of the conductivity (impedance as well) ratio \(h\), whereas \(\delta p_2\) corresponds to finite-size effects on the geometrical percolation problem.

### 2.1.5 Complex Dielectric Function

Before we start to discuss the ac properties of composite material, complex dielectric function (or complex conductivity) is an important concept to be understand. The transport of electromagnetic waves through a binary composite is described by the macroscopic Maxwell equations. Assuming monochromatic fields of the form \(\vec{E}(\vec{r}, t) = \vec{E}(\vec{r}) \exp(-i\omega t)\), and adopting the usual convention that the physical fields are the real parts of complex quantities, these take the form (in MKSA units)

\[ \nabla \cdot (\varepsilon' \varepsilon_0 \vec{E}) = \rho, \quad (2.16) \]

\[ \nabla \cdot \vec{B} = 0, \quad (2.17) \]

\[ \nabla \times \vec{E} = \frac{i\omega}{c} \vec{B}, \quad (2.18) \]

\[ \nabla \times \vec{H} = \sigma' \vec{E} - i\omega \varepsilon' \varepsilon_0 \vec{E}, \quad (2.19) \]

which are to be supplemented by the continuity equation

\[ \nabla \cdot (\sigma' \vec{E}) - i\omega \rho = 0 \quad (2.20) \]

Substituting (2.20) into (2.16) gives

\[ \nabla \cdot \vec{D} = 0, \quad (2.21) \]

where the free current density and polarization current have been combined into single effective displacement field.
\[ \vec{D} = (\varepsilon' \varepsilon_\infty + \frac{i \sigma'}{\omega}) \vec{E} \]  

(2.22)

With introduction of a complex dielectric function

\[ \varepsilon(\vec{r}, \omega) = \varepsilon_\infty \varepsilon'(\vec{r}, \omega) + \frac{i}{\omega} \sigma'(\vec{r}, \omega) \]  

(2.23)

2.1.6 Predictions on AC conductivity

Recently, there has been interest in the AC conductivity \( \sigma(\omega, p_c) \) and the dielectric constant \( \varepsilon(\omega, p_c) \). Since we aim at studying frequency-dependent AC electrical properties, the elementary conductances of the bonds will in general be considered as arbitrary complex 'admittances'. We shall nevertheless still call them conductances. The only restriction on the values \( \sigma_\text{M}(\omega) \) and \( \sigma_\text{I}(\omega) \) of both types of conductances comes form their nature as passive dipoles: they are bound to dissipate electrical power. Since the amount of loss in a conductor in a steady ac regime is proportional to the real part of its conductance, we shall assume that the conditions

\[ \text{Re}(\sigma_\text{I}) \geq 0, \quad \text{Re}(\sigma_\text{M}) \geq 0 \]

are satisfied. The limiting cases of elements without losses, such as perfect inductance coils and capacitors, correspond to purely imaginary conductances, \( \sigma = (iL\omega)^{-1} \) and \( \sigma = iC\omega \) respectively.

The results from section 2.1.2 can now be used for modeling the AC conductivity and dielectric properties of a disordered composite near \( p_c \). Many authors have studied this problem, and among the earliest ones we should mention Efros and Shklovskii (1976), and Bergman and Imry (1977).
Consider a percolation network in which a fraction $p$ of the bonds are purely resistive, while a fraction $q = 1 - p$ of the bonds behave as perfect capacitors. Thus we set $\sigma_m = \frac{1}{R}$, $\sigma_l = iC\omega$, where $R$ is the resistance of the bond, $C$ is the capacitance, $\omega$ is the frequency. The conductance ratio $h$ is then given by

$$ h = \frac{i\omega}{\omega_0}, $$

(2.24)

where $\omega_0 = 1/(RC)$. This model is usually referred to as the R-C model. The key result is that if $p$ is close to $p_c$, the R-C model possess scaling properties. Replacing $\sigma_l = 0$ by $\sigma_l = iC\omega$, we get from (2.4)

$$ \sigma_e \sim \frac{1}{R} \left| p - p_c \right|^\nu \int \left( \frac{i\omega}{\omega_0} \left| p - p_c \right|^{-\nu} \right) $$

(2.25)

The real and imaginary parts of $\sigma_e$ give us directly the critical behavior of the (real) bulk effective conductivity and the (real) bulk effective dielectric constant $\sigma_e$. In particular, the dielectric constant is predicted to diverge as $\Delta p^x$ when $p_c$ is approached from either side, and at $p_c$ both Re($\varepsilon$) and Re($\sigma$) acquire a peculiar frequency dependence.

By using general analytic properties of the effective complex dielectric constant of a random composites, Bergman and Imry (1977) derived the following scaling relations, which can be obtained by substitute $\varepsilon(p, \omega) = \frac{\sigma_e}{i\omega}$ to (2.25)

$$ \sigma_e \sim \omega^x, $$

(2.26)

$$ \varepsilon_e \sim \omega^{-y}, $$

(2.27)

where the sum of exponent $x$ and $y$ is one. This is a direct consequence of the fact that a complex conductivity is an analytic function of $i\omega$. Bergman and Imry argued that
the main contribution to the AC properties is due to polarization effects between various percolation cluster and proposed that

\[ x = \frac{t}{t + s}, \]  
(2.28)

\[ y = \frac{s}{t + s}, \]  
(2.29)

2.2 Magnatotransport Properties in Ferromagnetic Metal

2.2.1 Giant Magneto Resistivity

Magnetotransport properties starts to attract attention in the last decade since the discovery of the giant magnetoresistance (GMR) in magnetic multilayers, such as Fe/Cr or Co/Cu [15-18]. Subsequently, the GMR was also observed in immiscible magnetic granular composite films with a metallic matrix, such as Co-Cu and Fe-Ag or with an insulating matrix such as Co-Al₂O₃, when the volume fraction of the magnetic component \( p \) is below the percolation threshold \( p_c \). It is well believed that the GMR in such inhomogeneous system results from a spin-dependent scattering.

The mechanisms of electronic transport in metallic and dielectric granular magnets are different and the magnetoresistance effects were so far treated separately.

