Long Intrinsic Radiative Lifetimes of Excitons in Quantum Wires

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(Received 8 September 1992)

Calculations of exciton radiative lifetimes in quantum wires free of defects are presented. Only excitons near zero wave number can spontaneously undergo radiative decay while conserving energy and momentum. The lifetime of the lowest-energy exciton in 100-Å-diam GaAs quantum wires, $\tau_{\rm spon} = 150$ ps, is an order of magnitude longer than in quantum wells. This is due to the finite spatial coherence in the lateral direction. Thermalization effects lead to an effective lifetime of $\tau(T) = 140\sqrt{T}\ \mathrm{ps\,K^{-1/2}}$.

PACS numbers: 78.47.+p, 71.36.+c, 73.20.Dx, 78.65.–s

Low-temperature photoluminescence (PL) decay times in quantum wells (QW) spectrally integrated over the $1s$ heavy-hole peak are in the range 0.2–1 ns [1, 2]. Theoretical calculations show that the intrinsic decay times, i.e., in a structure free of defects, are shorter, on the order of 10 ps [3, 4]. Because only excitons near zero wave number can decay spontaneously into a photon due to the requirement of energy-momentum conservation, only a small fraction of the thermal exciton population can decay via the purely electrodynamic path [1, 3]. The picture is further complicated by the presence of excitons bound to interface defects, particularly in QWs narrower than the exciton Bohr radius. The localization mixes nonradiative large-wave-vector components into the exciton wave function for states near the zone center and so increases the radiative decay time.

Similar effects are expected for quantum wires (QWRs). In this case, however, the detection of intrinsic effects is expected to be more elusive because of the typically low interface quality of QWRs and because the ratio of interface area to volume is greater than for QWs. Moreover, whereas even in the best of cases QW interface morphology is ill characterized and its interaction with excitons is not well understood, even less is known about QWRs. There are lifetime measurements in QWRs grown by lithography and etching [5], but the aforementioned problems make difficult the extraction from the data of an intrinsic decay time. These difficulties necessitate the use of epitaxially grown QWRs in order to study the intrinsic decay mechanism. Recently, cathodoluminescence decay times in V-groove QWRs were measured [6]. The QWRs are GaAs/AlGaAs in composition and are crescent shaped with dimensions of 28 Å thick and ~200 Å across. A radiative lifetime of 310 ps at 5 K was measured. Another study [7] performed on serpentine superlattices gives a PL-decay time of 379 ps at 1.4 K. In this Letter we present results of a Green's function based calculation which gives, e.g., for a GaAs QWR with a diameter of 100 Å, an intrinsic exciton radiative decay time of ~150 ps and a thermally averaged decay time of $140\sqrt{T}$ ps (T in kelvins). In contrast, the radiative lifetimes in a 100-Å GaAs/AlGaAs QW are ~13 ps and $17T$ ps [4]. A comparison of the radiative lifetimes in QWs and QWRs of similar confinement dimensions shows that the long lifetimes are due to finite spatial coherence in the lateral direction in the QWR [8]. The slow radiative lifetimes, together with the predicted slow dephasing times in QWRs due to low scattering rates and bottlenecks in the carrier populations [9, 10], indicate that the time response of QWR-based devices incorporates a more stringent limitation than any present for QW-based devices.

The details of the theoretical treatment by which the intrinsic decay rate is obtained are presented elsewhere [4]. Consider a QWR oriented along the z axis. The electron coordinate is denoted by $(\boldsymbol{\rho}, \zeta)$ and the hole coordinate by $(\boldsymbol{\rho}', \zeta')$. $\boldsymbol{\rho}$ and $\boldsymbol{\rho}'$ are the coordinates in the plane perpendicular to the z axis. $\mathbf{r} = (\mathbf{r}_\perp, r_z) = (\boldsymbol{\rho} - \boldsymbol{\rho}', \zeta - \zeta')$ is the relative coordinate. We assume Wannier excitons formed from two doubly degenerate subbands. The QWR single-particle Bloch states in the conduction (valence) subband are labeled c_σ ($v_{\sigma'}$) where σ (σ') is a spin index. The label $(cv)_s$ means an electron-hole pair $c_\sigma v_{\sigma'}$. For an exciton of spin state $s = (\sigma\sigma')$, the interacting Green's function, which is proportional to the dipole autocorrelation function, is

