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Dynamics of exciton transfer between the bound and the continuum states in GaAs-$Al_xGa_{1-x}As$ multiple quantum wells

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Experimentally observed two-exponential decay of excitonic transitions in GaAs-$Al_xGa_{1-x}As$ multiple quantum wells has been successfully interpreted in terms of the exciton transfer between the continuum (free carriers) and the bound states. The calculation results obtained from this exciton-transfer model are in excellent agreement with experimental observations. The rates of the exciton transfer and the free-carrier recombination have been obtained. We have demonstrated that the emission-energy dependence of the decay time constant of the slower decay component is caused by the variation in exciton binding energy induced by interface roughness in the quantum wells.

Optical properties of semiconductor quantum wells (QW's) and superlattices (SL's) have been intensively investigated during the past few years because of their novel properties.[1-3] The decay of excitonic transitions in QW's was found to show exactly two-exponential behavior.[4-6] Nevertheless, the physical origin of this two-exponential decay and the dynamic processes of excitons in QW's are not yet well understood. In this paper, the origin of the two-exponential decay of excitonic transition in a QW has been studied by using time-resolved photoluminescence. We demonstrate that the origin of the two-exponential decay of excitonic transitions in QW's is caused by exciton transfer between the bound and the continuum states (free carriers) via acoustic phonon-exciton (phonon-free carriers) interaction.

The sample used for this study was a GaAs-$Al_{0.5}Ga_{0.5}As$ MQW which was grown by molecular-beam epitaxy on a GaAs(100) substrate without growth interruption. It consists of alternate 250-Å well layers and 278-Å $Al_{0.5}Ga_{0.5}As$ barrier layers with a total of ten periods. Experimental details have been described previously.[6]

Figure 1 shows a semilogarithmic plot of temporal responses of photoluminescence of a GaAs-$Al_{0.5}Ga_{0.5}As$ MQW measured at three representative emission energies around the heavy-hole exciton transition peak. The inset shows the time-integrated emission spectrum with heavy-hole and light-hole exciton luminescence peaks at 1.5253 and 1.5297 eV, respectively. These values are consistent with those calculated by using the transfer-matrix method with the conduction-band offset parameter being 0.65 and the binding energies of heavy- and light-hole excitons being 5.8 and 6.2 meV, respectively.[7] The shoulder at about 1 meV below the heavy-hole exciton peak is due to either impurity-bound exciton or biexciton transitions.[8,9] It is known that the exciton emission linewidth is predominantly caused by interface roughnesses.[10] Therefore, luminescence signals measured in the vicinity of the exciton transition peak correspond to exciton recombination occurring in different QW domains.[6,10] In Fig. 1, the nonexponential decay of photoluminescence is clearly observed with different decay rates occurring at different emission energies.

Figure 2 plots the luminescence as a function of delay time t_d measured at the heavy-hole exciton transition peak. The rise part of the luminescence is not shown here and $t_d = 0$ has been chosen at the peak position in the luminescence temporal responses of Fig. 1. Crosses show the measured values while the dashed and solid lines are fittings using one- and two-exponential decay, respectively. From Fig. 2, it is clear that the decay of the heavy-hole excitons cannot be described by a single-exponential

