# Bose-Einstein statistics in thermalization and photoluminescence of quantum-well excitons

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Quasiequilibrium relaxational thermodynamics is developed to understand LA-phonon-assisted thermalization of Bose-Einstein distributed excitons in quantum wells. We study quantum-statistical effects in the relaxational dynamics of the effective temperature of excitons $T=T(t)$. When $T$ is less than the degeneracy temperature $T_0$, well-developed Bose-Einstein statistics of quantum-well excitons leads to nonexponential and density-dependent thermalization. At low bath temperatures $T_b \to 0$, the thermalization of quantum statistically degenerate excitons effectively slows down and $T(t) \propto 1/\ln t$. We also analyze the optical decay of Bose-Einstein distributed excitons in perfect quantum wells, and show how nonclassical statistics influences the effective lifetime $\tau_{opt}$. In particular, $\tau_{opt}$ of a strongly degenerate gas of excitons is given by $2\tau_R$, where $\tau_R$ is the intrinsic radiative lifetime of quasi-two-dimensional excitons. Kinetics of resonant photoluminescence of quantum-well excitons during their thermalization is studied within the thermodynamic approach and taking into account Bose-Einstein statistics. We find density-dependent photoluminescence dynamics of statistically degenerate excitons. Numerical modeling of the thermalization and photoluminescence kinetics of quasi-two-dimensional excitons are given for GaAs/$Al_xGa_{1-x}$As quantum wells. [S0163-1829(99)05507-1]

## I. INTRODUCTION

The fundamental features of relaxation and photoluminescence (PL) of the excitons in quantum wells (QW's) originate from the quasi-two-dimensionality (quasi-2D) of the system. In the last decade these basic processes in GaAs QW's have attracted continual attention.[1–11] The formation of QW excitons through LO-phonon cascade emission, LA-phonon, and carrier-carrier scattering completes within 20 ps after the initial excitation of electron-hole pairs.[4,7,12] The created hot QW excitons then thermalize through low-energy acoustic-phonon scattering. This process occurs in a sub-ns time scale. In GaAs QW's the relaxation of hot excitons can be observed through resonant PL from the optically active bulk modes which refer to a small in-plane momentum. Thus the rise and decay times of excitonic PL relate to the thermalization process. Furthermore, the PL kinetics of long-lifetime indirect excitons in a high-quality GaAs/$Al_xGa_{1-x}$As coupled quantum well (CQW) is now the subject of experimental study.[13] There has been recent progress in the investigation of thermalization of quasi-2D excitons in ZnSe QW's.[14] In the latter case the picosecond LA-phonon-assisted kinetics of QW excitons is visualized through LO-phonon-assisted PL for all in-plane modes $\mathbf{p}_\parallel$.

While the importance of interface disorder and the localization effects were recognized in the very first experimental[1] and theoretical[15] studies on the relaxation kinetics of QW excitons, the quality of GaAs QW's continuously improves from an inhomogeneous excitonic linewidth of about 7.5 meV (Ref. 1) toward a homogeneous one between 80 and 120 $\mu$eV.[8] In the present work we investigate LA-phonon-assisted relaxation kinetics in a perfect QW emphasizing the importance of Bose-Einstein (BE) statistics of quasi-2D excitons. In particular, we attribute the density-dependent PL kinetics reported, e.g., in Refs. 4 and 7, to nonclassical statistics of QW excitons. In the past, most theoretical modeling of the relaxation kinetics in QW's (Refs. 2,3,8,10,15 and 16) dealt with a classical gas of Maxwell-Boltzmann distributed excitons. Quantum-statistical effects were included in numerical simulations of the relaxation kinetics of a trapped quasi-2D exciton gas[17] and in the study of the exciton-biexciton law of mass action in QW's.[18]

Crossover from classical to quantum statistics occurs near the degeneracy temperature $T_0$, given by

$$k_B T_0 = \frac{2\pi}{g}\left(\frac{\hbar^2}{M_x}\right)\rho_{2D}, \tag{1}$$

where $M_x$ is the in-plane translational mass of a QW exciton, $\rho_{2D}$ is the 2D concentration of excitons, and $g$ is the spin degeneracy factor of QW excitons. We put $g=4$, because for GaAs QW's the exchange interaction is rather weak.[19] Furthermore, this interaction is completely suppressed for indirect excitons in CQW's. One has a classical gas of QW excitons at temperatures $T \gg T_0$. BE statistics smoothly develops with decreasing $T \sim T_0$, leading to occupation numbers $N_{\mathbf{p}_\parallel} \geqslant 1$ of the low-energy in-plane modes $\mathbf{p}_\parallel$. We will show that at helium temperatures nonclassical statistics already affects the thermalization process at moderate densities of QW excitons $\rho_{2D} \geqslant 3-5\times 10^{10}$ cm$^{-2}$.

In this paper we develop *relaxational thermodynamics*, and within this approach study how BE statistics influences thermalization and photoluminescence of quasi-2D excitons. Relaxational thermodynamics requires that exciton-exciton interaction is much stronger than exciton–LA-phonon cou-

 

pling, and is appropriate if the concentration $\rho_{2D}$ of QW excitons is larger than some critical density $\rho_{2D}^{c}$. In this case, the the exciton system establishes a quasiequilibrium temperature $T$. For GaAs QW's we will obtain an estimate $\rho_{2D}^{c} \simeq 1-3\times 10^{9}$ cm$^{-2}$. Equation (11), which is the basic equation of the relaxational thermodynamics of QW excitons, provides us with a unified description of the thermalization process. The thermodynamic approach yields the temporal evolution of the effective temperature of QW excitons $T=T(t)$ from the initial value $T_i=T(t=0)$ to the bath temperature $T_b$. While we study the thermalization process in both limits, classical and degenerate, of a gas of QW excitons, the most interesting results refer to $T \leqslant T_0$. In this case one finds a density-dependent nonexponential relaxation of quasi-2D excitons. Both acceleration and slowing down of the thermalization kinetics may occur due to BE statistics. However, with a decrease of the bath temperature $T_b$, slowing down of the relaxation starts to dominate in a quantum gas. In particular, for $T_b \to 0$ we derive $1/\ln t$ cooling law for QW excitons.

The thermalization kinetics of excitons in GaAs QW's can be observed through resonant PL from the in-plane radiative modes. Therefore we generalize the PL theory[3,16] to well-developed BE statistics of QW excitons, and numerically model the PL process in GaAs QW's. It will be shown that at low temperatures nonclassical statistics changes the law $\tau_{opt} \propto T$, where $\tau_{opt}$ is the effective radiative lifetime of QW excitons. Furthermore, we calculate a temperature-independent correction to the classical linear behavior of $\tau_{opt}(T)$. A density dependent component of the correction originates from BE statistics and can be traced even at high temperatures.

Relaxational thermodynamics refers to the phonon-assisted thermalization kinetics of QW excitons. We will analyze the relaxation kinetics due to bulk LA phonons, assuming the initial distribution of hot QW excitons is below the threshold for optical phonon emission. The bulk LA phonons are due to a semiconductor substrate of a QW. While the lifetime of long-wavelength acoustic phonons are on a sub-$\mu$s time scale, the scattering LA phonons leave and enter the QW area within a time $\sim L_z/v_s \sim 1-10$ ps ($L_z$ is the thickness of a QW, and $v_s$ is the longitudinal sound velocity). Therefore, we assume the Planck distribution of the LA phonons interacting with QW excitons. The LA-phonon-assisted kinetics will be treated for an isolated band of ground-state QW excitons. Being applied to heavy-hole excitons in GaAs QW's with $L_z \leqslant 100$ Å, this assumption means QW exciton energies $E \leqslant 10$ meV and temperatures less than 100 K. Only the exciton–LA-phonon deformation potential interaction is included in our model.

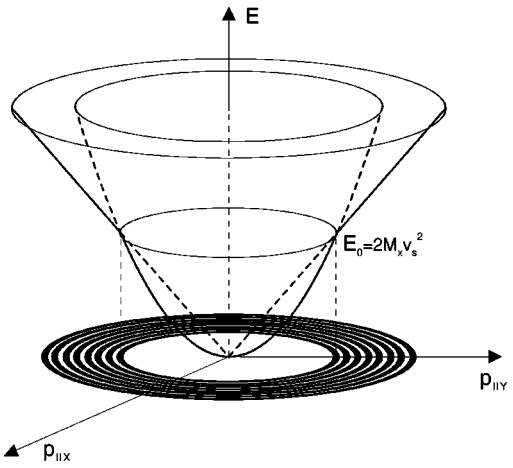


FIG. 1. Schematic picture of the energy-momentum conservation in scattering of QW excitons by bulk LA phonons. The parabolic and conical surfaces refer to the excitonic $E=\hbar^2 p_{\|}^2/2M_x$ and acoustic $E=\hbar v_s p_{\|}$ dispersions, respectively. The bold rings show the states $\hbar p_{\|} \geqslant 2M_x v_s$ of QW excitons which couple to the ground-state mode $\mathbf{p}_{\|}=0$.

The in-plane momentum is conserved in the scattering of QW excitons by bulk LA phonons. The momentum conservation in the $z$ direction (the QW growth direction) is relaxed. As a result, a scattered QW exciton interacts with a continuum spectral band of scattering LA phonons of a given direction of propagation. In contrast, an exciton in bulk semiconductors couples in Stokes or anti-Stokes scattering only with one phonon mode of a given direction. In Fig. 1 we depict a schematic picture of the states which are involved in the scattering of a QW exciton with in-plane momentum $\mathbf{p}_{\|}=0$. The energy state $E=0$ couples to the continuum energy states $E \geqslant E_0 = 2M_x v_s^2$, i.e., to the QW states which lie inside the acoustic cone given by $E=E(p_{\|})=\hbar v_s p_{\|}$. The energy $E_0$ is an important parameter of the relaxational thermodynamics of QW excitons. For GaAs QW's with $M_x=0.3m_0$ ($m_0$ is the free electron mass) and $v_s=3.7\times 10^5$ cm/s, one has $E_0=46.7$ $\mu$eV and $E_0/k_B=0.54$ K.

The relaxation kinetics of QW excitons coupled to thermal bulk LA phonons is given by the Boltzmann equation (see, e.g., Ref. 20)

$$\frac{\partial}{\partial t}N_{\varepsilon'} = -\frac{2}{\tau_{sc}}\int_0^\infty d\varepsilon\,\varepsilon^2 \int_{-1}^{1} du |F_z(a\varepsilon u)|^2 \Bigg\{ \frac{1}{\sqrt{4\varepsilon'(1-u^2)-(\varepsilon+1-\varepsilon u^2)^2}} [N_{\varepsilon'}(1+n_\varepsilon^{ph})(1+N_{\varepsilon'-\varepsilon})$$
$$-(1+N_{\varepsilon'})n_\varepsilon^{ph} N_{\varepsilon'-\varepsilon}]\Theta[2\sqrt{\varepsilon'(1-u^2)}-\varepsilon-1+\varepsilon u^2]\Theta(\varepsilon'-\varepsilon) + \frac{1}{\sqrt{4\varepsilon'(1-u^2)-(\varepsilon-1-\varepsilon u^2)^2}}$$
$$\times [N_{\varepsilon'} n_\varepsilon^{ph}(1+N_{\varepsilon'+\varepsilon})-(1+N_{\varepsilon'})(1+n_\varepsilon^{ph})N_{\varepsilon'+\varepsilon}]\Theta[2\sqrt{\varepsilon'(1-u^2)}+\varepsilon-1-\varepsilon u^2]\Theta[2\sqrt{\varepsilon'(1-u^2)}-\varepsilon$$
$$+1+\varepsilon u^2]\Bigg\}, \tag{2}$$

where the dimensionless energy is $\varepsilon = E/E_0 = E/(2M_x v_s^2)$; $n_\varepsilon^{ph} = 1/[\exp(\varepsilon E_0/k_B T_b) - 1]$ and $N_\varepsilon$ are the distribution functions of bulk LA phonons and QW excitons, respectively. $\Theta$ is the Heaviside step function. The integration variable $u$ is given by $u = \cos\theta$, where $0 \leqslant \theta \leqslant \pi$ is the angle between the $z$ axis and the wave vector of a scattering bulk LA-phonon. The scattering time is $\tau_{sc} = (\pi^2\hbar^4\rho)/(D^2 M_x^3 v_s)$, where $\rho$ is the crystal density and $D$ is the deformation potential. The form factor $F_z(\chi) = [\sin(\chi)/\chi][e^{i\chi}/(1-\chi^2/\pi^2)]$ refers to an infinite QW confinement potential.[21] This function describes the relaxation of the momentum conservation in the $z$ direction, and characterizes a spectral band of LA phonons, which effectively interact with a QW exciton. The dimensionless parameter $a \sim 1$ is defined by $a = (L_z M_x v_s)/\hbar$.

The kinetic equation (2) deals with an isotropic in-plane distribution of QW excitons, i.e., the occupation number $N_{\mathbf{p}_\parallel}$ of the in-plane mode $\mathbf{p}_\parallel$ relates to the distribution function $N_E$ by $N_{\mathbf{p}_\parallel} = N_{E_{\mathbf{p}_\parallel}} = N_E$. This approximation corresponds to the experimental conditions of Refs. 1–10. The first (second) term in the figure brackets on the right-hand side of Eq. (2) describes the phonon-assisted Stokes (anti-Stokes) scattering. In accordance with Eq. (2), the relaxation kinetics into the ground-state mode $\mathbf{p}_\parallel = 0$ is given by

$$\frac{\partial}{\partial t} N_{E=0} = \frac{2\pi}{\tau_{sc}} \int_1^\infty d\varepsilon\, \varepsilon \sqrt{\frac{\varepsilon}{\varepsilon - 1}} |F_z(a\sqrt{\varepsilon(\varepsilon-1)})|^2 \times [N_\varepsilon(1 + n_\varepsilon^{ph}) - N_{E=0}(n_\varepsilon^{ph} - N_\varepsilon)]. \tag{3}$$

The integral on the right-hand side of Eq. (3) characterizes the coupling of the ground-state mode $E = 0$ to continuum of the energy states $E \geqslant E_0$ (see Fig. 1).

In Sec. II, we develop the relaxational thermodynamics of QW excitons coupled to bulk LA phonons. A basic equation for the effective temperature $T$ of quasiequilibrium QW excitons is derived from LA-phonon-assisted kinetics given by Eq. (2). The conditions of the validity of the thermodynamic picture are analyzed and tested for excitons in GaAs QW's.

In Sec. III, the thermalization law $T = T(t)$ is studied for both classical ($T \gg T_0$) and well-developed BE statistics ($T \ll T_0$) of quasiequilibrium QW excitons. We demonstrate that the BE statistics strongly influences the thermalization process and leads to the density-dependent characteristic thermalization time $\tau_{th} = \tau_{th}(\rho_{2D})$ for $|T - T_b| \ll T_b$ and to the *nonexponential* density-dependent relaxation at $T - T_b \geqslant T_b$. In particular, for low bath temperatures $T_b \leqslant E_0/k_B$, we find a very slow thermalization $T(t) \propto 1/\ln t$ of quantum degenerate quasi-2D excitons. The numerical simulations of the relaxational dynamics are given for excitons in GaAs QW's.

