

$(\text{In}_{0.52}\text{Al}_{0.48})_{1-x}\text{Mn}_x\text{As}$  diluted magnetic semiconductor grown on InP substrates

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## LETTER TO THE EDITOR

**(In<sub>0.52</sub>Al<sub>0.48</sub>)<sub>1-x</sub>Mn<sub>x</sub>As diluted magnetic semiconductor grown on InP substrates****W N Lee<sup>1</sup>, Y F Chen<sup>2</sup>, J H Huang<sup>2</sup>, X J Guo<sup>3</sup>, C T Kuo<sup>1</sup>, T S Chin<sup>2</sup> and H C Ku<sup>4</sup>**<sup>1</sup> Department of Materials Science and Engineering, National Chiao Tung University, Hsinchu 300, Taiwan<sup>2</sup> Department of Materials Science and Engineering, Materials Science Center, National Tsing Hua University, Hsinchu 300, Taiwan<sup>3</sup> Institute of Physics, Academia Sinica, Taipei 11529, Taiwan<sup>4</sup> Department of Physics, National Tsing Hua University, Hsinchu 300, TaiwanE-mail: [wnlee@mx.nthu.edu.tw](mailto:wnlee@mx.nthu.edu.tw) and [jihhuang@mx.nthu.edu.tw](mailto:jhhuang@mx.nthu.edu.tw)

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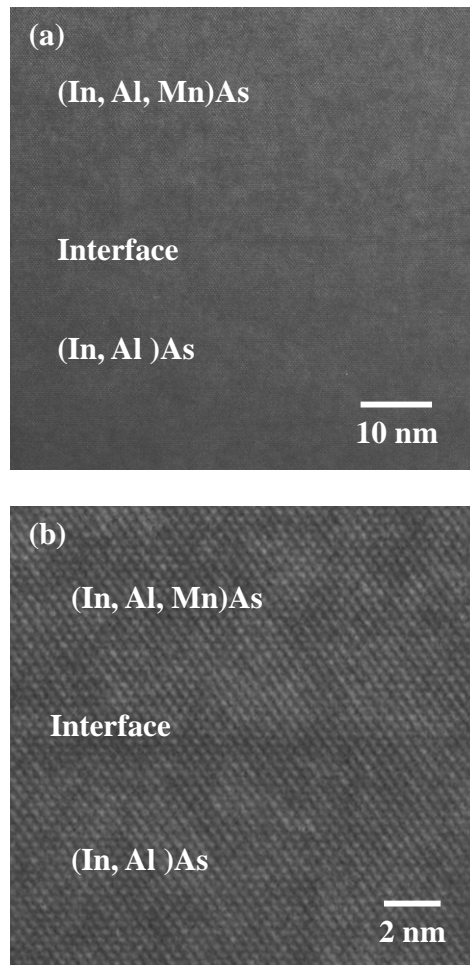
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Online at [stacks.iop.org/JPhysCM/18/L15](http://stacks.iop.org/JPhysCM/18/L15)**Abstract**

A series of diluted magnetic semiconductors, (In<sub>0.52</sub>Al<sub>0.48</sub>)<sub>1-x</sub>Mn<sub>x</sub>As ( $0 < x \leq 0.11$ ), was successfully grown on InP substrate by low-temperature molecular beam epitaxy. Our results indicate that (In<sub>0.52</sub>Al<sub>0.48</sub>)<sub>1-x</sub>Mn<sub>x</sub>As exhibits interesting magnetic behaviours at 5 K: it shows paramagnetic-like behaviour when  $x \leq 0.05$  and ferromagnetic behaviour when  $x \geq 0.06$ .

**1. Introduction**

Recently, III–V diluted magnetic semiconductors (DMSs), e.g. (In, Mn)As and (Ga, Mn)As, have attracted intense attention due to their significance in basic physics and potential application to spintronics devices [1–5]. Since the hole-mediated ferromagnetism [6] in III–V semiconductors is sensitive to both electronic and less understood structural properties, investigation of new diluted magnetic semiconductors is interesting and rewarding [7, 8]. It is well known that most III–V DMSs are ternary and grown on GaAs substrates. Demonstrating the magnetic properties of new quaternary DMSs, e.g. (In, Ga, Mn)As and (In, Al, Mn)As, grown on InP substrates is significant for the study of spin transport effects in devices built on InP substrates. Moreover, such a quaternary magnetic semiconductor has many potential advantages: for example, it is easy to adjust the bandgap energy, easy magnetization axis, and band structure by changing the indium content, which cannot be realized by ternary alloy magnetic semiconductors [8]. Very recently, several groups have focused on Mn-doped (In, Ga)As, and a Curie temperature of 100–130 K, similar to that of (Ga, Mn)As, has been reported in (In, Ga)<sub>1-x</sub>Mn<sub>x</sub>As with  $x \geq 0.10$  [9, 10]. However, there are no reports about Mn doping in (In, Al)As, another important ternary semiconductor grown on InP substrates.



