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High T_C ferromagnetism in diluted magnetic semiconducting GaN:Mn films

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Abstract

Wurtzite GaN:Mn films with an extremely high Curie temperature of around 940 K and Mn concentration of only 3 to 5% are successfully grown on sapphire substrates by molecular beam epitaxy. Magnetization measurements are carried out using magnetic fields of up to 7 T parallel to the film surface. The magnetization process reveals the coexistence of ferromagnetic and paramagnetic contributions at low temperatures, with characteristic ferromagnetic magnetization at high temperatures. The observed transport characteristics demonstrate a close relation between magnetism and impurity conduction. The double exchange mechanism of the Mn-impurity band is presented as a possible models for the high-T_C ferromagnetism in GaN:Mn.

Keywords: Gallium Nitride, Diluted Magnetic Semiconductor, Ferromagnetism

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1. Introduction

Research on diluted magnetic semiconductors has attracted much interest since the first fabrication of Mn-doped InAs (InAs:Mn) [1]. That result has led to intensive investigation of III-V-based diluted magnetic semiconductors (DMS) [2-4]. Certain ferromagnetic semiconductors are expected to be applicable in spin polarizers [5] as a ferromagnetic material for spin injection into non-magnetic semiconductors. However, the highest Curie temperature T_C of present GaAs-based DMSs is 110 K. These functions are therefore not yet available at room temperature. Recent developments in growth techniques for wurtzitic gallium-nitrides has led to the fabrication of GaN-based optical and electrical devices [6, 7].

There have also been some theoretical predictions of the possibility of GaN-based DMS with Curie temperatures exceeding room temperature [8, 9]. Although a few experimental studies on the magnetic characteristics of GaN-based DMSs have been reported [10-14], a sample with a T_C far exceeding room temperature has yet to be presented. Notable, an observed anomalous "offset magnetization" has been observed at room temperature related to ferromagnetism, but intrinsic ferromagnetism was not confirmed [14]. The present authors have reported the ferromagnetic characteristics of GaN:Mn film based on observation of the magnetization process, which revealed a clear hysteresis loop attributable to the ferromagnetic domain structure [15]. Clear step-like magnetization around zero-field also strongly indicated ferromagnetism. A Curie temperature of much higher than 400 K was estimated based on the temperature-dependent saturation moment.

The main purpose of this study is to discuss the magnetic characteristics of DMS films. We also discuss the coexistence of ferromagnetic and paramagnetic parts, and the origin of the high Curie temperature of GaN:Mn, and present a tentative but plausible model

for the high T_C based on experimental results.

2. Experimental

2.1 Sample preparation

Wurtzite GaN:Mn films exhibiting ferromagnetic behavior and a Curie temperature above RT were successfully grown on sapphire(0001) substrates by molecular beam epitaxy (MBE) using an ammonia nitrogen source (NH_3-MBE) system (MBC-100, ULVAC) equipped with a reflection high-energy electron diffraction (RHEED) apparatus. Solid-source effusion cells were used for Ga and Mn sources. GaN:Mn films of 1300 to 6000 Å in thickness were grown at temperatures between 850 and 1020 K with various Ga/Mn flux ratios on wurtzite GaN buffer layers on the sapphire substrates [15]. Observed RHEED pattern revealed that single-crystal wurtzite GaN:Mn films were obtained at a higher growth temperatures and higher Ga/Mn flux ratios. Zinc-blende crystals were included in the films grown at a lower growth temperatures and lower Ga/Mn flux ratios. The relationship between growth conditions and the crystal structures of the grown films are presented in detail in Ref. [16]. After growth of the GaN:Mn film, a pure GaN layer of thickness 200 Å was grown as a cap layer to prevent oxidation of the film. The quality of the GaN:Mn films was examined by RHEED and X-Ray diffraction (XRD) before measuring the magnetic and electrical properties. Table I lists the growth conditions and the RHEED patterns observed during the growth of the GaN:Mn film and the GaN buffer layer. XRD measurements revealed that the film had a wurtzite structure without phase separation [16]. The wurtzite crystal structure of the GaN:Mn film was confirmed by coaxial impact collision ion scattering spectroscopy (CAICISS), which clearly denied any possibility of segregation on the surface of the GaN:Mn film within the resolution limit. The results of the detailed

