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Biexciton Luminescence from Individual Isoelectronic Traps in Nitrogen δ-Doped GaAs

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We report on the observation of biexciton luminescence from single isoelectronic traps formed by nitrogen–nitrogen pairs in nitrogen δ-doped GaAs. The biexciton luminescence intensity showed a quadratic dependence on the excitation power while the exciton luminescence intensity increased linearly with increasing excitation power. The biexciton binding energy was found to be 8 meV, which is considerably larger than that reported for single InAs quantum dots in GaAs. We have also found that both the biexciton and exciton emission lines show completely unpolarized and no fine-structure splitting. This is suitable for the application to polarization-entangled photon pairs.

Single photons and entangled photon pairs are expected to play an important role in the field of quantum information science and technology, such as quantum key distribution[1–3] and quantum computation.[4] In order to obtain single photons or entangled photon pairs, various candidates have been proposed, for instance, single molecules,[5] single nitrogen-vacancy centers in diamond,[6] single quantum dots,[7,8] and single acceptors in semiconductors.[9] Single isoelectronic traps formed by nitrogen–nitrogen (NN) pairs in GaAs[10–15] or GaP[16,17] are also promising candidates for generating single photons or entangled photon pairs. Since the emission energy is determined by the atomic configuration of NN pairs, sharp emission lines with highly reproducible photon energies are readily available, which is advantageous to the design of distributed Bragg reflectors or filters for enhancing and selecting photons with specific energies. In our previous papers,[11–14] we reported the polarization properties and level splitting of exciton luminescence from single isoelectronic traps formed by NN pairs in GaAs. The present paper reports on the observation of biexciton emission from individual isoelectronic traps in nitrogen δ-doped GaAs. We also present the polarization properties of exciton and biexciton luminescence observed from single isoelectronic traps.

The sample used in this study was a nitrogen δ-doped GaAs layer grown on a semi-insulating undoped GaAs(110) substrate by low-pressure metalorganic vapor phase epitaxy. Trimethylgallium (TMG), tertiarybutylarsine (TBA), and dimethylhydrazine (DMHy) were used as the Ga, As, and N sources, respectively. The growth temperature was 630 °C. The nitrogen δ-doped layer was sandwiched between a 300-nm-thick GaAs buffer layer and a 40-nm-thick GaAs cap layer. To perform the nitrogen δ-doping into GaAs, we supplied DMHy flow for 5 s during the interruption of only TMG flow. The nitrogen coverage is estimated to be $\sim 10^{-4}$ monolayers from the preliminary examination.[10] We measured micro-photoluminescence (PL) spectra at 4.2 K using a diode-pumped solid-state laser ($\lambda = 532$ nm) as the excitation source to observe the emission from single isoelectronic traps. The luminescence was dispersed by a 75 cm monochromator and detected by an intensified charge-coupled device. The spatial and energy resolutions of the micro-PL measurement system used in this study were ~1 μm and ~30 μeV, respectvely. We also performed polarization measurements of PL spectra.

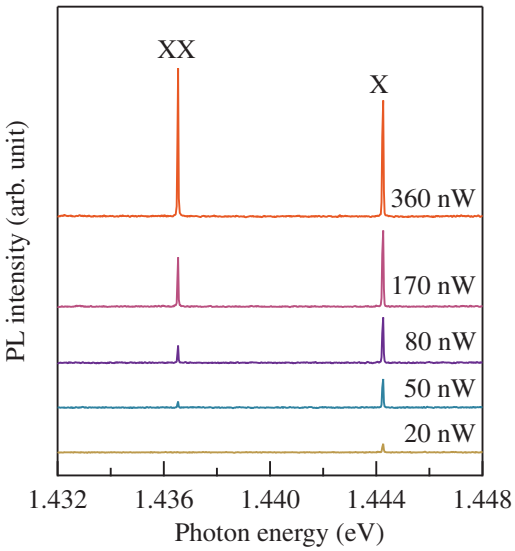

Fig. 1. Excitation-power-dependent micro-PL spectra obtained from an individual isoelectronic trap in nitrogen δ-doped GaAs.

