

Epitaxial growth and properties of n-type magnetic semiconductor (In,Co)As

Nguyen Thanh Tu, Le Duc Anh, Pham Nam Hai, and Masaaki Tanaka

Department of Electrical Engineering & Information Systems, The University of Tokyo

7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan.

Phone: +81-3-5841-6773 E-mail: nguyen@cryst.t.u-tokyo.ac.jp

Abstract

A new n-type magnetic semiconductor ($\text{In}_{1-x}\text{Co}_x$)As has been successfully grown by low-temperature molecular beam epitaxy (LT-MBE) on GaAs (001) substrates. Reflection high energy electron diffraction (RHEED) patterns indicate that (In,Co)As layers with the Co concentration $x = 3 - 21\%$ remain zinc-blende crystal structure. The electron concentration can be controlled between $2.4 \times 10^{18} - 4.9 \times 10^{19} \text{ cm}^{-3}$ by changing the Co concentration. The insulator-metal transition is observed at $x = 5\%$. Large negative magnetoresistance (up to 17.5% at 9.5kG) is observed at 5K and can be attributed to spin-disorder scattering.

1. Introduction

Magnetic semiconductors (MSs) have attracted much attention due to their ability of exploiting both magnetic and semiconducting properties for future low-power consumption electronic devices, such as non-volatile magnetic memory or reconfigurable logic using spin transistors. Among MS families, Mn doped III-V based MS such as (In,Mn)As [1] and (Ga,Mn)As [2][3] are intensively studied. However, these materials are only p-type and their Curie temperatures are still much lower than room temperature. For realizing spintronic devices, not only p-type but also n-type MS materials are required. Recently, Fe-based n-type III-V ferromagnetic semiconductor (In,Fe)As was successfully grown and shown to exhibit surprisingly large s - d exchange interaction [4], but its origin is still unclear. In this paper, we present the growth and properties of a new n-type MS, (In,Co)As. We compare its magnetic and transport properties with other InAs based MS to give a possible explanation for the observed trend of magnetism in InAs based MSs.

2. Molecular beam epitaxial growth

($\text{In}_{1-x}\text{Co}_x$)As layers were grown on semi-insulating GaAs (001) substrates by LT-MBE. First we grew a 10nm-thick InAs buffer layer at high temperature (520°C) to help relax quickly the lattice mismatch between InAs ($a = 0.605\text{nm}$) and GaAs ($a = 0.565\text{nm}$) and obtain a smooth InAs surface. Then, we grew a (In,Co)As layer (thickness $d = 30 - 100\text{nm}$) at a typical growth rate of $0.5\mu\text{m/h}$ under In-stabilized conditions. The substrate temperature was kept constant at 200°C . Finally, a 5nm-thick InAs cap layer was grown to prevent oxidation of the (In,Co)As layer. The cobalt concentration x was changed in the range of 3 - 21%.

RHEED patterns of the (In,Co)As layers after the MBE growth are spotty as shown in Fig. 1. Due to the low growth temperature, the surface becomes rough and the

growth mode changes from two to three dimensional as the (In,Co)As thickness increases. Nevertheless, the RHEED patterns clearly show zinc-blende crystal structure without any second phase or polycrystals. This result indicates that high Co concentration far above the Co solubility limit can be realized in (In,Co)As by LT-MBE.

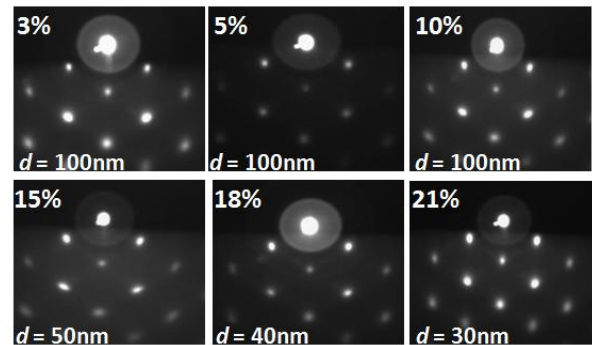


Fig.1 RHEED patterns of the (In,Co)As layers (thickness $d = 30 - 100\text{nm}$) after the MBE growth on GaAs substrates.

