Radiative lifetimes of excitons in quantum wires

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The excitonic population lifetime and its temperature dependence is measured in nanometer-scale quantum wires. The measurements were performed on two cleaved-edge overgrown GaAs quantum-wire systems, in which the lateral dimensions are comparable to the dimensions of a reference quantum well and an easy comparison between them is straightforward. The excitonic population decay time in the wires is less sensitive to temperature changes than that in the wells. Whereas the latter is linear with temperature, the former is roughly proportional to the square root of the temperature. Consequently, the decay times of the photoluminescence from the wires are longer than those from the wells at low temperature, but shorter at higher temperatures. From the temperature dependence of the lifetimes, which is an unambiguous signature of a one-dimensional system, we find the intrinsic radiative lifetime of excitons in quantum wires. They are an order of magnitude longer than in quantum wells.

It follows from energy and crystal momentum conservation that in intrinsic bulk semiconductors the interaction of free excitons with the electromagnetic radiation field leads to a stationary state called an excitonic polariton. This elementary excitation, which is a coherent state of the entire macroscopic three-dimensional (3D) crystal, does not decay radiatively.[1] In quantum wells (QW's) the situation is different. Here, due to the removal of the translational symmetry in one direction, the interaction between the 3D electromagnetic radiation field and the 2D excitons results in some low in-plane crystal momentum exciton modes that have finite width and they do decay radiatively.[2–5] The reciprocal of the width of such an exciton mode, or a spin state, is its intrinsic radiative lifetime.[4] The radiative lifetime of the excitonic population in a given mode equals half of its intrinsic lifetime.[5] Andreani showed that in a typical $GaAs/Ga_{1-x}Al_xAs$ QW this intrinsic quantity amounts to a few tens of picoseconds.[4] He explained the considerably longer decay times observed in time-resolved photoluminescence (PL) experiments[6] in terms of the thermal distribution of the excitonic population, where only a small fraction populates the optically active part of the available phase space.[4,5] Due to the characteristic 2D density of states of a QW, this fraction is inversely proportional to the system temperature. Thus, if excitons can reach thermal equilibrium[4,5] faster than they can decay radiatively, and if recombination from long-lived extrinsic centers that break the translational symmetry can be neglected,[5] the model perfectly explains the linear dependence between the PL decay times and the QW's temperature, as was observed in numerous experiments.[6–8]

In a recent Letter, Citrin applied similar considerations to an axially symmetric $GaAs/Ga_xAl_{1-x}As$ quantum wire (QWR) of nanometer scale.[9] He found that the intrinsic radiative lifetime of such a system is longer than the radiative lifetime of a QW with comparable width. He explained this counterintuitive conclusion in terms of the decrease of the excitonic coherence length imposed by the lateral confine-

ment. The decrease in the coherence length predominates over the enhancement of the QWR oscillator strength, which is the result of the increase in the electron-hole wavefunction overlap. Similar calculations performed by others arrived at somewhat different results.[10,11]

The 1D nature of the excitonic density of state in QWR's should thus reveal itself in a characteristic square-root dependence of the PL decay times on the QWR's temperature.[9] The low-temperature PL decay times in QWR's were recently measured by several groups.[12–15] In two cases, where comparison with a QW reference system was available, it was indeed reported that the PL decay times from the QWR's are longer than those from the QW.[14,15]

In this paper we report on a measurement of PL decay times and their temperature dependence in nanometer-scale quantum wires. We show that in our QWR structures, which were fabricated by cleaved-edge epitaxial overgrowth, the intrinsic radiative lifetime amounts to more than 100 ps, and it depends on the dimensions of the structure. This is a few times longer than the corresponding lifetime of a reference QW. We show that in the QWR's the PL decay time increases with temperature (T) like T^β, where β is roughly 0.5. This is in marked contrast with the corresponding QW's, where β is roughly 1.0 at the same temperature range.

