Gate-Controlled Transport Properties in Dilute Magnetic Semiconductor (Zn, Mn)O Thin Films

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Ionic liquid (IL) gating of functional oxides has drawn significant attention, since it can provide reversible changes in carrier concentration (∼10^{14} cm^{-2}) at the interface, permitting the manipulation of electrical and magnetic properties of oxide films with low voltages. In this paper, we demonstrated the electric-field manipulation of transport properties in the dilute magnetic semiconductor of $Zn_{0.98}Mn_{0.02}O$ (MZO), using an electric-double-layer transistor geometry through the IL electrolyte gating. The MZO layer exhibited reversible control of resistance up to 33% at 230 K. Moreover, magnetoresistance (MR) measurements revealed the influence of applied gate voltage (V_g) on the magnetotransport behavior, which exhibited a positive MR in the low-field region and a negative MR in high magnetic field (up to 9 T). An increase in low-field positive MR (<1 T) upon switching V_g from −2 to 2 V implied an enhanced ferromagnetic state of MZO due to an increased electron carrier concentration. The results demonstrated that a controllable carrier concentration by electric-field effect played an important role in the manipulation of magnetism in MZO.

Index Terms—**Dilute magnetic semiconductor (DMS) and magnetism, electric effect, Mn-doped ZnO (MZO).**

I. INTRODUCTION

ELECTRIC-FIELD manipulation of magnetic properties provides a promising way for preparing fast and compact data storage devices [1]–[4]. Recently, intensive studies were made on the control of magnetism in artificial multiferroic heterostructures through the manipulation of charge carriers or strain in the samples [4]–[11]. However, fabrication of such heterostructures is often limited by the compatibility between the ferromagnetic and ferroelectric components, such as lattice mismatch issue and interfacial diffusion due to high deposition temperatures [7], [11]. Moreover, for observable tunability of magnetism, one often requires thin ferroelectric layers or large-switching voltages for the complete reversal of the polarization directions [12], which may result in the breakdown of the ferroelectric layer [6].

For low-switching voltages and high tunability of carrier concentrations at the ferromagnetic interface, ionic-liquid (IL)-gating modulation has demonstrated interfacial carrier concentration changes up to 10^{14} cm^{-2} in ferromagnets, which is larger than the tuning by electrostatic doping via magnetoelectric coupling [13]–[17]. The IL-gating technique has been widely deployed for achieving high tunability in different systems such as the metal-to-insulator transition in the complex oxides [13], [14], controlling superconductivity in 2-D materials [18], and exchange-bias effect in antiferromagnets [19].

Generally, the electric-field control of carrier density is more effective in semiconductors as compared with metallic systems, since the carrier density in semiconductors is two to three orders of magnitude smaller than metals [1]. A high tunability in carrier density is expected as the screening depth is increased. Among candidates of ferromagnetic semiconductors, Mn-doped ZnO has the advantage that the carrier concentration is independent of the total moment of the film, since the zinc atom is isovalent with manganese [20]. Wang *et al.* [20], [21] showed that the magnetic moment of Mn-doped ZnO can be controlled via Mn doping or the oxygen content inside the film.

In this paper, we report the fabrication and characterization of electric-double-layer transistors (EDLTs) for electric-field manipulation of magnetotransport in n-type Mn-doped ZnO. The devices were characterized by X-ray diffractometry (XRD), atomic force microscopy (AFM), Hall measurements, resistance-switching tests, and magnetotransport measurements. Our results showed the possibility of low-magnitude (≤2 V) gate voltage (V_g) manipulation in the accumulations and depletion of charge carriers in the Mn-doped ZnO films, and the low-temperature magnetotransport can also be manipulated by V_g.

II. EXPERIMENTAL PROCEDURE

The 10 nm-thick $Zn_{0.98}Mn_{0.02}O$ (MZO) thin films were deposited on (0001) Al_2O_3 single crystal substrates by pulsed laser deposition (PLD). The film deposition was performed at 300 °C with an oxygen pressure of 5×10^{-4} Pa. As-grown films were cooled down to room temperature in the same oxygen ambient at a rate of ∼10 °C/min for promoting the oxygen stoichiometry of deposited films. The thickness of as-grown films was estimated by the number of laser pulses used and was further confirmed with AFM.

For electric-field manipulation of MZO devices, the films were patterned into Hall bars (channel width: 50 μm and channel length: 110 μm) by photolithography and wet etching, using dilute HCl (1% HCl, etching rate: 60 nm/min) as etchant [22]. Fig. 1(a) depicts the schematic of the

Manuscript received March 13, 2018; accepted June 16, 2018. Date of publication July 23, 2018; date of current version October 17, 2018. Corresponding author: C. W. Leung (e-mail: dennis.leung@polyu.edu.hk).

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Digital Object Identifier 10.1109/TMAG.2018.2850067