Thermalization Effect on Radiative Decay of Excitons in Quantum Wires

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The radiative decay of one-dimensional (1D) excitons is studied in novel GaAs quantum wires (QWIs) prepared by molecular beam epitaxy. The temperature dependence of the radiative lifetime $\tau(T)$ is measured, and is compared with that of 2D excitons in quantum wells (QWs): $\tau(T)$ of 1D excitons grows much more slowly with T than that of 2D excitons, resulting in shorter lifetime for $T > 20$ K. This is ascribed to thermalization in the concentrated 1D density of states in QWIs. Analysis based on a theory has revealed that $\tau(T)$ is expressed as $92\sqrt{T}$ ps K$^{-1/2}$ in QWIs and as $24T$ ps K^{-1} in QWs.

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State-of-the-art crystal growth technology has opened the door to the study of intrinsic properties of quantum wires (QWIs) as well as quantum wells (QWs). The recent intensive study of optical properties of QWIs is motivated by two expectations. First, the concentrated electronic density of states (DOS) should make optical transition highly efficient, which would lead to high differential gain in semiconductor lasers [1]. Second, the Coulomb interaction should be very effective in QWIs and create excitons with large binding energy and enhanced optical transition probability [2–4].

Early experiments on QWIs, however, have not provided clear evidences to support these basic expectations. Christen *et al.* [5] obtained cathodoluminescence (CL) decay time of 310 ps at 5 K for crescent-shaped GaAs QWIs. Miller *et al.* [6] obtained photoluminescence (PL) decay time of 379 ps at 1.4 K for serpentine superlattice QWIs. Comparing these values with those of reference QWs, they claimed that the radiative lifetime of one-dimensional (1D) excitons is long.

To understand all these reports consistently, we have examined theories [7–12] on the radiative decay of 1D and 2D excitons, and realized the importance of further experiments at various temperatures.

The point of the theories is as follows. Because of the k-conservation rule in the direction of free motion, the exciton with large center-of-mass wave vector $k > k_0$ cannot decay radiatively, where k_0 is the wave vector of light in the crystal with the same energy as the exciton. Instead, excitons with small $k < k_0$ decay radiatively; their lifetime is short because of their spatial coherence, and is characterized by the intrinsic radiative lifetime τ_0 of the $k \sim 0$ excitons. However [9], when the localization or the homogeneous broadening Δ^* of the excitons is so large that the uncertainty of the wave vector is comparable to k_0, it limits the spatial coherence of the excitons to the coherence length L^* ($\Delta^* \sim h^2/2ML^{*2}$; M is the exciton mass). Then, the $k \sim 0$ exciton lifetime τ_0 is replaced by a longer lifetime τ_0^* determined by L^*, and the maximum wave vector k_0 of the radiative excitons is replaced by $2\pi/L^*$. In experiment, the lifetime observed is the average over those exciton states which are quasithermally distributed.

In view of these theories, the long lifetime observed at low temperatures [5,6] should be regarded essentially as τ_0^*, which depends on the coherence length L^*. Furthermore, the lifetime of 1D excitons may well become shorter than 2D at high temperatures. Hence, it is important to study the temperature dependence of radiative lifetime $\tau(T)$. Most importantly, this measurement of $\tau(T)$ allows one to determine the intrinsic radiative lifetime τ_0 free from L^* or Δ^*, as will be proved later.

The measurement of radiative lifetime over a range of temperatures is not easy because it requires a high-quality sample with negligible nonradiative decay processes even at high temperatures. We have recently fabricated by MBE novel self-forming QWIs [13] as well as the reference QWs of equivalent quality, which are suited for the above purpose; these QWIs and reference QWs have shown PL efficiency which changes little for T up to 60 K.

In this Letter, we present our work on the temperature dependence of the radiative lifetime $\tau(T)$ of 1D and 2D excitons (denoted shortly by τ_{1D} and τ_{2D}, respectively). We show, in particular, that τ_{1D} and τ_{2D} are almost the same at low temperatures below 20 K, whereas τ_{1D} gets shorter than τ_{2D} at higher temperatures. This behavior is explained by the difference of thermalization within 1D and 2D DOS. The radiative lifetimes τ_{1D} and τ_{2D} are expressed as $92\sqrt{T}$ ps K$^{-1/2}$ and $24T$ ps K^{-1}, respectively, from which the intrinsic radiative lifetime τ_0 of the $k \sim 0$ excitons is found to be 110 ps (1D) and 26 ps (2D).

