Enhancing redshift phenomenon in time-resolved photoluminescence spectra of AlGaN epilayer*

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AlGaN epitaxial layer has been studied by means of temperature-dependent time-integrated photoluminescence (PL) and time-resolved photoluminescence (TRPL). An enhancing redshift phenomenon in TRPL spectra with increasing temperature was observed, and the localized excitons behaved like quasi two-dimensional excitons between 6 K and 90 K. We demonstrated that these behaviors are caused by a change in the carrier dynamics with increasing temperature due to the competition of carriers' localization and delocalization in the AlGaN alloy.

Keywords: AlGaN, time-resolved photoluminescence, localized excitons

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1. Introduction

AlGaN ternary compound has attracted much interest due to its wide application in light-emitting devices,[1,2] solar-blind ultraviolet (UV) detectors,[3,4] and high-power and high-temperature electronic devices.[5,6] With the band gap varying from 3.4 eV to 6.2 eV, it is an ideal semiconductor for UV applications, especially light-emitting devices. Although AlGaN light-emitting devices have been made successfully, the AlGaN epilayer suffers from a high density of threading dislocations due to the lattice mismatch and thermal mismatch, which act as nonradiative recombination centers. The nonradiative recombination centers lower the external quantum efficiency of the AlGaN devices. Some researchers[7,8] pointed out that carriers' localization was favorable for efficient light emission due to preventing the nonequilibrium carriers from reaching the nonradiative recombination centers. Although many researchers[9–13] have reported carriers' localization induced by potential fluctuations in AlGaN epilayers, the variation of carriers transferring with temperature is not fully understood. A further study of the carriers' dynamics of the AlGaN alloys is essential, not only for the physical interest but also for the design and the optimization of the device structures.

In this paper, we report temperature-dependent photoluminescence (PL) studies of AlGaN by means of time-integrated PL and time-resolved PL (TRPL). By combining the steady-optical measurement results with temporal response results, localized excitons' recombination dynamics are analyzed.

2. Experiment

AlGaN epilayer with a thickness of 0.5 μm was grown on *c*-plane sapphire substrate by metalorganic chemical vapor deposition (MOCVD). Trimethygallium (TMG), trimethylaluminum (TMAl), and ammonia were used as the precursors in the AlGaN growth. Prior to the AlGaN growth, 20-period $Al_{0.54}Ga_{0.46}N$/AlN short-period superlattices were deposited as the buffer layer. The Al content in the AlGaN epitaxial layer was determined by x-ray diffraction (XRD) measurements and estimated to be 54%.

PL was performed by a home-made vacuum spectrometer, with an excitation source of a 177 nm solid-state fs laser. A streak camera was used for the TRPL measurement, and the overall time resolution of the DUV laser spectroscopic system was higher than 16 ps.

3. Results and discussion

Figure 1 shows the temperature-dependent PL spectra from 6 K to 240 K. At $T = 6$ K, an asymmetric PL emission at 4.778 eV with a relatively sharp high-energy cutoff and low-energy tail is observed. This transition is due to the recombination of excitons trapped by potential fluctuation at the band edge caused by the random composition distribution. The S-shape temperature-dependent variation of the localized excitons is also shown in Fig. 1, the peak of the localized excitons decreases in the temperature range of 6–120 K, increases for 120–180 K, and decreases again for 180–240 K. This anomalous temperature dependence of the PL peak energy caused by

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carriers' localization due to the potential fluctuation is common for the AlGaN epitaxial layer. Similar results have been reported and are well explained by another group.[14]

