Excitonic transitions in GaAs-Al$_x$Ga$_{1-x}$As multiple quantum wells affected by interface roughness

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Time-resolved photoluminescence has been used to study the effects of interface roughness on excitonic transitions in GaAs-Al$_x$Ga$_{1-x}$As multiple quantum wells. In addition to the luminescence linewidth broadening and Stokes red shift, the interface roughness also strongly affects the dynamic process of optical transitions so that the excitonic transition peak shifts with delay time. However, the heavy-hole exciton transition has red shifts at short delay times and exhibits a turnover at longer delay times. A maximum shift of about 0.1 meV at a delay time of 4 ns was obtained. We have demonstrated that the peak shift is caused by interface roughness in the quantum wells. Furthermore, the decay of the excitonic transition is found to fit a two-exponential form. Based on a model involving interface roughness and two-exponential decay, we calculated the position of the excitonic transition peak as a function of delay time. Our calculations are consistent with experimental results.

I. INTRODUCTION

Recently, quantum-well (QW) and superlattice (SL) structures have attracted a great deal of attention because of their novel properties.[1-4] Since the proposal of these exciting new structures,[5,6] they have been studied extensively, and thus many important features have been discovered. For fundamental physics, quantum-well and superlattice structures have been used to explore the physical properties of a whole new field of low-dimensional systems and quantum effects. Many novel phenomena in the quantum regime have been discovered, such as resonant tunneling of double-barrier quantum wells with negative differential resistance.[7,8] By separating the impurities with charge carriers by modulation doping, a significant mobility enhancement in GaAs-Al$_x$Ga$_{1-x}$As quantum wells has been achieved.[9,10] Applications of these quantum wells and superlattices include high-speed electronics, optoelectronics and photonic devices, such as quantum-well lasers,[11,12] modulation-doped field-effect transistors[13] (MODFET), photodetectors,[14] etc.

Although much work has been done in this field, there are only a few investigations that concentrate on the dynamic processes of excitonic transitions in QW and SL. The lifetimes of excitons in GaAs-Al$_x$Ga$_{1-x}$As quantum wells were first reported by Christea et al.[15] An increase of almost one order of magnitude in the transition rate of excitonic recombination in GaAs quantum wells with a well width of 52 Å compared with bulk GaAs has been observed. They also found that the decay of the excitonic recombination is nonexponential.

Molecular beam epitaxy (MBE) techniques can be used to grow QW and SL with very high quality but roughness at the interfaces of two materials of QW and SL can still not be completely eliminated. It is of interest and importance to know how this roughness affects the optical processes in QW's. The interface roughness (or interface defects) in QW's has been studied previously by low-temperature continuous-wave (cw) photoluminescence and by photoluminescence excitation spectroscopy.[16,17] The main effects so far observed due to the interface roughness were exciton linewidth broadening and the red shift (Stokes shift) of the emission: the emission of the lowest heavy-hole exciton generally is slightly shifted to low energy (typically a few meV) with respect to the absorption or excitation spectrum maximum. This was attributed to localization of excitons within potential fluc-

tuations due to interface roughness. The cw luminescence and photoluminescence excitation spectroscopy used to date to study the interface roughness of the QW yields very little understanding about the role of the interface roughness in the dynamic process of optical transitions.

In this paper we investigate the effects of interface roughness on the dynamic process of optical transitions. Low-temperature time-resolved photoluminescence of GaAs-Al$_x$Ga$_{1-x}$As multiple QW's has been studied. The peak positions of heavy-hole exciton luminescence shifting with delay time has been observed. The shift is accounted for by interface roughness. A calculation based on interface roughness and a two-exponential decay forms fits the experimental results very well.

