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Photoluminescence Spectroscopy Studies of InGaAsN Quantum Wells*

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Abstract: The photoluminescence (PL) spectroscopy in InGaAsN/InGaAs/GaAs quantum wells (QWs) with low-nitrogen composition has been measured in the temperature range 13-300K. The peak position energies of InGaAsN as measured by PL exhibit anomalous inverted S-shape-like temperature dependence. Fitting with Varshni empirical relation to PL data, we have found that carriers in InGaAsN QWs are localized at low temperatures. Moreover, Time-resolved photoluminescence (TRPL) measurements, performed for various temperatures and different PL energy position, further proved that the broad PL emission of InGaAsN at low temperature is mainly dominated by strong localization, which arises from nitrogen-induced fluctuating potential in quantum wells.

Key words: InGaAsN; Photoluminescence; Quantum wells

InGaAsN 量子阱的光致发光谱研究

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摘　要：我们测量了低 N 组分的 InGaAsN/InGaAs/GaAs 量子阱材料的光致发光(PL)谱，测量温度范围从 13K 到 300K。实验结果显示，InGaAsN 的 PL 谱的主峰值的能量位置随温度的变化呈现出反常的 S 型温度依赖关系。用 Varshni 经验公式对实验数据进行拟合之后，发现在低温下 InGaAsN 量子阱中的载流子是处于局域态的。此外，我们还测量了样品在不同的温度、不同的能量位置的瞬态谱，结果进一步证实了：在低温下，InGaAsN 的 PL 谱谱峰主要是局域态激子的复合发光占据主导地位，而且 InGaAsN 中的载流子局域态主要是由 N 等电子缺陷造成的涨落势引起的。

关键词：InGaAsN；光致发光；量子阱
中图法分类号：O472.3　　　文献标识码：A

Introduction

Recently, the quaternary material - InGaAsN has attracted a great deal of attention for both theoretic and experimental investigations due to its large bowing coefficients and the potential applications for long wavelength optoelectronic devices based on GaAs[1]. Most attractive is the realization of 1.3μm vertical cavity surface emitting lasers (VCSELs) on GaAs using the well-established AlAs/GaAs distributed Bragg reflector (DBR) techniques[1-2]. This is thanks to the incorporation of a small amount of nitrogen

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in the InGaAs. In fact, a dramatic redshift of the alloy band-gap is commonly observed, about 120meV percent of N[3]. The band-anticrossing (BAC) model in which localized N states interact with the conduction band has well explained the physical origin of such band-gap shrinkage[4-5]. For optical communications, it is important to obtain photoluminescence (PL) wavelength equal or longer than 1.3μm, but achieving PL emission at longer wavelength is considerably more difficult and requires a systematic investigation.

Therefore, the knowledge of influence of nitrogen on optical properties and recombination mechanisms in InGaAsN/GaAs quantum wells is very important due to its potential applications. At the same time, the PL and TRPL measurements carried out in the broad temperature can provide useful information about the recombination mechanisms and the character of the electronic states. In this paper, we report high-resolution x-ray diffraction (HRXRD), the temperature-dependent and time-resolved photoluminescence (PL) measurement of several InGaAsN/InGaAs/GaAs quantum wells (QWs) samples with different indium and nitrogen content.

Experimental

The investigated $In_xGa_{1-x}As_{1-y}N_y/In_xGa_{1-x}As/GaAs$ QWs were grown on semi-insulating GaAs (100) substrate by molecular beam epitaxy (MBE) using a plasma rf nitrogen source. The four samples discussed here were grown at approximately 500 ℃ and consist of a 6.5nm thick InGaAsN layer and a 200nm thick InGaAs layer. These two layers have the same In concentration, where the InGaAs layer was used as a reference. The indium and nitrogen contents of the samples are: 28# (x = 0.1, y=0.009), 29# (x=0.107, y=0.011), 30# (x=0.135, y=0.015), and 01# (x=0.1, y=0.009). The high-resolution x-ray diffraction (HRXRD) and room temperature PL were used to determine the layer thickness, the indium, and nitrogen contents by comparing measured, calculated, and simulated rocking curves. Photoluminescence measurements were performed at temperature range between 13 and 300K by using a variable-temperature close-cycle cryostat under the excitation of 638nm line from a pulse semiconductor laser, whose average excitation power was about 5mW, working at 20MHz. PL and TRPL were carried out in the FL920 fluorescence lifetime spectrometer, in which spectra were dispersed by a M300 monochromator, and then were detected using a liquid-nitrogen cooled Hamamatsu R5509 near infrared (NIR) PMT.

