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A comparative study on magnetic and magnetotransport properties in (Ga, Mn)N epitaxial films grown on undoped and n-type GaN by PEMBE

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Abstract

We present a comparative study on the magnetic and magnetotransport properties of (Ga, Mn)N epitaxial films grown on undoped GaN and n-type GaN templates by plasma-enhanced molecular beam epitaxy. Regardless of the kind of substrate, the (Ga, Mn)N epitaxial films were found to obviously exhibit ferromagnetic ordering with a Curie temperature exceeding room temperature. However, the negative magnetoresistances of the (Ga, Mn)N films grown on undoped GaN and n-type GaN templates show a remarkable difference. Negative magnetoresistance for the (Ga, Mn)N films grown on undoped GaN templates was observed up to room temperature, while negative magnetoresistance for the (Ga, Mn)N grown on n-type GaN templates was observed only below 100 K. Considering that the Mn concentrations in the (Ga, Mn)N films are the same, it is believed that the content of Mn spins in the (Ga, Mn)N films grown on n-type GaN templates is not sufficient to order all the carriers in the n-type GaN and the film itself, since the Mn spins in (Ga, Mn)N interact with the carriers in both.

1. Introduction

Diluted magnetic semiconductors (DMSs) have attracted great attention, since they offer novel functionalities by introducing a spin degree of freedom in traditional semiconductors [1, 2]. Conventional electronic and optoelectronic semiconductor devices are controlled by means of the charge of the carriers while spintronic semiconductor devices combined with DMSs are manipulated by means of the spin in addition to the charge of the carriers, leading to spin-polarized light emitting diodes (LEDs) [3], spin field effect transistors (FETs) [4], and spin resonant tunnelling diodes (RTDs) [1].

In recent years, (Ga, Mn)As has been intensively studied as a representative DMS, but its Curie temperature (T_C) stays below ~140 K [5], far from room temperature. It is necessary to obtain T_C for the DMSs exceeding room temperature for practical device applications. In this context, wide bandgap semiconductors are of particular importance, since Dietl *et al* [6] predicted T_C above room temperature for Mn-doped GaN and ZnO on the basis of the mean-field Zener model of ferromagnetism. In particular, the host GaN has been used in various electronic and optoelectronic devices such as blue LEDs and laser diodes (LDs) [7]. Moreover, according to the prediction of Kronik *et al*, (Ga, Mn)N forms a 100% spin-polarized impurity band due to hybridization of Mn 3d and N 2p orbitals [8]. Therefore, it is believed that GaN is a very suitable material for spintronic semiconductor device applications. More recently, (Ga, Mn)N DMSs with T_C exceeding room temperature have been reported by several groups [9–12].

In this paper, we report on a comparative study on the magnetic and magnetotransport properties of (Ga, Mn)N epitaxial films grown on undoped GaN and n-type GaN templates by plasma-enhanced molecular beam epitaxy (PEMBE). The (Ga, Mn)N epitaxial films were found to exhibit n-type conductivity and ferromagnetic ordering above room temperature. However, the negative magnetoresistances of the (Ga, Mn)N films grown on undoped GaN and n-type GaN templates exhibited a significant difference.

2. Experiment

The growth of (Ga, Mn)N epitaxial films on undoped GaN and n-type GaN templates was carried out in a PEMBE system under ultrahigh-vacuum (UHV) conditions with a base pressure of $\sim 1 \times 10^{-10}$ Torr. The source materials were high-purity Ga and Mn metals (6N and 5N5, respectively). As the nitrogen source, high-purity (6N) nitrogen gas was supplied through a radio-frequency (rf) plasma source. For the epitaxial growth and examining the indirect influence of the conductivity, 2 μm thick undoped GaN and n-type GaN ($n \approx 3 \times 10^{18}$ cm^{-3}) templates on (0001) sapphire, grown by metal–organic chemical vapour deposition (MOCVD) prior to the growth of the (Ga, Mn)N layers, were used as substrates. In particular, the growth of (Ga, Mn)N films on n-type GaN templates is essential, since DMS thin films used in fabricating spintronic devices such as spin LEDs and spin FETs were grown on conducting or semiconducting materials such as metals or n-type semiconductors. The (Ga, Mn)N films were grown at 700 °C with Ga cell temperatures of 1000 °C. The Mn concentration in the films was controlled by changing Mn cell temperature between 650 and 750 °C. The thickness of the (Ga, Mn)N films was approximately 500 nm.