In Sec. IV, we develop a theory of resonant PL of statistically degenerate QW excitons. An effective radiative lifetime $\tau_{opt}$ of a quasi-2D excitonic gas is calculated for BE-distributed QW excitons. We show that the law $\tau_{opt} \propto T$, which is valid for classical statistics of QW excitons, is violated at low temperatures and at $T \to 0$ the effective decay time $\tau_{opt}$ tends to $2\tau_R$, where $\tau_R$ is the intrinsic radiative lifetime of a QW exciton. The PL kinetics of QW excitons is described in the thermodynamic approach within three coupled equations for $T(t)$, $\rho_{2D}(t)$, and $\tau_{opt}(T, \rho_{2D})$. We give a numerical modeling of the $T$- and $\rho_{2D}$-dependent PL kinetics of quantum-degenerate quasi-2D excitons.

In Sec. V, we discuss the influence of interface polaritons and QW biexcitons on the relaxation kinetics of statistically degenerate excitons in perfect QW's. Our theory is always appropriate for thermalization of indirect excitons in CQW's, where even at small densities $\rho_{2D}$ the interface polariton effect is rather weak and biexciton states are unbound. For single QW's we argue that QW polaritons and biexcitons are considerably weakened at large densities, due to particle-particle scattering. In this case our model is applicable to QW's with direct excitons.

In Appendix A, in order to estimate the critical density $\rho_{2D}^c$ of QW excitons for the development of relaxational thermodynamics we calculate the characteristic equilibration time $\tau_{x\text{-}x}$ due to exciton-exciton scattering. The equilibration rate $1/\tau_{x\text{-}x}$ is found in both limits of classical and well-developed BE statistics. In Appendix B, we analyze how a relatively large homogeneous linewidth $\sim \hbar/\tau_{x\text{-}x} \gg \hbar/\tau_{th}$, $\hbar/\tau_R$ of high-density QW excitons in a perfect QW influences the PL process.

## II. RELAXATIONAL THERMODYNAMICS OF QW EXCITONS

In this section we summarize the thermodynamic relations for an ideal two-dimensional gas of bosons, and derive the basic equation for relaxational dynamics of QW excitons.

### A. Thermodynamic relations for quasi-2D excitons

The thermodynamic equation $\mu = \mu(T, \rho_{2D})$ for a quasi-equilibrium gas of QW excitons can be derived from the condition

$$\rho_{2D} = \frac{1}{S}\sum_{\mathbf{p}_\parallel} N_{\mathbf{p}_\parallel}^{eq} = \frac{2gM_x k_B T}{\pi\hbar^2} \int_0^\infty \frac{dz}{e^{-\mu/k_B T} e^z - 1}. \tag{4}$$

From Eq. (4), one obtains

$$\mu = k_B T \ln(1 - e^{-T_0/T}), \tag{5}$$

where the degeneracy temperature $T_0$ is given by Eq. (1) with $g = 4$. With the chemical potential $\mu$ of Eq. (5), the equilibrium distribution function of QW excitons is

$$N_E^{eq} = \frac{1 - e^{-T_0/T}}{e^{E/k_B T} + e^{-T_0/T} - 1}. \tag{6}$$

In particular, the occupation number of the ground-state mode is given by $N_{\mathbf{p}_\parallel = 0}^{eq} = N_{E=0}^{eq} = \exp(T_0/T) - 1$.

Classical, Maxwell-Boltzmann, statistics of QW excitons is realized for $T \gg T_0$. In this case Eqs. (5) and (6) reduce to

$$\mu = k_B T \ln(T_0/T) \quad \text{and} \quad N_{E=0}^{eq} = T_0/T \ll 1, \tag{7}$$

i.e., the occupation numbers of QW modes $\mathbf{p}_\parallel$ are much less than unity. In the opposite limit $T < T_0$ of well-developed BE statistics one has

$$\mu = -k_B T e^{-T_0/T} \quad \text{and} \quad N_{E=0}^{eq} = e^{T_0/T} \gg 1. \tag{8}$$

According to Eq. (8), the chemical potential of a degenerate gas of QW excitons approaches zero much faster than the temperature $T$. While the chemical potential $\mu<0$ for $T>0$ and the BE condensation of the QW excitons is absent within the thermodynamic approach for temperatures above zero, the occupation number of the ground-state mode $\mathbf{p}_\parallel=0$ increases exponentially with decreasing $T\ll T_0$.

Equations (5)–(8) can be applied to an arbitrary two-dimensional quasi-ideal gas of Bose particles with quadratic dispersion. The specific characteristics of the bosons, like the spin degeneracy factor $g$ and the translational mass $M_x$, enter the thermodynamic relationships only through the degeneracy temperature $T_0$ defined by Eq. (1).

For GaAs QW's one estimates $T_0=(\pi\hbar^2\rho_{2D})/(2M_xk_B)=4.6$ K ($k_BT_0=399.5$ $\mu$eV) for $\rho_{2D}=10^{11}$ cm$^{-2}$. This density of quasi-2D excitons corresponds to the mean interparticle distance $\sim 1/\sqrt{\rho_{2D}}\simeq 320$ Å and to the Mott parameter $\rho_{2D}[a_x^{(2D)}]^2\simeq 0.04$, i.e., the QW excitons are still well-defined quasiparticles. Here $a_x^{(2D)}$ is the Bohr radius of a quasi-2D exciton. In Sec. II B we will use Eqs. (5)–(8) to develop the relaxational dynamics.

### B. Thermalization equation for quasiequilibrium excitons in QW's

The thermalization of QW excitons occurs through a sequence of quasiequilibrium thermodynamic states, which are characterized by an effective temperature $T=T(t)$ and chemical potential $\mu=\mu(t)$, provided that the exciton-exciton interaction in a QW is much stronger than the coupling of QW excitons with bulk LA phonons. The initial interaction is conservative, and equilibrates the system of QW excitons without change of total energy. The characteristic equilibration time $\tau_{x\text{-}x}$ depends on the density $\rho_{2D}$ of QW excitons. For excitons distributed below the threshold for LO-phonon emission, energy (effective temperature) relaxation results from QW exciton–bulk LA-phonon scattering, and is characterized by an effective thermalization time $\tau_{th}$. The hierarchy of interactions means that in a large and interesting range of density we have $\tau_{x\text{-}x}\ll\tau_{th}$. In this case the equilibration and thermalization kinetics can be naturally separated (see, e.g., Ref. 22), and the thermodynamic approach is correct.

The condition $\tau_{x\text{-}x}\ll\tau_{th}$ will hold for $\rho_{2D}>\rho_{2D}^c$, where $\rho_{2D}^c$ is some critical density for QW excitons. For a classical distribution of quasi-2D excitons one has $T_0/T\ll 1$ and $N_E^{eq}=(T_0/T)\exp(-E/k_BT)\ll 1$, according to Eq. (6). In this limit the characteristic equilibration time $\tau_{x-x}$ is estimated by Eq. (A4) of Appendix A, while the thermalization time $\tau_{th}$ is given by Eq. (15) of Sec. III. The comparison of Eqs. (A4) and (15) yields $\tau_{x\text{-}x}\ll\tau_{th}$, provided that

$$k_BT_0>k_BT_0^c=\frac{4\hbar}{\pi}\left(\frac{\mu_x}{M_x}\right)^2\frac{C_{2D}}{\tau_{sc}}, \tag{9}$$

where $\mu_x$ is the reduced mass of a QW exciton and the constant $C_{2D}=C_{2D}(a)\gg 1$ is defined by Eq. (15). For a classical gas of excitons in GaAs QW of thickness $L_z=100$ Å we estimate $\rho_{2D}^c=1.2\times 10^9$ cm$^{-2}$ and the corresponding temperature scale $T_0^c=56$ mK. For comparison, the critical density of about $4\times 10^9$ cm$^{-2}$ has been estimated in experiments[4] with high-quality GaAs QW's of $L_z=80$ Å. For well-developed BE statistics of QW excitons one has $T_0/T\gg 1$, and the occupation numbers of the energy states $E\gg k_BT\exp(-T_0/T)$ are given by the Planck function, i.e., $N_{E>0}^{eq}=1/[\exp(E/k_BT)-1]$, according to Eq. (6). In this case $\tau_{x\text{-}x}$ and $\tau_{th}$ are given by Eq. (A5) of Appendix A and Eq. (18) of Sec. III, respectively. The condition $\tau_{x\text{-}x}\ll\tau_{th}$ reduces to

$$E_0\left(\frac{T_0}{T}\right)e^{T_0/T}>\frac{4\hbar}{(1-\pi/4)}\left(\frac{\mu_x}{M_x}\right)^2\frac{\tilde{C}_{2D}}{\tau_{sc}}, \tag{10}$$

where the constant $\tilde{C}_{2D}=\tilde{C}_{2D}(a)\gg 1$ is given by Eq. (18). Inequality (10) always holds for strongly degenerate QW excitons.

Criteria (9) and (10) of the validity of the thermodynamic picture at $T\gg T_0$ and $T\ll T_0$, respectively, are independent of the scattering length $\sim a_x^{(2D)}$ of exciton-exciton interaction. This is due to the quasi-two-dimensionality of QW excitons (for details, see Appendix A). In contrast, in a three-dimensional gas the equilibration time $\tau_{x\text{-}x}$ due to particle-particle interaction depends explicitly on the scattering length.[23] In further analysis we assume the hierarchy of interactions, i.e., that $\rho_{2D}>\rho_{2D}^c$, so that inequality (9) is valid. The initial density of photogenerated excitons in GaAs QW's is usually larger than $10^9$ cm$^{-2}$ (see Refs. [3–10]), indicating that the thermodynamic picture of relaxation is adequate for typical experimental conditions. We will omit the superscript in $N_E^{eq}$, because within our approach $N_E=N_E^{eq}$.

The thermalization dynamics of QW excitons is determined by the slowest elementary LA-phonon-assisted relaxation process of the kinetic equations (2) and (3). The joint density of states for Stokes and anti-Stokes scattering from the energy mode $E$ continuously decreases with decreasing $E$, according to the right-hand side of Eq. (2) (note, that the density of quasi-2D excitonic states is constant given by $16\pi^2M_x/\hbar^2$ for $g=4$). Moreover, the low-energy states $E\leqslant E_0/4$ are not active in Stokes scattering, and couple with the corresponding LA-phonon-separated states $E\geqslant E_0$ only through the anti-Stokes process. The above arguments show a particular status of Eq. (3), which describes the relaxation kinetics into the ground-state mode. Furthermore, the ground-state mode $\mathbf{p}_\parallel=0$ refers to the lowest energy $E=0$ and to the largest occupation number $N_{E=0}$. For well-developed BE statistics of QW excitons one has $N_{E=0}\gg 1$. As a result, the population of the state $E=0$ generally requires an additional time in comparison with that for the high-energy states $E\gg k_BT$ with $N_E\ll 1$. Thus LA-phonon-assisted occupation of the ground-state mode is the slowest, ''bottleneck,'' relaxation process, which determines the thermalization kinetics of quasiequilibrium QW excitons. A similar picture holds for thermalization of excitons in bulk semiconductors.[24,25]

With the substitution of $N_E=N_E^{eq}$ given by Eq. (6), in Eq. (3), we derive

$$\frac{\partial}{\partial t}T=-\frac{2\pi}{\tau_{sc}}\left(\frac{T^2}{T_0}\right)(1-e^{-T_0/T})\int_1^\infty d\varepsilon\,\varepsilon\sqrt{\frac{\varepsilon}{\varepsilon-1}}|F_z[a\sqrt{\varepsilon(\varepsilon-1)}]|^2\frac{e^{\varepsilon E_0/k_BT_b}-e^{\varepsilon E_0/k_BT}}{(e^{\varepsilon E_0/k_BT}+e^{-T_0/T}-1)}\frac{1}{(e^{\varepsilon E_0/k_BT_b}-1)}. \quad (11)$$

Equation (11) describes the thermalization dynamics $T=T(t)$ of QW excitons from the effective temperature $T_i=T(t=0)$ to the bath temperature $T_b$. A finite lifetime $\tau'$ of excitons, due to radiative and nonradiative recombination, can be incorporated into Eq. (11) by the degeneracy temperature $T_0(t)\propto\rho_{2D}(t=0)\exp(-t/\tau')$. In Sec. III we apply Eq. (11) to the thermalization kinetics of QW excitons in the absence of their recombination ($\tau'\to\infty$). This analysis refers to the case when the characteristic thermalization time $\tau_{th}\ll\tau'$. In Sec. IV we will use Eq. (11) in order to develop a theory of resonant photoluminescence of quantum degenerate excitons in perfect QW's, when $\tau'=\tau_{opt}$ is determined by the intrinsic radiative lifetime $\tau_R$ of QW excitons.

## III. THERMALIZATION KINETICS OF QUASIEQUILIBRIUM QW EXCITONS

In this section the thermalization dynamics of QW excitons is analyzed within Eq. (11) for $\rho_{2D}=$const. First we linearize Eq. (11) about some bath temperature $T_b$ to study the exponential relaxation of QW excitons with the effective temperature $|T-T_b|\ll T_b$. Linearization is not appropriate for the thermalization kinetics at $T_b=0$. Here we show that the thermalization becomes very slow, with $T(t)\propto 1/\ln t$. In Sec. III C, the cooling of hot QW excitons with $T-T_b\geqslant T_b$ is treated for both classical and strongly degenerate limits of BE statistics. There are four characteristic temperature scales $T$, $T_0$, $T_b$, and $E_0/k_B$, and the precise form of the relaxation will depend on all four. Note, however, that if $T\gg T_0$, the degeneracy temperature is not a relevant parameter, and the most relevant aspects of phonon bottleneck effects are captured by the ratio $E_0/k_BT_b$. In practice the relaxational dynamics is controlled by two parameters $T_0/T$ and $E_0/k_BT_b$.

### A. Relaxation kinetics between nearby thermodynamic states

If the effective temperature $T$ of QW excitons is close to the bath $T_b$, the basic thermodynamic (11) reduces to

$$\frac{\partial}{\partial t}T=-\frac{1}{\tau_{th}}(T-T_b), \quad (12)$$

where the effective thermalization time $\tau_{th}$ is given by

$$\frac{1}{\tau_{th}}=\left(\frac{2\pi}{\tau_{sc}}\right)\left(\frac{E_0}{k_BT_0}\right)(1-e^{-T_0/T_b})\int_1^\infty d\varepsilon\,\varepsilon^2\sqrt{\frac{\varepsilon}{\varepsilon-1}}|F_z(a\sqrt{\varepsilon(\varepsilon-1)})|^2\frac{1}{(e^{\varepsilon E_0/k_BT_b}+e^{-T_0/T_b}-1)}\frac{e^{\varepsilon E_0/k_BT_b}}{(e^{\varepsilon E_0/k_BT_b}-1)}. \quad (13)$$

The linear approximation of Eq. (11) by Eq. (12) is valid for $|T-T_b|/T_b<k_BT_b/E_0$, and can be done for any $T_b>0$. Equation (12) corresponds to the *exponential* thermalization law $T(t)=T_b+\Delta T\exp(-t/\tau_{th})$, where $\Delta T=T_i-T_b$ and $T_i=T(t=0)$ is the initial temperature of QW excitons. The thermalization time $\tau_{th}$ uniquely describes all quasiequilibrium relaxation processes in a gas of quasi-2D excitons. For example, the relaxation kinetics into the ground-state mode $E=0$ is given by $N_{E=0}(t)=N^f_{E=0}+(N^i_{E=0}-N^f_{E=0})\exp(-t/\tau_{th})$, where $N^i_{E=0}=\exp(T_0/T_i)-1$ and $N^f_{E=0}=\exp(T_0/T_b)-1$. We will analyze Eq. (13) in the limit of classical and well-developed BE statistics of QW excitons, respectively.