II. EXPERIMENT

The sample used for this study was a GaAs-Al$_{0.5}$Ga$_{0.5}$As multiple quantum well (MQW) which was grown by molecular beam epitaxy on a GaAs(100) substrate without growth interruption. It consists of alternate 278-Å Al$_{0.5}$Ga$_{0.5}$As barrier layers and 250-Å well layers with a total of ten periods. The x-ray diffraction data indicate a period length of 525 Å, which is in good agreement with the design parameters.

Excitation pulses of about 7 ps in duration at a repetition rate of 1 MHz were provided by a cavity-dumped ultrafast dye laser (Coherent 702-2CD) with an average power of 10 mW, which was pumped by a yttrium aluminum garnet (YAG) laser (Quantronix 416) with a frequency doubler. The pulse duration was continually monitored by using a rapid-scan autocorrelator.[18] The Lansing photon energy was 2.125 eV with a spectral width of 2 meV. A time-correlated single-photon counting system with a double monochromator (Jarell Ash 25-100) and a computer were used for the measurements. The effective time resolution of the system is about 0.2 ns. The sample was mounted strain free inside a closed-cycle He refrigerator and maintained at a temperature of 8.5 K.

III. RESULTS AND DISCUSSION

Experimental results of low-temperature (8.5-K) time-resolved photoluminescence at three different delay times for a GaAs-Al$_{0.5}$Ga$_{0.5}$As MQW are plotted in Fig. 1. The exciting photon energy was 2.125 eV at an average power density of $\sim$50 mW/cm^2. The luminescence at different delay times has been rescaled for presentation. The peaks at 1.5252 and 1.5295 eV are ascribed, respectively, to transitions of 1s heavy-hole ($n=1$, e-HH) and light-hole ($n=1$, e-LH) excitons, which are composed of an electron and a heavy (light) hole belonging to the lowest state ($n=1$) in the QW. These values are consistent with those calculated by using the transfer-matrix method[19,20] with the conduction-band offset parameter being 0.65 and the binding energies of heavy- and light-hole excitons being 6.0 and 6.2 meV, respectively. The observed spectral width (about 1.78 meV) is attributed to the interface roughness of the QW.[16,17] In Fig. 1 there is a shoulder at about 1 meV below the heavy-hole exciton
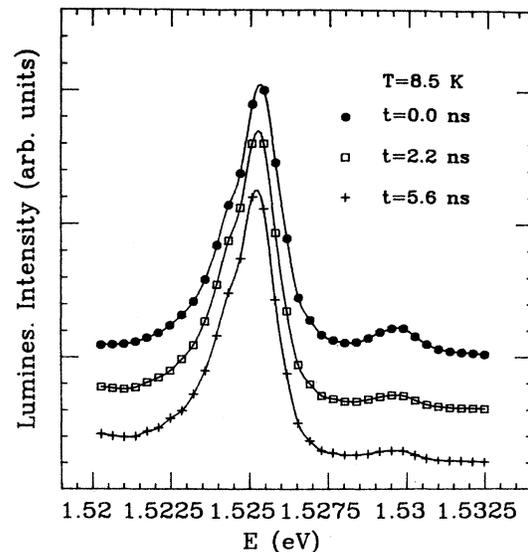

FIG. 1. Low-temperature (8.5 K) time-resolved photoluminescence at three different delay times for GaAs-Al$_{0.5}$Ga$_{0.5}$As multiple quantum wells with thicknesses of the well and barrier being 250 and 278 Å, respectively. The excitation energy was 2.125 eV with an average power density about 50 mW/cm^2.

peak which has also been reported by Miller and co-workers.[21,22] Based on the excitation intensity, temperature, and polarization dependencies of this low-energy peak (shoulder here), they concluded that it was due to a biexciton transition with a binding energy of about 1 meV. By using excitation-intensity-dependent luminescence and time-resolved spectroscopy, Charbonneau and co-workers[23] recently showed that the lower-energy components of heavy-hole excitonic transitions have different origins in different samples and can be attributed either to biexcitons or to impurity-bound excitons.