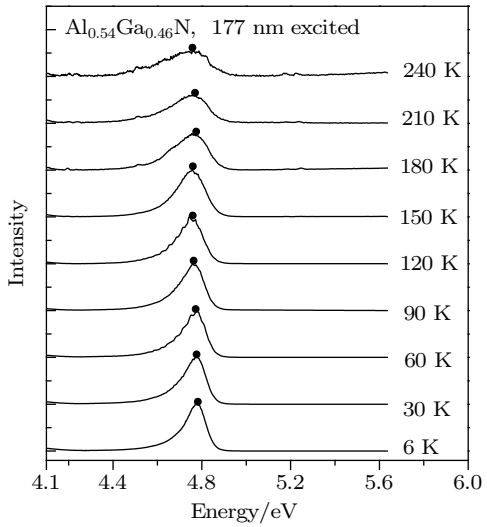


Fig. 1. Temperature dependence of PL spectra of $Al_{0.54}Ga_{0.46}N$ epilayer from 6 K to 240 K.

Figure 2 shows the temporal responses of the PL emission of the $Al_{0.54}Ga_{0.46}N$ epilayer around the peak position of the localized excitons. All of the PL decay curves can be fitted by a single exponential. At the energy range from 4.694 eV to 4.794 eV, the decay time decreases monotonously from 749 ps to 585 ps. The inset exhibits the measured PL decay time as a function of the emission energy at 6 K. We can clearly see that the decay time decreases with increasing emission energy, which is regarded as a hallmark of localized excitons.[12,15] We find that the case is also the same for the high temperature, and the decay time decreases with increasing emission energy at high temperature. At low temperature, radiative recombination is dominant, the carriers are localized in the localized states. On the one hand, compared with the low localized states, the deep localized states provide a stronger constraining effect, the separation of electron and hole wave functions is more serious, and the overlap of the wave functions is smaller, resulting in a weaker oscillator strength of the transition, and the radiative recombination lifetime of the carriers is longer. On the other hand, the high-energy localized excitons decay not only via radiative recombination but also through the additional channel of the transfer process to low-energy states. Thus, at low temperature, the decay time decreases with increasing emission energy. At high temperature, on the one hand, the high-energy localized excitons have lower localized energy, the carriers in the high-energy states can more easily escape from the localized states to free states, compared with the localized states, the overlap of the wave functions is bigger in the free states, resulting in a stronger oscillator strength of the transition, and the radiative recombination lifetime of the carriers is shorter; on the other hand, the nonradiative recombination is dominant at high temperature, compared with the low-energy localized excitons, the high-energy localized excitons are more likely to escape to free states and be captured by the nonradiative recombination centers. As a result, the decay time also decreases with increasing emission energy at high temperature.

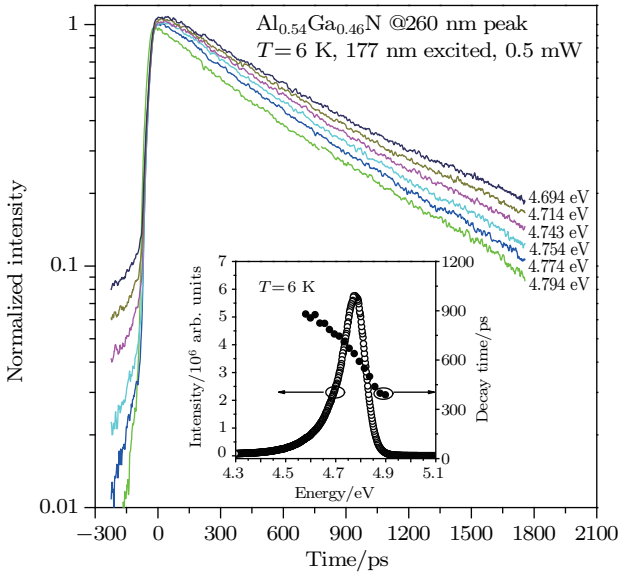


Fig. 2. Temporal responses of the PL emission of $Al_{0.54}Ga_{0.46}N$ epilayer over the energy range from 4.694 eV to 4.794 eV at 6 K. The inset shows the time-integrated PL spectra and the PL decay time as a function of emission energy for $Al_{0.54}Ga_{0.46}N$ epilayer at 6 K.

