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Evidence for Carrier-Induced High-T_C Ferromagnetism in Mn-doped GaN film

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Abstract. - A GaN film doped with 8.2 % Mn was grown by the molecular-beam-epitaxy technique. Magnetization measurements show that this highly Mn-doped GaN film exhibits ferromagnetism above room temperature. It is also revealed that the high-temperature ferromagnetic state is significantly suppressed below 10 K, accompanied by an increase of the electrical resistivity with decreasing temperature. This observation clearly demonstrates a close relation between the ferromagnetism with extremely high-T_C and the carrier transport in the Mn-doped GaN film.

The discovery of ferromagnetism in Mn-doped InAs and GaAs [1–3] has stimulated intensive research on the III-V based diluted magnetic semiconductors (DMSs), because DMSs are considered to be potential materials for future spintronics devices which combine both charge and spin degrees of freedom in semiconductor electronics. Since practical spintronics devices are expected to work around and above room temperature, DMSs should have Curie temperatures T_C in excess of 400 K and considerable efforts have been devoted in the recent decade to synthesize such films. As to GaN-based DMSs, progress in GaN growth [4] and device development [5] have been remarkable in the past decade and high-conductivity, p-type epitaxial layers can be produced [6]. Blue-light-emitting diodes are already commercial products [7], and blue-laser diodes [8], ultraviolet detectors [9], and high-power, high-temperature field-effect transistors [10] have been achieved and have been improved rapidly. Accordingly, the technological importance of room-temperature ferromagnetic GaN for spintronics is very large. Recently, some of the present authors have reported the successful growth of Mn-doped GaN (GaMnN) films showing room-temperature ferromagnetism [11–14]. So far, the highest T_C is around 940 K in 5.7 % Mn-doped GaN. Although the origin of high-temperature ferromagnetism in these GaMnN systems is still under discussion, conducting carriers are thought to play a substantial role in mediating a strong ferromagnetic interaction between the magnetic moments.

Similar to the importance of the control of the carrier density for achieving the best semiconducting properties, the carrier density plays a crucial role in determining the magnetic properties of the material as well. Previously, we have studied the relation between the transport properties of GaMnN film and the high-T_C ferromagnetism [14, 15]. For a GaMnN film with 5.7 % Mn, a steep decrease of the spontaneous magnetization with increasing temperature was found below 10 K, while the ferromagnetism persists up to above 750 K. Furthermore, a steep decrease of the resistivity, or an increase of carrier density, with increasing temperature was found in the same low-temperature region. These results have been discussed in terms of the double-exchange mechanism due to the hopping electrons of the Mn-impurity band, where the decrease of the spontaneous moment with increasing temperature below 10 K was attributed to delocalization of localized carriers that also play a principal role in high-T_C ferromagnetism.

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These studies have well established the close relation between the carrier transport and the magnetism in high-T_C GaMnN film. However, the precise role of carrier transport in the development of the high-temperature ferromagnetic state is still unclear.

In this paper, we demonstrate evidence for carrier-induced ferromagnetism in GaN film doped with 8.2 % Mn, grown by the molecular-beam-epitaxy (MBE) technique. Magnetization measurements point out that the GaMnN film is ferromagnetic above room temperature. Surprisingly, we found that the high-temperature ferromagnetism is significantly suppressed below 10 K, accompanied by a steep increase of the electrical resistivity. This observation strongly suggests that the high-T_C ferromagnetic state is driven by carrier conduction.

The fabrication of the GaMnN film was carried out by an ammonia-MBE technique. Details of the sample preparation have been described elsewhere [11]. The wurtzite crystal structure of the obtained GaMnN film was confirmed by X-ray diffraction (XRD). The XRD study also showed that there is no secondary phase within the resolution limit. An X-ray absorption fine structure (XAFS) study confirmed that the Mn atoms are substituted for Ga atoms. By means of electron probe micro analysis (EPMA), the Mn concentration in the film was established to be 8.2 %.

The temperature and field dependence of the magnetization were measured by using a superconducting quantum interference device (SQUID) magnetometer (MPMS-XL, Quantum Design Co. Ltd.). The temperature variation of the in-plane electrical resistivity was measured by a conventional four-probe dc method.

