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Mn-doped GaN films on sapphire (0001) substrates were grown by molecular beam epitaxy system using ammonia as nitrogen source. The result of magnetization measurement gives Curie temperature as high as 940 K. The field and temperature dependencies of the magnetization show coexistence of ferromagnetic and paramagnetic phases. In addition, the temperature dependencies of electrical resistance and carrier concentration were measured to investigate the relation between the ferromagnetism and transport property. Below about 10 K, a similar anomalous increase of magnetization and resistance is observed.
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I. INTRODUCTION

Since the first fabrication of the Mn doped InAs, (InAs:Mn),[1] research on the diluted magnetic semiconductors (DMSs) has attracted much interest as they combine the two large branches in condensed matter physics; semiconductor and magnetism. The result has led to intensive investigations on the group III–V based DMSs successively.[1–5] Moreover, there are some theoretical predictions of the possibility of GaN-based DMS having Curie temperature exceeding room temperature.[6–8] In a recent work, Sonoda *et al.*[9] reported that ferromagnetic like Mn-doped GaN film has the Curie temperature much higher than 400 K. In this study, we report on the magnetic and transport characteristics of the Mn-doped GaN film. A relation between magnetism and the impurity conduction is shown as well as the coexistence of ferromagnetic and paramagnetic phases.

II. EXPERIMENT RESULTS

The wurtzite Mn-doped GaN films were grown on the sapphire (0001) substrates by the molecular beam epitaxy (MBE) as reported in the previous work.[9] The MBE in this work was of the model ULVAC MBC-100, equipped with a reflection high energy electron diffraction (RHEED). The Mn-doped GaN films with thickness ranging from 1300 to 3600 Å were grown at each growth temperature. The RHEED observation revealed that the single crystal films of the wurtzite Mn-doped GaN are grown. The growth condition of the crystal structures in the films is discussed in detail in Ref. 9. The Mn-concentration in the samples used in this work was about 3%. The concentration was estimated by the secondary mass spectroscopy experiment (Model ims-5f, CAMECA Co. Ltd.) at JAIST. Magnetization measurements of the Mn-doped GaN films were carried out by the use of a superconducting quantum interference device magnetometer (MPMS-XL, Quantum Design Co. Ltd.). The magnetic field was applied parallel to the film plane in this work because of the easy-plane type anisotropy of the film.

Figure 1 shows the applied field dependency of the magnetization, M-H curves at 1.8 and 300 K. Each loop shows clear hysteresis. In a recent M-H measurement at 400 K, the hysteresis has been observed. Because of the increased noise level in the magnetization measurement above 400 K, due to the unstable temperature, the appearance of the hysteresis loop is ambiguous. However, temperature dependent magnetization (M-T) was carried out up to 750 K at a constant applied magnetic field of 0.1 T. The result is presented in the Fig. 1(a). The field 0.1 T corresponds to the magnetization just above the hysteresis loop of the M-H curve of 1.8 K. A

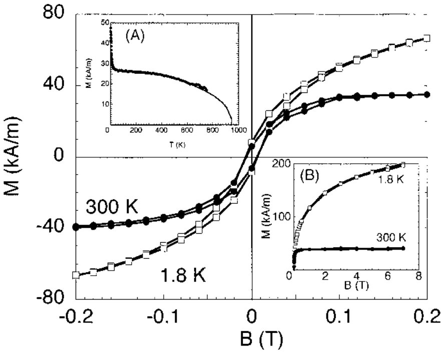

FIG. 1. Field dependency of the magnetization $(M\text{-}H)$ curves at 1.8 and 300 K. The inset (a) shows the temperature dependence of the magnetization $(M\text{-}T)$ up to 750 K and the solid line represents the calculation curve by use of the molecular field approximation. The ferromagnetic transition temperature T_C is estimated to be 940 K. (b) shows the magnetization processes up to 7 T.

