Radiative lifetimes of excitons in quantum wells: Localization and phase-coherence effects

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A theory for intrinsic radiative decay of excitons in quantum wells including the effects of excitons bound to width fluctuations and finite phase coherence is presented. The approach is based on Green's functions at nonzero temperature. In $GaAs/Al_xGa_{1-x}As$ quantum wells of widths 50–150 Å, lifetimes on the order of 100 ps are predicted for localized excitons. We show that the localized excitons play an important role in the photoluminescence (PL) decay times over the entire temperature range $T<150$ K. The theoretical results explain the large scatter in the published experimental data. The relationship between the homogeneous linewidth $\hbar\Gamma_h$ and the spectrally integrated decay rate at nonzero temperature for the free-exciton population is derived using a Green's-function approach. The free-exciton contribution to the PL-decay rate reduces to the infinite-lifetime result of L. C. Andreani *et al.* [Solid State Commun. **77**, 641 (1991)] for $k_BT \gg \hbar\Gamma_h$. In the limit $k_BT \ll \hbar\Gamma_h$, the result of J. Feldmann *et al.* [Phys. Rev. Lett. **59**, 2337 (1987)] assuming finite phase coherence is obtained.

I. INTRODUCTION

Time-resolved photoluminescence (PL) spectroscopy has proved to be a powerful tool for the study of exciton lifetimes[1–4] and exciton-population relaxation.[5] This paper is a theoretical investigation of radiative lifetimes of free and localized excitons in quantum wells (QW's) and the effect on the decay time of the PL intensity integrated over the lowest exciton peak including finite temporal and spatial coherence effects. The mixed modes in solids involving excitons and light are polaritons. The interaction of Frenkel excitons and light was first investigated in bulk and it was found that the resulting polaritons are stationary. Thus bulk excitons have no purely electrodynamic radiative-decay path available which conserves both energy and momentum.[6] Radiative decay must be associated with phonons or translational-invariance breaking defects, impurities, or interfaces. QW's, however, inherently break full translational invariance and so the electrodynamic decay channel is open for radiative recombination of excitons lying below the crossing with the photon line [i.e., with center-of-mass in-plane wave number $k_\parallel$ less than $\kappa = E\sqrt{\epsilon_\infty}/(\hbar c_0)$ where $E/\hbar$ is the angular frequency of light, ϵ_∞ is the high-frequency dielectric constant of the medium, and c_0 is the *in vacuo* speed of light].[7–9] Thus QW excitons that lie below the crossing with the photon line are quasistationary.

Andreani, Tassone, and Bassani have recently calculated the radiative lifetime and the temperature dependence of the PL decay time for free excitons in $GaAs/Al_xGa_{1-x}As$ QW's.[9] For an optically active $1s$ heavy-hole exciton in a 100-Å $GaAs/Al_xGa_{1-x}As$ QW, decay times of 25.5 ps for the $k_\parallel = 0$ states were found for decay into longitudinal (L) and transverse (T) photons.[9] (The polarization direction refers to that of the emitted photon with respect to the exciton wave vector.) For the Z polarization, the oscillator strength vanishes for the heavy-hole exciton and so its decay rate is zero for all $\mathbf{k}_\parallel$. Such short lifetimes have been observed recently in time-resolved PL studies of a GaAs/AlAs QW;[10] however, typically the measured decay times at low temperature are in the range 0.25–1 ns.[1,2] It is argued in Ref. 9 that for a thermal distribution of excitons, only a small fraction of the states lie below the crossing with the photon line and thus the measured decay time should be larger than the $k_\parallel = 0$ value. If the exciton population is near thermal equilibrium, then the decay time for the spectrally integrated PL intensity coincides with the inverse of the thermally averaged decay rate. For a 100-Å $GaAs/Al_xGa_{1-x}As$ QW, the PL decay time assuming only free excitons was found to be[9] $34T$ ps (here T is the temperature in kelvins) in the temperature range 5 K$< T <$150 K above which scattering with LO phonons[2,11] becomes important.[12] In what follows we shall refer to this as the high-temperature regime. It is argued that at lower temperatures a significant fraction of the exciton population becomes trapped due to interface roughness and impurities.[13] The model of Ref. 9, which predicts the linear temperature dependence of the PL decay time, assumes rapid scattering of the excitons (rapid on the time scale of the radiative decay) and neglects the role of excitons bound to inhomogeneities in the QW. In the following we consider the effects on the PL decay time due to excitons weakly localized by well-width fluctuations, finite scattering lifetime (dephasing), and finite spatial coherence. By weakly localized we mean that the internal-motion wave function of the exciton is adequately approximated by the expression in a perfect QW. The model for localization is similar to that of Ref. 14; however, here we also use the model to predict the

lifetimes of the states. For QW's of width $L_z < 150$ Å we find that the localized states have an important effect on the PL decay time over the entire temperature range $T < 150$ K. In wider QW's, the defects are not sufficiently strong to give rise to a large density of localized states. Instead, finite temporal-coherence effects on the free-exciton recombination due to the dephasing are expected to be important.

This paper is organized as follows. In Sec. II we outline the Green's-function method by which the radiative self-energy is obtained neglecting the dephasing processes. Section III is concerned with the role of localized excitons in determining the PL decay time. In Sec. IV we discuss the effect of nonzero homogeneous linewidth (finite phase coherence) on the PL decay time. In Sec. V we present our conclusions.

II. RADIATIVE SELF-ENERGY

In this section we derive the proper radiative self-energy of a QW exciton interacting with the electromagnetic (em) field. We assume a Wannier exciton formed from two doubly degenerate subbands. The QW single-particle Bloch states in the conduction (valence) subband are labeled c_σ ($v_{\sigma'}$) where σ (σ') is a spin index. We use the label $(cv)_s$ to denote electron-hole pair $c_\sigma v_{\sigma'}$. Thus s is a spin index for the exciton. We begin with the many-body Hamiltonian for excitons (without exciton-exciton interactions) interacting with the em field $\mathcal{H}=\mathcal{H}_{\rm ex}+\mathcal{H}_f+\mathcal{W}$. $\mathcal{H}_{\rm ex}$ is the Hamiltonian for the noninteracting excitons,

$$\mathcal{H}_{\rm ex}=\sum_{\mathbf{k}_\parallel s} E_{\rm ex}(\mathbf{k}_\parallel) b^*_{\mathbf{k}_\parallel s} b_{\mathbf{k}_\parallel s},$$

where $b^*_{\mathbf{k}_\parallel s}$ is an exciton creation operator for a QW exciton with center-of-mass wave vector $\mathbf{k}_\parallel$ and of spin state s. $E_{\rm ex}(\mathbf{k}_\parallel)$ is the exciton dispersion. $\mathcal{W}$ is the interaction which can be written as[15,16]

$$\mathcal{W}=-\frac{e}{c_0}\sum_i \mathbf{A}(\mathbf{R}_i)\cdot\dot{\mathbf{R}}_i + \frac{1}{2i\hbar}\left(\frac{e}{c_0}\right)^2\sum_i \mathbf{A}(\mathbf{R}_i)\cdot[\mathbf{R}_i,\mathbf{A}(\mathbf{R}_i)\cdot\dot{\mathbf{R}}_i],$$

where $\mathbf{R}_i$ are the electron coordinates and $\mathbf{A}(\mathbf{r})$ is the vector potential. The sum over i is over all the electrons in the solid and the velocity $\dot{\mathbf{R}}_i=i\hbar^{-1}[\mathcal{H}_{\rm ex},\mathbf{R}_i]$. $\mathcal{W}$ is written in second-quantized form in terms of electron field operators which are expanded as a sum of single-particle QW eigenstates. One finally obtains

$$\mathcal{W}=\sum_{\mathbf{k}_\parallel s} i(b^*_{\mathbf{k}_\parallel s}-b_{-\mathbf{k}_\parallel s})\mathbf{C}_s\cdot\int dz\, f_c(z)f_v(z)\mathbf{A}(z,\mathbf{k}_\parallel) + \sum_{\mathbf{k}_\parallel s}\frac{1}{E_{\rm ex}(\mathbf{k}_\parallel)}\mathbf{C}^*_s\cdot\int dz_1\int dz_2\, f_c(z_1)f_v(z_1)\mathbf{A}(z_1,\mathbf{k}_\parallel)\mathbf{A}(z_2,-\mathbf{k}_\parallel)f_c(z_2)f_v(z_2)\cdot\mathbf{C}_s,$$

where

$$\mathbf{C}_s=\frac{e}{c_0}F_{\rm ex}(\mathbf{0})\langle(cv)_s|\mathbf{R}|0\rangle E_{\rm ex}(\mathbf{k}_\parallel)/\hbar.$$

Here $F_{\rm ex}(\mathbf{0})$ is the exciton envelope function evaluated at zero electron-hole separation and $f_c(z)$ [$f_v(z)$], chosen to be real, is the single-particle envelope function for the conduction (valence) subband. $\langle(cv)_s|\mathbf{R}|0\rangle$ is the dipole matrix element between the pair state $|(cv)_s\rangle$ and the crystal ground state $|0\rangle$ (filled valence band and empty conduction band). $\langle(cv)_s|\mathbf{R}|0\rangle$ gives the spin selection rules for the polarization given by the direction of $\mathbf{R}$. We consider the four excitons formed out of the two doubly degenerate subbands and we neglect the exchange interaction which partially lifts this degeneracy by a small amount. $\mathcal{H}_f$ is the Hamiltonian for the noninteracting em field.

