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Highly conductive GaAsNSe alloys grown on GaAs and their nonalloyed ohmic properties

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Doping properties of Se in GaAsN alloys grown on GaAs (001) substrates by metalorganic-molecular-beam epitaxy were studied. Ditertiarybutylselenide (DtBSe) precursor was used as a Se source. It was found that Se was incorporated into GaAs and GaAsN layers up to a considerable concentration of $\sim 15\%$. It was also suggested that the N concentrations in GaAsNSe layers were increased by the DtBSe supply. The GaAsNSe layers were heavily doped *n* type, and the maximum electron concentration was as high as $\sim 1 \times 10^{20} \text{ cm}^{-3}$. With the increase of the carrier concentrations, the resistivity of GaAsNSe dramatically decreased to $1.2 \times 10^{-4} \Omega \text{ cm}$. This made it possible to have ohmic contacts without thermal annealing, which indicates that GaAsNSe alloys are an attractive candidate for the formation of nonalloyed ohmic contacts. © 2001 American Institute of Physics. [DOI: 10.1063/1.1418449]

Nitrogen-incorporated III–V alloys such as GaAsN have recently attracted considerable attention from both experimental and theoretical viewpoints due to their extremely large band gap bowing.^{1–7} Active studies have been performed on basic properties such as the N concentration dependence of the band gaps in GaAsN and GaInAsN alloys to realize long-wavelength laser diodes with high-temperature stability for optical-fiber communications.^{8,9} Recently, a 1.3 μm continuous-wave lasing operation of GaInAsN quantum-well lasers was demonstrated at room temperature.¹⁰ It was achieved by increasing the N concentration up to 1% in a GaInAsN quantum-well layer. However, there still remain difficulties to fabricate high-quality GaInAsN layers with large N concentrations due to the miscibility gap problem, and threshold current densities of GaInAsN semiconductor lasers still remain high.

In addition to the improvement of the crystalline quality of the GaInAsN active layers in semiconductor lasers, the improvement of ohmic contacts is another important issue for improving the laser performance. Recently, Yu *et al.* reported the large increase of the electron concentrations in heavily Se-doped GaInAsN alloys¹¹ and in the near-surface region of S and N coimplanted GaAs.¹² The maximum electron concentrations of $7 \times 10^{19} \text{ cm}^{-3}$ and $1.5 \times 10^{19} \text{ cm}^{-3}$ were observed, respectively. The large increase of the activation efficiency of Se and S atoms was explained by a band anti-crossing model. However, more detailed studies of transport and ohmic properties have not been studied.

In this letter, Se doping in GaAsN alloys was studied and the formation of quaternary GaAsNSe alloys with substantial Se concentrations was identified. It was also found that the presence of Se in the alloys significantly increased the N incorporation in GaAsNSe. The measured maximum electron concentration in GaAsNSe alloys was as high as $\sim 1 \times 10^{20} \text{ cm}^{-3}$. The resistivity of GaAsNSe dramatically decreased to $1.2 \times 10^{-4} \Omega \text{ cm}$ with the increase of the carrier

concentrations. This made it possible to realize Au/GaAsNSe ohmic contacts without thermal annealing.

GaAsNSe layers were grown on semi-insulating GaAs (001) substrates by metalorganic-molecular-beam epitaxy. The metalorganic precursors used were triethylgallium, trisdimethylaminoarsenic, monomethylhydrazine (MMHy), and ditertiarybutylselenide (DtBSe). Prior to the growth of GaAsNSe layers, $\sim 1\text{-}\mu\text{m}$ -thick GaAs buffer layers were grown on the substrates. All the epitaxial layers were grown at 600 °C. Crystallinity of the samples was analyzed by high-resolution x-ray diffraction (XRD) method. The XRD data demonstrated that these layers were indeed single-phase alloys with zinc blende structures. The cubic-phase lattice constant and the solid concentration were determined from (004), (115), and ($\bar{1}$ 15) XRD results to take into account the lattice strains induced in the layers.⁵ Optical properties of the samples were studied by absorption spectrum measurements at room temperature.⁴ Electrical properties of the samples were studied by Hall-effect measurements in the Van der Pauw geometry using In electrodes. The In/GaAsNSe contacts were usually formed by thermally alloying small pieces of In attached to the surfaces. The contact resistivity and specific contact resistance of Au/GaAsNSe contacts were measured by the transfer length method (TLM).¹³ Prior to the deposition of the Au electrode layers, the sample surfaces were chemically cleaned in a solution of HCl for 1 min and then immediately blow dried using dry N_2 to remove native oxides.

