TEMPERATURE DEPENDENT TIME-RESOLVED EXCITON LUMINESCENCE IN GaAs/AlgaAs QUANTUM WIRES AND DOTS

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Transient photoluminescence of GaAs/AlGaAs quantum wires and quantum dots formed by strain confinement is studied as a function of temperature. At low temperature, luminescent decay times of the wires and dots correspond to the radiative decay times of localized excitons. The radiative decay time can be either longer or shorter than that of the host quantum well, depending on the size of the wires and dots. For small wires and dots (~ 100 nm stressor), the exciton radiative recombination rate increases due to lateral confinement. Exciton localization due to the fluctuation of quantum well thickness plays an important role in the temperature dependence of luminescent decay time and exciton transfer in quantum wire and dot structures up to at least ~ 80 K. Lateral exciton transfer in quantum wire and dot structures formed by laterally patterning quantum wells strongly affects the dynamics of wire and dot luminescence. The relaxation time of hot excitons increases with the depth of strain confinement, but we find no convincing evidence that it is significantly slower in quasi 1-D or 0-D systems than in quantum wells.

1. Introduction

Although the precise definition of the radiative decay time for a free exciton in a bulk semiconductors is still a matter of dispute[1-3], much work has been done to determine the radiative decay time in quasi two dimensional (2-D) systems such as GaAs/AlGaAs quantum wells[4-6]. Work has also been extended to lower dimensional systems, including GaAs/AlGaAs quantum wires (1-D)[7-12] and quantum dots (0-D)[11-14]. The decay rate of the luminescence is not necessarily the true radiative decay time because of various other dynamical processes[4-6]. To obtain the radiative decay time from the measured luminescence decay, careful analysis is required, and the luminescence decay rate can be helpful in understanding other properties of semiconductors, such as nonradiative recombination and exciton localization.

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There have been several time-resolved studies of the luminescence of GaAs/AlGaAs quantum wires and dots at low temperature [7-9,11], and one as a function of temperature[10]. In most cases[7-9,11,12], the observed decay rate was interpreted as the intrinsic radiative decay of the free excitons, and the possible effect of exciton localization was not considered, even though localization is known to be very important in quantum wells[4,6]. In the low temperature limit, the luminescent decay times in various types of wires and dots were found to be of the same order of magnitude as in quantum wells[8-10,11]. Nevertheless, it was found that when nonradiative decay is suppressed, as in "V-groove" wires[10] and in strain confined quantum dots[11], the luminescence decay rate is reduced relative to the corresponding quantum well. The decrease has been ascribed to slower carrier relaxation[13]. However, the decay rate can also be reduced by the reduction in spatial coherence in the lateral direction(s)[12,14], which is sufficient to overcome the effect of the increased electron-hole overlap due to lateral confinement[12]. Furthermore, in strain-confined dots and wires the electron-hole overlap may be reduced rather than increased, because strain can spatially separate the electron and hole. The slower decay in 20 nm diameter InGaAs/GaAs quantum dots has been attributed to this[15].

If we assume that the excitons are free, with a Boltzmann distribution in a single parabolic band, the decay time in a quantum wire varies as T^{1/2}[12], as a consequence of the 1-D density of states. This temperature dependence was reported for the "V-groove" quantum wire of ref.[10]: however, there are problems in the analysis of those data, which will be discussed in Section 4. In 20 nm InGaAs/GaAs quantum dots the luminescence decay time was found to be independent of temperature up to 30 K[15]. This was explained as the effect of 0-D density of states with large sublevel spacing due to strong confinement.

In our previous report on a 350 nm strain confined quantum wire[16], we found that the luminescence decay times showed a different temperature dependence from the parent quantum well, but not the temperature dependence given by ref.[12]. While the sublevel spacings in this wire (~1meV) are too small for the 1-D theory to hold, our data showed that the exciton localization was important in determining the temperature dependence of the wire decay time.

Another relevant process is energy transfer from the adjacent 2-D region[9,16,17]. We have found that thermally activated transfer significantly reduces the decay time in the 2-D region[16], and suppresses nonradiative recombination in this region, so that the overall luminescence efficiency is higher in strain confined quantum wire and dot structures than in quantum wells[17].

In our previous work[18,19], the mechanism of strain confinement has been discussed in detail. A

systematic study of the temperature dependence of the steady-state luminescence is in press[17]. In this paper, we report an investigation of the time-resolved luminescence after pulsed excitation of strain confined quantum wires and dots, as a function of temperature from 4K to 80K. Wires and dots based on different host quantum wells and with different lateral confinement scales are used. It is important to note that although the stressors that produce the confining strain are rather large (100-800 nm) by the standards of quantum confinement, the width of the exciton wave function in its ground state is much less, typically of order 20 nm, and the energy level spacings are 1-4 meV[18,19].

In this paper we show that at low temperature the luminescent decay times in our wires and dots are mainly determined by the radiative decay of localized excitons. The decay time in a wire or dot made on a given quantum well tends to increase with lateral scale, but it may be either shorter or longer than the decay time of the host quantum well, depending on the specific confinement potential. With increasing temperature, the decay time increases, as it does in the parent quantum well, but the temperature dependence is different. The difference from the quantum well is greater in a quantum dot than in the same sized wire. Transfer from quantum well to quantum wire or dot is thermally activated, and the transfer time becomes comparable to the exciton decay time at 50-80K. However, exciton localization by interface fluctuations plays an important role over the entire temperature range. The transient results are consistent with the temperature dependence of the steady-state luminescence in the same samples[17].

