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Relaxation process of photoexcited carriers in GaAs structures with low-temperature-grown layers

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Spatial relaxation processes of photoexcited carriers in GaAs structures are studied by means of photoluminescence spectroscopy. A single GaAs/$Al_{0.3}Ga_{0.7}As$ quantum well and a low-temperature-grown GaAs (LT-GaAs) layer containing a high concentration of excess arsenic are placed in a GaAs structure as optical markers; the former serves as the radiative recombination site, while the latter as the trapping site of photoexcited carriers. The photoluminescence intensity from the quantum well is significantly reduced by the presence of a LT-GaAs layer immediately next to a barrier layer. The effect of the LT-GaAs layer is exponentially enhanced as a thickness of the barrier layer decreases. The results suggest that once an excess As point defect is placed within an extent of a wave function of a photoexcited carrier, trapping of the photoexcited carrier occurs at an extremely fast rate. In a structure where a LT-GaAs is placed at a distant location from the quantum well, the photoluminescence intensity from the quantum well is weakly dependent on the location of the LT-GaAs layer as expected from thermal diffusion of photoexcited carriers to trap sites as semiclassical particles. © *2005 American Institute of Physics*. [DOI: 10.1063/1.2030408]

I. INTRODUCTION

A GaAs layer grown by molecular-beam epitaxy (MBE) at a low-temperature-grown GaAs (LT-GaAs) has been considered as an important material for ultrafast optoelectronic applications such as ultrafast detectors, pulse generators, and photomixers.[1] Among a number of properties of LT-GaAs, the most important is a very short lifetime of photoexcited carriers, which results from trapping of carriers by a high concentration of excess As point defects in as-grown layers or As clusters in annealed layers. Relaxation processes of photoexcited carriers in LT-GaAs, hence, have been extensively studied in the past decade.[2–7] These studies have shown that photoexcited electrons are trapped by antisite As ions, As_{Ga}^{+},[5] while photoexcited holes are trapped predominantly by neutral antisite As atoms.[7]

In spite of numerous studies on the fast relaxation of photoexcited carriers in LT-GaAs to date, spatial processes of the relaxation in LT-GaAs have been scarcely understood. In earlier studies on LT-GaAs,[8,9] the Shockley:Read:Hall [SRH] model,[10,11] which was originally developed for the description of slow relaxation processes of photoexcited carriers in indirect band-gap semiconductors, was used for the analysis of the carrier lifetime as a function of the excess As concentration. In a recent study,[12] Rath *et al.* have investigated the relaxation process in GaAs/AlAs multiple quantum well structures grown at low temperatures. They found that the trapping rate of photoexcited carriers increased linearly with the $As_{Ga^{+}}$ concentration in a low-concentration range as expected from the SRH model. The trapping rate in a high-concentration range, where the average spacing of excess As point defects is in the order of a few nanometers was found to change superlinearly with the As_{Ga}^{+} concentration; the trapping rate increased at a faster rate than expected from the SRH model. According to the SRH model, the carrier lifetime, which is the reciprocal of the trapping rate, is expected to become shorter as the concentration of trapping sites increases, but the results obtained by Rath *et al.* suggest that the origin of the very short carrier lifetime in LT-GaAs cannot be explained only with the semiclassical picture on which the SRH model is based.

In the present paper we report a photoluminescence study on spatial relaxation processes of photoexcited carriers in LT-GaAs. Photoluminescence occurs as a result of a number of elementary processes, and, hence, this method alone is not suitable for investigation of complex spatial processes of the fast relaxation. In order to overcome this disadvantage we have grown samples with highly controlled structures by fully utilizing the capability of MBE and have compared photoluminescence intensities of samples in which only one structure parameter varies. In the samples a thin LT-GaAs layer with a high concentration of excess As is placed at a selected position with respect to a single GaAs/$Al_{0.3}Ga_{0.7}As$ quantum well; the quantum well and LT-GaAs layer serve as optical markers where the former gives rise to radiative recombination sites and the latter trap sites of photoexcited carriers. The main focus of the study is the effect of the LT-GaAs layer on the photoluminescence intensity in the structures where the LT-GaAs layer is placed immediately next to the $Al_{0.3}Ga_{0.7}As$ barrier layer. The thickness of the barrier layer was changed from 5 to 10 nm among the samples. The photoluminescence intensity from a quantum well in the sample with a LT-GaAs layer was found to be significantly weaker than that without a LT-GaAs layer. The effect of the LT-GaAs layer was found to increase exponentially as the barrier thickness is reduced. The results suggest