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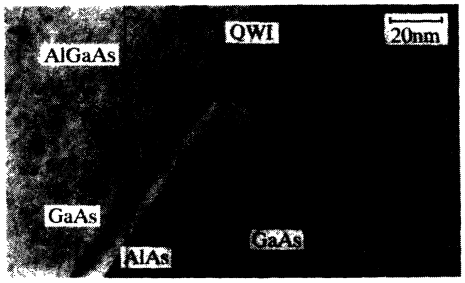

FIG. 1. Cross-sectional TEM micrograph of the top part of the facet structures. GaAs (black) ridge quantum wires 9 nm thick and 18 nm wide are formed between $Al_{0.3}Ga_{0.7}As$ (dark gray) and AlAs (bright gray) barriers.

The QWIs studied here were grown by MBE on a patterned GaAs (001) substrate, on which line patterns of reverse mesa structures had been formed along the ⟨110⟩ direction (1.6 μm width, 2 μm height, 4.0 μm period). First, a GaAs buffer was grown on this substrate. Because of the crystallographic selectivity of epitaxy, sharp ridge structures with two adjacent (111)B facets were formed on top of the reverse mesas. Then, we deposited AlAs (barrier), GaAs (wire), $Al_{0.3}Ga_{0.7}As$ (barrier), and GaAs (cap) layers. Figure 1 is a cross-sectional transmission electron microscope (TEM) photograph of the top part of the facet structure, which clearly shows that GaAs ridge QWIs 9 nm thick and 18 nm wide are successfully formed between the top $Al_{0.3}Ga_{0.7}As$ barrier and the bottom AlAs barrier. In another region in the same wafer, nonuniformity of the initial mesa width had caused incomplete ridge structures with the (001) surface remaining at the top. In such a region, QWs (10 nm thickness, 120 nm width) were formed on this (001) surface (top QWs), which are used as reference. The details of the growth procedures are reported elsewhere [13].

Figure 2 shows PL spectra at various temperatures measured for the two regions denoted by A (thick lines) and B (thin lines) where QWIs and reference top QWs are formed, respectively. Excitons were excited initially in the $Al_{0.3}Ga_{0.7}As$ barrier by a He-Ne laser (632.8 nm, 3 mW, 500 μm spot diameter). The origin of PL peaks in the spectra are assigned by CL measurements [13], and shown in the figure. We observed not only the PL peaks of 1D and 2D excitons from the ridge QWIs and the top QWs, but PL peaks from the bottom QWs formed in the valley region of mesas (a), the side QWs on the (111)B facet plane (b), and bulk GaAs (c,d). Compared with the bottom QWs and the side QWs, the QWIs and the top QWs show stronger PL which survives above 200 K. This indicates the good quality of the top region of the facet structures. Moreover, the PL peaks of the QWIs remain sharp up to 220 K, which shows stability of 1D excitons in the QWIs and their uniformity. To check the PL efficiency, the PL peak intensities of the QWIs and the top QWs are plotted against temperature in Fig. 3. The weakness of the temperature dependence below 60 K

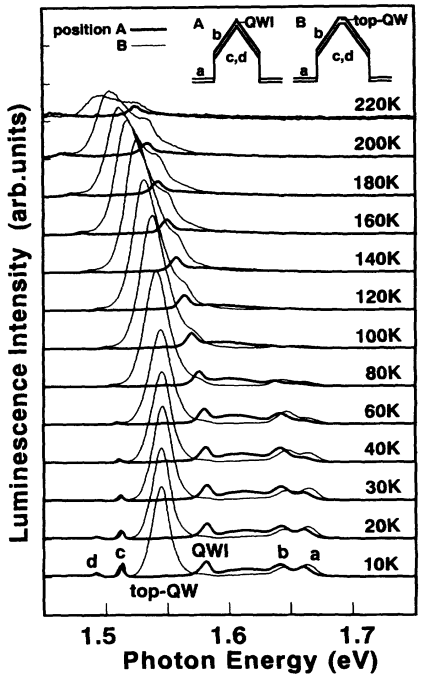

FIG. 2. PL spectra at various temperatures excited by He-Ne laser measured for the two regions, A (thick lines) and B (thin lines). The peaks are assigned as PL from ridge quantum wires (QWI), top quantum wells (top QW), bottom quantum wells (a), side quantum wells (b), and bulk GaAs (c, d).

suggests that the contribution of the nonradiative decay process is small in the low temperature region.

We then measured the time-resolved PL intensities excited by the second harmonics of yttrium-lithium-fluoride (YLF) laser pulses (526 nm, 5×10^9 photons/cm^2) with the time-correlated photon counting method.