160 nm thick amophous carbon wire

30 ı	ım GaAs	cap	
20 n	m AlGa	As barri	ег
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20 n	m AlGa		ег
	2 nm Ga	As well	
20 n	m AlGa	As barri	er
	4 nm Ga	As well	
20 n	m AlGa	As barri	er
500 nm	GaAs b	uffer lay	/ег
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100 nm thick amophous carbon wire or dot

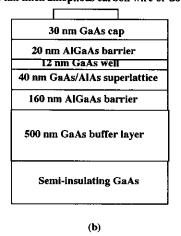


Fig. 1. Schematic cross sections of strain confined wire or dot structures (not to scale), (a) for sample A, (b) for sample B. "AlGaAs" means Al_{0.3}Ga_{0.7}As, and the superlattice in sample B has the same Al/Ga ratio.

(a)

2. Samples and Experimental Setup

Two samples, labelled A and B, are used in this work. Their structures are shown in Fig.1. The host quantum wells were grown by molecular beam epitaxy on [100] oriented semi-insulating GaAs. Briefly, the quantum wires or dots were made in the following way[20]: on the top of a quantum well structure, an amorphous carbon layer was patterned and etched to form 40-50 µm square arrays of wires or dots. These carbon wires or dots are called stressors because they strain the semiconductor underneath. The wires are strips along the [100] direction, while the dots are squares with sides in the same orientation. In sample A, whose host structure consists of three GaAs wells of 9, 2 and 4 nm thickness, there are two arrays of wires, labelled here W2 and W3. Both have wires 350 nm wide, but with period 1μm and 2μm respectively. We will use the label A-W3(350nm/9nm) for the spectrum from the 9 nm quantum well under the wire array W3(350 nm wire width) in sample A, and so on. Dots are similarly labelled with D replacing W. Sample B has a single 12 nm GaAs well, and five arrays of wires and five arrays of dots with widths 160, 200, 320, 460 and 780 nm. The corresponding periods are 300, 600, 900, 1200 and 2400 nm. More details of the sample fabrication are given in ref.[20].

A time-resolved single photon counting system was used to measure the transient photoluminescence. The

system consists of a mode-locked 5145 Å argon laser giving 100 ps pulses at 81 MHz, a 1-m double grating monochromator, a multichannel plate photomultiplier (Hamamatsu R1564U-01), a timing amplifier (Ortec 574), constant-fraction discriminators (Ortec 934), a time-to-amplitude converter (Ortec 457 Biased Time to Pulse Height Converter), and a multichannel analyzer (Hewlett Packard 54-22A/16B/10A). Transient luminescence data were taken at 47 ps/channel resolution on the multichannel analyzer.

The laser was focussed to a spot of ~12 µm diameter centered on the 40 µm x 40 µm patterned area. A small and uncontrolled fraction of the exciting light leaked to the surrounding unpatterned area, and the contribution from this area produces scatter in some of the data on the time decay of the luminescence from the quantum well in the patterned region. The average excitation power was typically 20 µW, or 0.25 pJ/pulse, corresponding to an incident density of 18 W/cm² or 0.22 µJ/cm²/pulse. The absorption coefficient of Alo.3Gao.7As at 5145Å is about 5x10⁴ cm⁻¹, so about 10% of the exciting light is absorbed in the 20 nm barrier layer, producing a carrier density ~5x10¹⁰ cm⁻². However, not all this excitation reaches the quantum well, and the excitation density in the well was low enough to avoid any nonlinear effects[21] except as noted below.

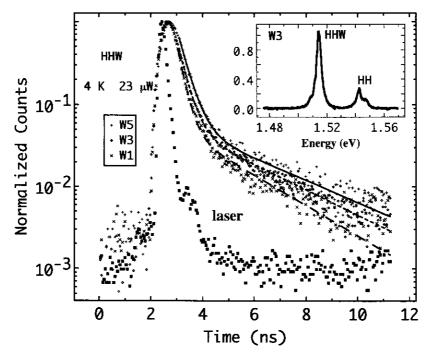


Fig.2. Transient luminescence of the heavy hole excitons in quantum wires of different widths in sample B (12 nm well). W1: 100nm; W3: 300nm; W5: 800nm. Insert is the luminescence spectrum of W3.

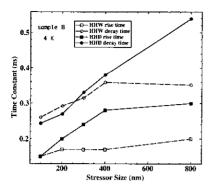


Fig.3. Rise and decay times of wire (HHW) and dot (HHD) excitons in sample B at 4 K, as a function of stressor width. The rise and decay times for the unpatterned quantum well are 150 ps and 320 ps respectively.

3. Experimental Results

3.1. Low Temperature Measurements

First, we present the stressor size dependence of the luminescence rise and decay times, and the effect of exciton transfer, for our quantum wires and dots at about 4K. Fig. 2 shows the transient luminescence of the heavy hole (HH) excitons in quantum wires of different widths in sample B. The insert shows the photoluminescence spectrum measured at the same time. HHW stands for the exciton in the wire and HH for that in the quantum well between wires. The decay curves were taken at the peak of the HHW line. Similar data are obtained from quantum dots (HHD). As in the cases of quantum wells, the decay is generally nonexponential. The three-exponential form below (Eq.1) is convolved with the system response (obtained from the measured laser pulse) to fit the data.

$$I(t) = a_1 e^{-t/\tau_0} + a_2 e^{-t/\tau_1} + a_3 e^{-t/\tau_2}$$
(1).