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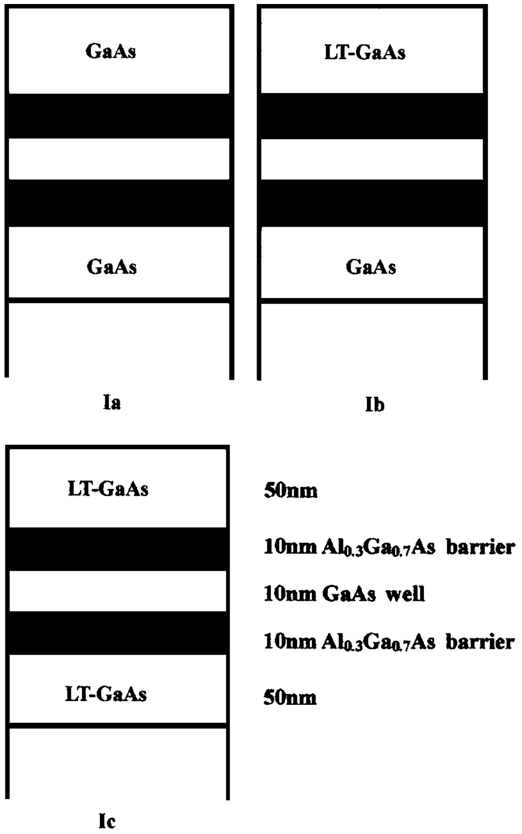

FIG. 1. Layer configurations of three samples Ia, Ib, and Ic of the first set.

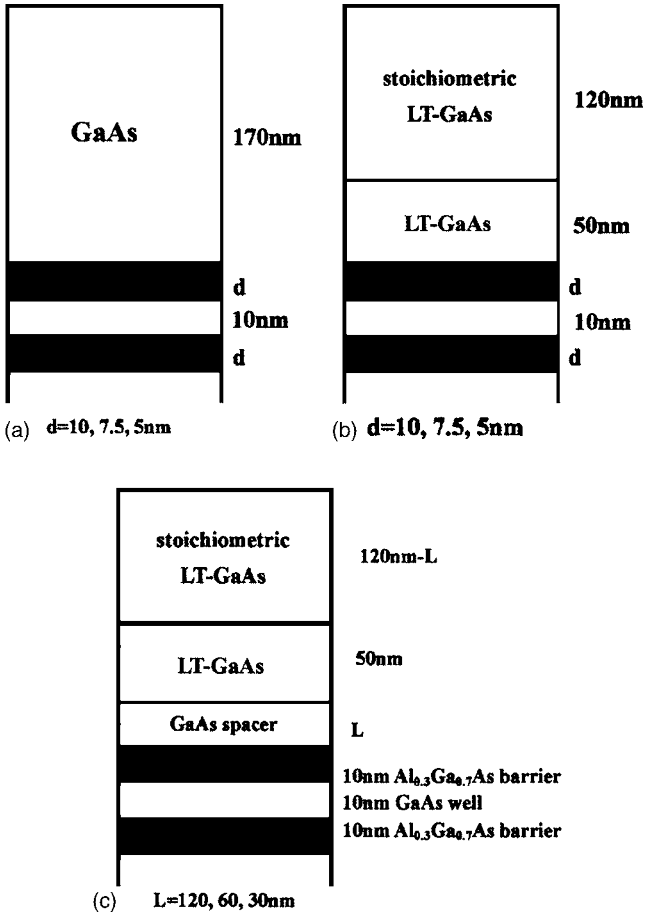

FIG. 2. Layer configurations of a pair of samples HT (a) and LT (b) of the second set for a given barrier layer thickness, and a representative structure of the third set (c).

that once an excess As point defect is placed within an extent of a wave function of a photoexcited carrier, trapping of the carrier occurs at a significantly faster rate than expected from the semiclassical picture.

II. EXPERIMENT

GaAs/$Al_{0.3}Ga_{0.7}As$ single quantum well samples were grown on semi-insulating epiready (100)GaAs substrates by utilizing a conventional MBE system. The Ga and As fluxes were 5.2×10^{-7} and 2.5×10^{-5} Torr in the ionization gauge reading, respectively. The Al cell temperature was set on the basis of lattice parameter measurements of thick $Al_xGa_{1-x}As$ layers with the x-ray diffraction method. After desorption of an oxide layer of a substrate surface and growth of a 150-nm-thick GaAs buffer layer at 580 °C, a structure with a selected layer configuration was grown.

