

# Ambipolar transport and modulation of electronic properties of $\text{Mn}_2\text{CoAl}$ films by ionic liquid gating

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## Abstract

We demonstrate ambipolar transport and modulation of electronic properties in  $\text{Mn}_2\text{CoAl}$  (MCA), one of the most promising candidates for spin gapless semiconductors (SGSs), by using ionic liquid gating in electronic double-layer transistors. The carrier concentration and mobility of MCA films were systematically changed with carrier polarity inversion from p- to n-type by the gating technique. The ambipolar transport is one of the most significant properties of SGSs and strongly promotes the gapless features of MCA.

## 1. Introduction

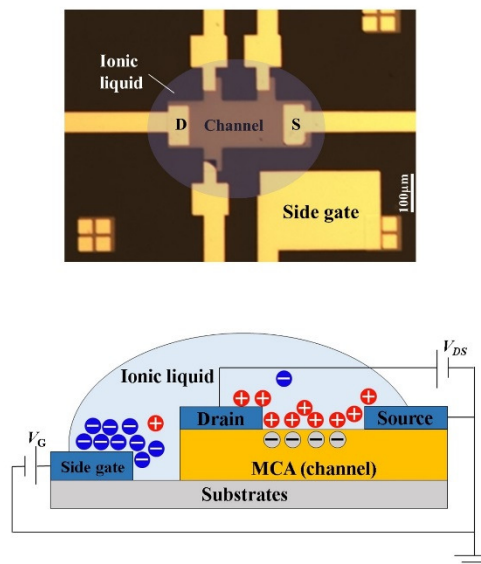
Spin gapless semiconductors (SGSs) are magnetic semiconductors with zero gap at the Fermi level ( $E_F$ ) in one spin channel and the usual energy gap in another spin channel [1]. Because of their special band structures, they have the properties of both gapless semiconductors and half-metals, and show high potential for the fabrication of novel spintronic devices. The main advantages of using SGSs as spintronic devices is that the energy necessary to excite electrons from the valence to the conduction band is negligibly small. The carriers are fully spin-polarized and expected to have very high mobility owing to the large band curvature around  $E_F$ . Highly spin-polarized electrons or holes can be produced by tuning the  $E_F$  of SGSs because they are expected to have ambipolar characteristics. Theoretical calculations have predicted many compounds to be SGSs, however despite all the theoretical calculations, no material has yet been established as an SGS, although SGS-like magnetic and transport properties have been reported for several Heusler alloys.

The inverse Heusler compound  $\text{Mn}_2\text{CoAl}$  (MCA) is one of the most promising candidates for an SGS because SGS-like transport properties (linear MR, high mobility, temperature-independent conductivity, etc.) have already been reported for bulk MCA [2]. We believe SGS-like transport properties do not constitute strong enough evidence for an SGS; rather, the main indicators of an SGS, namely ambipolar transport and full spin-polarization, must be sought after. In this study, we have observed the ambipolar transport properties of MCA by using the ionic liquid gating technique for the first time [3].

## 2. Experimental

MCA films were fabricated on  $\text{MgAl}_2\text{O}_4$  (MAO) substrates at 400°C by Ar ion beam assisted sputtering (IBAS)

[4, 5]. The lattice mismatch between MCA films and MAO is  $\sim 1.5\%$ . Hall bar patterns using thin MCA films ( $\sim 15\text{-}25\text{ nm}$ ) were prepared using a conventional photolithographic process. The channel areas and side-gate electrodes were covered by small droplets of an ionic liquid (DEME-TFSI) to form liquid-gated electronic double-layer transistors (Fig. 1). Transport properties such as longitudinal ( $\rho_{xx}$ ) and Hall resistivity ( $\rho_{xy}$ ) were measured by using the same Hall bar patterns under a magnetic field of 0 - 5 T.



**Fig. 1** Optical micrograph (upper) and schematic (lower) of electric-double-layer transistors (EDLTs) using MCA films.

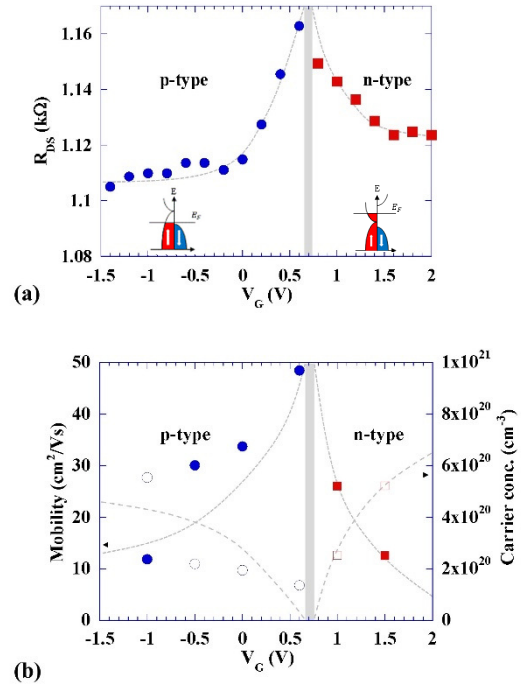
## 3. Results and Discussion

In the out-of-plane X-ray diffraction (XRD) patterns of the MCA films, only (00 $l$ ) peaks of the MCA films, except for substrate peaks, were observed, indicating c-axis orientation of the MCA films. Low glancing angle in-plane XRD measurements ( $\phi$ -scan) revealed that both (400) peaks of the MCA films and MAO substrates have four-fold symmetry, and the  $\phi$  values of the MCA (400) peaks are shifted by 45° with respect to those of the MAO peaks. The  $c$ - and  $a$ -axis lattice constants of the MCA films were estimated to be 0.5764 and 0.5798 nm, respectively, which agree well with that of bulk MCA ( $a = 0.5798\text{ nm}$ ) [2]. The MCA films were almost completely relaxed, presumably because of the effects of the Ar

