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Optical transitions in GaInNAs/GaAs multi-quantum wells with varying N content investigated by photoluminescence excitation spectroscopy

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We report on the nitrogen-concentration dependence of optical transitions between quantized states of electrons and holes in GaInNAs/GaAs multi-quantum wells. Using low-temperature photoluminescence excitation spectroscopy, systematic studies have been performed on a series of samples with nitrogen concentrations in the range 0%–1.14%. The observed data were compared with theoretical fitting based on the band anticrossing model in which the localized N states interact with the extended states in the conduction band, taking strain effects into account. Our results are consistent with the band anticrossing model, but with differing coupling strength between different quantum states of electrons in the quantum wells. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1539921]

Dilute nitride alloy III–V semiconductors have recently attracted a great deal of attention. The incorporation of a small amount nitrogen gives rise to unusual physical properties favorable for optoelectronic devices in optical fiber communications.[1,2] Various devices operating near 1.3 μm wavelength based on GaInNAs/GaAs quantum wells (QWs), including edge emitting lasers,[3–7] vertical cavity surface emitting lasers,[8] and saturable Bragg reflectors for mode locking,[9] have been extensively demonstrated. Meanwhile, the unusually large bowing coefficient due to N incorporation has drawn strong interest to the investigation of its physical origin. A theoretical model based on a band-anticrossing (BAC) interaction between the highly localized nitrogen-derived state and the zone-center conduction band Bloch states has been constructed and used to explain the band-gap behavior, including pressure and temperature dependence, for bulk dilute nitrides including GaNAs, GaInNAs, and GaNP.[10–13] However, experimental studies of the detailed electronic band structure of related quantum wells are still limited.[14–16] Such investigation is very important because most practical optoelectronic devices adopt quantum well structures. In this letter we investigate the dependence of the electronic band structure of GaInNAs/GaAs multi-quantum wells (MQWs) on nitrogen concentration by photoluminescence excitation spectroscopy (PLE). We have obtained systematic results on optical transition energies for a series of samples, all having the same In concentration ($\sim$0.27) but differing in N concentration over the range 0–0.014. In the framework of the BAC model the QW transition energies for various N concentrations have been calculated, taking into account strain effects. The theoretical results agree well with the experimental data. Our work was undertaken with the particular aim of deducing the effect of quantum confinement on the interaction strength between the localized N states and the extended states of semiconductor matrix. We found that the interaction strength in QWs is not the same for the different quantum states. This result is significant for the understanding of practical device structures.

The samples in this study were grown by gas-source molecular beam epitaxy on semi-insulating GaAs (100) substrates. The structures consisted of five GaInNAs quantum wells embedded in GaAs barriers and capped by a 60 nm layer of GaAs. The nominal thickness for the well and barrier layers is 10 and 40 nm, respectively. The arsenic flux is obtained from thermally cracked AsH_3 at 950 °C. Ultrahigh purity N_2 was injected through a radio frequency plasma source operating at a frequency of 13.56 MHz to generate active N species. The indium content in the quantum wells is 27.2% as measured on the GaInAs reference sample, while the N content varied from 0% to 1.14% as determined by high resolution x-ray diffraction analysis. For the measurement of PLE, the photoluminescence (PL) signal was dispersed by a 0.46-m-grating monochromator and detected by a thermoelectrically cooled Si/InGaAs detector using standard lock-in techniques. The tunable excitation source was provided by a 250 W tungsten halogen lamp dispersed by a 0.27-m-grating monochromator, scanned under computer control. The samples were mounted in a helium flow cryostat, and measurements were taken at 5 K.

Figure 1 shows typical PL and PLE spectra for the $Ga_{0.718}In_{0.272}N_{0.005}As_{0.995}/GaAs$ multi-quantum well sample of well width 10 nm. The PLE spectra show clear excitonic features due to the respective optical transitions between the quantized electron-, heavy-, and light-hole states. The Stokes shift between the PL peak energy and the lowest absorption peak is about 20 meV, indicating the high quality of the

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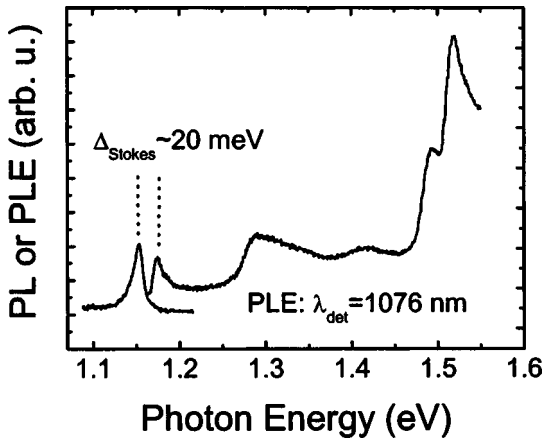

FIG. 1. Typical spectra of PL and PLE for $GaInN_xAs_{1-x}$/GaAs quantum wells with $x=0.5\%$.

samples and that the PL is due to the band edge excitonic emission.