CAICISS measurement are discussed in Ref. [17], and are consistent with the observed RHEED patterns. From the depth profile measured by secondary ion mass spectroscopy (SIMS) and spectra measured by X-ray absorption fine structure (XAFS) analysis, it was confirmed that the Mn concentration in the GaN:Mn film was uniform within the resolution limit of 30 nm, and Mn atoms were incorporated at the Ga sites [16, 18]. A 3000-Å GaN film on a GaN buffer layer was prepared for a comparative study of the magnetic properties by subtracting the stray magnetization of the GaN layer on the sapphire substrate from the observed magnetization data for GaN:Mn film. The Mn-concentration in the samples used in this work was about 3~5%, as estimated by secondary ion mass spectrometry (SIMS; IMS-5F, Cameca Co. Ltd.) at JAIST.

2.2 Magnetization measurement

Magnetization measurements were carried out using a superconducting quantum interference device (SQUID) magnetometer (MPMS-XL, Quantum Design Co. Ltd.) at temperatures from 1.8 to 750 K. Magnetic fields of up to 7 T were applied parallel to the film plane (with in-plane spin). Curie temperatures higher than 400 K were examined by heating the samples in an oven with precise temperature control in the SQUID magnetometer. The background contribution to the magnetization originated from the GaN buffer layer, the cap layer, and the diamagnetism of the sapphire substrates. These effects were eliminated from the original sample data using experimental results.

2.3 Transport measurement

Transport measurements have already been reported for the composite GaN-buffer layer/GaN:Mn film/GaN-cap layer [24]. In order to obtain more precise transport characteristics for GaN:Mn films, samples without a cap layer, a relatively thick

GaN:Mn film and high-resistance buffer layer were specially prepared. The room-temperature ferromagnetism of these samples was confirmed before transport measurement. Sn was used to make the ohmic contact between the sample and lead wire.

The magnetoresistance was measured by the four terminal method, and the net carrier concentration was estimated from the measured Hall resistance. The raw data revealed a symmetric field dependence for the zero-field, which is unusual for the Hall resistance. It is considered that the magnetoresistance interfered with the Hall resistance data due to unavoidable misalignment of electrodes. This stray magnetoresistance was eliminated by comparing two Hall resistance data taken using opposite magnetic fields while leaving the other parts of the circuit unchanged.

3. Results and discussion

3.1 Magnetization

Figure 1 shows the temperature dependence of the magnetization (*M*-*T* curve) at 0.1 and 7 T. The 0.1-T magnetic field, which induces a negligible change in paramagnetism, is just above the field corresponding to the top of the hysteresis loop measured at 1.8 K. Therefore, the *M*-*T* curve in Fig. 1 is considered to show the temperature dependence of the spontaneous polarization, M_{Sp}, in the ferromagnetic state. The *M*-*T* curve is observed at up to 750 K to estimate the value of T_{C} precisely.

The field dependence of the magnetization, that is, the magnetization process (*M*-*H* curve), was measured at up to 400 K. Typical data are shown in Fig. 2. The hysteresis loop is visible in the *M*-*H* curve at these temperatures. The value of the coercive field at 300 K shown in the inset of Fig. 2 is about 6 mT.

A steep increase in the *M*-*T* curve at 4.2 K appears to indicate the coexistence of

paramagnetic and ferromagnetic components. The features of coexistence are also seen in the *M-H* curve, which presents a paramagnetic-like magnetization process on the ferromagnetic magnetization. However, the steep increase in the *M-T* curve indicates more than simple coexistence: the saturation magnetization, M_{Top}, at the top of the hysteresis loop also has the steep increase, as indicated by the open squares in Fig. 1. The steep increase in saturation magnetization was confirmed directly by the observation of an anomalous increase in the remanent moment in the same low temperature range.

The magnetization and susceptibility data will yield much information, however, careful analysis is required before quantitative discussion because the ferromagnetism generated by doping Mn ions is not a simple coexistence of paramagnetic and ferromagnetic components. These phenomena prevent a clear decomposition of the paramagnetic and ferromagnetic components. A careful analysis of the detailed temperature dependence of the magnetization process will be given by Suga *et al.* in the near future [19]. The results of that study suggest that the ferromagnetic saturation moment varies with temperature or a change in the exchange interaction. Such coexistence characteristics have been commonly observed in experiments on other similar DMS materials [20]. We believe that these characteristics are an intrinsic problem of DMS materials, and have yet to be systematically discussed in any detail.

