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Twin photoluminescence peaks from single isoelectronic traps in nitrogen δ -doped GaAs

Y. Endo^{a,*}, Y. Hijikata^a, H. Yaguchi^a, S. Yoshida^a, M. Yoshita^b, H. Akiyama^b, F. Nakajima^c, R. Katayama^c, K. Onabe^c

^aDepartment of Electrical and Electronic Systems, Saitama University, 255 Shimo-Okubo, Sakura-ku, Saitama 338-8570, Japan
 ^bInstitute for Solid State Physics, The University of Tokyo, 5-1-5 Kashiwanoha, Kashiwa, Chiba 277-8581, Japan
 ^cDepartment of Advanced Materials Science, The University of Tokyo, 5-1-5 Kashiwanoha, Kashiwa, Chiba 277-8583, Japan

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Abstract

We report on twin emission peaks observed from single isoelectronic traps formed by nitrogen pairs in nitrogen δ -doped GaAs by high-energy resolution micro-photoluminescence (PL) spectroscopy. The twin PL peaks show almost the same intensity with narrow linewidths of $20-50\,\mu\text{eV}$, which are considerably smaller than that ever reported for isoelectronic traps in GaAs:N. The higher and lower energy luminescence transitions were linearly polarized in the [1 $\bar{1}$ 0] and [1 1 0] directions, respectively, indicating that the two exciton states have completely orthogonal relations with each other. The fact that any pair of the twin PL peaks has the same polarization properties suggests that the splitting is due to the anisotropy between [1 $\bar{1}$ 0] and [1 1 0] directions in the host crystal and is possibly explained by the strain anisotropy in the sample. © 2007 Elsevier B.V. All rights reserved.

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1. Introduction

Single-photon sources are expected to play a key role in the field of quantum information science and technology, such as quantum cryptography [1,2] and quantum computing [3]. In order to realize the single-photon sources, a single molecule [4] and a single quantum dot [5,6] have been investigated. A single isoelectronic trap in semiconductors is also a promising candidate for single-photon sources because sharp emission lines are obtained from isoelectronic traps, for instance, formed by nitrogen pairs in dilute III–V–N alloys (GaAs:N [7–9] and GaP:N [10]). Thus, we have studied luminescence from single isoelectronic traps formed by nitrogen pairs in GaAs to demonstrate the potential for the single-photon sources. In our previous paper [11], we have succeeded in observing exciton emission lines from single isoelectronic traps

formed by nitrogen pairs in nitrogen δ -doped GaAs layers with low nitrogen concentrations. In this work, we have observed twin emission peaks from single isoelectronic traps by micro-photoluminescence (PL) spectroscopy with highenergy resolution. We have also studied the polarization properties of the twin PL peaks from single isoelectronic traps, and discuss the origin of the peak splitting.

2. Experimental procedure

The sample used in this study was a nitrogen δ -doped GaAs layer grown on a 350- μ m-thick semi-insulating undoped GaAs(001) substrate by low-pressure metalorganic vapor phase epitaxy. The sources were trimethylgallium (TMG), tertiarybutylarsine (TBA) and dimethylhydrazine (DMHy). The growth temperature was 630 °C. The nitrogen δ -doped layer was sandwiched between a 200-nm-thick GaAs buffer layer and a 40-nm-thick GaAs cap layer. To perform nitrogen δ -doping into GaAs, we supplied DMHy

^{*}Corresponding author. Fax: +81488583841.

E-mail address: endo@opt.ees.saitama-u.ac.jp (Y. Endo).

flow during the interruption of TMG flow for several seconds, where the nominal coverage of nitrogen atoms impinging on the surface corresponds to 0.88 monolayers (ML). Since most of nitrogen atoms desorb from the surface, however, the actual coverage is estimated to be $\sim 10^{-4}\,\mathrm{ML}$ from our preliminary experiments on uniformly N-doped GaAs.

We have measured micro-PL spectra at 4 K using a diode-pumped solid-state laser (532 nm) as the excitation source. The luminescence was dispersed by a 0.75-m monochromator and detected by a charge-coupled device. The spatial resolution and energy resolution of the micro-PL measurement system were $\sim\!1\,\mu m$ and $\sim\!30\,\mu eV$, respectively. We have also carried out the polarization measurement of the PL spectrum.

3. Results and discussion

Fig. 1 shows a PL intensity map of nitrogen δ -doped GaAs obtained by scanning the sample in one direction. As shown in this figure, several PL lines with narrow linewidths of less than 50 μ eV are observed at specific positions, which clearly indicate that the emission from a single isoelectronic trap can be successfully detected. It is also found that the emission lines at one specific position always make a pair with energy separations of around 200 μ eV. The twin PL peaks shown in Fig. 1 are located at 1.477 and 1.486 eV, which agree well with the energies reported for emission lines due to nitrogen pairs [7–9], NN_A and Y₃, respectively. Therefore, one pair of the twin PL peaks is attributed to two splitting transitions related to one single isoelectronic trap formed by a specific nitrogen pair.