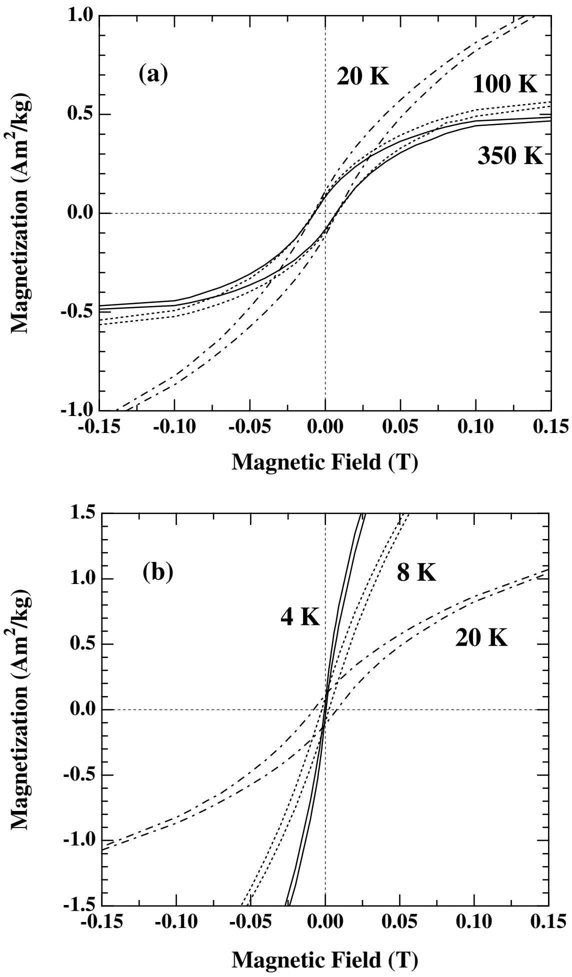

Fig. 1: Magnetization process at various temperatures.

The field dependence of the magnetization has been investigated at various temperatures up to 350 K. Figure 1 shows representative results of M-H curves in a magnetic field parallel to the film plane. At fields below 0.1 T, hysteretic behavior is quite pronounced in the 20 K, 100 K and 350 K curves (fig. 1(a)), where both the coercive field B_c and the remanent magnetization M_r are almost independent of temperature above 20 K. This result clearly shows the high-T_C ferromagnetism of the present heavily-doped GaMnN film with a Curie temperature far above room temperature. Upon further cooling below 20 K (fig. 1(b)), however, the hysteresis is considerably reduced and also M_r decreases as can be seen in the 4 K curve.

In order to clarify the evolution of the high-T_C ferromagnetic state with temperature, the temperature dependence of the remanent magnetization M_r has been investigated. In fig. 2, the M_r-T curve, as recorded during a temperature cycle 350 K $\rightarrow$ 2 K $\rightarrow$ 350 K, is shown. The measurement was performed in zero field, after a field of 1 T had been applied along the film plane for a few minutes at 350 K. The nearly T-independent behavior of M_r confirms the presence of a ferromagnetic state with a T_C value of more than 350 K. Surprisingly, below about 10 K, a steep decrease of M_r occurs and there is no substantial difference between the curves recorded with descending and ascending temperature. It should be noted that the difference between the two curves around 100 K is due to experimental error and not intrinsic. We also emphasize that a similar T-dependence of M_r has been observed when the measurement is performed after applying a field perpendicular to the film plane.

The observed significant suppression of the high-temperature ferromagnetic state below 10 K convincingly excludes a contribution of a possible secondary phase, such as ferromagnetic Mn_xN_y or Ga_xMn_y as a plausible origin of the observed high-T_C ferromagnetism, because these ferromagnets do not show such a reduction of the ferromagnetism at low temperatures [16, 17]. The reversible behavior of M_r upon temperature variation further clarifies that the decrease of M_r does not arise from rotation of magnetic domains or freezing of Mn spins because these effects should cause an irreversible behavior of M_r.

The high-T_C ferromagnetism observed in the present GaMnN film is clearly related to the carrier transport. Figure 2 shows that the temperature dependence of the electrical resistivity ρ is only gradual at temperatures above 20 K, while it shows a steep increase with decreas-

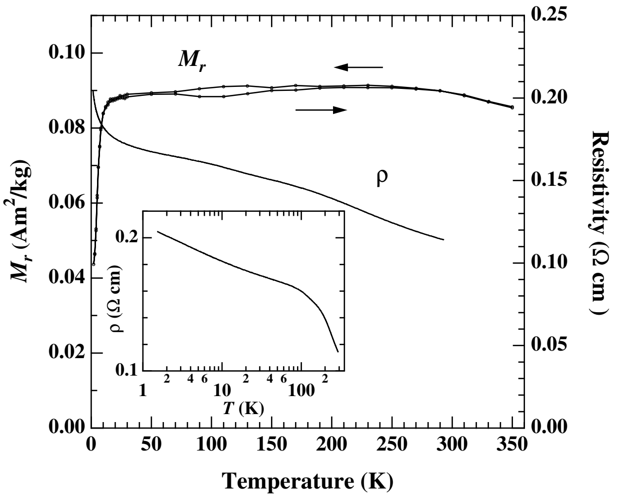

Fig. 2: Temperature dependence of the remanent magnetization M_r and the electrical resistivity ρ. M_r and ρ were measured after keeping the sample in applied field of 1 T for a few minutes at 350 K and 270 K, respectively. (Inset) Electrical resistivity plotted on a logarithmic temperature scale.

