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Selectively excited photoluminescence of $GaAs_{1-x}N_x$ single quantum wells

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GaAsN bulk and GaAsN/GaAs single quantum wells grown by molecular beam epitaxy are studied by selectively excited photoluminescence (PL) measurements. A significant difference is observed in the PL spectra when the excitation energy is set below or above the band gap of GaAs for the GaAsN/GaAs quantum well samples, while the spectral features of GaAsN bulk are not sensitive to the excitation energy. The observed difference in PL of the GaAsN/GaAs quantum well samples is attributed to the exciton localization effect at the GaAsN/GaAs interfaces, which is directly correlated with the transfer and trap processes of the photogenerated carriers from GaAs into GaAsN through the heterointerfaces. This interface-related exciton localization effect can be greatly reduced by a rapid thermal annealing process, making the PL be dominated by the intrinsic delocalized transition in GaAsN/GaAs. © *2003 American Institute of Physics.*

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I. INTRODUCTION

Ga(In)NAs 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.[1–8] Understanding the origin of photoluminescence (PL) in GaAsN bulk and quantum wells (QWs) is very important, not only from the viewpoint of physical interest but also for the device design. Generally, low-temperature PL spectra of GaAsN alloys in the near band-gap spectral region are dominated by a rather asymmetric PL band with a sharp high-energy cutoff and an exponential low-energy tail. This asymmetric PL line shape is usually correlated with strong exciton localization by potential fluctuations, i.e., localized exciton (LE) recombination.[9–12] The sharp high-energy cutoff corresponds to the mobility edge, separating the localized and delocalized states. In our previous work,[12] we used short laser pulse to excite the sample and observed a PL peak from GaAsN/GaAs single quantum wells (SQWs) at the high energy side of the LE exciton emission. The observed PL peak was attributed to the recombination of delocalized excitons in QWs. In this article, we further explore the PL properties of GaAsN SQWs by selectively excited PL measurement. A significant difference is observed in the PL spectra when the excitation energy is set below or above the band gap of GaAs. The observed difference was ascribed to the exciton localization effect at the GaAsN/GaAs interface. This interface-related exciton localization effect can be greatly reduced by a rapid thermal annealing process.

II. EXPERIMENT

The samples under investigation were grown by molecular beam epitaxy (MBE) on undoped GaAs (100) substrates using dc active nitrogen plasma as the nitrogen source. A 500 nm GaAs buffer layer was grown at first, followed by a strained GaAsN quantum well layer of different thickness and a 100 nm GaAs cap layer. The growth temperature was around 500 °C. The detailed growth procedure was described elsewhere.[13] In PL measurements, a tunable Ti:sapphire laser, either in continuous wave (cw) mode or in mode-locked mode, was used as an excitation source. The detection system consists of an HR250 monochromator and an InGaAs photomultiplier tube or cooled Ge detector. Rapid thermal annealing (RTA) was carried out in a flowing N_2 gas ambient on the samples covered with a protective GaAs wafer in a homemade RTP-300 rapid thermal processor.

III. RESULTS AND DISCUSSION

Figure 1 shows the PL spectra at 12 K of the as-grown sample No. 323 ($GaAs_{0.985}N_{0.015}$/GaAs SQW with 3 nm well width) under different excitation energies (E_{ex}). When E_{ex} (1.55 eV) is higher than the band gap of GaAs (E_{GaAs}, 1.516 eV at 12 K), the PL spectrum exhibits a typical asymmetric line shape of the localized exciton emission.[9–12] As seen in Fig. 1, this asymmetric PL contains approximately two parts: a low energy peak at 1.36 eV (labeled as X) and a high energy peak at 1.385 eV (labeled as M). When E_{ex} $(1.49\ \mathrm{eV}) < E_{GaAs}$, however, a strong PL peak at 1.405 eV (labeled as B in Fig. 1) dominates the spectrum and the PL peak M at 1.385 eV almost disappears. Peak B is 20 meV higher than M. Compared to our previous results,[12] the above observed M and X can be identified as N-related localized transitions, while the band B is consistent with the delocalized transition in GaAsN/GaAs QWs. In Fig. 1, we

