Reduction of S-parameter by the Introduction of Nitrogen in GaNAs: Positron Annihilation and Its Comparative Study with Photoluminescence Spectroscopy

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1. Introduction

The nature of the defects in dilute nitrides has been a controversial issue.[1,2] Positron annihilation spectroscopy was employed for the analysis the vacancy-type defects in GaNAs. The dependence of S-parameter on annealing temperature have been reported, and its impact on photoluminescence characteristics was reported.[3,4] So far, the introduction of nitrogen into GaAs has been reported to increase S-parameter compared to the substrate GaAs. Even at the best case, that keeps equivalent amount, which is assumed to be the preserved material quality.[3] On the contrary, we here report that the introduction of nitrogen *reduces* the S-parameter. That is thoroughly observed for GaNAs with its nitrogen composition between 0.05 and 2.2%. The phenomena suggest a characteristic behavior of positron annihilation processes in this material system.

2. Experimental

We investigate samples grown on undoped semi-insulating GaAs(001) substrate with its etch pit density smaller than 2000 cm^{-2} . The growth was carried out by plasma-assisted solid-source molecular beam epitaxy. A 500 nm thick undoped GaNAs layer, four samples having 0.05, 0.7, 1.4 and 2.2% N, respectively, were grown at 450 °C. As₂ beam equivalent pressure was 3.0 x 10^{-5} Torr during the growth. After the growth, samples were annealed in situ under As₂ beam pressure at 600°C or 680°C for 30 min. The samples were characterized by positron annihilation spectroscopy and photoluminescence (PL) spectroscopy.

3. Result and Discussion

Figure 1 shows the results of positron annihilation spectroscopy for the samples having 0.7% and 1.4% N. Compared to the GaAs substrate, as-grown GaNAs samples show slightly smaller S-parameter. That largely decreases when annealed at 600°C, and further modest reduction is observed for the samples annealed at 680 °C. The positron lifetime of the substrate GaAs was observed to be 230 ps. That is in good agreement with the value for high-quality standard GaAs. Hence, the reduction of S-parameter by the introduction of nitrogen cannot straightforwardly be characterized as a reduction of vacancy type defects. Consequently, that is a specific behavior of positrons in GaNAs.



Figure 1 S parameter as a function of positron incident energy. The plots show the data for the GaNAs samples annealed at different temperatures, and the result for reference semi-insulating GaAs substrate. The positron energy around 7 keV mostly reflects the feature of 500 nm GaNAs bulk.

The impact of the nitrogen on the S-parameter indicates that the introduction of nitrogen forms particular regions where positrons preferably exist. However, at the region, the positrons annihilate with greater Doppler broadening. That is contrary to the situation at simple vacancy type defects, the existence of which shows the smaller Doppler broadening.



Figure 2 Plots of PL intensity as a function of N composition for the samples annealed at 600° C and 680° C. The measurement was carried out with the excitation power of 400 W/cm² at room temperature. Luminescence from as-grown samples was not observed.



Figure 3 Plots of PL peak energy as a function of N composition for the samples annealed at 600° C and 680° C. The measurement was carried out with the excitation power of 400 W/cm^2 at room temperature.

Figure 2 shows plots of PL intensity for the series of the samples after annealing. The PL was not observed for the as-grown samples. The intensity was greatly improved at the annealing temperature 680°C compared to 600 °C. This result can be the great reduction of defects by the annealing at 600 °C, or there is no correlation between S-parameter and PL intensity.[3] Figure 3 shows the plots of PL peak energy. The energy deference was almost negligible between the samples annealed at 600 °C and 680 °C for the series. Figure 4 shows temperature dependence of PL peak energy for the samples having 1.4% N. The s-shape behavior was observed even after annealing at 680°C which has obviously smaller S-parameter than GaAs.



Figure 4 Temperature dependence of the PL peak energy measured at different excitation density. Filled circles are data for the samples annealed at 680°C. Open squares are those for the samples annealed at 600°C. The colors indicate the excitation density at the measurement. The theory curve is a fit using Pässler's formula.

The samples studied have typical optical characteristics of GaNAs; showing the improvement of luminescence intensity, negligible blueshift, and s-shape behavior depending on the annealing condition. The s-shape behavior is stemmed from carrier localization within the material, which are observed for the samples having smaller S-parameters than that of high-quality GaAs.

The above results suggest that the possibility of correlation between the carrier localization and the annihilation process of the positrons, which can reduces S-parameter.

4. Summary and Conclusion

We have comparatively studied the annealing behavior of GaNAs with positron annihilation spectroscopy and photoluminescence spectroscopy. The introduction of nitrogen reduces S-parameter of the material and that was further lowered by annealing. The samples showed typical optical characteristics of GaNAs system. The results suggest that the possibility of correlation between the carrier localization and the annihilation process of positrons.

Acknowledgements

This work was partly supported by a Grant-in-Aid for Scientific Research on Priority Areas from MEXT, and a Grant-in-Aid for Scientific Research (B) from JSPS, Japan.

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