1. *Classical gas of QW excitons* ($T_b\gg T_0$)

In this case Eq. (13) yields

$$\frac{1}{\tau_{th}}=\left(\frac{2\pi}{\tau_{sc}}\right)\left(\frac{E_0}{k_BT_b}\right)\int_1^\infty d\varepsilon\,\varepsilon^2\sqrt{\frac{\varepsilon}{\varepsilon-1}}\frac{|F_z[a\sqrt{\varepsilon(\varepsilon-1)}]|^2}{(e^{\varepsilon E_0/k_BT_b}-1)}. \quad (14)$$

From Eq. (14) one concludes, as expected, that the characteristic thermalization time of the Maxwell-Boltzmann distributed QW excitons is indeed independent of their concentration $\rho_{2D}$. In Fig. 2 we plot $\tau_{th}=\tau_{th}(T_b)$ calculated by Eq. (14) for GaAs QW of thickness $L_z=100$ Å. The ratio $E_0/k_BT_b$ determines the high- and low-temperature limits of $\tau_{th}$.

A. *High-temperature limit* ($k_BT_b\gg E_0$). The characteristic thermalization time, which in this case we will designate by $\tau_H$, is given by

$$\frac{1}{\tau_H}=\frac{C_{2D}}{\tau_{sc}}, \quad (15)$$

where

$$C_{2D}=2\pi\int_1^\infty d\varepsilon\,\varepsilon\sqrt{\frac{\varepsilon}{\varepsilon-1}}|F_z(a\sqrt{\varepsilon(\varepsilon-1)})|^2.$$

In the high-temperature limit of a classical gas of QW excitons, the thermalization time $\tau_H$ is independent of the bath temperature $T_b$. The constant $C_{2D}=C_{2D}(a)$, which completely determines $\tau_H$ in terms of $\tau_{sc}$, is much larger than unity. For example, for a GaAs QW with $L_z=100$ Å, the dimensionless parameter $a=0.096$ and the constant $C_{2D}=2530$. The corresponding thermalization time of the QW excitons is $\tau_H=5.5$ ps. For comparison, the high-temperature thermalization time of a classical gas of excitons

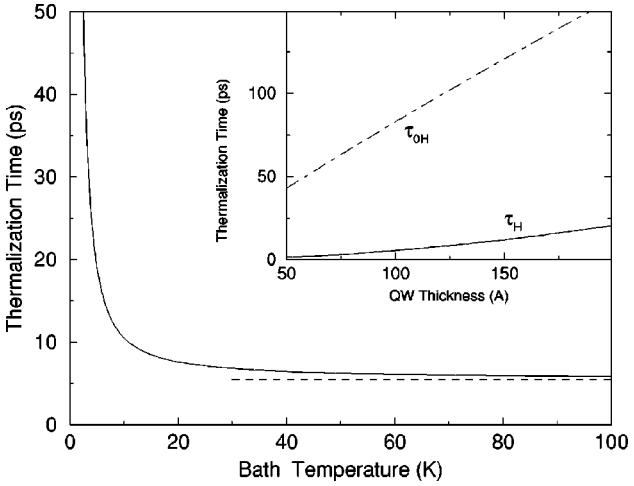


FIG. 2. The thermalization time $\tau_{th}=\tau_{th}(T_b)$ of a classical gas of QW excitons, a GaAs QW with $L_z=100$ Å and $\tau_{sc}=13.9$ ns [see Eq. (14)]. Inset: the high-temperature thermalization time $\tau_H=\tau_H(L_z)\propto L_z^2$ of Maxwell-Boltzmann distributed QW excitons (solid line) and the high-temperature thermalization time $\tau_{0H}=\tau_{0H}(L_z)\propto L_z$ of strongly statistically degenerate QW excitons (dot-dashed line). The latter dependence refers to $T_0=(k_BT_b^2)/E_0=1$ $(T_0\gg T_b)$.

in a bulk semiconductor is given by $\tau_H^{(3D)}=(3/8\pi)\tau_{sc}$.[25] This estimate yields $\tau_H^{(3D)}=1.66$ ns for bulk GaAs, i.e., $\tau_H^{(3D)}\gg\tau_H^{(2D)}=\tau_H$. The ratio $\tau_H^{(2D)}/\tau_H^{(3D)}=(8\pi)/(3C_{2D})\simeq 3.3\times 10^{-3}$, which refers to bulk GaAs and GaAs QW with $L_z=100$ Å, clearly demonstrates the effective cooling of QW excitons in the presence of a bath of bulk phonons. Due to the relaxation of momentum conservation in the $z$ direction, a ground-state QW exciton couples to the continuum states $E\geqslant E_0$ rather than to the single-energy state $E=E_0$ as occurs in bulk materials. With decreasing bath temperature $T_b$, the effective thermalization time $\tau_{th}$ of QW excitons monotonously increases, starting from its high-temperature limit given by $\tau_H$ (see Fig. 2). For $T_b=5$ K one finds from Eq. (15) that $\tau_{th}=19.1$ ps in GaAs QW with $L_z=100$ Å.

There is an uncertainty in values of the deformation potential $D$ of exciton–LA-phonon interaction, published in literature (see, e.g., Refs. 2, 15, and 21), from $D_{min}=7.0$ eV to $D_{max}=18.1$ eV. In our numerical evaluations we use $D=15.5$ eV. According to Eq. (13), the deformation potential contributes to $\tau_{th}$ only through $\tau_{sc}\propto 1/D^2$. Therefore the numerical calculations of $\tau_{ph}$ can be straightforwardly re-scaled to another value of $D$ [we will use this procedure in calculations of Fig. 9(a) to reproduce qualitatively the experimental data of Ref. 4]. The value $D=15.5$ eV corresponds to $\tau_{sc}=13.9$ ns.

B. *Low-temperature limit* $(k_BT_b\leqslant E_0)$. In this case Eq. (14) reduces to

$$\frac{1}{\tau_L}=\frac{2\pi^{3/2}}{\tau_{sc}}\left(\frac{E_0}{k_BT_b}\right)^{1/2}e^{-E_0/k_BT_b}, \tag{16}$$

where we designate the low-temperature limit of $\tau_{th}$ by $\tau_L$. According to Eq. (16) the thermalization time $\tau_L$ increases exponentially with decreasing bath temperature $T_b$ below $E_0/k_B$. The origin of this result can be understood from the initial Eq. (3) of the phonon-assisted relaxation kinetics into the ground-state mode $E=0$. For a classical gas of QW excitons one has $N_E\ll 1$. As a result, both the spontaneous (independent of $N_{E=0}$) and stimulated (proportional to $N_{E=0}$) processes contribute to the population of the ground-state mode. The first, Stokes, process is $\propto N_{E\geqslant E_0}(1+n^{ph}_{E\geqslant E_0})$, and increases $N_{E=0}$, while the second, anti-Stokes, process is $\propto -N_{E=0}n^{ph}_{E\geqslant E_0}$, and decreases the occupation number of the mode $E=0$. For the high-temperature limit of a classical gas of QW excitons both opposite fluxes are intense because $n^{ph}_{E\geqslant E_0}=k_BT_b/E\gg 1$. In the low-temperature limit $n^{ph}_{E\geqslant E_0}=\exp(-E/k_BT_b)\ll 1$, and both the spontaneous process, which is proportional to $N_{E\geqslant E_0}(1+n^{ph}_{E\geqslant E_0})\simeq N_{E\geqslant E_0}=(T_0/T)\exp(-E/k_BT)$, and the stimulated process, which is proportional to $-N_{E=0}n^{ph}_{E\geqslant E_0}=-(T_0/T)\exp(-E/k_BT_b)$, are exponentially weak.

The plot $\tau_{th}=\tau_{th}(T_b)$ (see Fig. 2) shows that for excitons in GaAs QW's the high-temperature limit given by Eq. (15) means $T_b\geqslant 30$–$50$ K. On the other hand, a strong increase of $\tau_{th}$ occurs already at $T_b\simeq 5$–$10$ K, i.e., at temperatures much above the low-temperature range determined by $T_b\leqslant E_0/k_B\simeq 0.54$ K, and where the approximation of $\tau_{th}$ by Eq. (16) is valid.

2. *Quantum gas of QW excitons* $(T_0\gg T_b)$

In this case Eq. (13) reduces to

$$\frac{1}{\tau_{th}}=\left(\frac{2\pi}{\tau_{sc}}\right)\left(\frac{E_0}{k_BT_0}\right)\int_1^\infty d\varepsilon\,\varepsilon^2 \times\sqrt{\frac{\varepsilon}{\varepsilon-1}}|F_z[a\sqrt{\varepsilon(\varepsilon-1)}]|^2\frac{e^{\varepsilon E_0/k_BT_b}}{(e^{\varepsilon E_0/k_BT_b}-1)^2}. \tag{17}$$

Equation (17) shows that the thermalization time of quantum degenerate quasi-2D excitons depends on the concentration $\rho_{2D}$, i.e., $\tau_{th}\propto\rho_{2D}$.

A. *High-temperature limit* $(k_BT_b\gg E_0)$. In this limit Eq. (17) yields$(\tau_{th}=\tau_{0H})$:

$$\frac{1}{\tau_{0H}}=\left(\frac{k_BT_b^2}{T_0E_0}\right)\frac{\tilde{C}_{2D}}{\tau_{sc}}, \tag{18}$$

where

$$\tilde{C}_{2D}=2\pi\int_1^\infty d\varepsilon\sqrt{\frac{\varepsilon}{\varepsilon-1}}|F_z(a\sqrt{\varepsilon(\varepsilon-1)})|^2.$$

From Eqs. (15) and (18) we find

$$\frac{1}{\tau_{0H}}=\left(\frac{k_BT_b^2}{T_0E_0}\right)\left(\frac{\tilde{C}_{2D}}{C_{2D}}\right)\frac{1}{\tau_H}. \tag{19}$$

The constant $\tilde{C}_{2D}=\tilde{C}_{2D}(a)$ is much larger than unity, but much less than $C_{2D}$. For example, $\tilde{C}_{2D}=170$ so that $\tilde{C}_{2D}/C_{2D}=0.07$ for a GaAs QW with $L_z=100$ Å. According to Eq. (19), one has $\tau_{0H}<\tau_H$ for $T_0\gg T_b>[(T_0E_0C_{2D})/(k_B\tilde{C}_{2D})]^{1/2}$ and $\tau_{0H}>\tau_H$ for $T_b<[(T_0E_0C_{2D})/(k_B\tilde{C}_{2D})]^{1/2}$. The acceleration or slowing down of the relaxation kinetics in comparison with that in a classical gas of QW excitons originates from BE statistics.

Because for the quasiequilibrium degenerate QW excitons $N_{E=0}=\exp(T_0/T)\gg N_{E\geqslant E_0}=1/[\exp(E_0/k_BT)-1]$, only the stimulated processes $\propto N_{E=0}(N_{E\geqslant E_0}-n^{ph}_{E\geqslant E_0})$ contribute to occupation kinetics of the ground-state mode $E=0$. From Eqs. (7) and (8), one obtains $\delta T/T=-\delta N_{E=0}/N_{E=0}$ for classical statistics and $\delta T/T=-(T/T_0)(\delta N_{E=0}/N_{E=0})$ for well-developed BE statistics of the ground-state mode, i.e., the same relative change of the occupation number $N_{E=0}$ is accompanied at $T\ll T_0$ by the much less relative change of the effective temperature than that at $T\gg T_0$. Therefore, BE occupation of the ground-state mode with $N_{E=0}\gg 1$ slows down the thermalization kinetics of QW excitons. On the other hand, one has $N_{E\geqslant E_0}-n^{ph}_{E\geqslant E_0}=n^{ph}_{E\geqslant E_0}(\delta T/T_b)$ for well-developed BE statistics of the energy modes $E\geqslant E_0$ and $N_{E\geqslant E_0}-n^{ph}_{E\geqslant E_0}=n^{ph}_{E\geqslant E_0}(E/k_BT_b)(\delta T/T_b)$ for a classical distribution of QW excitons at $E\geqslant E_0$. As a result, the BE occupation numbers $N_{E\geqslant E_0}\gg 1$ enhance the relaxation dynamics by the factor $\sim k_BT_b/E_0$, which is much larger than unity in the high-temperature limit. With decreasing bath temperature $T_b$ the slowing down of thermalization, which results from $N_{E=0}\gg 1$, starts to dominate over the acceleration of relaxation due to BE statistics of the modes $E\geqslant E_0$.

The thermalization time $\tau_{th}=\tau_{th}(a)$ is very sensitive to the dimensionless parameter $a\propto L_z$ through the form-factor function $F_z$ on the right-hand side of Eq. (13). With decreasing $L_z$ the spectral width of $F_z[a\sqrt{\varepsilon(\varepsilon-1)}]$ increases, indicating a stronger relaxation of the momentum conservation in the $z$-direction. From Eqs. (15) and (18) we conclude that for the infinite square QW confinement potential $\tau_H\propto L_z^2$ and $\tau_{0H}\propto L_z$, respectively. The dependences $\tau_H=\tau_H(L_z)$ and $\tau_{0H}=\tau_{0H}(L_z)$ are plotted in the inset of Fig. 2.

B. *Low-temperature limit* ($k_BT_b<E_0$). In this case, from Eq. (17) we obtain

$$\frac{1}{\tau_{0L}}=\frac{2\pi^{3/2}}{\tau_{sc}}\left(\frac{E_0T_b}{k_BT_0^2}\right)^{1/2}e^{-E_0/k_BT_b}=\left(\frac{T_b}{T_0}\right)\frac{1}{\tau_L},\qquad(20)$$

where we designate the low-temperature thermalization time of degenerate QW excitons by $\tau_{0L}$, and the corresponding thermalization time of Maxwell-Boltzmann distributed QW excitons $\tau_L$ is given by Eq. (16). The slowing down of the thermalization process by the factor $T_0/T_b\propto\rho_{2D}$, which is much larger than unity, stems from well-developed BE statistics of the ground-state mode, i.e., from $N_{E=0}\gg 1$. The distribution function of QW excitons at $E\geqslant E_0$ is classical, i.e., $N_{E\geqslant E_0}=\exp(-E_0/k_BT)<1$, and does not enhance the relaxation processes into the ground-state mode $E=0$.

The influence of BE statistics on the thermalization of QW excitons is demonstrated in Fig. 3 for $\tau_{th}=\tau_{th}(T_b)$, $\rho_{2D}=$const, and in Fig. 4 for $\tau_{th}=\tau_{th}(\rho_{2D})$, $T_b=$const, respectively. The numerical evaluations of $\tau_{th}$ refer to Eq. (13).

