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Electrical spin injection from room-temperature ferromagnetic (Ga, Mn)N in nitride-based spin-polarized light-emitting diodes

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Abstract
We present the electrical spin injection from room-temperature ferromagnetic (Ga, Mn)N in nitride-based spin-polarized light-emitting diodes. The electroluminescence spectra from the spin LED indicate the existence of the spin polarization via optical polarization of emitted light up to room temperature. This demonstrates that the spin injection from the (Ga, Mn)N layer into (In, Ga)N quantum wells was achieved persisting up to room temperature by comparing it with the magnetic field dependence of the Hall resistance, which is proportional to the out-of-plane magnetization. These results support that (Ga, Mn)N is an appropriate material for a spin injection source in room-temperature operating semiconductor spintronic devices.

1. Introduction

In semiconductor spintronics, a rapidly growing field in the framework of condensed matter physics, the use of the quantum mechanical spin states is expected to lead to conceptual devices with fundamentally multiple functionalities, enabling quantum computation [1]. The electrical injection of spin-polarized carriers from ferromagnetic materials into semiconductors is a prerequisite for realizing such devices [2–5]. As spin injection sources, diluted magnetic semiconductors (DMSs) especially with a Curie temperature (T_C) above room temperature represent the most promising candidates due to minimizing the mismatch in conductivity between materials and being compatible with existing semiconductor devices [6]. Since Dietl *et al* predicted a T_C above room temperature for Mn-doped GaN [7], comprehensive studies have focused on (Ga, Mn)N [8, 9]. Whereas theoretical studies are based on p-type (Ga,

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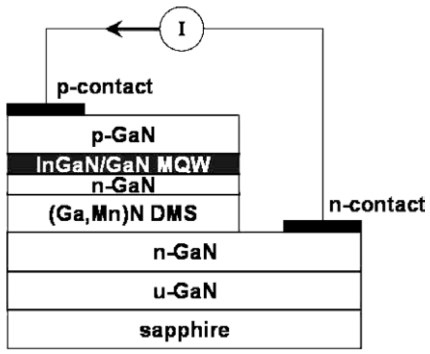

Figure 1. Schematic cross section of the spin LED heterostructure with the (Ga, Mn)N layer as a spin injection source.

Mn)N having Mn and hole concentrations of 5% and 3.5 × 10^{20} cm^{-3}, respectively [7], most experiments have demonstrated that (Ga, Mn)N is ferromagnetic at room temperature with n-type conductivity [8, 9]. Because of this contradiction between theory and experiment, ferromagnetism in n-type (Ga, Mn)N is not completely understood yet [8–10]. Spin-dependent transport phenomena such as an anomalous Hall effect provide direct evidence that the ferromagnetism in DMSs is due to an intrinsic property and not ferromagnetic precipitates [11, 12]. It is further confirmed by the injection and manipulation of spin-polarized currents in spin-based devices fabricated using DMSs [2, 11–13], which are ultimate goals in spintronics [1]. A spin-polarized light-emitting diode (LED) provides a simple and quantitative method to assess electrical spin injection efficiency by measuring the polarization state of light, whereas a spin-polarized transistor should consider the contact resistance and detects the spin injection signal along with contributions from anisotropic magnetoresistance and local Hall effect. Importantly, DMSs should exhibit ferromagnetic ordering at room temperature for realizing room-temperature operating spintronic devices. In this study, we present the electrical spin injection from room-temperature ferromagnetic (Ga, Mn)N in nitride-based spin LEDs through electroluminescence (EL) spectra, persisting up to room temperature. It is assured by comparing it with the anomalous Hall resistance proportional to the out-of-plane magnetization.