Ferromagnet-insulator mixtures

Following the early experiments of Gittleman, Goldstein, and Bozowski[13] on discontinuous Ni films, the magnetoresistance of ferromagnet-insulator mixtures
was derived by Helman and Abeles [14] in the framework of the tunneling theory of spin-polarized electrons. Here, the probability of an electron tunneling across an intergranular barrier was taken account of additional magnetic exchange energy arising when the magnetic moments of the neighboring grains are not parallel and electron spin is conserved in tunneling. In this model, the conductivity of the system is proportional to the product of the nonmagnetic and the spin-dependent terms. Magnetoresistance is defined as \( \Delta \rho / \rho = -[\sigma(H,T) - \sigma(0,T)]/\sigma(H,T) \), and by keeping only linear terms in the magnetic energy \( E_m \) the normalized magnetoresistance can be approximated by

\[
\frac{\Delta \rho}{\rho} = \frac{[E_m(H) - E_m(0)]P}{2kT},
\]

(2.30)

where \( P \) is polarization of the tunneling electrons. The magnetic exchange energy \( E_m \) can be expressed in terms of the spin correlation function of two neighboring grains. In the case of the ferromagnetically coupled grains the magnetoresistance is given by

\[
\frac{\Delta \rho}{\rho} = -(JP/4kT)[M^2(H,T) - M^2(0,T)],
\]

(2.31)

where \( J \) is the exchange coupling constant and \( M \) is magnetization. Within this model the normalized magnetoresistance does not depend on the total resistivity of the sample.

*Metallic-magnetic system*

In metallic magnetic systems the GMR is regarded as the extra electrical resistance due to scattering from nonaligned ferromagnetic entities. At finite temperatures, the fractional GMR is given by
\[ \frac{\Delta \rho}{\rho} = -\rho_m(T)/[\rho_o + \rho_{ph}(T) + \rho_m(T)] F(M/M_s), \]  

(2.32)

where \( \rho_o \) is the temperature independent contribution from defects, \( \rho_{ph}(T) \) is the phonon contribution, and \( \rho_m \) is the magnetic resistivity responsible for GMR. \( F(M/M_s) \) is usually well described by \( F = \alpha(M/M_s)^3 \). Although both magnetic and nonmagnetic resistivities depend on the density of charge carriers, it is not a priori obvious that the ratio \( \Delta \rho/\rho \) is independent of the total resistivity value in the range from \( 10^{-5} \) to \( 10^3 \, \Omega \text{cm} \) [1-3].

### 2.2.2 Hall effect in bulk ferromagnetic materials

![Diagram of Hall effect resistivity vs. field](image)

**Fig. 2.1 Typical shape of the Hall resistivity as a function of field in bulk ferromagnetic material.**

The Hall effect in bulk ferromagnetic materials is fairly understood and fig. 2.2.1 shows a typical shape of the Hall resistivity as a function of field. A steep increase at low fields followed by a slow variation at higher fields. This is usually described by a phenomenological equation [4]:

\[ \rho_{xy} = 2 \rho_{sys} \]

slope = \( R_o \)

\[ 2 \rho_{sys} \]

H
\[ \rho_H = R_o B + R_s M, \quad (2.33) \]

where the two terms are the ordinary Hall effect proportional to the magnetic field and the extraordinary Hall effect proportional to the magnetization. \( R_o \) describes the number of conduction carriers per atom as non-magnetic system. The additional term \( R_s \) is called the extraordinary Hall coefficient (EHE) which is characteristic of magnetic materials. This is the term characterize the magnetic materials in the Hall resistivity. Moreover, \( R_s \) is usually much larger than \( R_o \) and more sensitive to the temperature. The Hall resistivity \( \rho_H \) has been widely studied and results can be generalize as:

\[ \rho_H \propto (\rho(T))^n, \quad (2.34) \]

where the power \( n \) varies between 1 and 2 depending on the material and temperature range used in the experiment (which is very different for our result). It is suggested that this kind of dependence is related with electron scattering.

It is already seen from the relationship (2.33) that a theory of the EHE should reflect the interconnection between the magnetization (electron spin) and transport properties of the material, i.e., spin-orbit interaction should play a role in the EHE effect. Let us consider an electron approaching a scattering potential. For a symmetric scattering potential the direction of the electron motion is unchanged. Spin-orbit interaction, however, results in asymmetry between right and left, which leads to the trajectory after scattering differing from the one before scattering. The new trajectory may be at the direction of angle \( \theta \) with respect to the old one (fig. 2.2.2a). The electron acquires transverse momentum from scattering and the effect is called skew scattering. The new trajectory may also be displaced by a finite distance from the old trajectory (fig. 2.2.2b). This transverse displacement is called side jump. Detailed
calculations show that the side-jump per collision is independent of the strength, range, or sign of the scattering potential and is predicted to be $10^{-11} - 10^{-10}\text{m}$. This value is large enough to account for the experimental data of the Hall effect in ferromagnetic materials like Fe or Ni. Furthermore, the side-jump mechanism predicts $n = 2$. The skew scattering alone, predicting $n = 1$, cannot explain the EHE of ferromagnets above 100K, where the side jump mechanism appears to be dominant.

\[ \rho_{yy} \propto \rho_{xx}^n \]

Fig. 2.2 Mechanism of extraordinary Hall effect in: (a) skew scattering, which is left-right asymmetry in scattering cross section with $n = 1$, and (b) side jump, which believed as quantum mechanical effect with $n = 2$. 
Chapter 3

Experimental Techniques

3.1 Sample preparation:

3.1.1 Film Deposition

Granular metals can be made by co-sputtering, co-deposition, laser ablation, co-evaporation and other methods. Sputtering has been demonstrated to be the most versatile. In sputtering, one can employ a variety of target configurations: two homogeneous targets, co-sputtering from two separated targets, or a composite target with slabs of materials pieced together. The co-sputtering method was adopted in this case. A metal target (e.g. Ni, Pt, Au) and a SiO$_2$ target were used in conjunction with magnetron sputtering whose schematic configuration is shown in Fig. 3.1. All the samples were deposited by rf-sputtering with chamber pressure about $10^{-7}$ torr with the substrate temperature at room temperature. The film thickness was kept at 0.5-1µm for all films. By alternating the sputtering rate of metal target and keeping the constant sputtering rate of insulating target, we can vary the metal volume fraction.