### B. Thermalization of QW excitons at $T_b=0$

The linearization of the basic Eq. (11) at $T_b>0$, developed in Sec. III A, does not hold when $|T-T_b|>k_BT_b^2/E_0$, and especially at zero bath temperature. Because $N^{eq}_{E=0}\to\infty$ at $T_b=0$, the effective temperature $T$ of QW excitons is a

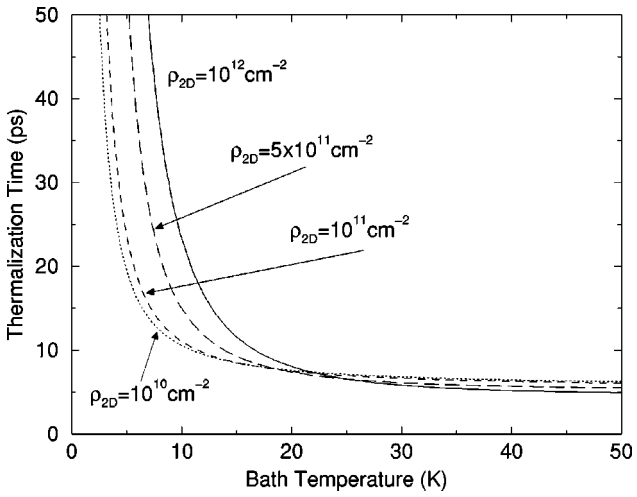


FIG. 3. The thermalization time $\tau_{th}=\tau_{th}(T_b)$ of BE-distributed QW excitons of the densities $10^{12}$ cm$^{-2}$ (solid line), $5\times10^{11}$ cm$^{-2}$ (long dashed line), $10^{11}$ cm$^{-2}$ (dashed line), and $10^{10}$ cm$^{-2}$ (dotted line). The calculations with Eq. (13) refer to GaAs QW with $L_z=100$ Å and $\tau_{sc}=13.9$ ns.

nonanalytic point of the right-hand side of Eq. (11) at $T=T_b=0$. For $T_b=0$, Eq. (11) reduces to

$$\frac{\partial}{\partial t}T=-\frac{2\pi}{\tau_{sc}}\left(\frac{T^2}{T_0}\right)(1-e^{-T_0/T})\int_1^\infty d\varepsilon\,\varepsilon\times\sqrt{\frac{\varepsilon}{\varepsilon-1}}\frac{|F_z(a\sqrt{\varepsilon(\varepsilon-1)})|^2}{(e^{\varepsilon E_0/k_BT}+e^{-T_0/T}-1)}.\qquad(21)$$

Equation (21), which describes how the QW excitons with effective temperature $T$ cool down toward $T_b=0$, can be further simplified for $k_BT\leqslant E_0$:

$$\frac{\partial}{\partial t}T=-\frac{2\pi^{3/2}}{\tau_{sc}}\left(\frac{T^2}{T_0}\right)\left(\frac{k_BT}{E_0}\right)^{1/2}e^{-E_0/k_BT}.\qquad(22)$$

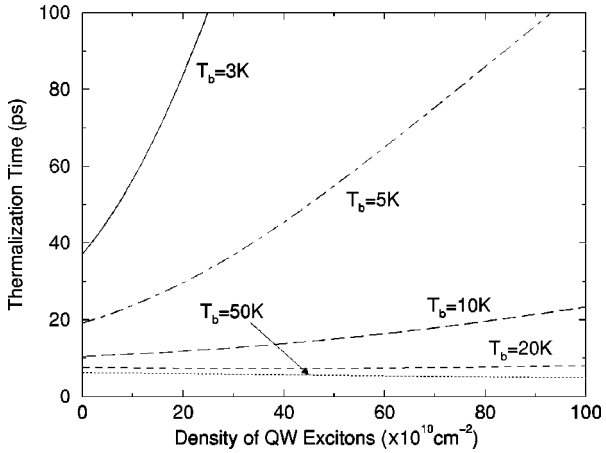


FIG. 4. The thermalization time $\tau_{th}=\tau_{th}(\rho_{2D})$ of BE-distributed QW excitons at bath temperatures of 3 K (solid line), 5 K (dot-dashed line), 10 K (long dashed line), 20 K (dashed line), and 50 K (dotted line). The calculations with Eq. (13) deal with a GaAs QW with $L_z=100$ Å and $\tau_{sc}=13.9$ ns. Note that numerical evaluations of Figs. 2–4 refer to effective temperatures of QW excitons close to the bath temperature, i.e., $|T-T_b|\ll T_b$.

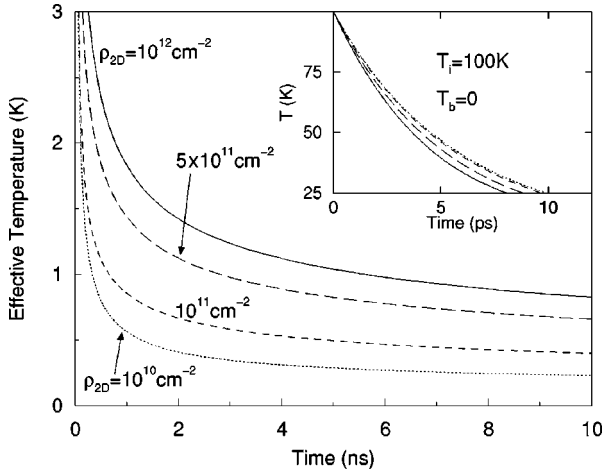


FIG. 5. Thermalization dynamics $T=T(t)$ of QW excitons at $T_b=0$ and $\rho_{2D}=10^{12}$ cm$^{-2}$ (solid line), $5\times10^{11}$ cm$^{-2}$ (long dashed line), $10^{11}$ cm$^{-2}$ (dashed line), and $10^{10}$ cm$^{-2}$ (dotted line). Inset: the initial transient thermalization of hot QW excitons BE distributed at $t=0$ with $T_i=100$ K.

In dimensionless time and temperature units, $\tau=(2\pi^{3/2}E_0t)/(k_BT_0\tau_{sc})$ and $\tilde{T}=k_BT/E_0$, and Eq. (22) takes the canonical form $d\tilde{T}/d\tau=-\tilde{T}^{5/2}\exp(-1/\tilde{T})$. The solution $T=T(t)$ of Eq. (22) is given by the transcendental equation

$$F(k_BT/E_0)=\frac{2\pi^{3/2}}{\tau_{sc}}\left(\frac{E_0}{k_BT_0}\right)t+A_i, \tag{23}$$

where $F(x)=[1/\sqrt{x}-Ds(1/\sqrt{x})]e^{1/x}$, $Ds(y)=e^{-y^2}\int_0^y dt\, e^{t^2}$ is Dawson's integral, and the integration constant $A_i$ is defined by the initial condition $T_i=T(t=0)\leqslant E_0/k_B$, i.e., $A_i=F(k_BT_i/E_0)$. For $t\gg[(k_BT_0)/(2\pi^{3/2}E_0)]\tau_{sc}$ the asymptotic solution of Eq. (22) is

$$k_BT(t)=\frac{E_0}{\ln[(2\pi^{3/2}E_0t)/(k_BT_0\tau_{sc})]}, \tag{24}$$

i.e., $\tilde{T}(\tau)=1/\ln(\tau)$ for $\tau\gg1$.

The asymptotic law [Eq. (24)] characterizes the *nonexponential* and extremely slow thermalization kinetics of a quantum gas of QW excitons. Because the phonon occupation numbers $n_{\mathbf{q}}^{ph}=0$ at $T_b=0$, only Stokes scattering of the QW excitons determines the relaxation process. In this case the integrand on the right-hand side of Eq. (3) is proportional to $N_{E\geqslant E_0}(N_{E=0}+1)\simeq N_{E\geqslant E_0}N_{E=0}$. At $k_BT\leqslant E_0$ the energy states $E\geqslant E_0$, which couple with the ground-state mode $E=0$ [see Eq. (3)], are weakly populated by the QW excitons, while $N_{E=0}\gg1$. The Maxwell-Boltzmann distribution function $N_{E\geqslant E_0}=\exp(-E/k_BT)$ gives rise to the factor $\exp(-E_0/k_BT)\ll1$ on the right-hand side of Eq. (22). This term, together with the need to accumulate a huge number of QW excitons in the ground-state mode $\mathbf{p}_\parallel=0$, due to BE statistics, is responsible for $1/\ln(\tau)$ thermalization law at $T_b=0$.

In Fig. 5 we plot the results of numerical evaluation of the relaxational dynamics at $T_b=0$ from Eq. (21) for various concentrations $\rho_{2D}$ of QW excitons and for an initial effective temperature $T_i=100$ K. The simulations refer to the long-lived quasi-2D excitons in GaAs/Al$_x$Ga$_{1-x}$As CQW's. The first hot relaxation (see the inset of Fig. 5) is completed within $\Delta t\leqslant30$–$100$ ps. At the end of the transient stage the effective temperature $T$ of QW excitons is still much higher than $E_0/k_B\simeq0.54$ K, so that $N_{E\simeq E_0}>1$ for $T\leqslant T_0$. As a result, the duration of hot thermalization decreases with increasing concentration $\rho_{2D}$ of QW excitons. Here one has an acceleration of the relaxation kinetics due to BE statistics of energy modes $E\geqslant E_0$. The further thermalization kinetics of cold QW excitons refers to $t\geqslant100$ ps and reveals $1/\ln(\tau)$ law (see Fig. 5). According to Eq. (24), a critical slowing down of the phonon-assisted relaxation dynamics develops with increasing degeneracy temperature $T_0\propto\rho_{2D}$. An extension of the numerical evaluations presented in Fig. 5 on a $\mu$s time scale shows that even at $\Delta t=1$ $\mu$s the effective temperature of QW excitons is still a few hundred mK for $\rho_{2D}\geqslant10^{11}$ cm$^{-2}$, i.e., $T(t=1\ \mu\text{s})=89$ mK for $\rho_{2D}=10^{10}$ cm$^{-2}$, $T(t=1\ \mu\text{s})=129$ mK for $\rho_{2D}=10^{11}$ cm$^{-2}$, $T(t=1\ \mu\text{s})=181$ mK for $\rho_{2D}=5\times10^{11}$ cm$^{-2}$, and $T(t=1\ \mu\text{s})=213$ mK for $\rho_{2D}=10^{12}$ cm$^{-2}$. On a time scale much longer than the duration of the initial hot thermalization the effective temperature $T$ is nearly independent of $T_i=T(t=0)$, provided that $k_BT_i\gg E_0$. Recently, a strong increase of the thermalization times in a highly degenerate quantum gas of quasi-2D excitons has indeed been observed in GaAs/Al$_x$Ga$_{1-x}$As CQW's.[26]

However, our treatment has neglected possible low-temperature collective states due to interactions between QW excitons. At a critical temperature $T_c=\alpha T_0$, where $\alpha=\alpha(a_x^{(2D)}\rho_{2D}^{1/2})<1$, a system of quasi-2D excitons may undergo a phase transition to a superfluid state. Our calculations of relaxational thermodynamics become invalid at $T\leqslant T_c$, i.e., one cannot trace with Eq. (21) the transition to the excitonic superfluid phase and how, further, a collective ground state of QW excitons arises at $T\to0$ (for $\rho_{2D}[a_x^{(2D)}]^2<1$ the ground state can be interpreted in term of BE condensation of QW excitons[27,28]). While in the dilute limit $\ln(\rho_{2D}[a_x^{(2D)}]^2)\ll1$ the parameter $\alpha\ll1$, i.e., $T_c\ll T_0$,[29] there is still no first-principles theory to estimate $\alpha$ for the densities $\rho_{2D}[a_x^{(2D)}]^2\leqslant1$. With the above-mentioned restrictions, we find that cooling high-density QW excitons to very low temperatures is rather difficult. This conclusion may have some bearing on the search[30] for the collective ground state of indirect excitons in GaAs/Al$_x$Ga$_{1-x}$As CQW's.

### C. Thermalization dynamics of hot QW excitons ($T\gg T_b$)

In experiments[1–9] with nonresonant excitation of heavy-hole excitons in GaAs QW's, the initial effective temperature $T_i\gg T_b$. While in this case the thermalization kinetics can be analyzed numerically from the basic Eq. (11), we will clarify the various relaxation scenarios by analytic approximations of Eq. (11).

1. *Classical gas of QW excitons* ($T_i\gg T_b\gg T_0$)

A. *High-temperature limit* ($k_BT_b\gg E_0$). In this case Eq. (11) reduces to

$$\frac{\partial}{\partial t}T=-\frac{1}{\tau_H}(T-T_b), \tag{25}$$

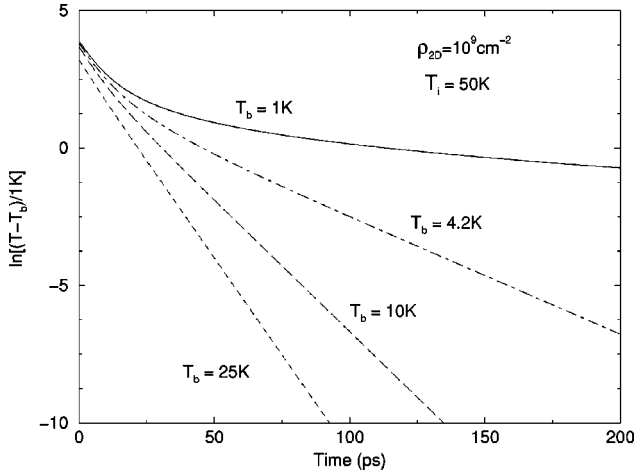


FIG. 6. The relaxational dynamics of a classical gas of hot QW excitons at $T_b=1$ K (solid line), 4.2 K (dot-dashed line), 10 K (long dashed line), and 25 K (dashed line). The latter straight line refers to the exponential kinetics $\ln|(T-T_b)/(T_i-T_b)|=-t/\tau_H$, which is valid in the high-temperature limit $k_BT_b\gg E_0$.

where the characteristic thermalization time $\tau_H$ is given by Eq. (15). Thus, in the high-temperature limit $k_BT_b\gg E_0$ the exponential law $T(t)=T_b+(T_i-T_b)\exp(-t/\tau_H)$ is valid even for $(T_i-T_b)/T_b\gg 1$.

B. *Low-temperature limit* ($k_BT_b<E_0$). The initial hot thermalization down to $k_BT\simeq E_0$ is approximated by

$$\frac{\partial}{\partial t}T=-\frac{T}{\tau_H}. \tag{26}$$

This exponential cooling $T(t)=T_i\exp(-t/\tau_H)$ completes at $t'\simeq\tau_H\ln(T_i/T_b)$. The hot thermalization refers to the high-temperature limit given by Eq. (25). At $t>t'$ the cold QW excitons relax to the bath temperature according to

$$\frac{\partial}{\partial t}T=-\frac{1}{\tau_L}(T-T_b), \tag{27}$$

where the low-temperature thermalization time $\tau_L$ is defined by Eq. (16). Approximation (27) is valid for $(T-T_b)/T_b<k_BT_b/E_0$, yields the exponential relaxation with $\tau_L$, and corresponds to the linearization of Eq. (11) at $T=T_b$.

