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Magnetotransport Properties of a Room-Temperature Ferromagnet (Ga, Mn)N

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Heavily Mn-doped GaN thin films were fabricated by using solid-state diffusion. The magnetic properties of the samples were determined by Hall effect measurements and by using a superconducting quantum interference device. The results show ferromagnetic behavior even above room temperature, the Curie temperature being 330 K. The samples were of n-type and the estimated electron concentration was $\approx 1 \times 10^{20}$ cm^{-3}. Due to the spin-disorder scattering, a negative magnetoresistance was observed and it was largest at Curie temperature. The measured sheet resistance versus temperature could also be explained by the spin disorder scattering model for ferromagnetic metals. From the measurement results, the value of the exchange interaction parameter between the electron spins and the magnetic moments of the Mn atoms was estimated to be about 1 eV. The material properties were studied also by using secondary ion mass spectroscopy and X-ray diffraction, which showed some segregation of $Ga_x Mn_y$.

Index Terms—**Hall effect, magnetic semiconductors, manganese compounds.**

I. Introduction

Magnetic semiconductors have been studied since the 1960s. However, a significant breakthrough in the field occurred as late as in 1989, when Mn-doped InAs fabricated by using low-temperature molecular beam epitaxy (MBE) turned out to be ferromagnetic [1]. When the same method succeeded in the case of GaAs, it was realized that it is possible to integrate spintronic devices with conventional semiconductor devices. Unfortunately, the Curie temperatures of Mn-doped InAs or GaAs are well below room temperature. One of the most promising solutions to this problem is Mn-doped GaN, where the Curie temperature may even be as high as 940 K [2], or at least well above room temperature [3]–[7], which is the critical temperature for applications. In addition, the fabrication technique for GaN devices is well developed, and therefore Mn-doped GaN has become one of the most interesting new materials for semiconductor spintronics.

The ferromagnetism in Mn-doped GaAs can be explained by using a mean field model based on the exchange interactions mediated by holes [8]. However, in the case of Mn-doped GaN it is difficult to explain the mechanisms behind the ferromagnetism with the same model since, e.g., the $Mn^{3+/2+}$ acceptor level is very deep ($\sim E_v + 1.8$ eV) [9]. The ferromagnetic (Ga, Mn)N thin films have been fabricated by using various methods such as solid-state diffusion (SSD) [4], ion implantation [11], and plasma enhanced MBE [2], [3], [5]–[7], [10]. In the present work we have chosen the SSD technique, and studied especially the transport properties of the (Ga, Mn)N samples. Furthermore, we have aimed at a quantitative analysis of the measured transport properties based on a spin disorder scattering model.

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II. Sample Fabrication

Our (Ga, Mn)N samples were fabricated by using a solid-state diffusion of Mn into a 2-μm-thick undoped GaN layer, which was grown on a sapphire (0001) substrate by using MOCVD at ATMI Ltd. Then, a 50-nm-thick Mn layer was deposited on GaN at room temperature by using our VG100H MBE system. The Mn diffusion was activated by annealing the samples under UHV conditions for 14 h at 600 °C. Then, the rest of the Mn layer was removed from the GaN surface by dipping the samples in HCl. The ohmic contacts to the samples were made by using copper (98%)/beryllium (2%) wires.

III. Results and Discussion

According to the secondary ion mass spectroscopy (SIMS) measurements, the thickness of the heavily Mn-doped layer in our (Ga, Mn)N thin films is approximately 200–300 nm. The mole fraction of Mn could not be estimated from the SIMS results, since no Mn reference sample was available. However, with the aid of the results from the Hall and SIMS measurements, the electron concentration can be estimated as $\approx 1 \cdot 10^{20}$ cm^{-3}, but the origin of the defects which donate the charge carriers cannot be determined from our measurement.

Previous studies have pointed out that it is important to study the crystallographic structure of the (Ga, Mn)N samples, since there are many compounds of Ga, N, and Mn that also are ferromagnetic at high temperatures [4], [10]. An X-ray diffractometer with incident beam mirror and monochromator was used to study possible phase segregation in our thin films. The reference sample for the X-ray diffraction (XRD) measurements was similar to the original sample excluding the Mn doping. The peaks at 34.6° and 72.9° in Fig. 1 are due to the 2-μm-thick GaN layer and the peak at 41.7° is due to the sapphire substrate. In addition, small additional peaks at $\theta = 31.9°$ and $\theta = 66.6°$ are observed in the Mn-doped sample. Our XRD