One important feature depicted in Fig. 1 is that the exciton transition peak shifts toward lower energy as the delay time increases. This is manifested as the intensity ratio of two data points at the exciton luminescence maximum changing with increasing delay time. Here, delay times were measured from the end of the excitation pulses. The origin of this shift cannot be correlated with the filling state phenomena[24,25] because of the temporal behavior as will be discussed later.

In order to fully explore the fact that the exciton transition peak shifts with delay times, we have performed a high spectral resolution experiment around the peak position of the heavy-hole exciton transition. The results are shown in Fig. 2, where all the parameters and experimental conditions are the same as those in Fig. 1. The solid lines in Figs. 1 and 2 are a guide to the eye. Figure 2 clearly demonstrates that the peak position of the heavy-hole exciton transition shifts toward lower energy with increasing delay times. We have used a least-squares fit for the experimental data to find the peak positions at different delay times. The effect of the lower energy
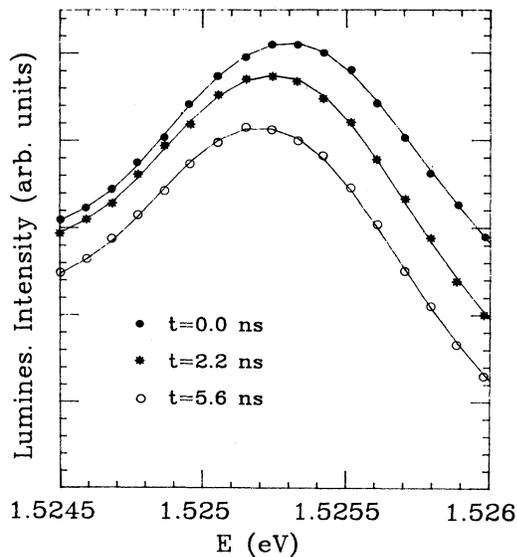
FIG. 2. High resolution time-resolved photoluminescence spectra around the peak position of the heavy-hole exciton transition at three different delay times. The spectral slit width and step width were, respectively, 0.4 and 0.5 Å. Other parameters and experimental conditions are the same as those in Fig. 1.

shoulder on the peak position of the heavy-hole exciton transition have been eliminated by deconvolution. This effect is shown to be negligible. Only a few data points around the maximum intensity of the excitonic transition were sufficient to obtain the peak position at different delay times. The results for the heavy-hole exciton peak position at differnt delay times are shown in Fig. 3 (circles). From Fig. 3 we see that the maximum shift is only about 0.1 meV at about 4 ns. Another feature is that the
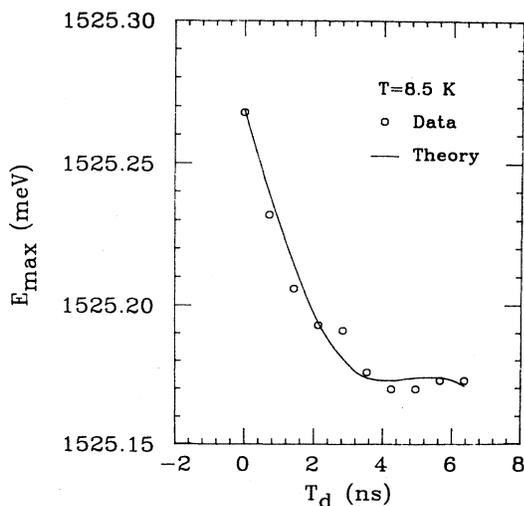
peak positions shows a red shift at the short delay time (0–4 ns), and then turns over beyond 4 ns. One notices that the shift at short delay times is almost linear. The amount of shift is small; nevertheless, it contains very important information. The experimental results shown in Fig. 3 can be interpreted in terms of the interface roughness in QW's and a two-exponential decay of the excitonic transition.