Our samples were prepared by two stages of epitaxial growth in two orthogonal directions, as shown schematically in the inset of Fig. 1. In the first stage a 150-period $In_xGa_{1-x}As/GaAs$ strained-layer superlattice (SLS) was grown on a (001)-oriented GaAs substrate. Each period contained 71 Å of $In_{0.063}Ga_{0.937}As$ and 240 Å of GaAs. The SLS was capped with a 2-μm-thick GaAs layer. In the second stage the samples were inserted into a molecular-beam-epitaxy machine. They were then cleaved *in situ* and two $Al_xGa_{1-x}As/GaAs$ QW's of 80 and 34 Å, respectively, were grown on the (110)-oriented cleaved facet.[16] The QW's were separated from the cleaved facet and from each other by 200-Å-thick layers of $Al_{0.3}Ga_{0.7}As$. More details on the
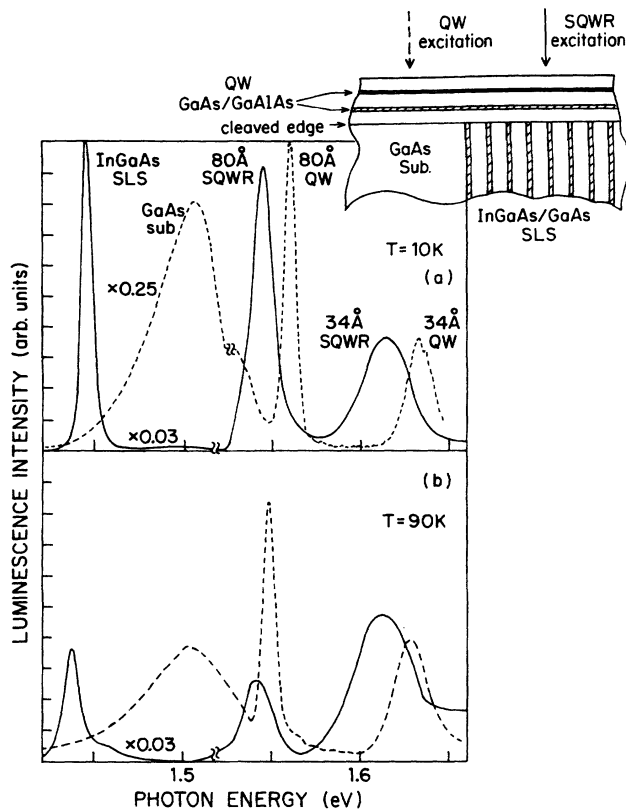
FIG. 1. Temporal integrated PL spectra of the QW's (dashed line) and SQWR's (solid line) at (a) 10 K and (b) 90 K. The cleaved-edge overgrown SQWR's and the positions of the exciting laser beams are schematically described in the inset.

sample growth and the characterization of these strain-induced QWR's (SQWR's) are given elsewhere.[17] An additional T-shaped QWR sample[18] (TQWR) was studied. The sample, which was also grown on a superlattice cleaved edge, is described in detail elsewhere.[19] The nanometer-scale lateral confinement in these wires is produced by the intersection of two orthogonal 70-Å QW's, not by the strain.

For the PL decay-time measurements, light from a picosecond pulsed Ta:sapphire laser was focused to a spot of roughly 3 μm diameter at normal incidence onto the (110) overgrown facet of the sample, using a $\times 40$ microscope objective mounted in a helium-flow cryostat. The emitted light was collected from the sample side and it was analyzed by a 0.25-m monochromator and a cooled multichannel plate photomultiplier. The temporal decay of the PL was measured using conventional time-correlated single-photon counting electronics. The repetition rate of the pulsed laser was 76 MHz. The temporal resolution of the system at this rate was roughly 60 ps.

In Fig. 1 we display the time-integrated PL spectra from the SQWR's (solid line) and from the reference QW's (dashed line). The spectra are given for sample temperatures of 10 and 90 K in Figs. 1(a) and 1(b), respectively. In order to facilitate an experimental comparison between the emission from the SQWR's and that of the QW's, change of a few micrometers in the microscope objective position was needed, as illustrated in the inset to Fig. 1, while everything else remained unchanged. The observed spectral features are identified in Fig. 1(a).