Three sets of samples were grown for the present study. Figure 1 shows the structures of three samples Ia, Ib, and Ic of the first set, each of which has 10-nm-thick $Al_{0.3}Ga_{0.7}As$ barrier layers, a 10-nm-thick GaAs well layer, and a 50-nm-thick GaAs cap layer. The GaAs well and $Al_{0.3}Ga_{0.7}As$ barrier layers of all three samples were grown at 580 °C. The entire layers of the sample Ia were grown at 580 °C, and, hence, this sample does not contain any LT-GaAs layer. The sample Ib has a LT-GaAs cap layer which was grown at 240 °C with a high As flux and, hence, contains a high concentration of excess As point defects. According to an earlier study,[13] a LT-GaAs layer grown at 240 °C under the present As and Ga flux conditions is expected to contain 1×10^{20}-cm^{-3} As_{Ga} atoms. The sample Ic has a LT-GaAs cap layer similarly to the sample Ib but also has a 50-nm-thick LT-GaAs layer grown at 240 °C below the quantum well. Since the $Al_{0.3}Ga_{0.7}As$ barrier layers and GaAs well layer were grown at 580 °C, excess As point defects in the lower LT-GaAs layer are expected to have changed into As clusters during the high-temperature growth of the well and barrier layers.

Six samples were grown as the second set. Their single quantum wells have three different barrier layer thickness, 10, 7.5, and 5 nm, with the well layers being 10 nm. For each barrier layer thickness, a pair of samples high temperature (HT) and LT were grown, as shown in Figs. 2(a) and 2(b). One sample, HT, was grown entirely at 580 °C including a 170-nm-thick GaAs cap layer. In the other sample, LT, a LT-GaAs cap layer was grown at 240 °C with its first 50-nm-thick part containing a high concentration of excess As point defects and the remaining 120-nm-thick part being nearly stoichiometric. The stoichiometric LT-GaAs layer was grown with a 6.8×10^{-6}-Torr As flux on the basis of the results of an earlier study.[14]

The structure of the samples of the third set is shown in Fig. 2(c). In this structure a GaAs spacer layer was grown at 580 °C prior to a LT-GaAs cap layer. The thickness of the spacer layer was changed among the samples; they are 30, 60, and 120 nm. In order to keep the depth of the quantum

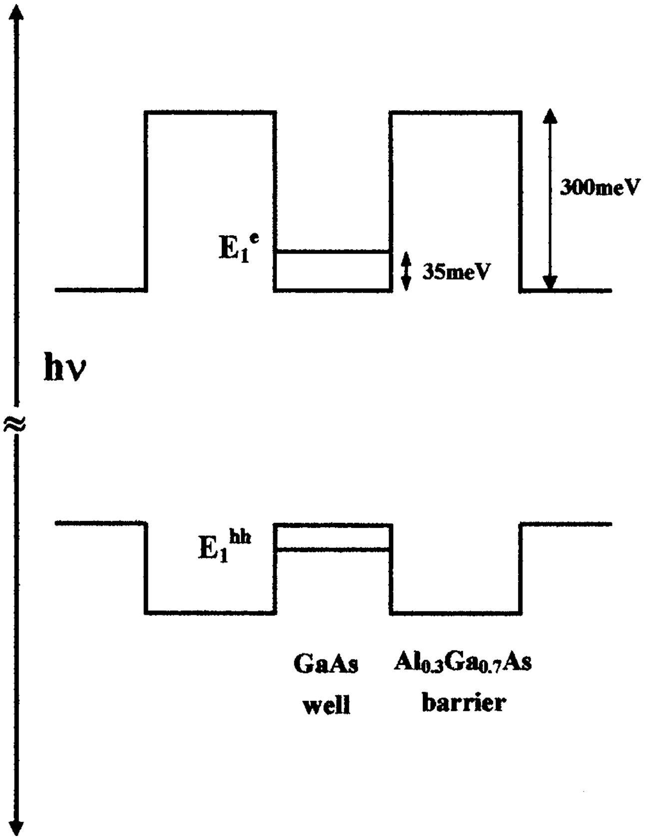

FIG. 3. Energy-band diagram of a $GaAs/Al_{0.3}Ga_{0.7}As$ single quantum well with a photoexcitation energy $h\nu$.

well from the sample surface at 170 nm for all samples of the third set, the thickness of the stoichiometric LT-GaAs layer was also changed among the samples.