In (1) τ_0 is the rise time, τ_1 the fast decay time, τ_2 the slow decay time, and a₁, a₂ and a₃ are constants, with $a_2+a_3=$ - $a_1.$ The fitted values of $\,\tau_0\,$ and $\,\tau_1\,$ are shown as a function of wire or dot size in Fig.3. We see that with increasing size, both τ_0 and τ_1 increase. Because the magnitude of the strain at the center of the wire or dot increases with size of stressor, there is a similar correlation of τ_0 and τ_1 with the redshift of the exciton. In general, the changes in decay time and rise time from their values in the unpatterned quantum well are found to be greater in a dot than in a wire with the same stressor size. We see from Fig. 3 that in the wires the risetime is not significantly different from the unpatterned value, while the decay times approach the unpatterned values at large stressor width. For dots, on the other hand, they are both substantially longer.

The slow decay component τ_2 (typically $\sim 1\text{--}2$ ns) could be due to impurity related luminescence[22] or

exciton migration among localized sites[23], and will be discussed later. It is weaker in the wires and dots than in the parent quantum well.

We discuss elsewhere mechanisms[17] for the lateral exciton transfer: tunneling and "capture" at low temperature, and thermal activation at higher temperature. Strain confinement produces a potential barrier separating the wire or dot region from the surrounding well region. The barrier for a 350 nm wire, for example, is found experimentally to be about 3.5 meV high and 100 nm wide[17]. With above band gap excitation, hot carriers or hot excitons generated in the well region can be captured into the wire or dot before they relax to the lowest quantum well state. Once they are fully relaxed, the probability of tunnelling through the barrier is small. As a result, the decay time of HH in the patterned quantum well at low temperature is the same as that of the unpatterned quantum well[16]. However, for small wires or dots, both the barrier height and thickness are smaller, and transfer by tunneling is significant even for fully relaxed excitons. Fig.4 shows that the decay of HH is indeed faster in a 100 nm dot than in the unpatterned quantum well. A similar result is found for a 100nm wire.

Another distinction is that quantum dots are more easily saturated than wires. Fig.5 shows the time resolved spectra of B-D2(200nm/12nm), with excitation density ten times what we normally used. At time t = 560 ps (measured from the peak of the laser pulse), the dots are saturated and "hot luminescence" from excited states is seen. There is no corresponding change in the quantum well luminescence (HH) at this excitation density. Since the risetimes are almost the same, the hot luminescence cannot be due to slow relaxation of the hot excitons, but must be due to filling of the low energy states. At a later time, 2.24 ns from the peak, the hot excitons have relaxed to the low energy states, so that these states appear to have a longer lifetime. Similar effects are seen in unpatterned quantum wells, but at higher excitation density[21]. The decay of the patterned quantum well luminescence is slower than that of the hot luminescence of the dot, which is just below it in energy, confirming that at low temperature the relaxed excitons in the quantum well are not in communication with states in the dot.

3.2. Temperature Dependence

With increasing temperature, the exciton decay rate in the unpatterned quantum wells decreases initially, but then increases when the luminescent efficiency drops due to nonradiative recombination. The same trend is found for excitons in the quantum wires and quantum dots, but with different temperature dependences for each dimensionality. Fig.6 shows the transient photoluminescence from excitons in the wire in sample A-W3(350nm/2nm) at various temperatures. Similar measurements were made on both the quantum well and the wire or dot luminescence in all the samples, and were fitted to a convolved tri-exponential as in the previous section. The fitted fast decay times τ_1 are shown as a function of temperature in Figs.7(a)-(c) for the different wires and dots, together with the corresponding decay times in the unpatterned quantum well (open circles)[24].

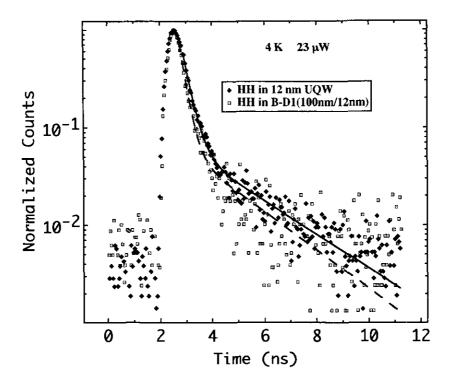


Fig.4. Time decay of the HH exciton in the unpatterned quantum well (UQW: full line) and in the quantum well between 100 nm dots (B-D1: dashed line). Background has been subtracted.

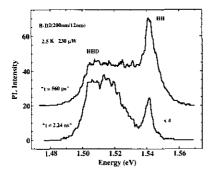


Fig.5. Time-resolved spectra of B-D2(200nm/12nm) at two delay times (time is measured from the peak of laser pulse).

Note that because the focusing and positioning of the laser spot is not perfect, luminescence from the adjacent unpatterned area may contribute to the HH peak supposedly from the inter-wire region. This can cause error and scatter in the decay time.

At high temperature, the decay times are all longer in the wire or dot than in the unpatterned quantum well.

One obvious reason is that nonradiative recombination is suppressed to some degree in the wires and dots[17]. The degree of suppression is always greater in the dots and depends on the width of the host quantum well, so that onset of nonradiative recombination occurs at a lower temperature for wider quantum wells, in agreement with the CW result[17] that the quantum efficiency drops off at a lower temperature in a wider quantum well.

The slow (τ_2) component gets weaker as the temperature is raised, as expected if it is related to excitons bound to impurities[22], or to transfer amongst localized sites [23].