The proper radiative self-energy $\hbar\Sigma_s(k)$ for a given exciton spin state is obtained using standard techniques.[17] A similar treatment has been applied to Frenkel excitons.[18] The only modification is that we treat Wannier excitons of reduced dimensionality. We consider the correlation function $\mathcal{D}_s(\tau,\mathbf{k}_\parallel)$ for the operator $a_{\mathbf{k}_\parallel s}=b_{\mathbf{k}_\parallel s}-b^*_{-\mathbf{k}_\parallel s}$ at nonzero temperature. The temperature correlation functions for the noninteracting and interacting systems are, respectively,

$$\mathcal{D}^{(0)}_s(\tau,\mathbf{k}_\parallel)=-\langle T_\tau \bar{a}_{\mathbf{k}_\parallel s}(\tau)\bar{a}^*_{\mathbf{k}_\parallel s}(0)\rangle,$$
$$\mathcal{D}_s(\tau,\mathbf{k}_\parallel)=-\langle T_\tau a_{\mathbf{k}_\parallel s}(\tau)a^*_{\mathbf{k}_\parallel s}(0)\rangle$$

where

$$\bar{a}_{\mathbf{k}_\parallel s}(\tau)=e^{\tau\mathcal{H}_{\rm ex}/\hbar}a_{\mathbf{k}_\parallel s}e^{-\tau\mathcal{H}_{\rm ex}/\hbar},$$
$$a_{\mathbf{k}_\parallel s}(\tau)=e^{\tau\mathcal{H}/\hbar}a_{\mathbf{k}_\parallel s}e^{-\tau\mathcal{H}/\hbar}.$$

T_τ is the ordering operator and $\langle\cdots\rangle$ denotes the thermodynamical average. Detailed definitions of the symbols are given in Ref. 17. The noninteracting correlation function in the frequency domain is

$$\mathcal{D}^{(0)}_s(k)=\frac{2E_{\rm ex}(\mathbf{k}_\parallel)}{(i\omega)^2-E_{\rm ex}(\mathbf{k}_\parallel)^2}=\mathcal{G}^{(0)}_s(k)+\mathcal{G}^{(0)}_s(-k),$$

where $\mathcal{G}^{(0)}_s(k)=[i\omega-E_{\rm ex}(\mathbf{k}_\parallel)]^{-1}$ is the zeroth-order exciton Green's function and $k=(i\omega,\mathbf{k}_\parallel)$. The S-matrix expansion[19] of $\mathcal{D}_s(k)$ is easily carried out. One obtains

$$\mathcal{D}_s(k)=\mathcal{D}^{(0)}_s(k)+\frac{2}{E_{\rm ex}(\mathbf{k}_\parallel)}\mathcal{K}'_s(k)\mathcal{D}^{(0)}_s(k) + \mathcal{D}^{(0)}_s(k)\mathcal{K}'_s(k)\mathcal{D}^{(0)}_s(k)+\cdots, \quad (1)$$

where $\mathcal{K}'_s(k)=\mathbf{C}_s^*\overleftrightarrow{V}(k)\mathbf{C}_s$ and

$$\overleftrightarrow{V}(k)=\int d^3r_1\int d^3r_2\; f_c(z_1)f_v(z_1)e^{-i\mathbf{k}_\parallel\cdot\mathbf{r}} \times\overleftrightarrow{\mathcal{K}}(\mathbf{r}_\parallel,z_1,z_2;i\varepsilon)f_c(z_2)f_v(z_2). \quad (2)$$

Here $\mathbf{r}=\mathbf{r}_1-\mathbf{r}_2$. The tensor function $\overleftrightarrow{\mathcal{K}}(\mathbf{r}_\parallel,z_1,z_2;i\varepsilon)$ in Eq. (2) is the photon Green's function and is derived in the Appendix.

We now choose axes so that the tensor $\overleftrightarrow{V}(k)$ is diagonal in the polarization index. We can then separate the self-energy into terms for the different polarizations ϵ and we have $V_{\epsilon\epsilon'}(k)=V_\epsilon(k)\delta_{\epsilon\epsilon'}$. In QW's $\hbar\Sigma_s(k)=\hbar\Sigma_{sL}(k)+\Sigma_{sT}(k)+\Sigma_{sZ}(k)$. The integrals in Eq. (2) for polarization ϵ are performed to give $V_\epsilon(k)=2\pi U_\epsilon(k)/\alpha$, where $\alpha=[k_\parallel^2-(i\omega/\hbar c)^2]^{1/2}$. In QW's there are three possibilities for ϵ, namely T, L, and Z,[20,21] which give

$$U_T(k)=-\mathcal{P},\qquad U_L(k)=\frac{\alpha^2}{(i\omega/\hbar c)^2}\mathcal{P},$$
$$U_Z(k)=-\frac{k_\parallel^2}{(i\omega/\hbar c)^2}\mathcal{P}+2\frac{\alpha}{(i\omega/\hbar c)^2}\mathcal{I} \quad (3)$$

with

$$\mathcal{P}=\int dz_1\int dz_2 f_c(z_1)f_v(z_1)f_v(z_2)f_c(z_2)e^{-\alpha|z|},$$
$$\mathcal{I}=\int dz_1|f_c(z_1)|^2|f_v(z_1)|^2.$$

Here $z=z_1-z_2$.[22]

The two terms explicitly given in Eq. (1) are the only types that arise. The series (1) can then be summed exactly giving the Dyson equation $\mathcal{D}_s(k)=\mathcal{D}_s^{(0)}(k)+\mathcal{D}_s^{(0)}(k)\hbar\Sigma_s(k)\mathcal{D}_s(k)$. Finally, one obtains the interacting Green's function

$$\mathcal{D}_s(k)=\frac{\mathcal{D}_s^{(0)}(k)}{1-\mathcal{D}_s^{(0)}(k)\hbar\Sigma_s(k)}=\frac{2E_{\rm ex}(\mathbf{k}_\parallel)}{(i\omega)^2-E_{\rm ex}(\mathbf{k}_\parallel)^2-2E_{\rm ex}(\mathbf{k}_\parallel)\hbar\Sigma_s(k)}, \quad (4)$$

where the proper radiative self-energy $\hbar\Sigma_s(k)$ is

$$\hbar\Sigma_s(k)=\frac{(i\omega)^2}{E_{\rm ex}(\mathbf{k}_\parallel)^2}\mathcal{K}'_s(k)=\frac{2\pi}{\alpha}\frac{(i\omega)^2}{E_{\rm ex}(\mathbf{k}_\parallel)^2}\sum_\epsilon|\mathbf{C}_s\cdot\hat{\mathbf{n}}_\epsilon|^2U_\epsilon(k). \quad (5)$$

