Electric field induced room temperature ferromagnetism in transition metal doped oxide semiconductor

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1. Introduction

Ferromagnetic semiconductor possesses both ferromagnetic and semiconducting characters, leading to the controllability of both spin and charge degrees of freedom [1]. In previous studies on established ferromagnetic semiconductors like (Ga,Mn)As and (In,Mn)As, various spintronic functionalities have been reported such as the spin polarized light emitting diode [2], the electric field effect on ferromagnetism [3], the optical control of ferromagnetism [4], and the electrical magnetization rotation [5]. However, the Curie temperatures of those ferromagnetic semiconductors are lower than 200 K, hampering the operation of the spintronic devices at room temperature. In order to raise the Curie temperature, the other host compounds may be needed for a larger exchange interaction between band carriers and localized spins. In wide gap oxide semiconductors, the larger exchange interaction could be expected because of the heavy carrier mass and the large electron carrier density. We have proposed various magnetic oxide semiconductors [6-8], and discovered room temperature ferromagnetism in (Ti,Co)O₂ [9]. If the ferromagnetism is originated from an extrinsic source like the segregation of Co metal, this compound will not be useful for semiconductor spintronics. In order to confirm the presence of a carrier mediated exchange interaction, the electric field control of ferromagnetism at room temperature in (Ti,Co)O₂ is quite useful, as was demonstrated in (In,Mn)As and (Ga,Mn)As at low temperature [3]. The electrically controlled ferromagnetism at room temperature is also important for the implementation of spintronic devices.

2. Experiments and results

Pulsed laser deposition was used for epitaxial growth of anatase $(Ti,Co)O_2$ thin films. $Ti_{0.90}Co_{0.10}O_2$ (001) films were grown on LaAlO₃ (100) substrates buffered with 5 unit cell thick TiO₂. The amount of oxygen vacancy was varied to control the electron carrier density, by changing the oxygen pressure during growth, where the oxygen vacancy serves as an electron donor. The detail conditions are described elsewhere [10,11]. The optimization of sample quality was indispensable for reproducible properties.

In $(Ti,Co)O_2$, both ordinary and anomalous Hall terms are observed from the Hall effect measurement. Hence, it is possible to evaluate the relation between the carrier density and the magnetization, in contrast with (Ga,Mn)As. The anomalous Hall conductivity σ_{AH} was examined to evaluate the magnetization from the anomalous Hall term,, since σ_{AH} is monotonically increasing functions of the magnetization *M* and the longitudinal conductivity σ_{xx} , where the latter is proportional to the carrier density [10,11].

For electrical induction of the ferromagnetism in $(Ti,Co)O_2$, electric double layer transistor was used to apply high electric field for heavy carrier doping > 10^{14} cm⁻². This method is a powerful tool for the application of high electric field ~50 MV/cm without dielectric breakdown [12]. By applying positive gate voltage < 4 V, a sufficient amount of electron carriers was accumulated to transform insulating and paramagnetic (Ti,Co)O₂ channel into metallic and ferromagnetic channel at room temperature [10]. Figure 1 shows a schematic diagram of the observed phenomena. The amplitude of the magnetization is enhanced electrically at room temperature. Such enhancement is usually difficult for ferromagnetic metals due to the very high carrier density except for the ultrathin film form [13].



Fig. 1 A schematic diagram representing electrically induced ferromagnetism in this study. The itinerant electron carriers (sphere) accumulated by electric field induce ferromagnetic magnetization (arrows) [10].

3. Discussion

Co ions in (Ti,Co)O₂ was confirmed to be divalent from x-ray photoemission and x-ray magnetic circular dichroism spectroscopies [14–16]. Ferromagnetic responses of Co²⁺ ions in different samples were confirmed from the x-ray magnetic circular dichroism spectroscopy, in which significantly reduced surface magnetization was observed possibly due to the surface depletion [17]. From these results, any effect of the metallic segregation on the ferromagnetism was ruled out. Several studies reported previously the observation of superparamagnetism accompanied with the presence of Co nanoclusters [18,19]. Such superparamagnetism could be attributed to an excessively reduction of the sample preparation [11].

The electric field experiment also rules out the possibility of the defect-mediated ferromagnetism which does not need the mobile carriers [20–22]. In the chemical doping study, ferromagnetic insulator phase with smaller magnetization and lower carrier density was observed, however, such ferromagnetic insulator phase tuned into ferromagnetic metal phase with further increasing the carrier density [11]. Therefore, recently observed ferromagnetic insulator phase could be attributed to an insufficient carrier density.

4. Conclusion

The electric field effect and chemical doping experiments clarified that the carrier-mediated exchange interaction is responsible for the high temperature ferromagnetism in $(Ti,Co)O_2$. The electrically induced ferromagnetism at room temperature in this compound is promising for room temperature semiconductor spintronics. Further exploration of materials and the device implementation will pave the way to the development of room temperature semiconductor spintronics.

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