Figure 1 shows the excitation-power-dependent micro-PL spectra obtained from an individual isoelectronic trap in nitrogen δ-doped GaAs. As shown in this figure, sharp emission lines with full width at half maximum of about 30 μeV, which may be restricted by the energy resolution of the measurement system, are seen at 1.444 eV (labeled X) and 1.436 eV (labeled XX). The PL line labeled X is assigned to the excitonic emission due to a NN pair named Z_2[18] because its photon energy is 1.444 eV. On the other hand, the emission with a photon energy of 1.436 eV has never been reported before for NN pairs in GaAs. With increasing excitation power, the lower-energy emission line labeled XX rapidly becomes stronger than the excitonic emission labeled X. It should be noted that the PL spectra are background-free even if the excitation power increases, which allow us to observe the lower-energy emission labeled XX that had never been noticed in our previous studies.[11–13]

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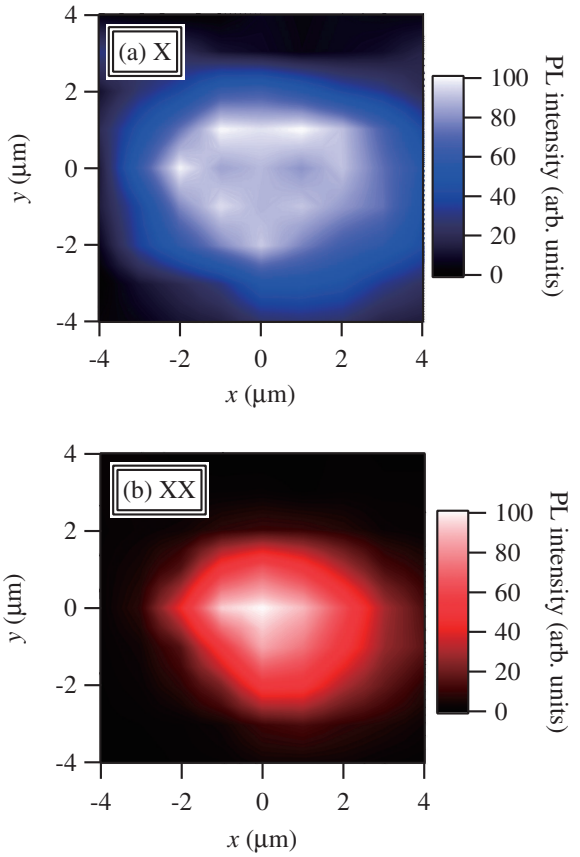

Fig. 2. Micro-PL intensity maps of the emission lines labeled X and XX.

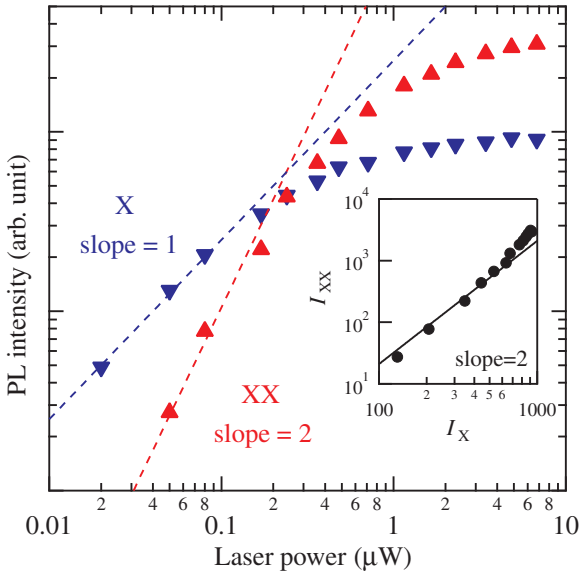

Fig. 3. PL intensity for the X and XX emission lines as a function of laser power. The PL intensity of XX is plotted as a function of that of X in the inset.

We have obtained micro-PL intensity maps of the emission lines labeled X (1.444 eV) and XX (1.436 eV) at the same position coordinate as shown in Figs. 2(a) and 2(b) by scanning the laser spot over the sample surface. In this observed area, no emission lines except 1.444 and 1.436 eV lines were found. As shown in Fig. 2(a), the excitonic luminescence labeled X is observed in a narrow region, showing that the emission from an individual isoelectronic trap can be successfully detected. The diameter of the narrow region is about 4 μm, which is believed to correspond to the diffusion length of photoexcited carriers.[19] The coincidence of the center positions between X and XX suggests that these emission lines are attributed to the same origin, i.e., a single isoelectronic trap. Compared with the excitonic emission labeled X, the emission labeled XX is observed in a narrower region. This result is consistent with the excitation power dependence of the PL intensity shown in Fig. 1 because the PL labeled XX is weaker than that labeled X when photo-excited carriers diffusing into the centers are fewer. Although we show the experimental results only for the NN pair named Z_2 in this paper, we have also observed a similar tendency for other kinds of NN pairs, such as NN_B and NN_D.[18]

