Optical Study of Localized and Delocalized States in GaAsN/GaAs

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ABSTRACT

Taking advantages of short pulse excitation and time-resolved photoluminescence (PL), we have studied the exciton localization effect in a number of GaAsN alloys and GaAsN/GaAs quantum wells (QWs). In the PL spectra, an extra transition located at the higher energy side of the commonly reported N-related emissions is observed. By measuring PL dependence on temperature and excitation power along with PL dynamics study, the new PL peak has been identified as a transition of the band edge-related recombination in dilute GaAsN alloy and delocalized transition in QWs. Using selective excitation PL we further attribute the localized emission in QWs to the excitons localized at the GaAsN/GaAs interfaces. This interface-related exciton localization could be greatly reduced by a rapid thermal annealing.

INTRODUCTION

GaAsN semiconductor alloys grown on GaAs substrate have attracted much attention due to their unusual physical properties and potential applications in long wavelength optoelectronic and photonic devices [1-5]. Understanding the emission mechanism in these materials is very important, not only from the viewpoint of physical interest but also for the device design. In recent optical studies, the strong action of N atoms in GaAs and the large strain between GaAsN and GaAs have been shown to make the absorption and photoluminescence (PL) very complicated [6-9]. In PL measurements, N-related pair states, cluster states, localized band-tail states and delocalized states were reported in various experiments. In this work, we use photoluminescence (PL) and time-resolved PL to study exciton localization and delocalization effects in GaAsN/GaAs system.

EXPERIMENTAL DETAILS

The diluted $GaAs_{1-x}N_x$ samples investigated here were grown by gas-source MBE on semi-insulating (001) GaAs substrates using a RF nitrogen radical beam source. The epilayer thickness of the samples is nominally 400 nm. Four samples (#2658 (x~0.10%), #2846 (x~0.22%), #2847 (x~0.36%), and #2848 (x~0.62%)) were used in this study. GaAsN/GaAs quantum well (QW) samples were also grown by MBE using a DC active nitrogen plasma as the nitrogen source. The structure contains a 500 nm GaAs buffer layer, a $GaAs_{0.985}N_{0.015}$ QW layer of different thickness and a 100 nm GaAs cap layer. Detailed growth process was described elsewhere[7,10]. Rapid thermal annealing (RTA) was carried out in a flowing N_2 gas ambient on the samples covered with a protective GaAs wafer. In the PL measurements, a Ti:sapphire laser, in either cw mode or mode-locked mode, was used as the excitation source for continuous wave (cw) and pulse wave (pw) excitations. For the time-resolved PL, a Ti:sapphire mode-locked laser was used and the time-correlated signal was analyzed by a two dimensional (2D) synchroscan streak camera with an overall resolution better than 20 ps.

RESULTS AND DISCUSSION

1. Diluted $GaAs_{1-x}N_x$ Alloy (x<1%)

Fig.1 shows the PL spectra of #2658 $GaAs_{1-x}N_x$ sample (x=0.1%) under cw excitation at different temperatures. It is found that a number of N-related peaks (denoted as NN_E, NN_D, NN_B, 1.441, 1.408, 1.400, and so on) appear in all spectra. These peaks have been previously reported and attributed to different NN_x pairs or N-clusters[7,8]. Note that, at the high-energy side of the N-related transitions, an extra peak, labeled as E_b, is observed. The intensity of E_b increases with the increase of temperature, and it completely dominates the PL spectra at 110 K. Meanwhile,

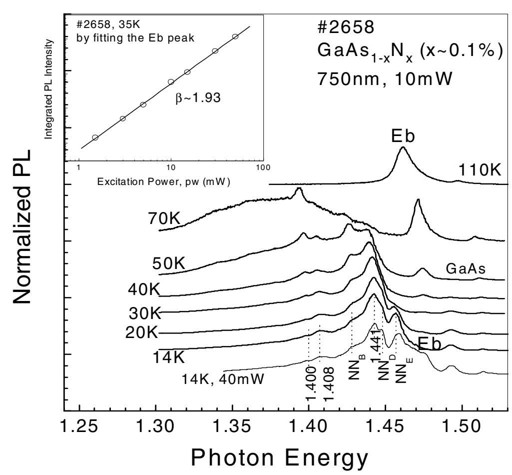

Fig.1. PL spectra of the $GaAs_{1-x}N_x$ sample (x=0.1%) under cw excitation at various temperatures. The inset shows the integrated PL intensity as a function of excitation power.

