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Observation of defect complexes containing Ga vacancies in GaAsN

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Positron annihilation spectroscopy was used to study GaAsN/GaAs epilayers. GaAsN layers were found to contain Ga vacancies in defect complexes. The density of the vacancy complexes increases rapidly to the order of $10^{16}$ cm$^{-3}$ with increasing N composition and decreases after annealing at 700°C. The anticorrelation of the vacancy concentration and the integrated photoluminescence intensity suggests that the Ga vacancy complexes act as nonradiative recombination centers. © 2003 American Institute of Physics. [DOI: 10.1063/1.1533843]

Quaternary alloy $\text{Ga}_{1-x}\text{In}_{x}\text{N}_{y}\text{As}_{1-x}$ is a novel material for low-band-gap applications on gallium arsenide. The alloy is lattice matched to GaAs when $y \approx 3x$, and only a few percent of N ($x < 0.05$) is needed to decrease the band gap below 1 eV due to huge band-gap bowing. This material is used in applications such as infrared lasers, multijunction solar cells, and heterojunction bipolar transistors. Because of the large size mismatch between As and N atoms, high quality arsenide-nitride alloys are difficult to fabricate. The epitaxial growth is typically performed at the temperature range of 400 to 600°C. The effects caused by N are more evident as an increase in the low-momentum parameter $S$ and a decrease in the high-momentum parameter $W$. The measurement is sensitive in the vacancy density range approximately from $10^{16}$ to $10^{19}$ cm$^{-3}$.

The GaAsN layers were grown by MOVPE on seminsulating GaAs (001) substrates using TMGa, TBAs, and DMHy precursors and hydrogen carrier gas. A detailed description of the growth parameters can be found in Ref. 16. The samples consist of a 50-nm-thick GaAs buffer, a GaAs$_{1-x}$N$_x$ layer with a thickness of 170 nm and composition $x$ ranging from 0 to 0.05, and a 3-nm-thick GaAs cap layer. One of the samples was cut in pieces, which were annealed in the MOVPE reactor for 10 min under different annealing conditions. The GaAsN layer thicknesses and compositions were determined by high-resolution x-ray diffraction and photoluminescence (PL) at a temperature of 10 K was used to characterize the optical quality of the layers. The unintentionally doped epilayers were $p$-type with hole concentrations between $10^{16}$ and $10^{17}$ cm$^{-3}$ as determined by Hall measurements. The hole concentration decreased with increasing N composition, and the effect of annealing to the hole concentrations was negligible.

When nitrogen is introduced into the GaAsN epilayer, an increase in the $S$ parameter and a decrease in the $W$ parameter are observed at positron incident energies of 0 to 5 keV, indicating the presence of vacancy defects in the 170-nm-thick overlayer (Fig. 1). By increasing the nitrogen composition $x$ of the GaAs$_{1-x}$N$_x$ layer higher than 1%, the value of the $S$ parameter saturates. A $p$-type GaAs sample was used to obtain the reference levels $S_B = 0.5374$ and $W_B = 0.0361$ for the GaAs lattice.

The linear dependency between the $S$ and $W$ parameters in our as-grown GaAsN epilayers (Fig. 2) shows that the change in the positron annihilation signal is caused by only a single type of a vacancy. To identify this defect, we compare the slopes of the $(S, W)$ data to those expected for $V_{\text{Ga}}$ and $V_{\text{As}}$. The parameters of $S_{V,\text{Ga}}/S_B = 1.019$ and $W_{V,\text{Ga}}/W_B = 0.76$ were estimated for Ga vacancies by measuring the highly Si-doped and Si $\delta$-doped GaAs samples, where complexes of $V_{\text{Ga}}$ have been previously identified. The parameters for neutral As vacancies have been estimated to be $S_{V,\text{As}}/S_B = 1.030$ and $W_{V,\text{As}}/W_B = 0.80$. The $(S, W)$ points of GaAsN layers appear between the values of GaAs

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lattice and Ga vacancy, which allows us to identify the defects as Ga vacancies. In fact, As vacancies are not typically seen by positron annihilation spectroscopy in $p$-type and semi-insulating GaAs, because of their positive charge. 15 According to electron irradiation studies, isolated Ga vacancies are not stable in the GaAs lattice at temperatures above 300 °C.20 Thus, the vacancies in our samples grown at $>500 \degree C$ probably belong to defect complexes with some other defects or impurities. The C and H impurity concentrations in MOVPE-grown GaInNAs are typically $10^{17}-10^{18} \text{cm}^{-3}$ and $10^{18}-10^{20} \text{cm}^{-3}$, 8,21 respectively. However, the complex of $V_{Ga}$ and C impurity is unlikely due to the negative charge of both of them and vacancies decorated by hydrogen are usually not positron traps.22 Molecular-beam-epitaxy-grown GaAsN has been shown to contain interstitial nitrogen (N$_i$).12–14 However, we consider it unlikely that complexes of N$_i$ and $V_{Ga}$ could be stable, since the N interstitials could easily annihilate the Ga vacancy. The As$_{Ga}$ antisite is another typical defect arising at group-V-rich growth conditions, and it has already been identified in GaAsN.10,11 Indeed, the densities of positive As$_{Ga}$ and negative $V_{Ga}$ have been found to correlate in As-rich low-temperature-grown GaAs,23 suggesting that they form neutral complexes.24 We thus infer that the complexes involving $V_{Ga}$ constitute intrinsic defects, and their likely assignment is that formed of the acceptor $V_{Ga}$ and donor antisites such as As$_{Ga}$ or N$_{Ga}$.

The vacancy concentration of an epilayer can be estimated by applying a positron trapping model with a single type of vacancies.15 Figure 3 shows the experimental $V_{Ga}$ concentration and the integrated PL intensity at 10 K of the GaAsN samples. The $V_{Ga}$ concentration increases with N composition up to the order of $10^{18} \text{cm}^{-3}$. Interestingly, the $V_{Ga}$ concentration anticorrelates with the intensity of the photoluminescence.

A post-growth thermal annealing process is typically used to enhance the optical quality of the arsenide-nitrides. Figure 4 shows the $S$ parameter data of the GaAs$_{0.975}$N$_{0.025}$ layers annealed at different conditions. Annealing at 650 °C decreases the $S$ parameter and the sample annealed at 700 °C exhibits an even more pronounced effect. The $S$ parameter, however, remains high compared with the vacancy-free level $S_B$.

Figure 5 shows the PL efficiency and the $V_{Ga}$ concentra-
The density of the vacancies was found to increase rapidly up to the order of $10^{18}$ cm$^{-3}$ with increasing N composition, and to decrease by a factor of 5 after the thermal annealing procedure optimized for the PL intensity. The anticorrelation between the vacancy density and the integrated PL intensity suggests that the defect complex acts as a nonradiative recombination center.

In summary, we have found vacancies in MOVPE-grown GaAsN that are attributed to Ga vacancies in defect complexes. The density of the vacancies was found to increase rapidly up to the order of $10^{18}$ cm$^{-3}$ with increasing N composition, and to decrease by a factor of 5 after the thermal annealing procedure optimized for the PL intensity. The anticorrelation between the vacancy density and the integrated PL intensity suggest that the defect complex acts as a nonradiative recombination center.