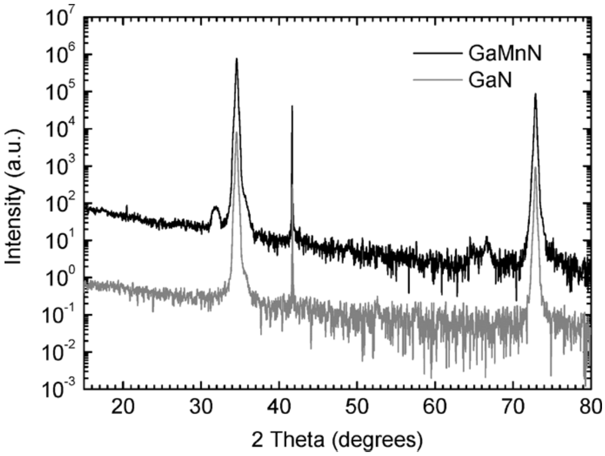

Fig. 1. XRD measurement results: upper graph is the (Ga, Mn)N thin film sample and lower graph is the GaN without Mn. Lower scan is offset for clarity.

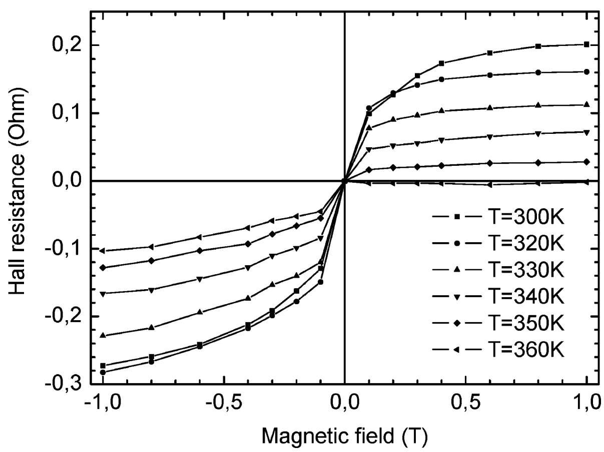

Fig. 2. Hall resistance versus magnetic field at various temperatures in (Ga, Mn)N.

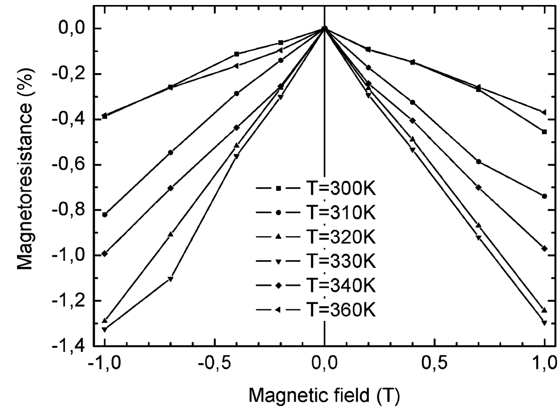

Fig. 3. Magnetoresistance versus magnetic field at various temperatures in (Ga, Mn)N.

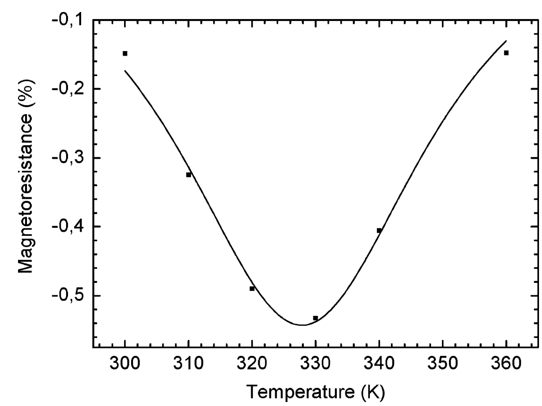

Fig. 4. Magnetoresistance as a function of temperature in (Ga, Mn)N in the magnetic field 0.4 T.

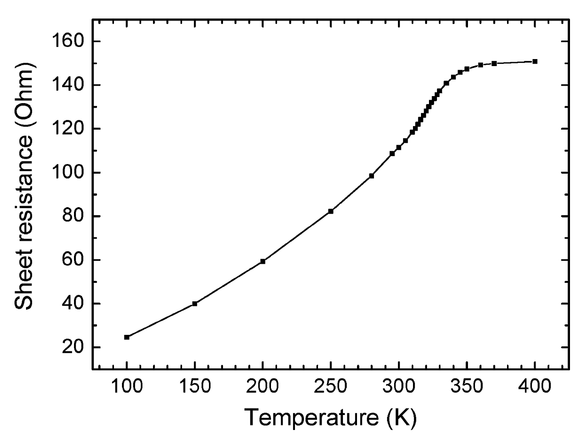

Fig. 5. Sheet resistance as a function of temperature in (Ga, Mn)N.

results (Fig. 1) and SIMS results indicate that some segregation of Ga_xMn_y in (Ga, Mn)N is present in our sample. Generally, the lattice constant of (Ga, Mn)N changes as a function of Mn concentration [12]. However, typically the samples with high Mn concentration, as ours, show secondary phase peaks [10], but no shift in GaN peak related to the variation of the lattice constant.