Figure 3 shows the intensity of the PL labeled X and XX as a function of laser power. The intensity of the excitonic emission labeled X increases linearly with increasing laser power while the PL intensity labeled XX clearly shows a quadratic dependence on the laser power under weak excitation conditions. We also plotted the PL intensity of XX, I_{XX}, as a function of that of X, I_X, in the inset, indicating that I_{XX} is almost proportional to the square of I_X. These results show that the emission labeled XX is due to a radiative transition from a biexciton state to an exciton state.[20] When the laser power exceeds 0.2 μW, the PL intensity tends to saturate, which also indicates that the emission is observed from a single isoelectronic trap.[14–16]

As shown in Fig. 1, the energy of the biexciton emission is 8 meV lower than that of the exciton emission. Thus, the biexciton binding energy for the individual isoelectronic trap Z_2 is found to be 8 meV. The biexciton binding energy is considerably larger for single isoelectronic traps than for single InAs quantum dots in GaAs, which depends on the size or shape of quantum dots, and varies between 2 and −9 meV.[21,22] Unlike single InAs quantum dots, the biexciton binding energy for isoelectronic traps is found to be almost always constant from the observation of emission lines from several single NN pairs named Z_2. As in the case of exciton emission lines, therefore, biexciton emission lines with highly reproducible photon energies can be readily obtained from individual NN pairs in GaAs.

Figure 4(a) shows polarized PL spectra with different polarization angles obtained from an individual isoelectronic trap named Z_2. The PL spectra show a single peak both for the X and XX emission lines at any polarization angle. In addition, the peak positions do not shift with varying polarization angle. Figures 4(b) and 4(c) show the polarized PL intensity of the X and XX emission lines as a function of polarization angle. As can be found from these figures, both the polarized exciton and biexciton emission intensities are almost constant at any polarization angle. We found, therefore, that random polarization with no fine-structure splitting can be obtained for both the biexciton and exciton emission lines. This result is suitable for the application to polarization-entangled photon pairs using the biexciton cascade.

We have reported that the polarization properties of the emission from single isoelectronic traps are determined by the pairing direction of NN pairs when GaAs(110) substrates are used.[14] Thus, the unpolarized emission is obtained

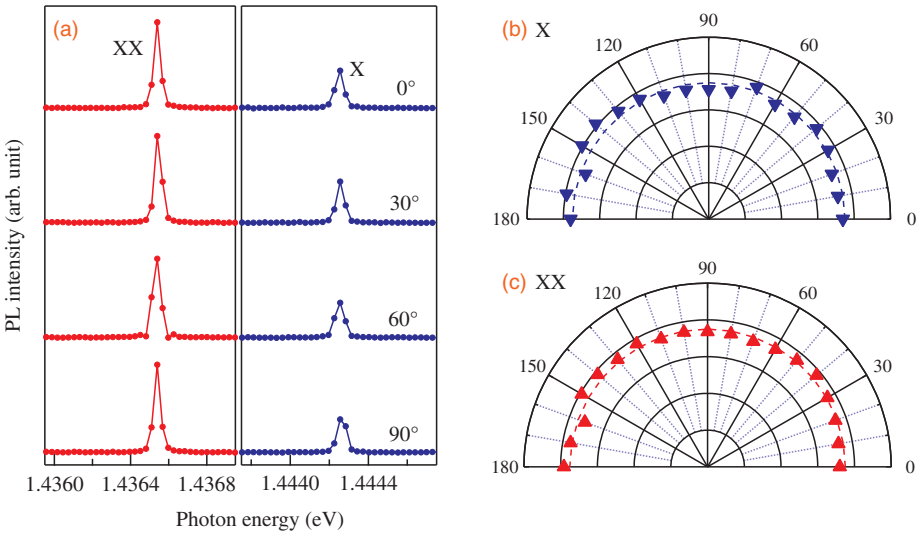

Fig. 4. (a) Polarized PL spectra observed from a single isoelectronic trap named Z_2. (b, c) Polarized PL intensity of the X and XX emission lines as a function of polarization angle.

probably because the paring direction of the observed single NN pair (1.444 eV) is perpendicular to the (110) surface, in other words, parallel to the [110] direction. Magneto-PL spectra[23] revealed that the 1.444 eV line is attributed to the first-nearest neighbor NN pair, which has the pairing direction along the [110] direction. This is consistent with our result that the pairing direction of the NN pair observed in this study is parallel to the [110] direction.