The magnetization curve for 7 T is close to that of ferromagnetic spontaneous magnetization for 0.1 T at room temperature. These features arise primarily from the temperature dependence of the paramagnetic component. Analysis of the inverse susceptibility plot is not appropriate in the case of high-T_C ferromagnetics like GaN:Mn because the high-temperature expansion of the plot is not applicable to the magnetic coexistence system with temperature-dependent saturation moment. From these results,

it can merely be stated that the GaN:Mn film exhibits ferromagnetism with a Curie temperature higher than 750 K and the coexistence of paramagnetism in some form. The Curie temperature of the present sample is quite high in comparison with the ferromagnetic DMS materials reported to date. The T_C was estimated to be 940 K using the usual theoretical curve given by the molecular field approximation [21].

3.2 Segregation

The high-T_C ferromagnetism of this sample may be attributed to segregation of ferromagnetic compounds. However, this possibility is clearly refuted by the following observations: 1) Small segregated ferromagnets should exhibit superparamagnetism characteristics. However, the observed ferromagnetic *M*-*H* curve in this study differs completely from that characteristic of superparamagnetism, and is in fact a typical ferromagnetic curve above room temperature. 2) Mn was confirmed to be uniformly distributed in the sample within the resolution limit of the SIMS apparatus and Rutherford back scattering (RBS; 1700H, Nisshin-High Voltage Co. Ltd.) measurements at JAIST. The resolution limits of these equipments are approximately 30 and 20 nm, respectively. The results of the CAICISS experiment also clearly show a uniform surface without segregation. 3) The strong fluorescence of GaN observed at 3.3 eV (in the ultraviolet region) is entirely non-existent in GaN:Mn. If segregation occurs as a result of the inclusion of 3% Mn in the sample, the optical effects of segregation should be apparent as a blocking fluorescence of the same wavelength. However, such fluorescence is not observed. Instead of the blocking band, a weak band is observed at different wavelengths around 2 eV (orange region). This result strongly suggests an intrinsic change in the electronic state as a result of Mn-doping because such a change in the electronic state cannot occur through the simple coexistence of GaN and

segregated materials. 4) The change in the carrier type and the large variation in carrier density from *n*-type GaN to *p*-type GaN:Mn support the intrinsic change in the electronic state as a result of Mn-doping. 5) The Curie temperature of the present sample is much higher than the highest T_C (748 K) in the Mn-Ga alloy system [22] and the T_C (720 K) of Mn_4N [23]. These are the possible materials formed form Ga, N and Mn. α-Mn metal and ionic MnO are also possible compounds, but are antiferromagnetic. These experimental results are strong evidence that the GaN:Mn films grown by MBE in this work are new intrinsic ferromagnets of DMS with high T_C.

3.3 Transport property

As GaN:Mn is a DMS materials, the origin of the ferromagnetism is considered to be closely related to the properties of the carrier. Carrier transport may be dominated by hopping conduction via the impurity band of Mn, as suggested by the temperature range of the trapping effect of the carriers, which is consistent with that of the anomalous increase in magnetization.

The measured magnetoresistance and Hall resistance are shown in Figs. 3(A) and (B), as measured in fields stronger than 7 T. The high magnetic field is applied in order to avoid domain and ferromagnetic demagnetization effects in the hysteretic field dependence region. As mentioned above, due to experimental difficulties, a component of magnetoresitance was included in the measurements of Hall resistance. This stray magnetoresistance has been eliminated in the main data set shown in Fig. 3.

Figure 4(A) shows the temperature dependences of the resistance and carrier density. The carrier density is estimated from the Hall resistance. As in Ref. [8], the majority carrier in this work was found to be positive, forming a hole-dominant *p*-type conductor. It is worth mentioning the coincidence between the anomalous increase of spontaneous

magnetization and the decrease in carrier density n below approximately 10 K. The result suggests that there is a close relation between magnetism and the carrier characteristics. The plots of the resistivity and carrier density and their logarithmic plots are shown in Fig. 4. The temperature dependence of the plots indicates trapping characteristics at liquid-He temperature and the existence of trapping states in GaN:Mn. The temperature dependence of the charge density is relatively gentle in the high-temperature region. As the intrinsic gap energy of 3.4 eV in GaN is quite large, it can be considered that the high-temperature region corresponds to the exhaustion range for impurity conduction [25].