Figure 4 presents a schematic diagram of the interface roughness in a QW along the growth axis (a), and the concomitant effect on the photoluminescence linewidth broadening (b) and exciton lifetimes (c). The fluctuation in well (barrier) thickness can only be integral multiples of one monolayer. In the quantum-well layer plane, there are domains formed by the different well thicknesses with sizes varying from a few hundred angstroms to a few micrometers. Different emission energies around the principle excitonic peak correspond to excitons recombined in different spatial domains. As we have shown in Fig. 4, line A represents the lower photon energy which corresponds to excitons recombined at the location of a wider quantum well while line B represents the higher photon energy which corresponds to the excitons recombined at the location of a narrower quantum well. In Fig. 4(b) FWHM indicates the full linewidth at the half maximum. There was no investigation carried out previously ad-

FIG. 3. Energy position of the emission intensity maximum of heavy-hole exciton as a function of delay time for GaAs-$Al_{0.5}Ga_{0.5}As$ multiple quantum wells. The quantum well parameters are the same as those in Fig. 1.
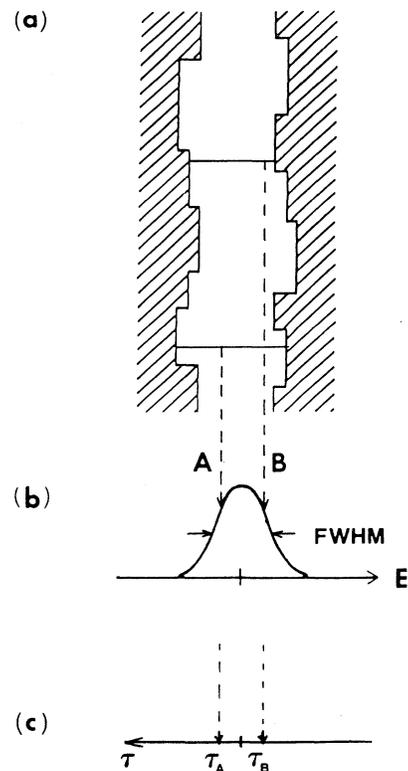

FIG. 4. A schematic diagram of the interface roughness in a quantum well shown along the growth axis (a). (b) and (c) show effects of roughness on the linewidth broadening and exciton lifetimes.

dressing the fact that excitons recombined at line A will also have a larger lifetime than those recombined at line B. Thus near the excitonic transition intensity maximum, the lifetime of the exciton, τ, will have a dependence on emission energy E, which can be written as $\tau(E)$. Because the well fluctuation normally is within a few monolayers the change in lifetime of excitons around the peak position is small. This is probably the reason that this effect has been previously neglected. However, the change of exciton lifetime with respect to the emission energy E, $d\tau/dE$, can provide very important physical information, since the recombination rate of the exciton depends on the QW thickness.[26,27] The larger the quantum well width, the longer the exciton lifetime, as indicated in Fig. 4(c).

First, let us prove that energy-dependent exciton lifetimes can cause the transition peak to shift with the delay time. Because the total amount of peak shift is very small (0.1 meV) we can write τ as a function of E, near the energy of maximum intensity E_0, as

$$\tau(E)=\tau(E_0)+(E-E_0)\alpha \; ; \tag{1}$$

here $\tau(E_0)$ is the lifetime of excitons measured at the energy of maximum intensity of delay time $t_d=0$ and α is the lifetime change rate with respect to energy, $d\tau/dE|_{E_0}$. Assuming the line-shape intensity distribution around the peak is a Gaussian with a single exponential decay, the luminescence of the excitonic transition can be written as a function of delay time t_d and energy E as

$$I(E,t_d)=I_0\exp\left[-\frac{(E-E_0)^2}{\sigma^2}-\frac{t_d}{\tau(E)}\right] . \tag{2}$$