Photoluminescence intensities of samples which are grown by MBE aimed at obtaining identical structures with a long period interval between their growth tend to be different from one another because of a change in the condition of a MBE growth chamber. For this reason, the samples whose photoluminescence intensities are directly compared with one another in one figure of this paper were grown successively in order to minimize an effect of such a change.

Photoluminescence measurements were carried out in the temperature range from 4.5 K to room temperature by using the 488-nm line of an Ar-ion laser as the excitation source and an InGaAs detector.In order to compare photoluminescence intensities among different samples, one sample was attached to an examined sample as a reference for each measurement. In Fig. 3 a band diagram of a single quantum well structure with a 10-nm-thick GaAs well and 10-nm-thick $Al_{0.3}Ga_{0.7}As$ barrier layers is schematically shown along with the energy $h\nu$ corresponding to the 488-nm excitation line. In the well the ground-state energies of electron and hole subbands are shown with respect to the band edges E_c and E_v. Note that the excitation energy which is 2.54 eV is far greater than the $Al_{0.3}Ga_{0.7}As$ band-gap energy, 1.89 eV, and, hence, excited carriers are not confined by the barrier layers immediately after the excitation.

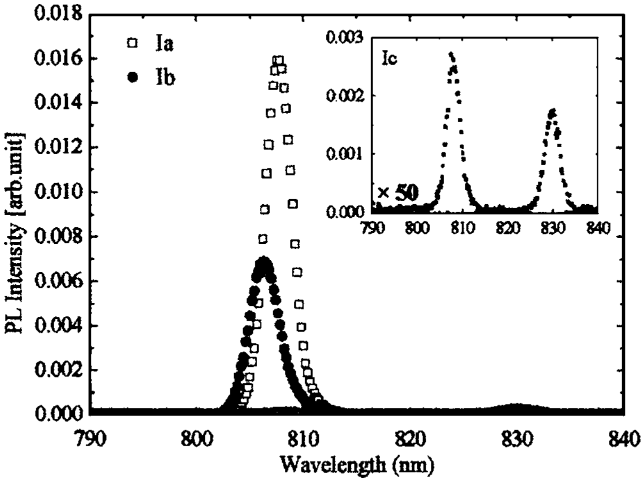

FIG. 4. Photoluminescence spectra of the sample Ia, Ib, and Ic which were observed at 4.5 K.

III. RESULTS AND DISCUSSION

Figure 4 shows the photoluminescence spectra of the three samples of the first set Ia, Ib, and Ic which were observed at 4.5 K. The peaks at approximately 808 nm correspond to the width of quantum wells which is approximately 10 nm with slight variations among the samples. The weak peak at 830 nm is considered to result from radiative recombination of carriers via the carbon acceptor level in thick GaAs layers which is known to occur commonly in the MBE-grown GaAs layers[16] and also in LT-GaAs.[17] As seen in the figure, the peak intensity of the quantum well in the sample Ib is lower than that of the sample Ia by nearly 60% of the latter intensity. The peak intensity of the sample Ic is far lower than those of the other two samples being nearly one-hundredth of that of the sample Ia. The most likely cause of the extremely low peak intensity of the sample Ic is migration of excess As through an $Al_{0.3}Ga_{0.7}As$ barrier layer into the quantum well from the previously grown LT-GaAs layer during the growth of the well and barrier layers at 580 °C. Such migration of excess As through an $Al_{0.3}Ga_{0.7}As$ layer was suggested by the results of an earlier study.[15] The reduction of the intensity from samples Ia to Ib, on the other hand, is considered to result from migration of photoexcited carriers through an $Al_{0.3}Ga_{0.7}As$ barrier layer and their trapping in the LT-GaAs layer, which will be further analyzed in the following with the results of the samples of the second set.