The PLE spectra of samples with various N concentrations are shown in Fig. 2. For comparison, the PLE spectrum of an InGaAs/GaAs MQW sample of the same indium concentration is also plotted. Except for the absorption peak of GaAs at 1.519 meV, the other absorption peaks are attributed to the quantum well interband optical transitions e_1-hh_1, e_2-hh_2, and e_1-lh_1. The peak energies of these interband transitions shift to lower energy systematically and the excitonic features persist as the N concentration increases to 1.14% though the excitonic peaks broaden. The increasing Stokes shift and broadening of the absorption peaks with increase of N content is attributed to exciton localization effects induced by the local fluctuations of N distribution.[17] The transition energies of e_1-hh_1, e_2-hh_2, and e_1-lh_1 as a function of N composition are summarized in Fig. 3. It is noted in Fig. 2 that, besides the transitions attributed to the

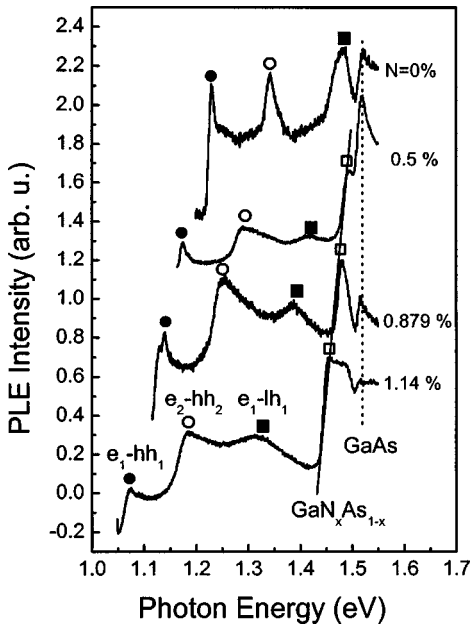

FIG. 2. PLE spectra of GaInNAs/GaAs quantum wells with various N composition. The absorption peaks marked by closed circles, open circles, and closed squares are attributed to the interband transitions e_1-hh_1, e_2-hh_2, and e_1-lh_1, respectively, while the open squares are due to the absorption of GaNAs barriers.

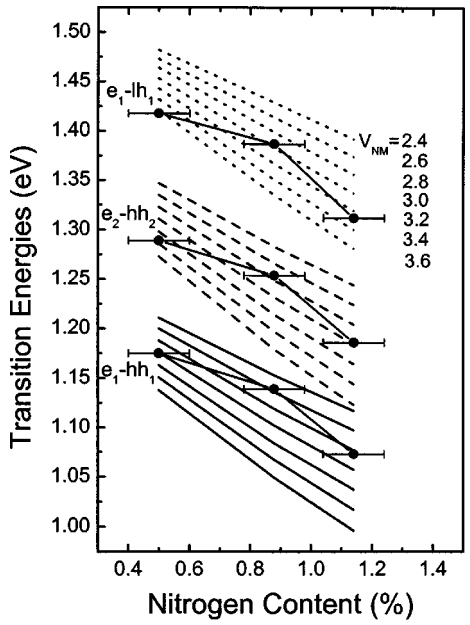

FIG. 3. Dependence of optical transition energies on N content. The curves (solid, dashed, and dotted) represent the calculated relationships of transitions e_1-hh_1, e_2-hh_2, and e_1-lh_1, respectively, with different C_{NM}. The values of C_{NM} from upper to lower are from 2.4 to 3.6 eV stepped by 0.2 eV.

GaInNAs/GaAs quantum wells, there appears a new absorption feature where the energy decreases as the N concentration increases. In view of the strong absorption characterization, we attribute this new feature to GaNAs barriers to the quantum wells due to the leakage of nitrogen when the shutter is closed. This assertion has been confirmed by the high resolution x-ray diffraction analysis.[18] The N concentration in the barriers should correlate to the N concentration in the wells, because the leakage rate of N (with shutter closed) should be dependent on the gas flow when the shutter is open which determines the N concentration in $GaInN_xAs_{1-x}$ wells. This explains qualitatively the appearance and behavior of GaNAs barrier absorption peaks.

