Ionic Liquid Gating Modulation of Diluted Magnetic Semicon-ductor (Zn, Mn)O Thin Films.

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Ionic liquid (IL) gating on functional oxide has drawn significant attention, since it can provide reversible changes in carrier concentration (~1014 cm⁻³) at the interface, permitting the manipulation of electrical and magnetic properties via a low voltage [1-3]. In this work, we demonstrate the electric-field manipulation of transport properties in the dilute magnetic oxide (DMS) of $Zn_{0.98}Mn_{0.02}O$ (MZO) using an electric-double-layer transistor (EDLT) geometry through IL of N,N-diethy1-N-(2-methoxyethy1)-N-methylammonium (DEME+) and bis(trifluoromethylsulfony1)-imide (TFSI-). By the application of different gate voltages (V_g) through the top electrolyte, the accumulated and depleted charge carrier in the MZO channel lead to a reversible control in the transport phenomena at the interface. 10 nm-thick MZO thin films were deposited on (0001) Al₂O₃ single crystal substrates by pulsed laser deposition. The growth of MZO films was conducted at 300°C with an oxygen pressure of 5 x 10⁻⁴ Pa. To study electrical-field manipulation of MZO devices, the films were patterned into Hall bar patterns (channel width: 50 μm, channel length: 110 μm) by photolithography and wet etching using dilute HCl as etchants. Au (50 nm)/Ti (5 nm) coplanar electrodes for IL and contact electrodes for MZO were prepared by electron beam evaporation. Prior to the gating experiment, IL was baked at 80 °C in a high vacuum chamber to get rid of water contamination. A drop of IL was placed on the top of the as-grown film, serving as the top gate electrode. The devices were immediately cooled down to 230 K for the gating process. By applying different V_{σ} (-2, 0 and 2 V), the charge carriers were accumulated and depleted in the channel surface. Then the devices were immediately cooled down to 180 K (below the freezing point of IL at 230 K) before V_g was removed. After the transport measurements, the devices were heated up to 230 K before changing the V_g for another measurement. Fig. 1 illustrates the profile of the longitudinal resistance (R_{xx}) of MZO EDLTs with alternating V_g between -2 and 2 V at 230 K. R_{xx} increases (decreases) sharply upon the application of $V_{\sigma} = -2 \text{ V } (2 \text{ V})$, which is consistent with the scenario of accumulated (depleted) electron charge carrier at the MZO interfaces [4]. Such modulations of R_{xx} are due to electron charge movement in MZO rather than the contribution of gate current; the drain-source current is higher than the gate-source current by at least two orders of magnitude. Magnetotransport behavior of MZO at 10 K after the application of different V_{σ} (-2, 0 and 2 V) are shown in Fig. 2, which shows the magnetoresistance (MR) with out-of-plane applied field for MZO EDLT. Here MR is defined as $MR = (R_{xx}(H) - R_{xx}(0))/R_{xx}(0)$, where $R_{xx}(H)$ and $R_{xx}(0)$ are the R_{xx} values with external magnetic fields of H and zero, respectively. The peak positive MR increases from 0 to 1.8% and the negative-MR (measured at 9 T) decreases from -4.5% to -0.6% when V_{σ} increases from -2 to 2 V. Enhancement in positive MR in the low field regime (<1 T) implies that the ferromagnetic state of MZO is enhanced, as the electron carrier concentration in MZO increases upon switching the $V_{\rm g}$ from -2 to 2 V [5]. The present results, therefore, demonstrate controllable movement of anions and cations in IL by electric-field effect plays an important role in the manipulation of magnetism in the MZO. Financial support by RGC, HKSAR (PolyU 153015/14P) and PolyU (1-ZE25) are acknowledged.

[1] C. Ge, K.J. Jin, L. Gu, L.C. Peng and Y.S. Hu *et al.*, Adv. Mater. Interfaces, 2, 1500407 (2015). [2] B. Cui, C. Song, G. Wang, Y. Yan and J. Peng *et al.*, Adv. Funct. Mater., 24, 7233 (2014). [3] A. Verma, K. Nomoto, W.S. Hwang, S. Raghavan and S. Stemmer *et al.*, Appl. Phys. Lett., 108 183509 (2016). [4] H. Yuan, H. Shimotani, A. Tsukazaki, A. Ohtomo and M. Kawasaki *et al.*, Adv. Funct. Mater., 19, 1046 (2009). [5] L.T. Chang, C.Y. Wang, J. Tang, T. Nie and W. Jiang *et al.*, Nano Lett., 14 1823 (2014).

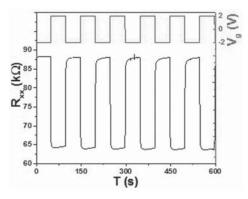


Fig. 1. Time profile of longitudinal resistance (R_{xx}) of MZO EDLTs, during the application of different V_{σ} (profile at top).

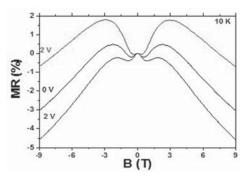


Fig. 2. Magnetoresistance of MZO EDLTs for V_g = -2, 0 and 2 V measured at 10 K.