Figure 5 shows the photoluminescence spectra of the samples of the second set which were observed at 4.5 K. In Figs. 5(a)–5(c), the intensities of three pairs of samples LT and HT with and without a LT-GaAs layer, respectively, are compared for the same barrier thickness. The comparison of intensities among samples with different barrier thickness can be made with values indicated at the vertical axis of each figure. The weak peak at 820 nm in each figure is considered to result from radiative recombination via bound excitons in a GaAs cap layer grown at 580 °C. A photoluminescence peak due to bound excitons is known to occur in a MBE-grown GaAs layer[16] but has not been observed in a LT-GaAs layer which is grown at a substrate temperature below

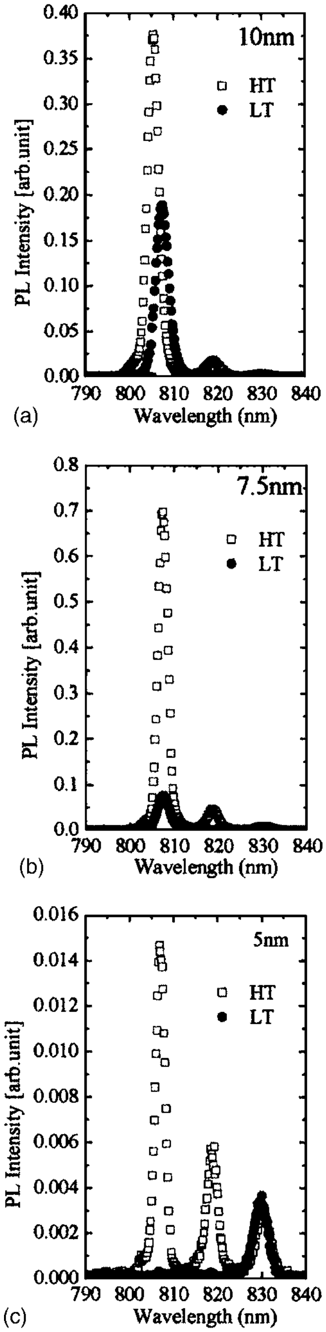

FIG. 5. Photoluminescence spectra of a pair of samples for (a) 10-, (b) 7.5-, and (c) 5-nm-thick barrier layers which were observed at 4.5 K.

300 °C.[17] The 830-nm peaks of the samples with 5-nm-thick barrier layers appear to be far higher than those of other samples, but such appearance is due to the very low 808-nm peak of the sample. From the sample with a LT-GaAs layer and 5-nm-thick barrier layers, no detectable peak is observed at 808 nm as shown in Fig. 5(c), although a quantum well with similar structure quality to other samples has formed in this sample. In Fig. 6, the cross-sectional transmission electron microscope (TEM) images of two samples are compared; one is the sample having 5-nm-thick barrier layers with a LT-GaAs layer, and the other is the sample of the third set, Fig. 2(c), with a 60-nm-thick spacer layer. The latter sample was annealed at 600 °C for 15 min in order to show excess As in the LT-GaAs layer as As clusters in the TEM image. Note that the quantum well structure of the former sample has similar quality with that of the latter sample which gives rise to a significant photoluminescence intensity of the quantum well as explained later. As seen in Fig. 5, the photoluminescence intensity from the quantum well is reduced by the presence of a LT-GaAs layer for all three cases of the barrier thickness. The degree of the reduction becomes far more significant as the barrier layer becomes thinner from 10 to 5 nm. The intensity of a sample with an LT-GaAs layer is one-half, one-tenth, and less than one-hundredth of a sample without a LT-GaAs layer for each barrier layer thicknesses of 10, 7.5, and 5 nm, respectively.

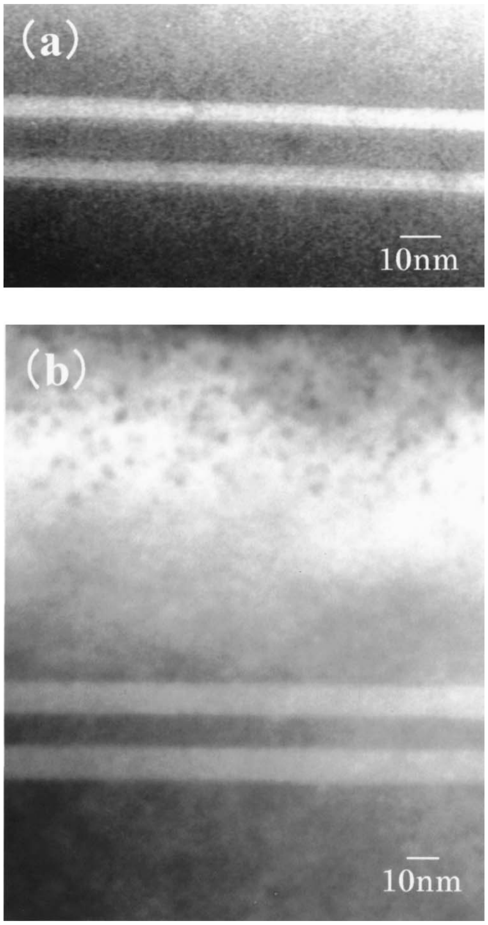

FIG. 6. Transmission electron microscope images of (a) the sample with the 5-nm-thick barrier layers and a LT-GaAs cap layer and (b) the sample with the 60-nm-thick spacer layer. The latter sample was annealed in order to show excess As as As clusters in the TEM image.