We note that the PL peaks, which are due to exciton recombination in the SQWR's, are shifted toward lower energies than the PL peaks from their parent QW's. In addition to these large shifts, which amount to more than 20 meV in the 10-K PL spectrum, similar shifts and drastic changes in the polarization selection rules of higher excitonic transitions were observed in the PL-excitation (PLE) spectra.[14,17]

The lowest-energy PL peaks are Stokes shifted from the lowest-energy excitonic resonances observed in the PLE measurements (not shown). These Stokes shifts, typically of the order of the PL linewidth at low temperature, and their temperature dependence are comparable in both the QW's and the SQWR's. This is indicative of the high crystallographic quality of the SQWR's, where the strain-induced lateral confinement does not create a tail of low-energy states, which is typical of other, more conventional fabrication methods.

We note that the exciton resonances of the QW's shift faster toward lower energies with the temperature than those of the wires. We believe that these trends, observed in both the PL and PLE spectra, are similar in nature to the insensitivity of the QWR exciton energy to excitation density variations.[19] Both are probably consequences of the QWR's 1D density of states.

We have discussed the spectroscopic evidence for the strong strain-induced lateral confinement previously.[14,17] In the following, we concentrate on the temperature dependence of the temporal decay of the PL from these nanometer scale SQWR's, and from the TQWR's.

In Fig. 2 we show by dots the measured transients of the PL from the 80-Å QW [Figs. 2(a) and 2(c)] and the associated SQWR [Figs. 2(b) and 2(d)]. The transients were recorded at a sample temperature of 15 K [Figs. 2(a) and 2(b)] and 80 K [Figs. 2(c) and 2(d)]. For the analysis of the measured temporal transients, a single-exponential-decay model, convoluted with the system response function, which is described by the dashed line in Fig. 2(a), was used. The solid lines in the figure represent the best fits to the data. For these measurements the PL was excited using 2-ps-long pulses of 710-nm wavelength and energy of about 10 pJ per pulse. Assuming $\sim 0.5\%$ absorption in the quantum structures, this amounts to carrier densities of roughly 10^{11} cm^{-2} and 3×10^5 cm^{-1} in the QW's and the QWR's, respectively. The results presented here were essentially excitation-intensity independent over more than two orders of magnitude centered around this excitation level. The intensity of the emission from the quantum structures was linearly dependent on the excitation intensity. It is clearly seen that the PL decay times of the SQWR's are longer than the PL decay times of the corresponding QW's at 15 K but much shorter at 80 K.

In Fig. 3(a) [3(b)] we plot on a logarithmic scale the temperature dependence of the PL decay times of the 80-Å (34-Å) QW by squares and of the associated QWR by circles. It is seen that at this range (10–90 K) the decay times follow roughly a temperature power-law dependence. The decay times from the QW's increase almost linearly with temperature, as demonstrated by the dashed line of the first unity slope, which is drawn through the measured points. The best-fitted slope obtained for the 80-Å QW was 1.05 ± 0.1 (not shown). The fitting to the lifetimes of the narrower well is more problematic. At low temperature it
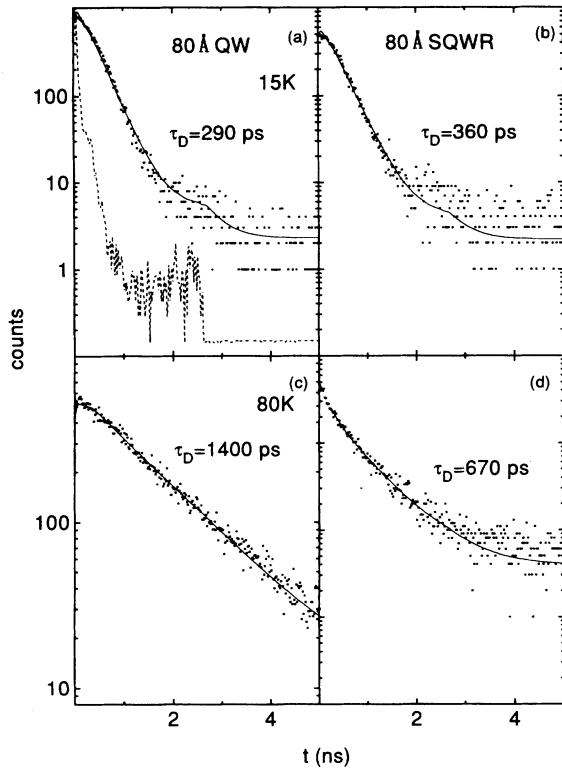
FIG. 2. PL transients from the 80-Å QW and its associated SQWR. The solid lines represent the single-exponential-decay model convoluted with the system response function, which is given by the dashed line in (a).