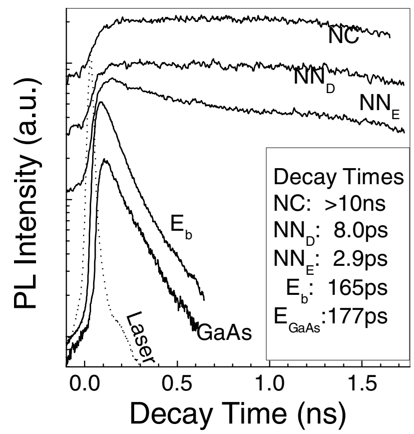

Fig.2. PL decay curves of the x=0.1% sample monitored at GaAs PL, E_b, NN_E, NN_D, and NC.

the PL intensity of E_b is found to increase superlinearly with the increase of excitation power, as shown in the inset of Fig.1. These results indicate that the E_b emission is an intrinsic band-edge emission of the GaAsN alloy[11]. Therefore, the observed superlinear increase of the PL intensity and the increase of the relative intensity of the E_b emission can be attributed to the enhanced free carrier population.

Figure 2 shows the PL decay curves monitored at different emissions for the GaN_xAs_{1-x} (x=0.1%) sample at 14K. One can clearly distinguish two types of carrier dynamics. The decay time of the E_b emission is ~165 ps, comparable to that of the GaAs band edge emission (177 ps). The observed short lifetime rules out the possibility of the associate recombination being related to either nitrogen or other impurities bound states. Whereas the PL decay times of the N bound states are much longer: 3 ns for NN_E, 8 ns for NN_D and more than 10 ns for NC. In general, excitons bound in potential minima will be *frozen up* in mobility, resulting in a reduction of the spatial coherence, and consequently have a longer lifetime[12].

2. GaAsN/GaAs Quantum Wells

Similar results are observed in GaAsN/GaAs QWs[13]. Under short pulse laser excitation, we have observed an extra high-energy PL emission in our QW samples. It dominates the PL spectra under high excitation and/or high temperature, as labeled as B in Fig.3. Fig.3(a) shows the excitation dependence of the PL at 10 K of a $GaAs_{0.985}N_{0.015}$ SQW sample. It is found that at low excitation power the PL spectra are dominated by band M, very similar to those under cw excitation[5,9,13]. With the increase of excitation power, band M shows a significant blueshift due to the saturation effect of localized states. When the excitation power further increases, band B comes into sight and becomes dominant at highest excitation power of 17 mW. It is noted that

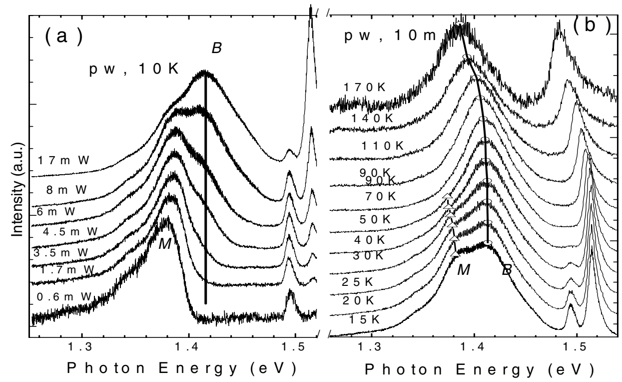

Fig.3. PL dependence on (a) excitation power and (b) temperature, under pulse excitation.

the peak energy of band B keeps almost unchanged with the excitation power, and its PL intensity increases quickly with increasing the excitation power. It is a typical behavior of delocalized states. Fig.3(b) is the temperature-dependent PL under the pw excitation. At 10K, both bands M and B are observed with comparable intensity. With the increase of temperature, however, band M is quickly quenched and becomes unnoticeable above 70 K. It also shows a fast red-shift respect to band B. All these experimental features demonstrate that the emission mechanism of band B differ significantly from that of band M, and verify that band B is an emission of delocalized excitons. The above assignment is further supported by PL decay time measurements[13].