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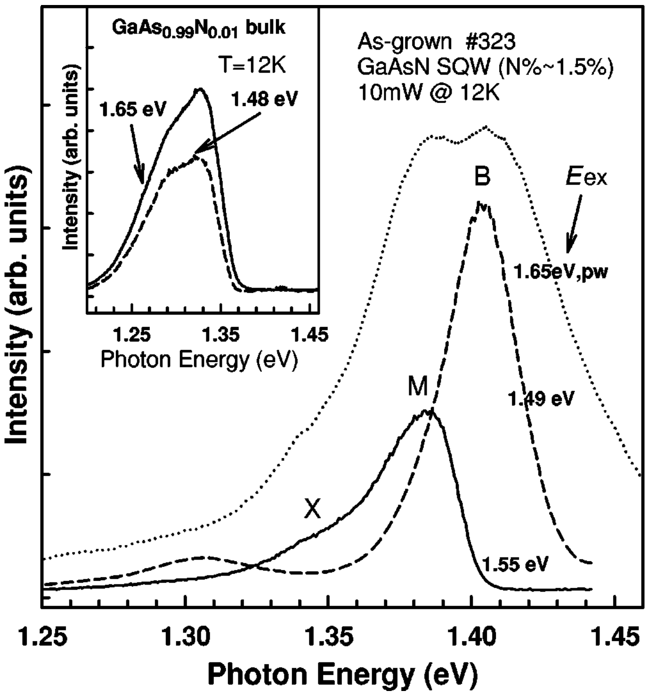

FIG. 1. 12 K PL spectra of the as-grown $GaAs_{0.985}N_{0.015}/GaAs$ SQW sample under different excitation energies. The excitation energy is indicated near the respective PL spectrum. The dotted curve is the PL spectrum of the sample under the short pulse excitation at 1.65 eV. The inset shows the selectively excited PL of $GaAs_{0.99}N_{0.01}$ bulk.

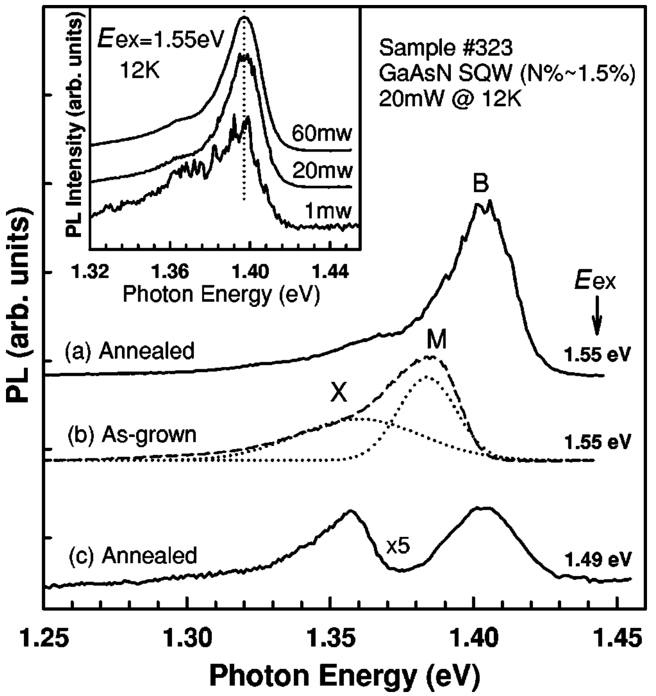

FIG. 3. Comparison of the low-temperature PL spectra of the as-grown and annealed $GaAs_{0.985}N_{0.015}/GaAs$ SQW under different excitation energies. The inset shows the 12 K PL spectra of the annealed sample under different excitation power.

