Large magnetoresistance of Ge_{1-x}Mn_x single films and heterostructures with magnetic nanocolumns

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1. Introduction

For Si-based spin-electronic applications, group-IV compatible materials, such as Si- and Ge-based ferromagnetic semiconductors (FSs), have been investigated for the past decade. Since Ge plays an important role for advanced CMOS devices [1], Mn doped Ge ($Ge_{1-x}Mn_x$) [2-7] has been investigated as a Ge based FS with considerable attention.

In our previous study, it was shown that the origin of ferromagnetism in $\text{Ge}_{1-x}\text{Mn}_x$ is nano-sized columnar precipitation of amorphous $\text{Ge}_{1-y}\text{Mn}_y$ with high Mn content *y* [3], which means that $\text{Ge}_{1-x}\text{Mn}_x$ films investigated so far were not FS. Here, *x* is the average Mn content in the whole film and *y* is the local Mn content in the precipitated nanocolumns, and x < y. Nanocolumnar structures in GeMn were widely reported [4-7], where specific features were observed such as positive magnetoresistance (MR) [4, 5], high T_C [4], and controllability of the magnetization of nanocolumns [6]. Although $\text{Ge}_{1-x}\text{Mn}_x$ with nanocolumns is not FS, it may be used as a group-IV based functional spin-electronic material.

In this paper, we investigate the magneto-transport properties of single $Ge_{1-x}Mn_x$ films and magnetic-tunnel-junction-like heterostructures, which consist of $Ge_{1-x}Mn_x$ / SiGe / $Ge_{1-x}Mn_x$, where $Ge_{1-x}Mn_x$ contains $Ge_{1-y}Mn_y$ nanocolumns.

2. Sample preparation

We prepared single $\text{Ge}_{1-x}\text{Mn}_x$ films and heterostructures using $\text{Ge}_{1-x}\text{Mn}_x$ with nanocolumns, which are grown on Ge (001) substrates ($p \sim 1 \times 10^{18} \text{ cm}^{-3}$) by low temperature molecular beam epitaxy (LT-MBE). A 30-nm-thick single $\text{Ge}_{1-x}\text{Mn}_x$ film with x = 0.11 and 0.28 was grown on a 15-nm-thick Ge buffer layer at a growth rate $R_G = 150$ nm/h and a substrate temperature $T_S = 100^{\circ}\text{C}$, in which $\text{Ge}_{1-y}\text{Mn}_y$ nanocolumns are known to be formed. Fig. 1 shows (a) a schematic sample structure of the single films and (b) a cross-sectional TEM image of a single $\text{Ge}_{0.94}\text{Mn}_{0.06}$ film grown at $R_G = 150$ nm/h and $T_S = 100^{\circ}\text{C}$, where amorphous $\text{Ge}_{0.91}\text{Mn}_{0.09}$ nanocolumns and surrounding $\text{Ge}_{0.97}\text{Mn}_{0.03}$ matrix are formed.

We also grew heterostructures using $Ge_{1,x}Mn_x$, whose schematic structures (S0367 and S0373) are shown in fig. 2. As the bottom magnetic layer, a 30-nm-thick $Ge_{0.94}Mn_{0.06}$ film was grown at $R_G = 150$ nm/h and $T_S = 100^{\circ}C$. The barrier layer was a 6-nm-thick $Si_{0.2}Ge_{0.8}$ film grown at $R_G =$ 30 nm/h and $T_S = 100^{\circ}C$. The top magnetic layer was a 30-nm-thick Ge_{0.89}Mn_{0.11} (S0367) and Ge_{0.72}Mn_{0.28} (S0373) films grown at $R_{\rm G} = 150$ nm/h and $T_{\rm S} = 200^{\circ}$ C. (In this growth condition, Mn₅Ge₃ intermetallic clusters with surrounding Ge matrix are formed in a Ge_{1-x}Mn_x film.) After growing the single films and heterostructures, we fabricated mesa diodes 200-µm in diameter with a Pt contact and a 50-nm-thick SiO₂ passivation layer.

3. Magneto-transport of the single Ge_{1-x}Mn_x films

Fig. 3 shows *I-V* characteristics of the single film $(Ge_{0.89}Mn_{0.11} \text{ and } Ge_{0.72}Mn_{0.28})$ mesas at 15 K. Although the *I-V* characteristics at room temperature showed ohmic contact behavior, strong rectification was observed at 15 K, which is probably caused by the Schottky contact between the Ge_{1-x}Mn_x and Pt layers.

