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Anomalous Hall- and magneto- resistances on Cr-doped Ge in high magnetic fields observed up to room temperature

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Abstract. Ge$_{1-x}$Cr$_x$ films grown by MBE show quite large magneto-transport effects in the temperature range up to the room temperature. Especially, Hall resistance in DC- fields up to 10 Tesla and magneto-resistances in pulsed fields up to 60 Tesla show significant nonlinear field dependence. The magnetic field effects on the transport properties are usually quite small in conducting materials and phonon plays main role above liq. N$_2$ temperature. ESR measurements suggest that antiferromagnetism up to $T_N$ around 70 K exists and the critical spin fluctuation might affects to the nonlinear field dependence in magneto-transport phenomena in high temperatures.

1. Introduction

Various phenomena of spin electronics have been expected as the interesting effects having high potentiality to application. The phenomenon also gathers much scientific interest, because the basic phenomena between conduction electron and localized spin system in semiconductors are also attractive subjects. Diluted Magnetic Semiconductors (DMS) are the most possible materials for these problems. For example, ferromagnets of (Ga,Mn)N with $T_C$ exceeding room temperature have been already reported [1]. Another famous example of the effect on spin electronics is Giant Magneto-Resistance (GMR). Though ferromagnetic metals, such as Fe, Ni and Co do not have so large magnetoresistance, GMR device has large MR-ratio at room temperature [2].

Here, we investigate new DMS materials showing large magneto-transport effect in high temperature. Germanium is one of the key elements to add new additional functions to Si and we focused our attention to Ge based DMS materials. Existence of ferromagnetism is also expected in Ge based DMS, but a theory predicts low critical temperature $T_C$ for Mn doped Ge [3]. However, the detailed characterizations for Cr doped Ge (Ge:Cr) have not been carried out, so far. It is also predicted that Cr has shallow acceptor level (~ 1 meV) with energy gap of Ge and it gives us possibility of a nice p-type semiconductor [4]. In this work, we report experiments and discussions on the anomalously large magnetoresistance and non-linear Hall effect of Ge:Cr in high magnetic fields against strong lattice vibration in high temperature region.
2. Experimental procedures and results

2.1. Sample Preparation
Samples were prepared by use of Molecular Beam Epitaxy system. GaAs (001) and Ge (001) wafers were used as the substrates. By using these two substrates, the magnetic coupling through the substrate could be checked. In the RHEED observation, we confirmed the streak like interference patterns kept the Ge crystal structure in the growth process. The X-ray diffraction patterns also show single phase crystallization of Ge:Cr. Another powerful method to check the secondary phase is cross section observation by High Resolution TEM. We could confirm no clustering of Cr or Ge-Cr alloys by the method of HRTEM. Especially, ferromagnetic cluster of CrGe$_2$ (Cr$_{11}$Ge$_{19}$) has not been observed.

2.2. Magnetic measurements
For the Ge$_y$Cr$_x$-type crystal family, AF-magnetism of Ge:Cr ($T_N = 62$ K) and ferromagnetism of CrGe$_{2}$ (Cr$_{11}$Ge$_{19}$; $T_C \approx 100$ K) have been reported. The magnetization process and the temperature dependence were observed on Ge$_{1-x}$Cr$_x$ films: $x = 0.022 \sim 0.079$ by using SQUID magnetometer. The field dependence could be observed up to 7 T by use of SQUID. The concentration of Cr was determined by use of EPMA. Existence of hysteresis curve in magnetization process is the most standard criterion of ferromagnetism, because the hysteresis curve includes existence of remnant moment at zero-field which directly means existence of domain wall and spontaneous spin polarization. Figure 1 shows the magnetization curves of samples with various concentrations.

![Figure 1](image.png)

**Figure 1.** (A) Magnetization curves at 1.8 K for various concentration of Cr in Ge$_{1-x}$Cr$_x$. The concentrations of $x$ are indicated in the figure. (B) Magnetization curves of Ge$_{0.921}$Cr$_{0.079}$ in various temperatures.

