Effect of carrier for magnetic and magnetotransport properties of Si:Ce films

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Epitaxial Si:Ce films with smooth surfaces were prepared by low temperature molecular beam epitaxy. Although as-deposited films showed positive magnetization due to the existence of Ce having been substituted in Si lattice, the conduction was *n* type. The conduction changed to *p* type as a result of hydrogen termination, indicating that the film had contained dangling bonds. The magnetization behavior of the as-deposited *n*-type sample is completely identical to that of the *p*-type sample. The temperature dependence of resistivity ($\rho - T$) for each sample with *n*-type or *p*-type conduction has a cusp at 150 K, which is related to the spin−glass transition. But the cusp observed in the $\rho - T$ curve of the sample with *n*-type conduction is broader than that of the sample with *p*-type conduction. It seems to have originated from the spin dynamics in Si:Ce with different carrier types. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1556116]

I. INTRODUCTION

Magnetic semiconductors have attracted much attention in recent years because of their application to spintronic devices. Mn doped III–V, e.g., (Ga,Mn)As, and II–VI, e.g. (Cd,Mn)Te etc., compound semiconductors have especially been extensively investigated.[1,2] On the other hand, we are also interested in Si based dilute magnetic semiconductors (DMS),[3] which should become promising materials for microelectronics applications. In Ce doped Si films, although various phenomena such as superparamagnetism, spin glass behavior, and giant magnetoresistance (MR) have been observed,[4,5] these films have a rough surface with root-mean square (rms) of 50 nm and Ce may inhomogeneously distribute in the film, which will disturb transport measurements. To eliminate the compositional inhomogeneity and surface roughness, the films were prepared by low temperature molecular beam epitaxy (LT-MBE). Si:Ce epitaxial films were successfully grown on a Si (100) substrate even at growth temperature of 300 °C. The rms roughness measured by atomic force microscopy (AFM) was approximately 0.5 nm. Although Ce should work as an acceptor in a Si matrix, the conduction was *n* type, probably due to the existence of dangling bonds in the Si:Ce films. In spintronic devices, it is very important to investigate if the physical properties can be controlled by the carrier, which in turn can be controlled by the external electric field. Therefore, this article focuses on the effect of carrier type for magnetic and magnetotransport properties of Si:Ce films having *n*-type or *p*-type conduction.

II. EXPERIMENT

Si:Ce epitaxial films were deposited on a (001) Si substrate by a solid source MBE system. Simultaneous evaporation of Si and Ce was carried out using high-temperature Knudsen cells. The evaporation temperature of Si was fixed at 1350 °C. The cell temperature of the Ce source was varied from 950 to 1150 °C to change the Ce concentration of the film. The deposition rates of Si and Ce were individually measured by a thickness monitor. To introduce a large amount of magnetic elements into Si, nonequilibrium growth methods (LT-MBE) were performed. The growth temperature (T_s) and the film thickness were fixed at 300 °C and 160 nm, respectively. The composition of the films was evaluated by Rutherford backscattering (RBS). The surface morphology was evaluated by AFM. Structural analysis of the films was performed with *in situ* reflection high energy electron diffraction (RHEED), conventional x-ray diffraction (XRD) (at 30 kV and 40 mA), and rotating target type x-ray diffraction (RT-XRD), (at 50 kV and 200 mA). To terminate the dangling bond in the LT-MBE film, some samples were annealed at 300 °C in forming gas ($5\%\ H_2 + 95\%\ N_2$). A superconducting quantum interference device (SQUID) was used to evaluate the magnetic properties. The resistivity and the Hall effect were measured using the van der Pauw method in the temperature range of 4–270 K.

III. RESULTS AND DISCUSSION

Throughout this work, all the Si:Ce films were prepared at $T_s = 300\,°C$. The *in situ* RHEED observation was performed so as to maintain the 2×1 surface reconstruction structure. The Ce concentration was changed from 0.01 to 2.5 at. %. The rms roughness measured by AFM is approximately 0.5 nm regardless of the Ce concentration. In films with various Ce concentrations, silicide precipitation was not observed even by RT-XRD. Although all the films have an expanded lattice due to the substitution of Ce ions for Si atoms, the film with Ce content of 0.2 has a maximum lattice constant. The magnetization of the samples also increases with an increase in the Ce content increase to 0.2. Therefore, this article focuses on films with Ce concentrations of 0.2 at. %, which is the largest magnetization.