starts to be temperature independent due to the larger effect of extrinsic centers,[5] and at higher temperatures it behaves like bulk ($\beta = 1.5$), probably due to thermionic emission of carriers from the narrow well. The temperature dependence of the lifetimes of the QW's is in clear contrast to the decay times from the QWR's. Here, the solid lines of slope $\frac{1}{2}$ describe the temperature dependence of the decay times much better. The best-fitted slopes obtained were 0.4 ± 0.1 and 0.33 ± 0.1 for the 34-Å and 80-Å SQWR's, respectively (see
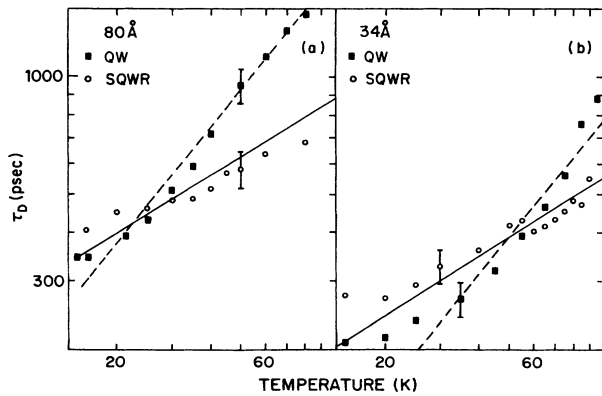

FIG. 3. PL decay times vs temperature for the QW's (squares) and the SQWR's (circles). The dashed lines describe best linear temperature-dependence fit to the QW decay times and the solid lines describe best square-root temperature-dependence fit to the SQWR decay times.

Table I). In particular, we note that as a result of this difference in the temperature dependence, the decay times of the PL from the QWR's become shorter than those from the corresponding QW's above a certain temperature.

In Table I we summarize the information gained from the fitting to a temperature power-law dependence of the measured decay times. The table also includes information from similar measurements performed on the TQWR's.[19]

We assume that photoexcited carriers form excitons that reach thermal equilibrium and lose their spin coherence on a time scale much shorter than the intrinsic radiative lifetime. Then, one can calculate the temperature dependence of the excitonic population lifetime by summing over all the allowed radiative decay rates of the thermal distribution and averaging over all the possible spin states.

For a QW one gets[4,5]

$$\tau_D^{QW}(T) = \frac{3 M_X K_B T}{\hbar^2 k_0^2} \tau_0, \tag{1}$$

where M_X is the lowest-energy exciton in-plane mass, K_B is the Boltzmann's constant, $\hbar$ is the Planck's constant, k_0 is the photon wave vector in the crystal, and $\tau_0 = 1/2\Gamma_0$ is the radiative lifetime of the lowest-energy exciton-state population at zero crystal momentum. The intrinsic radiative width of each one of the two (out of four) optically allowed modes of the lowest-energy exciton state at zero crystal momentum is given by Γ_0.

