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Formation and Characterization of Self-Organized Ferromagnetic Nanostructures in Epitaxially Grown Mn-Doped Ge Thin Films

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1. Introduction

Over recent years, Si- and Ge-based ferromagnetic semiconductors (FSs) have attracted considerable attention not only in the research field of semiconductor spintronics but also integrated electronics, owing to their suitability and compatibility with Si CMOS technology. In particular, Ge-based FSs would be a key material for future spintronics, since Ge plays an important role for advanced CMOS devices [1].

After the first report on ferromagnetism in Mn-doped Ge thin film [2], many investigations have been done for Ge-based FSs, including controversy over the origin of ferromagnetism in Mn-doped Ge films. Recently, we reported that highly-Mn-contained ferromagnetic *amorphous* $Ge_{1-x}Mn_x$ (a-$Ge_{1-x}Mn_x$) nanocolumns with high Mn content (x~20%) were precipitated in the pure Ge matrix when Mn-doped Ge thin films were epitaxially grown on Ge(001) by low-temperature molecular beam epitaxy (LT-MBE). We also found that precipitated a-$Ge_{1-x}Mn_x$ (that behaves as an amorphous ferromagnetic semiconductor [3]) is the origin of ferromagnetism in the Mn-doped Ge films [4]. In this paper, we investigate formation and characterization of self-organized a-$Ge_{1-x}Mn_x$ nanocolumns in epitaxially grown Mn-doped Ge thin films.

2. MBE Growth and Structural Characterization

We prepared $Ge_{1-x}Mn_x$ films grown on Ge (001) substrates by LT-MBE technique with a growth rate R_G = 30 nm/h at various substrate temperatures T_S (= 35 - 300°C) and with various growth rate R_G (= 30 - 150 nm/h) at a substrate temperature T_S = 100°C. Note that our previously investigated $Ge_{0.94}Mn_{0.06}$ film (R_G = 30 nm/h and T_S = 100°C) contained the precipitation of a-$Ge_{0.80}Mn_{0.20}$ nanocolumns in the pure Ge matrix [4]. Fig. 1 shows magnetic circular dichroism (MCD) spectra of $Ge_{0.94}Mn_{0.06}$ films with R_G = 30 nm/h at several T_S, measured with reflection configuration at 10 K. For T_S = 100°C, two MCD peaks at E_1 critical point of Ge and 2 eV originating from nanocluster were observed, indicating phase separation occurred in the film. By lowering T_S from 100°C, these two peaks were merged into one peak at 2.3 eV as shown in the figure. However, both ferromagnetic and paramagnetic components were clearly observed in the magnetic-field dependence of MCD intensity even for the films grown at T_S = 75°C and 35°C (Fig. 2). From TEM observations, self-organized nanocolumnar ordering was found to be formed in the T_S range between 35°C and 100°C. On the other hand, when T_S increased to 300°C, ferromagnetic Mn_5Ge_3 clusters were segregated in the film, and no nanocolumnar structure was formed in the film.

Fig. 3 shows a TEM image of $Ge_{0.94}Mn_{0.06}$ film with R_G = 150 nm/h at T_S = 100°C (where R_G was 5 times larger than that for the samples shown in Fig. 1). The film contained the precipitation of a-$Ge_{0.91}Mn_{0.09}$ nanocolumns in the $Ge_{0.97}Mn_{0.03}$ matrix. The diameter of the nanocolumns was ~5 nm, which is almost the same as the above described cases for R_G = 30 nm/h and T_S = 35 - 100°C. It was found that the Mn distribution was changed by increasing R_G; the Mn concentration x_{nc} of the nanocolumns decreased with increasing R_G, but that of the matrix region increased with R_G. From MCD measurements, we also observed that the contribution of the paramagnetic component described above was weaken with increasing R_G and disappeared at R_G = 150 nm/h.

Fig. 4 shows Curie temperature T_C and Mn concentration x_{nc} of nanocolumns in $Ge_{0.94}Mn_{0.06}$ films as a function of R_G. The x_{nc} of the nanocolumns decreased with increasing R_G, and also T_C decreased with increasing R_G. Fig. 5 shows coercive force H_C of the $Ge_{0.94}Mn_{0.06}$ films with R_G = 30 - 150 nm/h. When R_G > 90 nm/h, H_C was suddenly dropped and kept almost a constant value, as shown in Fig. 5.