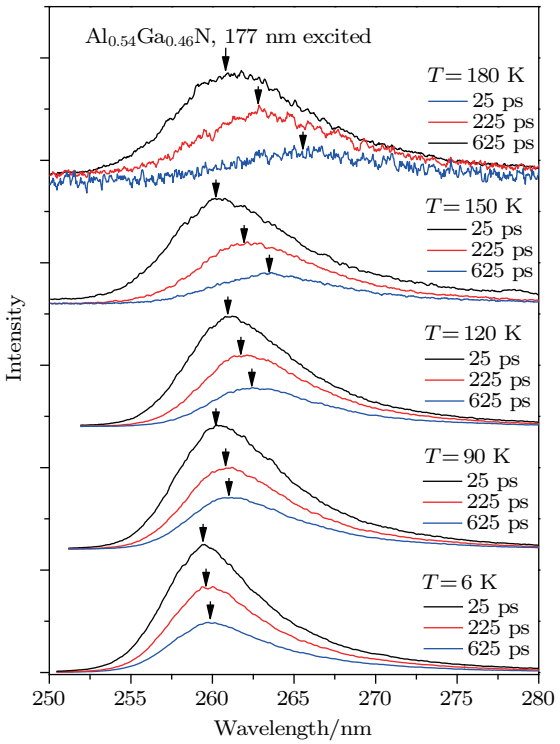


Fig. 3. TRPL spectra of localized excitons for several representative delay times at $T = 6$ K, 90 K, 120 K, 150 K, and 180 K, respectively.

In Fig. 3, we present the TRPL spectra of the localized excitons obtained at $T = 6$ K, 90 K, 120 K, 150 K, and 180 K,

respectively. The delay time $t = 0$ is chosen at the position of the maximum intensity in the luminescence temporal responses. In the TRPL spectra, there is a small redshift as the decay time increases at low temperature, and the redshift is enhancing with increasing temperature.

We would like to emphasize that the PL peak has a blueshift from 120 K to 180 K in the time-integrated PL, and the blueshift is attributed to excitons beginning to occupy different higher energy states due to increasing thermal energy.[8,9,14] We previously believed that there would be a blueshift as the decay time increases in the TRPL spectra between 120 K and 180 K, however, in the TRPL spectra, there was no blueshift as the decay time increased. It is an interesting and strange phenomenon.

According to the result reported by Li *et al.*,[16] carrier transfer time τ_{tr} caused by thermal activation is 22 fs in their InGaN films. Xu *et al.* has demonstrated that τ_{tr} is 30 fs in their InAs QDs.[17] We have also reported that τ_{tr} is 83 fs in our previous InAlN film.[18] So we think that carrier transfer time τ_{tr} is also probably a few tens of fs in this AlGaN sample, although we cannot well fit the τ_{tr} for this sample due to the irregular S-shape temperature dependence of the peak energy. We believe that the carrier transfer time caused by thermal activation is very fast, it is beyond the detection limit of our detection system, and at the delay time $t = 0$, the carriers relaxation and transfer have finished. So we cannot observe the blueshift in the TRPL spectra. For $t > 0$, the carriers at various localized states have different decay time. As we have mentioned above, the low-energy localized excitons have longer lifetime than the high-energy localized excitons. So the high-energy localized excitons decay faster, as a result, the peak redshifts as the delay time increases. The enhanced redshift with increasing temperature can be explained as follows: at low temperature, most of the carriers recombine in the low-energy localized states, so there is only a slight redshift. When the temperature increases, part of the carriers transfer from the low-energy localized states to the high-energy localized states, and the distribution of carriers expands. Since the high-energy localized excitons decay faster than the low-energy localized excitons, so the difference of recombination lifetime of carriers at various localized states is larger at high temperature. As a result, the redshift is more prominent as the temperature increases. At higher temperature, nonradiative recombination is dominant, the carriers in the high-energy states are affected by nonradiative recombination and the recombination lifetime is short; meanwhile part of the carriers in the low-energy states may be still in the localized states and have a longer recombination lifetime. Thus, the redshift is most obvious at higher temperature as the decay time increases.