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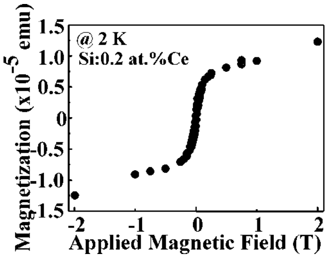

FIG. 1. Magnetization curve at 2 K of n-type Si:0.2at%Ce sample prepared at 300°C.

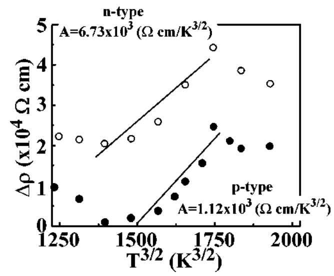

FIG. 3. $T^{3/2}$ dependence of $\Delta\rho$ for *n* and *p* type samples.

Figure 1 shows the magnetization curves ($M-H$) of the as-deposited Si:0.2 at.% Ce sample measured at 2 K. The magnetic field was applied normal to the film surface. The magnetic susceptibility of the sample at the applied magnetic field below 0.2 T is larger than that of a Ce^{3+} free ion calculated using the Brillouin function. Although it indicates that the sample has a superparamagnetic nature, an experimental saturated magnetization at 1.0 T was 0.15 emu/g which is about 13.0% of the theoretical saturated magnetization ($M_s = g_J \mu_B$).

Although we reported that Ce substitution is expected to make an acceptor level,[4] the film prepared by LT-MBE exhibits *n*-type conduction with carrier concentration of around $10^{15}\,\mathrm{cm}^3$. In the case of amorphous Si:Ce films that were previously fabricated,[4] although the conduction was *n* type, positive magnetization was not recognized. Therefore, the positive magnetization of this LT-MBE Si:0.2 at.% Ce films indicates that Ce was incorporated into the Si site. In this case, however, the film should be *p* type. The *n*-type conduction in the film originates from the generation of electrons due to the existence of dangling bonds in the low temperature grown epitaxial film. To justify this supposition, the sample was annealed at 300 °C in the forming gas. The conduction changed to *p* type with carrier concentration (N_c) of $1.6\times10^{16}\,\mathrm{cm}^{-3}$. The $M-H$ behavior of *p*-type hydrogen terminated film is almost the same as that of *n*-type Si:Ce at all temperature ranges. This suggests that the static magnetic structure is not influenced by the carrier type in this range of carrier concentration.

Figure 2 shows the temperature dependence of the resistivity for *n*-type (open circle) [N_c: $2.2\times10^{15}\,\mathrm{cm}^{-3}$, mobility ($\mu$): 848 cm^2/V s at room temperature (RT)] and *p* type (closed circle) (N_c: $1.6\times10^{16}\,\mathrm{cm}^{-3}$, μ: 210 cm^2/V s at RT) Si:Ce films. Either *p*-type or *n*-type Si:Ce film exhibits semiconductor-like $\rho-T$ behavior, having a cusp (T_g) at around 150 K, which is caused by the spin–glass transition.[6]

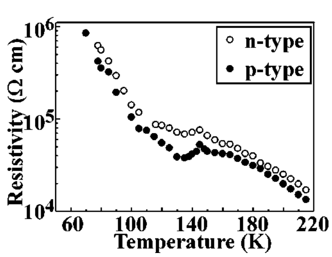

FIG. 2. Temperature dependence of resistivity for *n*-type (open circle) and *p*-type (closed circle) Si:Ce films.

But the cusp observed in the $\rho-T$ curve of the sample with *n*-type conduction is broader than that of the sample with *p*-type conduction. Since these cusps in $\rho-T$ curves originate from spin fluctuations just above the spin-freezing temperature,[7] the difference in $\rho-T$ behavior is considered to be caused by differences in the interaction between the carrier and magnetic spin. Since the $M-H$ curves for both *p*-type and *n*-type Si:Ce films were almost identical, the dynamical carrier–spin interaction on the spin–glass transition is considered to play an important role for $\rho-T$ behavior of samples with different carrier types.