The migration of carriers through a barrier layer occurs predominantly via tunneling at this low measurement temperature, and the tunneling probability is exponentially dependent on the barrier thickness. The results shown in Fig. 5, hence, appear to be explained with a simple physical picture of the carrier tunneling. Careful examination, however, is necessary in order to clarify the role of a LT-GaAs layer in this carrier trapping process. As shown in Fig. 3, the height of the $Al_{0.3}Ga_{0.7}As$ barrier for electrons is 300 meV, and the lowest level of the subband in the 10-nm-thick quantum well is 35 meV higher than the conduction-band edge. Because of the energy difference between the lowest levels of conduction electrons in the quantum well and the LT-GaAs layer,

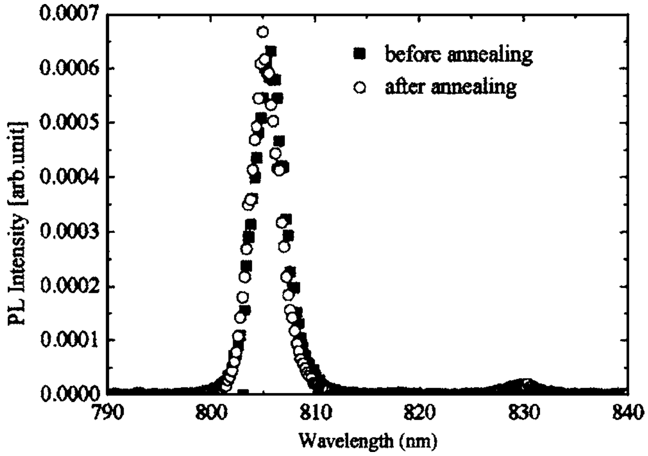

FIG. 7. Photoluminescence spectra of the sample with 10-nm-thick barrier layers and a LT-GaAs layer which were observed at 4.5 K before and after annealing at 240 °C for 3 h.

the carrier tunneling between the lowest levels occurs at 4.5 K only in the direction towards the outside of the quantum well. The population of photoexcited carriers in the lowest level of the quantum well and, hence, the photoluminescence intensity are expected to decrease as the barrier becomes thinner regardless of the presence of a LT-GaAs layer outside the quantum well. The low photoluminescence intensity of the sample with 5-nm-thick barrier layers and without a LT-GaAs layer in comparison to those with 10-nm-thick or 7.5-nm-thick barrier layers may be attributed to this process. The photoluminescence intensity of the sample having 7.5-nm-thick barrier layers without a LT-GaAs layer, however, is twice higher than that of the sample having 10-nm-thick barrier layers without an LT-GaAs layer. This opposite trend is considered to be due to different conditions of the MBE growth chamber when these two samples were grown. Two samples in each of Fig. 5, however, were grown successively in order to make direct comparison of their photoluminescence intensities as described earlier.

One possible process of the reduction of the photoluminescence intensity due to the presence of a LT-GaAs layer in the samples of the second set is diffusion of excess As point defects into the quantum well from the LT-GaAs layer through a thin $Al_{0.3}Ga_{0.7}As$ layer during the growth of the LT-GaAs layer and cap layer at 240 °C. Although significant diffusion and redistribution of excess As point defects are known to occur only at temperatures far higher than 240 °C, one cannot ignore this possibility without experimental verification. For this reason, the samples of 7.5-nm-thick barrier layers and 10-nm-thick barrier layer with LT-GaAs layers were annealed at 240 °C for 2 h and 3 h, respectively, and their photoluminescence intensities were measured before and after the annealing. If diffusion of excess As point defects through the barrier layer occurs at this temperature, further reduction of the photoluminescence intensity should be observed after annealing. For both samples, no significant intensity reduction by the annealing was found as shown in Fig. 7, indicating negligible diffusion of excess As during the growth of layers at 240 °C. The low photoluminescence intensity due to the presence of a LT-GaAs layer, therefore, should be attributed to a process other than diffusion of excess As point defects into the quantum well.