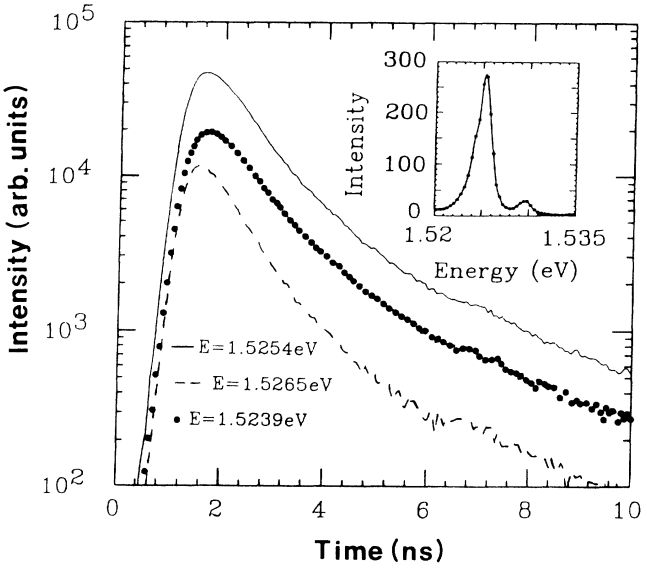

FIG. 1. Semilogarithmic plot of photoluminescence intensity vs delay time for three representative emission energies around the heavy-hole exciton transition peak of a GaAs-$Ga_{0.5}Al_{0.5}As$ multiple quantum well. Well and barrier thicknesses are 250 and 278 Å, respectively. The excitation energy was 2.125 eV with an average power density of 0.2 W/cm^2. The inset shows the low-temperature (8.5 K) time-integrated photoluminescence emission spectrum showing the heavy- and light-hole exciton emission bands.

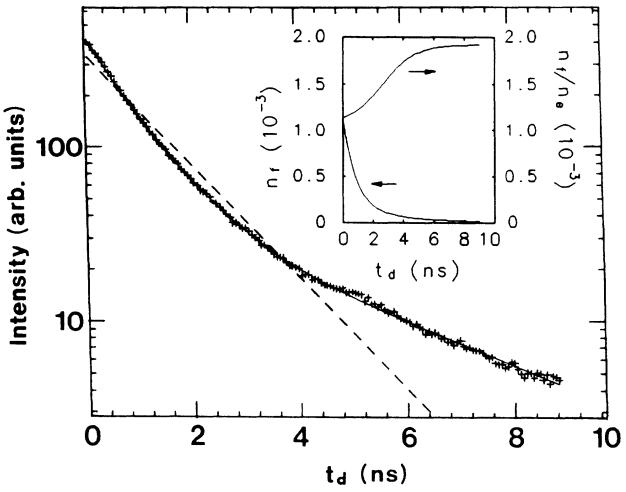

FIG. 2. Luminescence as a function of delay time t_d measured at the heavy-hole exciton transition peak. Crosses show the measured values while the dashed and solid lines are fittings using one- and two-exponential decay, respectively. $t_d=0$ has been chosen at the peak position in the luminescence temporal responses of Fig. 1. The inset shows the relative free carrier population n_f and the ratio of n_f to the exciton population n_e as functions of delay time t_d. n_f+n_e has been normalized to unity at $t_d=0$.

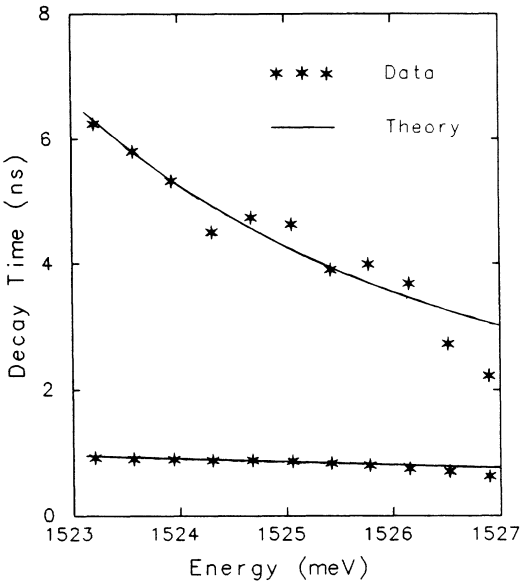

FIG. 3. Decay time constants of photoluminescence vs emission energies in the emission band of the heavy-hole exciton luminescence. The asterisks represent the experimental measured τ_1 (lower values) and τ_2 (higher values). The solid line of τ_1 is a least-squares fitting (see text). The solid line for τ_2 is the calculation results.

form. However, the two-exponential decay fits perfectly with the experimental data. Therefore, in general, the decay of the heavy-hole exciton transition in QW can be written as

$$I(t)=A\exp(-t/\tau_1)+B\exp(-t/\tau_2)\,, \tag{1}$$

where τ_1 and τ_2 are the two decay time constants representing faster and slower decay components and are about 0.9 and 4.2 ns measured at the heavy-hole exciton transition peak, respectively. Furthermore, decays of the exciton recombination in entire heavy- and light-hole exciton emission bands are also exactly two exponential. We have obtained the two decay time constants as functions of emission energies in the heavy-hole exciton emission band shown in Fig. 3. We see that τ_1 to be shown mainly due to the radiative decay of excitons depends only weakly on the emission energies. In contrast, τ_2 depends strongly on the emission energies and decreases monotonically with increase of emission energy from 6.4 ns at 1.5232 eV to about 2.2 ns at an energy of 1.5268 eV. We want to indicate here that the time constants of the slow decay component at the lower-energy side are even larger than the exciton lifetime in the bulk GaAs (3.3 ns).[11]