Fig.8 shows the time decay of the exciton in the wire (HHW), in the well between the wires (HH), and in the unpatterned quantum well (UQW), for sample A-W3(350nm/9nm) at 72K. All three decay times are similar at 4K, but they differ greatly in their temperature dependence. Comparison of the patterned with the unpatterned quantum wells shows the effect of transfer: the decay time in the patterned quantum well is reduced at high temperature because of thermally activated exciton transfer to the wires. In A-W3(350nm/9nm), a significant difference due to transfer is seen at ~50 K, whereas in A-W3(350nm/2nm) a higher temperature is needed. This is consistent with the CW result[17] that thermally activated

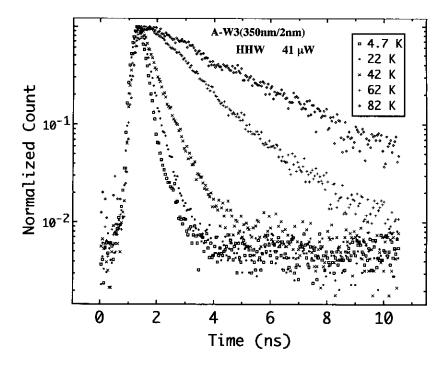


Fig.6. Temperature dependence of the transient luminescence of the heavy hole exciton in a 350 nm quantum wire.

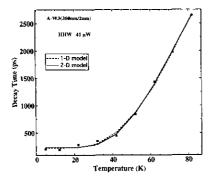
exciton transfer has a much lower onset temperature in A-W3(350nm/9nm) than in A-W3(350nm/2nm). The difference is qualitatively what one would expect from the stronger exciton localization due to well width fluctuations in the narrower quantum well.

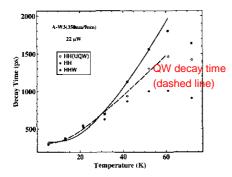
Exciton transfer from the well region to the wire or dot also affects the rise time of the wire or dot luminescence. To describe the contribution of the transfer accurately, extra terms should be added to (1)[16]. For simplicity, we fit the luminescence to (1), which yields effective rise and decay times. The effective decay time should be reasonably close to the true exciton decay time in the wire or dot, since at low temperature the transfer is weak while at high temperature the decay time is determined from the long time behavior, when transfer is complete. On the other hand, at high temperature the transfer contributes strongly to the photoluminescence and will affect the rise time, while at low temperature transfer is not important and the rise time is primarily determined by hot carrier or exciton relaxation. Capture transfer (transfer of excitation from well to wire or dot before thermalization in the well) probably has a time constant similar to that of the direct excitation to the wire or dot, since the barrier is very low compared to the energy difference between the exciting and emitted photons. This is illustrated in Fig.9, which shows the effective rise time (i.e. τ_0 in the fit to Eq. 1) as a function of temperature for A-W3(350nm/9nm). Up to about 30 K, direct excitation or capture transfer from the inter-wire region is dominant and τ_0 is determined by the relaxation rate of hot excitons, which decreases with temperature as in an unpatterned quantum well[25]. Above 30 K exciton transfer is significant in this sample, as is shown by the CW data[17], and τ_0 increases, being now partially determined by the lifetime of excitons in the well which is much longer than the relaxation time.

4. Discussion

4.1. Exciton Decay Time in Quantum Wires and Quantum Dots

The radiative decay time of a free exciton in a quantum well is reduced from the bulk value[3,26], because the extra confinement enhances the electron-hole wave function overlap[26]. Thus one might expect the decay time to decrease still further in quantum wires or dots. On the other hand, Citrin[12,14] argues that the competing effect of finite spatial coherence in quantum wires and dots produces a net increase in decay time. Citrin calculates a radiative decay time of 150 ps at 0 K for the ${\bf k}=0$ exciton in a 10 x10 nm GaAs/AlGaAs quantum wire[12] and 107 ps for a dot with 20x20x10 nm quantum dot[14], compared to 25 ps calculated for the parent 10 nm quantum well[12,14]. However, in general this calculation does not give the actual luminescence decay





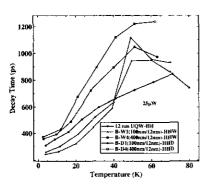


Fig.7. Temperature dependence of the heavy hole exciton decay times in unpatterned quantum well (UQW), quantum wires and dots. In (a) and (b), the lines are fitted to (5).

(a) A-W3(350nm/2nm). Fits to quantum wire data (HHW). Full line: n = 2; dashed line: n=1.

(b) A-W3(350nm/9nm). Full line: fit to quantum wire data (HHW) with n = 2; dashed line: fit to unpatterned quantum well data (UQW) with n = 2.

(c) B-W1(100nm/12nm), B-D1(100nm/12nm), B-W4(400nm/12nm) and B-D4(400nm/12nm). Lines in (c) are guides to the eye.

time because of various complications such as exciton localization[4-6]. In particular, Citrin has argued[4] that since only k=0 excitons can decay radiatively, well width

fluctuations will increase the exciton radiative decay time by broadening the exciton wavefunction in k-space, mimicking the effect of non-zero temperature.

In our work, exciton decay times in quantum dots[11,15] and in our relatively large wires and dots are found to be longer than in the parent quantum wells. Our wires or dots are formed by strain confinement, which tends to confine the electron and hole in separate regions, at any rate if the Coulomb interaction can be neglected[18,27]. This spatial separation of the electron and hole, and consequent reduction in the electron-hole overlap, might account for the increasing decay time with size (see Fig.3). In principle lateral confinement of the wire or dot can modify the fluctuation potential, but one would expect this effect to be most pronounced for the smallest wires, contrary to observation. Be that as it may, it is clear that the longer decay time in some quantum wires and dots cannot necessarily be attributed to the reduced dimensionality.