$$\mathcal{D}_s(k) = \frac{2E_{\rm ex}(k_z)}{(i\omega)^2 - E_{\rm ex}(k_z)^2 - 2E_{\rm ex}(k_z)\hbar\Sigma_s(k)}\ , \qquad (1)$$

where $k = (i\omega, k_z)$, k_z is the wave number (along the QWR), $i\omega$ is the discrete complex frequency, and $E_{\rm ex}(k_z)$ is the exciton dispersion. The basic ingredients that go into $\mathcal{D}_s(k)$ are the coupling strengths of the exciton with all the modes of the optical field having k_z as wave number projected in the z direction. $\hbar\Sigma_s(k)$ is the proper radiative self-energy which is given by

$$\begin{aligned} \hbar\Sigma_s(k) &= \sum_\epsilon \hbar\Sigma_{s\epsilon}(k), \\ \hbar\Sigma_{s\epsilon}(k) &= B_{s\epsilon}V_\epsilon(k), \qquad (2) \\ B_{s\epsilon} &= \frac{e^2|F_{\rm ex}(0)|^2\bar{\kappa}^2}{\epsilon_\infty}\,|\langle\langle (cv)_s|\mathbf{R}\cdot\hat{\mathbf{n}}_\epsilon|0\rangle|^2\ , \end{aligned}$$

where $\hat{\mathbf{n}}_\epsilon$ is a unit vector in the direction of the light

polarization ϵ. For cylindrical symmetry, the polarizations are longitudinal ($\hat{\mathbf{n}}_L \cdot \hat{\mathbf{z}} = 0$) denoted by $\epsilon = L$ and two transverse ($\hat{\mathbf{n}}_\pm \times \hat{\mathbf{z}} = \mathbf{0}$) denoted by $\epsilon = \pm$ for the two circular light polarizations. The other symbols are defined as follows: e is the electronic charge, $\bar{\kappa} = i\omega/\hbar c$, $c = c_0/\sqrt{\epsilon_\infty}$ is the speed of light in the medium, c_0 is the *in vacuo* speed of light, and ϵ_∞ is the high-frequency dielectric constant. $F_{\rm ex}(r_z)$ is the exciton envelope function and $\langle\langle (cv)_s|\mathbf{R}|0\rangle$ is the dipole matrix element between the electron-hole pair state and the crystal ground state. $V_\epsilon(k)$ is given by

$$V_\epsilon(k) = U_\epsilon \mathcal{P} + 2\pi\delta_{\epsilon\pm}\bar{\kappa}^{-2}\mathcal{I}$$

with

$$U_\pm = -(\bar{\kappa}^2 + k_z^2)/2\bar{\kappa}^2, \quad U_L = -(\bar{\kappa}^2 - k_z^2)/\bar{\kappa}^2,$$

$$\mathcal{P} = \int d^2\rho \int d^2\rho' f_c(\boldsymbol{\rho}) f_v(\boldsymbol{\rho}) f_v(\boldsymbol{\rho}') f_c(\boldsymbol{\rho}') \times \int_{-\infty}^{\infty} \frac{dz}{r} e^{i\bar{\kappa}r} e^{-ik_z z},$$

$$\mathcal{I} = \int d^2\rho \, |f_c(\boldsymbol{\rho})|^2 |f_v(\boldsymbol{\rho})|^2.$$

f_c and f_v are conduction- and valence-band (single-particle) envelope functions and are chosen to be real. The Green's function is continued analytically to real energies $i\omega \to E + i\delta$ [$\bar{\kappa} \to \kappa + i\delta = E/(\hbar c) + i\delta$] where δ is a positive infinitesimal. The self-energy gives the real energy shift $\hbar\Pi_s(E, k_z) = \mathrm{Re}\hbar\Sigma_s(E, k_z)$ and the intrinsic radiative decay rate $\Gamma_s(E, k_z) = -\mathrm{Im}\Sigma_s(E, k_z)$. [Note that the rate for the probability that the exciton has not decayed is given by $2\Gamma_s(E, k_z)$.] In the sequel, we need the energy shift and decay rate evaluated on energy shell, $E = E_{\rm ex}(k_z) \approx E_{\rm ex}(0)$. In order to evaluate $\mathcal{P}$ and $\mathcal{I}$, we require the single-particle envelope functions. A convenient choice that facilitates the evaluation of the integrals is $f_b(\boldsymbol{\rho}) = [2/(\lambda_b\pi)]^{1/2} \exp(-\rho^2/\lambda_b)$ ($b = c, v$). Although these functions do not solve the Schrödinger equation for the single-particle states, they provide a model which allows the evaluation of the integrals. A comparison of the true envelope function for a cylindrical QWR with infinite barriers with the model shows that $\sqrt{\lambda/2}$ is roughly half the wire radius ρ_0 where $\lambda = 2\lambda_c\lambda_v/(\lambda_c + \lambda_v)$. We define the parameters