3. Summary

The above described results show that the Mn concentration x_{nc} of the self-organized nanocolumns in the epitaxially grown Mn-doped Ge films can be manipulated, i.e., by lowering T_S or increasing R_G, x_{nc} was decreased and Mn atoms were distributed into the Ge matrix, and eventually magnetic property was changed, with keeping the size of nanocolumns and their spacing. The nanocolumnar ordering is explained by spinodal decomposition in ferromagnetic semiconductor [5]. The columnar ferromagnetic clusters presented here could be used as a magnetization-manipulable nano-magnet in Ge, which may lead to advanced nanoscale spin devices based on group-IV semiconductors.

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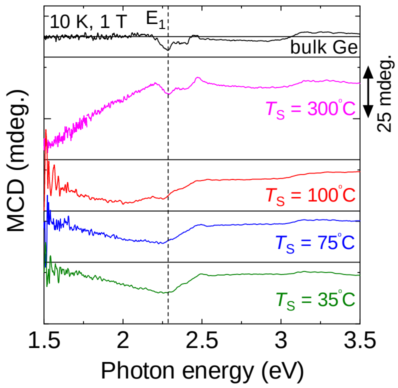

Fig. 1 MCD spectra of $Ge_{0.94}Mn_{0.06}$ films with R_G = 30 nm/h at T_S = 35, 75, 100, 300°C measured at 10 K under applied field of 1 T.

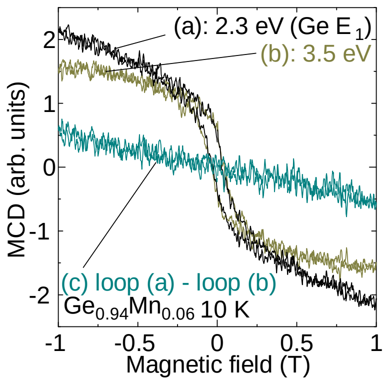

Fig. 2 MCD hysteresis of $Ge_{0.94}Mn_{0.06}$ films with R_G = 30 nm/h at T_S = 35°C measured at 2.3 eV (Ge E_1) and 3.5 eV. At the energy of E_1, paramagnetic component (curve (c)) was superimposed.

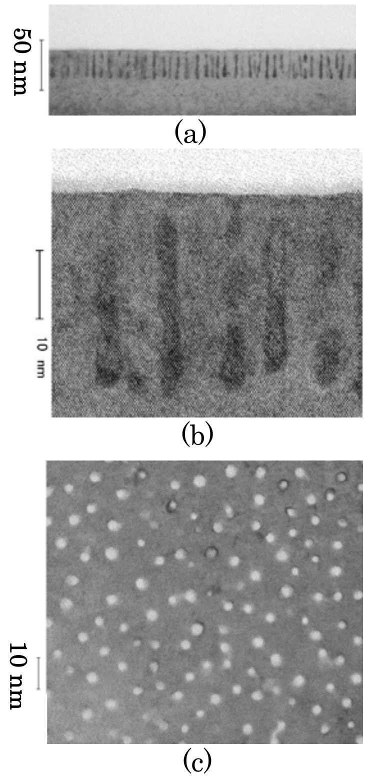

Fig. 3 TEM image of $Ge_{0.94}Mn_{0.06}$ film with R_G = 30 nm/h and T_S = 100°C. Mn was segregated into amorphous clusters. (a, b): cross-sectional view (c): plan-view

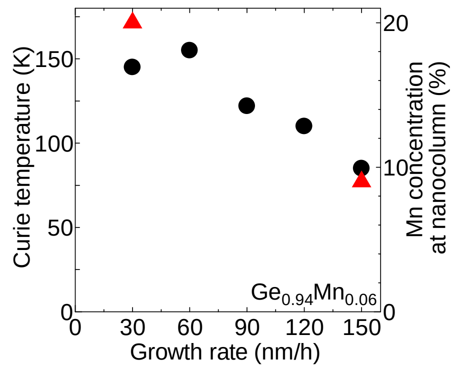

Fig. 4 T_C (circle plots) and Mn concentration at a-$Ge_{1-y}Mn_y$ nanocolumn (triangle plots) of $Ge_{0.94}Mn_{0.06}$ films grown at T_S = 100°C as a function of growth rate R_G.

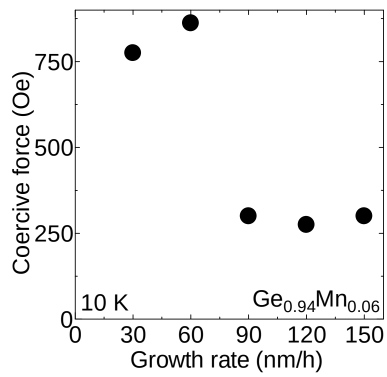

Fig. 5 Coercive force (Hc) of $Ge_{0.94}Mn_{0.06}$ films grown at T_S = 100°C as a function of growth rate R_G