The poles of $\mathcal{D}_s(k)$ give the single-exciton excitation energies (polariton dispersion) of the interacting system. $\mathcal{D}_s(k)$ is analytically continued to real values of $i\omega$ via $i\omega\to E+i\delta$ with δ a positive infinitesimal. From the self-energy we can extract the real energy shift $\hbar\Pi_s(E,\mathbf{k}_\parallel)=\mathrm{Re}\hbar\Sigma_s(E,\mathbf{k}_\parallel)$ and the intrinsic radiative decay rate $\Gamma_s(E,\mathbf{k}_\parallel)=-\mathrm{Im}\Sigma_s(E,\mathbf{k}_\parallel)$. We shall only require the decay rate evaluated on energy shell, $E=E_{\rm ex}(\mathbf{k}_\parallel)\approx E_{\rm ex}(\mathbf{0})$. (The real part of the self-energy is small and shall be neglected.) This gives the decay rate $\Gamma_s(k_\parallel)=\Gamma_s[E_{\rm ex}(\mathbf{0}),\mathbf{k}_\parallel]$ for spin state s. For $k_\parallel<\kappa_{\rm ex}=E_{\rm ex}(\mathbf{0})/(\hbar c)$, we have

$$\hbar\Gamma_{sT}(k_\parallel)=2\pi|F_{\rm ex}(\mathbf{0})|^2\,|\langle(cv)_s|\mathbf{R}\cdot\hat{\mathbf{n}}_T|0\rangle|^2\times\epsilon_\infty^{-1}\kappa_{\rm ex}^2\alpha_{\rm ex}^{-1}\mathrm{Re}\mathcal{P},$$
$$\hbar\Gamma_{sL}(k_\parallel)=-2\pi|F_{\rm ex}(\mathbf{0})|^2\,|\langle(cv)_s|\mathbf{R}\cdot\hat{\mathbf{n}}_L|0\rangle|^2\times\epsilon_\infty^{-1}\alpha_{\rm ex}\mathrm{Re}\mathcal{P},$$
$$\hbar\Gamma_{sZ}(k_\parallel)=2\pi|F_{\rm ex}(\mathbf{0})|^2\,|\langle(cv)_s|\mathbf{R}\cdot\hat{\mathbf{n}}_Z|0\rangle|^2\times\epsilon_\infty^{-1}k_\parallel^2\alpha_{\rm ex}^{-1}\mathrm{Re}\mathcal{P},$$

where $\hat{\mathbf{n}}_\epsilon$ is a unit vector in the direction of the polarization and $\alpha_{\rm ex}=(k_\parallel^2-\kappa_{\rm ex}^2-i\delta\,\mathrm{sgn}\kappa_{\rm ex})^{1/2}$. For $k_\parallel>\kappa_{\rm ex}$, $\hbar\Gamma_s(k_\parallel)=0$. For the experimental data we shall discuss below, the four different spin states of the excitons are equally likely. Therefore we must average expression (5) over s. We denote the resulting rates $\Gamma_\epsilon(k_\parallel)=\sum_s\Gamma_{s\epsilon}(k_\parallel)/4$:

$$\Gamma_T(k_\parallel)=\frac{1}{4}\Gamma_{0,\parallel}\frac{\kappa_{\rm ex}}{\sqrt{\kappa_{\rm ex}^2-k_\parallel^2}},$$
$$\Gamma_L(k_\parallel)=\frac{1}{4}\Gamma_{0,\parallel}\frac{\sqrt{\kappa_{\rm ex}^2-k_\parallel^2}}{\kappa_{\rm ex}}, \quad (6)$$
$$\Gamma_Z(k_\parallel)=\frac{1}{4}\Gamma_{0,\perp}\frac{k_\parallel^2}{\kappa_{\rm ex}\sqrt{\kappa_{\rm ex}^2-k_\parallel^2}},$$

where

$$\Gamma_{0,\parallel}=\frac{\pi}{m_0c_0\sqrt{\epsilon_\infty}}e^2f_\parallel,\quad \Gamma_{0,\perp}=\frac{\pi}{m_0c_0\sqrt{\epsilon_\infty}}e^2f_\perp.$$

Here f_j is the oscillator strength per unit area given by $f_j=2m_0E_{\rm ex}(\mathbf{0})|F_{\rm ex}(\mathbf{0})|^2\sum_s|\langle(cv)_s|\mathbf{R}\cdot\hat{\mathbf{n}}_j|0\rangle|^2\mathrm{Re}\mathcal{P}/\hbar^2$ with $j=\parallel,\perp$. $\parallel$ ($\perp$) denotes photon polarization parallel (perpendicular) to the QW plane. For narrow QW's, $\mathrm{Re}\mathcal{P}$ is the squared overlap between the electron and hole single-particle envelope functions. In particular, note that for heavy-hole excitons, we have $\sum_s|\langle(cv)_s|\mathbf{R}\cdot\hat{\mathbf{n}}_\parallel|0\rangle|^2=|\mu_{cv}|^2$ and $\sum_s|\langle(cv)_s|\mathbf{R}\cdot\hat{\mathbf{n}}_\perp|0\rangle|^2=0$, where μ_{cv} is the dipole matrix element between bulk Bloch states. For narrow QW's, $\mathcal{P}$ reduces to the squared overlap of the electron and hole single-particle envelope functions, and the expression for the oscillator strength takes its usual form. The decay time for the exciton is defined as that time over which the probability the exciton has not decayed is reduced by a factor of e. Thus the decay time, which is defined for the particles, is given by half the reciprocal of the rate, which is defined for the wave function. The *particle* decay rate of an optically active state is then given by $2\sum_s\Gamma_{s\epsilon}(k_\parallel)/2$, which gives the results of Ref. 9. [Note that in Eq. (3) of Ref. 9 the decay rates are defined as particle decay rates for the optically active states.] As an example of Eq. (6) we consider the $1s$ heavy-hole exciton in a 100-Å $GaAs/Al_xGa_{1-x}As$ for which $f_\parallel=5\times10^{-4}$ Å^{-2}.[23] Then $\Gamma_{0,\parallel}^{-1}=25.5$ ps.[9] The decay time for a non-spin-polarized state at $k_\parallel=0$ is then

$[2(\frac{1}{4}+\frac{1}{4})\Gamma_{0,\|}]^{-1}=25.5$ ps while for an optically active state it is half this value.

III. EFFECT OF LOCALIZED EXCITONS

In this section we consider the effect of exciton localization on the temperature dependence of the PL decay time. We assume an interface defect due to a well-width fluctuation of thickness $\frac{a}{2}$ and lateral size b. The model potential is then taken to be $\mathcal{H}_d=-V_e\exp[-(x_e^2+y_e^2)/(2b^2)]\Xi(z_e)-V_h\exp[-(x_h^2+y_h^2)/(2b^2)]\Xi(z_h)$, where $\Xi(z_i)=1$ for $\frac{L_z}{2}\le z_i\le\frac{L_z}{2}+\frac{a}{2}$ and 0 otherwise. V_e and V_h are the offsets in the conduction and valence bands and L_z is the well width. For GaAs/AlAs QW's, a monolayer fluctuation gives $\frac{a}{2}=2.83$ Å—half the bulk lattice constant. We assume a variational wave function for the localized exciton of the form[14]

$$\psi_{\rm loc}(\mathbf{r}_e,\mathbf{r}_h)=\eta(\pi)^{-1/2}e^{-R_\|^2\eta^2/2}F_{\rm ex}(\mathbf{r}_\|)\times f_c(z_e;x_e,y_e)f_v(z_h;x_h,y_h)\quad\text{(case I)},$$

$$\psi_{\rm loc}(\mathbf{r}_e,\mathbf{r}_h)=\eta(\pi)^{-1/2}e^{-R_\|^2\eta^2/2}F_{\rm ex}(\mathbf{r}_\|)f_c(z_e)f_v(z_h)\quad\text{(case II)}$$

where $\mathbf{R}_\|$ and $\mathbf{r}_\|$ are the in-plane center-of-mass and relative coordinates and η is the variational parameter. Here $f_c(z_e;x_e,y_e)$ $[f_v(z_h;x_h,y_h)]$ is the envelope function for the electron (hole) in a well of width $L_z+\frac{a}{2}\exp[-(x_e^2+y_e^2)/(2b^2)]$ [or $L_z+\frac{a}{2}\exp\{-(x_h^2+y_h^2)/(2b^2)\}$]. Case I is applied to QW's where L_z and b are sufficiently large so that the single-particle states are essentially relaxed into the interface defect. For narrow QW's with small b and small band offsets, the single-particle state does not relax into the interface defect and case II is applicable. We write $\langle\psi_{\rm loc}|\mathcal{H}_d|\psi_{\rm loc}\rangle$ as

$$\langle\psi_{\rm loc}|\mathcal{H}_d|\psi_{\rm loc}\rangle=\frac{\eta^2}{\pi}\int d^2R_\|\,e^{-R_\|^2\eta^2}V_{\rm eff}(R_\|),$$

where

$$V_{\rm eff}(R_\|)=\int d^2r_\|\int dz_e\int dz_h\mathcal{H}_d|F_{\rm ex}(\mathbf{r}_\|)\times f_e(z_e;x_e,y_e)\times f_v(z_h;x_h,y_h)|^2\quad\text{(case I)},$$

$$V_{\rm eff}(R_\|)=\int d^2r_\|\int dz_e\int dz_h\mathcal{H}_d|F_{\rm ex}(\mathbf{r}_\|)\times f_e(z_e)f_v(z_h)|^2\quad\text{(case II)}.$$