3. Electrical and magneto-optical properties of (In,Co)As

Figure 2 shows the Hall resistance R_{Hall} measured at room temperature for many samples with different Co concentrations. The result shows that all the samples are n-type. The estimated electron concentration n , mobility μ , and resistivity ρ are listed in Table I. One can see that the electron concentration can be controlled from 2.4×10^{18} to $4.9 \times 10^{19} \text{ cm}^{-3}$ by increasing the Co concentration. This result may be explained by the increasing of defects such as anti-site As due to the high Co concentrations. The highest electron concentration was obtained at $x = 15\%$.

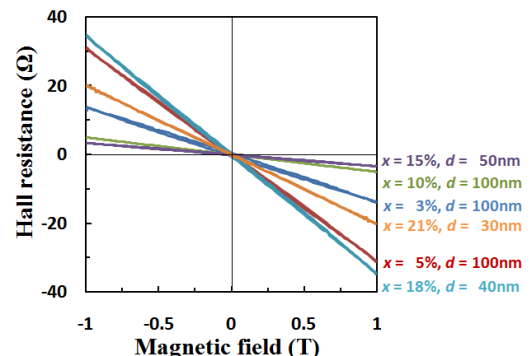


Fig. 2 The Hall resistance at 300K for (In,Co)As samples with different Co concentrations ($x = 3 - 21\%$).

Resistivity was measured at 5 - 300 K. All the samples showed metallic behavior except for the sample with $x = 3\%$. This indicated that the insulator-metal transition occurs at $x = 5\%$.

Table I Electron concentration n , mobility μ , resistivity ρ at 300K, and magnetoresistance MR (defined as $[\rho(9.5 \text{ kG}) - \rho(0)]/\rho(0)$) at 5K of $(\text{In}_{1-x}\text{Co}_x)\text{As}$ with various Co concentrations ($x = 3 - 21\%$), where $\rho(H)$ is the resistivity at the magnetic field of H .

x (%)	n (cm^{-3})	μ (cm^2/Vs)	ρ (Ωcm)	MR (%)
3	5.5×10^{18}	16.3	7.0×10^{-2}	-
5	2.4×10^{18}	134.7	1.9×10^{-2}	-1.5
10	1.4×10^{19}	25.8	1.8×10^{-2}	-5.8
15	4.9×10^{19}	19.2	6.6×10^{-3}	-7.6
18	5.3×10^{18}	20.3	5.8×10^{-2}	-17.5
21	1.2×10^{19}	18.7	2.7×10^{-2}	-5.0

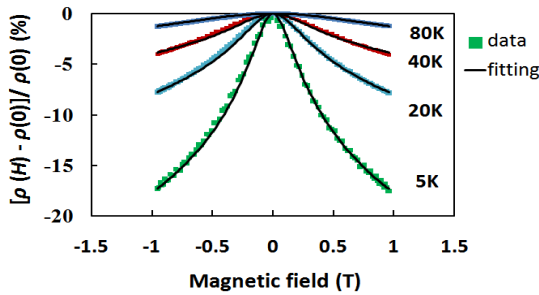


Fig. 3 Magnetoresistance $[\rho(H) - \rho(0)]/\rho(0)$ as a function of applied magnetic field for $(\text{In}_{1-x}\text{Co}_x)\text{As}$ with $x = 18\%$.

Table II Fitting parameters and MR of the sample with $x = 18\%$ at different temperatures.

$T(\text{K})$	MR (%)	B_1	B_2 (T^{-1})
5	-17.5	4.5	50.0
20	-7.8	2.5	23.0
40	-4.0	1.4	15.9
80	-1.2	1.2	2.0

Figure 3 shows the magnetoresistance (MR), that is, resistance as a function of applied magnetic field at different temperatures for a sample with $x = 18\%$. At 5K, the negative MR is as large as -17.5% at 9.5kG. The magnitude of MR decreased as the temperature increased. We fit the MR curves using the spin-disorder scattering theory. The negative MR is fitted by the semiempirical equation (1) proposed by Khosla and Fischer[5] based on the scattering between carriers and localized spin of impurity atoms.

$$\frac{\Delta\rho}{\rho} = -B_1^2 \ln(1 + B_2^2 H^2). \quad (1)$$

$$B_1 \propto J\rho_F[S(S+1) + \langle M^2 \rangle]. \quad (2)$$

$$B_2 \propto \frac{1}{T}. \quad (3)$$

Here, B_1 and B_2 are fitting parameters, H is magnetic field, J is the exchange interaction energy, ρ_F is the density of states at the Fermi energy, $\langle M \rangle$ is the average magnetization, T is temperature and S is the spin of the localized magnetic moment. The fitting parameters at different temperatures were summarized in Table II. The fitting reproduced the observed MR almost perfectly at all temperatures, suggesting that the MR was induced by spin disorder scattering. The magnitude of negative MR at 5K for different samples was summarized in Table I.