Here I_0 and E_0 are the maximum intensity of the excitonic transition and the energy position of the intensity peak at delay time $t_d=0$, respectively, while σ defines the linewidth which correlates the QW thickness fluctuation parameter.[28] In Eq. (2), $I_0\exp[-(E-E_0)^2/\sigma^2]$ represents the Gaussian intensity distribution at $t_d=0$, and $\exp[-t_d/\tau(E)]$ is the decay factor which depends on energy. The peak positions at different delay times can be obtained by setting

$$\frac{dI(E,t_d)}{dE}=0 , \tag{3}$$

which gives

$$E_{\max}(t_d)=E_0+\frac{\sigma^2}{2[\tau(E)]^2}\frac{d\tau}{dE}t_d . \tag{4}$$

Here we have assumed that the linewidth of the excitonic transition is independent of the delay time, which is consistent with the experimental results shown in Fig. 1. Because the second term in Eq. (1) is much smaller than the first, we obtain from Eq. (4)

$$E_{\max}(t_d)=E_0+\frac{\sigma^2}{2[\tau(E_0)]^2}\alpha t_d . \tag{5}$$

Equation (5) indicates that the peak position of the excitonic transition will shift linearly to lower energies as the delay time increases, since $\alpha=d\tau/dE=(d\tau/dL)/(dE/dL)<0$, i.e., $d\tau/dL>0$ and $dE/dL<0$. Here L is the average thickness of the QW's.

The above calculation shows that the peak position of the excitonic transition will shift as the delay time increases because of the presence of interface roughness in the QW's. However, Eq. (5) only gives a linear red shift of peak position with the delay time, which is inconsistent with experimental observation depicted in Fig. 3. The experimental results are more complicated and cannot be fully described by Eq. (5). As we will see, by using the above treatment with the assumption of a two-exponential decay for luminescence, the experimental results can be well accounted for. This is obtained by rewriting the luminescence intensity as a function of emission energy E and delay times t_d, around the peak position, as

$$I(E,t_d)=\exp[-(E-E_0)^2/\sigma^2]$$
$$\times\{A\exp[-t_d/\tau_1(E)]$$
$$+B\exp[-t_d/\tau_2(E)]\} , \tag{6}$$

where A and B are constants. Here τ_1 and τ_2 are time constants which are both functions of energy E. From Eq. (3) we get

$$E_{\max}(t_d)=E_0+\frac{\sigma^2}{2}\frac{f(E,t_d)}{g(E,t_d)} , \tag{7}$$

with

$$f(E,t_d)=\sum_{i=1}^{2}\frac{C_it_d}{\tau_i^2(E)}\left[\frac{d\tau_i}{dE}\right]\exp\left[-\frac{t_d}{\tau_i(E)}\right] \tag{7a}$$

and

$$g(E,t_d)=\sum_{i=1}^{2}C_i\exp\left[-\frac{t_d}{\tau_i(E)}\right] , \tag{7b}$$

where

$$C_i=\begin{cases}1 & (i=1)\\ B/A & (i=2) .\end{cases} \tag{7c}$$

One notices that $C_2=B/A$ is the ratio of two luminescence components at $t_d=0$. For energies near that of the excitonic transition peak, we can write

$$\tau_i(E)=\tau_i(E_0)+(E-E_0)\alpha_i \quad (i=1,2) . \tag{8}$$

By using Eqs. (7), (7a), (7b), and (8) we obtain

$$E_{\max}(t_d)=E_0+\frac{\sigma^2}{2}\frac{\sum_{i=1}^{2}[C_i\alpha_it_d/\tau_i^2(E_0)]\exp[-t_d/\tau_i(E_0)]}{\sum_{i=1}^{2}C_i\exp[-t_d/\tau_i(E_0)]} . \tag{9}$$