4.2. Carrier Relaxation in Quantum Wires and Ouantum Dots

It has been suggested[13,28] that in 1-D or 0-D systems with 100-200 nm lateral quantization, intersubband electron relaxation by acoustic phonon-electron scattering at low temperature is slow compared to that in 3-D and 2-D systems. Furthermore, Rota et al[29] pointed out that the thermalization rate due to electron-electron interactions may also be reduced in semiconductor quantum wires. On the other hand, it is possible that electron-hole correlation enhances the exciton-phonon scattering rate relative to the electron-phonon rate, at least in small quantum dots (≤ 100 nm)[30]. It has also been argued[31] that a dense electron-hole plasma in the 2-D region can permit hot electrons in 0-D states to relax rapidly by Auger processes, even if relaxation by phonon scattering alone were as slow as predicted in ref.[28]. However, while the total photo-generated carrier density in our samples is of order 5x10¹⁰/cm², most of the excitons relax quickly to 1-D or 0-D states, so that the carrier density in the 2-D states is much lower, and the Auger mechanism should not be significant in our work. In conclusion, it is not apparent from theoretical studies whether the relaxation should be slower or faster, even in ideal structures.

Experimentally, in an etched 150 nm wire[32], selectively excited hot excitons were found to have a relaxation time of 150 ps when the wire was excited at low excitation density within the lowest heavy hole exciton line. This is much slower than in quantum wells excited below the lowest subband edge[33,34], and the explanation given was that intraband exciton-exciton scattering cannot conserve both energy and quasi momentum in an ideal 1-D system. For excitation 8.5 meV above the exciton energy, where intersubband scattering is possible, the relaxation time falls to 85 ps, comparable with that in quantum wells. In another experiment on wires 40 - 200 nm wide, made by ionimplantation[8], the decay time of the hot luminescence ~12 meV above the exciton line was found to increase by ~25% from the widest to the narrowest wire. This change was attributed to a reduced energy relaxation rate in quasi 1-D systems. However, one should note that even

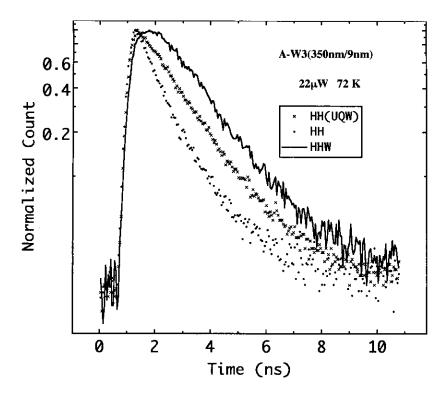


Fig. 8. Time resolved luminescence of excitons in a 350 nm wire (HHW), in the 9 nm host quantum well between the wires (HH) and in the unpatterned quantum well (UQW), at 72 K.

though the excitation density per unit length was kept constant in this experiment, one would expect narrower wires to saturate more easily than wide ones, since there are fewer sublevels within 12 meV of the lowest level. In the narrower wires, the high excitation density will tend to saturate the lower levels and hence increase the decay time of the higher levels. Thus, apparently slower energy relaxation does not necessarily imply an intrinsically slower energy relaxation rate.

In most cases, such as the quantum wire or dot structure in this work, there are two types of hot exciton relaxation: relaxation from 2-D states to 1-D or 0-D states, and relaxation within 1-D or 0-D states. Most theoretical work has focussed on the latter process. Experimentally, for excitation above the band gap, relaxation from the initial state to the luminescent state includes contributions from both types of relaxation. While one can avoid this by looking at the decay time on the high energy side of the luminescent peak, as in ref.[8], this decay is affected by filling of lower states, which can only empty by radiative decay, so it does not necessarily give the true relaxation time. A more reliable and direct way for obtaining the inter-subband or inter-sublevel relaxation time would be to use selective excitation at low excitation density within the

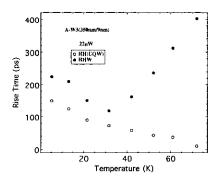


Fig.9. Rise time of exciton luminescence of excitons in a 350 nm wire (HHW) and in the 9 nm unpatterned quantum well (UQW) versus temperature.

1-D or 0-D states. In summary, out of the previous experimental work on quantum wires, only ref. [32], where the relaxation is within a single subband, provides evidence for relaxation significantly slower than in the parent quantum well.

Fig. 3 shows that at 4K in our strain-confined wires and dots, the luminescence rise time, which is a measure of the relaxation time, decreases rather than increases as the stressor size is reduced. However, the structure assumed in the theoretical models discussed above is quite different from the structures studied in this work: in particular the theoretical model assumes infinitely high and abrupt barriers in lateral directions, so that the quantum dot states are above the quantum well ground Fig. 3 shows that the relaxation time in dots increases with size; it also tends to increase with redshift (the energy difference between the exciton in the dot and in the parent quantum well), which is correlated with the width. Thus we can interpret our data in two ways. One possible interpretation is that relaxation is spatial, and the time to taken to reach the lowest state in the center of the dot increases with the distance the exciton has to move. A semiclassical model of an exciton moving in a parabolic potential well gives $\tau_0 \sim \tau_s [1 + (\omega_0 \tau_s)^{-2}]$, where ω_0 is the oscillation frequency of the exciton in the well and τ_s is the elastic scattering time. Since ω_0^{-1} and τ_s are both ~1ps, this gives too short a time, but the process could be greatly slowed down by well width fluctuations. The other, perhaps more physically realistic model, is to treat energy relaxation of excitons as a multiphonon process so that $\tau_0 = C \Delta E$, where ΔE is the energy to be dissipated and C is a constant. In quantum wells this dependence, with C ~ 5ps/meV, has been observed by selective excitation for $\Delta E < 30 \text{meV}$ [33,34]. Interpreted this way, our data suggest that the effective value of ΔE increases with redshift. If excitation were via the lowest exciton state in the quantum well, this is what one would expect. However, it has been established that except for the smallest wires and dots, excitation at low temperature is predominantly by capture, in which an exciton created in the quantum well transfers to the wire or dot before it can relax to the lowest quantum well state (see Section 3 and ref. 17). The dependence on redshift (or, what is much the same thing, on the depth of the well below the barrier) suggests that relaxation to the barrier is fast, but becomes slow once the exciton (or free carrier) is within the wire or dot with an energy below the barrier. It is not clear why this should be the case. Time resolved selective excitation measurements are now under way which may cast light on this problem.