X-ray diffraction (XRD) and transmission electron microscopy (TEM) [13] analyses revealed that the wurtzite (Ga, Mn)N films were substitutional single-phase solid solutions without nanoclusters and secondary phases. This indicates that manganese atoms effectively replaced gallium atoms located at the substitutional sites in the (Ga, Mn)N structure. Mn concentrations were measured by secondary ion mass spectroscopy (SIMS), and they were $x = 0.2\%$ and 0.92% in the $(Ga_{1-x}Mn_x)N$ films with Mn cell temperature of 650 and 750 °C, respectively. The magnetic behaviour was studied using a superconducting quantum interference device (SQUID) magnetometer and a high-sensitivity (10^{-8} emu) alternating gradient magnetometer (AGM). The van der Pauw Hall and magnetoresistance (MR) measurements were performed with a physical properties measurement system (PPMS) applying a magnetic field up to 9 T in the temperature range of 5–300 K.

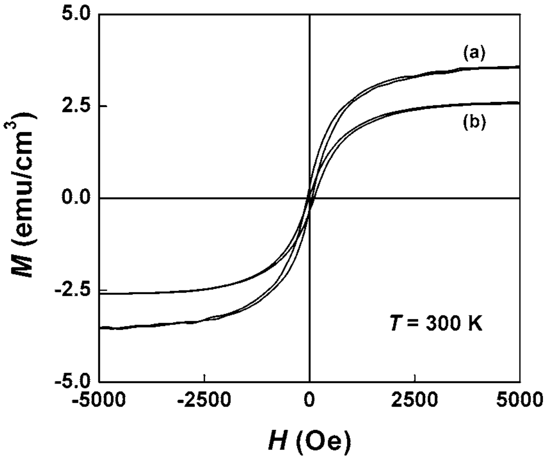

Figure 1. Hysteresis loops for the $(Ga_{1-x}Mn_x)N$ films with $x = 0.92\%$ grown on (a) undoped GaN and (b) n-type GaN templates, obtained with magnetic fields applied parallel to the plane of the samples by an AGM at room temperature. For the (Ga, Mn)N films, the non-magnetic contributions of the GaN template, sapphire substrate, and experimental apparatus have been subtracted from the data.

3. Results and discussion

The magnetic properties of the (Ga, Mn)N films grown on undoped GaN and n-type GaN templates were investigated at room temperature using an AGM that is appropriate for evaluating small magnetization because of its high sensitivity. Figure 1 shows magnetization versus magnetic field (M–H) curves for the $(Ga_{1-x}Mn_x)N$ films with $x = 0.92\%$ grown on undoped GaN and n-type GaN templates at room temperature. Regardless of the conductivity difference between the substrates, clear hysteresis loops for all the (Ga, Mn)N films were observed, indicating that all the samples exhibit ferromagnetic ordering. Even though the two (Ga, Mn)N films have the same Mn concentration, the saturation magnetization (M_s) for the (Ga, Mn)N film grown on the undoped GaN template is 3.6 emu cm^{-3} while M_s for the (Ga, Mn)N film grown on the n-type GaN template is 2.6 emu cm^{-3}—smaller than that for the film grown on the undoped GaN template. The effective magnetic moments calculated from M_s are 1.28 μ_B/Mn and 0.93 μ_B/Mn for (Ga, Mn)N films grown on undoped GaN and n-type GaN templates, respectively. Considering that each Mn atom within GaN has the theoretical magnetic moment of 4 μ_B [8], these findings indicate that the ferromagnetically activated Mn contents in (Ga, Mn)N films grown on undoped GaN and n-type GaN templates are 32% and 23%, respectively, resulting in the occurrence of saturation at fields higher than the coercive fields. The degradation of the magnetic properties in the (Ga, Mn)N films grown on n-type GaN templates as compared with the case of (Ga, Mn)N films grown on undoped GaN templates might be attributed to electrons in n-type GaN templates interacting with Mn spins or carriers in the (Ga, Mn)N films grown.