In order to explore the origin of the exciton localization in GaAsN/GaAs QWs, we have performed the selectively excited PL measurement. Fig.4 shows the PL spectra of the sample #323 ($GaAs_{0.985}N_{0.015}$/GaAs SQW with 3nm well width) under different excitation energies (E_{ex}). It is found that when E_{ex} (1.55 eV)> E_{GaAs} (1.516eV at 12 K), the PL spectrum displays a typical line-shape of the localized exciton emission. Careful study shows that this asymmetric PL contains two parts: a low energy peak X at 1.36 eV and a high energy peak M at 1.385 eV. When E_{ex} (1.49 eV)<E_{GaAs}, however, a strong PL peak at 1.405 eV dominates the PL and the peak M at 1.385 eV almost disappears. Compared to the results in Fig.3, the above observed M and X can be attributed to N-related localized transitions, while band B is the delocalized transition in GaAsN /GaAs QWs. In Fig.4, we also display the PL of the sample under the pw excitation at 1.65 eV. As expected, both band B and band M are simultaneously observed. It is important that the selective excitation does not change the PL line shape for a bulk $GaAs_{0.99}N_{0.01}$ sample (inset of Fig.4). Therefore, it is reasonable to suggest that the observed change of the PL line-shape in the QW sample could be related to the carrier transfer process through the GaAsN/GaAs interface, as both GaAs barrier and GaAsN well are excited in the case of E_{exc}>E_{GaAs}. In other

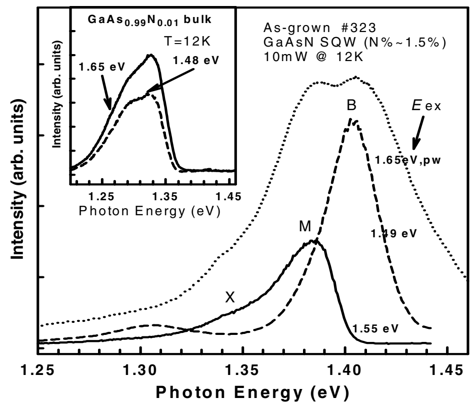

Fig.4. 12 K PL spectra of the $GaAs_{0.985}N_{0.015}$/GaAs SQW under different excitation energies. The inset shows the selectively excited PL of $GaAs_{0.99}N_{0.01}$ bulk sample.

words, band M is the interface-related localized exciton emission.

Finally, we have carried out the rapid thermal annealing on the sample #323 at 850 ^{0}C for 30 s, and compared the low-temperature PL spectra of the annealed and as-grown samples. The results are shown in Fig.5. It is found that for the annealed sample, the PL spectrum under 1.55 eV excitation is no longer dominated by the localized state M. Instead, it is dominated by the delocalized exciton emission B at 1.404 eV. This result significantly differ from that of the as-grown sample, where the PL is dominated by band M. The missing of band B in the annealed sample can be attributed to the improvement of the interface quality of GaAsN/GaAs heterointerface by reducing the localization traps.

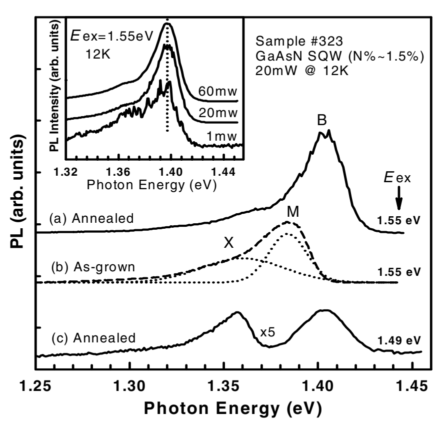

Fig.5. Comparison of the low-temperature PL spectra of the as-grown and annealed $GaAs_{0.985}N_{0.015}$/GaAs SQW under different excitation energies. The inset shows the 12 K PL spectra of the annealed sample under different excitation power.

CONCLUSIONS

We have investigated a set of $GaAs_{1-x}N_x$ samples with small nitrogen composition (x<1%) by cw and pw excitation PL, and time-resolved PL. In the PL spectra, an extra transition located at the higher-energy side of the commonly reported N-related emissions was observed. By measuring the PL dependence on temperature and excitation power, the new PL peak was identified as a transition of band-edge-related recombination in GaAsN. The PL dynamics further confirms its intrinsic nature as being associated with the band edge rather than N-related bound states. Similar results are observed in GaAsN/GaAs QWs. Under short pulse laser excitation, we have observed an extra high-energy PL emission in our QW samples and attributed it as the recombination of delocalized excitons in QWs. The selective excitation PL shows that the origin of the exciton localization in GaAsN/GaAs QWs is interface-related localized exciton emission.

ACKNOWLEDGEMENT

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