$$B_\epsilon = \sum_s B_{s\epsilon} \approx \frac{(\hbar c_0)\alpha E_{\rm ex}(0)}{2m_0 c_0^2} f_\epsilon |S|^{-2},$$

where $\alpha = 1/137$ is the fine-structure constant, m_0 is the free-electron mass, S is the overlap of f_c and f_v, and $f_\epsilon = 2m_0 E_{\rm ex}(0)|S|^2 \sum_s |\langle\langle (cv)_s|\hat{\mathbf{n}}_\epsilon \cdot \mathbf{R}|0\rangle|^2/\hbar^2$ is the oscillator strength per unit length for polarization ϵ. For the lowest-lying exciton, the only nonvanishing dipole matrix elements are $|\langle\langle (cv)_{\pm\mp}|\hat{\mathbf{n}}_L \cdot \mathbf{R}|0\rangle|^2 = \frac{2}{3}\mu^2$ and $|\langle\langle (cv)_{\pm\pm}|\hat{\mathbf{n}}_\pm \cdot \mathbf{R}|0\rangle|^2 = \frac{1}{3}\mu^2$, where μ^2 is the squared bulk dipole matrix element. The ratio of the oscillator strengths for the different polarizations is then $f_L : f_+ : f_- = \frac{4}{3} : \frac{1}{3} : \frac{1}{3}$ [11].

The self-energy is evaluated in the QWR ($\kappa^2\lambda \ll 1$) and long-wavelength ($k_z^2\lambda \ll 1$) limits. This gives

$$\begin{aligned} \hbar\Sigma_{sL}(E, k_z) &= -B'_{sL}\lambda(\kappa^2 - k_z^2)\mathcal{P}(E, k_z), \\ \hbar\Sigma_{s\pm}(E, k_z) &= -\tfrac{1}{2}B'_{s\pm}[\lambda(\kappa^2 + k_z^2)\mathcal{P}(E, k_z) - 4], \end{aligned} \tag{3}$$

where $B'_{s\epsilon} = B_{s\epsilon}/(\lambda\kappa^2)$ is independent of κ and

$$\mathcal{P}(E, k_z) = |S|^2[-\ln\lambda|\kappa^2 - k_z^2| + 2\ln 2 - 2\gamma + i\pi\Theta(\kappa - k_z)e^{\kappa^2\lambda/4}].$$

The real energy shift $\hbar\Pi_{s\epsilon}(E, k_z) = \mathrm{Re}\hbar\Sigma_{s\epsilon}(E, k_z)$ and the decay rate $\Gamma_{s\epsilon}(E, k_z) = -\mathrm{Im}\Sigma_{s\epsilon}(E, k_z)$ are then

$$\begin{aligned} \hbar\Pi_{sL}(E, k_z) &= -B'_{sL}\lambda(\kappa^2 - k_z^2)\mathcal{P}_r(E, k_z), \\ \hbar\Pi_{s\pm}(E, k_z) &= -\tfrac{1}{2}B'_{s\pm}[\lambda(\kappa^2 + k_z^2)\mathcal{P}_r(E, k_z) - 4], \end{aligned} \tag{4}$$

$$\begin{aligned} \Gamma_{sL}(E, k_z) &= \tfrac{\pi|S|^2}{\hbar}B'_{sL}\lambda(\kappa^2 - k_z^2)\Theta(\kappa - k_z)e^{-\kappa^2\lambda/4}, \\ \Gamma_{s\pm}(E, k_z) &= \tfrac{\pi|S|^2}{2\hbar}B'_{s\pm}\lambda(\kappa^2 + k_z^2)\Theta(\kappa - k_z)e^{-\kappa^2\lambda/4}. \end{aligned} \tag{5}$$

