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Micro-photoluminescence study of nitrogen delta-doped GaAs grown by metalorganic vapor phase epitaxy

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Abstract

We have measured micro-photoluminescence (PL) spectra of nitrogen delta (δ)-doped GaAs with various concentrations. In nitrogen δ-doped GaAs with low concentrations, the micro-PL spectra measured at different positions were individually different. In contrast, almost the same spectra were obtained for nitrogen δ-doped GaAs with higher concentrations and uniformly nitrogen-doped GaAs. The micro-PL study shows that there are no more than a few isoelectronic traps within a diameter of $\sim$1 μm. This indicates a good prospect for applications of isoelectronic traps to single photon emitting devices.

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1. Introduction

III–V–N alloys have been expected to be promising materials for optoelectronic device applications because of their unique properties, such as large band gap bowing and isoelectronic traps. In dilute-impurity regions, nitrogen atoms are substituted for matrix group V atoms and act as isoelectronic traps. As one of the features of isoelectronic traps, sharp luminescence lines are observed [1–5]. The luminescence energy due to isoelectronic traps is dependent on the number of related nitrogen atoms and the distance between nitrogen atoms. The luminescence energy due to nitrogen pairs in GaAsN has been identified in several reports [3,4]. If the luminescence can be obtained from a single isoelectronic trap, it is one of the prospective candidates for single photon emitting devices, which is expected to play a key role in the field of quantum information technology, such as quantum cryptography and quantum computing. Since the size of nitrogen pairs is smaller than $\sim$1 nm [3], a single nitrogen pair can trap only one electron, or only one exciton, which is suitable for single photon emitters. In addition, compared with using a quantum dot [6], utilizing a single isoelectronic trap is advantageous to the design of multiple-layer mirrors or filters because one can obtain luminescence with specific energies from isoelectronic traps. In this paper, we have performed micro photoluminescence measurements of nitrogen delta (δ)-doped GaAs to examine the prospect for applications to single photon emitting devices. The purpose of this work is to observe a sharp photoluminescence (PL) line from a limited number of isoelectronic traps, ideally speaking, from a single isoelectronic trap. As an approach, we have adopted δ doping of low nitrogen concentrations to restrict nitrogen atoms to a limited region. In addition, we have used micro-PL method to examine whether it is possible to make a specific single nitrogen pair in a limited area with a size of $\sim$1 μm, which is of great importance to the fabrication of single photon emitting devices.

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2. Experimental procedure

Samples used in this study were nitrogen δ-doped GaAs layers grown on semi-insulating undoped GaAs (0 0 1) substrates by low-pressure metalorganic vapor phase epitaxy using trimethylgallium (TMG), tertiarybutylarsine (TBA), and dimethylhydrazine (DMHy) as the sources. The growth temperatures were 600 and 630 °C. A 200-nm-thick GaAs layer was grown as a buffer layer. To perform the nitrogen δ doping into GaAs, only TMG flow was stopped and DMHy was supplied for 5 s. The flow rate of DMHy was changed from 250 to 4000 μmol/min to obtain various nitrogen concentrations. A 40-nm-thick GaAs layer was grown as a cap layer. We measured micro-PL spectra at 4 K using a diode-pumped solid-state (DPSS) laser (532 nm) as the excitation source. The spatial resolution of the micro-PL measurement system used in this study was ~1 μm. Luminescence was detected with an intensified charge-coupled device. In order to observe sharp PL lines, we have measured PL spectra under weak excitation conditions because the density of isoelectronic traps is much less than the density of states of host GaAs. We have performed the PL measurements twice to distinguish sharp PL lines from occasional cosmic rays.

3. Results and discussion

Fig. 1 shows the micro-PL spectra for nitrogen δ-doped GaAs with various nitrogen concentrations. The number of sharp PL lines is found to decrease with decreasing nitrogen concentration. For lower nitrogen concentrations, all the sharp lines observed in PL spectra can be identified as emissions due to nitrogen isoelectronic traps [2–5]. According to previous reports, Z_1, NN_A, Y_1, Y_2, and Y_3 lines shown in Fig. 1 are due to nitrogen pairs. For higher N concentrations, some sharp lines cannot be identified as those due to nitrogen pairs, but are located near the PL lines due to nitrogen pairs. This suggests that these lines are probably due to isoelectronic traps formed by three or more nitrogen atoms. Sharp PL lines are observed at higher energies with decreasing nitrogen concentration, indicating the separations between nitrogen atoms become larger [3].