The trapping energy in the lowest temperature range is estimated to be about 0.04 meV, representing a shallow trapping potential. In the p-d scattering model of a delocalized carrier system, such a decrease of carrier density with decreasing temperature doesn't occur, although the resistance increases with decreasing temperature due to the Kondo effect. The following concept of a weak localization mechanism could be applicable for this shallow localized level: The randomness of the distribution of impurity ions gives rise to a sharp localized level at low temperatures. At higher temperatures, thermal motion results in the broadening and hybridization of the localized level, and the impurity band forms. The experimental data, in which the conductivity is proportional to $\log T$ below 20 K, as shown in the inset of Fig. 3(A), supports this weak localization model.

Generally, ferromagnetism in high-T_C 3d conductors is closely related to the strong magnetic interaction between carriers in the narrow 3d-band originating from localized magnetic cores. In the case of GaN:Mn, the most plausible origin of the shallow level is the formation of an impurity band from acceptors of Mn in GaN and the hopping carriers on Mn ions in the higher temperature region. The spin-dependent hopping band

of Mn impurities near the Fermi level in GaN:Mn is considered to be the origin of the ferromagnetism. The shallow energy gap of 0.04 meV is readily explained by the weak localization model, and the hopping conduction mechanism on Mn impurities implies relatively large magnetic scattering. In fact, as is seen in Fig 3(A), the large change in magnetoresistance predicted by this model was observed in the low temperature range.

3.4 Origin of ferromagnetism in GaN:Mn

The coexistence of ferromagnetic and paramagnetic parts is characteristic of DMS materials [13], and remains a general and intrinsic problem. The most characteristic property of DMS materials is the random distribution of magnetic impurities, including lone impurities and other magnetic impurities with ferromagnetic couplings. Such lone magnetic impurities are considered to be the origin of the paramagnetism. The most important and interesting problem is why the inclusion of these magnetic impurities results in such high-T_C ferromagnetism in DMS materials. It is expected that this high-T_C ferromagnetism is related to strong exchange coupling between magnetic impurities in the impurity band.

The localized properties of the carriers appear remarkably at low temperature. In fact, as discussed in the previous section, an anomalous increase in spontaneous magnetization is observed in the lowest temperature range below 10 K, consistent with the decreasing carrier density at those temperatures. This indicates that electron trapping in Mn impurities increases the magnetization.

Although the origin of the high-T_C ferromagnetism has yet to be confirmed clearly, a tentative model is proposed here that offers a consistent explanation of the experimental results. The model is based on DMS band theory given by Sato and Katayama [9]. According to the theory, the narrow 3*d*-band of Mn impurities in GaN is generated near

the Fermi level. The impurity band originates from the anti-bonding orbits of the t_{2g}-level in the Mn impurities. It should be emphasized that the anti-bonding orbit adopts a spin-triplet state that allows ferromagnetic spin transfer between Mn ions, affording the double-exchange mechanism. The theory predicts a slight energy gap between the Fermi energy and the anti-bonding t_{2g}-band for ideal GaN crystal. However, it is noted that actual GaN tends toward an *n*-type semiconductor. In fact, GaN produced by NH_3-MBE has an electron density of 10^{26} m^{-3}. As the electron density in *n*-type GaN is about 10 times the hole density of *p*-type GaN:Mn, it is reasonable that the excess electrons cause the Fermi energy to pull up to the t_{2g}-band, resulting in magnetic scattering that produces the ferromagnetism.

A general model of this high-T_C ferromagnetism would then be as follows. Spin splitting or shift between the up- and down-spin bands of hopping carriers is generated by double-exchange interaction. Ferromagnetic parallel spin hopping then originates from anti-bonding orbitals in Mn impurities. The characteristics of the strong exchange interaction in GaN:Mn arises from the mechanism of the double-exchange interaction, being a direct exchange interaction between Mn spins. This interaction is a lower rank perturbation compared to indirect exchange interactions such as the *p*-*d* (or *s*-*d*) interaction discussed in Ref. [8] in that direct exchange interaction does not occur via the conduction electron.