Parameters σ, τ_1, τ_2, and C_2 can be deduced from experimental measurements. One sees that σ is related to the FWHM by FWHM$=2(\ln 2)^{0.5}\sigma$. The value of FWHM obtained from Fig. 1 is about 1.78 meV, which gives $\sigma = 1.07$ meV. $\tau_1(E_0)$ (-0.9 ns) is the time constant of the fast decay component of exciton luminescence measured at the energy of the intensity maximum occurring at $t_d = 0$, which corresponds to the exciton lifetime of radiative recombination. $\tau_2(E_0)$ is the time constant of the slow decay component, which is measured to be 4.2 ns. One notices that this time constant does not represent the lifetime of exciton recombination since it is even larger than the exciton lifetime in bulk GaAs (3.3 ns).[29] (The physical origin for this two-exponential decay is under investigation.) Now C_2 is measured to be 0.07. We insert these values into Eq. (9) and adjust α_1 and α_2 to obtain the least-square fit with experimental data. We find that, with $\alpha_1 = -0.06$ ns/meV and $\alpha_2 = -0.42$ ns/meV, the plot of Eq. (9) shown as the solid line in Fig. 3 is in excellent agreement with experimental results (circles). Here α_1 represents the change rate of exciton lifetime with respect to emission energy, and thus the fitting value (-0.06 ns/meV) gives a total lifetime change of the fast decay component of about $0.06 \times 1.8 \approx 0.1$ (ns) within the FWHM. However, as we will show later, one does not need to measure the exciton lifetime of QW's with different thicknesses in order to obtain the change rate of exciton lifetime with respect to well thickness, $d\tau/dL$. A much easier way to obtain $d\tau/dL$ is to measure the peak position shift as a function of delay time.

We studied the temporal response of the luminescence at the excitonic transition peak as shown in Fig. 5. The circles are the experimental values which have been deconvoluted to account for the temporal response of the
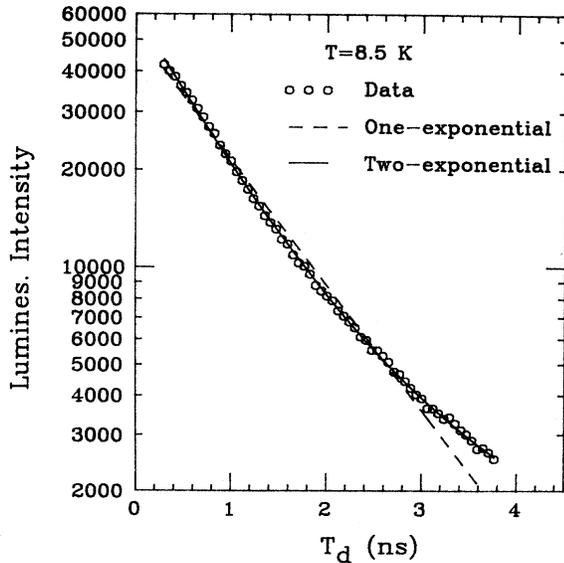

FIG. 5. Luminescence of heavy-hole exciton as a function of delay time. The circles show the measured values while the dashed and solid lines are fit using one- and two-exponential decay models, respectively (see text).

detection system. The rising part of the luminescence is not shown here. The dashed and solid lines are the theoretical fit obtained using one- and two-exponential decay models, respectively. From Fig. 5 it is clear that the decay of heavy-hole excitons cannot be described by a single exponential form. The two-exponential decay fits the experimental results very well. The decay time constants obtained from Fig. 5 are 0.9 and 4.2 ns, respectively, for the fast and slow components. This two-exponential decay behavior has been observed recently by other groups.[30-32]