Fig. 2 shows a PL spectrum obtained at the position where the intensity of the 1.477-eV twin PL peaks is maximized. The inset shows the same PL spectrum in a wider energy range. Although the spacing between the experimental data points is as small as ~30 µeV, the points are too sparse to represent such narrow PL peaks, as can be seen from this figure. Thus, we fitted the twin PL peaks by two Lorentzian functions because the emission from a single isoelectronic trap can be described by homogeneous broadening. The twin PL peaks show almost the same intensity. The full-width at half-maximum (FWHM) of the

peaks is smaller than or equal to $25 \,\mu\text{eV}$, and the energy difference ΔE between twin PL peaks is $120 \,\mu\text{eV}$. Since the estimated FWHM is less than the energy resolution ($\sim 30 \,\mu\text{eV}$) of the micro-PL measurement system, the true linewidth cannot be determined, but the linewidth of $25 \,\mu\text{eV}$ is considerably smaller than that ever reported for isoelectronic traps in GaAs:N [12,13]. As a result of searching for twin PL peaks due to single isoelectronic traps, we found that twin PL peaks are located at 1.497, 1.490, 1.486, 1.477, 1.474, 1.459, 1.450, 1.447, and 1.441 eV, which are assigned to isoelectronic traps labeled X_2 , NN_F , Y_3 , NN_A , A_3 , NN_E , NN_D , and Z_2 [7–9], respectively. The FWHM and ΔE for these twin PL peaks were found to be in the range of 20– $50 \,\mu\text{eV}$ and 120– $290 \,\mu\text{eV}$, respectively, and be independent of the PL energy.

Fig. 3(a) and (b) show polarized PL spectra around $1.477\,\text{eV}$ and the intensity of the twin PL peaks as a function of polarization angle. It is found from these figures that the higher and lower energy PL transitions are linearly polarized in the $[1\,\bar{1}\,0]$ and $[1\,1\,0]$ directions,

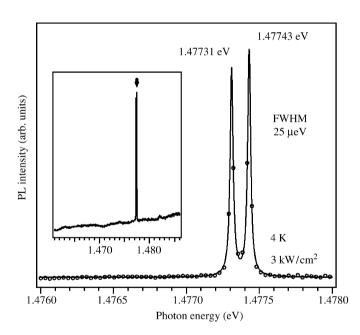


Fig. 2. PL spectrum obtained at the position where the intensity of the 1.477-eV twin PL peaks is maximized. The inset shows the same PL spectrum in a wider energy range.

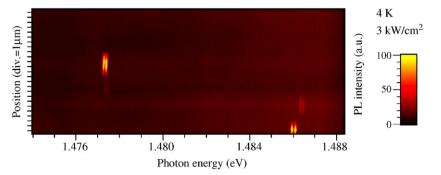


Fig. 1. PL intensity map of nitrogen δ-doped GaAs.

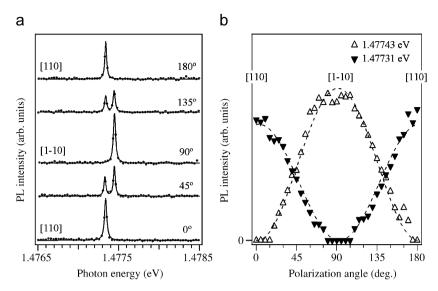


Fig. 3. Polarized PL spectra and the twin PL peak intensity as a function of polarization angle.

respectively. This indicates that the exciton states related to a single isoelectronic trap split into two states which have orthogonal relations with each other. Therefore, this splitting cannot be explained by the exchange interaction (J=1, 2) [13]. Also for the other twin PL peaks, the same polarization properties were observed, that is, the higher and lower energy transitions were linearly polarized in the [1 \(\bar{1} \) 0] and [1 \(1 \) 0] directions, respectively. The fact that all the twin PL peaks show the same polarization properties without relation to what kind of nitrogen pairs suggests that the splitting is due to not the pair arrangements [12] but anisotropy between the [1 \bar{1} 0] and [1 1 0] directions in the host crystal, which is possibly caused by strain anisotropy in the sample. If there is the strain anisotropy between the [1 $\bar{1}$ 0] and [1 1 0] directions of $\sim 10^{-5}$ for some reason, the valence bands split into two states with the [1 \(\bar{1} \) 0] and [1 \(1 \) 0] components whose energy differences is estimated to be ~100 µeV based on the shear deformation potential of GaAs [14]. In addition, the assumption that the lattice spacing in the $[1\bar{1}0]$ direction would be smaller than that in the [110] direction leads to the polarization anisotropy that the higher and lower energy PL transitions are linearly polarized in the [1 \(\bar{1} \) 0] and [1 \(1 \) 0] directions, respectively. Nevertheless, further experiments are needed to clarify the origin of the polarization properties of the twin PL peaks observed from single isoelectronic traps in δ-doped GaAs.

4. Conclusions

We have successfully observed twin PL peaks with almost the same intensity from single isoelectronic traps in nitrogen $\delta\text{-doped}$ GaAs by high-energy resolution micro-PL measurements. The linewidth of the emission lines was in the range of 20–50 μeV , which is smaller than any other reported values for the isoelectronic traps in GaAs:N. For

all the twin PL peaks, the higher and lower energy luminescence transitions were linearly polarized in the $[1\ \bar{1}\ 0]$ and $[1\ 1\ 0]$ directions, respectively. The polarization property is possibly explained by the strain anisotropy in the sample.

Acknowledgments

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