**Figure 1.** Microstructure analysis of sample D: (a) bright-field TEM image and (b) lattice image.

In this letter, we report the growth and magnetic properties of a series of quaternary DMSs,  $(\text{In}_{0.52}\text{Al}_{0.48})_{1-x}\text{Mn}_x\text{As}$ , grown on InP substrates.

## 2. Experiment

The samples used in this study were grown on (001) semi-insulating, epi-ready InP substrates by a Varian Modular GEN-II MBE system. A growth rate of  $0.3 \mu\text{m h}^{-1}$  and a V/III beam equivalent pressure (BEP) ratio of 20 were used. Following native oxide desorption, a 100 nm  $\text{In}_{0.52}\text{Al}_{0.48}\text{As}$  buffer layer was first grown at  $460^\circ\text{C}$  to smoothen the surface. Then, the substrate temperature was lowered to  $220\text{--}230^\circ\text{C}$ . Subsequently, the 100 nm-thick  $(\text{In}_{0.52}\text{Al}_{0.48})_{1-x}\text{Mn}_x\text{As}$  ( $0 < x \leq 0.11$ ) active epilayer was grown. A, B, C, D, E denote five  $(\text{In}_{0.52}\text{Al}_{0.48})_{1-x}\text{Mn}_x\text{As}$  epilayers with  $x = 0.03, 0.05, 0.06, 0.08,$  and  $0.11$ , respectively. The growth was monitored *in situ* with reflection high-energy electron diffraction (RHEED), and a  $(2 \times 4)$  pattern was observed for the  $\text{In}_{0.52}\text{Al}_{0.48}\text{As}$  buffer layers, while it changed to  $(1 \times 2)$  during and after growth of the  $(\text{In}_{0.52}\text{Al}_{0.48})_{1-x}\text{Mn}_x\text{As}$  epilayer. After growth, the wafers

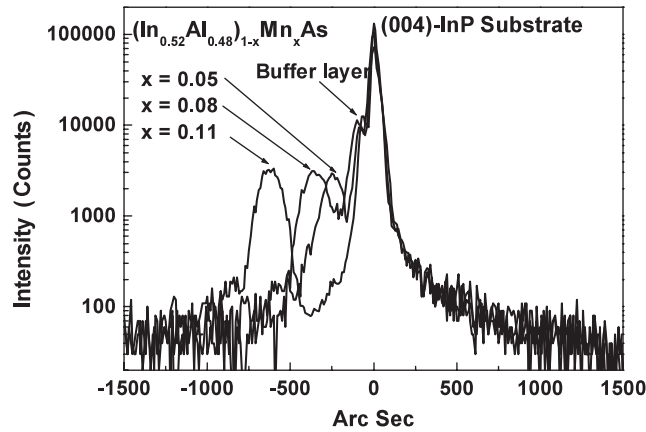


Figure 2. The DXRD rocking curves of (004) for samples B, D, and E.

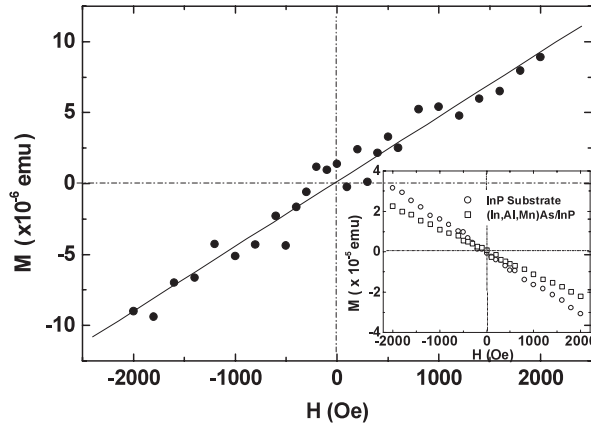
were cleaved into a number of pieces for various characterizations. The crystalline structure of  $(\text{In}_{0.52}\text{Al}_{0.48})_{1-x}\text{Mn}_x\text{As}$  epilayers was examined by double-crystalline x-ray diffraction (DXRD) and transmission electron microscopy (TEM). Cross-sectional samples parallel to the (110) plane were prepared conventionally by mechanical thinning and Ar-ion milling for TEM observation. Mn concentrations were determined by electron microprobe analysis (EMPA). Magnetic measurements were carried out in a commercial superconducting quantum interference device (SQUID) magnetometer.