The temperature dependences of the magnetization for the $(Ga_{1-x}Mn_x)N$ films with $x = 0.92\%$ grown on undoped GaN and n-type GaN templates were measured by a SQUID magnetometer in a magnetic field of 500 Oe and the results are shown in figure 2. The magnetization in both films decreases slowly and continuously with increasing temperature. Both films were clearly observed to exhibit a ferromagnetic ordering in the temperature range of 5–300 K, indicating that T_C for both films exceeds at least room temperature. Moreover,

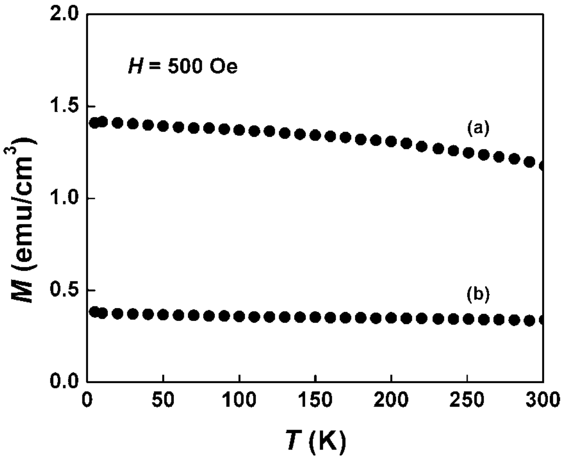

Figure 2. The temperature dependence of the magnetization for the $(Ga_{1-x}Mn_x)N$ films with $x = 0.92\%$ grown on (a) undoped GaN and (b) n-type GaN templates, obtained by a SQUID magnetometer in a magnetic field of 500 Oe.

because Mn in GaN forms a deep acceptor level at E_v+1.42 eV [14] and the Mn concentration in the (Ga, Mn)N films is not high enough to compensate for the native defects, the films exhibit n-type characteristics. Therefore, our results support the assertion that the ferromagnetic ordering in the (Ga, Mn)N is due to the indirect Ruderman–Kittel–Kasuya–Yosida (RKKY) exchange interaction between localized Mn moments mediated by the electron gas [1], as theoretically predicted for the low-carrier-density regime [15].

The magnetotransport properties of the (Ga, Mn)N films were examined with a magnetic field up to 9 T applied perpendicular to the plane of the films in the temperature range of 5–300 K. Figure 3 shows the variation of the magnetoresistance (MR; $\Delta R/R$) for the $(Ga_{1-x}Mn_x)N$ films with (a) $x = 0.2\%$ and (b) $x = 0.92\%$ grown on undoped GaN templates. The contact geometry used for the MR measurements is shown elsewhere [16]. The negative MR in the $(Ga_{1-x}Mn_x)N$ films with $x = 0.2\%$ and 0.92% is found to gradually increase with decreasing temperature, and the maximum values of the negative MR at 5 K reach ~9% and ~14%, respectively. In particular, the $(Ga_{1-x}Mn_x)N$ film with $x = 0.92\%$ exhibits a negative MR in the temperature range of 5–300 K and maintains the negative MR of ~1% up to room temperature. However, the MR shows a slowly saturating behaviour at even higher magnetic fields. Such MR behaviour is probably related to the paramagnetism. So this indicates that not all the Mn spins within (Ga, Mn)N are coupled ferromagnetically, and corresponds to the result from the M–H curve in figure 1. In spite of the coexistence of ferromagnetism and paramagnetism in our (Ga, Mn)N, our results appear to exhibit room temperature ferromagnetic ordering and room temperature negative MR, suggesting that room temperature spintronic devices are feasible. Since the Mn concentration in the (Ga, Mn)N films is very low, the films have low spin correlation among the Mn moments. The interaction between Mn spins and electrons induces spin-disorder scattering which is understood to be the origin of the negative MR in the (Ga, Mn)N films [17, 18]. The difference in MR behaviour between the non-Mn-doped GaN and $(Ga_{1-x}Mn_x)N$ films with $x = 0.92\%$ grown on undoped GaN templates is shown in the inset of figure 3(b). The non-Mn-doped GaN is unlikely to show a negative MR at 5 and 300 K and this difference obviously reveals the effect of Mn doping on GaN.