It is known that at room temperatures, optical nonlinearities in absorption spectra of GaAs-$Al_xGa_{1-x}As$ multiple and single quantum well structure under high laser excitation intensity is due to the ionization of excitons after their creation,[12,13] and the transfer between the bound and the continuum states becomes important.[14] The excited states of excitons (including the continuum states corresponding to the ionization threshold of excitons) in QW's have also been observed at low temperatures under low laser intensity excitation.[15,16] In this Rapid Communication, we interpret that even at low temperatures excitons are not only occupying the bound states of lowest energies, but also higher dissociated states (ionized excitons or free carriers) due to thermal ionization. Here, we concentrate on the behavior of the free exciton decay after $t_d=0$, which is about 500 ps after laser excitation. It is known that hot carriers initially generated by above band-gap excitation relax to the bottom of the subband and thermally distributed to generate excitons within about 200 ps at 1.8 K,[17] and thus our system is in thermal equilibrium at $t_d=0$. Under the above consideration, the rate equations for the exciton populations in the bound and the continuum states at delay time $t_d>0$ can be written as

$$\begin{aligned}\frac{dn_e}{dt}&=-\gamma_e n_e-Un_e+Dn_f\,,\\ \frac{dn_f}{dt}&=-\gamma_f n_f-Dn_f+Un_e\,,\end{aligned} \tag{2}$$

where n_e and n_f denote, respectively, the exciton population in the bound state ($1S$ ground state) and the free-carrier population in the conduction band. γ_e and γ_f are the recombination rates of excitons and free carriers. $U(D)$ is the rate of exciton transfer from the bound (continuum) to the continuum (bound) states. In writing Eq. (2), we have included all other excited states into continuum states since experimentally observed transition from excited $2S$ exciton state is corresponding to the continuum edge and the transfer time of the continuum to excited states is expected to be negligible. Effect due to impurity-bound excitons or biexcitons have been neglected since their populations are very small in our case. The solution of Eq. (2) is an exact two-exponential form of Eq.

(1) with

$$\tau_{1,2}^{-1} = \tfrac{1}{2}\{\gamma_e+\gamma_f+D+U \pm [(\gamma_e+U-\gamma_f-D)^2 + 4DU]^{1/2}\} . \quad (3)$$

In order to compare with experimental results, we need to know two more quantities among four unknowns, γ_e, γ_f, U, and D. Exciton transfer rates U and D can be calculated by using quantum theory and consider that the exciton transfer process is assisted by emission or absorption of acoustic phonons. D and U thus can be written as

$$D = (2\pi/\hbar)\sum_{\mathbf{k}_1}\sum_{\mathbf{k}_2}\sum_{\mathbf{q}} |M|^2 f(\mathbf{k}_1)[n(\mathbf{q})+1] \times \delta(\mathbf{k}_1-\mathbf{k}_2-\mathbf{q})\delta(E_1-E_2-\hbar\omega) , \quad (4)$$

$$U = (2\pi/\hbar)\sum_{\mathbf{k}_1}\sum_{\mathbf{k}_2}\sum_{\mathbf{q}} |M|^2 f(\mathbf{k}_2)n(\mathbf{q}) \times \delta(\mathbf{k}_2-\mathbf{k}_1+\mathbf{q})\delta(E_2-E_1+\hbar\omega) .$$

Here M is the matrix element depending on the interaction, $n(q) = \{\exp[\hbar\omega(\mathbf{q})/kT-1]\}^{-1}$ is the probability of finding an acoustic phonon of wave vector $\mathbf{q}$ at temperature T, and $f(\mathbf{k}_1)$ the probability of the initial exciton state being occupied has been assumed to be a Boltzmann distribution. E_1 and E_2 are the exciton energy and $\mathbf{k}_1$ and $\mathbf{k}_2$ are the two-dimensional wave vectors of excitons in the plane perpendicular to the growth z axis in the initial and final states, respectively. Here the bound state presents initial (final) state for $U(D)$. The detailed calculation procedure will be published in a forthcoming paper.[18] However, an important result obtained is that D and U are related by the following expression:

$$D = U\exp(\Delta E/kT) , \quad (5)$$

where ΔE is the energy difference between the continuum and the bound states, which is the binding energy of exciton E_b. With an approximation of the matrix element M being a constant and independent of $\mathbf{k}_i$ $(i=1,2)$, from Eqs. (3)–(5), we can calculate τ_1 and τ_2.