![Schematic diagram of Denton magnetron sputtering system](image)

*Fig. 3.1 Schematic configuration of Denton magnetron sputtering system*
3.1.2 Composition analysis

The deposition rate of metal granules is generally different from that of the matrix material in which they are imbedded. The metal volume fraction in our case is quite different from the nominal value calculated by assuming linear relation between the rate and the power. In our experiment, the metal volume fraction was controlled by the relative ratio of plasma powers on the metal and insulator targets, and was determined using energy-dispersive x-ray analysis (EDX) (Philips EDAX XL30). The EDX techniques were carried out for all films with Kapton substrates. For quantitative analysis, pure elemental gold, platinum, nickel, iron, silicon were used as standards. The results in atomic percentage were transferred to volume fraction by using atomic volume of 6.8cc/mol for Ni, 9.1cc/mol for Pt, 10.2cc/mol for Au and 6.6cc/mol for SiO₂. The nominal value and the corresponding determined value metal volume fraction p in all deposited films were listed in following table.

Table 3.1 NiFe-SiO₂, Pt-SiO₂ and AuSiO₂ co-sputtered films nominal sputtering rate and the metal volume fraction by EDX.

<table>
<thead>
<tr>
<th>Pt vol %</th>
<th>nominal</th>
<th>Au vol %</th>
<th>nominal</th>
<th>NiFe vol %</th>
<th>nominal</th>
</tr>
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<tr>
<td>35.23</td>
<td>45</td>
<td>57.75</td>
<td>72.50</td>
<td>51.60</td>
<td>39</td>
</tr>
<tr>
<td>34.56</td>
<td>50</td>
<td>56.36</td>
<td>75.00</td>
<td>51.83</td>
<td>40</td>
</tr>
<tr>
<td>34.66</td>
<td>55</td>
<td>60.73</td>
<td>77.50</td>
<td>54.72</td>
<td>41I</td>
</tr>
<tr>
<td>40.12</td>
<td>60</td>
<td>59.84</td>
<td>82.50</td>
<td>53.16</td>
<td>42</td>
</tr>
<tr>
<td></td>
<td></td>
<td>66.97</td>
<td>85.00</td>
<td>57.56</td>
<td>43</td>
</tr>
<tr>
<td></td>
<td></td>
<td>63.93</td>
<td>87.50</td>
<td>60.57</td>
<td>44</td>
</tr>
<tr>
<td></td>
<td></td>
<td>68.87</td>
<td>90.00</td>
<td>63.31</td>
<td>45II</td>
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<tr>
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<td></td>
<td>60.85</td>
<td>80.00</td>
<td>63.51</td>
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</tr>
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<td>54.90</td>
<td>70.00</td>
<td>63.72</td>
<td>47</td>
</tr>
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<td>60.00</td>
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</tr>
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<td></td>
<td></td>
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<td></td>
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</tr>
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<td></td>
<td></td>
<td></td>
<td></td>
<td>83.62</td>
<td>80</td>
</tr>
</tbody>
</table>
3.1.3 Distribution size of the metallic granules

The distribution of sizes $\Phi$ of metal particles for NiFe-SiO$_2$ films has been studied by Transmission Electron Microscopy in [4,20]. The distribution functions for three of the samples are shown in Fig. 3.3 [4]. The composites consist of very small crystalline metal granules (the minimum diameters down to less than 1 nm) immersed in the amorphous insulating matrix. The granule sizes $\Phi$ follow approximately an exponential distribution. A considerable fraction of metal may be probably present in a form of dissolved impurity atoms.

![Graph showing distributions of particle sizes $\Phi$ in (NiFe)-SiO$_2$ films. Lines are the fits to $A \exp[-\Phi/\Phi_0]$ where $\Phi_0$ of $p=0.63, 0.65$ and $0.72$ is found to be 14.7, 15.7 and 24.3 respectively.]

*Fig. 3.2 Distributions of particle sizes $\Phi$ in (NiFe)-SiO$_2$ films. Lines are the fits to $A \exp[-\Phi/\Phi_0]$ where $\Phi_0$ of $p=0.63, 0.65$ and $0.72$ is found to be 14.7, 15.7 and 24.3 respectively.*
3.2 Measurement techniques:

3.2.1 DC resistivity measurement

For the DC resistivity measurement, two kinds of configuration were applied. The first one was conventional four-probe resistance measurements. The films were performed as functions of temperature and magnetic field as shown schematically in Fig. 3.3. A Keithley-model-236 source measure unit given a constant current (10nA–100mA) to the film and a high precision Keithley-model-182 nano-voltmeter was used to record the voltage difference between the two inner leads of the sample. All the equipment were controlled by a computer through a GPIB card. In an effort to avoid the error from thermal gradient or geometry, the resistivity was measured in both positive and negative current. The advantage of this method is that can provide a high accuracy of resistance under the field in a particular sample, however the geometrical factor can affect the results versus different samples.

Fig. 3.3 Schematic representations of circuit used to measure the resistance of the film.
Therefore, van der Pauw's method, which is mentioned below, was applied to determine the resistivity without knowing the geometrical factor. Bio-Rad-model-HL5500PC Hall effect measurement system was also used in measuring of resistivity in both nitrogen temperature and room temperature. Samples of size 1cm × 1cm square with four indium contact about 0.05 mm was measured. The experimental arrangement for a resistivity measurement using van der Pauw's method is shown in Fig. 3.4. The resistivity is then give by:

\[
\rho = \frac{\pi \cdot d}{2 \cdot \ln(2)} \left[ \frac{V_{43}}{I_{12}} + \frac{V_{23}}{I_{14}} \right] \cdot F(Q) \quad (\Omega\text{-cm})
\]

\[
Q = \frac{V_{43} \cdot I_{14}}{I_{12} \cdot V_{23}}
\]

where \( t \) is the thickness of the active layer, \( Q \) and \( F \) the symmetry and correction factors respectively. More detail of correction factors is shown in Ref [19].

![Fig. 3.4 Resistivity measurement using van der Pauw's method](image-url)
3.2.2 Impedance measurement

Rectangular samples with length of 10-20 mm and width 1-2 mm was used. Four 1 cm long, 0.05 mm thick gold or platinum wire terminals were attached to the sample surface with the silver paste or pressed with indium. We found that contacts can introduce considerable complex impedance. So the drive and sense contacts were well separated shown in Fig. 3.5. The measurements were done on an HP 4284A and HP 4285 LCR meter with frequency range 20-1M Hz and 75k-30M Hz respectively. Standard “short” and “open” corrections were use to eliminate the effects of parasite impedance. To confirm the results, we also tried measurements on samples with different distances between the sense terminals.