This tentative model also explains the field and temperature dependence of the magnetization in the lowest temperature region. Such a localization of hopping electrons prevents hopping and suppresses the ferromagnetic double exchange interaction, resulting also in prevention of ferromagnetic band splitting. This localization also increases the magnetic moment. This effect is reasonable if the hopping electrons become trapped and are fed back to the divalent Mn states. The trapping

process itself increases the localized moment because spin polarization per electron in the conduction band is usually lower than spin polarization of the localized_electrons. This satisfactorily explains why the exchange interaction between Mn ions is suppressed at low temperatures, while the localized moment is larger than for the ferromagnetic phase. This model also explains the coincidence of the temperature regions in which carrier density decreases and magnetization increases.

For this tentative model to hold, the Curie temperature and magnetization should exhibit an Mn concentration dependence. In fact, the saturation moment does exhibit a large concentration dependence, however, experimental error in the magnetization curve prevents exact determination of T_C. Therefore, the relationship between Mn concentration and T_C remains to be captured satisfactorily. The experimental error is attributed primarily to the low sensitivity of measurement using the oven system in the SQUID magnetometer. In order to make further clarify this high-T_C ferromagnetism, a more detailed and sensitive experimental method will need to be developed for examination of samples with various Mn compositions.

4. Conclusions

In summary, the magnetic and transport properties of Wurtzite GaN:Mn films have been reported. The magnetic experiments on GaN:Mn revealed the coexistence of paramagnetic and ferromagnetic components. The highest Curie temperature of the ferromagnetism observed in this work was about 940 K, with a clear hysteresis loop as high as 400 K. The transport properties exhibited a close relation between magnetism and impurity conduction. An anomalous increase in spontaneous magnetization observed below 10 K was consistent with the decreasing carrier density at those temperatures, suggesting a localization of hopping electrons. The double exchange

mechanism of the Mn impurity band was proposed to account for the high ferromagnetic transition temperature in this film.

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Table caption

Table 1. Growth conditions and RHEED pattern of GaN:Mn film and GaN buffer layer.

	GaN	GaN:Mn
Cell temperature (K)		
Ga	1170	1120
Mn	-	850
NH_3 flow (10^{-3}Pa·m^3/s)	8.4	8.4
Substrate temperature (K)	990	990
Thickness(Å)	2000	3600
RHEED pattern		

Figure captions

Fig. 1. Temperature dependence of magnetization (*M*-*T*) at 0.1 (closed circles) and 7 T (open circles) in the temperature range 1.8 to 300 K. Open squares represent magnetization at the magnetic field at the top of the hysteresis curve measured at each temperature. (Inset) Temperature dependence of magnetization up to 750 K. Solid line represents curve calculated by molecular field approximation. The estimated ferromagnetic transition temperature T_C is 940 K.

Fig. 2. Magnetic field dependence of magnetization (*M*-*H*) at 1.8 and 300 K. Broken line represents the magnetic field at the top of the hysteresis loops. (Inset) Magnetization processes up to 7 T.

Fig. 3. (A) Magnetoresistance data at various temperatures between 1.8 and 200 K. The magnetic field was applied parallel to the film plane and swept between –10 and +10 T. (Inset) Logarithmic temperature dependence of conductivity. Broken line is the fitting curve representing the log*T* dependence. (B) Hall resistance data at various temperatures between 1.8 and 200 K with correction for included magnetoresistance. (Inset) Raw data.

Fig. 4. (A) Temperature dependence of resistivity and carrier density as estimated from Hall resistivity data. (B) Logarithm of resistivity and carrier density as a function of reciprocal temperature.