The key factor which causes τ_2 to be strongly dependent on emission energy is the emission-energy dependence of the exciton binding energy $\Delta E(=E_b)$ as a result of interface roughnesses. The observed exciton emission linewidth corresponds to a well width fluctuation of about 20 Å at 250 Å, we therefore use an approximation of a linear relation for E_b as a function of well thickness L, $E_b(L) = E_b(L_0) - \alpha(L-L_0)$, where $L_0 = 250$ Å is the average well thickness and α is about 0.012 meV/Å in this region of L.[19] We also use an expression, $\tau_1 = \tau_0 - \beta(L-L_0)$, to account for the changing of the radiative recombination rate with L, where $\beta = 3.08\times10^{-3}$ ns/Å and $\tau_0 = 0.85$ ns, which are consistent with those deduced from the exciton transition peak position shift with delay time.[6] However, taking τ_1 as a constant will not alter the behavior of τ_2. Strong dependence of τ_2 on emission energies is a direct consequence that the transfer rates D and U are functions of exciton binding energies, E_b, which depend on emission energies as a result of interface roughness. The recombination rate of free carriers (exciton in the continuum states) γ_f is assumed to be independent of well thickness in this region. τ_2 as a function of emission energy has been calculated and the result is plotted as a solid line in Fig. 3, which is in good agreement with experimental data. The best fitting between experimental data and calculation yields $\gamma_f^{-1} = 20$ ns consistent with the value obtained previously[20] and the matrix element $M = 1.39\times10^{-9}$ meV. Based on these results, the physical origin of the two exponential decays have been completely resolved. The fast decay rate is mainly due to the radiative recombination of excitons. The slow decay component is determined by the recombination rate of free carriers and the rates of exciton transfer between the bound and the continuum states.

At $L = 250$ Å, the transfer rates U and D are 7.52×10^4 and 2.06×10^8 s^{-1}, respectively. Although the transfer rate of D is 3 orders of magnitude larger than that of U, the absolute numbers of excitons transferred between the bound (continuum) and the continuum (bound) states are compatible, since the population of excitons in the bound state (n_e) is about 3 orders of magnitude larger than those in the continuum state (n_f) in our case, as shown in the inset of Fig. 2. In the inset of Fig. 2, we plot the variations of n_f and n_f/n_e as functions of delay time t_d. Here, n_e+n_f has been normalized to unity at $t_d = 0$. We see that the relative population of excitons in the continuum state (free carriers) increases with the increase of t_d. This is caused by the fact that the transfer rates U and D, as well as the free-carrier recombination rate, are much smaller than the exciton radiative recombination rate in the bound state, which also leaves τ_1 being hardly affected by the exciton transfer between the bound and the continuum states. Another point we want to indicate is that the population of excitons in the bound and the continuum states are in thermal equilibrium only at $t_d = 0$ due to the transfer rate being smaller than the radiative decay rate in the bound state. The discrepancy between the calculated and experimental results of τ_2 in the high-energy side of Fig. 3 is caused by the luminescence intensity overlap between the heavy- and light-hole exciton transitions. The absence of the transition line from the free carriers, especially at high temperatures, may be due to the free carrier recombination rate being much smaller than the transfer rate D, yet remains to be investigated.

In conclusion, the origin of two-exponential decay of excitonic transition in GaAs-$Al_xGa_{1-x}As$ MQW's has been investigated. Our results demonstrate that the two-exponential decay of exciton transition is a direct consequence of the exciton transfer between the bound and continuum states (free carriers) via acoustic phonon-exciton (phonon–free carriers) interaction. The recombination rate of free carriers as well as exciton transfer rates are obtained.

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