![Diagram](image)

*Fig. 3.5 Schematic representations of circuit used to measure the impedance of the film.*
3.2.3 Hall resistivity measurement

Five-probe Hall resistance measurement of the films as functions of magnetic field and temperature were performed using a DC bridge arrangement shown in Fig. 3.6. By choosing a suitable current, the potentiometer is adjusted so that there is zero voltage detected in zero field, the Hall voltage generated by the applied magnetic field is measure between the two sides of the film. The value of variable bridge resistance was chosen at least ten times larger than that of the sample resistance in order to reduce any experimental errors.

*Fig. 3.6 Schematic representations of circuit used to measure the Hall resistance of the film.*
Chapter 4

Results and Discussions

4.1 Resistivity, Hall effect and magnetoresistivity

In granular systems, the overwhelming majority of the studies on the GMR have been focused on the ferromagnet-metal granular system, whereas only a few work was done on the ferromagnet-insulator granular systems. Follow the works on NiFe-SiO$_2$ [3], where measured both $\rho_{xy}$ and $\rho_{xx}$ were measured in liquid Helium temperature, we have measured both quantities at 300K in order to see the dependence of temperature. Fig. 4.1 is the plot of $\rho_{sys}$, $\rho_{sys}$, and $\rho_{xx}$ versus its metal volume fraction at 5K and 300K respectively.

![Graph showing resistivity as a function of metal volume fraction]

*Fig. 4.1* The saturated extraordinary Hall resistivity $\rho_{sys}$, which is determined by extrapolation to $H=0$, ordinary Hall resistivity $\rho_{sys}$ at saturation field $H = 0.6T$, and zero field resistivity $\rho$ as functions of NiFe volume fraction.
Following Ref. [4], we define the percolation threshold $p_c$ as a concentration below which DC resistivity of the samples follows an exponential dependence on temperature: $\rho \propto \exp[-T_c/T]$ [20]. The values of $p_c$ were determined to be in the range 0.5 - 0.55 for different compositions of the films. We can easily observed that samples with $p > p_c$ do not behave as pure metals (Fig. 4.2). The temperature coefficient of resistivity (TCR) is negative in a wide range of temperatures, which extends to room temperature for samples close to the percolation threshold. It is believed that the negative TCR may be related to a contribution of tunneling junctions to conduction.

![Fig 4.2(a) Temperature dependence of resistivity for Au-SiO$_2$ and Pt-SiO$_2$ samples normalized at 5K.](image-url)
Fig 4.2(b) Temperature dependence of resistivity for (NiFe)-SiO$_2$ samples normalized at 5K.

### 4.2 Phenomenological Model

Physical model available to describe such kind of metal-insulating granular composite is limited. Percolation model was usually used in many previous works. However, it cannot explain very well on the critical exponent, because those found in experiment are several times larger than that predicted by the model. To interpret this result, a phenomenological model was proposed and used to explain these discrepancies.

It is found that the universal behaviors associated with giant Hall effect GHE [3] observed in NiFe-SiO$_2$ and Fe-SiO$_2$ are most likely related to their common percolating granular structure, even they have very different microscope characters (e.g. with different charge carriers)

According to [4], NiFe-SiO$_2$ and SiO$_2$ films exhibit -log(T) like dependence for annealing below 300°C, whereas its becomes metallic above 300°C. The change of granule size was accounted for this. Moreover,
magnetoresistivity of these samples have the same negative value for films near the percolation threshold. As mentioned in Chapter 2, the magnetoresistance is a result of spin-dependent transport. Fig. 4.3(b) shows the magnetoresistivity $\Delta \rho$ versus resistivity in a log-log scale for Ni-rich NiFe-SiO$_2$ (filled circle) and Fe-SiO$_2$ (open circle) samples measured at 300K. (The value of Fe-SiO$_2$ $\Delta \rho$ was divided by 4 to take into account the difference in the saturation magnetization because $\Delta \rho$ scales with $M^2$ for a spin-dependent process.) Dashed line in this figure is the fit of $\Delta \rho \sim \rho^b$, with $b = 1.2$. It shows that within experimental error the same power law fit both NiFe-SiO$_2$ and Fe-SiO$_2$ systems, which indicates that they have the similar spin-dependent transport.

![Graphs showing temperature dependent part of resistivity and magnetoresistivity](image)

**Fig. 4.3** (a) Temperature dependent part of resistivity, $\Delta \rho'$, versus $\rho$ for NiFe-SiO$_2$ in a log-log plot. (b) Magnetoresistivity, $-\Delta \rho$, as functions of resistivity, $\rho$, for NiFe-SiO$_2$ (filled circle) and Fe-SiO$_2$ (open circle).
mechanism, irrespective to the sign of the carriers and the details of the microstructures.

In an effort to further investigate the universal behaviors, we should know the condition that the particle of size $s^3$ can be considered as to be conducting. In general, a metallic particle can be considered as conducting if there is no energy barrier against transporting electrons in and out of the particle. In view of this, we can consider the thermal energy $\sim k_B T$ of electrons, which is larger than the energy spacing inside the particle, $\Delta E(s) \sim \frac{\hbar^2}{2m' s^2}$, where $m'$ is the electron effective mass, or $\Delta E(s) \sim \frac{e^2}{\epsilon s}$, where $\epsilon$ is the effective dielectric constant, if Coulomb blockade effect is important. Fig. 4.4 shows that in our granular systems there exist a large number of small particles smaller or equal to 3nm, therefore, it is expected that the finite size effect would be important. It can be easily expected that this large amount of small particles behave as metals at high temperature and would become insulating at temperature $k_B T \leq \Delta E(s)$, and the effective conducting volume fraction decreases with decreasing temperature, which leads to increasing resistance at low temperature. In the case when the particles are magnetized and with magnetization pointing in random directions, there is an additional energy barrier between particles with magnetization pointing in opposite direction, and these particles become insulating with respect to each other. This energy barrier is removed when the magnetization of the particles are aligned, as is in the case when a large magnetic field is applied, leading to an increase in effective volume fraction and negative magnetoresistance. Due to this kind of
effective volume fraction change, we assume that effective classical percolation models near percolation threshold can describe the transport properties of the magnetic granular systems under consideration, where the resistivity is given by the formula

\[ \rho(T,H) = \frac{\rho_o}{(p_{\text{eff}}(T,H) - p_c)'}, \]  