If the high-temperature limit does not hold, the thermalization kinetics of Maxwell-Boltzmann distributed QW excitons is exponential only locally in time, i.e., $\tau_{th}=\tau_{th}(t)$. The described above picture of the relaxation in the low-temperature limit in fact deals with a continuous increase of the instant thermalization time $\tau_{th}(t)$ from $\tau_H$ to $\tau_L$. As a result, for $T_i-T_b\gg T_b$ the total relaxation process can show a nonexponential behavior. In Fig. 6 we illustrate how a single-exponential thermalization kinetics with $\tau_{th}=\tau_H$ of a classical gas of QW excitons initially distributed at $T_i\gg T_b$ develops with the decreasing bath temperature toward the nonexponential relaxation.

2. *Quantum gas of QW excitons* ($T_i\gg T_0\gg T_b$)

A. *High-temperature limit* ($k_BT_b\gg E_0$). The initial hot relaxation to the temperature $T\simeq T_0$ is given by Eq. (26), and completes at $t'\simeq\tau_H\ln(T_i/T_0)$. At $t\geqslant t'$ QW excitons become degenerate, and the BE statistics starts to influence the thermalization process. The relaxation kinetics is approximated by

$$\frac{\partial}{\partial t}T=-\left(\frac{T}{T_b}\right)^2\frac{(T-T_b)}{\tau_{0H}}, \tag{28}$$

where $\tau_{0H}\propto\rho_{2D}$ is given by Eq. (18). Equation (28) yields the nonexponential transient thermalization from $T\simeq T_0$ to $T\simeq T_b$:

$$T(t)=\frac{T_0}{[1+2(T_0/T_b)^2[(t-t')/\tau_{0H}]]^{1/2}}. \tag{29}$$

The nonexponential transient lasts for a time $\Delta t\simeq\tau_{0H}/2$. The relaxation kinetics from $t\geqslant t''\simeq t'+\tau_{0H}/2$ refers to $T-T_b<T_b$, and recovers the exponential law with $\tau_{0H}$.

B. *Low-temperature limit* ($k_BT_b<E_0$). At $0\leqslant t\leqslant t'\simeq\tau_H\ln(T_i/T_0)$ the first hot thermalization from $T=T_i$ to $T\simeq T_0$ is characterized by the exponential law of Eq. (26). The next nonexponential transient relaxation from $T\simeq T_0$ to $T\simeq E_0/k_B$ is given by Eq. (29), and corresponds to the time interval $t'\leqslant t\leqslant t''\simeq(k_BT_b/E_0)^2(\tau_{0H}/2)$. These two stages are similar to those found in the high-temperature limit. For the thermalization from $T\simeq E_0/k_B$ to $T\simeq T_b$ one recovers Eq. (22), and the corresponding $1/\ln(\tau)$ kinetics of Eq. (24) treated in Sec. III B for $T_b=0$. This stage is rather long, and completes at $t'''\simeq(k_BT_0\tau_{sc}/2\pi^{3/2}E_0)\exp(E_0/k_BT_b)=(k_BT_b/E_0)^{1/2}\tau_{0L}$. The final exponential relaxation refers to $(T-T_b)/T_b<k_BT_b/E_0$, and is given by $T(t\geqslant t''')=T_b+(k_BT_b^2/E_0)\exp[-(t-t''')/\tau_{0L}]$, where the low-temperature thermalization time $\tau_{0L}$ of the degenerate QW excitons is defined by Eq. (20).

In order to demonstrate how BE statistics can influence the thermalization kinetics of QW excitons at $T_i\gg T_b$, in Fig. 7 we plot the numerical solution $T=T(t)$ of Eq. (11) for $T_i=50$ K, $T_b=4.2$ K, and various values of the concentration $\rho_{2D}$. The development of the nonexponential relaxation with the increasing degeneracy temperature $T_0\propto\rho_{2D}$ is clearly seen. The inset of Fig. 7 illustrates the transient thermalization given by Eq. (29).

## IV. RESONANT PHOTOLUMINESCENCE OF QUANTUM DEGENERATE QUASI-2D EXCITONS

In high-quality GaAs QW's the decay of quasi-2D excitons is mainly due to radiative recombination.[3–10] Moreover, following the first studies[31] of the optical properties of excitons in GaAs/$Al_xGa_{1-x}$As CQW's, very recent experiments[13] show that the radiative recombination channel can also be dominant for the long-lived indirect excitons. In this section we generalize the theory[3,16] of steady-state resonant PL of a quasiequilibrium classical gas of QW excitons to well-developed BE statistics, and exploit the relaxation thermodynamics of Eq. (11) in order to analyze the PL kinetics of QW excitons.

In perfect QW's an exciton can emit a bulk photon only from radiative modes, which are located inside the photon cone given by $k=k(\omega)=(\sqrt{\epsilon_b}\omega)/(\hbar c)$. Here $\omega$ is the frequency of light, and $\epsilon_b$ is the background dielectric constant for an exciton line. This means that the radiative zone

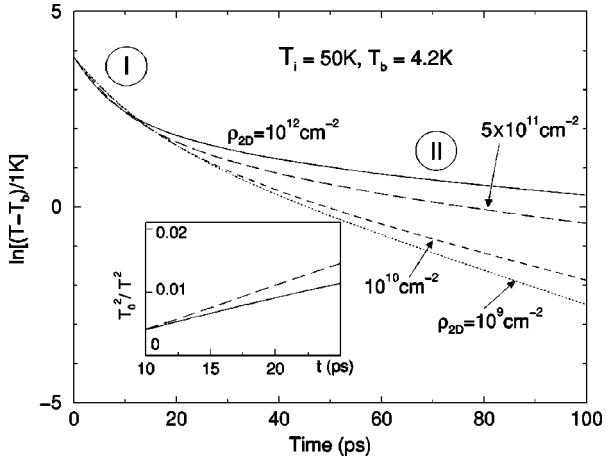


FIG. 7. Thermalization kinetics of hot QW excitons with densities $10^{12}$ cm$^{-2}$ (solid line), $5\times10^{11}$ cm$^{-2}$ (long dashed line), $10^{10}$ cm$^{-2}$ (dashed line), and $10^{9}$ cm$^{-2}$ (dotted line). The initial effective temperature of QW excitons is $T_i=50$ K, and the bath temperature is $T_b=4.2$ K, a GaAs QW with $L_z=100$ Å and $\tau_{sc}=13.9$ ns. The first transient thermalization occurs in the time interval $I$ ($t\leqslant 20-40$ ps). The density-dependent quasi-steady-state relaxation develops in time domain $II$, where $T-T_b<T_b$. Inset: verification of the approximation by Eq. (29) of the nonexponential transient relaxation of strongly degenerate QW excitons.

of QW excitons is given by $p_\parallel\leqslant k_0$, where $k_0=k(\omega_t)$ corresponds to crossover of the photon and exciton dispersions, and $\hbar\omega_t$ is the exciton energy. The intrinsic radiative rates for in-plane transverse ($T$-polarized) and in-plane longitudinal ($L$-polarized) dipole-active QW excitons are given by[16]

$$\Gamma_T(p_\parallel)=\Gamma_0\frac{k_0}{\sqrt{k_0^2-p_\parallel^2}} \tag{30}$$

and

$$\Gamma_L(p_\parallel)=\Gamma_0\frac{\sqrt{k_0^2-p_\parallel^2}}{k_0},$$

respectively. The intrinsic radiative lifetime $\tau_R$ of a QW exciton is defined by $\tau_R=1/\Gamma_0$, where $\Gamma_0=\Gamma_0(L_z)=\Gamma_T(p_\parallel=0)=\Gamma_L(p_\parallel=0)$ is the radiative rate for the ground-state mode $\mathbf{p}_\parallel=0$. $Z$-polarized heavy-hole QW excitons are forbidden. Here we use the polarization notations of Ref. 16.

The total radiative decay rate $\Gamma_{opt}=1/\tau_{opt}$ of a gas of quasiequilibrium BE-distributed QW excitons is

$$\Gamma_{opt}=\frac{1}{\rho_{2D}}\int_0^{k_0}N_{p_\parallel}[\Gamma_T(p_\parallel)+\Gamma_L(p_\parallel)]\frac{p_\parallel dp_\parallel}{2\pi}, \tag{31}$$

where the occupation number $N_{p_\parallel}=N^{eq}_{E_{p_\parallel}}$ is given by Eq. (6). Equation (31) takes into account the equal probabilities of the $T$ and $L$ polarizations and twofold spin degeneracies, $\sigma^+$ and $\sigma^-$ of dipole-active QW excitons. Using Eqs. (6) and (30) we find, from Eq. (31),

$$\Gamma_{opt}=\frac{\Gamma_0}{2}[J_T(T,T_0)+J_L(T,T_0)],$$

$$J_T(T,T_0)=\left(\frac{E_{k_0}}{k_BT_0}\right)\int_0^1\frac{dz}{Ae^{-z^2E_{k_0}/k_BT}-1}, \tag{32}$$

$$J_L(T,T_0)=\left(\frac{E_{k_0}}{k_BT_0}\right)\int_0^1\frac{z^2dz}{Ae^{-z^2E_{k_0}/k_BT}-1},$$

where $A=A(T,T_0)=e^{E_{k_0}/k_BT}/(1-e^{-T_0/T})$ and $E_{k_0}=\hbar^2k_0^2/2M_x$ (for GaAs QW's with $\epsilon_b=12.9$ and $\hbar\omega_t=1.55$ eV, one has $E_{k_0}\simeq 101$ $\mu$eV and $E_{k_0}/k_B\simeq 1.17$ K). The contribution of twofold dipole-inactive (triplet) QW excitons to a total density $\rho_{2D}$ is included in Eq. (32) through the degeneracy temperature $T_0\propto\rho_{2D}$.

For Maxwell-Boltzmann distributed QW excitons at an effective temperature $T$ much larger than $T_0$ and $E_{k_0}/k_B$, we find from Eq. (32) that $J_T=3J_L=(\hbar^2k_0^2)/(M_xT)$ and

$$\Gamma^{cl}_{opt}=\left(\frac{\hbar^2k_0^2}{3M_xk_BT}\right)\Gamma_0. \tag{33}$$

In the limit $T\ll T_0$ of a strongly degenerate gas of QW excitons, one approximates $J_T=1+(T/T_0)\ln(4E_{k_0}/k_BT)$ and $J_L=1+(T/T_0)[\ln(4E_{k_0}/k_BT)-2]$. In this case $\Gamma_{opt}$ of Eq. (32) reduces to

$$\Gamma^q_{opt}=\left[1-\frac{T}{T_0}+\frac{T}{T_0}\ln\left(\frac{2\hbar^2k_0^2}{M_xk_BT}\right)\right]\frac{\Gamma_0}{2}. \tag{34}$$

The expansion of $J_L(T,T_0)$, $J_T(T,T_0)$, and $\Gamma_{opt}$ in terms of the ratio $T/T_0\ll 1$ is valid for the effective temperature of QW excitons $T\leqslant E_{k_0}\exp(-T_0/T)$. The second and third terms in the square brackets on the right-hand side of Eq. (34) are small corrections, i.e., $\Gamma^q_{opt}(T\to 0)\to\Gamma_0/2$. The nonzero limit of $\Gamma_{opt}$ at $T\to 0$, which is completely determined by the intrinsic radiative rate $\Gamma_0$ of the ground-state mode $\mathbf{p}_\parallel=0$, is due to the effective accumulation of low-energy QW excitons with $N_E\gg 1$ in the radiative zone $p_\parallel\leqslant k_0$.

In the limit $T\gg T_0,E_{k_0}$ of a classical behavior, one derives, from Eqs. (31) and (32),

$$\tau^{cl}_{opt}=\left(\frac{3M_xk_BT}{\hbar^2k_0^2}\right)\tau_R+\left(\frac{9}{10}-\frac{3M_xk_BT_0}{2\hbar^2k_0^2}\right)\tau_R, \tag{35}$$

where in the expansion of $\tau^{cl}_{opt}=1/\Gamma_{opt}$ we keep not only the leading term $1/\Gamma^{cl}_{opt}\propto T$ [see Eq. (33)], but also the temperature-independent correction. The next terms of the expansion are proportional to $1/T^{n\geqslant 1}\to 0$, and can indeed be neglected. The first term on the right-hand side of Eq. (35), i.e., $1/\Gamma^{cl}_{opt}=(3M_xk_BT\tau_R)/(\hbar^2k_0^2)$, is a well-known result of Ref. 16. The temperature-independent correction, which is given by the second term on the right-hand side of Eq. (35) consists of the density-independent and -dependent contributions of the opposite signs. The density-dependent contribution originates from the BE distribution function used in Eq. (31) and can be much larger than $\tau_R$ if $k_BT\gg k_BT_0\gg E_{k_0}$. Note that the density-independent term (9/10) $\tau_R$ can be derived even with Maxwell-Boltzmann statistics. In the leading

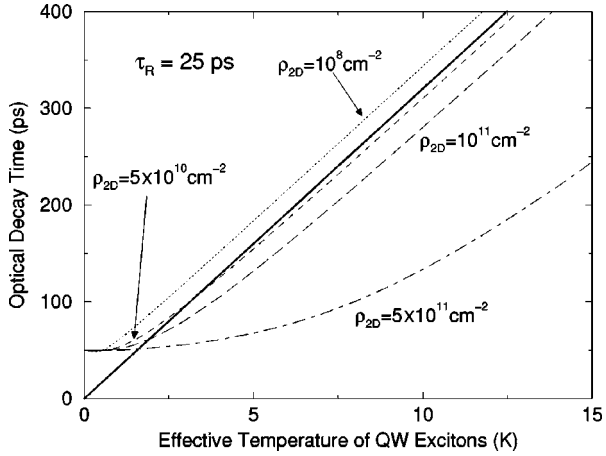


FIG. 8. The optical decay time $\tau_{opt}$ vs the effective temperature $T$ of QW excitons: $\rho_{2D}=10^8$ cm$^{-2}$ (dotted line), $5\times10^{10}$ cm$^{-2}$ (dashed line), $10^{11}$ cm$^{-2}$ (dot-dashed line), and $5\times10^{11}$ cm$^{-2}$ (solid line). The intrinsic radiative lifetime of QW excitons is $\tau_R=25$ ps. The bold straight line is given by $1/\Gamma_{opt}^{cl}=(3M_xk_BT)/(\hbar^2k_0^2)$.

term $1/\Gamma_{opt}^{cl}\propto T$, one has that the optical decay time $\tau_{opt}^{cl}\gg\tau_R$, because only a small fraction of QW excitons occupy the radiative modes $p_\parallel\leqslant k_0$ at $T\gg T_0,E_{k_0}/k_B$.[3,16]

In Fig. 8 we plot the effective decay time of quasiequilibrium QW excitons $\tau_{opt}=\tau_{opt}(T)$, calculated numerically with Eq. (32) for various values of the concentration $\rho_{2D}$. Following Ref. 16, for a GaAs QW with $L_z=100$ Å, we use $\tau_R=25$ ps. For $T\gg T_0,E_{k_0}/k_B$, the linear behavior given by Eq. (35) is clearly seen (dotted, dashed, and long-dashed lines in Fig. 8). The reference bold solid line indicates the $1/\Gamma_{opt}^{cl}\propto T$ law. Note that the linear asymptotics of $\tau_{opt}=\tau_{opt}(T,\rho_{2D})$ from the classical range $T\gg T_0,E_{k_0}/k_B$ down to low temperatures $T\to0$ reveal nonzero density-dependent values. Thus one concludes that even at high temperatures $T\gg T_0$ the influence of nonclassical statistics can be found in the PL of QW excitons. At $T\to0$ the limit $\tau_{opt}^{cl}\to\tau_R[9/10-(3k_BT_0)/(4E_{k_0})]$ given by Eq. (35) breaks, and all curves approach the double intrinsic radiative lifetime $2\tau_R$. The density-dependent deviation from the linear classical law of Eq. (35) develops with increasing $\rho_{2D}$. For example, at $\rho_{2D}=5\times10^{11}$ cm$^{-2}$ the quantum asymptotics $\tau_{opt}^q(T\to0)=2\tau_R$ can already be traced for the effective temperature $T\simeq10$ K (see Fig. 8).