We assume a Gaussian form for the $1s$ excitonic envelope function $F_{\rm ex}(\mathbf{r}_\|)=(\lambda\sqrt{\pi})^{-1}\exp(-r_\|^2/2\lambda^2)$. The parameter λ is determined variationally. This gives

$$V_{\rm eff}(R_\|)=e^{-R_\|^2/2b^2}\sum_{i=e,h}\frac{\delta E_i}{\alpha_i\lambda^2}e^{\beta_i^2R_\|^2/4\alpha_i},$$

$$\alpha_i=\frac{1}{2}\left(\frac{m_j^*}{Mb}\right)^2+\frac{1}{\lambda^2},\qquad\beta_i=\frac{m_j^*}{Mb^2},$$

where $j\neq i$. Here m_e^* (m_h^*) is the in-plane electron (hole) subband effective mass. The potential strengths δE_e and δE_h are given by

$$\delta E_i=-\frac{\hbar^2\pi^2a}{2m_{i,\perp}^*L_z^3}\quad\text{(case I)},$$

$$\delta E_i=-V_iP_i\quad\text{(case II)}$$

where $m_{e,\perp}^*$ $(m_{h,\perp}^*)$ is the perpendicular electron (hole) effective mass and $P_i=\int dz_i\;\Xi(z_i)|f_i(z_i)|^2$ is the probability that the single-particle state is in the width fluctuation. The expression for case I is the result of an effective-medium approximation. Neglecting the kinetic energy due to the deformation of the single-particle states (only relevant for case I), one obtains the binding-energy enhancement $E_{\rm loc}=\langle\psi_{\rm loc}|\mathcal{T}+\mathcal{H}_d|\psi_{\rm loc}\rangle$ due to localization,

$$E_{\rm loc}=\frac{\hbar^2\eta^2}{2M}+\sum_{i=e,h}\frac{\delta E_i}{\lambda^2\alpha_i}\frac{\eta^2}{\eta^2+a_i^2},\tag{7}$$

where $a_i^2=1/(2b^2)-\beta_i^2/(4\alpha_i)$. Here $\mathcal{T}$ is the kinetic-energy operator for the center-of-mass motion. Minimizing the energy gives the optimal value of the variational parameter. η is determined by

$$0=\frac{\hbar^2}{2M}+\sum_{i=e,h}\frac{\delta E_i}{\lambda^2\alpha_i}\frac{a_i^2}{(\eta^2+a_i^2)^2}.\tag{8}$$

As Eq. (8) is quartic in η^2, it can be solved exactly; however, the values of the coefficients make it more convenient to obtain the roots numerically. For sufficiently small values of b (with $\frac{a}{2}$ fixed), η^2 in Eq. (8) is negative. Although the approach is variational, this indicates that there is no bound state associated with sufficiently small well-width fluctuations. Thus, for a given pair b and $\frac{a}{2}$, there is a critical well thickness L_c greater than which the fluctuation does not give rise to localized states. The scattering states associated with the defect and the effect on the PL decay time is considered in Sec. IV. For large b, the localized state approaches that of a free exciton in a well of width $L_z+\frac{a}{2}$. Using the variational envelope function for the center-of-mass motion, we find the decay rate of the localized exciton:

$$\Gamma_{L(T)}^{(\rm loc)}=\frac{1}{\eta^2\pi}\int_{k_\|<\kappa}d^2k_\|\;e^{-k_\|^2/\eta^2}\Gamma_{L(T)}(k_\|).\tag{9}$$

The interface-defect potential $V_{\rm eff}(R_\|)$ mixes free-exciton states of both small $k_\|$ (radiative) and large $k_\|$ (nonradiative) leading to long decay times compared with the free excitons. Thus

$$\Gamma_T^{(\rm loc)}=2\mu\Gamma_{0,\|}D(\mu),\qquad\Gamma_L^{(\rm loc)}=\Gamma_{0,\|}\left[1-\mu^{-1}D(\mu)\right].\tag{10}$$

$D(\mu)=\exp(-\mu^2)\int_0^\mu dt\exp(t^2)$ is tabulated in Ref. 24. The dimensionless quantity μ is defined by $\mu=\kappa/\eta$. Note that $\Gamma_Z^{(\rm loc)}=0$ for the heavy-hole exciton since $f_\perp=0$. Summing over the decay channels L and T, we obtain the effective decay rate for localized-exciton states $\Gamma^{(\rm loc)}=\Gamma_T^{(\rm loc)}+\Gamma_L^{(\rm loc)}=\frac{1}{4}H(\mu)\Gamma_{0,\|}$, where $H(\mu)=$

$[(2\mu-1/\mu)D(\mu)+1]$. For b small or large, we expect the lifetimes to approach the value for a non-spin-polarized exciton at $k_{\|}=0$. For b small, as mentioned above, there is no bound state. For large values of b, $\mu\rightarrow\infty$ in Eq. (8) and $H(\mu)\sim 2$ giving the expected lifetime for free excitons at $k_{\|}=0$. Figure 1 shows the lifetime $\tau=1/(2\Gamma^{(\mathrm{loc})})$ of a localized exciton as a function of b and $\frac{a}{2}$ for several L_z. The solid curves are for the model of case I and the dashed curves for case II. For the 50-Å QW case I is inaccurate because we expect the penetration of the single-particle envelope functions into the barrier to be large, while for the 150-Å QW case II is inaccurate. For the 150-Å QW, for $\frac{a}{2}=2.83$ and 5.66 Å the exciton is only barely localized for any b ($|E_{\mathrm{loc}}|<1$ meV in all cases) and thus the increase in the radiative decay time is quite small. Recently it was found experimentally[25] that for very high quality GaAs/AlAs QW's the inhomogeneous broadening is negligible for $L_z>150$ Å. The calculated lifetimes for the chosen values of b and $\frac{a}{2}$ are all on the order of 100 ps. Some of the values are slightly lower than the observed low-temperature PL decay times (typically in the range 0.25–1 ns).

For somewhat stronger binding to the interface defect, the approximations leading to the forms we use for $V_{\mathrm{eff}}(R_{\|})$ are invalid. Nevertheless, Eqs. (9) and (10), which express the radiative decay rate in terms of the parameter η, are still expected to hold qualitatively. η, however, must then be determined according to a more appropriate model. In such cases, μ is small. In the limit $\mu\rightarrow 0$, we have $H(\mu)\sim\frac{8}{3}\mu^2$. Then, $\Gamma^{(\mathrm{loc})}\propto\kappa^2/\eta^2$ and η^{-2} can be interpreted as a coherence area. In general, however, the form of $H(\mu)$ is such that no simple interpretation along these lines is possible. In the opposite limit of very weak or no binding appropriate for $L_z>L_c$, the resonances associated with the interface defects lie near or in the free-exciton continuum. In such a case scattering with the defects is expected to contribute to the lifetime of the exciton. These effects are discussed below in Sec. IV.

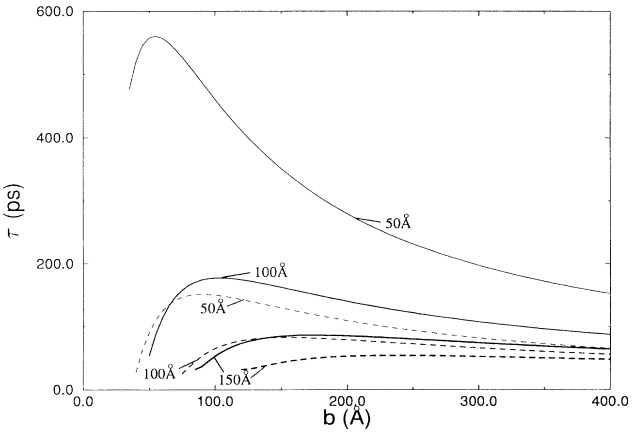

FIG. 1. The lifetime τ of a localized exciton as a function of b for $a/2=2.83$ for L_z =50-, 100-, and 150-Å GaAs/$Al_{0.3}Ga_{0.7}$As QW's. The solid curves are for case I and the dashed curves for case II. $\Gamma_{0,\|}^{-1}=16$, 26, and 53 ps for the L_2 = 50-, 100-, and 150-Å QW's, respectively, and indicate the lifetimes of non-spin-polarized free excitons at $k_{\|}=0$.