Fig. 2(a) shows micro-PL spectra at different positions on a nitrogen δ-doped GaAs with a low nitrogen concentration. The spectra are individually different. Although GaAs-bound-exciton and free-exciton peaks are seen in every spectrum, the sharp PL lines due to isoelectronic traps are not always seen. For example, the Y_4 peak seen in the second spectrum from the top disappears in the other spectra; the NN_A peak observed in the third spectrum from the top is not seen in the other spectra. In contrast, almost the same micro-PL spectra are obtained at different positions for the sample with a higher nitrogen concentration, as shown in Fig. 2(b). Furthermore, we measured micro-PL spectra for a uniformly nitrogen-doped GaAs (N~0.099%) and also observed almost the same spectra at different positions. These results clearly show that δ doping of low nitrogen concentrations is crucial for observing a sharp PL line from a limited number of isoelectronic traps.

Near-field PL study [7] reported similar results that different PL spectra were observed at different positions of a GaAsN (N~0.8%) epilayer because there exists alloy fluctuations. A key difference between this study and the near-field PL study, however, is that the size of our observed area (~1 μm) is much larger than the size observed by near-field PL (~150 nm). The minimum size for obtaining a single isoelectronic trap is crucial to the fabrication of single photon emitting devices.

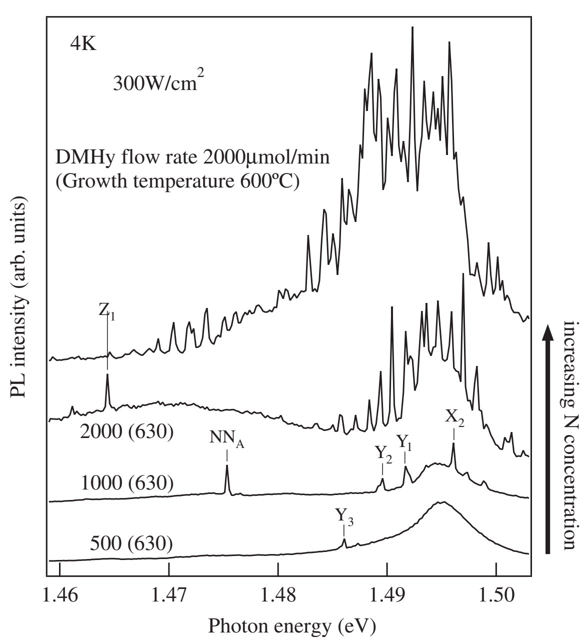

Fig. 1. Micro-PL spectra of N δ-doped GaAs with various N concentrations.

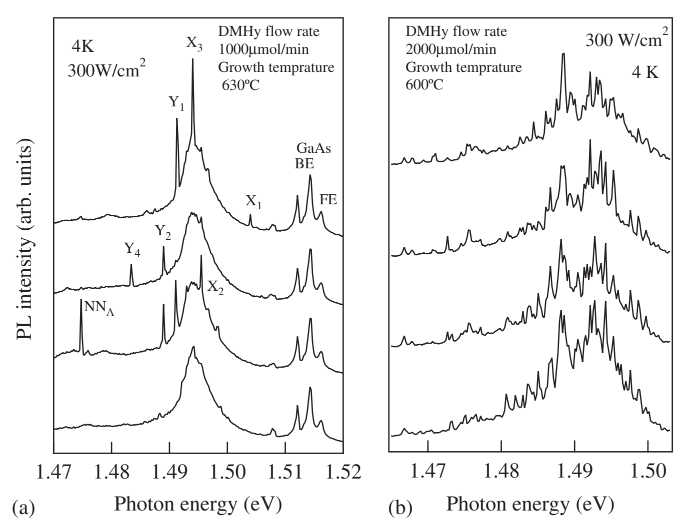

Fig. 2. Micro-PL spectra at different positions. (a) Low N concentration: DMHy flow rate 1000 μmol/min, growth temperature 630 °C and (b) high N concentration: DMHy flow rate 2000 μmol/min, growth temperature 600 °C