Here $\mathcal{P}_r(E, k_z) = \mathrm{Re}\mathcal{P}(E, k_z)$. Thus, the states between the bottom of the exciton band and the crossing with the photon line ($k_z < |\kappa|$) are quasistationary with respect to radiative decay, similar to the case for QWs [3, 4]. Equation (4) shows that the polariton dispersions for $\epsilon = L$ and $\epsilon = \pm$ are qualitatively distinct near the photon line $\kappa = k_z$. The L mode is continuous with a cusp at $\kappa = k_z$ while the $\pm$ modes are discontinuous and composed of two branches each. This type of behavior for the L and T modes has been predicted for exciton polaritons in QWs [12–15].

For non-spin-polarized excitons, the decay rates must be averaged over the four spin states and summed over the decay channels [16]. The self-energy is evaluated on energy shell $E = E_{\rm ex}(k_z) \approx E_{\rm ex}(0)$. Thus

$$\begin{aligned} \Gamma(E_{\rm ex}(0), 0) &= \frac{1}{4}\sum_{s\epsilon}\Gamma_{s\epsilon}(E_{\rm ex}(0), 0) \\ &= (4\hbar)^{-1}(B_L + \tfrac{1}{2}B_+ + \tfrac{1}{2}B_-)|S|^2\pi e^{-\kappa_{\rm ex}^2\lambda/4}, \end{aligned}$$

where $\kappa_{\rm ex} = E_{\rm ex}(0)/\hbar c$. We consider a $2\rho_0 = 4\sqrt{\lambda/2} = 100$ Å GaAs/AlGaAs QWR and take $f_L \approx 4 \times 10^{-2}$ Å^{-1} [17] and $E_{\rm ex}(0) = 1.577$ eV $+ E_0 - E_b$ with $E_b = 10$ meV (exciton-binding energy) and $E_0 = 120$ meV (sum

of the electron and hole single-particle confinement energies) [17]. The energy difference $|\hbar\Pi_{s\pm}(E_{ex}(0),0) - \hbar\Pi_{sL}(E_{ex}(0),0)| \sim 0.3$ meV (provided s is dipole active for the respective polarizations) between the L and $\pm$ modes sets the scale for the longitudinal-transverse splitting. The intrinsic radiative lifetime is $[2\Gamma(E_{ex}(0),0)]^{-1} \sim 150$ ps at $k_z = 0$. This value is almost an order of magnitude greater than the $k_\parallel = 0$ decay time of 25.5 ps for excitons in a QW of 100 Å width [3, 4].

This increase in the decay time compared with that for QWs comes about as follows. In terms of the expressions for the radiative decay rates in QWs and QWRs at zero wave vector, the ratio of the lifetimes for two cases is [4, 12]

$$\frac{\tau_{spon}^{(QWR)}}{\tau_{spon}^{(QW)}} \approx \frac{\sum_\epsilon f_\epsilon^{(QW)}}{\sum_{\epsilon'} f_{\epsilon'}^{(QWR)} \kappa_{ex}},$$

where the superscripts indicate whether the quantity is for the QWR or QW. Substituting typical values for the oscillator strengths [17, 18] gives $\tau_{spon}^{(QWR)}/\tau_{spon}^{(QW)} \approx 5$. To an approximation, $f_\epsilon^{(QWR)} \sim 2\rho_0 f_\epsilon^{(QW)}$. Thus, $\tau_{spon}^{(QWR)}/\tau_{spon}^{(QW)} \sim (2\rho_0\kappa_{ex})^{-1}$. The additional factor of $2\rho_0$ in the QWR oscillator strength compared with that of the QW is due to the finite spatial coherence in the QWR for the lateral direction imposed by the geometry rather than by disorder [8]. The argument from the point of view of spatial coherence is only approximate as the lateral confinement increases the electron-hole overlap and so leads to some enhancement of the QWR oscillator strength compared with the QW value normalized to the wire width. Nevertheless, the numerical values obtained from Eq. (5) show that the effect of the lateral coherence length dominates for structures of comparable confinement dimensions and consequently one expects long intrinsic radiative lifetimes of excitons in QWRs compared to QWs. The discussion also shows that smaller band-gap materials should be exploited to magnify the lifetime enhancement.