Results and discussions

We have investigated HRXRD and PL for all the four samples, and have used simulations of measured x-ray rocking curves and PL peaks from reference InGaAs layer to determine In concentrations and layer thickness. We show in Figure 1 the experimental and simulated HRXRD patterns of sample 28#. The simulated results of In concentration and the thickness of InGaAs layer are also shown in the figure. The N concentrations in InGaAsN QWs were calculated according to the shift from the PL peak of InGaAsN to that of InGaAs, assuming a band gap reduction of 120meV percent of N.

Figure 2 shows PL spectra of sample 01# at different temperatures and under an average excitation power of 5mW. First, we observed that compared to InGaAs peak, the PL intensity of InGaAsN QWs increases with increasing temperature[6], indicating that InGaAsN QWs has a larger carrier confinement. Second, we could find that at temperatures below 60K, the InGaAsN spectra show a characteristic asymmetric line shape. In fact, the InGaAsN spectra mainly consist of a symmetric high-energy structure and a low-energy band tail, labeled by '**A**' and '**B**', respectively. It is suggestive that two carrier recombination mechanisms are responsible for the PL emission of InGaAsN QWs at low temperatures. It is noted that peak ' **A**' shifts well following with a regular thermalisation of the carriers. So, we believed that peak ' **A**' originated from the recombination of free excitons in InGaAsN QWs. However, peak ' **B**' exhibits inverted S-shape-like temperature dependence (a red-blue-red-shift of emission energy with increasing temperature). The low-energy tail as peak ' **B**' is present in other InGaAsN QWs and epilayers and in a large number of semiconductor alloys[6, 8, 9]. It is shown that the low-energy tail dominates the spectra at temperatures below 60K. But it decreases rapidly with increasing temperature and diminishes finally for temperatures above 60K. Therefore, apparently peak ' **B**' appears so-called inverted S-shape-like temperature dependence. This anomalous behaviour was observed in all samples containing nitrogen, and also is supposed to occur only together with strong localization, which arises from nitrogen-induced fluctuating potential in quantum wells[6, 8, 9].

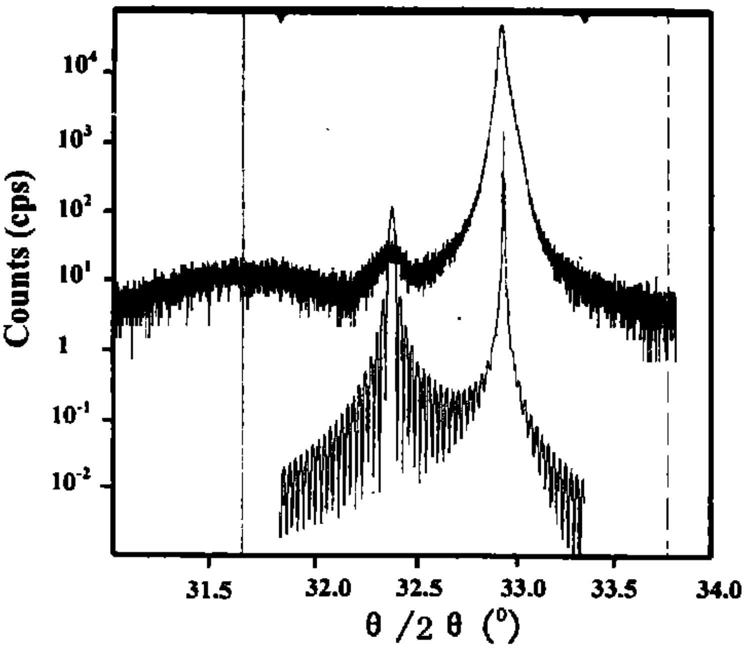

Fig. 1 HRXRD rocking curves taken from sample 28 #. The dark and light solid lines are the experiment and the simulation data, respectively.

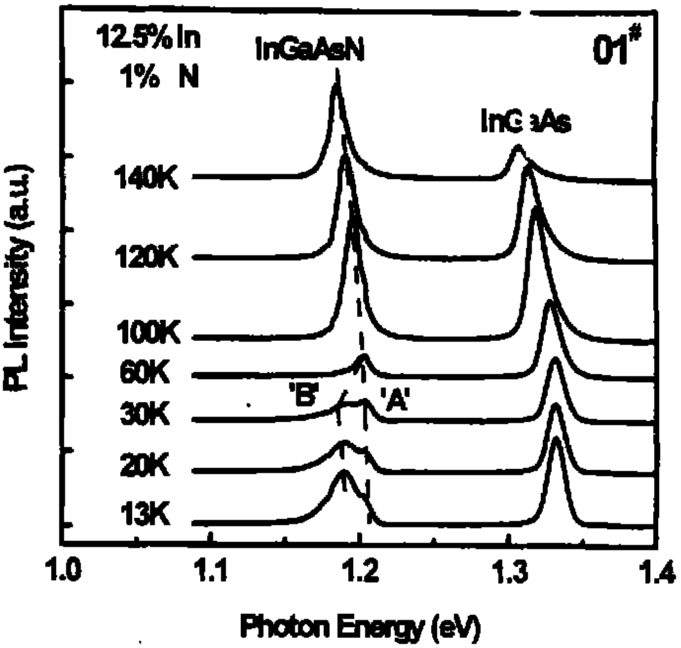

Fig. 2 the PL spectra of InGaAs and InGaAsN recorded at different temperatures and under average excitation power of 5 mW for sample 01 #.