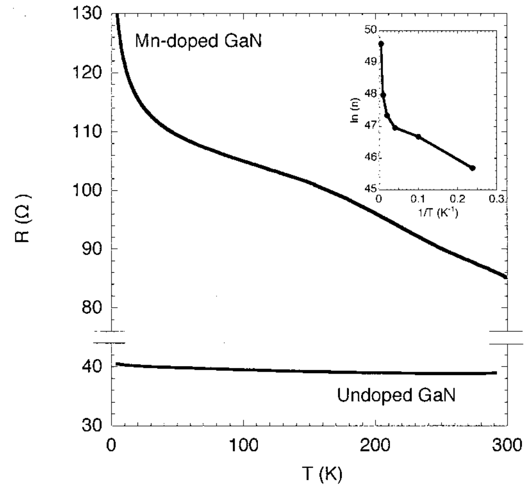

FIG. 3. Temperature dependency of the resistance of Mn-doped and undoped GaN. The inset shows the logarithmic plots of the carrier density of Mn-doped GaN versus $1/T$ of horizontal axis.

fitting to the theoretical curve by the mean field approximation predicts that the T_C is about 940 K.

Considering Fig. 2 and Fig. 1(b), the steep increase in $M\text{-}T$ curve at 0.1 and 7 T and the $M\text{-}H$ curve at liquid He temperature look like a ''coexistence'' of paramagnetic and ferromagnetic phases.[10] The magnetization curve at 7 T is getting close to the curve of the ''spontaneous magnetization'' in 0.1 T at temperature higher than the room temperature. In this case, however, the magnetization at the top of the hysteresis loop or at the critical field of the single domain also has the steep increase. The data is illustrated by open squares in Fig. 2. Moreover, the temperature dependency of the remanent moment also shows anomalous increase below 10 K (inset of Fig. 2). It means that the steep increase includes both ferromagnetic and paramagnetic phases in the present sample.

The electrical conductivity and the Hall measurements of Mn-doped GaN were performed in order to obtain the relation between the transport characteristic and the magnetism. The present sample exhibits negative type conduction but the contribution of the buffer layers is considerable. Figure 3 shows the temperature dependency of the resistance of Mn-doped and undoped GaN. The data of Mn-doped GaN was only estimated the anomalous increase of resistance below about 10 K. The inset of Fig. 3 shows the logarithmic of the carrier density n versus $1/T$ horizontal axis. The data shows characteristics of the typical impurity conduction of the hopping electrons in semiconductors.[11]

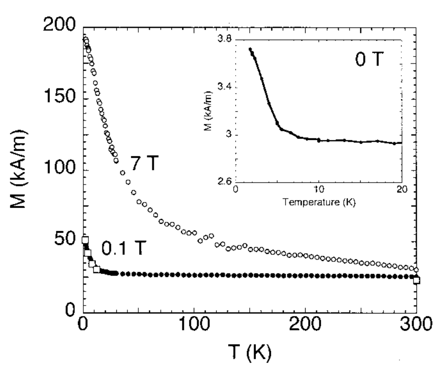

FIG. 2. Temperature dependency of the magnetization $(M\text{-}T)$ at 0.1 T (closed circles) and 7 T (open circles) in the temperature range between 1.8 and 300 K. Open squares represent the magnetization at the magnetic field of the top of the hysteresis curve measured at each temperature. The inset shows the temperature dependence of the remanent magnetization.

III. DISCUSSION AND CONCLUSION

The large field dependence of the magnetizations and the hysteresis loop of Mn-doped GaN are clearly observed up to 400 K.

