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Time-Resolved Photoluminescence Studies of AlInGaN Alloys *

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We study the two samples of AlInGaN, i.e., 1-μm GaN grown at 1030°C on the buffer and followed by a 0.6-μm-thick epilayer of AlInGaN under the low pressure of 76 Torr and the AlInGaN layer deposited directly on the buffer layer without the high-temperature GaN layer, by temperature-dependent photoluminescence (PL) spectroscopy and picosecond time-resolved photoluminescence (TRPL) spectroscopy. The TRPL signals of both the samples were fitted well as a stretched exponential decay at all temperatures, indicating significant disorder in the material. We attribute the disorder to nanoscale quantum dots or discs of high indium concentration. Temperature dependence of dispersive exponent β shows that the stretched exponential decay of the two samples comes from different mechanisms. The different depths of the localization potential account for the difference, which is illustrated by the results of temperature dependence of radiative recombination lifetime and PL peak energy.

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In the past few years, excellent progress has been made in the growth of III-nitride materials and their optical properties have been extensively studied.[1,2] However, the potential of this material system can be significantly enhanced with the ability to grow the quaternary alloy AlInGaN. By varying In and Al compositions in AlInGaN, one can change the energy band gap while keeps the lattice matched with GaN, which can be used to reduce dislocation density as well as piezoelectric field. Potential applications of AlInGaN as InGaN/AlInGaN quantum well light emitters,[3] GaN/AlInGaN heterojunction field-effect transistors,[4] and UV detectors have been demonstrated recently.[5] It is also found that the quantum efficiency of AlInGaN is enhanced significantly over AlGaN with a comparable Al content.[6] In spite of the fast development in device applications, the emission mechanisms of this material are still not fully understood and a detailed understanding of the physical origin of the emission exists in the material is very important for improvement of material quality as well as the design of optical devices.

In this Letter, we use temperature-dependent PL spectroscopy and picosecond TRPL spectroscopy to study the radiative recombination dynamics of AlInGaN alloys. The samples were grown by metal-organic chemical-vapour deposition (MOCVD) on sapphire (0001) substrate. A 20-nm-thick low-temperature GaN buffer layer was first deposited. For sample D8, 1 μm of GaN grown at 1030°C is deposited on the buffer, which was followed by a 0.6-μm-thick epilayer of AlInGaN under the low pressure of 76 Torr. While for sample D30, the AlInGaN layer was deposited directly on the buffer layer without the high temperature GaN layer. Trimethylgallium (TMG), trimethylaluminum (TMAl), ethyldimethylindium (EDMIn) and ammonia were used as sources for gallium, aluminium, indium, and nitrogen respectively; and SiH_4 was used for n-type doping. The composition of the samples was determined by energy dispersive spectroscopy (EDS), giving a result of $Al_{0.12}In_{0.095}Ga_{0.785}N$ and $Al_{0.1}In_{0.07}Ga_{0.83}N$ for D8 and D30, respectively. For TRPL measurements, the PL was excited by frequency-doubled ($\lambda = 360$ nm) laser pulses from a Ti:sapphire mode-locked femtosecond laser and the time correlated signals were analysed by a two-dimensional (2D) synchronous streak camera with an overall resolution of better than 15 ps. The PL measurement was performed in cw configuration by combination of a cooled GaAs PMT with a Data-Link electronic system.

Fig. 1 shows the temperature dependences of peak energy for both the samples. For D8, the peak energy redshifts quickly with the increasing temperature; while for D30 there is a slight blueshift at $T > 250$ K. In the insets we display the Arrhenius plots of PL intensity versus temperature of the two samples. The thermal activation energy E_A can be obtained by fitting the data with $I \propto \exp(E_A/k_BT)$. We obtain E_A equal to 30 meV for D8 and 18 meV for D30. The measured activation energies can be regarded as a measure of the exciton localization effect in the alloys.[7] Such large E_A may come from phase separation in the film. In D30 carriers are thermally activated from the potential minimum to band edge before recombination, which results in blueshift of peak energy at high temperature; while the localization potentials in D8 is too large for the carriers to

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be activated out of them until room temperature.