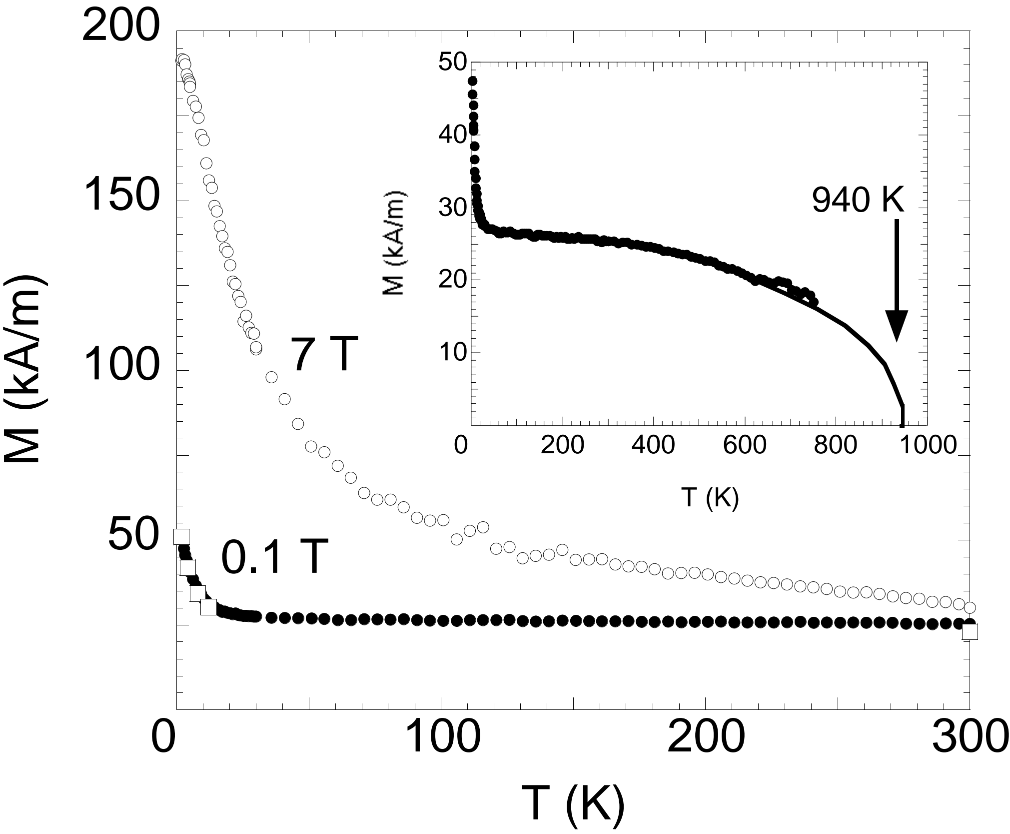


Fig.1 H. Hori et al.

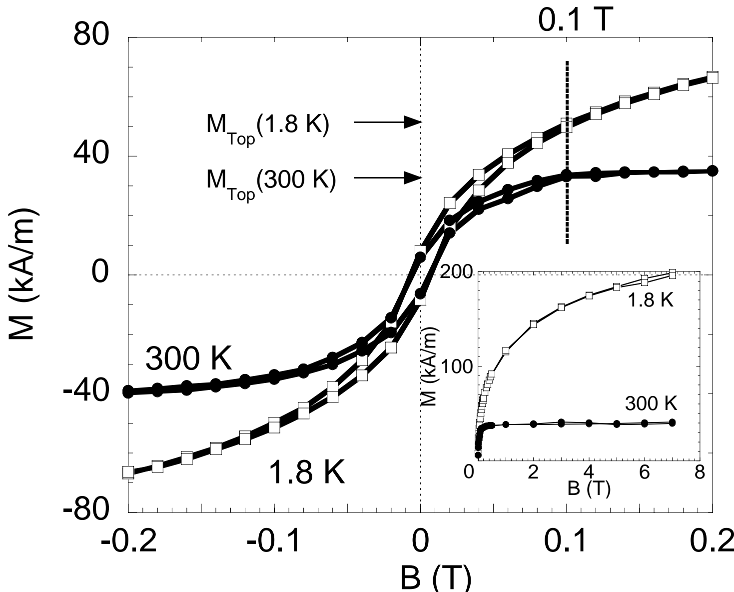
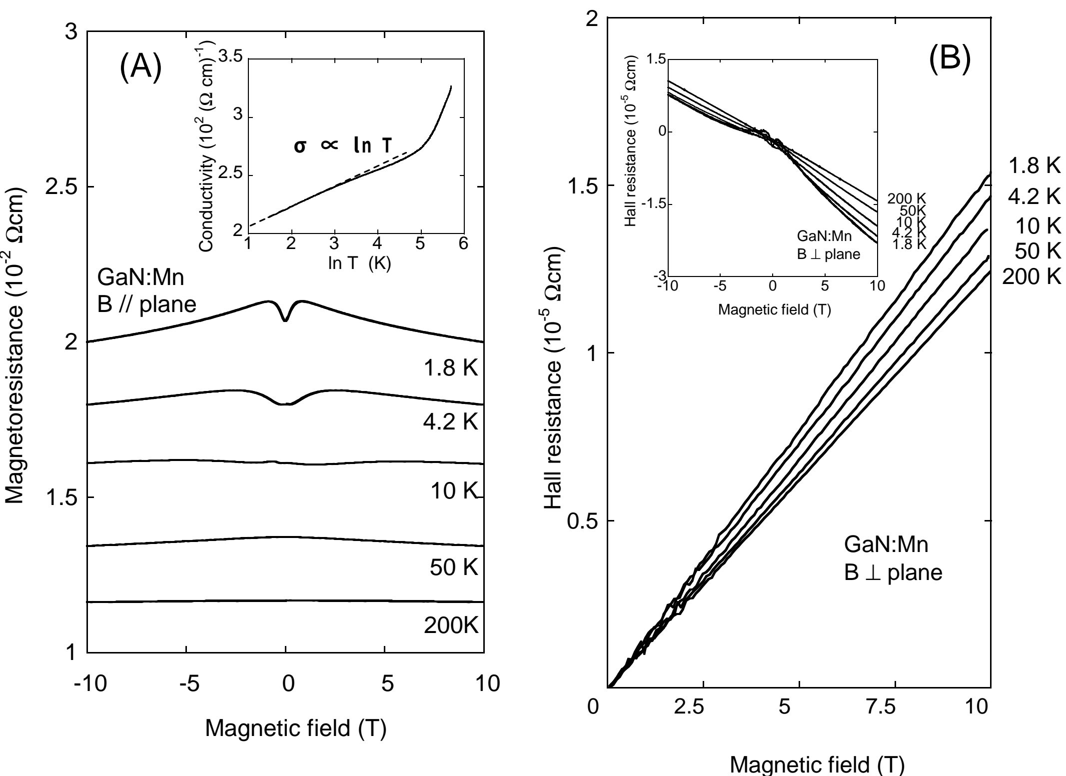