2. Experiment

The spin LED heterostructure shown in figure 1 was prepared through a combination of metallorganic chemical vapour deposition (MOCVD) and plasma-enhanced molecular beam epitaxy (PEMBE) methods on (0001) sapphire substrates. First, for the epitaxial growth, a 2 μm thick undoped semi-insulating GaN buffer layer was grown by MOCVD, followed by a 2 μm thick Si(n)-doped GaN layer. Subsequently, the samples were transferred into the PEMBE system, where a 100 nm thick (Ga, Mn)N layer as a spin injection source was grown with a Mn concentration of 1%. After this stage, the samples were returned into the MOCVD system. Then, a growth of a 100 nm thick Si(n)-doped GaN layer was performed, followed by the active layers consisting of three periods of 4 nm thick (In, Ga)N quantum wells (QWs) with In content of 10% separated by 10 nm thick GaN barriers and a 200 nm thick Mg(p)-doped GaN layer. The (Ga, Mn)N layer is sandwiched between two n-type GaN layers because the Mn concentration in the layer is not high enough to compensate the native defects, and electrons rather than holes have long spin lifetimes and high mobilities, which are favoured in

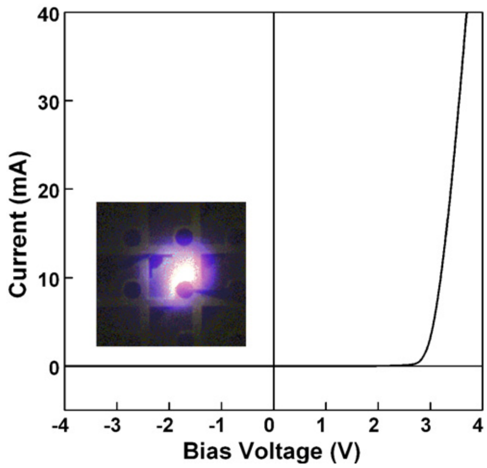

Figure 2. I–V characteristic of the spin LED at room temperature showing the rectifying diode behaviour. The inset shows its image after the turn-on.

(This figure is in colour only in the electronic version)

high-frequency and low-power device operation. Our n-type (Ga, Mn)N films were previously found to exhibit a ferromagnetic ordering above room temperature with in-plane magnetic anisotropy and have no secondary phases [9, 14]. The devices were fabricated using standard photolithography and dry etching techniques. Ni/Au and Ti/Al were used as Ohmic contacts for p-type and n-type GaN, respectively.

The electrical properties were characterized at room temperature with an HP4145B semiconductor parameter analyser. The EL measurements were performed in the Faraday geometry, i.e. with the magnetic field parallel to the light propagation direction. The circular polarization was analysed by passing the EL light through a photoelastic modulator (PEM) and a linear polarizer. The details of the experimental system have been described in [5]. For the comparison with EL results, Hall resistance measurements proportional to the out-of-plane magnetization were carried out with a quantum design physical properties measurement system (PPMS) applying a magnetic field up to 9 T.

3. Results and discussion

In order to investigate the electrical properties of the spin LEDs, the current–voltage (I–V) characteristic was measured at room temperature, and the results are shown in figure 2. The I–V characteristic of the spin LED exhibits the typical characteristic of a nonlinear and rectifying diode. It also shows a threshold voltage of ~3.1 V and a low reverse leakage current of ~10 nA. After the turn-on, the image of the device is shown in the inset of figure 2. This reveals that the device is electrically of high quality in spite of the insertion of the (Ga, Mn)N layer into the LED structure.

Figure 3(a) shows the representative EL spectrum from the spin LED taken under forward bias at 150 K. The spin LED produced intense EL emission with a peak dominated at 393 nm with the applied bias of 3.8 V. This indicates that the radiative recombination emerges from (In, Ga)N QWs in the spin LED. The EL spectra from the spin LED for applied magnetic fields of 0 and 8 T at 150 K, analysed for right (σ^+) and left (σ^-) circular polarizations, are shown in figure 3(b). At zero field, no EL polarization was observed. As the magnetic field is applied along the surface normal, electrons in the (Ga, Mn)N layer become spin polarized and are