It may be inferred that the segregation of some ferromagnetic compounds may be an origin of the high T_C ferromagnetism of this present sample. But the possibility is denied because of the following reasons: (1) The segregated small ferromagnets should show characteristics of the super paramagnetism. The observed $M\text{-}H$ curve, however, is completely different from the super paramagnetism. It is typically ferromagnetic above room temperature, (2) uniform distribution of Mn in the sample is confirmed by the measurement of Rutherford back scattering (Model 1700H, Nisshin-High Voltage Co. LTD) at JAIST, (3) the result of extended x-ray absorption fine structure measurement indicates that the Mn atoms in the present sample substitute for the Ga sites in a wurtzite GaN lattice,[12] (4) the Curie temperature of the present sample is much higher than 748 K comparing with another high T_C ferromagnet like Mn-Ga alloys and Mn_4N.[13,14] The α-Mn metal and the ionic compound MnO are also possible candidates but they are antiferromagnetic.

The magnetization process shows a ''coexistence'' of paramagnetic and ferromagnetic components but anomalous increase of the remanent magnetization is also observed in the localized electron region of the lowest temperature range. The result shows that the ''coexistence'' characteristic is not simple to analyze. On the other hand, the coincidence between the anomalous increase of the spontaneous magnetization and the resistance below about 10 K suggest a close relation between the magnetism and the carrier. These experimental results support that the Mn-doped GaN grown by the MBE in this work is new intrinsic ferromagnet of DMS.

In order to discuss the experimental results obtained in this work, a tentative model is considered based on the band theory of the ferromagnetism in DMS given by Sato and Katayama.[7] High T_C ferromagnetism is closely related with the thermally excited three-dimensional-electrons from the localized level on Mn impurity. The excited hopping electrons in Mn-doped GaN make a narrow spin polarized band because of the double exchange mechanism for t_{2g} level in Mn. The band magnetism in narrow band materials generally shows strong exchange coupling and high T_C ferromagnetism as seen in many iron family magnets.

[1] H. Munekata, H. Ohno, S. von Molnar, A. Segmullerr, L. L. Chang, and L. Esaki, Phys. Rev. Lett. **63**, 1849 (1989).
[2] H. Ohno, H. Munekata, T. Penney, S. von Molnar, and L. L. Chang, Phys. Rev. Lett. **68**, 2664 (1992).
[3] H. Munekata, A. Zaslavsky, P. Fumagalli, and R. J. Gambino, Appl. Phys. Lett. **63**, 2929 (1993).
[4] S. Koshihara, A. Oiwa, M. Hirasawa, S. Katsumoto, Y. Iye, C. Urano, H. Takagi, and H. Munekata, Phys. Rev. Lett. **78**, 4617 (1997).
[5] Y. Ohno, D. K. Young, B. Beschoten, F. Matsukura, H. Ohno, and D. D. Awschalom, Nature (London) **402**, 709 (1999).
[6] T. Diel, H. Ohno, F. Matsukura, J. Cibert, and D. Ferrand, Science **287**, 1019 (2000).
[7] K. Sato and H. K. Yoshida, Jpn. J. Appl. Phys., Part 2 **40**, L485 (2001).
[8] M. Zajac, R. Doradzinski, J. Gosk, J. Szczytko, M. Lefeld-Sosnowaka, M. Kaminska, A. Twardowski, M. Placzewska, E. Grzanka, and W. Gebicki, Appl. Phys. Lett. **78**, 1276 (2001).
[9] S. Sonoda, S. Shimizu, T. Sasaki, Y. Yamamoto, and H. Hori, J. Cryst. Growth (to be published) (cond-mat/0108159).
[10] F. Matsukata, H. Ohno, A. Shen, and Y. Sugawara, Phys. Rev. B **57**, R2037 (1998).
[11] K. Seeger, *Semiconductor Physics* (Springer-Verlag, New York, 1973), p. 4–8.
[12] M. Sato, H. Tanida, K. Kato, T. Sasaki, Y. Yamamoto, S. Sonoda, S. Shimizu, and H. Hori, Jpn. J. Appl. Phys. (to be published).
[13] T. A. Bither and W. H. Cloud, J. Appl. Phys. **36**, 1501 (1965).
[14] M. Mekata, J. Phys. Soc. Jpn. **17**, 796 (1962).