The effects of the DtBSe supply during GaAs and GaAsN growth were investigated. Figure 1 shows the DtBSe pressure (P_{DtBSe}) dependence of the lattice constant for GaAsSe and GaAsNSe layers. Although the lattice constant of GaAsSe layers was equal to that of GaAs at low DtBSe pressure ($< 1 \times 10^{-4}$ Torr), it was gradually decreased as the DtBSe pressure was increased, which suggests that Se is incorporated into GaAs as a part of the alloy constituents. When the DtBSe pressure was 7.4×10^{-4} Torr, the lattice constant of the GaAsSe alloy was 5.6203 Å. Since the lattice constant of Ga_2Se_3 in the zinc blende structure is 5.429 Å,¹⁴

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FIG. 1. Dependence of the lattice constant of GaAsSe and GaAsNSe on the pressure of the DtBSe precursor. The lattice constants of the GaAsNSe vary abruptly compared with that of the GaAsSe as the DtBSe pressure increases.

the Se concentration in this sample was estimated to be 14.7%. In comparison with the GaAsSe layers, the lattice constants of $GaAs_{1-x-y}N_xSe_y$ layers were more abruptly decreased from 5.6097 Å ($x=3.87\%$, $y=0$) to 5.4936 Å. When the N concentration in GaAsNSe layers is assumed not to depend on the DtBSe supply, the Se concentration in the GaAsNSe layers should be as high as 51.5%. This large reduction of the lattice constants in the GaAsNSe layers with the DtBSe supply will be attributed not to the single cause of the Se incorporation but to the increase of the N concentration in the layers as is evident in Fig. 1.

Figure 2(a) shows the MMHy pressure (P_{MMHy}) dependence of the lattice constant of GaAsNSe layers grown with the various DtBSe pressures. When the MMHy pressure was set to zero, the Se concentration in the GaAsSe layers grown with the DtBSe pressures of 1.6×10^{-4} and 4.0×10^{-4} Torr was 2.42% and 8.15%, respectively. The lattice constants of the samples were decreased with the increase of the MMHy pressure. The comparison of the GaAsN and GaAsNSe char-

FIG. 2. (a) MMHy pressure dependence of the lattice constant of the samples. (b) The measured band gap energy vs the lattice constant of the samples.

FIG. 3. Dependence of (a) the electron concentration and (b) resistivity of the samples on the pressure of the DtBSe precursor. The maximum electron concentration of GaAsNSe layer is $\sim 1 \times 10^{20} \text{ cm}^{-3}$.

acteristics shows that the decrease of the lattice constant was more enhanced for the higher MMHy supply by the presence of Se on the growth surfaces. When the Se concentration in GaAsNSe layers is assumed not to depend on the MMHy supply and the MMHy pressure was 3.2×10^{-3} Torr, the N concentrations in the layers were estimated to be 2.95%, 6.59%, and 8.97% for the DtBSe pressures of 0, 1.6×10^{-4} , and 4.0×10^{-4} Torr, respectively. These results indicate that the N concentration in GaAsNSe is increased by the mutual reactions between Se and N atoms on the surfaces during the growth.

The measured band gap energies of these samples are summarized in Fig. 2(b). In the case of the GaAsN alloys shown by the closed circles, the large band gap reduction was observed with the decrease of the lattice constant, i.e., with the increase of the N concentration. On the other hand, the band gap energies of the GaAsSe alloys grown without the MMHy supply, which are indicated by the dashed line in Fig. 2(b), did not show clear dependence on the lattice constant, i.e., on the Se concentration, and were almost constant at ~ 1.24 eV. The band gap energies of the GaAsNSe alloys were lowered with the decrease of the lattice constant, which will be mainly due to the increase of the N concentration as discussed on Fig. 2(a). The decrease of the energy gap of the GaAsNSe alloys was more enhanced with the higher DtBSe pressures. These results indicate that the N concentration in GaAsNSe will be increased by the mutual reactions between Se and N atoms on the surfaces during the growth. However, as is shown in Fig. 2(b), the variation of the GaAsNSe band gap is much more gradual than that of GaAsN. The details of this difference are now under study.