We first at 77 K compared the results for the QWIs and the side QWs. The intensity for the QWIs rises quickly and then decays with a time constant of 0.6 ns, whereas the intensity for the side QWs shows quick rise and decay with a time constant of 1.4 ns. This means that the

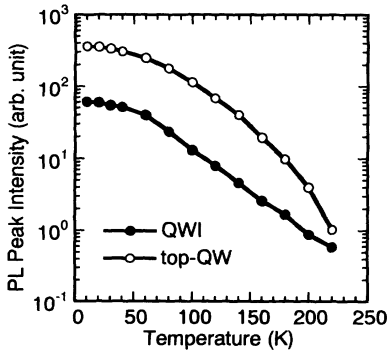

FIG. 3. Temperature dependence of PL peak intensities of QWIs and top QWs excited by He-Ne laser.

excitons created in the $Al_{0.3}Ga_{0.7}As$ barriers flow into the QWIs and the side QWs quickly and decay there, or that excitons flowing from the side QWs to the QWIs are negligible. This is also supported by the results of PL excitation spectrum measurements and CL measurements. Therefore, the decay time of each PL peak shows the lifetime of excitons in each structure.

Next, we compared the temperature dependence of the PL decay time of the QWIs and the top QWs, which is shown in Fig. 4. While the decay time in the high temperature region reflects nonradiative lifetime, that below 60 K shows mainly radiative lifetime, as expected from Fig. 3. Let us, therefore, concentrate on the temperature region below 60 K, and compare the radiative lifetime τ_{1D} and τ_{2D} of the 1D and 2D excitons in the QWIs and the top QWs: We see that τ_{2D} increases linearly with temperature, as is generally the case in QWs [9,11]. On the other hand, the increase of τ_{1D} in this temperature region is much weaker than τ_{2D}. We ascribe this small temperature dependence of τ_{1D} in QWIs to 1D DOS and thermalization within it. As explained in the introduction, the thermalization of excitons to large k states suppresses the radiative decay of the excitons. Since the DOS of 1D excitons is concentrated in the small k region, thermalization is less effective in 1D than in 2D.

We should comment here on the Stokes shifts of the QWI sample measured at various temperatures to see the validity of the interpretation based on the free exciton picture. PL and PL excitation spectra were measured using a synchronously pumped mode-locked dye laser for this purpose. The Stokes shift was 3–4 meV at 9 K, but it became as small as 1–1.5 meV at 30–60 K, which is much smaller than the spectral linewidth (about 10 meV). This indicates that the luminescence is dominantly from free excitons at the elevated temperatures, and supports the validity of the free exciton picture. Further refinement of the theory of free exciton recombination to estimate the correction due to the coexistence of localized states is a subject of future work, and is not treated here.

The effect of the thermalization is characterized by the following parameter $\zeta(T)$, which expresses the fraction of excitons with $k < k_0$. By assuming Maxwell-Boltzmann distribution for excitons [9], $\zeta(T)$ is given by

$$\zeta(T) = \frac{\int_0^{\Delta} D(E)e^{-E/k_BT}dE}{\int_0^{\infty} D(E)e^{-E/k_BT}dE}, \tag{1}$$

where $D(E)$ denotes DOS of excitons, and $\Delta = \hbar^2 k_0^2/2M$ the maximum kinetic energy of excitons which can decay radiatively. The values of $M = 0.27m_0$ (m_0 is the free electron mass) [14], refractive index $n = 3.3$, and the photon energy of the observed PL $\hbar\omega = 1.58$ (1.54) eV for the QWIs (the top QWs) give $\Delta = 0.098$ (0.094) meV. Substituting 1D and 2D DOS ($\propto 1/\sqrt{E}$ and =const, respectively), and assuming $T \gg \Delta/k_B = 1.1$ K, we obtain

$$\zeta_{1D}(T) = \sqrt{\frac{4\Delta}{\pi k_B T}}, \quad \zeta_{2D}(T) = \frac{\Delta}{k_B T}. \tag{2}$$

The averaged radiative lifetime $\tau(T)$ is then given by

$$\tau(T) \simeq \tau_0 \zeta(T)^{-1} = \begin{cases} \tau_0 \sqrt{\dfrac{\pi k_B T}{4\Delta}} & \text{(1D)} \\ \tau_0 \dfrac{k_B T}{\Delta} & \text{(2D)}, \end{cases} \tag{3}$$

where τ_0 is the intrinsic radiative lifetime of the exciton at $k \sim 0$. Therefore, the radiative lifetime $\tau(T)$ is proportional to $\sqrt{T}$ for 1D, and proportional to T for 2D. Although τ_0 is replaced with τ_0^* and Δ with Δ^* in the presence of large homogeneous broadening or localization, one should note that Eq. (3) remains formally valid, as will be proved later, and is to be compared with the experimental results. The observed lifetime below 60 K, where radiative decay is dominant, is indeed fitted by $92\sqrt{T}$ ps $K^{-1/2}$ for the QWIs and $24T$ ps K^{-1} for the top QWs, as shown by the curves in Fig. 4. By comparing these results with Eq. (3), we find that the intrinsic radiative lifetime τ_0 of the $k \sim 0$ exciton is 110 ps in the QWIs and 26 ps in the top QWs; τ_0 is longer in 1D than in 2D.