The excitation energy of the 488-nm line is greater than the band gap of $Al_{0.3}Ga_{0.7}As$ barrier layers. Carriers are, hence, excited in the barrier layers and may contribute to photoluminescence intensities of the quantum well. This process also needs to be considered in explaining the change in the photoluminescence intensity with the change in the barrier layer thickness but is not directly relevant to the change in the photoluminescence intensity due to the presence of a LT-GaAs layer for a given barrier thickness. Photoexcited carriers occupy higher-energy levels immediately after their excitation. Carriers with the 2.54-eV photoexcitation energy have a greater energy than the $Al_{0.3}Ga_{0.7}As$ barrier energy, and, hence, they can move freely irrespective of the existence of the barrier layers, transferring to lower-energy levels. Carriers at energy levels closer to the top of a barrier, on the other hand, have greater tunneling probability through the barrier.[18] In the early stage of photoexcited carriers which is less than a few picoseconds after the excitation,[18] therefore, photoexcited carriers are expected to move out and move in the quantum well almost freely via tunneling. If trapping of the carriers by excess As point defects occurs significantly in this early stage, the presence of a LT-GaAs makes a significant difference in the photoluminescence intensity from the quantum well, and its effect becomes more significant for a sample with thinner barrier layers. The results shown in Fig. 5 strongly suggest the occurrence of such a process. Because of the direct involvement of carrier tunneling, the reduction of the photoluminescence intensity and, hence, the trapping rate of photoexcited carriers are exponentially dependent on the barrier thickness. This implies that if an excess As point defect is present within an extent of a wave function of a photoexcited carrier where the carrier can no longer be treated as a semiclassical particle, the trapping rate becomes far more sensitive to their distance than that expected from the SRH model.

In the samples of the third set a LT-GaAs layer is placed at a distant location from a quantum well by inserting a thick GaAs spacer layer. The thickness of a spacer layer was changed from 30 to 120 nm with the thickness of a $Al_{0.3}Ga_{0.7}As$ barrier layer being kept 10 nm. Figure 8 shows the photoluminescence spectra from quantum wells in these samples, which were observed at 4.5 and 75 K. At both temperatures the photoluminescence intensities of samples with thinner spacer layers are lower than those with thicker spacer layers. Figure 8, however, shows that the dependence of the intensity on the spacer layer thickness is far weaker than the case shown in Fig. 5. This weak dependence on the spacer layer thickness suggests that photoexcited carriers migrate to a LT-GaAs layer via thermal diffusion as semiclassical particles in the former set of samples and, therefore, their trapping rate can be better described in terms of the SRH model. Excitation of carriers in a GaAs spacer layer may also contribute to photoluminescence intensity of the quantum well, as they are not immediately trapped by excess As point defects and, hence, can migrate to the quantum well via ther-

