# 行政院國家科學委員會專題研究計畫　期中進度報告

## 利用磁性元素參雜研究金屬-非金屬與超導體-非導體相變

## (2/3)

計畫類別：個別型計畫
計畫編號：NSC92-2112-M-009-007-
執行期間：92 年 08 月 01 日至 93 年 07 月 31 日
執行單位：國立交通大學電子物理學系

計畫主持人：許世英

計畫參與人員：鍾廷翊、林佩真、張嘉顯、林玉敏、陳偉仁

報告類型：精簡報告

處理方式：本計畫可公開查詢

中　華　民　國 93 年 6 月 1 日

# 行政院國家科學委員會補助專題研究計畫期中報告

※※※※※※※※※※※※※※※※※※※※※※※※※
※利用磁性元素參雜研究金屬-非金屬與超導-非導體相變(2/3)※
※※※※※※※※※※※※※※※※※※※※※※※※※

計畫類別：■個別型計畫　　□整合型計畫

計畫編號：NSC92－2112－M－009－007

執行期間：92 年 8 月 1 日至　93 年 7 月 31 日

計畫主持人：許世英

共同主持人：

計畫參與人員：

鐘廷翊（博士班一年級生）

林佩真（博士班一年級生）

張嘉顯（碩士班二年級生）

林玉敏（碩士班一年級生）

陳偉仁（碩士班一年級生）

本成果報告包括以下應繳交之附件：

□赴國外出差或研習心得報告一份

□赴大陸地區出差或研習心得報告一份

□出席國際學術會議心得報告及發表之論文各一份

□國際合作研究計畫國外研究報告書一份

執行單位：國立交通大學電子物理系

中　華　民　國　93　年　05　月　29　日

# 行政院國家科學委員會專題研究計畫成果報告

## 利用磁性元素參雜研究金屬-非金屬與超導-非導體相變

## Metal-Insulator and Superconductor-insulator transitions in the magnetic materials

計畫編號：NSC 92-2112-M-009-007

執行期限：92 年 8 月 1 日至 93 年 7 月 31 日

主持人：許世英　　國立交通大學電子物理系

### 一、中文摘要

在二氧化矽與鈷共濺鍍的厚膜系統，藉由二氧化矽與鈷兩者的相對濺鍍速率控制，我們可以調變樣品內的金屬體積百分率，使樣品無序程度由弱到強分布，然由於不同的顆粒成長機制，顆粒間磁矩的耦合卻造成一般在磁多層膜與磁穿隧連接元件所發現的巨磁阻(遲滯)，而其他量子干涉效應反倒可能因磁矩導致破壞而未展現該有大小的磁阻；隨著樣品無序程度增加，樣品的遲滯磁電阻率變化率有上升的趨勢，在金屬-絕緣相變附近的樣品磁電阻由 GMR 效應主導，在溫度 10K 時，樣品最大磁電阻率變化率在 0.05%~2%之間。隨著樣品無序程度增加，在強無序地帶時，樣品磁電阻由 TMR 效應主導，其樣品最大磁電阻率變化率在 2.0%~10%之間。

另外在二維鈷膜可藉由自然氧化增加樣品的無序性，而且由於氧化鈷的反鐵磁性，發現異向性磁組會因無序程度（氧化程度）有所改變。

**關鍵詞**：量子干涉效應、無序程度、磁矩、異向性磁組、鐵磁、反鐵磁。

### Abstract

We have measured the magnetization and magneto-transport in a series of 3D magnetic $Co_x(SiO_2)_{1-x}$ samples to study the metal-insulator translation. By controlling the sputtering rates of Co and $SiO_2$, the degree of disorder of $Co(SiO_2)$ can be made from weakly disordered to strongly disordered. Co seems form grains embedded in $SiO_2$ sea and giant MR due to the couplings of magnetic moments between grains appears in these samples. GMR is usually observed in either a magnetic multilayer system or a magnetic tunneling junction. Moreover, MR due to quantum interference effects is greatly reduced. We think that magnetic moment in each grain is detrimental to the quantum interference effects. For sample near the metal-insulator transition, the magnetoresistance is due to spin-dependent scattering effect. The magnitude of GMR ratio is in the range of 0.05%~2% at 10K. For sample in strongly disordered region, MR comes from spin-dependent tunneling effect. The magnitude of TMR ratio is in the range of 2%~10% at 10K.