Figure 4 presents the temperature responses of the localized excitons recombination at the spectral peak position from 6 K to 240 K. The inset shows the PL decay curves at various temperatures. From 6 K to 120 K, the PL decay curves can be described well by a single exponential. At $T > 120$ K, by getting rid of the laser scattering effect, the PL decay curves can still be fitted by a single exponential. Then we obtain the lifetime of the localized excitons at various temperatures, as depicted in Fig. 4. We observe an interesting phenomenon that the lifetime increases with temperature according to $T^{1.15}$ between 6 K and 90 K. As we all know, the radiative recombination lifetime of three-dimensional (3D) excitons increases with temperature according to $T^{1.5}$, while the radiative recombination lifetime of two-dimensional (2D) excitons is proportional to the temperature T.[19–21] The results indicate that in this temperature range, radiative recombination is dominant; what is more, the localized excitons in our experiment behave more similarly to 2D excitons rather than 3D excitons. In other words, the carriers are partly trapped like being confined in quantum wells. For 90 K$< T <$ 210 K, the lifetime decreases sharply as the temperatures increases; this is due to the enhanced impact of nonradiative recombination.

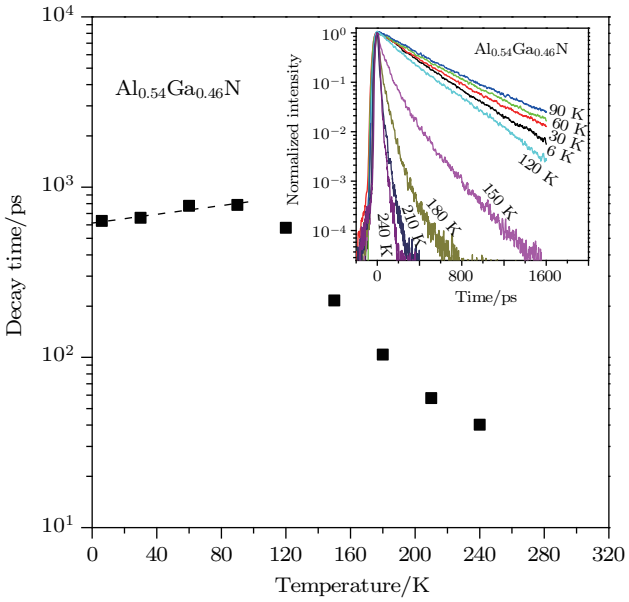


Fig. 4. Temperature dependence of the decay lifetime of localized excitons, the inset shows the temperature-dependent TRPL transients of localized excitons measured at the corresponding peak energy for the $Al_{0.54}Ga_{0.46}N$ films.

4. Conclusion

$Al_{0.54}Ga_{0.46}N$ epilayer has been studied by continuous-wave PL and TRPL measurements. S-shaped temperature dependence of the PL peak energy shift is observed, which is attributed to the localized excitons induced by potential fluctuations. TRPL spectra reveal that the redshift with increasing delay time enhances as the temperature increases, and it is probably due to the fast transferring of carriers caused by thermal activation. At the temperature range from 6 K to 90 K, radiative

recombination is dominant, and the decay time increases with temperature according to $T^{1.15}$, this result suggests that the carriers behave more similarly to 2D excitons. For $T > 90$ K, nonradiative recombination begins to play a more and more important role as the temperature increases. Due to the strong nonradiative recombination, the lifetime decreases quickly as the temperature increases. Therefore improving the growth technology and reducing dislocations which act as nonradiative recombination centers are still crucial for the further improvement of quantum efficiency for AlGaN light-emitting devices.

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