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Extended Abstracts of the 2015 International Conference on Solid State Devices and Materials, Sapporo, 2015, pp1220-1221

Magnetic properties and intrinsic ferromagnetism in narrow-gap ferromagnetic semiconductor (Ga,Fe)Sb

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Abstract

A p-type ferromagnetic semiconductor (Ga$_{1-x}$,Fe$_x$)Sb (x = 3.9 – 20%) has been grown by low-temperature molecular beam epitaxy (MBE). Crystal structure analyses by scanning transmission electron microscopy (STEM) and transmission electron diffraction (TED) indicate that the (Ga$_{1-x}$,Fe$_x$)Sb thin films maintain the zinc-blende crystal structure up to x = 20%. The intrinsic ferromagnetism was confirmed by magnetic circular dichroism (MCD) spectroscopy and anomalous Hall effect (AHE) measurements. The Curie temperature T_C of (Ga$_{1-x}$,Fe$_x$)Sb depends on x and the hole concentration as in the case of other ferromagnetic semiconductors with hole-induced ferromagnetism. The highest T_C reaches 230 K at x = 20%, which is the highest value so far reported in III-V ferromagnetic semiconductors.

1. Introduction

Development of new ferromagnetic semiconductors (FMSs) is an important issue in the emerging field of "semiconductor spintronics". Although Mn-doped III-V FMSs, such as (In,Mn)As and (Ga,Mn)As, have been intensively studied, the maximum Curie temperature T_C of (Ga,Mn)As (200 K) and (In,Mn)As (90 K) are still much lower than room temperature [1,2]. Furthermore, the origin of ferromagnetism of Mn-based FMSs is under debate [3]. Recently, a new Fe-based n-type III-V FMS (In,Fe)As was successfully grown and exhibited surprisingly large *s-d* exchange interaction [4,5]. Furthermore, very recently, we have successfully grown a new p-type Fe-doped FMS (Ga,Fe)Sb. Notably, T_C of (Ga$_{1-x}$,Fe$_x$)Sb reaches 140 K at x = 13.7%, which is the highest T_C in narrow-gap III-V FMSs [6]. The successful growth of n-type (In,Fe)As and p-type (Ga,Fe)Sb FMSs opens a new opportunity to fabricate all-FMS spintronic devices.

In this paper, we systematically investigate the crystal structure, magneto-optical properties, magnetization, and magneto-transport properties of (Ga$_{1-x}$,Fe$_x$)Sb (x = 3.9 – 20%). The highest T_C observed in (Ga,Fe)Sb reaches 230 K for the sample with x = 20%, indicating that Fe-based FMSs are very promising for realization of high-T_C FMSs.

2. Crystal structure analysis

The (Ga$_{1-x}$,Fe$_x$)Sb samples with various Fe concentration x = 3.9 – 20% and thicknesses d = 30 – 100nm were grown on GaAs substrate at 250°C by LT-MBE. The studied (Ga$_{1-x}$,Fe$_x$)Sb samples are listed in Table I. Reflection high-energy electron diffraction (RHEED) patterns of the (Ga,Fe)Sb layers during the MBE growth are streaky with surface reconstruction of (1×3), which is similar to that of GaSb grown at high temperature. This indicates that (Ga,Fe)Sb layers preserve the zinc-blende-type crystal structure and have an atomically smooth surface. Figures 1(a) and 1(b) show high-resolution STEM images of two representative samples with x = 13.7% and 20% projected along the [110] axis. The inset of Figs. 1(a) and 1(b) show the transmission electron diffraction (TED) patterns of these samples. The STEM images and TED patterns indicate that the crystal structure of (Ga,Fe)Sb layers are of zinc-blende type without any visible second phases.

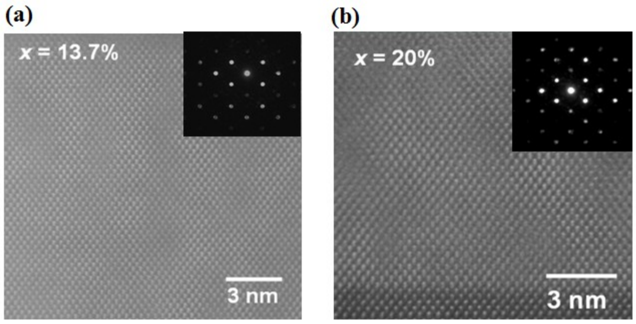

Fig. 1 (a) and (b) STEM images and TED patterns of (Ga$_{1-x}$,Fe$_x$)Sb samples with x = 13.7% and 20%, respectively.

Table I Thickness d, Curie temperature T_C, and hole concentration p at 300 K of (Ga$_{1-x}$,Fe$_x$)Sb samples with x = 3.9 – 20%.

x (%)	d (nm)	T_C (K)	p (cm^{-3})
3.9	100	20	4.4×10^{18}
6.7	100	27	7.8×10^{18}
9.0	100	50	1.3×10^{19}
11.4	100	80	4.0×10^{19}
13.7	100	140	4.6×10^{19}
17.0	40	180	-
20.0	30	230	-

3. Magneto-optical properties.

Figure 2(a) shows the MCD spectra of the (Ga$_{1-x}$,Fe$_x$)Sb samples (x = 3.9 – 20%) and a reference undoped GaSb sample at 5 K with a magnetic field H of 1 T applied perpendicular to the film plane. The MCD spectra of (Ga,Fe)Sb show strongly enhanced peaks at E_1 (2.19 eV) and $E_1 + \Delta_1$ (2.63 eV) corresponding to the optical critical point energies of the GaSb band structure [7], indicating that (Ga,Fe)Sb maintains the zinc-blende crystal structure with large spin-split band structure due to the *s,p-d* exchange interactions. Figures 2(b) and 2(c) show the MCD-*H* characteristics of the samples with x = 17% and 20%, respectively at different temperatures. Clear hysteresis curves were observed at low temperature, demonstrating the presence of ferromagnetic order at low temperature.