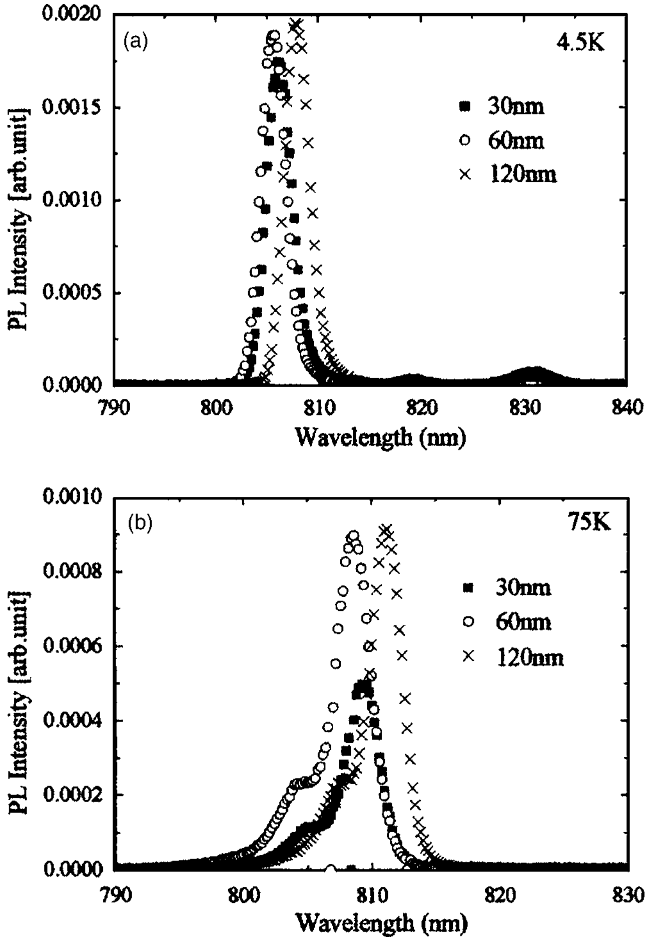

FIG. 8. Photoluminescence spectra of three samples of the third set with the 30-, 60-, and 120-nm-thick spacer layers which were observed at 4.5 K (a) and 75 K (b).

mal diffusion. This process is expected to give rise to a higher intensity in a thicker spacer layer similarly to the above process.

In summary, spatial relaxation processes of photoexcited carriers in GaAs structures were studied by means of photoluminescence spectroscopy with the samples where a single GaAs/$Al_{0.3}Ga_{0.7}As$ quantum well and a LT-GaAs layer containing a high concentration of excess As were placed at selected locations. The photoluminescence intensity from the quantum well is significantly reduced by the presence of a LT-GaAs layer immediately next to a barrier layer. The effect of the LT-GaAs layer is exponentially enhanced as the thickness of the barrier layer deceases. The results suggest that once an excess As point defect is placed within an extent of a wave function of a photoexcited carrier, trapping of the photoexcited carrier occurs at an extremely fast rate.

[1] J. F. Whitaler, in *Properties of Gallium Arsenide*, 3rd ed., edited by M. R. Brozel and G. E. Stillman (INSPEC, London, 1996), p. 693.

[2] S. D. Benjamin, H. S. Loka, A. Othonos, and P. W. E. Smith, Appl. Phys. Lett. **68**, 2544 (1996).

[3] P. Grenier and J. F. Whitaker, Appl. Phys. Lett. **70**, 1998 (1997).

[4] T. S. Sosnowski, T. B. Norris, H. H. Wang, P. Grenier, J. F. Whitaker, and C. Y. Sung, Appl. Phys. Lett. **70**, 3254 (1997).

[5] U. Siegner, R. Fluck, G. Zhang, and U. Keller, Appl. Phys. Lett. **69**, 2566 (1996).

[6] T. Okuno, Y. Masumoto, M. Ito, and H. Okamoto, Appl. Phys. Lett. **77**, 58 (2000).

[7] R. Admacicius, A. Krotkus, K. Bertulis, V. Sirutkaitis, R. Butkus, and A. Piskarskas, Appl. Phys. Lett. **83**, 5304 (2003).

[8] S. Gupta, M. Y. Frankel, J. A. Valdmanis, J. F. Whitaker, G. A. Mourou, F. W. Smith, and A. R. Calawa, Appl. Phys. Lett. **59**, 3276 (1991).

[9] A. J. Lochtefeld, M. R. Melloch, J. P. Chang, and E. S. Harmon, Appl. Phys. Lett. **69**, 1465 (1996).

[10] W. Shockley and W. T. Read, Phys. Rev. **87**, 835 (1952).

[11] R. N. Hall, Phys. Rev. **87**, 387 (1952).

[12] M. C. Rath, T. Araya, S. Kumazaki, K. Yoshihara, and N. Otsuka, J. Appl. Phys. **94**, 3173 (2003).

[13] A. Suda and N. Otsuka, Surf. Sci. **458**, 162 (2000).

[14] S. Fukushima, T. Obata, and N. Otsuka, J. Appl. Phys. **89**, 380 (2001).

[15] L. W. Yin, Y. Hwang, J. H. Lee, R. M. Kolbas, R. J. Trew, and U. K. Mishra, IEEE Electron Device Lett. **11**, 561 (1990).

[16] B. Hamilton, in *Properties of Gallium Arsenide*, 3rd ed., edited by M. R. Brozel and G. E. Stillman (INSPEC, London, 1996), p. 291.

[17] T. Obata, S. Fukushima, T. Araya, and N. Otsuka, J. Cryst. Growth **227–228**, 112 (2001).

[18] J. Shah, *Ultrafast Spectroscopy of Semiconductors and Semiconductor Nanostructures*, 2nd ed. (Springer, Berlin, 1999).