Fig. 4 shows magnetic field dependence of resistance (MR curve) of the single $Ge_{0.89}Mn_{0.11}$ and $Ge_{0.72}Mn_{0.28}$ film mesas measured at 15 K and applied bias of -5 mV. The vertical axis of fig. 4 shows MR ratio, which is defined as

$$(R - R_0)/R_0 \times 100(\%),$$
 (1)

where *R* is the resistance of the sample and R_0 is the minimum resistance in the measured range of a magnetic filed (-0.8 - 0.8 T). In fig. 4, large positive MR was observed in both single film samples, the MR ratio increased with increasing *x*. Bias voltage dependence of the MR ratio of the Ge_{0.89}Mn_{0.11} mesa is also shown in fig. 4. The MR ratio at 0.8 T is higher than 2000% at a bias of -1 mV.

Positive MR is often observed in granular systems and nano-/micro-composites. Although the mechanism of the present large positive MR is not clear, this is related to nanocolumnar structure of the $Ge_{1-x}Mn_x$ films.

4. Magneto-transport of the $Ge_{1-x}Mn_x$ heterostructures with nanocolumns

Fig. 3 also shows *I-V* characteristic of one of the heterostructure mesas (S0367) measured at 15 K. Negative bias rectification was also dominant in *I-V* characteristics.

Fig. 4 shows MR curves of the heterostructure mesa (S0367) measured at 15 K at various biases of -20, -30, and -40 mV. Large positive MR was also observed, which may originate from the bottom nanocolumnar layer. Fig. 5 shows MR curves of the heterostructure mesa (S0373) measured at 15 K at various biases of -5, -10, -15, -20, and -40 mV. (Note that offsets were added on some MR curves in fig. 5 for viewability.) In addition to the large

positive MR, hysteretic behavior was observed in the MR curves at the biases less than -20 mV. Black and gray arrows in fig. 5 indicate scanning directions of magnetic field for the MR curve at a bias of -5 mV.

The origin of the hysteretic behavior is not clear. However, some relation exists between the hysteretic behavior and coercive forces of the top and bottom layers. The coercive force of a $Ge_{0.94}Mn_{0.06}$ film grown with the same growth condition as the bottom layer of the heterostructures measured by magnetic circular dichroism (MCD) was very small (lower than 0.01 T), which corresponds to the sharp decrease of the MR with increase of magnetic field around 0 T. The coercive of a Ge_{0.72}Mn_{0.28} film with Mn₅Ge₃ precipitation, which is approximately considered as that of the top Ge_{0.76}Mn_{0.24} layer, was roughly 0.1 T, witch corresponds to the peak of the hysteretic behavior around 0.1 T. This indicates that the hysteretic behavior originates from, or is related to the magnetization of the top and bottom layers of the heterostructure mesa (S0373).

5. Summary

In the single $\text{Ge}_{1-x}\text{Mn}_x$ films and heterostructures, we observed large positive magnetoresistance with MR ratio over 2000% at 0.8 T and hysteretic behavior. Although the mechanism is not clear, these phenomena are caused by the nanocolumnar structure and may be used for advanced nanoscale spin devices based on group-IV semiconductors, such as magnetic sensors and magnetic recording devices.

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Fig. 1 (a) Schematic sample structure of the single $Ge_{1-x}Mn_x$ films and (b) a cross-sectional TEM image of a $Ge_{0.94}Mn_{0.06}$ film, where amorphous $Ge_{0.91}Mn_{0.09}$ nanocolumns are precipitated in the $Ge_{0.97}Mn_{0.03}$ matrix.



Fig. 2 Sample structures of the tri-layer heterostructures; S0367 (a) and S0373 (b).



Fig. 3 (left) I-V characteristics of the $Ge_{0.89}Mn_{0.11}$ and $Ge_{0.72}Mn_{0.28}$ single films, and heterostructure mesa (S0367) at 15 K.

Fig. 4 (right) Magnetic field dependence of the MR ratio of the $Ge_{0.89}Mn_{0.11}$ mesa measured at 15 K at -1 and -5 mV, and the $Ge_{0.72}Mn_{0.28}$ mesa at -5 mV.



Fig. 5 (left) Magnetic field dependence of the MR ratio of the heterostructure mesa (S0367) measured at 15 K at -20, -30 and -40 mV.

Fig. 6 (right) Magnetic field dependence of resistance of the heterostructure mesa (S0373) measured at 15 K at biases of -5, -10, -15, -20, and -40 mV.