also display the PL spectrum of the sample under the short pulse excitation at 1.65 eV. As expected, bands *B* and *M* are simultaneously observed. For comparison, the selectively excited PL was also performed on a bulk $GaAs_{0.99}N_{0.01}$ sample, and the results are shown in the inset of Fig. 1. It can be seen that the selective excitation does not change the PL line shape when the excitation energy varies from 1.65 eV ($>E_{GaAs}$) to 1.48 eV ($<E_{GaAs}$). This is in strong contrast to that observed in GaAsN/GaAs QWs. The above difference results from the change of the sample excitation. Figure 2 shows a schematic band diagram of our selective excitation. When the excitation energy is lower than the band gap of GaAs, only the GaAsN layer is excited [Fig. 2(a)]. The photoexcited carriers relax fast and contribute to the recombination of the delocalized exciton states (*B*) and bound states (*X*) in the GaAsN layer. In this case, the carrier excitation, relaxation, and recombination occur in the GaAsN layer only. However, when the excitation energy is higher than the band gap of GaAs [Fig. 2(b)], both the GaAs barrier and GaAsN well are excited. A large part of the photogenerated carriers in GaAs will transfer into the GaAsN layer through their interfaces and contribute to the localized exciton emission *M*, while a small part of the carriers recombine in the GaAs layer. This suggestion has been evidenced by the relative intensities between the GaAs-related PL and the PL of GaAsN.[12] As the most important difference between the two excitations is the involvement of the carrier transfer process from GaAs into GaAsN through their heterointerfaces, it is reasonable to suggest that the GaAsN/GaAs heterointerface plays a very important role in the optical properties of GaAsN QWs, and the physical origin of the band *M* is correlated with the localization effect at the interfaces. In other words, the band *M* is the interface-related localized exciton emission.

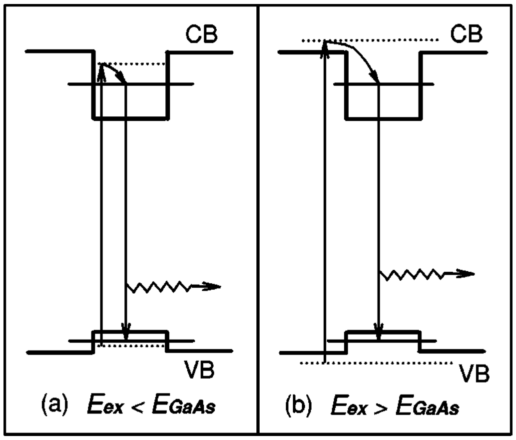

FIG. 2. Sketch band diagram of the selective excitation: (a) $E_{ex}<E_{GaAs}$ and (b) $E_{ex}>E_{GaAs}$.

In order to understand more about the properties of the transitions of *X*, *M*, and *B*, RTA was carried out on sample No. 323 at 850 °C for 30 s. Figure 3 compares the low-temperature PL spectra of the annealed and as-grown samples. It is found that for the annealed sample, the spectrum under 1.55 eV excitation is no longer dominated by the localized exciton emission *M* [Fig. 3(a)]. Instead, it is dominated by the delocalized exciton emission *B* at 1.404 eV. This result presents a striking contrast to the PL of the as-grown sample [Fig. 3(b)], where the PL is dominated by band *M*. Disappearance of the localized emission *M* in the annealed sample can be due to improvement of the interface

