

Chapter 1

Introduction

Spintronics [1], a new approach based on the up and down spins of charge carriers where the orientation of magnetization in ferromagnetic films determines electrical resistance [2-3], is one of the most attractive fields in nano-electronics. Especially, the discovery of the tunneling magnetoresistance (TMR) in magnetic multilayers has made a strong impact on scientific researches and the development of read heads or magnetic random access memories (MRAM) [4].

100% spin polarization makes the half metallic material so attractive in applications for spintronic devices. The so-called half metallic ferromagnets can provide electrons of one directional spin. According to Juilier's model, half-metal electrodes used for tunneling TMR junctions should result in an ultrahigh MR at room temperature. Applications to spintronic devices require high Curie temperature (T_C) due to the consideration of thermal stability. Hence, the Fe_3O_4 ($T_C = 850\text{K}$) may be superior to LaSrMnO_3 ($T_C = 360\text{K}$) or CrO_2 ($T_C = 395\text{K}$) [5-6]. In this dissertation, investigation and fabrication of half-metal Fe_3O_4 was one of the study topics.

A diluted magnetic semiconductor (DMS) is one of the promising candidates for spintronic devices due to its capability of manipulating both degrees of freedoms of electrons' spins and charges [7-8]. According

to theoretical calculations [9], doped-ZnO is one of the candidates for high Curie-temperature DMSs. In this dissertation, investigation and fabrication of DMS ZnCoO was one of the study topics.

Finally, in order to investigate the tunneling spin-transport behavior of the MTJs with Fe_3O_4 and ZnCoO as the FM layer, the MTJs stacks were in the form of $\text{Fe}_3\text{O}_4/\text{MgO}/\text{ZnCoO}$ fabricated with the modified underlayer at room temperature.

1.1 Motivation

The study of the TMR multilayer has drawn a great deal of attention in recent years due to its application in the storage industry, e.g., magnetoresistive read heads or magnetic random access memories (MRAM) [4].

The so-called half metallic ferromagnets can provide electrons of one directional spin. According to Juilier's model, half-metal electrodes used for TMR junctions should result in an ultrahigh MR at room temperature. Applications to spintronic devices require high T_C due to the consideration of thermal stability. Hence, the Fe_3O_4 ($T_C = 850\text{K}$) has more superiority when it to be used as FM layer in spintronic device.

Epitaxial growth of Fe_3O_4 , predicted to be a half-metal, on MgO single crystal has been reported by several groups [10-12]. Gong *et al.*

[10] succeeded in obtaining $M_s \sim 415 \text{ emu/cm}^3$ and $T_V \sim 120\text{K}$ on a MgO(100) substrate by using a pulsed laser deposition (PLD) system, but the deposition temperature was at 350°C , and the thickness was as thick as 6600\AA . To take advantages of half-metallic films for spintronic devices, for example, TMR or GMR devices, low temperature deposition is preferred. In addition, epitaxial Fe_3O_4 films may enable us to clearly investigate the effect of the half metal without grain boundary scattering. In addition, if Fe_3O_4 films are applied to the electrodes of TMR junctions, a highly conductive underlayer is required because the poor conductivity of Fe_3O_4 films may lead to a non-uniform current distribution. Therefore, the main purpose of this work is to grow epitaxial Fe_3O_4 films on the conducting underlayer at room temperature.

Dilute magnetic semiconductors (DMSs), which can manipulate both the spin and charge degrees of freedom, have attracted attentions in recent years. According to theoretical calculations [9], doped-ZnO is one of the candidates for high Curie-temperature DMSs. In order to integrate DMSs into devices, an exchange biasing scheme is quite important. The exchange coupling between a DMS ($\text{Ga}_{1-x}\text{Mn}_x\text{As}$) and an antiferromagnet (MnO) has been reported [13]. However, the Néel temperature T_N of the MnO is 118K which is far below the room temperature. Therefore, we used the high Néel temperature T_N material $\text{Ir}_{20}\text{Mn}_{80}$ ($T_N > 690\text{K}$) in this study as an antiferromagnetic (AFM) layer. On the other hand, in order to prevent the problems of interfacial reactions between the DMS and AFM layers, full-oxide exchange-bias system (ZnCoO/NiO) is also to be investigated.

The unidirectional anisotropy in a ferromagnetic (FM)/AFM system has been reported to be a consequence of interfacial exchange coupling between FM and pinned uncompensated AFM spins [14]. However, it is difficult to directly observe the vertical magnetization shift with conventionally measured FM/AFM hysteresis loops. This is because the typical magnetization of FM films is too large to reveal the existence of the pinned interfacial spins. On the other hand, vertical magnetization shifts have been reported in several nanoparticle core-shell systems due to the high interfacial area to volume ratio [15]. DMSs have much lower magnetization than typical FM films, which may enable us to investigate the role of the pinned spins in exchange-bias systems by measuring conventional hysteresis loops.

Finally, in order to investigate the tunneling spin-transport behavior of the Fe_3O_4 and ZnCoO material to be used as the electrode, the MTJs stacks were in the form of $\text{Fe}_3\text{O}_4/\text{MgO}/\text{ZnCoO}$ fabricated with the modified underlayer. We should remark that MgO has an excellent crystal lattice matching with Fe_3O_4 . High quality epitaxial Fe_3O_4 films could be grown on MgO substrate. Therefore, one can expect high tunneling MR in $\text{Fe}_3\text{O}_4/\text{MgO}/\text{Fe}_3\text{O}_4$ or $\text{Fe}_3\text{O}_4/\text{MgO}/\text{Fe}$ MTJs. Therefore, we choose the MgO as the tunneling barrier. Half metal – Fe_3O_4 and DMS – ZnCoO would be combined into MgO -based MTJs.

1.2 Outline of the Dissertation

The main part of this dissertation is a discussion of the deposition, characterization, related magnetic and spin-dependent transport properties of the fully oxides MTJ device composed of Fe_3O_4 and ZnCoO ferromagnetic layer with a crystalline MgO barrier. This chapter describes the motivation and the goals of the studies in the dissertation. Chapter 2 presents the background relevant to the discussions in this dissertation, including the theory of the TMR effect, theoretical models for exchange anisotropy, and a summary of related report about the half metal – Fe_3O_4 and DMS – ZnCoO . Then, the deposition method and some important measurements are described in the Chapter 3. The room-temperature fabrication and identification of half-metal Fe_3O_4 films and the epitaxial (111) Fe_3O_4 films with introducing Cu conducting underlayer was investigated in Chapter 4. The epitaxial (0002) ZnCoO films with introducing Cu conducting underlayer was prepared and an exchange bias behavior between quasi-epitaxial full-oxide exchange-bias system (ZnCoO/NiO) was investigated in Chapter 5. Chapter 6 is a discussion of deposition, fabrication, and spin-dependent transport properties of the fully oxides MTJ device composed of $\text{Fe}_3\text{O}_4 / \text{MgO} / \text{ZnCoO}$. Finally, the dissertation is summarized in Chapter 7.