For more appropriate treatment of the resistivity of the spin glass, the spin-diffusion theory is considered.[8,9] For purely diffusive modes, the resistivity below the cusp becomes $\rho(T,c)=c\rho(0)+A(c)T^{3/2}$. Here the residual $c\rho(0)$ is due to resonant scattering from the spin-split virtual bound state and $A(c)=\rho(\ )D^2(JS/\Gamma)^2(a^3/2\pi)(\pi k_B/2\Lambda a^{-2})^{3/2}$. $\rho(\infty)$ is the unitarity scattering limit $[=(1/2)c(m/e^2)x\Gamma(n/\Omega)^{-1}]$, with Γ the conduction bandwidth and n/Ω the electron concentration. Other parameters in the expression for $A(c)$ include D, the virtual bound state's $\pm$ displacement from the Fermi level to the width parameter; J, k_B, a, and Λ correspond to the spin (S)–spin coupling constant, Boltzmann constant, the lattice spacing, and diffusion constant, respectively. In addition to this theory, we need to consider the semiconducting nature of the sample. Therefore, the theoretical resistivity of Si with the same carrier concentration was subtracted from the experimental one. Figure 3 shows the $T^{3/2}$ dependence of $\Delta\rho$ ($\rho_{\mathrm{ex}}-\rho_{\mathrm{semi}}$) for both *n* (open circle) and *p* (closed circle) type samples. The slope of $T^{3/2}$ for the *p*-type sample is calculated to be $1.12\times10^3\,\Omega\,\mathrm{cm/K}^{3/2}$, which is larger than that from the *n*-type sample ($6.73\times10^2\,\Omega\,\mathrm{cm/K}^{3/2}$). The $T^{3/2}$ dependence in spin diffusion theory is very sensitive to the availability of long-wavelength diffusion modes. Therefore, the spin diffusion mode of the *p*-type sample corresponds to a longer wavelength than that of the *n*-type sample.

Figure 4 shows MR behavior at 130 K, which is just below T_g. The magnetic field was applied normal to the film surface. The MR ratio of the *n*-type sample is positive in a magnetic field of 1 T. In the *p*-type sample, on the other hand, negative MR is observed at a magnetic field of 0.2 T, which corresponds to the magnetic susceptibility in the $M-H$ curve. At high magnetic field, over 0.2 T, however, positive MR is observed. The negative MR of the *p*-type sample at low magnetic field is considered to be caused by the stabilization of long-wavelength carrier diffusion.

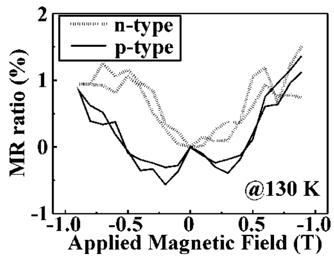

FIG. 4. Magneto-resistance measured at 130 K of *n*-type (dashed line) and *p*-type (solid line) Si:Ce films.

Although a distinct difference in $M-H$ behavior was not observed between samples with different carrier types in the carrier concentration range of $1\times10^{-16}\,\mathrm{cm}^{-3}$, the difference is recognized in the $\rho-T$ and MR behavior. This seems to originate from the spin dynamics in Si with different carrier types. Based on these results, we might be able to control the static magnetic structure by changing the large carrier concentration.

IV. CONCLUSIONS

The effects of carrier type on the magnetic and magnetotransport properties of Si:Ce films with surface roughness less than 0.5 nm were studied. Although, the $\rho-T$ curves of both *n*-type and *p*-type samples had a cusp at 150 K, the cusp of the *n*-type sample was broader compared with that of *p* type. An analysis of the resistivity below the cusp temperature suggested that the spin diffusion mode of *p*-type sample corresponded to longer wavelength than that of the *n*-type sample. The MR behavior also supports the notion that there is positive magnetic interaction in Si:Ce film with *p*-type conduction.

ACKNOWLEDGMENTS

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