4.3. Temperature Dependence Of Exciton Decay Times in Quantum Wires and Quantum Dots

In the absence of nonradiative recombination, the temperature dependence of the free exciton decay time has the following simple form[2,4,6,26,35]:

$$\tau(T) = \frac{\tau_{r}(T)}{r(T)} \tag{2}$$

where

$$r(T) = \frac{\int_0^{\Delta(T)} D(E) f(E) dE}{\int_0^{\infty} D(E) f(E) dE}$$
(3).

Here $\tau_r(T)$ is the exciton radiative decay time in the ground state, $\Delta(T)$ is the small energy interval (determined by k-conservation) within which excitons can decay radiatively, D(E) is the exciton density of states, f(E) is the Maxwell-Boltzmann distribution, and r(T) represents the fraction of excitons with energy smaller than $\Delta(T)$. This model is valid for any dimensionality. For a single parabolic subband or sublevel, the limit of (1) for kT >> Δ is $\tau(T) = 3\sqrt{\pi} (kT/\Delta)^{3/2} \tau_r$ in 3-D, $\tau(T) = (kT/\Delta)\tau_r$ in 2-D, $\tau(T) = [(\pi kT/(4\Delta)]^{1/2}\tau_r$ in 1-D and $\tau(T) = \tau_r$ in 0-D. Thus if τ_{r} and Δ are temperature independent, the exciton decay time varies as T^{n/2}, where n is the dimensionality. In this case the temperature dependence is determined only by the dimensionality, through the density of states, although the parameters τ_r and Δ depend on the microscopic model[4,12,35].

This ideal temperature dependence is obscured by various additional effects, such as well width fluctuations, nearby subbands or sublevels, and nonradiative recombination. Consider, for example, the measurements on calculated decay time in a 18 nm "V-groove" wire, which were reported to give $\tau(T) = C_1 T^{1/2}$ $C_1 = 92 \text{ ps/K}^{1/2}[10]$. This result is in excellent agreement with theory, which predicts $C_1 = 140 \text{ ps/K}^{1/2}$ for an ideal 10 nm quantum wire[12]. However, according to the authors' own analysis, only in the range 30-60 K was the luminescence dominated by free exciton radiative decay: nonradiative recombination becomes significant above 60 K, while the excitons are localized below 30K. Ref. 10 has only one data point, at about 40 K, which is affected neither by localization nor nonradiative recombination, and the temperature dependence obtained in ref.[10] cannot be compared with the prediction for a free exciton.

We will now describe a simple model that incorporates the effect of exciton localization in quantum wells and wires[4,36]. We assume that free and localized excitons are in thermal equilibrium with each other. The luminescent decay time (including the contribution of both free and localized excitons) is approximately

$$\tau(T) = \tau_{loc} \frac{\left(1 + \frac{N_f(T)}{N_{loc}(T)}\right)}{\left(1 + \frac{N_f(T)\tau_{loc}}{N_{loc}(T)\tau_f(T)}\right)} \tag{4}$$

where $\tau_{r}(T)$ is the free exciton decay time given by (2) and τ_{loc} is the localized-exciton radiative decay time. For an exciton of mass $M,\ N_{f}(T)=\left(MkT/2\pi\hbar^{2}\right)^{n/2}$ and $N_{loc}(T){=}N_{D}\text{exp}(E_{loc}/kT)]$, where N_{D} is the areal or lineal density of localization sites and E_{loc} is the localization energy[37]. If we write $N_{f}(T)/N_{D}=c_{1}T^{n/2}$ and $\tau_{f}(T)=c_{2}T^{n/2}$, (4) becomes

$$\tau(T) = \tau_{loc} \frac{(1 + c_1 T^{n/2} e^{-E_{loc}/kT})}{(1 + \frac{c_1 \tau_{loc}}{c_2} e^{-E_{loc}/kT})}$$
(5).

Structures	T Range	t _{loc} (ps)	¢ ₁ (K ⁻¹)	c ₂ (K ⁻¹)	E _{loc} (meV)	N _D (cm ⁻²)
9nm quantum well	< 52 K	307	0.0976	_	2.2	5x10 ¹⁰
A-W2(350nm/9nm)	< 52 K	314	0.130		3.3	1.2x10 ¹⁰
A-W3(350nm/9nm)	< 52 K	328	0.178	_	4.1	9x10 ⁹
A-W3(350nm/2nm)	< 82 K	242	2.36	63.8	16.0	6x10 ⁹

Table 1. Decay times of HH, HHW and HHD fitted to (5) in the 2-D approximation.