It was shown above that only free excitons with $k_z < \kappa$ have finite intrinsic radiative lifetimes (neglecting other processes). A thermal distribution of free excitons is expected to have a longer effective lifetime since only a small fraction of the occupied states satisfies $k_z < \kappa$. In order that thermal distribution of excitons be maintained, the quasielastic scattering (given by the energy-relaxation rate) of the excitons must be fast compared with the radiative process. Performing such a thermal average, one obtains for the temperature-dependent decay rate

$$\Gamma(T) = \frac{\sum_{s\epsilon} \int_0^\infty dE' e^{-E'/k_BT} \Gamma_{s\epsilon}(E_{ex}(0), k_z)}{\sum_s \int_0^\infty dE' e^{-E'/k_BT}},$$

where k_B is the Boltzmann constant and $E' = \hbar^2 k_z^2/2M$ with M the exciton effective mass. This gives for the lowest-lying exciton

$$\Gamma(T) = \frac{3B_L}{2\hbar}\sqrt{\frac{\pi E_1}{k_BT}} = \frac{3\alpha E_{ex}(0)}{4m_0c_0} f_L \sqrt{\frac{\pi E_1}{k_BT}} \quad (6)$$

for $1 < T < 150$ K. Above 150 K optical-phonon scattering is expected to become important [19]. Here $E_1 = \hbar^2\kappa_{ex}^2/2M$ is the exciton bandwidth below the crossing with the photon line. We take $M = 0.25m_0$. The temperature-dependent effective radiative lifetime of free excitons in a GaAs QWR ($2\rho_0 = 2\sqrt{2\lambda} = 100$ Å) is $\tau(T) = 1/2\Gamma(T) = 140\sqrt{T}$ ps (T in kelvins). Compared with the 100-Å QW [3, 4], the QWR exhibits considerably longer PL-decay time for $T < 100$ K. Furthermore, as a consequence of the different densities of states in one and two dimensions, the temperature dependences are different. (For a QW, the decay time is linear in T [4].) These temperature dependences of the PL-decay time are expected to be retained when excitons bound to width fluctuations are taken into account [4]. Thus, it is of interest to conduct temperature studies of the PL-decay time in QWRs for $T < 100$ K. Such experiments might serve as an analytic tool to identify high-quality QWRs.

An important assumption in the previous discussion is that the energy relaxation must be fast compared with the radiative lifetime. To restate this more accurately, the scattering mechanisms must be sufficiently fast so that a quasithermal distribution of excitons is maintained while the decay process takes place. In QWs, this assumption is probably satisfied [20]. In the early stages of the investigation of QWRs, however, it was predicted that scattering processes are suppressed in QWRs due to a reduction in the phase space for final states [21]. Also, one expects bottleneck effects in the scattering processes which may prevent rapid thermalization [9, 10]. Thus even in high-quality QWRs the effective PL decay might begin with a rapid decay (reflecting the intrinsic process) followed by a slow decay constrained by energy relaxation. A signature of the existence of rapid thermalization is the $\sqrt{T}$ dependence of the PL-decay time.

To conclude, it has been predicted that QWRs have long exciton radiative lifetimes compared with QWs for structures where the wire diameter is the same as the well width. The long lifetime in QWRs is due to the finite spatial coherence [8] of the QWR exciton in the lateral direction due to the geometric constraint. The present treatment describes the intrinsic radiative decay of free excitons. At very low temperature, however, a large fraction of the states is expected to be localized by interface defects. The localizing potential mixes a nonradiative large k_z component into the states and gives lifetimes longer than the intrinsic free-exciton lifetime. At high temperatures as well the localized excitons are expected to play an important role in the effective decay time [4]. Therefore, a study of the intrinsic radiative decay times in QWRs necessitates minimizing the density

of interface defects in addition to the elimination of nonradiative sites. Moreover, QWRs with very small cross section are required. Time-resolved cathodoluminescence spectrosopy has recently been used to study the radiative decay time of excitons in high-quality epitaxially grown GaAs/AlGaAs QWRs [6]. Radiative decay times of 310 ps were measured at 5 K. Although this value is close to the theoretically predicted decay time, such a value is also typical of low-temperature PL-decay times in QWs where recombination of excitons localized by well-width fluctuations dominates [1, 4]. This difficulty in interpretation applies for the QWRs as well. Similar comments also hold for the serpentine superlattices studied in Ref. [7].

We thank R. Eccleston for helpful conversations and a critical reading of the manuscript.

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