In order to examine semiquantitatively the temperature dependence of the PL decay time including both free and localized excitons, we have applied the simple model of Ref. 26 which assumes that the free- and localized-exciton populations are in thermal equilibrium with each other.[3,4] The temperature-dependent PL decay rate for localized and free excitons is then

$$\Gamma=(N_{\mathrm{loc}}\Gamma^{(\mathrm{loc})}+N_{\mathrm{fr}}\Gamma^{(\mathrm{fr})})/N, \tag{11}$$

where $N=N_{\mathrm{loc}}+N_{\mathrm{fr}}$ and N_{loc} and N_{fr} are the numbers of localized and free excitons, respectively. For $T>1$ K, $\Gamma^{(\mathrm{fr})}=2E_1\Gamma_{0,\|}/(3k_BT)$ [where $E_1=\hbar^2\kappa_{\mathrm{ex}}^2/(2M)$] is the thermally averaged decay rate of non-spin-polarized free excitons. (See Ref. 9 and Sec. IV below for the derivation.) This leads to a decay time of half the value given in Ref. 9. We assume that the density of localized states is narrow and centered on the energy of the free exciton less the binding-energy enhancement $|E_{\mathrm{loc}}|$ associated with particular values b and $\frac{a}{2}$. Furthermore we assume that, for the given values b and $\frac{a}{2}$, $L_z<L_c$. Thus $N_{\mathrm{fr}}=4Mk_BT/(2\pi\hbar^2)$ (the factor of 4 is due to the fourfold spin degeneracy) and $N_{\mathrm{loc}}=N_D\exp[|E_{\mathrm{loc}}|/(k_BT)]$ where $N_D\sim 10^{10}$–10^{11} cm^{-2} (Refs. 27 and 28) is the effective areal density of defects. At high temperature, $N_{\mathrm{fr}}\gg N_{\mathrm{loc}}$ and so the effective PL decay time in the high-temperature regime can be written

$$\tau(T)=\frac{1}{2\Gamma^{(\mathrm{fr})}}\left(1+\frac{N_D\Gamma^{(\mathrm{loc})}}{N_{\mathrm{fr}}\Gamma^{(\mathrm{fr})}}\right)^{-1}. \tag{12}$$

Using the expressions given above, $N_{\mathrm{fr}}\Gamma^{(\mathrm{fr})}$ is independent of temperature (above ~ 1 K).[9] For 50- and 100-Å GaAs/Al_xGa_{1-x}As QW's ($b=200$ Å, $\frac{a}{2}=2.83$ Å, case II), $\Gamma_{0,\|}^{-1}=17$ and 26 ps and $[2\Gamma^{(\mathrm{loc})}]^{-1}=109$ and 79 ps, respectively, for $b=200$ Å and $\frac{a}{2}=2.83$ Å. This gives $N_D\Gamma^{(\mathrm{loc})}/(N_{\mathrm{fr}}\Gamma^{(\mathrm{fr})})\approx 7.5\times10^{-11}N_D$ cm^2, $1.5\times10^{-10}N_D$ cm^2 for the 50- and 100-Å QW's. This number is typically on the order of unity or 10 given the values of N_D cited above.[27,28] (Other values of b and case II reduce these numbers by a factor of $\sim\frac{1}{2}$.) Thus, even in the high-temperature regime the PL decay time *depends on the density of interface defects* provided there is a significant density of localized states associated with defects.

The calculations in this section is variational in nature. The relaxation of the single-particle states into the defect has been taken into account only approximately.[29] Cases I and II can be viewed approximately as giving upper and lower bounds to the radiative lifetime.

IV. PHASE-COHERENCE EFFECTS

The decay rates associated with the free-excitons in the foregoing discussion were assumed to be quantum mechanically coherent spatially over the entire QW and temporally on time scales giving energy spreads small compared with E_1. Manifestly, these assumptions do not hold. The extended states undergo scattering among themselves and with phonons. In addition, the extended

states are expected to be influenced by interface roughness by introducing spatially random phase variations. The previous treatment assumed the existence of scattering among the excitons sufficiently rapid to maintain a thermal population while the radiative decay is taking place. In this section we consider[30] the effect on the free-exciton contribution to the PL decay time due to dephasing and show that in the proper limits the results reduce to models previously proposed in the literature.[1,9]

Early on it was suggested that the finite quantum-mechanical coherence plays an important part in the PL decay rate.[1] Subsequently, in the model of Refs. 31 and 32 the PL decay time is assumed to be determined by the smaller of an area[33] A_c [defined in terms of the temperature-dependent homogeneous linewidth (full width at half maximum) $\hbar\Gamma_h$] and the area A_l of localizing defects. A thermal-equilibrium model is used with populations of free and localized excitons. It is further assumed that only that fraction of the exciton population lying within the homogeneous linewidth can decay radiatively.[1] The decay time $\tau \propto 1/\min(A_c, A_l)$ in the high-temperature regime depends on the dephasing time $1/\Gamma_h$. In addition, the areal density of defects enters the model in analogy to Eq. (11). In many respects, the model of Ref. 31 is similar to the one presented above in Sec. III. The primary differences between the model of Refs. 1, 31, and 32 and of Ref. 9 are (1) exactly which states are optically active and (2) the inclusion of a coherence area. Here we present a unified picture of PL decay which reduces in the proper limits to the results of Ref. 9 and to results similar to the model of Refs. 1, 31, and 32. The results do not depend upon a definition of the coherence area, but are obtained directly from the homogeneous linewidth. Toward the end of this section we comment on the connection between the finite spatial and temporal coherence of the exciton due to interface defects.

We proceed to discuss the relationship between the homogeneous linewidth and the PL decay time. Because the microscopic relationship between the decay time and the dephasing time is expected to depend upon the dephasing mechanisms operating in a given sample, we adopt a phenominological approach based upon temperature Green's functions. We further assume, as in Sec. III, that the radiative decay rate is small compared to the dephasing rate. Consequently, equilibrium populations are employed and we identify the dephasing rate (for the particles) with Γ_h. The desired quantity is the thermal average of the imaginary part of the radiative self-energy $\hbar\tilde{\Sigma}_s(k)$ including the effects of dephasing. The dephasing is assumed to be caused by some unspecified quasi-elastic-scattering mechanism which we represent by $Q(k)$. For example, $Q(k)$ might represent an exciton-exciton interaction or the propagator and coupling contant for low-energy acoustic phonons. The associated proper self-energy $\hbar\Theta_s(k) = \int d^3q\, Q(k-q)\mathcal{D}_s^{(0)}(q)$ to lowest order where $\int d^3k = (2\pi)^{-2}\int d^2k_\parallel \beta^{-1}\sum_{i\omega}$. The radiative self-energy $\hbar\tilde{\Sigma}_s(k)$ is diagrammatically represented by the sum of $\hbar\Sigma_s(k)$, the diagram $\hbar\Theta_s(k)$ in which all bare propagators $\mathcal{D}_s^{(0)}(k)$ are replaced by $\mathcal{D}_s(k)$, and $-\hbar\Theta_s(k)$. (This last term is nonradiative.) The thermal average is then

$$\Gamma^{(\mathrm{fr})} = -\mathrm{Im}N^{-1}\sum_s \int d^3k\, \tilde{\Sigma}_s(k)\tilde{\mathcal{D}}_s(k),$$
$$N = \sum_s \int d^3k\, \tilde{\mathcal{D}}_s(k), \tag{13}$$

where $\tilde{\mathcal{D}}_s(k)$ is the Green's function including both the dephasing and the radiative effects. To lowest order in the dephasing, Eq. (13) becomes

$$\begin{aligned}\Gamma^{(\mathrm{fr})} &= -\mathrm{Im}N^{-1}\sum_s \int d^3k\, \tilde{\Sigma}_s(k)\mathcal{D}_s^{(0)}(k) \\ &= N^{-1}\sum_s \int d^3k\, \Gamma_s(k)\bar{\mathcal{D}}_s(k), \\ N &= \sum_s \int d^3k\, \mathcal{D}_s^{(0)}(k),\end{aligned}$$