The presence of localized excitons in InGaAsN can be inferred also from the temperature dependence of the PL spectra shown in Figure 3. The continuous lines in the same figure are the best fits to the PL data with Varshni empirical relation[7]:

$$E_g(T) = E_0 - \alpha \times T^2/(T+\beta), \quad (1)$$

where E_g is the energy gap, E_0 is its value at 0 K, and α and β are the adjustable parameters. The values of E_0, α and β obtained from the fitting procedure are given in Table 1. First, we found that a decrease of the thermal redshift of PL peak energy (value of α parameter decreases) with increasing N content, as proposed for GaNAs[8] and other InGaAsN[6, 9] based samples. Second, the evolution with temperature of the PL peak energy for the InGaAs is in agreement with regular thermalisation of the carriers, while for structures with N at temperatures below about 60K, the PL peak energies of InGaAsN are a little lower than the Varshni fit, indicating that the carriers are localized. At temperatures above about 60K, carriers are fully delocalized. The carrier localization energy is defined as[10]:

$$E_{loc}(T) = E(T) - E_{PL}(T), \quad (2)$$

where $E(T)$ is defined by Varshni model. The localization energies at 13K for all samples are also given in Table 1. Finally, we observed that both N and In content have an influence on carrier localization in the investigated InGaAsN QWs. Increasing N content increases density of defects, which lead to increase the number of carriers localized at potential fluctuations. On the contrary, Increasing In content would decrease the band-tail part and improve the luminescence.

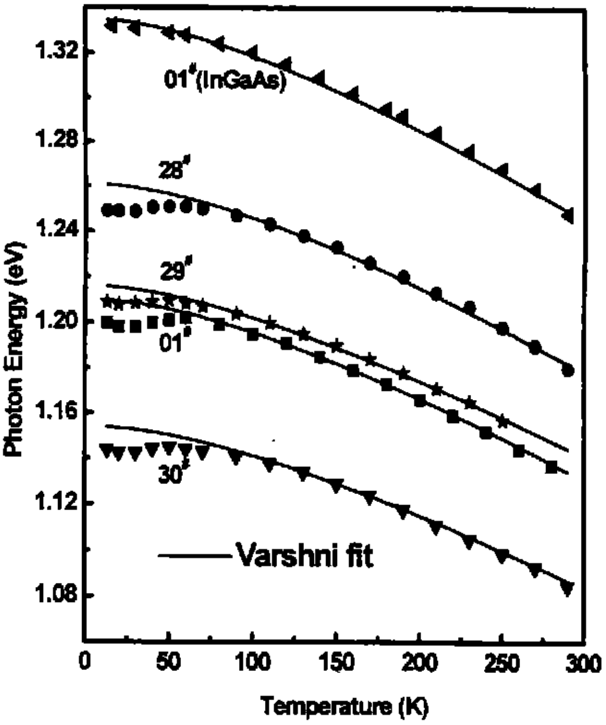

Fig. 3 the temperature dependence of the transition energy for all samples under study as measured by PL. The continuous lines are the best fits of Varshini empirical relation to the PL data.

Table 1　Value of the parameters obtained from a fit of Varshni equation

Sample	%In	%N	$E(0)$ (eV)	α (10^{-4}eVK^{-1})	β (K)	E_{loc}(13K) (meV)
01#	12.5	0.0	1.335	5.0	205	0
	12.5	1.0	1.210	4.5	210	10
28#	10.0	0.9	1.263	4.6	200	10
29#	10.7	1.1	1.216	4.2	210	9
30#	13.5	1.5	1.154	3.9	210	11

Time-resolved photoluminescence was used to obtain information about the dynamics of interband radiative recombination. Figure 4 depicts the decay time as a function of emission energy for sample 30# at a temperature of 13K. Also shown, is a representative PL spectrum at 13K. All of the measured decays were predominantly single exponentials, $I(t) = I_0 \exp(-t/\tau)$. The decay time (τ) is strongly energy dependent, particularly at low T. Generally, the decay times decrease with increasing energy, and vary over the range 0.33-0.58ns. The emission energy dependence of the PL decay is characreristic of a distribution of localized excitions[11]. Decay data in Figure 4 are fitted with the function:

$$\tau(E) = \tau_R / \{1 + \exp[(E - E_m)/E_0]\} \quad (3)$$

where τ_R is the radiative lifetime, E_m is the energy for which the radiative lifetime equals the lateral transfer time and E_0 is a characteristic energy for the density of states. From the least squares of equation (3) to the data, we get values for τ_R, E_m and E_0 are 580ps, 1.155eV and 14meV, respectively.