(4.1)

which has similar form mentioned in chapter 2, whereas resistivity \( \rho \) is used instead of conductivity \( \sigma \). The critical exponent \( t \) characterizing the effective classical percolation model, \( p_{\text{eff}}(T,H) \) is the effective conducting volume fraction. \( H \) is the external magnetic field and \( B = H + 4\pi M \) is the total magnetic field seen by electrons. In particular, we expect from the discussion above at zero external magnetic field the effective volume fraction:

\[ p_{\text{eff}}(T,0) \sim \int_0^T (s^3)n(s)ds = p_o - \int_0^T (s^3)n(s)ds, \]  

(4.2)

where \( n(s)ds \) is the number of particles in the sample with linear dimension between \( s \) and \( s + ds \), \( p_o = \int_0^T (s^3)n(s)ds \) is the metal volume fraction determined by EDX. \( s(T) \) is a size cutoff determined by the equation \( k_B T = \Delta E(s) \). At finite magnetic field, we expect that \( p_{\text{eff}} \) increases because of the alignment of magnetization between different particles. The precise magnetic field dependence of this effect is hard to estimate. However, at \( H > H_s \) where magnetization saturates, the magnetic field dependence should vanish and we expect

\[ p_{\text{eff}}(T,H > H_s) \sim p_{\text{eff}}(T,0) + \Delta p_M(T), \]  

(4.3)
where \( \Delta \rho_m(T) \sim A \int_{\lambda(T)}^\infty (\lambda s^3) n(s) ds \), and \( A \) is an unknown constant depending on the microscopic details of the material, \( \lambda \sim \) thickness of the magnetic domain wall separating particles with opposite magnetization orientation.

Assuming the change in \( p_{\text{eff}} \) is small, we can substitute Eq (4.2) into Eq (4.1), and the resistivity of zero magnetic field become

\[
\rho(T,0) \sim \rho_\infty \left( 1 + t \left( \frac{\rho_m}{\rho_o} \right)^i \int_{\lambda(T)}^\infty (s^3) n(s) ds \right) \tag{4.4}
\]

where \( \rho_\infty = \frac{\rho_o}{(p_o - p_e)^i} \) is the high temperature resistivity. By differentiate Eq (4.4) with respect to \( T \)

\[
\frac{d \rho(T,0)}{dT} = t \rho_\infty \left( \frac{\rho_m}{\rho_o} \right)^i \frac{1}{dT} ds(T) (s(T))^3 n(s(T)) \tag{4.5}
\]

which provides a precise relation between \( d\rho(T,0)/dT \) and particle size distribution \( n(s(T)) \) which can be tested experimentally. In particular, at the temperature region with -log(T) depends, we find correspondingly \( n(s) \sim s^{-d} \).

This kind of behavior is independent of the origin of \( \Delta E(s) \) and stays the same as long as \( \Delta E(s) \sim s^{-\alpha} \), with \( \alpha \geq 1 \). Rapid increase in \( n(s) \) as \( s \) decreases was indeed observed experimentally for samples with -log(T) dependent. According to [4], if the samples are annealed, the -log(T) behavior will be absent, it also found that the rapid increase in \( n(s) \) at small \( s \) is destroyed and the peak in \( n(s) \) was gradually shifted to higher values of \( s \) when annealing temperature is increased. The same qualitative behavior can also been obtain from using
experimental data \( \rho(T,0) \) into eqt (4.5). Fig. 4.4 shows the particle size distribution \( n(s) \) for (NiFe)_x-(SiO_2)_{1-x} films (a) as-deposited, and (b) annealed as 520°C. The solid lines are result of TEM measurement reproduced from Ref. 3,

![Particle size distributions](image)

**Fig 4.4** Particle size distributions \( \times 10^{11} / \text{mm}^2 \) determined by TEM and from using temperature dependent resistivity data for (a) (NiFe)_{0.55}-SiO_2 at 0.45 as deposited (O(TEM), \( \nabla \) (theory)), and (b) (NiFe)_{0.55}-SiO_2 annealed at 520°C ((TEM), \( \Delta \) (theory)).

whereas dash-lines are results obtained from the corresponding resistivity data using eqt(4.5) with \( \Delta E(s) = \frac{h^2}{2m_e s^2} \), where \( m_e \) is the free electron mass. The qualitative agreement between theory and experiment is apparent.

Other than resistivity changes due to temperature, using eqt(4.1) with eqt(4.3), it can also show that the saturated magnetoresistivity \( \Delta \rho \) is given by
\[
\Delta \rho = -t \rho \left( \frac{\rho_{\infty}}{\rho} \right)^{\frac{1}{t}} \Delta \rho (T) \tag{4.6}
\]

and
\[
\Delta \rho \sim (\rho_{\infty})^{b} \tag{4.7}
\]
where \( b = 1 + t^{-1} \).

In an effort to get the exponent \( b \), we can use 300K as the high temperature limit (\( \rho \rightarrow \rho_{\infty} \)). According to Fig. 4.5(a), \( t \) is similar to 4.2 \( \pm \) 0.5, which is consistent with experimental values of \( b \sim 1.2 \pm 0.2 \) within experimental accuracy. Also, eqn(4.5) can be modified to have similar form as (4.7) at \( H = 0 \). To avoid confusion, \( \Delta \rho' \) is used to indicate the resistivity changes at given \( T \) from high temperature, which define as:
\[
\Delta \rho' = \rho(T,0) - \rho_{\infty} \sim (\rho_{\infty})^{1+\frac{1}{t}} \tag{4.8}
\]

It is not surprise that both eqn(4.7) and eqn(4.8) have the same exponent \( b \) near percolation in this phenomenological model. Fig. 4.3 gives the temperature dependent part of resistivity \( \Delta \rho \) versus resistivity \( \rho(300K) \) in a log-log scale for (NiFe)$_{\lambda}$-(SiO$_{2}$)$_{\lambda}$ samples. Note that \( \rho(T,0) \) was taken at LN$_{2}$ (77K) temperature and only samples with \( \rho \sim 0.4\Omega cm \) or above were shown, because lower resistivity samples have minimum in the temperature dependence. It is found that this scaling behavior of \( \Delta \rho \) and \( \Delta \rho' \) versus \( \rho \) is apparent from the Fig. 4.3(b) and Fig. 4.3(a). It is pointed out that the temperature dependence of resistivity and magnetoresistance are usually results of very different physical processes, however, they result have very similar critical exponent. This observation supports our physical picture of temperature
and magnetic field dependent of effective conduction volume fraction, which dominates the physics of percolating magnetic nanostructures.