### 3. Results and discussion

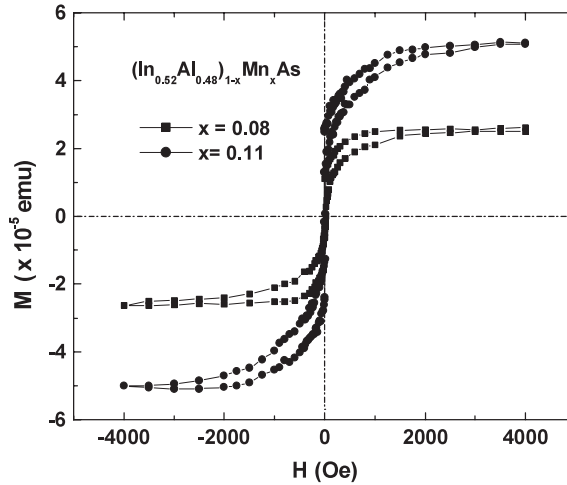
The cross-sectional image projected along the [110] zone of sample D is shown in figure 1. The quality of the interface between the  $(\text{In}_{0.52}\text{Al}_{0.48})_{0.92}\text{Mn}_{0.08}\text{As}$  epilayer and  $\text{In}_{0.52}\text{Al}_{0.48}\text{As}$  buffer layer is good. Both RHEED patterns and TEM measurements confirmed that the epitaxial  $(\text{In}_{0.52}\text{Al}_{0.48})_{1-x}\text{Mn}_x\text{As}$  active layer has a zinc blende structure.

Figure 2 selectively shows the DXRD rocking curves of (004) for samples B, D, and E with  $x = 0.05, 0.08,$  and  $0.11,$  respectively. Obvious peak separation due to the incorporation of Mn atoms was observed. The lattice expansion is about 0.20, 0.29, and 0.50% for samples B, D, and E, respectively, increasing with the increase in Mn content.

All samples of magnetization were measured with the  $[\bar{1}10]$  direction in-plane magnetic field. The total magnetization of  $(\text{In}_{0.52}\text{Al}_{0.48})_{1-x}\text{Mn}_x\text{As}/\text{InP}$  samples includes two components, i.e., the magnetization of the  $(\text{In}_{0.52}\text{Al}_{0.48})_{1-x}\text{Mn}_x\text{As}$  epilayer and the InP substrate. Therefore, in order to obtain the ‘net’ magnetization of the active layer, the magnetization of a ‘bare’ InP substrate of the same size must be separately measured and carefully subtracted from the total magnetization. The inset of figure 3 shows the magnetization as a function of applied field of  $(\text{In}_{0.52}\text{Al}_{0.48})_{0.95}\text{Mn}_{0.05}\text{As}/\text{InP}$  and corresponding ‘bare’ InP substrate of the same size. We can find that the  $M-H$  curve of the InP substrate shows almost a linear relationship, which suggests that the semi-insulating InP substrate exhibits diamagnetic behaviour, similar to that of a semi-insulating GaAs substrate. Compared with that of the InP substrate, the  $M-H$  curve of  $(\text{In}_{0.52}\text{Al}_{0.48})_{0.95}\text{Mn}_{0.05}\text{As}/\text{InP}$  shows a similar linear relationship with a smaller absolute slope. Figure 3 shows the ‘net’ magnetization as a function of applied field for the  $(\text{In}_{0.52}\text{Al}_{0.48})_{0.95}\text{Mn}_{0.05}\text{As}$  epilayer at 5 K after carefully subtracting the diamagnetic signal of the InP substrate. There is a good coincidence between the linear fit and