assist beam. These results indicate the films grew on the MAO substrates epitaxially in the relationship MCA (001) [110] // MAO (001) [100]. The MCA films showed magnetic hysteresis at room temperature. The saturation magnetization ( $M_s$ ) and coercive field ( $H_c$ ) of the MCA films were found to be  $\sim 295$  emu/cc and  $\sim 20$  Oe, respectively. The  $M_s$  and  $H_c$  values of the MCA films were comparable to those of bulk ferrimagnetic MCA [2]. The MCA films showed semiconducting behaviors with very small activation energy of  $\sim 3$  meV and relatively small resistivity of  $\sim 830$   $\mu\Omega\cdot\text{cm}$  for semiconductors, which agree with zero band-gap features of SGS. The electron concentration and mobility of the MCA films formed at  $300^\circ\text{C}$  deduced from Hall resistivity measurements ( $\rho_{xy}$ ) was  $6 \times 10^{20}$   $\text{cm}^{-3}$  and  $3.2$   $\text{cm}^2/\text{V}\cdot\text{s}$  at 4 K, respectively, which were comparable to the reported values for MCA films.

As mentioned above, high-quality epitaxial MCA films were obtained; however, they had lower mobilities and higher hole concentrations than bulk MCA. We believe that one of the most probable reasons for the lower mobility is the shift in the  $E_F$  of the MCA films to the valence band (p-type character) because of nonstoichiometry and atomic swap. The ionic liquid gating technique [6] was employed to control the  $E_F$  of them. MCA films that were thin enough (15–25 nm) to tune  $E_F$  by gating were used to fabricate electronic double-layer transistors (EDLT). Zero-field drain-source resistance ( $R_{DS}$ ) exhibited a sharp maximum at a gate voltage ( $V_G$ ) of  $\sim 0.6$  V, and the drain-source current changed its polarity from positive to negative at  $V_G \approx 0.6$  V [Fig. 2(a)]. This means that the carrier polarity changed from p-type to n-type upon gating. A similarly sharp maximum and current polarity change have often been observed for field-effect-induced ambipolar transport in graphene, which is a typical gapless semiconductor, and the maximum of the  $R_{DS}$ - $V_G$  curve corresponds to the charge neutrality points of gapless semiconductors. These results indicate that ambipolar transport was observed in MCA by the gating technique. At a  $V_G$  of  $\sim 0.6$  V, the mobility showed a maximum, while the carrier concentration showed a minimum [Fig. 2(b)]. The maximum mobility was  $\sim 45$   $\text{cm}^2/\text{V}\cdot\text{s}$ , roughly one order of magnitude higher than the minimum values, but smaller than those reported in bulk MCA [2]. Although the reason for smaller mobility than bulk MCA remains unclear, we believe this is due to interfacial and/or impurity scattering by defects such as Mn vacancies and the lower crystallinity of MCA. The change in carrier polarity from p- to n-type occurred at  $V_G \approx 0.6$  V as well. This behavior agrees well with the  $V_G$  dependence of  $R_{DS}$ , which exhibits cusp features with a sharp maximum at  $V_G \approx 0.6$  V. The Hall results corroborate the ambipolar transport properties of the MCA films.

From these magnetic and electrical measurements, we can conclude that MCA is a gapless semiconductor with finite spin-polarization ( $|P| > 0$ ). The results of this study are also important from the viewpoint of applications because spin-polarized electrons or holes can be generated by tuning the  $E_F$  of MCA through gating, which offers great advantages for the fabrication of novel spintronic devices, such as spin transistors and spin-polarized electron emitters.



**Fig. 2** (a) Zero-field drain-source resistance ( $R_{DS}$ ) as a function of gate voltage ( $V_G$ ) of EDLT using thin MCA films measured at RT. (b)  $V_G$  dependence of carrier concentration (solid circles and squares) and mobility (open circles and squares) of MCA films at RT. The charge neutrality point of MCA is expected to fall within the light gray area, at  $V_G \approx 0.6$  V.

#### 4. Conclusions

MCA films were fabricated and their magnetic and transport properties were investigated. MCA films were epitaxially grown on MAO by IBAS with the epitaxial relationship MCA (001) [110] // MAO (001) [100]. They showed temperature-independent conductivity and magnetic hysteresis, with an  $M_s$  of  $\sim 295$  emu/cc, which is comparable to the  $M_s$  of bulk MCA. The carrier concentration, mobility and polarity (p- or n-type) of the MCA films were effectively controlled by the ion-liquid gating technique and clear ambipolar characteristics were observed. The ambipolar characteristics are among the most significant features of SGS and strongly support the gapless features of MCA. These results indicate that MCA can be used as a spin source for providing both spin-polarized electrons and holes.

#### Acknowledgements

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#### References

- [1] X. L. Wang, Phys. Rev. Lett. **100** (2008) 156404.
- [2] S. Ouardi et al., Phys. Rev. Lett. **110** (2013) 100401.
- [3] K. Ueda et al., Appl. Phys. Lett., in press.
- [4] K. Ueda et al., Appl. Phys. Lett. **103** (2013) 052408.
- [5] M. Nishiwaki et al., J. Appl. Phys., **117** (2015) 17D719.
- [6] H. Du et al., J. Mater. Sci. **50** (2015) 5641.