Following the discussions presented above, $d\tau/dE$, the time-constant change rate with respect to emission energies E obtained from the peak shift, can be used to deduce the lifetime of the exciton in QW's of different well thickness, since we knew from previous work that the lifetime of the exciton increases almost linearly with well thickness.[26,27] From $\alpha = d\tau/dE = (d\tau/dL)/(dE/dL)$, we have

$$\frac{d\tau}{dL} = \alpha \frac{dE}{dL} . \tag{10}$$

Here

$$E = E_g(\text{GaAs}) + E_e + E_h - E_{\text{bind}} , \tag{11}$$

with $E_g(\text{GaAs})$ being the energy gap of GaAs material. E_e (E_h) is the confinement energy of the electron (hole) in the QW which is measured from the bottom of the conduction (top of the valence) band. E_{bind} is the binding energy of the exciton. By noting that the change in the binding energy of the exciton with respect to well thickness L, dE_{bind}/dL, is much smaller than the change in confinement energy of the electron or hole, we can neglect dE_{bind}/dL. In addition, the confinement energies of the ground-state electron and hole (of the order of 10 meV for our sample) are much less than those of the conduction and valence potential barriers (a few hundred meV here). Thus we can estimate E_e and E_h by treating electrons and holes as particles bound inside an infinitely deep QW. Therefore we have

$$E_e + E_h = \frac{\hbar^2\pi^2}{2m_e^* L^2} + \frac{\hbar^2\pi^2}{2m_h^* L^2} = \frac{\hbar^2\pi^2}{2\mu^* L^2} . \tag{12}$$

Here L is the average well thickness and $\mu^* = m_e^* m_h^*/(m_e^* + m_h^*)$ is the effective reduced mass of the exciton. From Eqs. (10)–(12) we obtain

$$\frac{d\tau}{dL} = -\frac{\hbar^2\pi^2}{2\mu^* L^2}\frac{2}{L}\alpha . \tag{13}$$

We used $m_e^* = 0.067m_e$ and $m_h^* = 0.45m_e$ for the electron and hole effective masses inside the QW,[33,34] where m_e is the effective mass of electron in the free space. With well thickness parameters of $L = 250$ Å and $\alpha_1 = -0.06$ ns/meV obtained in the above we find

$$\frac{d\tau}{dL} = 2.6 \times 10^{-3} \text{ ns/Å} .$$

This value is in good agreement with those extrapolated from the experimental results of Ref. 26, i.e., $\sim 2.5 \times 10^{-3}$ ns/Å. Here we want to indicate that in Ref.

26 the authors defined the time at which the intensity has dropped to $1/e$ of its maximum value, $\tau_{1/e}$, as the lifetime of the exciton. The real lifetime of the exciton radiative recombination will be different from this value. Nevertheless, $d\tau/dL$ deduced from their experimental results will be very close to the real value. Since exciton lifetime is approximately proportional to well thickness, one can deduce the exciton lifetime for different well thicknesses by measuring τ and $d\tau/dL$ for one quantum-well thickness.

IV. CONCLUSIONS

We have studied the effect of interface roughness on the time-resolved photoluminescence of the excitonic transition of GaAs-Al$_x$Ga$_{1-x}$As multiple QW's. We found that the energy position of intensity maximum of the excitonic transition shifted as the delay time increased. Based on a model involved interface roughness in the QW's and the form of a two-exponential decay for luminescence, we calculated the peak position as a function of delay times. The calculated results are in good agreement with experimental observations. From these measurements, we obtained the change rate in exciton lifetime with respect to the well thickness L. We showed that this is a very effective method to measure $d\tau/dL$ compared with the usual procedure of measuring exciton lifetimes at different well thicknesses directly. From the above discussions we see that besides the linewidth broadening and Stokes red shift, the interface roughness also strongly affects the dynamic process of the optical transition. The results obtained here are very important for fundamental research and also very useful for practical applications, such as narrowing the spectral and time pulse widths of QW lasers.

Obviously, the effect of QW thickness fluctuations on the dynamic process of optical transitions is very important. There are still many important questions to be answered. One of the most important questions here is why the decay of the exciton transition is composed of a two-exponential form. In future work, we will study the dynamic process of the optical transition by using QW's with different well thicknesses and at different temperatures. We hope that these studies will provide more understanding of the complicated effects of interface roughness.

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