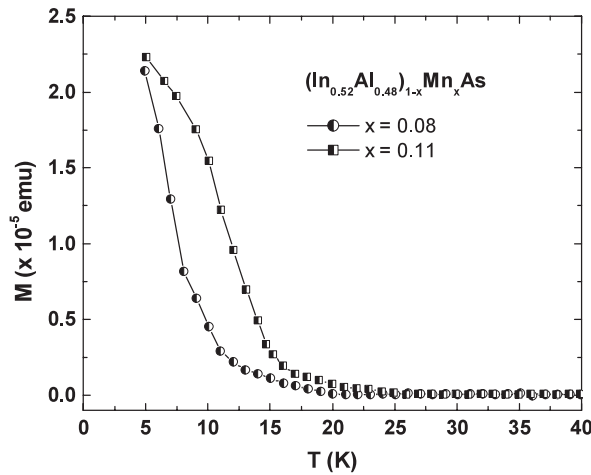
**Figure 3.** Magnetization as a function of in-plane applied field at 5 K for the  $(\text{In}_{0.52}\text{Al}_{0.48})_{0.95}\text{Mn}_{0.05}\text{As}$  epilayer. Closed circles stand for 'net' magnetization after subtracting the magnetization of the InP substrate and the solid line is the results of a linear fit of the experimental data. The inset shows the applied field dependence of magnetization at 5 K for the  $(\text{In}_{0.52}\text{Al}_{0.48})_{0.95}\text{Mn}_{0.05}\text{As}/\text{InP}$  substrate and 'bare' InP substrate of the same size.



**Figure 4.** Magnetization as a function of in-plane applied field at 5 K for samples D and E.

experimental data. Therefore, we can regard that sample B ( $(\text{In}_{0.52}\text{Al}_{0.48})_{0.95}\text{Mn}_{0.05}\text{As}$ ) shows paramagnetic-like behaviour at 5 K. It should be noted that a similar phenomenon was also observed in sample A with  $x = 0.03$ .

Samples with  $x \geq 0.06$  showed a ferromagnetic state at 5 K, as confirmed by the  $M-H$  curves. For example, the  $M-H$  curves of samples D and E are shown in figure 4. Obviously, both epilayers exhibit typical ferromagnetic state. In addition, the coercivity of  $(\text{In}_{0.52}\text{Al}_{0.48})_{1-x}\text{Mn}_x\text{As}$  is very low ( $< 15$  G). We also measured the field cooling  $M-T$  curves of the epilayers with the  $[110]$  direction in-plane applied magnetic field ( $H = 100$  Oe). Figure 5 selectively shows the  $M-T$  curves for samples D ( $x = 0.08$ ) and E ( $x = 0.11$ ) epilayers; the corresponding Curie temperatures are 20, and 25 K, respectively.



**Figure 5.** Temperature dependence of magnetization for samples D and E measured with the  $[\bar{1}10]$  direction in-plane applied magnetic field ( $H = 100$  Oe).

The reason for  $(\text{In}_{0.52}\text{Al}_{0.48})_{1-x}\text{Mn}_x\text{As}$  exhibiting paramagnetic-like behaviour for  $x \leq 0.05$  and ferromagnetic behaviour for  $x \geq 0.06$  can be simply analysed as follows. Liu *et al* reported a similar paramagnetic-like phenomenon observed in  $\text{Al}_{0.96}\text{Mn}_{0.04}\text{As}$  [11]. Their electrical transport measurements indicate a deep acceptor level of Mn inside AlAs, resulting in a lack of free carriers and semi-insulating and hence paramagnetic behaviour [11]. In the light of this viewpoint, we perceive that the Mn atoms in  $(\text{In}_{0.52}\text{Al}_{0.48})_{1-x}\text{Mn}_x\text{As}$  can act as deep-level acceptors depreciating the free holes and hence resulting in paramagnetic behaviour. When  $x \geq 0.06$ , the increased Mn atoms tend to form ‘shallow’ level acceptors and provide free holes, thus leading to ferromagnetic exchange interaction. Further electrical and magnetic transport measurements must be done to provide deeper insight into this phenomenon and prove the assumption.

#### 4. Conclusions

We have successfully grown a series of diluted magnetic semiconductors with zinc blende structure,  $(\text{In}_{0.52}\text{Al}_{0.48})_{1-x}\text{Mn}_x\text{As}$  ( $0 < x \leq 0.11$ ), on InP substrate by low-temperature molecular beam epitaxy. The lattice expansion increases with the increase in Mn content.  $(\text{In}_{0.52}\text{Al}_{0.48})_{1-x}\text{Mn}_x\text{As}$  reveals a paramagnetic-like state when  $x \leq 0.05$ , and a ferromagnetic state when  $x \geq 0.06$ .

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