In our strain confined structures, the localization energy is typically somewhat larger than the intersubband splitting due to the lateral confinement, though less than the quantum well splitting. Thus at a temperature where the excitons are free, several lateral subbands will be occupied, and the 2-D case applies. In Fig.7 a and b, (5) is fitted to the data on unpatterned wells and on wires in sample A over the temperature range in which nonradiative recombination is not significant. Note that (5) is valid only in the rather small range of temperature high enough for the localized and free excitons to be in thermal equilibrium, but not so high that free excitons thermally disassociate or occupy higher quantum well subbands, or that nonradiative recombination becomes significant. The fitting parameters, given in Table 1, are of the same order of magnitude as found in other studies [38,39]. For the 9 nm well and the wire based on it, $(N_t \tau_{loc})/(N_{loc} \tau_t) \ll 1$ for the temperature region where measurements were taken. In consequence free exciton states act only as "shelf" (nondecaying) states so that the luminescence is entirely from localized exciton states. The second term in the denominator of (5) is then negligible, and the data can give no information about the temperature dependence of the decay time of free excitons. In the 2 nm well (Fig 7a) this is not the case, but the fit is equally good for n = 1 or n = 2 (since the difference is only the pre-exponential factor) and the precise temperature dependence of the free exciton decay time cannot be distinguished.

With the same caveat, (5) can also account for the data on the wires and dots in sample B (Fig.7c) but a quantitative fit is not possible to the unpatterned quantum well because nonradiative recombination is present throughout the relevant temperature range.

The fitted localization energy for the 2 nm quantum well is unreasonably large: larger than the inhomogeneous linewidth of HHW (~10 meV). This may indicate that the single localization energy approximation is poor when the well width fluctuations are relatively large. The fitted $N_{\rm D}$ in the wires is somewhat smaller but close to that in the host quantum well.

We should emphasize that , while the fit to this approximate model may not give accurate values of the parameters, it does show that localization dominates the temperature dependence of the exciton decay time. The theory for free excitons can only be tested directly in samples of much higher quality than those used here or in most previous studies.

4.4. Thermally Enhanced Exciton Transfer from the Well Region to Wires

Since the difference in HH decay times between unpatterned and patterned quantum wells is due to thermally activated transfer, we might hope to get the transfer time τ_t from the difference. However, we must allow for the fact that the localized excitons do not contribute to the transfer and only free excitons should be considered. With this assumption, the lifetime τ_{HH} of excitons in the inter-wire region is given by

$$\frac{1}{\tau_{HH}} = \frac{1}{\tau_{tHH}^0} + \frac{1}{\tau_t} (1 + \frac{N_{loc}}{N_f})^{-1}$$
(6)

where τ_{HH}^0 is the decay time of the HH exciton in the unpatterned quantum well and τ_t is the transfer time. At low temperature, because the tunneling rate is expected to be very small[17] and most excitons are in localized states,

the difference between $\tau_{\rm HH}$ and $\tau_{\rm HH}^{\rm 0}$ is too small to detect. With increasing temperature, the transfer rate increases and more excitons are in free exciton states, so the difference can be obtained more accurately. However, it is still the difference between two large numbers which are subject to the errors discussed in Section 3, and an accurate value cannot be obtained. The transfer times for A-W3(350nm/9nm) and A-W2(350nm/9nm), obtained by fitting (6) to the data with the parameters given in Table 1, are shown in Fig.10. These data show the general trend with varying temperature. At low temperature, they are much longer than the radiative decay time of the HH excitons; at high temperature, they are comparable to the exciton decay time in the unpatterned quantum well.

The transfer time of A-W3 is consistently longer than that of A-W2. The only difference between the two samples is that A-W2 has a wire spacing L=650 nm, while for A-W2 L=1650 nm. If exciton diffusion is rapid in the well region, the transfer time is given by $\tau_t=2L/(\bar{\nu}\,P),$ where L is the wire spacing, P is the transmission probability and $\bar{\nu}$ is the average thermal velocity[17]. The wire-spacing dependence in Fig.10 agrees with this, if one allows for the large experimental uncertainty in these data. The values of τ_t obtained from the CW data of ref. 17 are considerably shorter than those

found here. The factor of five discrepancy is too large to be attributed to experimental error and shows that our kinetic model, while it provides a good qualitative account of the data, is too simple to be relied on quantitatively.

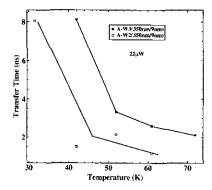


Fig. 10. Exciton transfer times versus temperature for A-W2(350nm/9nm) and A-W3(350nm/9nm). The samples differ only in wire spacing.

5. Conclusions

At low temperature, the luminescent decay times in the strain confined quantum wires and quantum dots are the radiative decay times of localized excitons. The radiative decay time can be either longer or shorter than that of the host quantum well, depending on the size of the wires and dots. For small wires and dots (~ 100 nm stressor), the exciton radiative recombination rate increases due to lateral confinement.

The relaxation time of hot excitons in the wires and dots appears to increase with the depth of strain confinement. We find no convincing evidence that it is significantly slower in quasi 1-D or 0-D systems than in quantum wells, as has been suggested[13].

Exciton localization due to the fluctuations of quantum well thickness plays an important role in the temperature dependence of the luminescent decay time and exciton transfer in quantum wire and dot structures up to a temperature of at least ~ 80 K.

Lateral exciton transfer in quantum wire and dot structures formed by laterally patterning quantum wells strongly affects the dynamics of wire and dot luminescence.