According to Eq. (30), the decay rate of $T$-polarized QW excitons diverges at $p_\parallel\to k_0$.[16] In Appendix B we show how the homogeneous linewidth of low-energy QW excitons $\hbar\Gamma_{hom}=\hbar/\tau_{x\text{-}x}+\hbar/\tau_{ph}+\hbar/\tau_R\simeq\hbar/\tau_{x\text{-}x}$ removes this divergence [see Eq. (B6)]. In the presence of homogeneous broadening, due to dominant exciton-exciton scattering, $\Gamma_T(p_\parallel)$ of Eq. (30) changes only in a very narrow band of QW states given by $0\leqslant k_0-p_\parallel\leqslant\tilde{\gamma}k_0$, where $\tilde{\gamma}=1/(\tau_{x-x}\omega_t)\ll1$. In particular, $\Gamma_T(p_\parallel\to k_0)=\Gamma_0/(2\tilde{\gamma}^{1/2})$ rather than diverges. Furthermore, homogeneous broadening of QW excitons from the radiative zone only slightly renormalizes the PL efficiency: $\Gamma_{opt}^{cl}$ changes on $\Gamma_{opt}^{cl}[1-(3/4)\tilde{\gamma}^{1/2}]$ (for details, see Appendix B). The correction is even less for $\Gamma_{opt}^q$ of Eq. (34). Therefore, we conclude that the corrections to the optical decay rate $\Gamma_{opt}$ due to the homogeneous linewidth $\Gamma_{hom}$ can indeed be neglected. Note, however, that inhomogeneous broadening strongly influences the PL process and changes our results obtained for a perfect QW.

The optical decay of QW excitons from the radiative zone $p_\parallel\leqslant k_0$ does not violate the assumptions of relaxational thermodynamics, provided that $\tau_{x\text{-}x}\ll\tau_R$. In this case the thermal quasiequilibrium distribution of the finite lifetime QW excitons at $E\leqslant E_{k_0}$ holds due to exciton-exciton scattering. Because both the minimal thermalization time $\tau_{th}$ given by $\tau_H$ of Eq. (15) and the intrinsic radiative time $\tau_R$ are on the same time scale of 5– 50 ps, the condition $\tau_{x\text{-}x}\ll\tau_H$ [see Eq. (9)] of the thermodynamic picture also nearly guarantees that $\tau_{x\text{-}x}\ll\tau_R$.

The radiative decay leads to the continuous decrease of the density of QW excitons (or the degeneracy temperature $T_0\propto\rho_{2D}$) and gives rise to the PL signal according to

$$\frac{\partial}{\partial t}\rho_{2D}=-\Gamma_{opt}(T_0,T)\rho_{2D}\equiv-\frac{\rho_{2D}}{\tau_{opt}(\rho_{2D},T)}, \tag{36a}$$

$$I_{PL}=-\hbar\omega_t\frac{\partial\rho_{2D}}{\partial t}\equiv\hbar\omega_t\frac{\rho_{2D}}{\tau_{opt}(\rho_{2D},T)}, \tag{36b}$$

where $I_{PL}=I_{PL}(t)$ is the photoluminescence intensity. Thus Eqs. (11), (32), (36a), and (36b) describe the PL kinetics of QW excitons cooling from the initial effective temperature $T_i$ to the bath $T_b$. For a high-temperature classical gas of QW excitons the optical decay time $\tau_{opt}^{cl}$ of Eq. (35) is approximated by the density-independent $1/\Gamma_{opt}^{cl}$ of Eq. (33) and the solution of Eq. (36a) is simply $\rho_{2D}(t)=\rho_{2D}^{(0)}\exp(-\int_0^t\Gamma_{opt}^{cl}(t)dt)$, where $\rho_{2D}^{(0)}$ is the initial concentration of QW excitons and $\Gamma_{opt}^{cl}$ is time dependent through the effective temperature of QW excitons $T=T(t)$. In the general case, however, Eq. (36a) gives a nonexponential decay of the density. The characteristic times of both the fundamental processes which contribute to the PL kinetics, LA-phonon-assisted thermalization and optical decay, generally depend upon the effective temperature and density of QW excitons, i.e., $\tau_{th}=\tau_{th}(T,T_0)$ and $\tau_{opt}=\tau_{opt}(T,T_0)$. As a result, the PL kinetics given by Eqs. (11), (32), (36a), and (36b) is also $T$ and $\rho_{2D}$ dependent. The thermalization process and radiative decay work in the opposite directions with respect to the quantum-statistical effects. Cooling of QW excitons is accompanied by an increasing number of low-energy particles toward well-developed BE statistics. In the meantime $\tau_{opt}$ decreases, and the optical decay of $\rho_{2D}$ speeds up, resulting in decreasing $T_0$. Thus the radiative processes interfere with the development of BE statistics.

For a classical gas of QW excitons in the high-temperature limit $k_BT_b\gg E_0$ the PL dynamics of Eqs. (11), (32), (36a), and (36b) can be analyzed analytically. In this case, $T(t)=T_b+(T_i-T_b)\exp(-t/\tau_H)$, where the temperature-independent $\tau_H$ is given by Eq. (15). At the first transient thermalization of hot QW excitons from the initial distribution at effective $T_i\gg T_b$ one has that $\tau_{opt}^{cl}(T_i)\gg\tau_H$, i.e., the optical decay is very slow and practically does not change the concentration of QW excitons. The transient stage lasts a few $\tau_H$ and for this time domain Eq. (36b) reduces to

$$I_{PL}(t\leqslant t_{tr})=\hbar\omega_t\left(\frac{\hbar^2k_0^2}{3M_xk_BT_i}\right)\rho_{2D}^{(0)}e^{t/\tau_H}\equiv\hbar\omega_t\frac{\rho_{2D}^{(0)}}{\tau_{opt}^{cl}(T_i)}e^{t/\tau_H}, \tag{37}$$

i.e., $I_{PL}(t\leqslant t_{tr})\propto\exp(t/\tau_H)$. Therefore, at the hot relaxation stage $t\leqslant t_{tr}$ the PL intensity increases exponentially with the rate $1/\tau_H$, due to the population of the radiative zone. The transient stage completes when $T(t_{tr})-T_b\leqslant T_b$. The PL intensity reaches its maximum at $t=t_0>t_{tr}$, when the population efficiency of the modes $p_\parallel\leqslant k_0$ is compensated by the increasing optical decay:

$$t_0=\tau_H\ln\left(\frac{3M_xk_BT_i}{\hbar^2k_0^2}\frac{\tau_R}{\tau_H}\right)\equiv\tau_H\ln\left[\frac{\tau_{opt}^{cl}(T_i)}{\tau_H}\right]. \tag{38}$$

The corresponding effective temperature of QW excitons is $T(t_0)=T_b[1+\tau_H/\tau_{opt}^{cl}(T_i)]$, i.e., $T(t_0)-T_b\ll T_b$. At $t=t_s>t_0$ the PL dynamics reaches a steady state, and is determined by the optical decay, because the QW excitons are already thermalized at $T=T_b$. For this time domain the PL kinetics is given by

$$I_{PL}(t\geqslant t_s)=\hbar\omega_t\left(\frac{\hbar^2k_0^2}{3M_xk_BT_b}\right)\rho_{2D}^{(s)}e^{-t/\tau_{opt}^{cl}(T_b)}$$

$$\equiv\hbar\omega_t\frac{\rho_{2D}^{(s)}}{\tau_{opt}^{cl}(T_b)}e^{-t/\tau_{opt}^{cl}(T_b)}, \tag{39}$$

where $\rho_{2D}^{(s)}=\rho_{2D}(t_s)\simeq\rho_{2D}^{(0)}$. Thus at the steady-state regime $I_{PL}(t\geqslant t_s)\propto\exp[-t/\tau_{opt}^{cl}(T_b)]$, i.e., the PL intensity decreases exponentially with the optical decay rate $\Gamma_{opt}^{cl}(T_b)$. Note that because usually $T_i\gg T_0$ and $T_0(t\to\infty)\to 0$, the very first transient stage and the very last steady-state decay of excitonic PL always follow Eqs. (37) and (39), respectively.

In Fig. 9(a) we plot numerical simulations of the PL dynamics done with Eqs. (11), (32), (36a) and (36b) for excitons of various initial densities in GaAs QW with $L_z=100$ Å. In order to model the experimental data on the density-dependent PL kinetics plotted in Fig. 2 of Ref. 4(a) we use $\tau_{sc}=85$ ns and $\tau_R=40$ ps. The change of the PL dynamics with increasing $\rho_{2D}^{(0)}$ shown in Fig. 9(a) reproduces qualitatively the corresponding experimental observations of Ref. 4(a). The decrease of the rise time $t_0$ of the PL signal with the increasing initial density of QW excitons is mainly due to increase of the optical decay rate at high densities [see the inset of Fig. 9(a)]. This effect originates from BE statistics of QW excitons.

In Fig. 9(b) we show numerical modeling of PL kinetics in $GaAs/Al_xGa_{1-x}As$ CQW's with the intrinsic radiative lifetime of long-lived excitons $\tau_R=30$ ns. Here the BE statistics strongly influences the PL temporal behavior in the steady-state regime at $t\geqslant t_s$, but still before a time when indirect excitons become Maxwell-Boltzmann distributed due to continuous decrease of $\rho_{2D}$. At this time domain the thermalization kinetics of high-density CQW excitons undergoes a critical slowing down, the effective temperature of CQW excitons is nearly stabilized at $T_0>T>T_b$, and the occupation numbers of the radiative modes $N_{p_\parallel\leqslant k_0}\gg 1$. As a result, with the increasing initial concentration $\rho_{2D}^{(0)}$ the decay rate of the PL signal approaches $\tau_{opt}^q=2\tau_R$, i.e., one has an

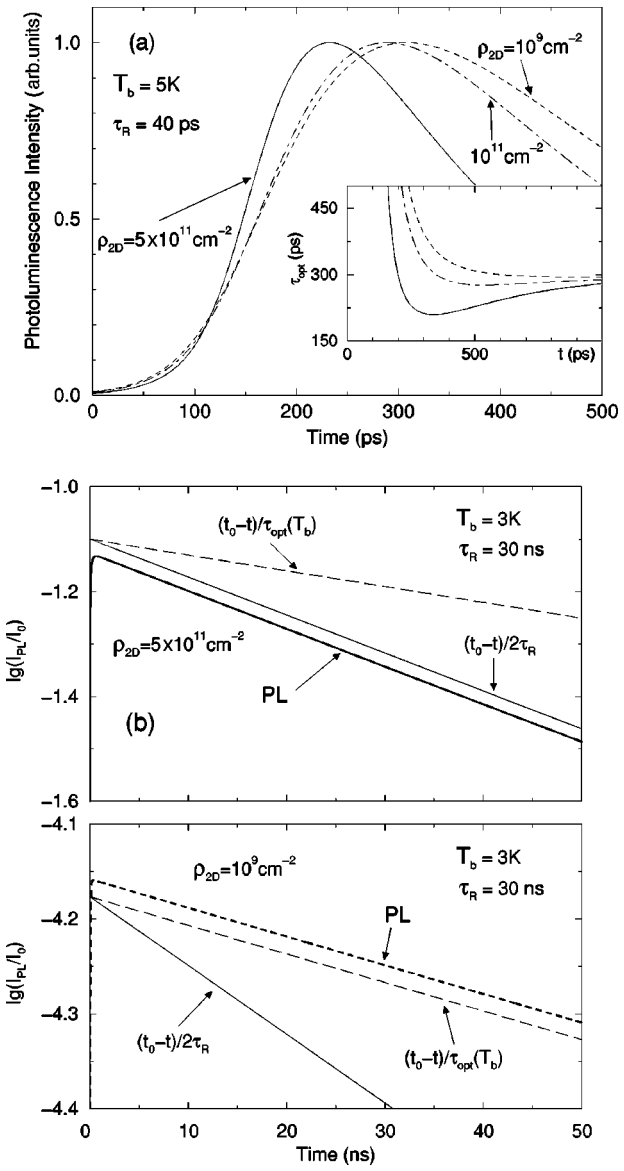


FIG. 9. Phototuminescence dynamics of QW excitons modeled with Eqs. (11), (32), (36a), and (36b). (a) GaAs QW with $L_z=100$ Å, $D=6.3$ eV, $\tau_{sc}=85$ ns, $\tau_R=40$ ps, $T_b=5$ K, $\rho_{2D}=10^9$ cm$^{-2}$ (dashed line), $10^{11}$ cm$^{-2}$ (dot-dashed line), and $5\times10^{11}$ cm$^{-2}$ (solid line). Inset: change of the optical decay time $\tau_{opt}=\tau_{opt}(t)$. (b) $GaAs/Al_xGa_{1-x}As$ CQW's with $\tau_{sc}=13.9$ ns, $\tau_R=30$ ns, $T_b=3$ K, $\rho_{2D}=10^9$ cm$^{-2}$ (bold dashed line), and $5\times10^{11}$ cm$^{-2}$ (bold solid line). The long dashed and solid straight lines refer to the exponential kinetics given by $I_{PL}(t)\propto\exp[-t/\tau_{opt}(T_b)]$ and $I_{PL}(t)\propto\exp[-t/(2\tau_R)]$, respectively. The PL intensities $I_{PL}(t)$ of (a) and (b) are normalized by $I_0=248.31$ kW/cm$^2$.

acceleration of the PL optical decay owing to well-developed BE statistics [see Fig. 9(b), where the long dashed and solid reference lines indicate $\ln(I_{PL}/I_0)=(t_0-t)/2\tau_{opt}(T_b)$ and $\ln(I_{PL}/I_0)=(t_0-t)/2\tau_R$, respectively].