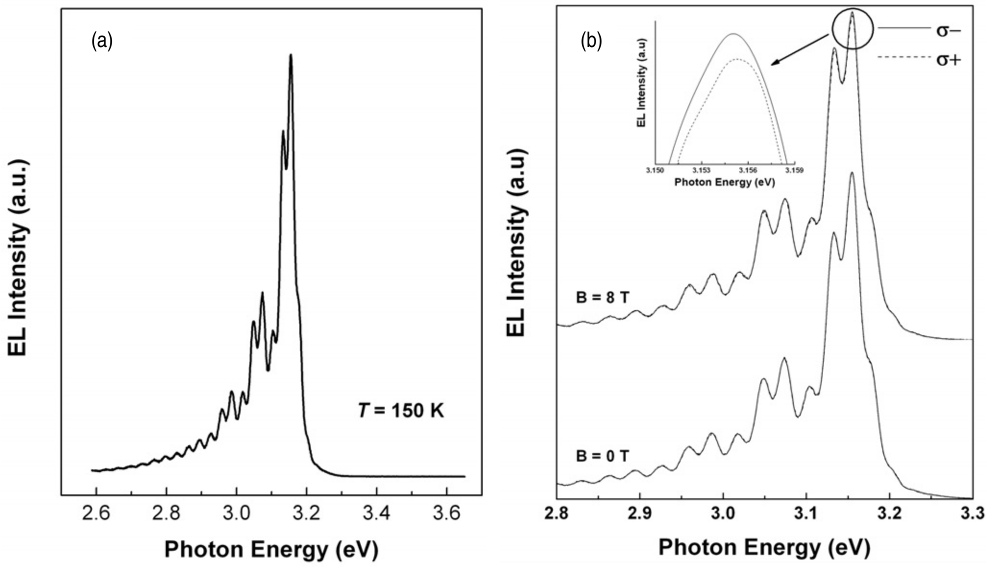

Figure 3. (a) Representative EL spectrum from the spin LED taken under forward bias at 150 K. (b) EL spectra from the spin LED for applied magnetic field of 0 and 8 T at 150 K, analysed for σ^+ (dashed line) and σ^- (solid line) circular polarizations. The peaks are normalized to align with the spectra at 0 T.

injected into the (In, Ga)N QWs. The radiative recombination of spin-polarized carriers, which obeys well known quantum mechanical selection rules, produces the emission of right or left circularly polarized light. A clear difference in intensity between the σ^+ and σ^- components is found for an applied magnetic field of 8 T, indicating the existence of spin polarization via optical polarization of light emitted from the spin LED. The EL signals from our spin LEDs were detected at low temperatures as well as room temperature in contrast to the previous work about spin LEDs with a (Ga, Mn)N layer [15], where EL measurements could not be performed below 230 K because of freeze-out of free carriers, but photoluminescence (PL) results at low temperature were obtained. In addition, these authors did not detect any optical polarization in the EL spectra at 300 K. This observation is related to the spin relaxation time in (In, Ga)N QWs, which is sensitive to the In contents. As reported by Beschoten *et al* [16], the spin coherence in GaN was found to yield spin lifetimes of ∼20 ns and ∼35 ps at 5 and 300 K, respectively, which are long enough to detect the spin polarization. Also, Julier *et al* [17] reported that the spin relaxation time became shorter with increasing In content in (In, Ga)N QWs. For nitride-based spin LEDs studied previously [15], the In content was as high as 40% in (In, Ga)N QWs, leading to a spin relaxation time, which is too fast to detect any spin polarization. However, our spin LEDs include (In, Ga)N QWs with an In content of 10%. Considering the experimental values for the spin relaxation time in [17], the spin relaxation time in our device is expected to be longer than 100 ps so that optical polarization in the EL spectra could be observed.

In order to elucidate the origin of the optical polarization of light emitted from the spin LED, EL polarization at 150 and 300 K is presented in figure 4 as a function of applied magnetic field. The EL polarization, P_{EL}, is obtained from the integrated intensity as $P_{EL} = (I^+ - I^-)/(I^+ + I^-)$, where I^+ and I^- are the integrated intensities of the most dominant peak for σ^+ and σ^- circular polarization, respectively. The EL polarization saturates around 8 T for both 150 and 300 K and reaches maximum values of approximately 1.3% and 0.8% at