The temperature dependence of the decay times for sample $30^{\#}$ is shown in Figure 5, in which a similar variation for sample $29^{\#}$ is also shown as a comparison. At lower thermal energies carriers are trapped in localized states with longer lifetimes. When the temperature is increased carriers gain enough thermal energy to populate the free exciton states in the QWs with a shorter lifetimes. The lifetime and PL integrated intensity decrease with temperature as might be expected for thermal activation to nonradiative recombination processes. The decay times decrease at temperatures from 13K to 130K, and increase again to room temperature. Taking into account the decrease of the integrated intensity of PL with increasing temperature, this shows that at room temperature the non-radiative processes in the InGaAsN QWs are reduced, which is important for laser application[12]. On the other hand, for temperatures from 13K to 50K, the decay times of sample $30^{\#}$ exhibits a little larger relative decrease with temperature than that of sample $29^{\#}$. This can be explained with a higher defect concentration in sample $30^{\#}$, since increasing nitrogen concentration, thus, leads to a stronger influence on defect-related non-radiative processes. This observation agrees with our discussion above. Thus, we propose that the dominant mechanism for the low-temperature PL in the investigated InGaAsN QWs is the localized excitons recombination.

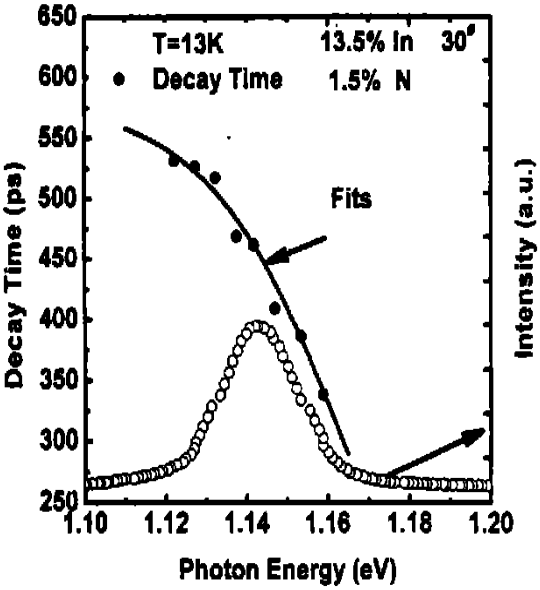

Fig. 4 The emission energy dependence of the decay time for InGaAsN QWs in sample $30^{\#}$ recorded at 13K. And a PL spectrum of InGaAsN at 13K is also shown.

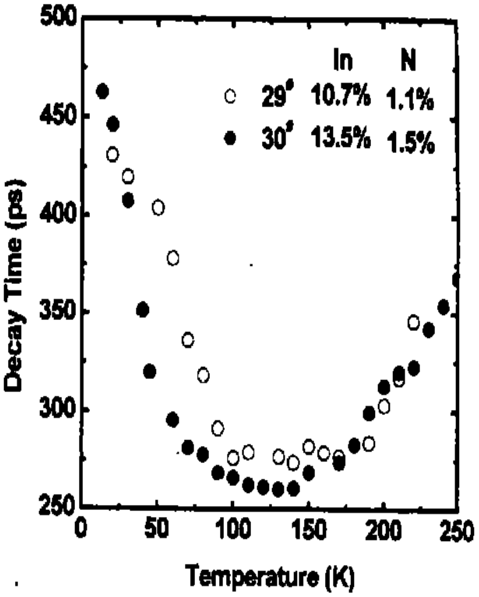

Fig. 5 The temperature dependence of the decay times for InGaAsN QWs in sample $29^{\#}$ (solid circles) and sample $30^{\#}$ (empty circles).

Conclusions

In summary, temperature-dependent and time-resolved photoluminescence experiments were employed to study the structural and optical properties of InGaAsN QWs. These include: (i) a characteristic asymmetric PL line shape of InGaAsN; (ii) inverted S-shape-like temperature dependence of PL peak

'**B**' in InGaAsN; (iii) decrease of the thermal redshift of the PL peak energy with increasing N content; (iv) strong dependence of the decay time of PL on energy at low temperatures; (v) a larger relative decrease with temperature of the decay time in samples with higher nitrogen content at low temperatures. Based on these observations we have suggested that the dominant PL mechanism in all investigated InGaAsN structures at low temperatures is the recombination of localized excitons, which is believed to arise from N-induced fluctuating potential in InGaAsN QWs, and InGaAsN incorporating larger levels of N will exhibit stronger localization effects and shorter recombination lifetimes, possibly degrading the performance of minority carrier devices and lasers.

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