To further clarify the meaning of these values, we follow Hanamura's theory [10] and obtain the following expressions for τ_0 for the 1D and 2D excitons at $k = 0$ with polarization parallel to the wires and wells, respectively,

$$\tau_{0,1D}^{-1} = \frac{2\pi\omega^2}{\hbar c^2}|\phi(0)|^2\mu_{cv}^2 = 6\pi^3\left(\frac{\lambda}{a}\right)\gamma_s, \tag{4}$$

$$\tau_{0,2D}^{-1} = \frac{2\pi\omega}{\hbar c}|\phi(0)|^2\mu_{cv}^2 = 6\pi\left(\frac{\lambda}{a}\right)^2\gamma_s, \tag{5}$$

where $\phi(r_e - r_h)$ is the 1D or 2D wave function for the exciton internal motion, μ_{cv} is the transition dipole moment between the relevant conduction band and valence band, λ is the wavelength of light, and $\gamma_s = 4\mu_{cv}^2/3\hbar\lambda^3$ is the contribution per unit cell to the band-to-band radia-

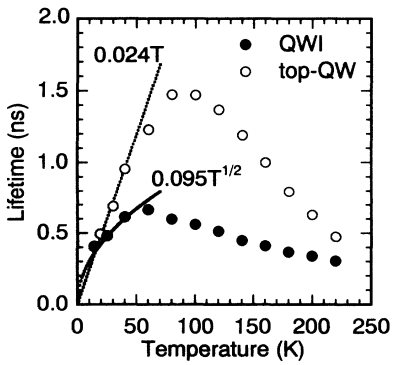

FIG. 4. Temperature dependence of PL decay time from ridge quantum wires (QWI) and top quantum wells (top QW) excited by the second harmonics of YLF laser pulses.

tive decay rate. The last expressions in Eqs. (4) and (5) are obtained by assuming a $1s$ wave function with a phenomenological parameter a for the Bohr radius. [Each a in Eqs. (4) and (5) depends on the structures of QWIs and QWs, and is not common.]

Equations (4) and (5) clearly explain why the observed τ_0 is longer in 1D than in 2D: $\tau_{0,1D}$ is enhanced by a factor of λ/a, whereas $\tau_{0,2D}$ is enhanced by $(\lambda/a)^2$. Although it is expected that the Bohr radius a of 1D excitons can become quite small, this effect is not sufficient to overcome the difference of the 1D and 2D enhancement factors. Our results together with this interpretation are essentially the same as the predictions by Citrin [12], who treated the radiative lifetime of 1D excitons using a more sophisticated theoretical framework.

We note here the validity of the determination of the intrinsic radiative lifetime τ_0 from the observed lifetime $\tau(T)$ using Eq. (3). It is true, according to the theory of Feldmann *et al.* [9], that the lifetime of the lowest exciton state is modified to be τ_0^*; τ_0^* is given by replacing λ/a with L^*/a in Eqs. (4) and (5). At the same time, Δ in Eqs. (1)–(3) should be replaced by Δ^*. Thus, τ_0^* and Δ^* are to be substituted into Eq. (3) instead of τ_0 and Δ. However, the final expression of $\tau(T)$ for $T \gg \Delta^*/k_B$ is unchanged, and the L^* dependence disappears, since $\Delta^* \sim h^2/2ML^{*2}$. This justifies our use of the original Δ in Eq. (3) in obtaining the intrinsic radiative lifetime τ_0 of the excitons at $k \sim 0$. At low temperatures as $T \simeq \Delta^*/k_B$, on the other hand, L^* and τ_0^* should play an important role, to which the observed deviation from Eq. (3) below 20 K in Fig. 4 is most likely attributed.

We finally remark again that the ineffective thermalization within 1D DOS makes the averaged radiative lifetime of excitons in QWIs shorter than in the reference QWs for $T > 20$ K, although the intrinsic radiative lifetime of $k \sim 0$ excitons is longer in the QWIs. This demonstrates an important advantage of QWIs for possible device applications, because such a device is commonly expected to operate at room temperatures.

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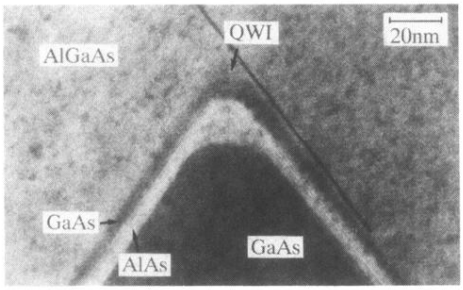

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