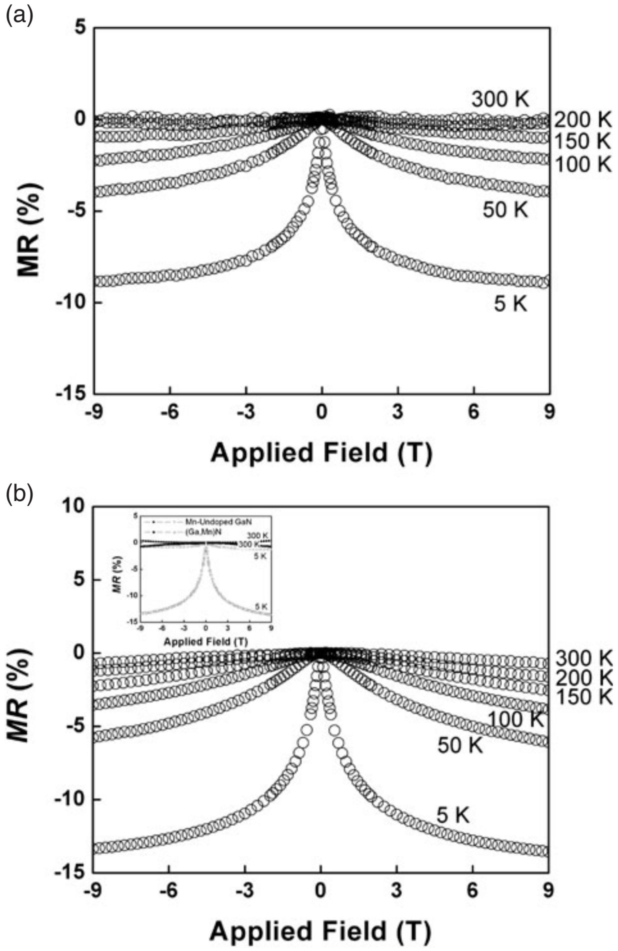

Figure 3. The variation of the magnetoresistance ($\Delta R/R$) against magnetic fields applied perpendicular to the plane of the samples for the $(Ga_{1-x}Mn_x)N$ films with (a) $x = 0.20\%$ and (b) $x = 0.92\%$ grown on undoped GaN templates in the temperature range of 5–300 K. The inset of (b) presents the MR behaviours for the non-Mn-doped GaN and $(Ga_{1-x}Mn_x)N$ films with $x = 0.92\%$ grown on undoped GaN templates.

The variations of the MR for the $(Ga_{1-x}Mn_x)N$ films with (a) $x = 0.2\%$ and (b) $x = 0.92\%$ grown on n-type GaN templates are shown in figure 4. In contrast to the aforementioned results in figure 3, here negative MR in the $(Ga_{1-x}Mn_x)N$ films with $x = 0.2\%$ and 0.92% is observed below 100 K, and the maximum values of the negative MR at 5 K reach ~3.8% and ~4.1%, respectively. Furthermore, positive MR in both films is observed in the temperature range 150–300 K. This positive MR is attributed to the ordinary MR due to the classical Lorentz force acting on the charge carriers [19]. In fact, the magnetotransport property of the (Ga, Mn)N films grown on n-type GaN templates is more important than that of the films grown on undoped GaN templates, since DMS thin films used in fabricating spintronic devices were grown on conducting or semiconducting materials. Nevertheless, the negative MR in the (Ga, Mn)N films grown on n-type GaN templates is not as large as that in the films grown on undoped GaN templates. The properties of the DMSs were appreciably affected by the carrier type and