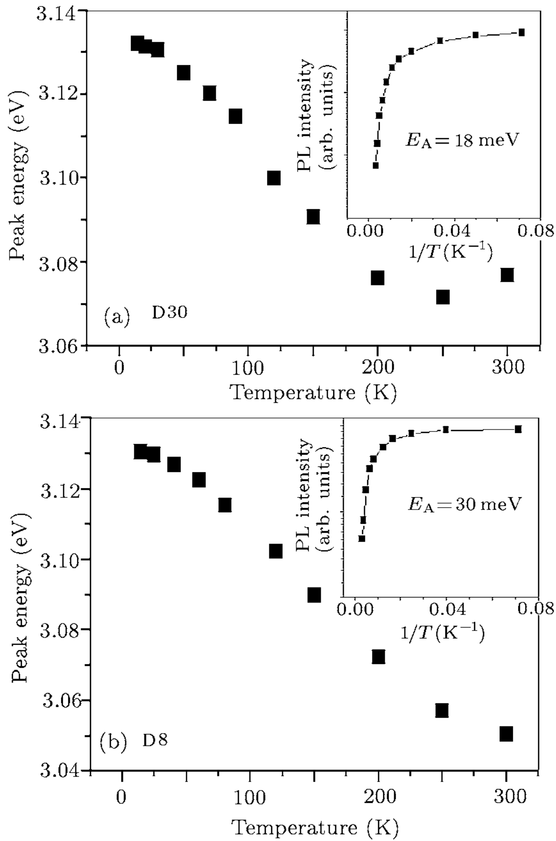

Fig. 1. PL peak energy versus temperature for sample D30 (a) and D8 (b). The insets show temperature dependences of PL intensity.

The TRPL signals from the both samples are nonexponential decay. Figure 2 presents some typical PL decays of D8 at different temperatures. A small but clear curvature from nonexponential decay behaviour is seen at all temperatures, which is a sign that the emission is from certain localized states. The PL kinetics of our samples from 14 to 250 K can be well described by a stretched exponential function:

$$I(t) = I_0 \exp[-(t/\tau)^\beta], \quad (1)$$

where $I(t)$ is the PL intensity at time t and I_0 is $I(0)$; $\beta \leq 1$ is a dispersive exponent; τ is the lifetime. This decay law is often encountered in disordered systems and is considered a consequence of the dispersive diffusion of the photoexcited carries, as observed in porous silicon.[8,9] Carrier diffusion among different spatial regions can pertain to the excitation of carriers from localized to extended states (shallow traps) or to the hopping (or tunnelling) of electrons among localized states (deep traps). In the former case, the localized states act as temporary traps and the disorder causes a distribution of release rates and of trap energies and the diffusion arises from a multiple trapping-detrapping (MTD) mechanism. In the latter one, the traps are so strong that diffusion can only come from the hopping (also tunnelling) of carries between localized states (H mechanism). In the presence of dispersive motion, e.g., H mechanism, the carries are temporarily frozen into localization centre. The late arrival of some electrons (holes) at the sites where holes (electrons) are present provides the long-time recombination, which modifies the delay law, yielding a stretched-exponential decay. Here we assume that the self-assembled quantum dots or discs formed by indium clusters act as the localization centre. The In clusters are easy to be formed in the AlInGaN alloy, which is similar to the case of InGaN.

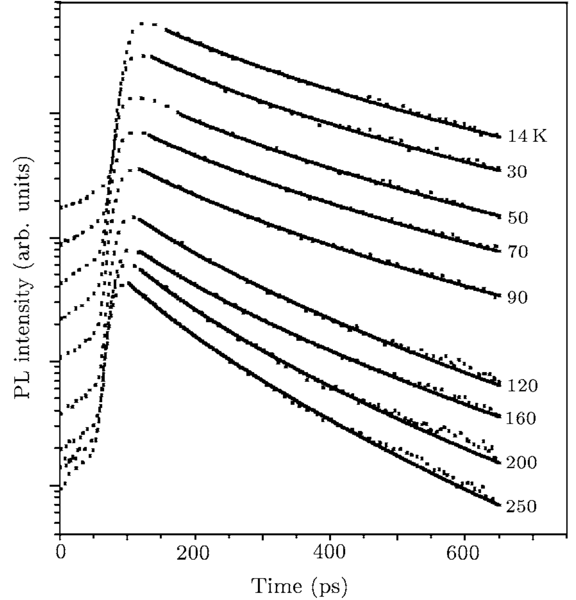

Fig. 2. TRPL from D8 at different temperatures. The dotted lines are experimental data and the full lines come from the fitting by Eq. (11).

Study of the localization effect is a good way to understand the disorder system. Here we use temperature dependence of β to determine the dominated mechanism of the dispersive diffusion. The full lines through the experimental data in Fig. 1 are the least-squares fits with stretched exponentials decay. This procedure yields a good fitting of the data and reliable τ and β parameters can be extracted in such a way. In Fig. 3 the temperature dependence of β is reported for both the samples. Obvious difference of the trend of β versus temperature lies between the two samples: for sample D30, β shows nearly a linear increase with temperature; while for sample D8, β within error 0.03 can be independent of temperature throughout the temperature range we studied. It is theoretically expected[10] that β increases linearly with temperature when the MTD mechanism dominates the diffusion, because the trapping effect is so weak that the release processes (detrapping) are thermally activated with the raising T. When localization potential is large (H mechanism dominates the diffusion), tunnelling rate is only sensitive to the topological structure of the localized centre. The exponent β can be independent of the temperature.