Besides, the disorder can be increased by oxidation in 2D Co films. Due to the antiferromagnetic property of CoO, the anisotropic MR can be changed by increasing disorder (oxidation).

**Keywords**: quantum interference effects, disorder, magnetic moment, anisotropic MR, ferromagnetism, antiferromagnetism.

### 二、緣由與目的

Disordered samples have a long history as systems in which new and interesting phenomena can be studied and uncovered. For studies of metal-insulator transition, numerous properties were well known by now. Recent interest is due to discover of giant magnetoresistance (GMR) in magnetic-nonmagnetic binary metal granular samples.[1] We would like to take consideration of magnetic moment into problem of disordered tuned phase transition to understand how it influence the dominant mechanisms that drives the transition.

In the previous project (1/3), we concluded that the evolution of temperature dependent resistivity with increasing disorder in granular $Co_x(SiO_2)_{1-x}$ sample is similar to that observed in non-magnetic CuGe systems. However, MR due to quantum interference effects is greatly decreased because that magnetic moment in each grain is detrimental to the quantum interference effects.

Two systems are employed in this work. One is granular $Co_x(SiO_2)_{1-x}$ samples (3D) and the other is oxidized Co films (2D). In the former topic, we made further experiments such as relative responses of annealing induced property change to disorder. We also set up a 5-probe Hall effect measurement system to complete the AHE study.[2,3] In the latter topic, the relation between the anisotropic MR and disorder R, was investigated.

## 三、實驗結果

### (I) Granular $Co_x(SiO_2)_{1-x}$ samples

As reported before, the behaviors of these samples can be cataloged into three regimes. Sample with $\rho_{10K}$ <500μΩcm, low temperature transport $\Delta\rho(T) \propto -T^{1/2}$. For sample with 500μΩcm<$\rho_{10K}$<100mΩcm, close to MIT, $\Delta\rho \propto \lambda n(T)$ is better than any other form to describe the transport behavior. [2] For sample with $\rho_{10K}$ >100mΩcm, ρ becomes much more sensitive to temperature and follows the hopping mechanism resulting in the form that $\Delta\rho \propto \exp[1/T^{1/2}]$. [4] However MR is mainly attributed to the couplings of magnetic moments between grains appears in these samples. MR due to quantum interference effects is greatly reduced. We plot data for sample with $\rho_{10K}$=70mΩcm, near MIT, in figure 1. As shown, both magnetic moment and magnetoresistance demonstrate clear hysteresis loops. MR cycles through a curve with maximum at the field near the coercive field of the sample ($H_c$=930Oe) and decreases with increasing field intensity until the saturation field. This observed hysteresis MR is similar to the observed GMR in magnetic multilayer systems and TMR in magnetic junction systems.

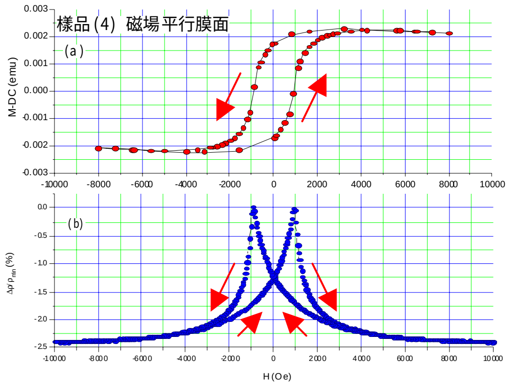


Figure1, normalized magnetic moment (a) and magnetoresistance (b) as functions of applied magnetic field for $Co_x(SiO_2)_{1-x}$ sample near the MIT.

The change of magnetoresistance ratio, $MR \equiv (R(H)-R_{min})/R_{min}$, of the samples in the intermediately disordered regime seems scale linearly with the disorder ρ. Data is shown in figure 2 and red line is obtained from least square root fit.