Note that our phenomenology in describing the dependencies of

\[ \rho_{xx}, \rho_{xy}(\Omega \text{cm}) \]

![Graph showing saturated Hall resistivity and ordinary Hall resistivity at 0.6T as functions of resistivity in 300K. Resistivity is altered by changing the metal volume fraction (p = 0.53, 0.55, 0.58, 0.61, 0.64, 0.78 and 0.84).](image)

resistivity on the temperature, the magnetic field, and the particle size distribution, in various percolating magnetic nanostructures, is reasonably successful. This leads us to ask the question of whether similar phenomenology can be applied to describe the Hall resistivity, and in particular, whether the physical origin of the GHE can be identified from our phenomenology.
Fig. 4.6 (a) The zero field resistivity $\rho(300K)$ for (NiFe)-(SiO$_2$) films with different $p$ as a function of $\Delta \rho = |\rho - p_c|$ and $p_c$ here is equal to 0.53. (b) The extraordinary Hall resistivity $\rho_{sys}$ (filled circle) obtained by extrapolation to $H=0$, ordinary Hall resistivity $\rho_{xyo}$ (open circle) obtained at $H=0.6T$ as functions of $\Delta \rho$.

Following our previous analysis, the Hall resistivity can be described by the formula

$$\rho_{xy}(T, H) = \frac{BR_o}{(p_{eff}(T, H) - p_c)^g} \quad (4.9)$$

where $R_o$ is a material dependent parameter and $B = H + 4\pi M$ is the total magnetic field seen by electrons in the system. The Hall number $R_{hi} \sim \rho_{xy}/H \sim (p_{eff}(T, H) - p_c)^g$ and extraordinary resistivity $\rho_{sys} \sim M/(p_{eff}(T, H) - p_c)^g$ are predicted to scale with $\rho(T, H)$ with the same exponent $\frac{g}{t}$ near the percolation
threshold in our phenomenology. Fig. 4.5 shows $\rho_{xx}$ against $\rho_{xxy}(0.6T)$ and $\rho_{sys}$ at $T = 300K$ in the log-log plot. The dash line is the fit of $-\rho_{xx} \propto \rho^n$ or $-\rho_{xxy} \propto \rho^n$, this model is consistent with existing data where $g/t \sim 0.7$ within the experimental accuracy. It can be predicted that scaling between two physical quantities are much more reliable, because it is insensitive to the determination of both the metallic volume fraction or its critical value separating the metal-insulator phases. This is particularly important for the percolating nanostructures where the critical value is not sharply defined as in the classical percolation system. Fig. 4.6(a) shows $\Delta \rho$ against $\rho(300K)$ in log-log plot for sample near percolation and Fig. 4.6(b) is $\Delta \rho$ against $\rho_{xxy}$ and $\rho_{sys}$. The critical exponent of $t$, $g_n$ and $g_s$ are 4.3, 2.9 and 3.3 respectively, which is slightly small than those found at 5K[4]. One can deduced that $g/t \sim 0.7$, which agree with our phenomenology, by the experimental results. Perhaps, these values are much larger than the corresponding values obtained from usual classical percolation models [13] $t \sim 2$ and $g \sim 0.4$, which suggested that the effective classical percolation models describing our experimental system are rather unusual. In particular, the large enhancement of the critical exponent $g$ is responsible for the large magnitude of Hall resistance observed near the percolation threshold, whereas the relatively smaller enhancement in $t$ seems to be the reason why the magnetoresistivity is not as drastic as the Hall effect. This is also suggested that the AC measurement given an insight of explaining smaller enhancement in $t$ but relative large in $g$. 
4.3 Impedance measurements in Ni-SiO₂, Pt-SiO₂ and Au-SiO₂

As we shown in Fig. 4.2, samples with \( p > p_c \) do not behave as pure metals. The temperature coefficient of resistivity (TCR) is negative in a wide range of temperatures, which extends to room temperature for samples close to the percolation threshold. As both weak localization and electron-electron interaction effects in metals are expected to be important only at very low temperatures, we predicted that the negative TCR may be related to a contribution of tunneling junctions to conduction.

In order to investigate this, the effective capacitance (\( \tilde{C} = C' + iC'' \)) of the samples were measured. The values of parallel capacitance (\( C_p = C' \)) and resistance (\( R_p = \frac{1}{2\pi f C''} \)) were measured in the parallel equivalent circuit mode at room temperature. We applied a geometry which is not conventionally used, which is perpendicular to the sample plane as shown in Fig. 3.5, for dielectric measurements, because of the high conductivity. This would result of no direct estimation of dielectric constant, instead we assume that real capacitance \( C' \) is proportional to the real part of dielectric constant. The apparent imaginary part of capacitance \( C'' \) contains contributions from both the true loss from dielectric and DC conductance, which are usually not distinguishable experimentally [21].

Fig. 4.7 is the plot of \( C' \) as a function of frequency on a double logarithmic scale for samples (NiFe)-SiO₂, whereas Fig. 4.8 is the plot of Au-SiO₂ and Pt-SiO₂, with \( p > p_c \). One can observed that the real capacitance decreases in the whole range of measurement and three different types of
behavior with frequency changes. At frequencies greater that $f_0 \sim 10^2 - 10^3$ Hz, $f_0$ here is referred to the cutoff frequency, but smaller than $f_c$, where $f_c$ is the crossover frequency, $C'$ is almost constant between this. The values of $f_0$ and $f_c$ are marked with arrows in Fig. 4.7. For the samples closest to the transition, a considerable change in the slope of the curves is observed near $f_c$. The crossover frequency is of the order of $10^5$ Hz for $p = 0.55$ and $p = 0.58$; it reaches $\sim 10^6$ Hz for $p = 0.63$, and is out of the measurement range for other samples. We approximated the behavior of the real capacitance at $f > f_c$ with a power law dependence of the form $C' \sim f^{-y}$. The estimations of the critical exponent $y$ give the values of $y \approx 0.21$ (at $f \sim 10^6$ Hz for $p = 0.55$), $y \approx 0.41$ (at $f \sim 10^6$ Hz for $p = 0.58$), and $y \approx 0.23$ (at $f \sim 10^7$ Hz for $p = 0.63$). The cutoff frequency $f_0$ is of the order of $10^2$ Hz for two samples closest to the percolation threshold, and increases to over $10^3$ Hz when $p = 0.72$. At low frequencies, $f < f_0$, the value of $C'$ increases sharply with decreasing frequency. One can also notice a kink in the dependence at a lower frequency.