In order to investigate the magneto-optical properties of $(\text{In},\text{Co})\text{As}$ layers, we measured the reflection magnetic circular dichroism (MCD) spectra, and the MCD spectrum

of the sample with $x = 5\%$ is shown in Fig.4. The spectrum shows two peaks corresponding to the optical critical point energies of the InAs band structure at E_1 (2.61eV), E_2 (4.74eV). We see no broad background which would be observed if metallic Co nanoclusters existed. However, the magnitude of MCD is small, and MCD intensity- H hysteresis shows no evidence of ferromagnetism or superparamagnetism up to $x = 21\%$, but all the samples are paramagnetic. This indicates that $(\text{In},\text{Co})\text{As}$ layers are free of ferromagnetic Co nanoclusters or other ferromagnetic Co-As intermetallic compounds. At the same time, this implies that the s - d exchange interaction is quite weak to induce ferromagnetism in $(\text{In},\text{Co})\text{As}$, and this is contrasting to the large s - d exchange interaction observed in $(\text{In},\text{Fe})\text{As}$. We suggest that the difference in magnetism between $(\text{In},\text{Co})\text{As}$ and $(\text{In},\text{Fe})\text{As}$ may be due to the difference in the relative energy position of the d level from the conduction band. In $(\text{In},\text{Fe})\text{As}$, the d level is close to the bottom of the conduction band so that the s - d exchange interaction is large and ferromagnetism can be induced [4]. In contrast, the d level in $(\text{In},\text{Co})\text{As}$ may lie much deep under the valance band of InAs as predicted by the first principles calculation [6]. Therefore, the s - d exchange interaction in $(\text{In},\text{Co})\text{As}$ is too small to induce ferromagnetic order. This means that the position of the d level of the transition metals in the host semiconductor band structure may play an important to induce ferromagnetism.

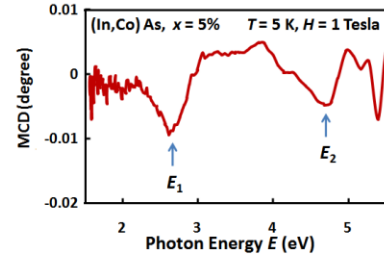


Fig. 4 Reflection MCD spectra of an $(\text{In},\text{Co})\text{As}$ sample with $x = 5\%$, measured at 5 K under a magnetic field of 1 Tesla.

4. Conclusions

We have successfully grown a new n-type magnetic semiconductor $(\text{In},\text{Co})\text{As}$. The electron concentration can be changed from 2.4×10^{18} up to $4.9 \times 10^{19} \text{ cm}^{-3}$ by changing the Co concentration. The $(\text{In},\text{Co})\text{As}$ layers with high Co doping level are metallic and have large negative MR. From the MCD analysis, $(\text{In},\text{Co})\text{As}$ is paramagnetic and has the bandstructure of zinc-blende type semiconductors.

Acknowledgments

This work is supported by Grant-in-Aids for Scientific Research including Specially Promoted Research, the Specially Promoted Research, the Project for Developing Innovation Systems of MEXT, and FIRST Program of JSPS.

References

- [1] H. Munekata *et al.*, Phys. Rev. Lett.63, 1849 (1989).
- [2] H. Ohno *et al.*, Appl. Phys. Lett. 69, 363 (1996).
- [3] T. Hayashi *et al.*, J. Cryst. Growth 175/176,1063 (1997).
- [4] P. N.Hai *et al.*, Appl. Phys. Lett.101, 252410 (2012); P. N.Hai *et al.*, Appl. Phys. Lett. 101, 182403 (2012); P. N. Hai *et al.*, Appl. Phys. Lett. 100, 262409 (2012).
- [5] R. P. Khosla and J. R. Fischer, Phys. Rev. B2, 4084 (1970).
- [6] K.Sato *et al.*, Rev. Mod. Phys. 82, 1633 (2010).