In conclusion, we have observed biexciton luminescence from single isoelectronic traps formed by NN pairs in nitrogen δ-doped GaAs grown on GaAs(110). The biexciton binding energy was found to be 8 meV and almost always constant. Thus, biexciton emission with a highly reproducible photon energy can be easily obtained from single NN pairs in GaAs as in the case of exciton emission. Both the biexciton and exciton emission lines show random polarization and no fine-structure splitting, which shows promise for the application to polarization-entangled photon pairs.

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1) C. H. Bennett and G. Brassard: Proc. IEEE Int. Conf. Computers, Systems and Signal Processing, 1984, p. 175.
2) C. H. Bennett: Phys. Rev. Lett. **68** (1992) 3121.
3) C. H. Bennett, G. Brassard, and N. D. Mermin: Phys. Rev. Lett. **68** (1992) 557.
4) E. Knill, R. Laflamme, and G. J. Milburn: Nature **409** (2001) 46.
5) B. Lounis and W. E. Moerner: Nature **407** (2000) 491.
6) C. Kurtsiefer, S. Mayer, P. Zarda, and H. Weinfurter: Phys. Rev. Lett. **85** (2000) 290.
7) C. Santori, M. Pelton, G. Solomon, Y. Dale, and Y. Yamamoto: Phys. Rev. Lett. **86** (2001) 1502.
8) R. M. Stevenson, R. J. Young, P. Atkinson, K. Cooper, D. A. Ritchie, and A. J. Shields: Nature **439** (2006) 179.
9) S. Strauf, P. Michler, M. Klude, D. Hommel, G. Bacher, and A. Forchel: Phys. Rev. Lett. **89** (2002) 177403.
10) Y. Endo, K. Tanioka, Y. Hijikata, H. Yaguchi, S. Yoshida, M. Yoshita, H. Akiyama, W. Ono, F. Nakajima, R. Katayama, and K. Onabe: J. Cryst. Growth **298** (2007) 73.
11) Y. Endo, Y. Hijikata, H. Yaguchi, S. Yoshida, M. Yoshita, H. Akiyama, F. Nakajima, R. Katayama, and K. Onabe: Physica E **40** (2008) 2110.
12) T. Fukushima, Y. Hijikata, H. Yaguchi, S. Yoshida, M. Okano, M. Yoshita, H. Akiyama, S. Kuboya, R. Katayama, and K. Onabe: Physica E **42** (2010) 2529.
13) H. Yaguchi: Proc. SPIE **7945** (2011) 79452F.
14) K. Takamiya, Y. Endo, T. Fukushima, S. Yagi, Y. Hijikata, T. Mochizuki, M. Yoshita, H. Akiyama, S. Kuboya, K. Onabe, R. Katayama, and H. Yaguchi: Mater. Sci. Forum **706–709** (2012) 2916.
15) M. Ikezawa, Y. Sakuma, L. Zhang, Y. Sone, T. Mori, T. Hamano, M. Watanabe, K. Sakoda, and Y. Masumoto: Appl. Phys. Lett. **100** (2012) 042106.
16) M. Ikezawa, Y. Sakuma, and Y. Masumoto: Jpn. J. Appl. Phys. **46** (2007) L871.
17) M. Ikezawa, Y. Sakuma, M. Watanabe, and Y. Masumoto: Phys. Status Solidi C **6** (2009) 362.
18) T. Makimoto, H. Saito, and N. Kobayashi: Jpn. J. Appl. Phys. **36** (1997) 1694.
19) D. Ritter, K. Weiser, and E. Zeldov: J. Appl. Phys. **62** (1987) 4563.
20) P. L. Gourley and J. P. Wolfe: Phys. Rev. B **20** (1979) 3319.
21) S. Rodt, R. Heitz, A. Schliwa, R. L. Sellin, F. Guffarth, and D. Bimberg: Phys. Rev. B **68** (2003) 035331.
22) R. J. Young, R. M. Stevenson, A. J. Shields, P. Atkinson, K. Cooper, D. A. Ritchie, K. M. Groom, A. I. Tartakovskii, and M. S. Skolnick: Phys. Rev. B **72** (2005) 113305.
23) Y. Harada, T. Kubo, T. Inoue, O. Kojima, and T. Kita: J. Appl. Phys. **110** (2011) 083522.