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Photoluminescence study of isoelectronic traps in dilute GaAsN alloys

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We have studied photoluminescence spectra in detail to clarify the character of the isoelectronic traps in dilute GaAsN alloys. Several sharp lines have been observed at the lower energy side of the GaAs bandgap and are in good agreement with the nitrogen pair-related emission lines previously reported. In addition to the nitrogen pair-related lines, some other emission lines have been also observed. Compared with the energies of these emission lines and the nitrogen pair-related emission lines, it was found that the energy differences agree with the longitudinal optical phonon energy at the Γ point of GaAs, showing that the character of isoelectronic traps due to nitrogen pairs in dilute GaAsN alloys is significantly contributed from the conduction band state at the Γ point. The temperature dependence of the peak energy of luminescence due to nitrogen pairs also indicates that the character of isoelectronic traps in dilute GaAsN alloys is due to the conduction band edge state at the Γ point of GaAs. For a dilute GaAsN alloy with lower nitrogen concentration, we have observed that the intensity of an emission line increased superlinearly with excitation power.

1 Introduction III-V-N alloys have bean expected to be promising materials for novel optoelectronic device applications because of their unique properties, such as large bandgap bowing, isoelectronic traps, and so on. Owing to the large bandgap bowing, for instance, InGaAsN is expected as a material for long-wavelength laser diodes with superior characteristics used in the optical fiber communications [1]. In dilute GaPN alloys, isoelectronic traps due to nitrogen pairs [2] were extensively investigated because they increase luminescence efficiency, and thus are utilized for visible light-emitting diodes. Since sharp luminescence lines are observed from isoelectronic traps in dilute GaAsN alloys, they are prospective candidates for single photon emitters [3, 4], which are expected to play an important role in the field of quantum information technology. However, isoelectronic traps in dilute GaAsN alloys have been diversely reported by several research groups [5–7], and are still controversial. For example, Liu *et al.* [6] reported nitrogen-related levels are located above the GaAs conduction band edge and that the isolated N atom forms a resonant state in the host GaAs, which is the basis for band anti-crossing model [8]. On the other hand, Schwabe *et al.* [5] observed nitrogen-related luminescence slightly below the conduction band edge of GaAs. For higher nitrogen concentrations (~10^{18} cm^{-1}), Saito *et al.* [7] reported that emis-

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sion lines due to N pairs are located well (~30-90 meV) below the GaAs conduction band edge. In this study, we have investigated photoluminescence (PL) spectra in detail to clarify the character of the isoelectronic traps in dilute GaAsN alloys.

2 Experimental The samples used in this study were grown on GaAs (001) substrates by low-pressure metalorganic vapor phase epitaxy [9]. Trimethylgallium, arsine and 1,1-dimethylhydrazine were used as the Ga, As and N sources, respectively. The nitrogen concentration in GaAsN alloys was determined using X-ray diffraction. PL measurements were performed using a He-Ne laser (632.8nm, 3 W/cm^2-10 kW/cm^2) at temperatures from 5 K to 70 K. PL was dispersed by a monochromator and detected with an intensified charge-coupled device.

3 Results and discussion Figure 1 shows the PL spectrum of GaAs$_{1-x}$N$_x$ (x = 0.1%) at 5 K. Several sharp lines can be seen at the lower energy side of the GaAs bandgap and are in good agreement with the optical transitions reported by Saito *et al.* [7]. This unequivocally indicates that isoelectronic traps due to nitrogen are located within the bandgap of GaAs. According to the previous work by Saito *et al.* [7], the emission lines observed at 1.4776, 1.4634, 1.4598, 1.4505, and 1.4299 eV are identified as the lines of isoelectronic traps due to nitrogen pairs and are labeled NN$_A$, NN$_C$, NN$_E$, NN$_D$, and NN$_B$, respectively.

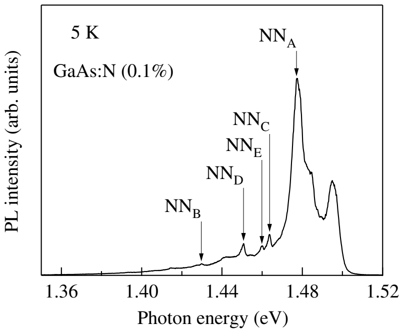

Fig. 1 PL spectrum of GaAs$_{1-x}$N$_x$ (x = 0.1%) at 5 K.

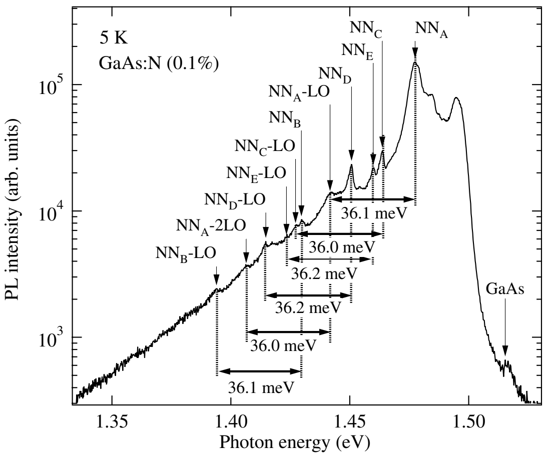

Fig. 2 Logarithmic plot of PL spectrum of GaAs$_{1-x}$N$_x$ (x = 0.1%) at 5 K.