ing temperature below 20 K. In addition, as shown in the inset of fig. 2, below 10 K a weakly-localized nature of the carriers is suggested by the ρ vs $\log T$ dependence. Comparison of the ρ vs T and M_r vs T curves in fig. 2 clearly demonstrates the close relation between the transport and magnetic properties. The suppression of the high-T_C ferromagnetic state below 10 K coincides with a decrease of the carrier conduction which should be attributed to an increased localization, while the high-T_C ferromagnetic state appears in the temperature regime of delocalized carriers. We emphasize again the simultaneous decrease of M_r and increase of ρ at low temperatures, and the T-reversible behavior of these both quantities, resulting in conclusive evidence that the observed high-T_C ferromagnetism is an intrinsic property of the present GaMnN film and that the carriers play a dominant role in the high-T_C ferromagnetism.

Theoretical investigations of the mechanism of the high-T_C ferromagnetism in GaMnN films have been carried out by several approaches, but the definitive answer has not yet been given. Dietl et al. [18, 19] have proposed a mean-field Zener p-d exchange-interaction model, which explains many characteristics of the GaMnAs system. This model is based on the idea that Mn 3d state is located within or near the valence p-band and that the polarization of the valence p-band, due to hybridization between Mn 3d-levels and p-band states, mediates the magnetic interaction between localized Mn spins. On the other hand, first-principles electronic-structure calculations have indicated that the Mn 3d state forms a pronounced impurity band in the wide band gap in GaN and that the Fermi energy is located in this impurity band [20–23]. A double-exchange type of mechanism was concluded to be dominant in ferromagnetic GaMnN, with hopping Mn d-electrons in the impurity band playing a crucial role in the ferromagnetic coupling between the Mn spins. The theoretical calculations also pointed out that the exchange interaction in GaMnN is short-range, whereas there are long-range correlations in GaMnAs.

At present, the lack of detailed knowledge of the electronic structure of the studied GaMnN film prevents a substantial discussion on the interaction mechanism that leads to the ferromagnetism with high T_C. However, formation of a Mn 3d impurity band in the gap was suggested by a XANES (X-ray absorption near edge structure) study, which has been performed on a GaMnN film with 5.7 % Mn and T_C of 940 K [13]. Furthermore, a recent NEXAFS (near edge X-ray absorption fine structure) study has revealed the coexistence of Mn atoms with valencies $Mn^{2+}$and Mn^{3+} in high-T_C GaMnN film [24]. With these pieces of experimental evidence, the intimate correlation between carrier trapping and the suppression of the high-T_C ferromagnetism below 10 K observed in the present study suggests that the ferromagnetic coupling between Mn spins is mediated by Mn 3d electrons hopping between Mn^{2+} and Mn^{3+}. In this scenario, the temperature evolution of the ferromagnetic state observed at low temperatures can qualitatively be understood as follows: the weak localization of carriers evidenced by the ρ vs $\log T$ dependence below 10 K, which must be caused by a random distribution of impurity ions, reduces the carrier conduction. Because of the short-range correlations as predicted by the theoretical calculations, reduction of the carrier conduction weakens the ferromagnetic interaction of the double-exchange mechanism between Mn spins, thus leading to the suppression of the high-T_C ferromagnetic state in the same temperature region.

Finally we would give comments on recent theoretical studies in which effects of inhomogeneity in DMSs are discussed as an origin of high Curie temperature [25, 26]. In the calculations, spinodal decomposition under layer by layer crystal growth condition forms quasi-one-dimensional "Konbu phase" which are connected on the surface of each layer, resulting in high-T_C ferromagnetism. From the present studies, unfortunately, we can not make conclusive remarks if a spinodal decomposition is really an origin of high-T_C ferromagnetism observed in our GaMnN film. However it is worth noting that an easy magnetization direction of the present GaMnN film exist in the film plane and the magnetic anisotropy between parallel and perpendicular to the film plane is small, which is not likely to the case predicted by the spinodal decomposition calculation. It is expected from the theory that the system under consideration possesses a considerable magnetic anisotropy with easy magnetization direction perpendicular to the film plane. We further stress that the decrease of remanent magnetization accompanied with the increase of resisitance upon cooling in zero field and the reversibility of the remanent magnetization against temperature can not be explained by the "Konbu phase" scenario, because such a ferromagnetic phase is usually stabilized at low temperatures.

In summary, we have investigated the ferromagnetic properties of GaMnN film with a high T_C value. By measuring the remanent magnetization M_r in different temperature cycles, it has been established that the high-T_C ferromagnetic state is weakened below 10 K. The observation that the localization of conducting carriers coincides with the suppression of the high-T_C ferromagnetism provides evidence that the ferromagnetic interaction responsible for the high T_C value of the GaMnN film is mediated by carrier conduction.

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