Abstract

Investigation of Fe_3O_4 and ZnCoO based Magnetic Tunnel Junctions

This research focuses on the investigation of Fe_3O_4 and ZnCoO based magnetic tunnel junctions (MTJs) with crystalline MgO barrier, especially on the fabrication of electrodes of Fe_3O_4 and ZnCoO and their magnetic and transport properties.

Three main topics are discussed in this dissertation. First of all, the growth of epitaxial Fe_3O_4 films is studied on MgO (100) substrates at room temperature by using reactive ion beam deposition (IBD). The Verwey transition, unique feature of stoichiometric Fe_3O_4, of 110K was observed on epitaxial (100) Fe_3O_4 films. To integrate epitaxial Fe_3O_4 films on Si substrates when the Fe_3O_4 are the electrode for MTJ, epitaxial (111) Fe_3O_4 films were grown on Si substrates with introducing Cu conducting underlayer at room temperature. X-ray φ-scans and TEM diffraction pattern revealed unusual 12-fold symmetry of the epitaxial (111) Fe_3O_4 films on Cu (001) underlayers due to the presence of two sets of epitaxial (111) grains in Fe_3O_4 films. In addition, a clear Verwey transition of room temperature growth (111) Fe_3O_4 films with Cu

conducting underlayers on Si substrate was also observed at around 116K.

The second topic is the fabrication of the dilute magnetic semiconductor – ZnCoO and the investigation of the exchange coupling between the AFM layer and ZnCoO. Epitaxial $ZnCo_{0.07}O$ films on the Cu underlayer were fabricated at room temperature by ion beam deposition on Si substrates, which revealed room-temperature ferromagnetic behavior with coercivity of 70 Oe. To integrate DMSs into spintronic devices, we studied exchange biasing between ZnCoO and NiO. In order to prevent the problems of interfacial reactions between the ZnCoO and NiO layers, the quasi-epitaxial full-oxide exchange-bias system (ZnCoO/NiO) was prepared at room temperature. In the epitaxial $ZnCo_{0.07}O$ /NiO system, exchange fields accompanying vertical magnetization shifts were observed after field cooling. Transitions of exchange fields and magnetization shifts were observed at 50 K, above which the magnetization shift disappeared and the exchange field was significantly reduced. Both the exchange field and the magnetization shift increased with increasing cooling-field strength at temperatures below 50 K, which might be attributed to the existence of "frozen" spins in ZnCoO. The observed linear dependence of the exchange field on the magnetization shift may directly elucidate the role of pinned spins on the exchange fields.

In the third topic, the fully oxides MTJ device composed of Fe_3O_4 and ZnCoO ferromagnetic layer with a crystalline MgO barrier was fabricated at room temperature. The novel structure of full stack MTJ, SiO_2// Ta 20nm/ ZnO 2.5nm/ MgO 1.2nm/ Fe_3O_4 50nm/ MgO 3nm/ ZnCoO 50nm/ Ta 10nm, by using an IBD system. This stack included a conducting electrode, Ta, suitable for lift-off process, a textured MgO barrier with sharp interface and highly (100) textured FM layer. The non-linear I-V curves for the fully oxides MTJs patterned by lift-off process were observed, which indicated that the electrical transport was dominated by tunneling. The MR of the full oxides stack MTJ was 2.68% at 150K. Furthermore, the clear plateau of the R-H curve was observed clearly in the low field region, which represented the typical pseudo-spin valve switching behavior. It was a strong evidence to prove that DMS could provide the spin-polarized electrons.