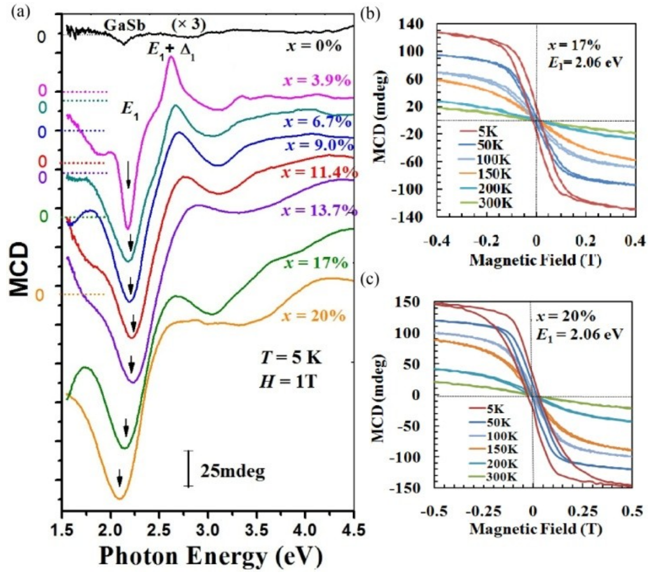

Fig. 2 (a) Reflection MCD spectra measured at 5 K under a magnetic field of 1 Tesla applied perpendicular to the film plane for ($Ga_{1-x}Fe_x$)Sb samples with x = 3.9 – 20%. The MCD spectrum of a reference undoped GaSb sample is also shown. (b) and (c) MCD-*H* characteristics measured at a photon energy of 2.19 eV of (Ga,Fe)Sb samples with x = 17% and 20%, respectively.

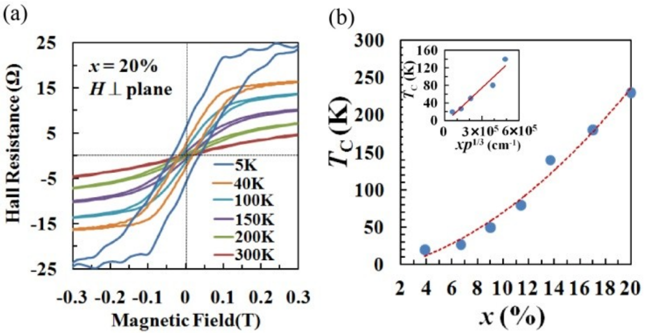

Fig. 3 (a) Hall resistance R_{Hall} vs. magnetic field H measured at different temperatures of the sample x = 20%. (b) T_C vs. Fe concentration x. Inset shows T_C vs. $xp^{1/3}$.

Next, we investigate the magneto-transport properties of the (Ga,Fe)Sb samples. The hole concentrations p estimated by Hall effect measurements at 300 K are listed in the 4th columns of Table I. For the samples with $x \geq 17\%$, we cannot estimate p due to the influence of the anomalous Hall effect even at 300 K. One can see that p increases from 4.4×10^{18} to 4.6×10^{19} cm^{-3} as x increases. This result may be explained by the increase of native acceptor defects due to the Fe doping, such as anti-site Ga. Figure 3(a) shows the Hall resistance vs. magnetic field (R_{Hall}–H) characteristics at various temperatures of the sample with x = 20%. At low temperatures, R_{Hall} are dominated by the anomalous Hall effect (AHE) with clear hysteresis curves, consistent with the MCD-*H* characteristics, thus supporting intrinsic ferromagnetism of (Ga,Fe)Sb. At 300 K, the R_{Hall}–H characteristics are linear with positive slopes, indicating that all the samples are p-type. The T_C values estimated by the Arrott plots of MCD – H characteristics and/or the AHE of all samples are listed in Table I and plotted as a function of x in Fig. 3(b). One can see that T_C increases as x increases. However, T_C is not linearly proportional to x. Instead, T_C is proportional to $xp^{1/3}$ as shown in the inset of Fig. 3(b). This indicates that T_C also depends on p as in the case of other FMSs with hole-induced ferromagnetism. Note that the obtained T_C (230 K) at x = 20% is the highest in III-V FMSs. To explain the ferromagnetism of (Ga,Fe)Sb, we have proposed a "resonant *s,p-d* exchange interaction" model in Fe-based narrow gap FMSs [4,5], in which the position of the *d*-level of the transition metals in the host semiconductor band structure plays an important role to induce the ferromagnetism [8].

4. Conclusions

P-type ferromagnetic semiconductor ($Ga_{1-x}Fe_x$)Sb (x = 3.9 – 20%) thin films were successfully grown by LT-MBE and show intrinsic ferromagnetism. The obtained T_C (230 K) of (Ga,Fe)Sb (x = 20%) is the highest in III-V FMSs, demonstrating that Fe-doped FMSs are promising for semiconductor spintronic devices. Our results suggest that the position of the *d*-level of the transition metals in the host semiconductor band structure plays an important role to induce ferromagnetism.

Acknowledgements

This work is supported by Grant-in-Aids for Scientific Research including the Specially Promoted Research and the Project for Developing Innovation Systems of MEXT. Part of this work was carried out under the Cooperative Research Project Program of RIEC, Tohoku University. P.N.H acknowledges supports from the Yazaki Memorial Foundation for Science and Technology, the Murata Science Foundation, and the Toray Science Foundation.

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