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REFERENCES

- 1. C. J. Hwang, Physical Review B8(1973) 646.
- 2. G. W. 't Hooft, W. A. J. A. van der Poel, L. W. Molenkamp and C. T. Foxon, Physical Review B35 (1987) 8281.
- 3. H. P. Hjalmarson, G. D. Gilliland, D. J. Wolford, T. F. Fuech and J. A. Bradley, Journal of Luminescence 60/61(1994) 830.
- 4. D. S. Citrin, Physical Review B47(1993) 3832, and references therein.
- 5. A. Vinattieri, Jagdeep Shah, T. C. Damen, D. S. Kim, L. N. Pfeiffer and L. J. Sham, Solid State Communications 88(1993) 189, and references therein,
- 6. Yong Zhang and M. D. Sturge, unpublished.
- 7. M. Oestreich, W. W. Rühle, H. Lage, D. Heitmann and K. Ploog, Physical Review Letters 70(1993)
- 8. G. Mayer, F. E. Prins, G. Lehr, H. Schweizer, H. Leier, B. E. Maile, J. Straka, A. Forchel and G. Weimann, Physical Review B 47(1993) 4060.
- 9. J. Christen, M. Grundmann, E. Kapon, E. Colas, D. M. Hwang and D. Bimberg, Applied Physics Letters 61(1992) 67.
- 10. H. Akiyama, S. Koshiba, T. Someya, K. Wada, H. Noge, Y. Nakamura, T. Inoshita, A. Shimizu and H. Sakaki, Physical Review Letters 72(1994) 924.
- 11. I-H. Tan, T. Yasuda, Y.-L. Chang, R. Mirin, E. L. Hu, Y. Segawa, J. Bowers and J. Merz, Applied Physics Letters 62(1993) 1378.
- 12. D. S. Citrin, Physical Review Letters 69(1993) 3393.
- 13. H. Benisty, C. M. Sotomayor-Torrès and C. Weisbuch, Physical Review B44(1991) 10945.
- 14. D. S. Citrin, Superlattices and Microstructures 13 (1993)303
- 15. G. Wang, S. Fafard, D. Leonard, J. E. Bowers, J. L. Merz and P. M. Petroff, Applies Physics Letters 64 (1994) 2815; S. Fafard, D. Leonard, J. L. Merz and P. M. Petroff, Applied Physics Letters 65(1994)
- 16. Yong Zhang, M. D. Sturge, K. Kash, B. P. Van der Gaag, A. S. Gozdz, L. T. Florez and J. P. Harbison, Journal of Luminescence 60/61(1994) 400.
- 17. Yong Zhang, M. D. Sturge, K. Kash, B. P. van der Gaag, A. S. Gozdz, L. T. Florez and J. P. Harbison, Physical Review B (in press) 18. Yong Zhang, Physical Review B49 (1994) 14352.
- 19. K. Kash, B. P. Van der Gaag, Derek D. Mahoney, A. S. Gozdz, L. T. Florez, J. P. Harbison and M. D. Sturge, Physical Review Letters 67(1991) 1326.
- 20. K. Kash, J. M. Worlock, Derek D. Mahoney, A. S Gozdz, B. P. Van der Gaag, J. P. Harbison, P. S. D. Lin and L. T. Florez, Surface Science 228(1990) 415.
- 21. R. Eccleston, B. F. Feuerbacher, J. Kuhl, W. W. Rühle and K. Ploog, Physical Review B45(1992)
- 22. J. Martinez-Pastor, A. Vinattieri, L. Carraresi, M. Colocci, Ph. Roussignol and G. Weimann, Physical Review B47(1993) 10456.
- 23. T. Takagahara, Journal of Luminescence 44(1989) 347.
- 24. Data are not available for the unpatterned 2 nm quantum well.

- Ph. Roussignol, C. Delalande, A. Vinattieri, L. Carraresi, M. Colocci, Physical Review B45(1992) 6965.
- J. Feldmann, G. Peter, E. O. Göbel, P. Dawson, K.J. Moore, C. Foxon and R. J. Elliott, Physical Review Letters 59(1987) 2337; Erratum in Physical Review Letters 60(1988) 243.
- I-Hsing Tan, Ming Yuan He, Jong Chang Yi, Evelyn Hu, Nadir Dagli and Anthony Evans, Journal of Applied Physics 72(1992) 546.
- U. Bockelmann and G. Bastard, Physical Review B 42(1990) 8947.
- L. Rota, F. Rossi, S. M. Goodnick, P. Lugli, E. Molinari and W. Porod, Physical Review B47(1993) 1632.
- Garnett W. Bryant, Bulletin of the American Physical Society 37(1992) 80, and private communication.
- 31. U. Bockelmann and T. Egeler, Physical Review B 46(1992) 15574.
- M Oestreich, W. W. Rühle, H. Lage, D. Heitmann and K. Ploog, Physical Review Letters 70(1993) 1682.

- B. Deveaud F. Clérot, N. Roy, K. Satzke, B Sermage and D. S. Katzer, Physical Review Letters 67(1991) 2355.
- Jun-ichi Kusano, Yusaburo Segawa, Yoshinobu Aoyagi, Susumu Namba and Hiroshi Okamoto, Physical Review B40(1989) 1685.
- L. C. Andreani, F. Tassone and F. Bassani, Solid State Communications 77(1991) 641.
- C. Delalande, M. H. Meynardier and M. Voos, Physical Review B31(1985) 2497.
- 37. Citrin (ref. 4) calls N_{loc} and N_f "the numbers of localized and free excitons respectively". This is incorrect, but the ratio N_{loc}/N_f is the ratio of these numbers, which is all that is needed.
- G. Guillemot, M. Gauneau, A. Regreny and J. C. Portal, Superlattices and Microstructures 2(1986) 445.
- 39. J. Hegarty and M. D. Sturge, Journal of the Optical Society of America B2(1985) 1143.