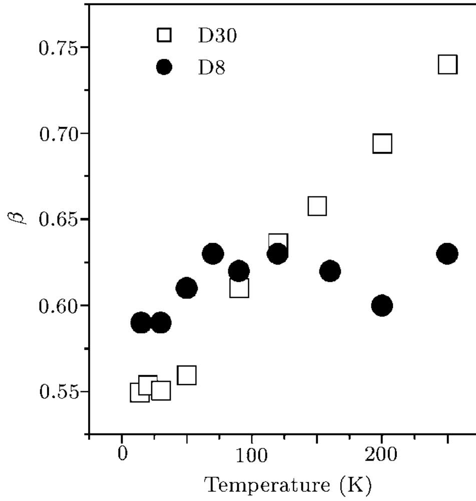

Fig. 3. Dispersive exponent β versus temperature for D8 and D30.

The dimension of localization centre is a very important parameter to reflect the property of localized excitons. Combining the PL and TRPL results, we obtain the values of the radiative lifetime $\tau_{\rm rad}$ and nonradiative lifetime $\tau_{\rm nonrad}$, as shown in Fig. 4. They are deduced from the longer component of PL decay and the PL intensity as a function of temperature using the relation $\eta_{\rm int} = 1/(1 + \tau_{\rm rad}/\tau_{\rm nonrad})$, where $\eta_{\rm int}$ is an internal quantum efficiency. The PL intensity of the both samples keeps nearly to be constant at temperatures lower than 30 K, demonstrating that the emission is dominated by radiative recombination. Therefore, $\eta_{\rm int}$ is set to be unity at low temperature of 14 K and then $\tau_{\rm rad}$ and $\tau_{\rm nonrad}$ can be expressed as

$$\tau_{\rm rad}(T) = \frac{I(14{\rm K})}{I(T)}\tau(T), \qquad (2)$$

$$\tau_{\rm nonrad}(T) = \frac{I(14{\rm K})}{I(14{\rm K}) - I(T)}\tau(T). \qquad (3)$$

The results show that strong difference lies in the behaviour of $\tau_{\rm rad}$ versus T. For D8, there is no much variation of $\tau_{\rm rad}$ from 14 K to 250 K, which is the characteristic of localized excitons of zero-dimension, strongly suggesting that emission comes from excitons localized in quantum-dot-like structures. For D30 $\tau_{\rm rad}$ increases linearly with T. It is theoretical indicated[11] that as the confinement increases (that is, the dimension is smaller), the temperature dependence of the radiative lifetimes decreases. If the dimension of density of states changes from zero to two, for example, from quantum dots to quantum discs, $\tau_{\rm rad}$ would increase linearly with temperature. In D8, the confinement is so large that electrons are still localized until room temperature. In D30, the localized excitons display characteristics of two dimensions, which implies that nanostructures such as quantum discs are formed in D30.

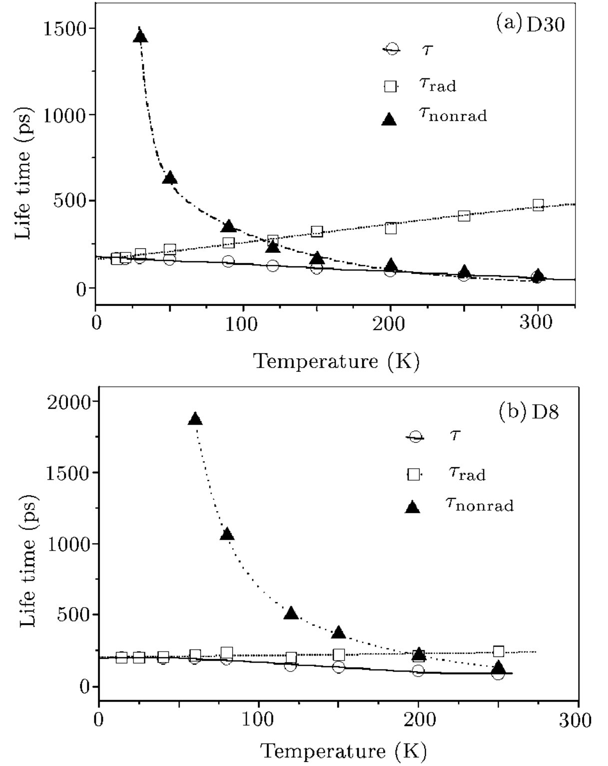

Fig. 4. PL lifetime τ_{PL} and radiative and nonradiative lifetimes $\tau_{\rm rad}$ and $\tau_{\rm nonrad}$.

In conclusion, we have studied the recombination mechanism of carriers in AlInGaN alloys by PL and TRPL. The PL intensity shows a stretched-exponential decay at all temperatures for both the samples. Temperature dependence of β shows that the stretched-exponential decay comes from different mechanisms for the two samples: MTD mechanism dominates the diffusion of carriers in D30, while H mechanism plays a role in D8. Further studies show that the dimension of localization centre in D8 is zero until room temperature, nearly two for D30. Emission energy dependence of lifetime and temperature dependence of peak energy for the two samples also displays different behaviour. All the observed experimental results are such strong evidence that nanostructures such as quantum dots or discs are formed by indium cluster in the material.

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