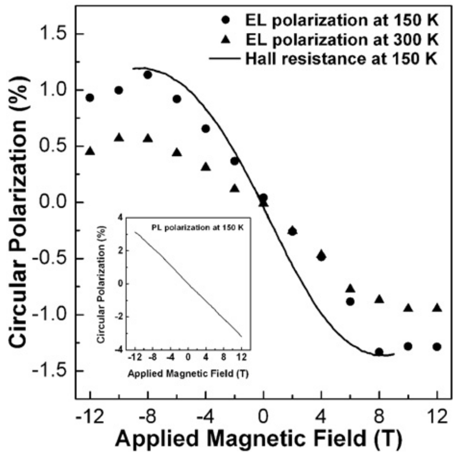

Figure 4. Applied magnetic field dependence of EL circular polarization from the spin LED measured in Faraday geometry at 150 K (circles) and 300 K (triangles). The Hall resistance (solid line) proportional to the out-of-plane magnetization of the (Ga, Mn)N film is shown as a function of the magnetic field applied perpendicular to the plane of the sample at 150 K for comparison in arbitrary units. To compare the Hall resistance with the EL polarization, the ordinary contribution of the Hall resistance has been subtracted from the data. The inset shows the PL polarization from the (In, Ga)N QWs as a function of applied magnetic field at 150 K.

150 and 300 K, respectively. The Hall resistance of the (Ga, Mn)N film grown under the same conditions obtained with the magnetic field applied perpendicular to the plane of the sample at 150 K is also shown in figure 4 for comparison as a solid line together with the EL polarization. As shown in figure 4, the anomalous Hall effect provides another proof of spin-dependent transport, confirming intrinsic ferromagnetic ordering in (Ga, Mn)N. The EL polarization and the Hall resistance ($R_{\text{Hall}} = R_0 B/d + R_s M/d$) proportional to out-of-plane magnetization [11] are found to exhibit an almost identical magnetic field dependence. This provides indisputable evidence that observed polarization is not intrinsic polarization of the (In, Ga)N QWs due to the Zeeman effect in the magnetic field [15, 18] and the spin-polarized electrons from (Ga, Mn)N participate in the radiative recombination in the QWs. In particular, the EL polarization persists up to room temperature even though its saturation value somewhat decreases with increasing temperature. In addition, the field dependence of the PL from the (In, Ga)N QWs at 150 K was measured to confirm that the observed polarization is not related to non-spin-injection effects and the result is shown in the inset of figure 4. It shows a linear characteristic without the saturation, which is different from the tendency of the EL polarization of the spin LED. It supports that the observed EL polarization is not background signals. However, the PL polarization is higher than the EL polarization of the spin LED, which might be due to population distribution between spin sublevels of the QWs [15]. After subtraction of the PL polarization from the raw data, the net spin polarization is 1.5% at 8 T. Our results reveal that spin injection from (Ga, Mn)N having intrinsic ferromagnetism into the QWs was achieved even at room temperature.

Surprisingly, the spin lifetime in GaN is three orders of magnitude larger than that in GaAs [19]. It has been predicted [20] that by the introduction of Mn in GaN a 100% spin-polarized impurity band is formed. Both properties suggest that GaN is a more suitable spin injection source. However, currently nitride-based spin LEDs have low spin injection efficiencies which might be attributed to some cancellation of the optical polarization due to the overlap of heavy-hole and light-hole transitions with opposite signs for circular polarization [18, 21] when the top of the valence band is split into two bands, the heavy-hole and light-hole states [4]. Room-temperature operating semiconductor spintronic devices could be realized by more research on nitride-based spin LEDs including systematic study about (In, Ga)N quantum wells in order to obtain high spin injection efficiencies and low spin switching fields at room temperature.

4. Conclusion

In summary, we have fabricated the nitride-based spin LED with the ferromagnetic (Ga, Mn)N layer as a spin injection source, from which EL measurements reveal that spin-polarized electrons from the (Ga, Mn)N layer into (In, Ga)N QWs were injected even at room temperature. This is supported by the magnetic field dependence of the Hall resistance in the (Ga, Mn)N layer. Our results suggest that (Ga, Mn)N possesses intrinsic ferromagnetism and is a promising candidate for spin injection. It is expected to open up opportunities towards the realization of practical semiconductor spintronic devices operable at room temperature.

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