## V. DISCUSSION

There are three neglected factors—the interface polariton effect, the low-energy biexciton states, and residual interface disorder—which can affect the developed model. In perfect

QW's, the radiative modes $p_\parallel < k(\omega)$, which are responsible for the optical decay of QW excitons in bulk photons, are accompanied by confined modes $p_\parallel > k(\omega)$ located outside the photon cone. These modes give rise to QW interface polaritons. While the QW polaritons cannot be seen in standard optical experiments, which deal with bulk photons, they can modify the quadratic in-plane dispersion of QW excitons and contribute to the total optics in a ''hidden'' way. Therefore in the general case both conjugated phenomena—the radiative decay into bulk photons and the QW polariton effect—should be treated simultaneously. The dispersion law of $T$-polarized QW polaritons is

$$\frac{c^2 p_\parallel^2}{\epsilon_b} = \omega^2 + \frac{\omega^2 R_c \sqrt{p_\parallel^2 - \epsilon_b \omega^2/c^2}}{\omega_t^2 + \hbar \omega_t p_\parallel^2/M_x - i\Gamma^x \omega - \omega^2},$$

where $R_c = (\pi e^2 f_{xy})/(\epsilon_b m_0)$, $f_{xy}$ is the oscillator strength of exciton-photon coupling per unit area of a QW (the subscript $xy$ refers to the in-plane polarization of the light), and $\Gamma^x$ is the rate of incoherent scattering of QW excitons.[19,32] Actually the dispersion equation can be applied to the both radiative and confined modes.[33] Within the photon cone the solution $\omega = \omega(p_\parallel)$ is $\omega = \omega_t - i\Gamma_T(p_\parallel)/2$, where $\Gamma_T(p_\parallel)$ is given by Eq. (30), with $\Gamma_0 = (\sqrt{\epsilon_b}/c)R_c$. The dispersion $\omega = \omega_T(p_\parallel)$ of $T$-polarized QW polaritons is located outside the photon cone, approaches the quadratic dispersion $\hbar p_\parallel^2/2M_x$ at $p_\parallel \gg k_0$, and continuously transforms to the photon dispersion $cp_\parallel/\sqrt{\epsilon_b}$ at $p_\parallel \ll k_0$. Thus during thermalization the QW excitons can accumulate at the ''bottleneck'' band of the $T$-polariton dispersion rather than in the radiative zone given by $\omega = \omega_t + \hbar p_\parallel^2/2M_x$ for $p_\parallel \ll k_0$.

The polariton effect will not influence the thermalization and PL kinetics provided that $E_{k_0} \gg (1/2)\hbar\Omega_c^{QW}$, where the effective QW polariton parameter $\Omega_c^{QW}$ is given by $(\Omega_c^{QW})^2 = (\sqrt{\epsilon_b}/c)R_c\omega_t$. In this case the QW polariton bottleneck band is very narrow, located at energies above $\hbar\omega_t$, and the mode $\mathbf{p}_\parallel = 0$ preserves the status of a ground-state mode. For a GaAs QW with $L_z = 100$ Å one has $f_{xy} \simeq 5\times 10^{-4}$ Å$^{-2}$.[16] This value of the oscillator strength yields $\hbar^2 R_c \simeq 1.2\times 10^{-2}$ eV$^2$ Å, $\tau_R = 1/\Gamma_0 \simeq 25$ ps, and $\frac{1}{2}\hbar\Omega_c^{QW} \simeq 3$ meV, i.e., $(1/2)\hbar\Omega_c^{QW} > E_{k_0} \simeq 0.1$ meV. In perfect CQW's the oscillator strength $f_{xy}$ can be made two or three orders smaller than the value used above. This means that indeed $E_{k_0} \gg (1/2)\hbar\Omega_c^{QW}$ and one can apply Eqs. (11), (32), (36a), and (36b) to the relaxational and PL dynamics of indirect CQW excitons.

The exciton-exciton interaction relaxes the polariton effect in perfect QW's and removes it, if $\Gamma^x = 1/\tau_{x-x} \gg \Omega_c^{QW}$. With the estimate of $1/\tau_{x-x}$ given by Eq. (A4) of Appendix A we conclude that for direct excitons in a single GaAs QW with $L_z = 100$ Å the interface polariton effect plays no role at concentrations $\rho_{2D} > 3\times 10^{10}$ cm$^{-2}$. Alternatively, in the presence of the well-developed polariton picture with $E_{k_0} < (1/2)\hbar\Omega_c^{QW}$, the relaxation kinetics should be reformulated in terms of ''QW polariton $\pm$ bulk LA phonon $\leftrightarrow$ QW polariton.'' Furthermore, in this case relaxational thermodynamics becomes invalid due to the absence of steady-state quasiequilibrium in a gas of QW excitons (QW polaritons).

Next we consider the formation of quasi-2D biexcitons. The potential for exciton-exciton interaction is repulsive for indirect excitons in CQW's. However, in a single QW two excitons in some spin configurations, e.g., $\sigma^+$ and $\sigma^-$ spin-polarized, attract each other due to the exchange Coulomb interaction. This interaction may lead to biexciton states which lie below the excitonic energies $E \geqslant 0$. While the biexciton states are absent in CQW's, they are presented in single GaAs QW's at low temperatures and moderate optical excitations. Typical values of the biexciton binding energy $E_m^{(2D)}$ are about 1–2 meV for $L_z = 200$–$100$ Å. The law of mass action[18] applied to QW excitons and biexcitons, which are Maxwell-Boltzmann distributed at the same effective temperature $T$, shows that the biexciton states are ionized in unbound excitons at $k_B T \geqslant E_m^{(2D)}$, i.e., at $T \geqslant 10$ K. At low temperatures, with increasing dimensionless parameter $[a_m^{(2D)}]^2\rho_{2D}$ the biexciton binding energy decreases due to screening by quasiequilibrium QW excitons, which remain well-defined quasiparticles for $[a_x^{(2D)}]^2\rho_{2D} \leqslant 1$. Here $a_m^{(2D)}$ $(> a_x^{(2D)})$ is the radius of a QW biexciton. The screening of the biexciton states is mainly due to the repulsive interaction between QW excitons of the identical spin structure, which is much stronger than the attractive potential.[34,35] The biexciton state is affected by excitons if $[a_m^{(2D)}]^2\rho_{2D}E_x^{(2D)} \geqslant E_m^{(2D)}$, where $E_x^{(2D)}$ is the two-dimensional excitonic Rydberg. This estimate refers to low effective temperatures $T \to 0$. The squared biexciton radius is approximated by $[a_m^{(2D)}]^2 = 2(\mu_x/M_x)(E_x^{(2D)}/E_m^{(2D)})[a_x^{(2D)}]^2 \simeq 2.4[a_x^{(2D)}]^2$, where $\mu_x$ is the reduced exciton mass. We estimate that the biexciton state starts to weaken at $[a_x^{(2D)}]^2\rho_{2D} \geqslant 0.05$–$0.1$, i.e., at $\rho_{2D} \simeq 10^{11}$ cm$^{-2}$. Certainly in the high-density limit $\rho_{2D} \simeq 2-3\times 10^{12}$ cm$^{-2}$, where the Mott parameter $\rho_{2D}[a_x^{(2D)}]^2$ approaches unity, the thermalization and photoluminescence of strongly degenerate QW excitons occurs in the absence of the biexciton states and can be analyzed with our approach. During the optical decay, at concentrations $\rho_{2D} \sim 10^{11}$ cm$^{-2}$, the QW biexcitons can contribute to the relaxation and PL processes. For these densities of QW excitons the relaxation thermodynamics given by Eq. (11) can be generalized to include the conversion of QW excitons to QW biexcitons and the LA-phonon-assisted relaxation within the both excitonic and biexcitonic bands. In this case the quasiequilibrium concentrations of QW excitons and biexcitons are connected by the law of mass action.[18]

The developed thermalization and photoluminescence kinetics refer to a perfect QW. In two dimensions, even weak disorder will formally lead to localization so that there are no truly unbound exciton states, although the localization length may be very long. Consequently for the weak disorder, relaxation can be ''disorder assisted,'' whereby momentum and energy conservation are satisfied by LA-phonon emission concomitant with scattering from the disorder potential. This will be relevant for relieving the phonon bottleneck if there are significant Fourier components of the disorder with wave vectors $p_d \geqslant 2M_x v_s/\hbar = 1.9\times 10^5$ cm$^{-1}$. For the opposite limit of strong interface and alloy disorder, the phonon-assisted relaxation kinetics of localized QW excitons was modeled in Ref. 15 and more recently in Ref. 36, where the PL process was also studied. The optical decay of local-

ized QW excitons was investigated in Ref. 37. Note that the number of localized QW excitons saturates due to Pauli blocking, if the concentration of the residual defect centers $N_D$ is much less than $\rho_{2D}$ [in perfect GaAs QW's $N_D \leqslant 10^{10}$ cm$^{-2}$ (Ref. 37)]. In this case the localization processes do not affect the BE kinetics of QW excitons.

At low bath temperatures $T_b \leqslant E_0$ the large thermalization times $\tau_L \propto \exp(-E_0/k_B T_b)$ and $\tau_{0L} = (T_0/T_b)\tau_L$ given by Eqs. (16) and (20) for statistically nondegenerate and strongly degenerate QW excitons, respectively, can be changed by two-LA-phonon-assisted relaxation processes ''QW exciton $\pm$ bulk LA phonon $\pm$ bulk LA phonon $\leftrightarrow$ QW exciton.'' While these processes are of the next order of smallness with respect to the considered QW-exciton–one-phonon scattering, they can be dominant in the presence of drastic slowing down of one-phonon thermalization. The energy-momentum conservation law in two-phonon-assisted relaxation allows for a QW exciton with energy $E \leqslant E_0$ to be scattered into the ground-state mode $\mathbf{p}_\parallel = 0$ only by first absorbing of a LA phonon and then re-emitting another LA phonon. The rate of the anti-Stokes–Stokes two-phonon scattering into the ground-state mode is given by

$$\frac{1}{\tau_{th}^{(2)}} = \left(\frac{2\pi}{\hbar}\right) \sum_{\mathbf{q},\mathbf{q}'} |\tilde{M}_{x\text{-}ph}(\mathbf{q})|^2 |\tilde{M}_{x\text{-}ph}(\mathbf{q}')|^2 \frac{N_{\mathbf{p}_\parallel} n_{\mathbf{q}}^{ph} (n_{\mathbf{q}'}^{ph} + 1)}{(E_{\mathbf{p}_\parallel} + \hbar\omega_{\mathbf{q}}^{ph} - E_{\mathbf{p}_\parallel + q})^2} \delta(E_{\mathbf{p}_\parallel} + \hbar\omega_{\mathbf{q}}^{ph} - \hbar\omega_{\mathbf{p}_\parallel + q}^{ph}) \delta_{\mathbf{p}_\parallel - q' + q}, \tag{40}$$

where $E_{\mathbf{p}_\parallel} = \hbar^2 p_\parallel^2 / 2M_x$ and $\hbar\omega_{\mathbf{q}}^{ph} = \hbar v_s q$ are the energies of a QW exciton and a bulk LA phonon, respectively. The matrix element of QW-exciton–bulk LA-phonon interaction is $\tilde{M}_{x\text{-}ph}(\mathbf{q}) = [(\hbar D^2 q)/(2 v_s \rho V)]^{1/2} F_z(q_z L_z/2)$, where $V$ is the volume which refers to bulk LA-phonons. For a quantum gas of excitons Eq. (40) yields the following estimate of the two-phonon-assisted relaxation rate:

$$\frac{1}{\tau_{0L}^{(2)}} = 8\pi \left(\frac{k_B T_b}{E_0}\right)^4 \frac{\hbar}{E_0 \tau_{sc}^2}. \tag{41}$$

This result can be motivated as follows. Due to the anti-Stokes component, only a small phase-space volume given by $p_\parallel \leqslant p_\parallel^{max} = (2 k_B T_b)/(\hbar v_s)$ contributes to the two-phonon relaxation. The dependence $1/\tau_{0L}^{(2)} \propto T_b^4$ stems from $N_{\mathbf{p}_\parallel} n_{\mathbf{q}}^{ph} (n_{\mathbf{p}_\parallel + q}^{ph} + 1) \propto T_b^3$ and from $p_\parallel^{max} \propto T_b$ on the right-hand side of Eq. (40). The two-phonon-assisted thermalization time $\tau_{0L}^{(2)}$ is proportional to $\tau_{sc}^2$, indicating the next order of smallness with respect to $\tau_{0L} \propto \tau_{sc}$. Equations (20) and (41) show that $\tau_{0L}^{(2)}(T_b)$ indeed increases with decreasing $T_b$ more slowly than $\tau_{0L}(T_b)$, i.e., the two-phonon scattering processes become dominant at $0 < T_b \leqslant T_b^c$, where $T_b^c$ is given by the equation $\tau_{0L}(T_b) = \tau_{0L}^{(2)}(T_b)$. For example, for $T_b/T_0 = 0.2$ the crossover occurs at $T_b^c \simeq 0.06 E_0/k_B = 31.3$ mK. Because many-phonon-assisted relaxation of QW excitons at the effective temperature $T \leqslant E_0/k_B$ into the ground-state mode occurs only through intermediate anti-Stokes scattering, the relaxation dynamics at $T_b = 0$ described by Eq. (21) is universal.

Thus our model refers to indirect excitons in perfect GaAs/Al$_x$Ga$_{1-x}$As CQW's, and to direct excitons in single QW's. In the latter case, however, the concentration of QW excitons should be $\rho_{2D} \geqslant 10^{11}$ cm$^{-2}$ in order to suppress the interface polariton and biexciton effects. Equations (11), (32), (36a), and (36b) provide us with a universal description of the thermalization and photoluminescence kinetics of QW excitons within the thermodynamic picture. Note that the concrete characteristics of QW-exciton–bulk-LA-phonon interaction enter the basic thermodynamic Eq. (11) only through the elementary scattering time $\tau_{sc}$ and the form-factor function $F_z(\chi)$. Our approach can also be applied to QW magnetoexcitons. In this case the quantum-statistical effects will be considerably enhanced, because the spin degeneracy factor $g$ of Eq. (1) reduces from $g = 4$ to $g = 1$.

## VI. CONCLUSIONS

In this paper we have studied the influence of Bose-Einstein statistics on the thermalization, radiative decay, and photoluminescence kinetics of a gas of QW excitons. The numerical calculations presented in the work deal with perfect GaAs/Al$_x$Ga$_{1-x}$As quantum wells. The following conclusions summarize our results.

(i) We have developed relaxational thermodynamics for QW excitons coupled to bulk thermal LA phonons. The thermodynamic picture implies that the concentration $\rho_{2D}$ of QW excitons is larger than the critical density $\rho_{2D}^c$ (for GaAs QW's with $L_z = 100$ Å, we estimate $\rho_{2D}^c \simeq 1.2 \times 10^9$ cm$^{-1}$). The relaxational thermodynamics is given by Eq. (11) for the effective temperature $T(t)$ of QW excitons. This equation describes the thermalization kinetics of a quasiequilibrium gas of QW excitons from the initial temperature $T_i$ to the bath $T_b$. For a quantum degenerate gas of quasi-2D excitons, when $T$ is less than the degeneracy temperature $T_0$, BE statistics yields nonexponential and density-dependent thermalization. In particular, for low bath temperatures $T_b \leqslant E_0/k_B \sim 1$ K the relaxation processes in a degenerate gas of QW excitons slow down dramatically, and the thermalization law is given by $T(t) \propto 1/\ln t$.

(ii) The effective radiative lifetime $\tau_{opt} = 1/\Gamma_{opt} = \tau_{opt}(T, T_0)$ of quasiequilibrium BE-distributed QW excitons is given by Eq. (32). The temperature-independent corrections, given by Eq. (35), to the well-known classical limit $\tau_{opt} \propto T$ show that BE statistics can be traced in the optical decay of a gas of QW excitons even at high effective temperatures $T$. Nonclassical statistics strongly influences the optical decay at $T \leqslant T_0$. In particular, at $T \to 0$ the lifetime $\tau_{opt}$ approaches $2\tau_R$ rather than zero as in the classical limit $\tau_{opt} \propto T$. Here the intrinsic radiative lifetime $\tau_R$ is determined

by the oscillator strength $f_{xy}$ of exciton-photon coupling in a QW.