In addition to the nitrogen pair-related lines, in fact, some other emission lines are also observed at 1.4415, 1.4274, 1.4236, 1.4143, and 1.3938 eV. Figure 2 shows a logarithmic plot of PL spectrum of $GaAs_{1-x}N_x$ (x = 0.1%) at 5 K. Compared with the energies of these emission lines and the nitrogen pair-related emission lines, it is found that the energy differences are always 36 meV. This value corresponds to the longitudinal optical (LO) phonon energy at the Γ point of GaAs [10]. Therefore, these emission lines can be identified as the LO phonon replicas of the emission from isoelectronic traps due to nitrogen pairs. This indicates that the character of isoelectronic traps due to nitrogen pairs in dilute GaAsN alloys is significantly contributed from the conduction band state at the Γ point, which is quite different from the fact that the isoelectronic traps due to nitrogen pairs in dilute GaPN alloys are mostly affected by the conduction band edge state in the vicinity of the X point of GaP [11]. It is also worth noting that PL was observed at 1.4055 eV, which is 72 meV (=36 meV × 2) lower than the NN_A emission energy. Therefore, this emission line can be identified as 2-LO phonon replicas of NN_A.

Figure 3 shows the temperature dependence of the peak energy of luminescence due to nitrogen pairs. For comparison, the temperature dependence of the E_0 gap of GaAs is also shown by solid curves. As can be seen from this figure, the peak energies of the nitrogen pair-related emission lines show almost the same temperature dependence of the E_0 gap of GaAs, which also indicates that the character of isoelectronic traps due to nitrogen pairs in dilute GaAsN alloys is mostly due to the conduction band egde state at the Γ point of GaAs. If the isoelectronic traps due to nitrogen were affected by the X-point conduction band state in GaAs, the temperature dependence of the PL peak energy for isoelectronic nitrogen traps would have been smaller than that obtained in this study. Because the energy difference between the X-point conduction and Γ-point valence band states tend to be less temperature-dependent than the direct bandgap energy E_0, as found from the case of GaP [12].

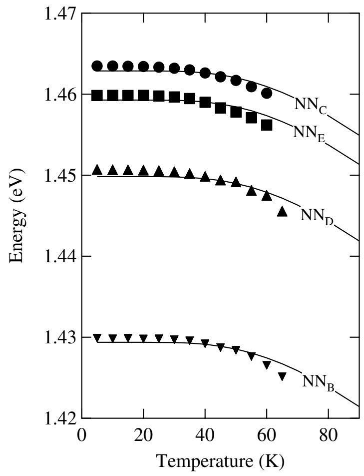

Fig. 3 Temperature dependence of the PL peak energy for isoelectronic nitrogen traps.

For a dilute GaAsN alloy with a lower nitrogen concentration, the PL spectrum is extremely different from that shown in Fig. 1. PL lines related to nitrogen pairs are not seen at all but a luminescence line is observed at 1.508 eV. This emission line can be identified as X_1 line reported by Schwabe *et al.* [5]. In Fig, 4 (a), the excitation power dependence of the PL spectrum of $GaAs_{1-x}N_x$ (x = 0.018%) at 5 K is shown. Incidentally, the PL peak seen around 1.49 eV is due to unintentionally doped C-related emission, which is composed of donor-acceptor pair (lower energy) and free-to-bound (higher energy) transitions. For lower excitation power, X_1 line is clearly observed. With increasing excitation power, the emission line labeled W_1 at the higher energy side of X_1 line becomes clear. As can be found from the spectra, the excitation power dependence of the intensity of W_1 emission line is nonlinear. Fig. 4(b) shows the relation between the intensity of W_1 and X_1 emission lines. As shown in this figure, the relation has a slope of 1.5, suggesting that W_1 emission may be due to excitonic molecules bound to isoelectronic traps reported for GaAs:N [3] andGaP:N [13]. In order to clarify the origin of W_1 emission, however, further investigation is required.

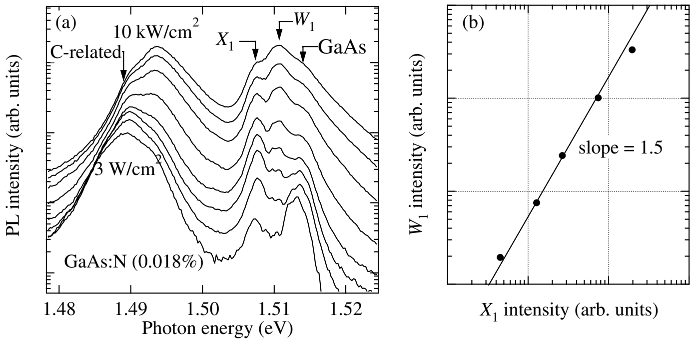

Fig. 4 (a) Excitation power dependence of the PL spectrum of $GaAs_{1-x}N_x$ (x = 0.018%) at 5 K, (b) relation between the intensity of W_1 and X_1 emission lines.

4 Conclusions We investigated PL in detail to clarify the character of the isoelectronic traps in dilute GaAsN alloys. Several sharp lines observed for $GaAs_{1-x}N_x$ (x = 0.1%) were in agreement with the nitrogen pair-related emission lines previously reported. In addition to the nitrogen pair-related lines, some other emission lines were also observed. Compared with the energies of these emission lines and the nitrogen pair-related emission lines, the energy differences were found to agree with the LO (Γ) phonon energy of GaAs, This shows that the character of isoelectronic traps in dilute GaAsN alloys is considerably contributed from the conduction band state at the Γ point. The temperature dependence of the PL peak energy also indicates that the character of isoelectronic traps is due tot the conduction band edge state at the Γ point of GaAs. For a dilute GaAsN alloy with lower nitrogen concentration, the intensity of the emission line at 1.51 eV was observed to nonlinearly increase with the excitation power

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