For a QWR with axial symmetry one gets[9]

$$\tau_D^{QWR}(T) = \frac{5}{16} \left(\frac{2\pi M_X K_B T}{\hbar^2 k_0^2} \right)^{1/2} \tau_0, \tag{2}$$

where $\tau_0 = 8/5\Gamma_0^L = 64/5\Gamma_0^{\pm}$ is the radiative lifetime of the lowest-energy-state exciton population at zero crystal momentum and Γ_0^L and Γ_0^+ (Γ_0^-) are the intrinsic radiative width for the longitudinal and positive (negative) helicity exciton modes with zero crystal momentum, respectively. These three (out of four) lowest-energy exciton modes are optically active in an axially symmetric QWR.[9,11] Similar modes are expected for quadratic cross-section QWR's.[10]

Using Eqs. (1) and (2) and a reasonable value for M_X (0.25 emu), we estimated from the measured PL decay curves the intrinsic radiative lifetimes of excitons in all the quantum structures studied. The values that we obtained are given in Table I. We note that the radiative lifetimes deduced for the QW's are in reasonable agreement with theory[5] and with experimental determination using resonant excitation.[8,20,21] It is clear that the temperature dependence of the population decay times and the radiative lifetimes of nanometer-scale QWR's are drastically different from those of QW's. As expected from simple density-of-state considerations, there is a temperature range in which the excitonic population decay time in a QW is roughly proportional to the ambient temperature, while at the same temperature range the decay time of a comparable QWR is roughly proportional to the square root of its temperature. In addition, we conclude from the table that the intrinsic radiative lifetime of excitons in QWR's is more than an order of magnitude longer than that of a comparable QW. We note also that the intrinsic lifetimes of the QWR's, like those of the QW's,

TABLE I. The experimentally determined intrinsic radiative lifetimes for the various QW's and QWR's studied in this work.

	34-Å QW	80-Å QW	34-Å SQWR	80-Å SQWR	TQWR
β	1.09 ± 0.2	1.05 ± 0.1	0.4 ± 0.1	0.33 ± 0.1	0.58 ± 0.1
A [a]	7 ± 3 (ps/K)	15 ± 5 (ps/K)	60 ± 20 (ps/K$^{1/2}$)	90 ± 27 (ps/K$^{1/2}$)	270 ± 50 (ps/K$^{1/2}$)
τ_0 (ps)	6 ± 3	12 ± 4	120 ± 40	180 ± 40	520 ± 100

[a]We fit $\tau_D(T) = A t^\beta$, where β equals 1 ($\frac{1}{2}$) for a QW (QWR).

scale with their quantum size. The intrinsic lifetime of the 80-Å SQWR is longer than that of the 34-Å SQWR, but much shorter than that of the TQWR, in which carriers do not see confining potential along the planes of the intersecting quantum wells that form the wire.

The departure of the temporal dependence of the population lifetime from the simple predictions of the model, as observed here for both the QW's and the QWR's can be explained in terms of additional contributions to the homogeneous width of the excitonic transition and their temperature dependence.[6-8] Densities of states which are not given exactly by the assumption of parabolic electronic bands should also be considered.[22] Disorder induced by interface roughness and composition fluctuations, as well as extrinsic effects caused by impurities and dislocations, should be included in the analysis in order to account for finer details of the experimental observations.[5] We leave these considerations for forthcoming research efforts. In conclusion, we demonstrated that in a nanometer-scale QWR the PL decay times are drastically different from those of a QW. They are intrinsically longer and less sensitive to temperature variations, in agreement with theory.

Note added. A recent paper by Akiyama *et al.*[23] reports a similar effect in PL transients from their self-formed QWR.

[1] J. J. Hopfield, Phys. Rev. **112**, 1555 (1958).

[2] V. M. Agranovitch and A. O. Dubovskii, Pis'ma Zh. Eksp. Teor. Fiz. **3**, 345 (1966) [JETP Lett. **3**, 223 (1966)].