Hysteresis have not been observed in the traces of the magnetization process on the samples in the concentration range of $x = 0.022 \sim 0.079$. It is noticeable that when the concentration of Cr becomes higher, the magnetizations are decreasing. The results point that the saturation moments do not increase with increase of the concentration. Moreover, the estimated saturation moments are extremely small for the concentration of Cr ions. The observed magnetization curves can be reproduced well by use of function form given by the sum of two Langevin functions. These Langevin functions correspond to two elemental magnets of $2\mu_B$ and $8\mu_B$, respectively. However, large reduction of the net magnetization for the concentration of Cr$^{+3}$ ions is necessary to fit to the experimental curve. The large reduction of magnetization in composite Brillouin function form suggests possibility of coexistence of both antiferro- (AF-) and superpara- magnetisms in the Ge:Cr film. In the case of DMS ferromagnetism, coexistence of ferro- and superpara- magnetisms has been already reported [5]. Similar discussion is also applicable to the coexistence of both AF- and superpara- magnetisms, because the elemental process of spin polarization is similar to DMS ferromagnetism. Assuming such coexistence and the AF-exchange interaction is strong enough, the superparamagnetism mainly contributes to the magnetization in low temperature. The possibility of the coexistence model should be checked by various experimental methods.
Focusing to highly concentrated Cr samples, temperature dependence of Ge\textsubscript{0.925}Cr\textsubscript{0.075} on Ge-coated Ge-wafer and that of Ge\textsubscript{0.921}Cr\textsubscript{0.079} samples on Ge-coated GaAs substrate show similar temperature dependence. Figure 1(B) shows the temperature dependence of magnetization process in Ge\textsubscript{0.925}Cr\textsubscript{0.075} on GaAs-wafer sample. As seen in figure 1(B), ferromagnetic hysteresis have not been observed and all other Ge:Cr samples on both substrates also do not show hysteresis. The clear non-linear magnetization process is observed at 1.8 K, but the non-linear dependence rapidly close to linear dependence in Figure 1(B). The temperature dependence is consistent with the characteristic of super paramagnetism. If there exists AF-magnetism in Ge:Cr and the AF-exchange interaction is strong enough, observation of slight linear field dependence in magnetization process can be expected. However, such linear dependence cannot be discriminated from other stray magnetizations. For example, diamagnetism and impurity paramagnetism in substrate etc. also have such a linear field dependence. Thus, ferromagnetism of Ge:Cr type samples has not been observed, so far.

2.3. ESR measurements

Figure 2(A) shows differential curves of ESR spectra in various temperatures. These traces seem to be largely different from usual signals of EPR absorption. If we can assume AF resonance (AFMR) having low anisotropy, the signals can be understood as shown in the schematic curves in figure 2(B). Assuming this model, we can interpret that there exist two branches of the ESR signals and the large temperature spectral shift for the stronger spectral branch. The large spectral shift and the temperature shift are explained by the temperature dependence of AF-exchange fields. The spectral shift is explained by change in exchange fields. Existence of two-branches is also characteristics of AFMR. Thus, these characteristics are just consistent with the AF-magnetism with low anisotropy. The low anisotropy effect is also confirmed by the angle dependence of ESR signals. As seen in figure 2(A), the weak ESR signal at 0.3 T is assigned as superparamagnetism, but the ESR signals cannot be observed above 10 K. From these results in ESR, the existence of AF-magnetism and $T_N$ of about 70 K are conjectured.

![Figure 2](image)

2.4. High field Hall- and Magneto- resistances

Figure 3(A) shows field dependence of the magnetoresistance at various temperatures. It is quite remarkable that the clear non-linear field dependence is observed in 200 K. This is also observed even in room temperature against the strong thermal fluctuation, whereas the magnetoresistance usually become less with increasing temperature. The effect is possible to apply to various applications at room temperature. Such clear non-linear field dependence was also observed in Hall resistance. The measurements were performed by use of magnetic fields up to 60 T, as shown in figure 3(B). The non-linear Hall resistance is called as anomalous Hall effect. The non-linear Hall- and non-linear
magnetoresistance effects have some concentration dependence. The most clear magneto-transport effect was observed on the sample with the Cr-concentration region of 7.5 ~ 8%.

![Figure 3](image)

**Figure 3.** (A) Field dependence in resistance on Ga$_{0.925}$Cr$_{0.075}$ at various temperatures. (B) Hall-resistance observed by pulsed magnetic field up to 60 T.

Anomalous Hall- and non-linear magneto-resistances are originated from some magnetic order effects, because the magneto-transport effects can be observed only in the collective spin phenomena against the strong thermal fluctuation in high temperature. The nonlinear curve of anomalous Hall resistance is getting close to the asymptotic straight line in high magnetic fields, where the straight line go through the origin. The anomalous Hall resistance subtracting from the linear part is shown in the inset of figure 3(B) and the asymptotic line is consistent with the normal Hall-resistance, because the spin fluctuation is suppressed in high magnetic fields. The carrier density can be estimated as $6 \times 10^{24} \text{ m}^{-3}$ and $1 \times 10^{25} \text{ m}^{-3}$ at 1.8 K and 200 K, respectively. Such non-liner dependence can be observed up to 40 T, as shown in the inset in figure 3(B).

3. Discussion
The origin of the anomalous Hall- and magneto- resistance in Ge:Cr is not understood well. However, the steep change in temperature dependence of resistance in the inset of figure 3(A) shows large decrease above 50 K. This change is completely different from the thermal activation type temperature dependence in semiconductors. One possible model is an effect on AF-magnetic spin fluctuation around $T_N$ of about 70 K. Such AF-spin fluctuation generally affects to electron scattering through the spin-orbit interaction and the region of the critical fluctuation spread out wide temperature range around $T_N$. In addition to the temperature dependence of the resistance, the large shifts of ESR spectra also support this model. Other methods, such as NMR or neutron scattering experiment are expected to confirm this model.

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