(iii) The PL kinetics of QW excitons is modeled by three coupled equations (11), (32), (36a), and (36b) for the effective temperature $T(t)$, effective radiative time $\tau_{opt}(T,\rho_{2D}) = 1/\Gamma_{opt}(T,\rho_{2D})$, and concentration $\rho_{2D}(t)$, respectively. This approach describes within the thermodynamic picture both the rise of PL, due to thermalization, and the decay of PL, due to the radiative recombination of thermalized QW excitons. For a classical gas of QW excitons the PL kinetics depends on the effective temperature $T$, but is independent of $\rho_{2D}$. With increasing density $\rho_{2D}$ of QW excitons the quantum-statistical effects build up and the PL kinetics becomes density dependent.


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## APPENDIX A: EQUILIBRATION DYNAMICS OF QW EXCITONS

The equilibration kinetics of QW excitons is mainly due to particle-particle scattering and refers to the Boltzmann-Uhlenbeck equation

$$\frac{\partial}{\partial t}N_{\mathbf{k}_\parallel} = -\frac{2\pi}{\hbar}\left(\frac{U_0}{S}\right)^2 \sum_{\mathbf{p}_\parallel,\mathbf{q}_\parallel} [N_{\mathbf{k}_\parallel}N_{\mathbf{p}_\parallel}(1+N_{\mathbf{p}_\parallel-\mathbf{q}_\parallel})(1+N_{\mathbf{k}_\parallel+\mathbf{q}_\parallel}) - (N_{\mathbf{k}_\parallel}+1)(N_{\mathbf{p}_\parallel}+1)N_{\mathbf{p}_\parallel-\mathbf{q}_\parallel}N_{\mathbf{k}_\parallel+\mathbf{q}_\parallel}]\delta(E_{\mathbf{p}_\parallel-\mathbf{q}_\parallel}+E_{\mathbf{k}_\parallel+\mathbf{q}_\parallel}-E_{\mathbf{k}_\parallel}-E_{\mathbf{p}_\parallel}), \tag{A1}$$

where $U_0/S$ is the potential of exciton-exciton Coulombic interaction in a QW, $S$ is the area. For CQW's the potential is repulsive, and independent of the spin structure of interacting excitons owing to a rather small contribution of the exchange interaction. For a single QW the potential depends upon the spin state of excitons and can be repulsive (e.g., for the QW excitons with an identical spin structure) as well as attractive (e.g., for $\sigma^+$ and $\sigma^-$ spin-polarized excitons).[34] However, the repulsive potential strongly dominates[35] and determines the equilibration dynamics of QW excitons at $\rho_{2D} \geqslant \rho_{2D}^c$.

In order to evaluate a characteristic equilibration time $\tau_{x\text{-}x}$ we analyze within Eq. (A1) the dynamics of a small fluctuation $\delta N_0^{(0)} = \delta N_0(t=0) \ll N_{\mathbf{p}_\parallel=0}^{eq}$ of the ground-state mode population, provided that the occupation numbers of all other modes are given by the BE distribution of Eq. (6). The linearization of the right-hand side of Eq. (A1) with respect to $\delta N_0$ yields an exponential law for the decay of the fluctuation, i.e., $d(\delta N_0)/dt = -\delta N_0/\tau_{x\text{-}x}$ and $\delta N_0(t) = \delta N_0^{(0)}\exp(-t/\tau_{x\text{-}x})$. The decay time of the fluctuation, which we attribute to the characteristic equilibration time, is given by

$$\frac{1}{\tau_{x\text{-}x}} = \left(\frac{U_0 M_x}{2\pi}\right)^2\left(\frac{k_B T}{\hbar^5}\right)e^{-T_0/T}(1-e^{-T_0/T})\int_0^\infty du\int_0^{2\pi} d\phi \times \frac{e^{2u}}{[e^{u(1-\cos\phi)}+e^{-T_0/T}-1][e^{u(1+\cos\phi)}+e^{-T_0/T}-1][e^{2u}+e^{-T_0/T}-1]}. \tag{A2}$$

In order to derive Eq. (A2) from Eq. (A1) we put $\mathbf{k}_\parallel = 0$ and introduce the dummy variable $\mathbf{q}_\parallel' = \mathbf{q}_\parallel - \mathbf{p}_\parallel/2$. The dimensionless integration variable $u$ on the right-hand side of Eq. (A2) is given by $u = (\hbar p_\parallel)^2/(4k_B M_x T)$, and $\phi$ is the angle between $\mathbf{q}_\parallel'$ and $\mathbf{p}_\parallel$.

While the repulsive potential $U_0$ for QW excitons in an identical spin state can be explicitly written in terms of an integral convolution of the different pair Coulomb potentials between the constituent electrons and holes with the excitonic wave functions (see, e.g., Ref. 38), here we give a scaling estimate of $U_0$. The potential $U_0$ is determined by $U_0 = \int U_{x\text{-}x}(r_\parallel)d\mathbf{r}_\parallel$, where $U_{x\text{-}x}(r_\parallel)$ is the real-space potential of exciton-exciton interaction and $r_\parallel$ is the distance between two interacting QW excitons. The characteristic energy and length scales for $U_{x\text{-}x}(r_\parallel)$ are given by the two-dimensional Rydberg $E_x^{(2D)} = 2\mu_x e^4/\hbar^2$ and the corresponding excitonic Bohr radius $a_x^{(2D)} = \hbar^2/(2\mu_x e^2)$, where $\mu_x$ is the reduced mass of a QW exciton. For simple model potentials $U_{x\text{-}x}(r_\parallel) = CE_x^{(2D)}\exp(-r_\parallel/a_x^{(2D)})$ and $U_{x\text{-}x}(r_\parallel) = CE_x^{(2D)}(a_x^{(2D)}/r_\parallel)\exp(-r_\parallel/a_x^{(2D)})$, where the constant $C \simeq 1$, one estimates

$$U_0 = 2\pi C E_x^{(2D)}[a_x^{(2D)}]^2 = \pi C\frac{\hbar^2}{\mu_x}. \tag{A3}$$

A remarkable feature of Eq. (A3) is that while with decreasing confinement in the $z$ direction (the QW growth direction) $E_x^{(2D)}$ and $a_x^{(2D)}$ start to approach the corresponding bulk values, their combination $E_x^{(2D)}[a_x^{(2D)}]^2$ does not change. Such a behavior of $U_0$ is due to the quasi-two-dimensionality of QW excitons. The constant $C$, which is of the order of unity, depends on the design of a QW, the shape of the model potential $U_{x\text{-}x}(r_\parallel)$, etc. In further analysis we put $C = 1$.

For a classical gas of QW excitons, when $T \gg T_0$, one obtains from Eqs. (A2) and (A3),

$$\frac{1}{\tau_{x-x}}=\frac{\pi}{4\hbar}\left(\frac{M_x}{\mu_x}\right)^2 k_B T_0, \tag{A4}$$

where $T_0$ is given by Eq. (1). According to Eq. (A4), the scattering rate due to particle-particle interaction is proportional to the concentration $\rho_{2D}$ of QW excitons. Furthermore, as a signature of the quasi-two-dimensionality of excitons in QW's, the characteristic equilibration time $\tau_{x\text{-}x}$ is independent of the temperature $T$ (for $T \gg T_0$) and of the scattering length $\sim a_x^{(2D)}$.

For a quantum gas of QW excitons ($T \ll T_0$) we find, from Eqs. (A2) and (A3),

$$\frac{1}{\tau_{x-x}}=\frac{\pi}{\hbar}\left(1-\frac{\pi}{4}\right)\left(\frac{M_x}{\mu_x}\right)^2 k_B T e^{T_0/T}. \tag{A5}$$

The considerable increase of the scattering rate given by Eq. (A5) in comparison with that of Eq. (A4) is due to the low-energy QW excitons with $N_E^{eq} \gg 1$ at $E \leqslant k_B T$.

## APPENDIX B: HOMOGENEOUS BROADENING IN RADIATIVE DECAY OF QW EXCITONS

The joint density of states $J(p_\parallel)$ for the resonant optical decay of a QW exciton with $p_\parallel \leqslant k_0$ into a bulk photon is given by

$$J(p_\parallel)=\left(\frac{1}{2\pi^2}\right)\int_{-\infty}^{+\infty} dk_\perp \frac{\Gamma_{hom}}{\Gamma_{hom}^2+[\omega_x(p_\parallel)-\omega_\gamma(p_\parallel,k_\perp)]^2}, \tag{B1}$$

where $\hbar\omega_x(p_\parallel)=\hbar\omega_t+\hbar^2 p_\parallel^2/2M_x$ and $\omega_\gamma(p_\parallel,k_\perp)=(c/\sqrt{\epsilon_b})\sqrt{p_\parallel^2+k_\perp^2}$ are the dispersions of QW excitons and bulk photons, respectively, $k_\perp$ is the $z$ component of the wave vector of the bulk photons, which resonantly interact with a QW exciton with in-plane wavevector $p_\parallel$, and $\Gamma_{hom}$ is the homogeneous linewidth of QW excitons. If $\partial\omega_\gamma(p_\parallel,k_\perp)/\partial k_\perp \neq 0$ at $k_\perp$, determined by the equation $\omega_x(p_\parallel)=\omega_\gamma(p_\parallel,k_\perp)$, the integrand on the right-hand side of Eq. (B1) preserves its Lorentzian shape in wave-vector coordinates, and the joint density of states $J(p_\parallel)$ is independent of $\Gamma_{hom}$. This is a regular case, which does not hold for $p_\parallel \to k_0$ ($p_\parallel \leqslant k_0$). In this case the solution of $\omega_x(k_0)=\omega_\gamma(k_0,k_\perp)$ is $k_\perp=0$ and $\partial\omega_\gamma(p_\parallel=k_0,k_\perp)/\partial k_\perp=0$, indicating a 1D van Hove singularity in the joint density of states. As a result, $J(p_\parallel)$ becomes $\Gamma_{hom}$ dependent in a close vicinity of $p_\parallel=k_0$. This is the only way that the homogeneous linewidth influences the optical decay of quasiequilibrium excitons in ideal QW's.

With substitution $k_\perp=p_\parallel\tan\phi$, where $\phi \ni [-\pi/2,\pi/2]$, Eq. (B1) reduces to

$$J(p_\parallel)=\frac{1}{\pi^2}\left[\frac{p_\parallel}{\omega_x(p_\parallel)}\right]\int_0^{\pi/2} du \frac{\tilde{\gamma}}{(1/4)u^4-(\tilde{\delta}^2/2)u^2+(\tilde{\delta}^4/4+\delta^2\tilde{\gamma}^2)}, \tag{B2}$$

where $\tilde{\gamma}=\Gamma_{hom}/\omega_t$, $\delta=\cos\phi_0=p_\parallel/k_0=(cp_\parallel)/(\epsilon_b\omega_t)$, $\tilde{\delta}=\sin\phi_0=\sqrt{1-\delta^2}$, and the integration variable $u=\phi-\phi_0+\sin\phi_0$. Straightforward integration of Eq. (B2) yields the joint density of states

$$J(p_\parallel)=\frac{\sqrt{2}}{\pi}\left[\frac{p_\parallel}{\omega_x(p_\parallel)}\right]\frac{\tilde{\gamma}}{(4\delta^2\tilde{\gamma}^2+\tilde{\delta}^4)^{1/2}[(4\delta^2\tilde{\gamma}^2+\tilde{\delta}^4)^{1/2}-\tilde{\delta}^2]^{1/2}}. \tag{B3}$$

For $\tilde{\delta} \gg \sqrt{2\tilde{\gamma}}$, which is identical to the condition $k_0-p_\parallel \gg \tilde{\gamma}k_0$, Eq. (B3) reduces to the standard formula[16]

$$J(p_\parallel<k_0-\tilde{\gamma}k_0)=\frac{1}{\pi}\left(\frac{\epsilon_b}{c^2}\right)\frac{\omega_t}{\sqrt{k_0^2-p_\parallel^2}}. \tag{B4}$$

In the opposite limit $\tilde{\delta} \ll \sqrt{2\tilde{\gamma}}$, i.e., for $p_\parallel \to k_0$, Eq. (B3) yields

$$J(p_\parallel=k_0)=\frac{1}{2\pi}\frac{k_0}{(\Gamma_{hom}\omega_t)^{1/2}}. \tag{B5}$$

Equation (B5) shows how the homogeneous linewidth $\Gamma_{hom}$ removes the van Hove singularity at $p_\parallel=k_0$. For $\tilde{\delta} \leqslant \sqrt{2\tilde{\gamma}}$ the joint density of states is $\Gamma_{hom}$ dependent.

With Eq. (B3) we derive the final expression for the intrinsic radiative rate of $T$-polarized QW excitons in the presence of homogeneous broadening:

$$\Gamma_T(p_\parallel)=\frac{(\sqrt{2}k_0^2 p_\parallel\tilde{\gamma})\Gamma_0}{[(k_0^2-p_\parallel^2)^2+4\tilde{\gamma}^2k_0^2p_\parallel^2]^{1/2}\{[(k_0^2-p_\parallel^2)^2+4\tilde{\gamma}^2k_0^2p_\parallel^2]^{1/2}-(k_0^2-p_\parallel^2)\}^{1/2}}. \tag{B6}$$

For $k_0-p_\parallel \gg \tilde{\gamma}k_0$ Eq. (B4) is identical to Eq. (30). The homogeneous linewidth $\Gamma_{hom}$ changes Eq. (30) for $\Gamma_T(p_\parallel)$ only at the band of states given by $\tilde{\gamma}k_0 > k_0-p_\parallel \geqslant 0$ and, in particular, $\Gamma_T(p_\parallel=k_0)=\Gamma_0/(2\tilde{\gamma}^{1/2})$ rather than diverging. Note that the band of states is rather narrow because $\tilde{\gamma}\simeq 1/(\tau_{x\text{-}x}\omega_t)\sim 10^{-3}$. The 1D van Hove singularity at $p_\parallel=k_0$ is absent for the $L$-polarized QW excitons.

Homogeneous broadening practically does not change the optical decay rate of a gas of QW excitons. The use of Eq. (B6) for calculation of $\Gamma_{opt}$ by Eq. (31) shows that for Maxwell-Boltzmann-distributed QW excitons one obtains only a small renormalization of $J_T$ of Eq. (32), i.e., $J_T \rightarrow J_T(1-\tilde{\gamma}^{1/2})$. As a result, the optical decay rate of a classical gas of QW excitons changes from $\Gamma_{opt}$ to $\Gamma_{opt}[1-(3/4)\tilde{\gamma}^{1/2}]$. This correction due to the homogeneous linewidth is very small and can indeed be neglected. The corresponding correction for statistically degenerate QW excitons is even smaller because the low-energy excitons tend to populate the ground-state mode $\mathbf{p}_{\|}=0$ rather than vicinity of the energy states $E \simeq E_{k_0}$.


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