Although as-grown GaAsN layers showed the hole concentration of $\sim 4 \times 10^{17} \text{ cm}^{-3}$ and the mobility of $\sim 80 \text{ cm}^2/\text{Vs}$ for the N concentration of $\sim 1\%$, the GaAsNSe layers were measured to be heavily doped *n* type. Figure 3 shows the DtBSe pressure dependence of (a) the measured carrier concentration and (b) the resistivity of some of GaAsSe and GaAsNSe samples indicated in Fig. 1. The electron concentration of the GaAsNSe was dramatically increased in comparison with the GaAsSe. The maximum electron concentration of GaAsNSe was as high as $\sim 1 \times 10^{20} \text{ cm}^{-3}$.

FIG. 4. Dependence of the mobility of GaAsNSe layers measured by Hall effect on the pressure of the DtBSe precursor.

$\times 10^{20} \text{ cm}^{-3}$, which is about four times larger than that of GaAsSe without N. As the DtBSe pressure is increased, a large decrease of the resistivity in GaAsNSe layers was observed as shown in Fig. 3(b). This is because the electron mobility in GaAsNSe was increased up to $\sim 500 \text{ cm}^2/\text{V s}$ in spite of the increase of the electron concentrations with the increase of the DtBSe pressure as shown in Fig. 4. The physical origin of this peculiar property is now under study.

Yu *et al.* reported the large increase of the electron concentration in heavily Se-doped $\text{Ga}_{1-3x}\text{In}_{3x}\text{As}_{1-x}\text{N}_x$ alloys.¹¹ The maximum electron concentration of $7 \times 10^{19} \text{ cm}^{-3}$ at $x=3.3\%$ and the electron mobility of 20–60 $\text{cm}^2/\text{V s}$ were observed in their Se-doped GaInAsN alloys. In our study, the electron concentration as high as 10^{20} cm^{-3} in GaAsNSe alloys was observed. It is well known that the band gap reduction in GaAsN alloys would lower the conduction band edge.² The higher electron concentrations in the present GaAsNSe alloys will be attributed to the enhancement of the conduction band lowering by the increase of the N concentration by the presence of Se. On the other hand, the electron mobility in GaAsNSe alloys is one-order-of-magnitude higher than that of GaInAsN alloys. Based on the present study, the Se-doped GaInAsN¹¹ may form GaInAsNSe alloys and the mobility in these alloy semiconductors may be reduced by the additional alloy-disorder scatterings.

Since the studies of the electrical properties of the GaAsNSe alloys showed the very low resistivity of $\sim 10^{-4} \Omega \text{ cm}$ due to the high electron concentration and the increase of the electron mobility, a possibility of fabricating low-resistance ohmic contacts was studied. Figure 5 shows the typical current–voltage (I – V) characteristic for Au electrodes deposited on GaAsNSe surface. The linear I – V characteristic shows that the Au/GaAsNSe contact is able to form nonalloyed ohmic contacts. The inset shows the resistance measured as a function of spacing between Au electrodes. This TLM measurements show that the specific contact resistance and sheet resistance determined from a straight-line curve fit to the measurements were $4.5 \times 10^{-4} \Omega \text{ cm}^2$ and 22 Ω/square , respectively.

In conclusion, Se doping in GaAsN was studied. It was found that the doped Se forms the quaternary GaAsNSe alloys with the Se concentration up to $\sim 15\%$. The increase of

FIG. 5. Typical I – V characteristics between adjacent Au electrodes deposited on the GaAsNSe. The Au/GaAsNSe contacts show ohmic behavior without the alloying reaction between the contact metal and GaAsNSe by annealing. The inset shows the contact resistance as a function of spacing between Au electrodes.

the N concentration in the GaAsNSe by the simultaneous DtBSe supply was indicated by the enhanced reduction of the lattice constant and the band gap energy. The electron concentration in GaAsNSe was dramatically increased up to as high as 10^{20} cm^{-3} . As the electron concentration is increased, the resistivity was decreased by the peculiar increase of the electron mobilities in GaAsNSe. The Au/GaAsNSe contacts exhibited good ohmic contact behavior.

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