where $\Gamma_s(E,\mathbf{k}_\parallel) = -\mathrm{Im}\Sigma_s(E,\mathbf{k}_\parallel)$. Here the Green's function $\bar{\mathcal{D}}_s(k)$ only includes the dephasing effects. $\bar{\mathcal{D}}_s(k)$ is used to define a spectral density function $A(E,\mathbf{k}_\parallel)$ independent of s via $A(E,\mathbf{k}_\parallel) = -2\,\mathrm{Im}\bar{D}_s^{(r)}(E,\mathbf{k}_\parallel)$ where $\bar{D}_s^{(r)}(E,\mathbf{k}_\parallel) = \lim_{i\omega\to E+i\delta}\bar{\mathcal{D}}_s(i\omega_n,\mathbf{k}_\parallel)$ and $\bar{D}_s^{(r)}(E,\mathbf{k}_\parallel)$ is the retarded Green's function for the operator $a_{\mathbf{k}_\parallel s}$ including the dephasing. We thus take

$$A(E,\mathbf{k}_\parallel) = \frac{\hbar\Gamma_h/2}{(E-E')^2 + (\hbar\Gamma_h/2)^2} - \frac{\hbar\Gamma_h/2}{(E+E')^2 + (\hbar\Gamma_h/2)^2},$$

where $E' = E_{\mathrm{ex}}(\mathbf{k}_\parallel)$. The radiative self-energy is evaluated on energy shell. With these approximations, we can write Eq. (13) as

$$\begin{aligned}\Gamma^{(\mathrm{fr})} &= N^{-1}\int_{-\infty}^{\infty}\frac{dE}{2\pi}\, n_B(E) \\ &\quad\times \int_{E_{\mathrm{ex}}(\mathbf{0})}^{E_{\mathrm{ex}}(\mathbf{0})+E_1} dE'\, A(E,\mathbf{k}_\parallel) \\ &\quad\times \sum_{\epsilon=L,T}\Gamma_\epsilon(\mathbf{k}_\parallel), \\ N &= \int_{E_{\mathrm{ex}}(\mathbf{0})}^{\infty}\frac{dE}{2\pi}\,[n_B(E) - n_B(-E)]\end{aligned} \tag{14}$$

where $n_B(E)$ is the Bose-Einstein distribution. We now assume classical statistics and that the temperature satisfies $k_BT \ll E_{\mathrm{ex}}(\mathbf{0})$. For GaAs-based structures, the latter requirement is amply satisfied. $\Gamma^{(\mathrm{fr})}$ in Eq. (14) is now evaluated for different ranges of the parameters.

A. $\hbar\Gamma_h/2 \ll E_1$. (Note, however, $\hbar\Gamma_h \gg 2\hbar\Gamma_{0,\parallel}$ in order that the assumption of rapid dephasing holds.) The homogeneous broadening is negligible and the Lorentzian $A(E,\mathbf{k}_\parallel) \sim \pi\delta(E-E') - \pi\delta(E+E')$. One obtains

$$\Gamma^{(\mathrm{fr})} = \frac{2E_1}{3k_BT}\Gamma_{0,\|}, \qquad T > E_1/k_B \sim 1K. \tag{15}$$

This corresponds by construction to the result obtained above for infinite phase coherence and in Ref. 9. Note that the PL decay time $1/(2\Gamma^{(\mathrm{fr})})$ is half the result given in Ref. 9.

B. $\hbar\Gamma_h/2 \gg E_1$. Here the spectral density function is approximately constant over the E' integration allowing the replacement $A(E,\mathbf{k}_\|) \to A(E,\mathbf{0})$. In the E integral, we treat $A(E,\mathbf{0})$ as a constant within the homogeneous linewidth $A(E,\mathbf{0}) = \pi(\hbar\Gamma_h)^{-1}\{\theta[E-E_{\mathrm{ex}}(\mathbf{0})]\theta[E_{\mathrm{ex}}(\mathbf{0}) + \hbar\Gamma_h - E] - \theta[-E-E_{\mathrm{ex}}(\mathbf{0})]\theta[E_{\mathrm{ex}}(\mathbf{0}) + \hbar\Gamma_h + E]\}$ where $\theta(E)$ is the Heaviside step function. Thus

$$\Gamma^{(\mathrm{fr})} = \frac{2E_1}{3\hbar\Gamma_h}\left(1 - e^{-\hbar\Gamma_h/k_BT}\right)\Gamma_{0,\|}. \tag{16}$$

This agrees with the expression for the PL decay time employed in Refs. 1, 31, and 32 where the lifetime is modeled as $\tau \propto \hbar\Gamma_h/[1-\exp(-\hbar\Gamma_h/k_BT)]$. The constant of proportionality depends upon the microscopic mechanism for radiative decay which is not discussed in Ref. 1. Thus the effects of a finite dephasing time are obtained without an *ad hoc* relation between A_c and $\hbar\Gamma_h$. For temperatures low compared with $\hbar\Gamma_h/k_B$ we expect that the requirement of a thermal distribution is relaxed.[32] In the limit $k_BT \gg \hbar\Gamma_h/2$, Eq. (15) is again obtained. Thus, *at high temperature the homogeneous broadening is unimportant.* Instead, the radiative decay is bandwidth (E_1) limited.

We therefore conclude that the relevant energy scales which govern the behavior of $\Gamma^{(\mathrm{fr})}$ are k_BT and $\hbar\Gamma_h$ regardless of E_1. Up to constant factors, Eq. (15) corresponds to that of Ref. 9 and Eq. (16) to Refs. 1, 31, and 32. In the high-temperature regime, Eq. (15) [or Eq. (16)] is valid and thus applies to the experiments of Refs. 1 and 2 subject to the previous discussion concerning the effects of bound excitons. The experiments of Ref. 32 are more fruitfully considered in the light of Eq. (16).

The present treatment of the effect of the homogeneous linewidth on the PL decay time does not so far entirely answer the question of the effect of finite spatial coherence of the exciton on the observed lifetimes. We now consider the connection between the interface defects and the scattering among the extended exciton states. From the theory of potential scattering, the Green's function for the exciton in the presence of interface defects is $\mathcal{G}_s^{(\mathrm{sc})}(k) = [i\omega - E_{\mathrm{ex}}(\mathbf{k}_\|) - \hbar\Sigma_s^{(\mathrm{sc})}(k)]^{-1}$. The imaginary part of the proper self-energy then contributes to the broadening of the spectral density function. We now obtain a rough estimate of $-2\,\mathrm{Im}\hbar\Sigma_s^{(\mathrm{sc})}(k)$. We assume the exciton scatters elastically from the interface defect according to the effective potentials $V_{\mathrm{eff}}(R_\|)$ of Sec. III. We proceed to obtain the interface-defect scattering contribution to the self-energy. On energy shell, the imaginary part of the self-energy can be expressed to first order in the interface-defect density N_{loc} as[17]

$$\begin{aligned} 2\hbar\Gamma^{(\mathrm{sc})}(\mathbf{k}_\|) &= -2\,\mathrm{Im}\hbar\Sigma_s^{(\mathrm{sc})}[E_{\mathrm{ex}}(\mathbf{k}_\|),\mathbf{k}_\|] \\ &= -2N_{\mathrm{loc}}\mathrm{Im}T_{\mathbf{k}_\|\mathbf{k}_\|}, \end{aligned} \tag{17}$$

where the T matrix in the first Born approximation is

$$\begin{aligned} T_{\mathbf{k}_\|\mathbf{k}'_\|} &= V_{\mathbf{k}_\|\mathbf{k}'_\|} + \frac{1}{A}\sum_{\mathbf{k}''_\|}\frac{V_{\mathbf{k}_\|\mathbf{k}''_\|}V_{\mathbf{k}''_\|\mathbf{k}'_\|}}{E_{\mathrm{ex}}(\mathbf{k}_\|) - E_{\mathrm{ex}}(\mathbf{k}''_\|) + i\delta}, \\ V_{\mathbf{k}_\|\mathbf{k}'_\|} &= \int d^2R_\|\, V_{\mathrm{eff}}(R_\|)e^{-i(\mathbf{k}_\|-\mathbf{k}'_\|)\cdot\mathbf{R}_\|}, \end{aligned} \tag{18}$$