The dependence of the real capacitance $C'$ of samples (NiFe)-SiO$_2$ on the value of $\Delta p = p - p_c$ on a double logarithmic scale is shown in the inset of Fig. 4.7. At intermediate frequency of 25 kHz (filled circles) a power law dependence of the form $C' \propto (\Delta p)^a$ is observed. The solid line in the plot shows the best fit to this dependence with $a = 0.7 \pm 0.1$. In contrast, the effective capacitance measured at a low frequency $f = 20$ Hz (shown by filled triangles) increases with $p$.

The frequency dependence of the imaginary part of capacitance $C''$ for the same set of samples is shown in Fig. 4.9. At low and intermediate frequencies,
At $f < f_c$, the values of $C''$ are inverse proportional to frequency within the experimental accuracy, $C'' \propto 1/f$. This is an indication of the dominant contribution of DC conductance [21]. For the samples closest to percolation, a dispersion of the form $C'' \propto f^{-\gamma}$ is observed at high frequency $f > f_c$, where $\gamma \approx 0.14, 0.33$ and $0.82$, respectively for samples with $p = 0.55, 0.58$ and $0.63$. In the latter case, the dispersion may not be well developed. The dependence of the real conductance $G = 2\pi f C''$ on $\Delta p$, at $f = 25$ kHz, is shown in the inset of Fig. 4.8 on a double logarithmic scale. The conductance can be represented as a power law function of metal volume fraction $G \propto \Delta p^t$. However, we notice that this dependence does not extend to the closest vicinity of the percolation threshold. The critical exponent $t$ has a value of $t \approx 4.2$, which is a little lower than that found in Ref. 4 for the same samples in low temperature DC measurements. One can find that measurements on several co-sputtered Pt-SiO$_2$ and Au-SiO$_2$ samples, where the dielectric behavior was found qualitatively similar to that of (NiFe)- (SiO$_2$) films.

The intermediate and high frequency behavior of the complex capacitance can be qualitatively understood as due to percolation type scaling. In the percolation models, for frequencies much smaller than the plasmon frequency of metal, the composite system is considered as a random mixture of metallic regions with real conductivity $\sigma_m$ and dielectric regions with real dielectric constant $\varepsilon_r$. The effective DC conductivity of the percolation system $\sigma(0)$ is determined by the infinite percolation cluster above the percolation transition, while the effective dielectric constant $\varepsilon(0)$ arises from charge polarization in the finite metallic clusters of the typical size $\xi_p$. The diverging percolation
correlation length $\xi_p \propto |\Delta p|^{-\nu}$, where $p$ is the concentration of metal, $\Delta p = p - p_c$ and $\nu \approx 0.9$ in 3d [22], is the scale of inhomogeneity of the percolation problem [23]. The static conductivity and dielectric constant are described as power functions of $\Delta p$, namely $\sigma(0) \propto (\Delta p)^t$ (for $p > p_c$) and $\varepsilon(0) \propto |\Delta p|^{-s}$, where $t$ and $s$ are the critical exponents. For a 3-dimensional array, in the simplest approximation, $\sigma(0)$ and $\varepsilon(0)$ scale with $\xi_p$ as $\sigma(0) \propto \varepsilon^{-3}_p$ (for $p > p_c$), and $\varepsilon(0) \propto \xi_p$. This gives a simple estimate for the values of the critical exponents $t \approx 1.8$, and $s \approx 0.9$, which are not too much different from the elaborate computer simulation results: $t \approx 2.0$ and $s \approx 0.76$ [22]. We note that the experimental dependence of the real capacitance $C'$ at $f = 25k$ Hz on concentration, shown in the inset of Fig. 4.7, is in agreement with the theoretical result, the experimental value of $s$ being $s = 0.7 \pm 0.1$. While the real effective dc conductance shows a non-universal behavior with $t \approx 4.2$. The non-universal values of $t$ were observed in cermets before and are sometimes attributed to tunneling effects [24].

Two well-known variations of the classical scaling theory of frequency dispersion close to the percolation transition, $|\Delta p| \ll 1$, are based on intercluster polarization approach [25,26], or on anomalous diffusion on non-interacting fractal clusters [27]. The frequency dependence of the complex effective conductivity $\sigma(\omega)$ and dielectric constant $\varepsilon(\omega)$ is determined by the value of the scaling parameter $z = h/\Delta p^{\nu t}$, where $h = (\omega \varepsilon_0 \varepsilon_\infty)/\sigma_m$ is the "conductivity ratio", $\omega = 2\pi f$, and $\varepsilon_\infty$ is the permittivity of free space. When the value of $z$ is small, $z \ll 1$, and $\Delta > 0$, the real part of dielectric constant is constant $\varepsilon'(\omega) = $ const, and the imaginary part is inverse proportional to
frequency, $\varepsilon''(\omega) \propto \omega^1$. The latter equation is the signature of dominant contribution of conduction via the infinite cluster. When $z \gg 1$, the dielectric constant follows a power law dependence on frequency, $\varepsilon(\omega) \propto \omega^y$, where the critical exponent $y = s/(s+t) \approx 0.27$, using the universal values of $s$ and $t$. This dispersive behavior can be observed when the frequency is greater than the crossover value, which can be estimated from the condition $z \equiv 1$, or $\omega_c = 2\pi f_c = \Delta n^{\circ}(\varepsilon_0, \varepsilon)$. It can be easily checked that this estimate exceeds considerably the values of $f_c$ which have been observed in many experiments. In our experiment (Figs. 4.7 and 4.8) the crossover frequency is also much lower that this estimate. The theoretical value of $y \approx 0.27$ can be compared to the experimental results discussed above. The experiment is qualitatively in line with the theory. However the values of the exponent deviate from the predictions, in particular for sample with $p = 0.63$, where the value of $y$ about 0.23 is found based on the $C'$ data, and is 0.82 based on the $C''$ data. A relatively narrow frequency range for evaluation of the exponents may be one reason for these discrepancies.