論文摘要

四氧化三鐵與鈷摻雜氧化鋅所組成之穿遂式磁阻元件的研究

本研究工作專注在穿遂式磁阻元件的製備以及自旋傳輸行為上，其中穿遂式磁阻元件包含了四氧化三鐵與鈷摻雜氧化鋅材料作為磁性層並搭配結晶化之氧化鎂作為絕緣層。此外，本論文特別對四氧化三鐵與鈷摻雜氧化鋅材料的制備、磁性質以及電性進行研究與討論。本論文中可分作三個主要的研究課題。

首先，我們利用反應式離子濺鍍系統在氧化鎂基版上室溫製備出磊晶成長的四氧化三鐵薄膜。在此磊晶四氧化三鐵薄膜中，我們在 110K 時觀察到顯著的 Verwey 轉換行為，而此轉換行為正是等化學劑量比之四氧化三鐵的重要表徵。另一方面，當四氧化三鐵作為磁穿遂電阻元件的電極時，為了能使用矽基版來取代氧化鎂基版，我們在室溫製程下先行製備良導電率的銅作為結構緩衝層於矽基版上，緊接在上面製備出（111）方向磊晶的四氧化三鐵薄膜。藉由 X 光繞射分析以及穿遂式電子顯微鏡的擇區繞射結果顯示出磊晶方向(111)的四氧化三鐵薄膜呈現了獨特十二軸對稱，此對稱乃是由於兩組磊晶方向為

（111）的四氧化三鐵晶粒所造成。此外，我們依然在良導電率之銅緩衝層上室溫成長的磊晶四氧化三鐵薄膜，在 116K 時觀察到 Verwey 轉換行為。

本論文的第二部份乃是專注在製備稀磁半導體—鈷摻雜氧化鋅以及研究反鐵磁層與鈷摻雜氧化鋅間的交互耦合行為。我們同樣利用離子槍濺鍍系統在矽基版上製備銅緩衝層，並在其上製備磊晶的鈷摻雜氧化鋅膜層。磊晶的鈷摻雜氧化鋅呈現室溫鐵磁性，其矯頑場達到 70 奧斯特。為了能更進一步低把鈷摻雜氧化鋅材料整合到自旋電子元件中，本論文對此材料的交互耦合行為進行研究。另一方面，為了根除稀磁半導體與反鐵磁材料間的介面反應問題，我們在室溫下製備了全氧化類型的交互偏壓系統（鈷摻雜氧化鋅 / 氧化鎳）。在此磊晶的鈷摻雜氧化鋅/氧化鎳系統中，倘若樣品經過場冷程序過後，我們發現樣品的交互偏壓場會伴隨著磁化量的垂直位移一齊發生。此特殊的行為在溫度為 50K 時，有一個劇烈的轉變行為，在 50K 以上，樣品的交互偏壓場以及磁化量的垂直位移都會呈現顯著的減少。在 50K 以下，樣品的交互偏壓場乃是隨著磁化量的垂直位移增加而增加，此現象有可能是來自於在鈷摻雜氧化鋅中凍結之自旋磁矩。此外，由於樣品的交互偏壓場與磁化量的垂直位移乃是呈現線性關係，代表此現象

可以協助我們釐清被固定之自旋磁矩在交互耦合行為中所扮演的角色。

在本論文的第三個課題，我們在室溫環境下製備出全部皆由氧化物所組成的磁穿遂磁阻元件，其中四氧化三鐵以及鈷摻雜氧化鋅作為磁性層，並搭配結晶化的氧化鎂絕緣層。此膜層結構為 SiO_2// Ta 20nm/ ZnO 2.5nm/ MgO 1.2nm/ Fe_3O_4 50nm/ MgO 3nm/ ZnCoO 50nm/ Ta 10nm，乃是由離子槍濺鍍系統在室溫下製備完成。值得強調的是此結構包含：適合於舉離製程的導電下電極鉭，擁有平整介面的織構化氧化鎂絕緣層以及高度織構化的鐵磁膜層。經由舉離製程所製備出來的氧化物類型穿遂式磁阻元件展現了非線性的電流-電壓曲線，此非線性曲線指出在元件中的電子傳輸行為主要是以穿遂行為為主。在 150K 時，元件的磁阻變化率也達到了 2.68%。我們更進一步地觀察到磁阻曲線中的低磁場範圍中，展現顯著的平台區，此正是典型的準自旋閥特徵曲線。上敘結果恰好為稀磁半導體亦能產生自旋極化電子提供了有力的證據。