The Hall resistance in (Ga, Mn)N thin films was measured by using the Van der Pauw configuration for the contacts. In ferromagnetic conductors, the Hall resistance is given by

$$R_{\text{Hall}} = \frac{R_0 B}{d} + \frac{R_S M}{d} \tag{1}$$

where B is the applied magnetic field, R_0 is the ordinary Hall coefficient, M is the magnetization of the film, R_S is the anomalous Hall coefficient, and d is the sample thickness. The results of the Hall resistance measurements (Fig. 2.) indicate, that our n-type Mn-doped GaN thin films are ferromagnetic, since a strong anomalous Hall effect (AHE) described by the last term in (1) is observed. The estimated Curie temperature, by using (1), is 330 K.

Fig. 3 shows the measured magnetoresistance at various temperatures in the field range 0–1.0 T. The negative magnetoresistance is largest ($\approx$1.3%) at around $\mathrm{T} = 330$ K, as shown in Fig. 4.

Fig. 5 shows the measured temperature dependence of the sheet resistance R_{sheet} in (Ga, Mn)N. At high temperatures above 330 K the temperature dependence is weak, whereas at lower temperatures sheet resistance decreases rather strongly with decreasing temperature.

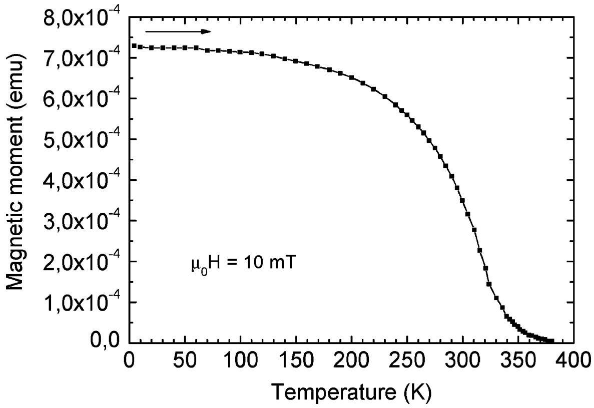

Fig. 6. Magnetization as a function of temperature in (Ga, Mn)N.

All the results shown in Figs. 2–4 can be explained by a model for spin disorder scattering in ferromagnetic metals [13]. The scattering is caused by the following exchange interaction between the carrier spin and the localized spins of the Mn atom

$$H_{\text{exch}} = -J_{\text{exch}} \sum_{\vec{R}} \delta(\vec{r} - \vec{R}) \vec{S}_{\vec{R}} \cdot \vec{s} \tag{2}$$

where J_{exch} is the exchange interaction parameter, $\vec{S}_{\vec{R}}$ the total spin of the Mn atom at site $\vec{R}$, and $\vec{s}$ is the spin of the carrier at site $\vec{r}$. The interaction (2) gives rise to the spin disorder scattering, which leads to the following expression for the resistivity:

$$\rho_s(T,B) = \rho_0[S(S+1) - \langle \vec{S}(T,B) \rangle^2] \tag{3}$$

with

$$\rho_0 = \frac{2\pi^2 k_F m*^2 \, J_{\text{exch}}^2 \Omega^2 C_{\text{Mn}}}{e^2 h^3 n} \tag{4}$$

where k_F is Fermi wave vector, Ω the volume of the unit cell, C_{Mn} concentration of Mn atoms, n the electron density, $S = 5/2$, and $\langle \vec{S} \rangle$ the thermal average of $\vec{S}$. The model (3) predicts that resistivity is constant at high temperatures $T > T_C$, where $\langle \vec{S} \rangle = 0$, and then ρ decreases with decreasing T, since below T_C $\langle \vec{S} \rangle$ starts to increase. This is just the behavior we found experimentally (Fig. 5). Also, in accordance with our experimental results shown in Fig. 4, the model (3) predicts that the magnetoresistance is largest at $T = T_C$. From the measured high temperature resistivity and (3) and (4), we can estimate the exchange interaction parameter $J_{\text{exch}} \approx 1$ eV for $C_{\text{Mn}} = 50\%$. To our knowledge, this is the first time this parameter has been estimated for electrons from the transport data in the new magnetic III-V semiconductors. The estimated value is smaller than the one for holes of (Ga, Mn)As, for which J_{exch} is 3.3 eV as estimated from (3) and (4), [13].

The observed AHE, the maximum of the magnetoresistance at $T = 330\ K$, and the kink in the observed R_{sheet} versus T behavior at the same temperature, all point to a Curie temperature of 330 K. This value was then confirmed by using direct magnetization measurements by superconducting quantum interference device, as shown in Fig. 6.

The tail of the magnetization curve is probably due to the nonuniformity and phase segregation in the (Ga, Mn)N thin films. Otherwise, the results in Fig. 6 show the typical Brillouin-like behavior of a ferromagnet.

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