with A a unit of area in the QW plane. This gives

$$\begin{aligned} 2\hbar\Gamma^{(\mathrm{sc})}(\mathbf{k}_\|) = \frac{\pi^2 N_{\mathrm{loc}}M}{\hbar^2\lambda^2}\Bigg[&\frac{2\delta E_e\delta E_h}{\alpha_e a_e^2\alpha_h a_h^2}e^{-k_\|^2(a_e^{-2}+a_h^{-2})/4}I_0[k_\|^2(a_e^{-2}+a_h^{-2})/4] \\ &+\frac{\delta E_e^2}{\alpha_e^2 a_e^4}e^{-k_\|^2/2a_e^2}I_0(k_\|^2/2a_e^2) + \frac{\delta E_h^2}{\alpha_h^2 a_h^4}e^{-k_\|^2/2a_h^2}I_0(k_\|^2/2a_h^2)\Bigg], \end{aligned} \tag{19}$$

where $I_0(x)$ is a modified Bessel function of the second kind. Note that this result is independent of the sign of the potential, i.e., the scattering rate is the same whether the barrier material penetrates into the well or vice versa. Provided $Mb/m_j^* \gg \lambda$, Eq. (19) for case I is similar to the model of Ref. 34 where the interface-defect scattering rate due to a slightly different potential is obtained by means of Fermi's golden rule. For $Mb/m_j^* \ll \lambda$ and $b < \kappa_{\mathrm{ex}}^{-1}$, the factor $\exp(-x)I_0(x)$ is near unity within the bandwidth E_1 and so $2\hbar\Gamma^{(\mathrm{sc})}(\mathbf{k}_\|)$ is nearly independent of $k_\|$ for the radiative states. Thus, although the effects under discussion are due to spatial inhomogeneities in the QW, they give rise to a contribution to the homogeneous linewidth of approximately Lorentzian shape. For $Mb/m_j^* \gg \lambda$ and $b > \kappa_{\mathrm{ex}}^{-1}$, the $k_\|$ dependence is more important. Then $2\hbar\Gamma^{(\mathrm{sc})}(\mathbf{k}_\|)$ is a decreasing function of $k_\|$. As temperature is increased and more excitons occupy states with larger $k_\|$, the interface-defect scattering contribution to the homogeneous linewidth (integrated over the spectral line) is reduced. Because of the uncertainty in the value of N_D and its dependence on b and $\frac{a}{2}$, a quantitative prediction of this effect is made difficult. A small reduction with increasing temperature ($T < 50$ K) in the homogeneous linewidth in $\mathrm{GaAs/Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$ QW's has been observed.[35] This effect was attributed to the thermalization of excitons bound to interface defects[35] and was strongest in a 142-Å QW where the typical value of the binding-energy enhancement due to the localization is $|E_{\mathrm{loc}}| \sim 0.5$ meV. The effect, however, was quite pronounced up to 50 K. This suggests that the origin of the narrowing of the line with increasing temperature is due primarily to the scattering of free excitons with well-width fluctuations. The actual situation is much more complicated due to the presence of several scatter-

ing mechanisms. In addition, the quoted homogeneous linewidth might also contain an inhomogeneous contribution.

Typical numerical results for $2\hbar\Gamma^{(sc)}(\mathbf{k}_{\|})$ are given in Ref. 34 where it is implicitly assumed that $Mb/m_j^* \gg \lambda$. Under the same assumption, Eq. (19) gives $2\hbar\Gamma^{(sc)}(\mathbf{k}_{\|}) \sim 1$ meV for $L_z = 100$ Å, $b = 200$ Å, $\frac{a}{2} = 2.83$ Å, and $N_{\rm loc} = 10^{11}$ cm^{-2} in a GaAs/AlAs QW in agreement with the results of Ref. 34. Unfortunately, however, numerical values for the interface-defect contribution to the homogeneous linewidth obtained from the models under consideration are limited in their ranges of validity and rather crude. The linewidth depends quartically (for $Mb/m_j^* \gg \lambda$) on b. In practice, there is a distribution as a function of b and $\frac{a}{2}$ of interface defects which is sample dependent and not accurately known. Also, the assumption of low interface-defect density which leads to the simple form of Eq. (17) is not justified for certain interface morphologies. We can, however, make the following tentative conclusions concerning the role of a finite coherence area on the radiative decay. For wide wells in which the density of localized states is expected to be small and interface defects cause scattering among the plane-wave exciton states, the interface defects contribute to the homogeneous broadening and the concept of a finite coherence area does not appear to be appropriate. The other extreme case is that of a narrow QW where the potentials associated with interface defects are strong enough to bind excitons on a length scale short compared with the wavelength of light in the medium. At very low temperatures, the populated states are primarily those bound by the interface fluctuations. Only in this case was it shown in Sec. III that the radiative decay can be characterized by a finite coherence area in the sense of Refs. 31 and 33.

V. CONCLUSION

Despite the uncertainty in some of the parameters in the calculations in this study, we can understand semiquantitatively the scatter in the published experimental data. PL decay-time measurements on various GaAs/AlAs and GaAs/Al_xGa_{1-x}As QW's yield very different results. We concentrate on two of the most extensive systematic studies of PL decay times.[1,2] Except at the lowest temperatures probed in these studies (~ 4 K), we do not expect phase-coherence effects on the free-excitons to dominate. In Ref. 1 the PL decay time τ was observed to increase with L_z and to have a temperature dependence moderately well descibed by the free-exciton result[9] with $\tau(5\text{ K}) \sim 0.5$–$1.5$ ns for QW's with 25 Å$\leq L_z \leq$150 Å. The results of Ref. 2 contrast with those of Ref. 1, where at low temperature, decay times of $\tau \sim 250$ ps were measured for several wells with $L_z = 20$, 40, and 80 Å. The decay times were about a factor of 5–10 less than in Ref. 1. Using Eq. (11), these discrepancies can be tentatively explained by assuming N_D of the QW's in Ref. 2 is $5-10$ times that in Ref. 1. Further comparison between the experimental data and the theory for QW's in which localized excitons and phase-coherence effects are important is hindered by the lack of detailed information on the interface morphology and the resulting density of localized states. Other helpful information could be provided by systematic studies of PL decay time as a function of temperature, well width, and interface quality (correlated with the Stokes shift between the PL and excitation spectra at low temperature). This information would provide impetus for more detailed theoretical investigations of the binding and scattering of excitons by interface defects and roughness.

It is important to state the limitations of the present theory. In order to apply the assumption of thermal equilibrium we have assumed that the dephasing takes place on a time scale much faster than the radiative decay. The dephasing time is typically a few ps.[34] The radiative decay time for optically active states is typically $\Gamma_{0,\|}^{-1} = 13$ ps (for a 100-Å GaAs/Al_xGa_{1-x}As QW). Therefore the assumption of rapid dephasing, and hence thermal distributions, might be near its limit of validity. Other omissions in the present treatment are the neglect of free carriers and the population of energetically higher states, whether excited exciton states or excitons formed from other subbands. The former contingency is treated in Ref. 25 by means of the Saha equation. The latter can be treated in the framework of the present study, though with more parameters.

To conclude, we calculate radiative lifetimes of localized excitons in GaAs/Al_xGa_{1-x}As QW's to be on the order of 100 ps. PL decay times are calculated assuming thermal equilibrium between localized and free states. We find that localized excitons play a role in the high-temperature regime for large N_D and small L_z. For wide QW's ($L_z > 150$ Å) the density of localized states is small and coherence effects are important for k_BT on the order of or smaller than the homogeneous linewidth. We find that the model of Refs. 1, 31, and 32 correlating the homogeneous linewidth and the PL decay time is justified at temperatures small compared with the homogeneous linewidth. Only for excitons localized on a scale short compared with the wavelength of light in the medium, however, does the coherence-area concept of Ref. 33 apply.

Note added. Results on interface-defect scattering times for electron (single-particle) states similar to Eq. (19) were obtained in R. Fierreira and G. Bastard, Phys. Rev. B **40**, 1074 (1989).