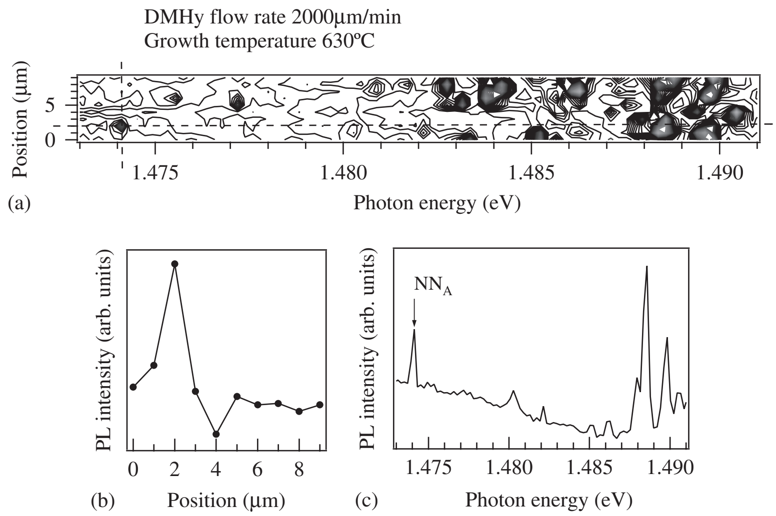

Fig. 3. (a) Contour plot of the micro-PL intensity of nitrogen δ-doped GaAs with a DMHy flow rate of 2000 µmol/min grown at 630 °C, (b) PL intensity due to NN_A as a function of position and (c) micro-PL spectrum at a position of 2 µm.

Fig. 3(a) shows a contour plot of the PL intensity of nitrogen δ-doped GaAs with a DMHy flow rate of 2000 µmol/min grown at 630 °C. These spectra were measured at positions with a separation of 1 µm in the 10-µm range. A lot of sharp PL lines are seen at higher energies. On the other hand, there are a few sharp PL lines including NN_A at lower energies, which indicates that the average separation between nitrogen atoms forming isoelectronic traps are much larger than 1.264 nm of NN_A [2]. Fig. 3(b) shows the PL intensity at 1.4741 eV designated as NN_A in Fig. 3(a) as a function of position. The maximum is located at a position of 2 µm, and the PL intensity is weak at the other positions. In addition, we show the micro-PL spectrum at a position of 2 µm in Fig. 3(c). As can be seen from this figure, a sharp PL line is observed at 1.4741 eV (NN_A). Thus, the PL results shown in Fig. 3 demonstrate that a single NN_A pair is located only at a position within a diameter of ~1 µm. There are extra emissions but NN_A as shown in Fig. 3(c), however, which are obstacles and should be eliminated in order to obtain a single photon. The use of a multiple layer band pass filter can select only a desired emission, and the design of the filter is feasible because the emission from an isoelectronic trap is well defined, which is an advantage of using an isoelectronic trap over using a quantum dot. Further experiments are necessary to investigate the quality of a single photon obtained from a single isoelectronic trap for the near future.

4. Conclusions

We measured micro-PL spectra of nitrogen δ-doped GaAs with various concentrations to examine the potential for applications of isoelectronic traps to single photon emitting devices. Sharp photoluminescence lines were observed under low excitation conditions. In nitrogen δ-doped GaAs with low concentrations, the micro-PL spectra measured at different positions were individually different. In contrast, almost the same spectra were obtained for nitrogen δ-doped GaAs with higher concentrations and uniformly nitrogen-doped GaAs. As a result, we have successfully observed sharp PL lines due to a limited number of isoelectronic traps within a diameter of 1 µm for nitrogen δ-doped GaAs with low nitrogen concentrations. This result shows good prospects for the single photon emitter utilizing a single isoelectronic trap.

Acknowledgments

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References

[1] D.G. Thomas, J.J. Hopfield, Phys. Rev. 150 (1966) 680.
[2] R. Schwabe, W. Seifert, F. Bugge, R. Bindemann, V.F. Agekyan, S.V. Pogarev, Solid State Commun. 55 (1985) 167.
[3] T. Makimoto, H. Saito, N. Kobayashi, Jpn. J. Appl. Phys. 36 (1997) 1694.
[4] T. Shima, Y. Makita, S. Kimura, T. Iida, H. Sandhu, Y. Hoshino, Nucl. Instrum. Methods Phys. Res. B 127/128 (1997) 437.
[5] Y. Zhang, A. Mascarenhas, J.F. Geisz, H.P. Xin, C.W. Tu, Phys. Rev. B 63 (2001) 085205.
[6] C. Santori, M. Pelton, G. Solomon, Y. Dale, Y. Yamamoto, Phys. Rev. Lett. 1502 (2001) 86.
[7] K. Matsuda, T. Saiki, M. Takahashi, S. Takagishi, Appl. Phys. Lett. 78 (2001) 1508.