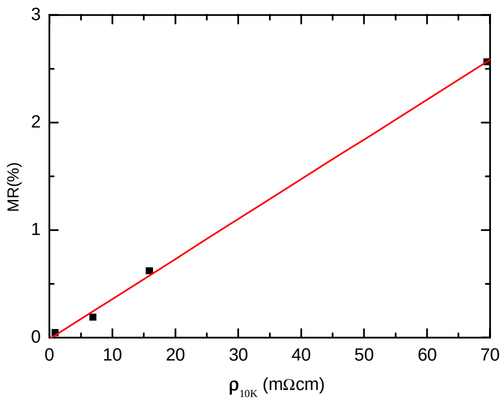


Figure2, normalized magnetoresistance (MR) as a function of $\rho_{10K}$ for four $Co_x(SiO_2)_{1-x}$ samples near MIT.

MR increases continuously with

increasing disorder, even in the strongly disordered regime where transport is dominated by hopping mechanism among isolated grains. However, the increasing rate seems slower instead of linear growing. We may conclude that tunneling effect limits the MR.

### (II) oxidized Co films

We fabricated 10nm Co thin films and controlled their degrees of disorder by nature oxidation. The temperature dependent sheet conductance can be well described by two-dimensional weakly disordered theorem and demonstrate that $\Delta G \propto A\frac{e^2}{2\pi^2\hbar}\ln(T)$ . We plot such relation for some samples in the inset of figure 3. A's obtained from the linear fit of data to the above predicted form for a series of samples are shown in figure 3. A is about 1 in consistent with most 2D homogeneous thin films. Disorder induced electron-electron interaction plays an important role. Films with severe oxidation have larger slope (A) and sheet resistance than slightly oxidative samples.

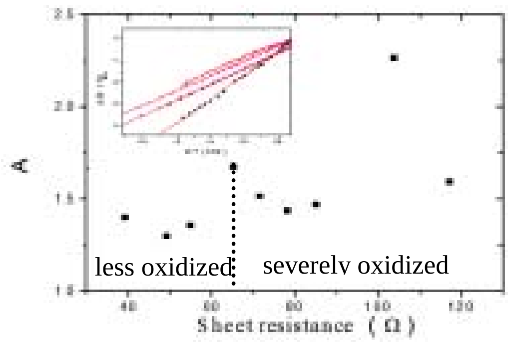


Figure 3, Inset: Sheet conductance vs. logarithmic temperature for some films. A obtained from theoretical fit vs. sheet resistance(15K) for a series of films.

For most magnetic materials, inverse MR occur when current is applied along magnetic field direction and normal MR occur when current is applied perpendicular to field direction. $\Delta R(H) \equiv R_{,}(H)-R_{saturation}$ at conditions that current is applied perpendicular or parallel with the magnetic field for a less oxidized Co film is shown in figure 4. This scenario can be attributed to the s-d scattering effect is usually referred as anisotropic magneto-resistance (AMR). [5] In the thin films, some may argue that quantum interference effect is very important for transport behavior such as ρ(T). However our data indeed show that there is a small symmetric positive MR in both curves due to the quantum interference effects at low temperature. The size is decreased with increasing temperature. To ignore this background, we analyzed data at T=15K. Such clear hysteresis MR results mainly from magnetic domain induced spin-dependent scattering.

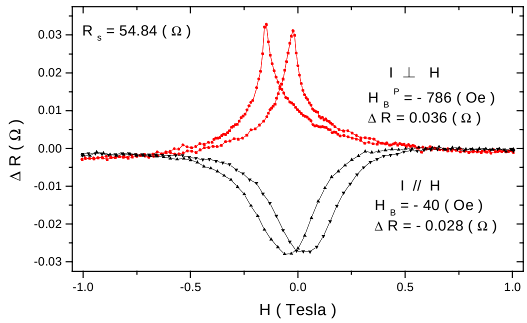


Figure 4, ΔR(H) vs. applied magnetic field for a less oxidized Co film at conditions that current is applied perpendicular to the magnetic field (top curve) and parallel with the magnetic field (bottom curve).

In a very thin magnetic film, surface oxidation can become very severe and affect its intrinsic properties. Hence, in this work we have also performed low temperature magneto-resistance and magnetization measurements for a series of Co thin film with different degrees of oxidation to investigate the role of surface oxidation. For thicker films and thin films with less oxidation, magneto-transports behave as theoretical expectation described above. However, for thin films with severe oxidation, the magneto-transports are completely reversed. The inverse MR is replaced by normal MR when current is applied along magnetic field direction. The normal MR is changed to inverse MR when current is applied perpendicular to field direction. We summarized magneto-transport results of all samples in figure 5**.** Normal MR is referred as the top curve in figure 4 and positive ΔR is defined as $R_{peak}$-$R_{saturation}$. Inverse MR is referred as the bottom curve in figure 4 and negative ΔR is defined as $R_{dip}$-$R_{saturation}$. Three samples with less disorder (R,<70Ω) demonstrate normal AMR and others with more disorder (R,>70Ω) demonstrate completely opposite behaviors, reversed AMR.