The sharp increase of the effective capacitance at low frequency (Fig. 4.7) cannot be explained by the percolation theory. We consider tunneling as a possible reason. The percolation problem with tunneling for metal-insulator composites below the percolation threshold has been considered both for DC conductivity [24], and AC conductivity [28]. We suggest that both the metallic spanning cluster and tunneling junctions have to be taken into account for ac properties of conducting ($p > p_c$) nanocomposite films. Qualitatively, it can be done as follows. Let us consider the system as consisting of two independent
components. The first, metallic sub-system, consists of the infinite percolation cluster and the finite clusters of metal particles discussed above. Its properties can be described by the classical percolation theory. The second component is composed of tunneling junctions between the finite clusters. The typical tunneling distances \( r \) may be distributed between \( d \) and \( \xi_p \), where \( d \) is the particle size. Neglecting the charging effects, the probability of phonon-assisted tunneling between metal clusters in unit time, or tunneling frequency, is [29]: \( \tau^{-1} \sim f_{ph} \exp[-2r/a] \), where \( f_{ph} \sim 10^{12} \) Hz is the typical phonon frequency and \( a \) of the order of several Angstroms is the localization length. As \( r \) is in general much greater than \( a \), the resistivity of the tunneling sub-system is much greater than that of the metal infinite cluster. Following Ref. [28], we can consider each finite cluster of size \( \sim \xi_p \) as a node for a new percolation problem where the resistance between adjacent nodes is \( R_{ij} = R_\infty \exp[\lambda] \). In our case, the values of \( \lambda \) are distributed between \( 2d/a \) and \( 2\xi_p/a \). For the system with the exponentially wide distribution of resistances [23], only a small fraction of tunneling junctions determine conductivity. These important junctions form a network with a typical separation \( \xi_m \), or the tunneling correlation length, which may be much greater than the percolation correlation length. A simple estimate [23] for this value gives \( \xi_m \sim \xi_p \times \lambda_m \sim \xi_p \sim 1/a \), where \( \lambda_m \) is the maximum value of the random parameter \( \lambda \). As noted in Refs. [23, 28], \( \xi_m \) is the true scale of inhomogeneity of field and current for the problem with tunneling. From which we assume that both the effective conductivity and dielectric constant of the tunneling sub-system should scale with \( \xi_m \), in particular, \( \epsilon_i \sim \xi_m \). Tunneling requires a wait time \( \tau \sim (f_{ph})^{-1} \exp[2r/a] \). This process is thus released only at

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low frequency $f << 1/r$. This is why the sharp increase of the effective capacitance is observed only at low frequency (Fig. 4.7). One can estimate the cutoff frequency as $f_c \sim f_0 \exp[-2d/a]$. Assuming $a \sim 2\text{Å}$, the cutoff frequency of $10^3 \text{ Hz}$ (Fig. 4.7) would correspond to a typical tunneling distance about 20 Å.

Fig. 4.7 Dependencies of the resistance real part of capacitance $C'$ on frequency for the (NiFe)-(SiO$_2$) samples. The metal volume fraction $p$ is shown in the figure. The insert show a dependence of capacitance at 20 Hz (circles) and at 25 kHz (triangles) on $\Delta p = p - p_c$ on a double logarithmic scale. The solid line is the best fit to a power law for the 25 kHz data. The dashed line is a lead to the eye.
For an intuitive understanding of why only the real part of capacitance, but not conductance is dominated by tunneling at low frequency, we can suggest a qualitative equivalent R-C circuit of the system, where the inter-cluster tunneling component with parameters $R_t$ and $C_t$ is in parallel with the metal component of $R_m$ and $C_m$. At low frequency, $R_t >> R_m$ and $C_t >> C_m$. Then capacitance is dominated by tunneling, and resistance is dominated by the metal channels.

Fig. 4.8 Dependencies of the capacitance $C'$ on frequency for the Au-SiO$_2$ and Pt-SiO$_2$ samples.
Fig. 4.9 Frequency dependencies of the effective imaginary part of capacitance $C''$ for (NiFe)-SiO$_2$ samples. The insert shows a dependence of $C''$ at 25 kHz on $\Delta p = p - p_c$ on a double logarithmic scale.
Chapter 5

Conclusion

To conclude, in transport properties in percolating magnetic nanostructures exhibiting giant Hall effect, we have identified a few unique correlated features, namely the correlation between temperature dependent resistivity and a particle size distribution having a large fraction of small nanometer sized particles, power law dependencies between the magnetoresistivity and the room temperature resistivity, and that of temperature dependent part of resistivity and room temperature resistivity. These unique features are explained by a phenomenological model developed in this work, assuming the presence of nanometer sized particles in the percolating conducting channels, whose contribution to the conduction is sensitive to the temperature and external magnetic field. We believe that the key to understand the origin of the giant Hall effect lies in the understanding of the unusually large critical exponents.

The dielectric measurements on NiFe-SiO$_2$, Pt-SiO$_2$ and Au-SiO$_2$ granular co-sputtered films close to the metal-insulator transition and in the metallic state suggest that tunneling dominates the real part of low frequency effective dielectric constant. We associate it with the frequency-dependent tunneling correlation length. A percolation-type behavior is observed at intermediate and high frequency. The behavior of the effective capacitance at frequencies lower than our measurement range would be an interesting experimental problem. The effective dielectric constant should be finite at $f \to 0$. We thus expect more interesting features at low frequency.
References


2472 (1988).


Appendix:

The Resistor Network Model Proposed by Miller and Abrahams

Starting with electron wave functions localized on individual donors, calculate the probability that an electron transition will occur between two donors i and j with the emission or absorption of a phonon. Then calculate the number of transitions i to j per unit time. In a weak electric field the forward and reverse transitions will give rise to a current proportional to the field. Evaluating this current yields the resistance $R_{ij}$ of a given transition, and then it will equivalent to calculating the electrical conductivity of network of random resistors. This kind of conductivity of a random network which has its vertices at the donors and which resistance connects each pair of vertices. According to [23], we have

$$R_{ij} = R_{ij}^o \exp(\xi_{ij})$$

where

$$\xi_{ij} = \frac{2r_{ij}}{a} + \frac{\epsilon_{ij}}{kT}, \ a \ is \ an \ effective \ Bohr \ radius$$

and

$$R_{ij}^o = \frac{kT}{e^2_y \gamma_{ij}^o}, \ \gamma_{ij}^o \ is \ the \ transition \ probability$$