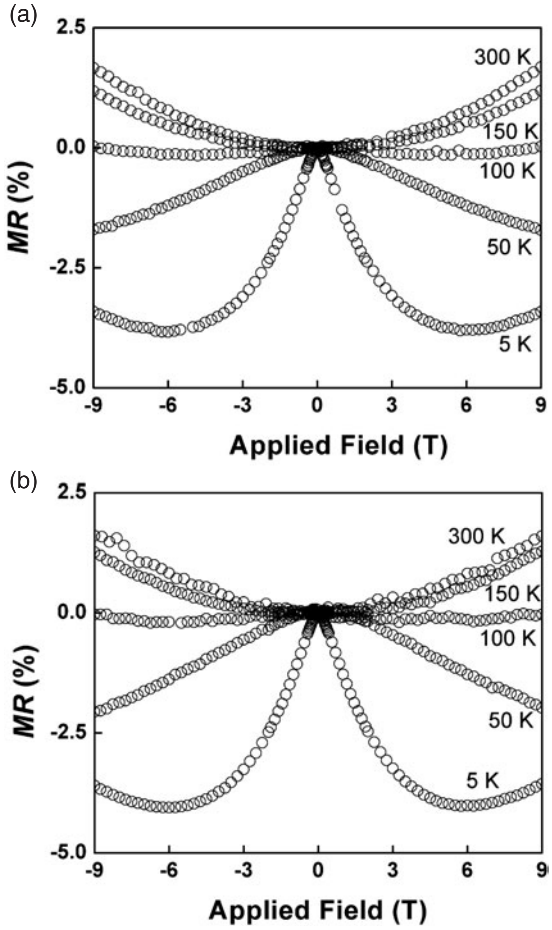

Figure 4. The variation of the magnetoresistance ($\Delta R/R$) against magnetic fields applied perpendicular to the plane of the samples for the ($Ga_{1-x}Mn_x$)N films with (a) $x = 0.20\%$ and (b) $x = 0.92\%$ grown on n-type GaN templates in the temperature range of 5–300 K.

concentration. The influence of the carriers on the magnetoresistance has been previously found in (Ga, Mn)As and the MR behaviour of (Ga, Mn)As varied with the conductivity of the (Ga, Mn)As itself [17]. The variation of the MR in semiconducting (Ga, Mn)As films was much larger than that in metallic (Ga, Mn)As films [17]. The MR behaviour of our (Ga, Mn)N varies according to the conductivity of the substrate rather than that of the (Ga, Mn)N itself. So it is thought that the Mn spins in (Ga, Mn)As interact with its own carriers, while the Mn spins in our (Ga, Mn)N interact with carriers in the n-type GaN as well as carriers in the film itself. Therefore, it is believed that the content of Mn spins in our (Ga, Mn)N films grown on n-type GaN templates is not sufficient to order all the carriers in the n-type GaN and (Ga, Mn)N film since the Mn concentration in the films is low. This leads to weakening of the ferromagnetic ordering in the (Ga, Mn)N films grown on n-type GaN templates. So far, the effect of carriers on MR in (Ga, Mn)N has been investigated by indirect methods using the variation of the conductivity in substrates, and further studies designed to enhance the properties of (Ga, Mn)N films grown on n-type templates are in progress.

4. Conclusion

In summary, we have investigated the magnetic and magnetotransport properties of (Ga, Mn)N epitaxial films grown on undoped GaN and n-type GaN templates by plasma-enhanced molecular beam epitaxy. Regardless of the kind of substrate, the (Ga, Mn)N epitaxial films were found to obviously exhibit n-type conductivity and ferromagnetic ordering with a Curie temperature above room temperature. The ferromagnetism in n-type (Ga, Mn)N is due to the RKKY interaction between the localized Mn moments mediated by the electron gas. For the (Ga, Mn)N films grown on undoped GaN templates, negative magnetoresistance was observed up to room temperature, and found to gradually increase with decreasing temperature. On the other hand, negative magnetoresistance for the (Ga, Mn)N films grown on n-type GaN templates was observed only below 100 K. Considering that the Mn concentrations in the (Ga, Mn)N films are the same, it is thought that the content of Mn spins in the (Ga, Mn)N films grown on n-type GaN templates is not sufficient to order all the carriers in n-type GaN and the film itself, since the Mn spins in (Ga, Mn)N interact with the carriers in both. Also, our results from the magnetization and MR behaviours reveal that not all the Mn spins within (Ga, Mn)N are coupled ferromagnetically—ferromagnetism and paramagnetism coexist in (Ga, Mn)N.

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