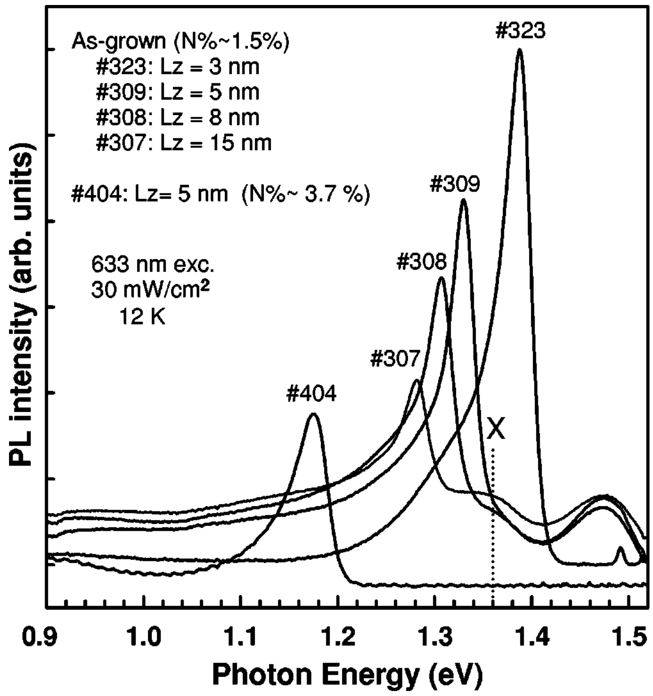

FIG. 4. 12 K PL spectra of the as-grown GaAsN/GaAs quantum wells with different well widths and N compositions under cw excitation at 633 nm. The PL peaks around 1.49 eV are related to the GaAs. The dashed line shows the position of band *X*.

quality of GaAsN/GaAs heterointerface by reducing the localization traps at the interfaces. It agrees with our previous hypothesis that the physical origin of band *M* is correlated with the localization effect at the interfaces. This interface-related exciton localization effect can be greatly reduced by a RTA process, causing the PL to be dominated by the intrinsic delocalized transition in GaAsN/GaAs. It is noted that the low energy peak *X* still exists in the annealed sample, especially when $E_{\mathrm{ex}}=1.49$ eV [Fig. 3(c)]. This is because the *X* emission originates from the "intrinsic" N-related localized states in the GaAsN layer itself, as discussed later.

We have also done PL experiments on the annealed sample under different temperatures and excitation intensities. It is found that the PL peak at 1.404 eV does not shift with increasing excitation intensity, as shown in the inset of Fig. 3 and its temperature variation follows closely the band gap of GaAs (not shown). All these results are in good agreement with the assignment of the delocalized transition of *B*.

With clear identification of *M* and *B*, we now turn to the emission peak *X*. In our previous work,[12] *X* and *M* have been attributed to the localized states, and the low energy tail *X* has much longer lifetime than the delocalized transition *B*. In order to further explore the origin of *X*, PL measurements are performed for a set of GaAsN SQW samples with different well widths and N compositions. The results are shown in Fig. 4. It can be seen that the PL peak energies shift to the low-energy with increasing well width, resulting from the quantum confinement effect in QWs. Meanwhile, a high-energy shoulder around 1.36 eV is observed for sample Nos. 307, 308, and 309, as denoted by a dashed line in Fig. 4. This energy position is consistent with the *X* emission observed in Figs. 1 and 3. In other words, all these four samples exhibit *X* emission and the peak position of the *X* emission is not affected by the quantum confinement effect. Further analysis shows that both N-related states in GaAsN and impurity states in GaAs could result in this nonshifted PL peak. If this peak comes from the GaAs layer, it should appear in all GaAsN/GaAs samples with similar growth conditions. However, it is not the case in our $\mathrm{GaAs}_{0.963}\mathrm{N}_{0.037}/\mathrm{GaAs}$ sample (No. 404). We did not observe any PL signal around 1.36 eV as shown in Fig. 4. Therefore, it is highly possible that the observed nonshifted PL peak *X* originates from the N-related localized states in GaAsN. It is well known that the PL peaks originated from NN pairs and/or N clusters in GaAsN have their unchanged energy positions,[10,14–16] as their PL energies are not determined by the N concentrations but rather by the distance between N atoms in a pair or clusters. Two dimensional quantum confinement effect has little effect on this kind of emission.

IV. CONCLUSIONS

In conclusion, we have investigated the photoluminescence of the GaAsN/GaAs SQWs by selective excitation measurements. Three kinds of optical emissions are observed in our samples. They are the intrinsic delocalized exciton emission *B*, N-related localized exciton emission *X*, and GaAsN/GaAs interface-related localized exciton emission *M*. The interface-related exciton localization effect can be greatly reduced by a rapid thermal annealing, causing the PL to be dominated by the intrinsic delocalized transition in GaAsN/GaAs. Our results can be very useful in fully understanding the mechanism of light emission in GaAsN/GaAs.

ACKNOWLEDGMENTS

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