Fig. 2 H. Hori et al.



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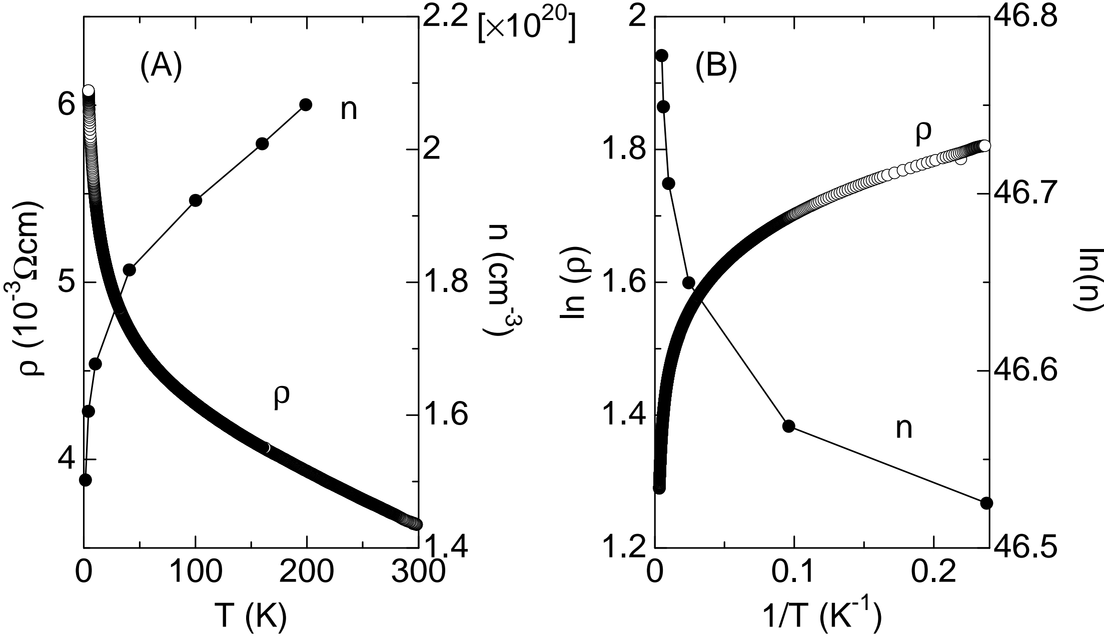

Fig. 4 H. Hori et al.