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APPENDIX

In this appendix we derive the photon Green's function for TE and TM optical modes. We consider a dielectric medium with in-plane translational symmetry and assume that the length scale for the variations in the di-

electric constant are large compared with L_z. The QW is taken to be at position z_0 in the structure and the local value of the speed of light and high-frequence dielectric constant are c and ϵ_∞, respectively. At the end of the calculation, we specialize the results to the case where the dielectric constant is the same throughout the medium. From Maxwell's equations, the wave equation for the electric field $\mathbf{E}$ is $(\boldsymbol{\nabla}\boldsymbol{\nabla} - \Delta \overleftrightarrow{1})\mathbf{E} = \frac{\omega^2}{c_0^2}\mathbf{D}$. Δ is the Laplacian operator and $\mathbf{D}$ is the displacement given by the constitutive relation $\mathbf{D} = \epsilon_\infty \mathbf{E} + 4\pi\mathbf{P}$. ϵ_∞ accounts for the high-frequency resonances of the medium, i.e., not including the exciton of interest, while $\mathbf{P}$ is the induced polarization of the exciton. We treat TE and TM modes separately.

1. TE modes

In this case the electric field satisfies $\boldsymbol{\nabla}\cdot\mathbf{E} = 0$. The wave equation takes the form

$$[(\hbar c)^2\Delta + \varepsilon^2]E_T = -\frac{4\pi}{\epsilon_\infty}\varepsilon^2 P_T,$$

where the subscript T denotes the in-plane component transverse to $\mathbf{k}_\parallel$. The Green's function for the vector potential near the location z_0 of the QW is obtained by considering a δ-function form for P_T. A simple calculation[36] shows it is given by

$$\mathcal{K}_T(\mathbf{r}_\parallel, z_1, z_2; i\varepsilon) = 4\pi(\hbar c)^2 \sum_{\nu\mathbf{k}_\parallel} \frac{\phi^*_{\nu\mathbf{k}_\parallel}(z_1)\phi_{\nu\mathbf{k}_\parallel}(z_2)}{(i\varepsilon)^2 - \varepsilon^2_{\nu\mathbf{k}_\parallel}} e^{i\mathbf{k}_\parallel\cdot\mathbf{r}_\parallel}.$$

The mode label ν runs over both confined and radiation modes. The z dependence of the modes is $\phi_{\nu\mathbf{k}_\parallel}(z)$ (assumed normalized). The in-plane dispersion of optical mode ν is given by $\varepsilon^2_{\nu\mathbf{k}_\parallel} = (\hbar c k_\parallel)^2 + (\hbar c\beta_{\nu\mathbf{k}_\parallel})^2$, where $\beta_{\nu\mathbf{k}_\parallel}$ is the local value of the propagation constant in the z direction at z_0.

2. TM modes

For TM modes, the divergence of the displacement vanishes, $\boldsymbol{\nabla}\cdot\mathbf{D} = 0$, and consequently the wave equation is

$$[(\hbar c)^2\Delta + \varepsilon^2]\mathbf{E} = -\frac{4\pi}{\epsilon_\infty}\varepsilon^2\left[\overleftrightarrow{1} + \frac{(\hbar c)^2}{\varepsilon^2}\boldsymbol{\nabla}\boldsymbol{\nabla}\right]\mathbf{P}.$$

For the L polarization we have

$$[(\hbar c)^2\Delta + \varepsilon^2]E_L = -\frac{4\pi}{\epsilon_\infty}\varepsilon^2\left[1 - \frac{(\hbar c k_\parallel)^2}{\varepsilon^2}\right]P_L$$

with the Green's function

$$\mathcal{K}_L(\mathbf{r}_\parallel, z_1, z_2; i\varepsilon) = 4\pi(\hbar c)^2 \sum_{\nu\mathbf{k}_\parallel}\left[1 - \frac{(\hbar c k_\parallel)^2}{(i\varepsilon)^2}\right] \times \frac{\phi^*_{\nu\mathbf{k}_\parallel}(z_1)\phi_{\nu\mathbf{k}_\parallel}(z_2)}{(i\varepsilon)^2 - \varepsilon^2_{\nu\mathbf{k}_\parallel}} e^{i\mathbf{k}_\parallel\cdot\mathbf{r}_\parallel}.$$

For the Z polarization, the appropriate wave equation is

$$[(\hbar c)^2\Delta + \varepsilon^2]E_Z = -\frac{4\pi}{\epsilon_\infty}\varepsilon^2\left[1 - \frac{(\hbar c\beta_{\nu\mathbf{k}_\parallel})^2}{\varepsilon^2}\right]P_Z.$$

The Green's function is

$$\mathcal{K}_Z(\mathbf{r}_\parallel, z_1, z_2; i\varepsilon) = 4\pi(\hbar c)^2 \sum_{\nu\mathbf{k}_\parallel}\left[1 - \frac{(\hbar c\beta_{\nu\mathbf{k}_\parallel})^2}{(i\varepsilon)^2}\right] \times \frac{\phi^*_{\nu\mathbf{k}_\parallel}(z_1)\phi_{\nu\mathbf{k}_\parallel}(z_2)}{(i\varepsilon)^2 - \varepsilon^2_{\nu\mathbf{k}_\parallel}} e^{i\mathbf{k}_\parallel\cdot\mathbf{r}_\parallel}.$$

The preceeding results are general and apply to cases both with and without dielectric mismatch.

For the case in which dielectric confinement is absent, the z dependence of the optical modes are given by planewaves. The replacements

$$\sum_\nu \phi^*_{\nu\mathbf{k}_\parallel}(z_1)\phi_{\nu\mathbf{k}_\parallel}(z_2) = \frac{1}{2\pi}\int_{-\infty}^{\infty} dk_z\, e^{-ik_z z}$$

(considered as an operator) and $\varepsilon^2_{\nu\mathbf{k}_\parallel} = (\hbar c k_\parallel)^2 + (\hbar c k_z)^2$ must be made in the Green's function. This gives

$$\mathcal{K}_T(\mathbf{r}_\parallel, z; i\varepsilon) = 4\pi(\hbar c)^2 \sum_{\mathbf{k}_\parallel} e^{i\mathbf{k}_\parallel\cdot\mathbf{r}_\parallel} I,$$

$$\mathcal{K}_L(\mathbf{r}_\parallel, z; i\varepsilon) = 4\pi(\hbar c)^2 \sum_{\mathbf{k}_\parallel} e^{i\mathbf{k}_\parallel\cdot\mathbf{r}_\parallel}\left[1 - \frac{(\hbar c k_\parallel)^2}{(i\varepsilon)^2}\right] I,$$

$$\mathcal{K}_Z(\mathbf{r}_\parallel, z; i\varepsilon) = 4\pi(\hbar c)^2 \sum_{\mathbf{k}_\parallel} e^{i\mathbf{k}_\parallel\cdot\mathbf{r}_\parallel} J,$$

where

$$I = \frac{1}{2\pi(\hbar c)^2}\int_{-\infty}^{\infty}\frac{dk_z\, e^{-ik_z z}}{(i\kappa)^2 - k_\parallel^2 - k_z^2} = -\frac{e^{-\alpha|z|}}{2\pi\alpha},$$

$$J = \frac{1}{2\pi(\hbar c)^2}\int_{-\infty}^{\infty}\left[1 - \frac{k_z^2}{(i\kappa)^2}\right]\frac{dk_z\, e^{-ik_z z}}{(i\kappa)^2 - k_\parallel^2 - k_z^2} = \frac{k_\parallel^2 I}{(i\kappa)^2} + \frac{\delta(z)}{(\hbar c)^2(i\kappa)^2}.$$

Carrying out the integrals over k_z gives

$$\mathcal{K}_T(\mathbf{r}_\parallel, z; i\varepsilon) = -\sum_{\mathbf{k}_\parallel}\frac{2\pi}{\alpha}e^{i\mathbf{k}_\parallel\cdot\mathbf{r}_\parallel}e^{-\alpha|z|},$$

$$\mathcal{K}_L(\mathbf{r}_\parallel, z; i\varepsilon) = \sum_{\mathbf{k}_\parallel}\frac{2\pi}{\alpha}e^{i\mathbf{k}_\parallel\cdot\mathbf{r}_\parallel}\frac{\alpha^2}{(i\kappa)^2}e^{-\alpha|z|},$$

$$\mathcal{K}_Z(\mathbf{r}_\parallel, z; i\varepsilon) = \sum_{\mathbf{k}_\parallel}\frac{2\pi}{\alpha}e^{i\mathbf{k}_\parallel\cdot\mathbf{r}_\parallel}\left(-\frac{k_\parallel^2}{(i\kappa)^2}e^{-\alpha|z|} + \frac{2\alpha}{(i\kappa)^2}\delta(z)\right).$$

The decomposition into TE and TM modes leads to a photon Green's-function diagonal in the mode number allowing us to drop the dyadic notation.

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