In order to fit the transition energies, we adopt the BAC model, which was proposed by Shan *et al.*[9] and modified by O'Reilly *et al.*[19] to calculate the band structure. The conduction band of the related quantum well is calculated by solving the eigenvalues of the quantum well with finite potential depth considering the perturbation of the N energy level.[20,21] The influence of the localized N states on valence band structure is small, hence is neglected in this model.[10,20] A Luttinger–Kohn Hamiltonian is used to calculate the valence band structure of the QW. Strain effects are also included.[22,23] All the parameters are obtained by linear interpolation between the parameters of relevant binary semiconductors.[24] The band offset in $Ga_{1-x}In_xAs$/GaAs system with $x=27.2\%$ is chosen to be 0.79/0.21.[25] We consider the interaction strength C_{NM} between the localized N states and the extended states of InGaAs matrix as a fitting parameter, and calculate the optical transition energies for various C_{NM}.

We focus on the comparison of N composition-dependent transition energies. The exciton binding energy should be considered in inferring the true optical transition energies, and in our samples is assumed to be 7 meV. The

results of the full calculations with a range of coupling strengths C_{NM} are shown in Fig. 3. One can see that the calculated transition energies are very sensitive to the values of C_{NM}. For the transition e_1-hh_1, when $C_{NM}=2.8$ eV the calculated dependence of the transition energies on N concentration agrees reasonably with the experiment. This value of C_{NM} is close to the value 2.7 eV reported for bulk GaInNAs.[26] However, for the transition e_2-hh_2, the best fitting value of C_{NM} is 3.0, somewhat higher than for the e_1-hh_1 transition. This implies that the coupling strength between the localized N states and the extended matrix states differs between quantum states in the QWs.

As is well known, C_{NM} is determined by the matrix element that describes interaction of wave functions of both localized N states and the extended states of the material matrix. That is why there are different values of C_{NM} for different matrix materials.[10–12] In the case of quantum wells, due to the quantum size effect, the bulk wave function of the material are modified by different quantum states, therefore it is understandable that in different quantum states the coupling strength may differ. Our results provide direct evidence on this issue. For comparison, we have calculated the optical transition energies based on the assumption that N is fully hybridized with the semiconductor matrix. As an example, for the sample with 1.14% N, the calculated transition energies for $e1-hh1$, $e2-hh2$, and $e1-lh1$ are 1.295, 1.447, and 1.642 eV, respectively. Compared with the corresponding experimental values 1.073, 1.186, and 1.312 eV, they are poorly consistent. Therefore, our results are supportive that the BAC model gives a better description of GaInNAs/GaAs multi-quantum wells.

As we have pointed out, in the BAC model N is assumed to influence only the conduction band. If the value of C_{NM} determined for the e_1-hh_1 transition is correct, it should apply to the e_1-lh_1 transition because they are related to the same electronic states. However, as can be seen in Fig. 3, $C_{NM}=2.8$ eV gives rise to higher theoretical transition energies for e_1-lh_1 transition. This is due to the influence of the GaNAs barriers. As discussed, the barriers in our samples are GaNAs rather than GaAs, thus the modification of band gap by the strain effect may be more complicated than in the case of GaAs as barriers. In addition, the introduction of N into the barriers may also change the band alignment between the well and the barriers,[27] but our model still assumes the band alignment of GaInAs/GaAs. These factors may influence both the heavy and light hole transition energies, but it is expected that light hole will show stronger influence. In any event, the adopted model still gives a good explanation for the transitions of e_1-hh_1 and e_2-hh_2.

In summary, the electronic band structure of GaInNAs/GaAs MQWs has been investigated by PLE spectroscopy at low temperature for various N concentrations from 0% to 1.14%. The experimental spectra show clear excitonic features, and the transition energies have been compared with theoretical calculations. The valence band is obtained by solving Luttinger–Kohn Hamiltonian using a standard approach taking into account the strain effect, while the conduction band is calculated using the BAC model. The theoretical calculation agrees with the experiment quite well, indicating that BAC model is applicable to quantum well structures. By fitting the multiple optical transitions with different interaction strength between the localized N states and the extended states of semiconductor matrix, we found that the coupling strength differs between the quantum states in QWs.

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