[3] E. Hanamura, Phys. Rev. B **38**, 1228 (1988).

[4] L. C. Andreani, Solid State Commun. **77**, 641 (1991).

[5] D. S. Citrin, Solid State Commun. **84**, 281 (1992); Phys. Rev. B **47**, 3832 (1993).

[6] J. Feldman, G. Peter, E. O. Gobel, P. Dawson, K. Moore, and C. Foxon, Phys. Rev. Lett. **59**, 2337 (1987).

[7] M. Gurioli, A. Vinattieri, M. Colocci, C. Deparis, J. Massies, G. Neu, A. Bosacchi, and S. Franchi, Phys. Rev. B **44**, 3115 (1991).

[8] J. Martinez-Pastor, A. Vinattieri, L. Carraresi, M. Colocci, Ph. Roussignol, and G. Weimann, Phys. Rev. B **47**, 10 456 (1993).

[9] D. S. Citrin, Phys. Rev. Lett. **69**, 3393 (1992).

[10] S. Jorda, Solid State Commun. **87**, 439 (1993), who calculates the effect for a quadratic cross-section QWR, gets an effect which is almost a factor of 3 smaller.

[11] F. Tassone and F. Bassani, Nuovo Cimento **14D**, 1241 (1992), whose calculation for a cylindrically symmetric wire disagrees with Ref. 7 on this point.

[12] M. Kohl, D. Heitmann, W. W. Ruhle, P. Grambow, and K. Ploog, Phys. Rev. B **41**, 12 338 (1990).

[13] J. Christen, M. Grundmann, E. Kapon, E. Colas, D. M. Hwang, and D. Bimbereg, Appl. Phys. Lett. **61**, 67 (1992).

[14] D. Gershoni, J. S. Weiner, E. A. Fitzgerald, L. N. Pfeiffer, and N. Chand, in *Optical Phenomena in Semiconductor Structures of Reduced Dimensions*, edited by D. J. Lockwood and A. Pinczuk (Kluwer Academic, Dodrecht, 1993), p. 337.

[15] M. S. Miller, H. Weman, C. E. Pryor, M. Krishnamurthy, P. M. Petroff, H. Kroemer, and J. L. Merz, Phys. Rev. Lett. **68**, 3464 (1992).

[16] L. N Pfeiffer, K. W. West, H. L. Stormer, J. Eisenstein, K. W. Baldwin, D. Gershoni, and J. Spector, Appl. Phys. Lett. **56**, 1697 (1990).

[17] D. Gershoni, J. S. Weiner, S. N. G. Chu, G. A. Baraff, J. M. Vandenberg, L. N. Pfeiffer, K. West, R. A. Logan, and T. Tanbun-Ek, Phys. Rev. Lett. **65**, 1631 (1990); **66**, 1375 (1991).

[18] A. R. Goni, L. N. Pfeiffer, K. W. West, A. Pinczuk, H. U. Baranger, and H. L. Stormer, Appl. Phys. Lett. **61**, 1956 (1992).

[19] W. Wegscheider, L. N. Pfeiffer, M. M. Dignam, A. Pinczuk, K. W. West, S. L. McCall, and R. Hull, Phys. Rev. Lett. **71**, 4071 (1993).

[20] B. Deveaud, F. Clerot, N. Roy, K. Satzke, B. Sermage, and D. S. Katzer, Phys. Rev. Lett. **67**, 2355 (1991).

[21] A. Vinattieri, J. Shah, T. C. Damen, D. S. Kim, L. N. Pfeiffer, and L. J. Sham, Solid State Commun. **88**, 189 (1993).

[22] G. A. Baraff and D. Gershoni, Phys. Rev. B **43**, 4011 (1991).

[23] H. Akiyama, S. Koshiba, T. Someya, K. Wada, H. Noge, Y. Nakamura, T. Inoshita, A. Shimizu, and H. Sakaki, Phys. Rev. Lett. **72**, 924 (1994).