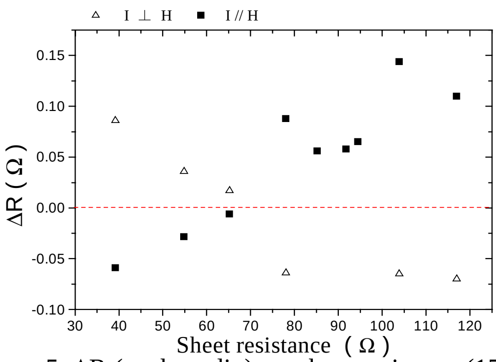

Figure 5, ΔR (peak or dip) vs. sheet resistance(15K) for a series of films. Δ: Current is applied perpendicular to the magnetic field. ■: Current is applied parallel with the magnetic field.

This reverse transition in AMR also occurs in Co films with decreasing sample thickness. As we know, the oxidized layer CoO is an antiferromagnetic material that may provide exchange coupling with the rest ferromagnetic Co layer. When the oxidation is severe relative to the Co layer that exchange coupling is strong to pin the magnetic moment of Co, AMR is reversed. [6]

## 四、結論

In the studies of $Co_x(SiO_2)_{1-x}$ systems, the MR demonstrates hysteresis behavior due to the magnetic couplings between Co grains. Near the metal-insulator transition with a $\lambda n$(T) dependent resistivity, scattering length of electron is more than numerous of gain distance, MR can be described by the two-current model as in multilayer systems. The magnitude of MR ratio is in the range of 0.05%~2% at 10K and scales nearly linear with disorder $\rho_{10K}$. For sample in strongly disordered region, MR comes from spin-dependent tunneling effect. The magnitude of MR ratio is in the range of 2%~10% at 10K. Negative MR due to quantum interference in hopping regime become importance and is comparable to spin-dependent MR. Besides, the disorder can be increased by oxidation in 2D Co films. The anisotropic MR can be changed by increasing disorder (oxidation) because of the antiferromagnetic property of CoO.

## 五、參考文獻


[1] J.Q. Xiao, J.S. Jiang, and C.L. Chien, Phys. Rev. Lett. **68**, 3749 (1992); J.Q. Wang and G. Xiao, Phys. Rev. B**49**,3982 (1994); X.N. Jiang, N. Wang, A.B. Pakhomov, K.K. Fung, and X. Yan, Phys. Rev. B**53**, 14032 (1996).

[2] B.A. Aronoz et al., JETP **70**, 90 (1999); P. Xiong et al., Phys. Rev. B**59**, R3929 (1999).

[3] C.C. Wan and P. Sheng, Phys. Rev. B **66**, 075309 (2002).

[4] T. Chui, G. Deutscher, P. Lindenfeld,and W. L. McLean, Phys. Rev. B**23**, 6172 (1981). X. N. Jing et al., Phys. Rev. B **53**, 14032 (1996).

[5] R.I. Potter, Phys. Rev. B**10**, 4626 (1974).

[6] B.H. Miller, Appl. Phys. Lett. **69**, 3932 (1996); T. Gredig, J. Appl. Phys. **91**, 7760 1851 (2002); I.N. Krivorotov et al., Phys. Rev. **B65**, 180406 (2002).


## 國內外會議：

1. T.Y. Chung and S.Y. Hsu, "Novel-AMR: anisotropic magneto-transport in 2D magnetic film", poster, 物理年會, Feb. 2004.
2. S. Y. Hsu**,** Y. T. Cheng, and C. H. Chang, "Electrical transport of granular Co samples near metal-insulator transition", oral, Taiwan International conference on superconductivity", July 2004.
3. P. J. Lin and S. Y. Hsu, "Upper critical field of granular SnGe samples near the superconductor-to-insulator transition", poster, Taiwan International conference on superconductivity", July 2004.