Exciton formation assisted by LO phonons in quantum wells

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Kinetics of exciton formation involving LO phonons is investigated in quantum wells. Considering the formation of an exciton from a free excited electron-hole pair due to LO-phonon emission, an expression is derived for the rate of formation of an exciton as a function of carrier densities, temperature, and wave vector $\mathbf{K}_\parallel$ of the center of mass of excitons in quantum wells, and the formation time of an exciton is also calculated. The theory is applied to GaAs quantum wells, in which it is found that the exciton formation dominantly occurs at $\mathbf{K}_\parallel \neq 0$.

I. INTRODUCTION

The dynamics of excitons in semiconductor nanostructures plays a very significant role in the ultrafast excitonic processes, which govern the basic performance characteristics of optoelectronic devices fabricated from these structures. Information on processes of exciton formation in such nanostructures is, therefore, very useful in studying their optoelectronic properties. When the energy of an incident photon on a nanostructure semiconductor is above its band-gap energy, the photon gets absorbed and a free electron-hole pair is excited. Such photogenerated electron-hole pairs can relax nonradiatively by emitting phonons and form excitons.[1] Excitons thus formed can also dissociate back into electron-hole pairs, form other excitonic complexes such as biexcitons and trions, or relax into other excitonic states through active participation of phonons. Such dynamical excitonic processes are very complicated to study, because they depend on many physical quantities, such as densities of excitons, electrons and holes, temperature, and interactions between charge carriers and charge carriers, charge carriers and excitons, excitons and excitons, excitons and phonons, and charge carriers and phonons. One of the most important interactions in the dynamics of excitons is the exciton-phonon interaction,[2] and its influence can be observed experimentally in the exciton linewidth and dephasing time,[3] time-resolved photoluminescence,[4] luminescence rise time,[5] mobility of excitons,[6] etc. It has been suggested[7,8] that at excitation energies larger than the band-gap energy, a photoexcited electron-hole pair can form an exciton first with a large total wave vector $\mathbf{K}_\parallel$, corresponding to its center-of-mass motion. The exciton then relaxes nonradiatively down to the $\mathbf{K}_\parallel \sim 0$ state by emitting phonons, and finally it recombines radiatively from the $\mathbf{K}_\parallel \sim 0$ excitonic state by emitting a photon to conserve energy and momentum. Therefore, in exciton luminescence experiments, the information on the formation time of an exciton as a function of exciton wave vector is crucial to study the luminescence rise time τ_R.

Experimentally, the first investigation of the dynamics of exciton formation was done by Damen *et al.*[7] in GaAs quantum wells. They found that the exciton formation time $\tau_f \leqslant 20$ ps after the creation of electron-hole pairs and suggested that excitons are formed in states with large wave vector (i.e., $\mathbf{K}_\parallel \neq 0$). After the work of Damen *et al.*, several groups have reported the values of formation time of excitons in GaAs quantum wells. Apparently, different approaches have been used to determine the formation time of excitons experimentally; some groups[5,7] have deduced it from the luminescence rise time, τ_R, and some[9,10] from the evolution of free-carrier luminescence using phenomenological rate equations. Strobel *et al.*[9] have reported $\tau_f = 14.4$ ps in a GaAs double-quantum-well structure. Blom *et al.*[5] have obtained LO-phonon-assisted exciton formation time $\tau_f = 1$ ps and acoustic-phonon-assisted $\tau_f = 12$ ps from their luminescence data. Deveaud *et al.*[11] have observed $\tau_f = 200$ ps from their time-resolved exciton luminescence experiments. Robart *et al.*[12] have obtained $\tau_f \lesssim 10$ ps, and Kumar *et al.*[10] obtained $\tau_f = 50$ ps. There have also been several values reported for the luminescence rise time τ_R,[13] e.g., $\tau_R \sim 400$ ps at an excitation density $< 5 \times 10^9$ cm^{-2}[7], $\tau_R = 25$–40 ps at a quantum-well width, $L_z = 26$ Å,[5] $\tau_R \sim 70$–160 ps,[4] and $\tau_R \sim 100$–350 ps,[14] depending on the carrier densities.

Theoretically, Thilagam and Singh[15] have calculated $\tau_f > 100$ ps for $L_z = 25$–200 Å by considering that excitons are formed by emitting an acoustic phonon via deformation-potential coupling. Selbmann *et al.*[16] have obtained $\tau_R = 50$–150 ps in bulk GaAs using an ensemble Monte Carlo approach. By solving the Boltzmann equation for an 80 Å GaAs quantum well, Zhang *et al.*[17] have obtained $\tau_f = 108$ ps (electron-hole density $n_{eh} = 4 \times 10^{10}$ cm^{-2} and electron-hole plasma temperature $T_{eh} = 50$ K), 113 ps ($n_{eh} = 4 \times 10^{10}$ cm^{-2} and $T_{eh} = 80$ K), and 192 ps ($n_{eh} = 2 \times 10^{10}$ cm^{-2} and $T_{eh} = 50$ K). Gulia *et al.*[18] have studied the exciton formation and relaxation in GaAs quantum wells using a Monte Carlo simulation of the coupled free carrier and exciton. They have obtained $\tau_R = 80$ ps for $L_z = 26$ Å and 120 ps for $L_z = 80$ Å. By analyzing their results of the formation coefficient,[18,19] they have shown that the exciton formation processes in quantum wells at low carrier densities are dominated by the LO-phonon emission during the first

few ps after the photoexcitation. Recently, an attempt at calculating the formation time of an exciton as a function of exciton wave vector has been made using the Fermi golden rule and fractional dimensional wave function.[20] However, a comprehensive theoretical development to study the dependence of the exciton formation time on the wave vector of the center of mass of the exciton has not yet been done.

In this paper, we have derived an expression for the rate of formation of an exciton as a function of the wave vector of its center of mass. The theory is then applied to calculate the formation time of an exciton in GaAs quantum wells. Here we have focused only on the exciton formation processes, without considering the exciton relaxation processes, which are published elsewhere.[21,22] We have found that the formation process of an exciton in GaAs quantum wells is dominant at a nonzero value of $\mathbf{K}_\parallel$ depending on charge-carrier density and temperature and also that the behavior of the formation of an exciton is very sensitive to the width of quantum wells. Our results demonstrate the square-law dependence of the formation rate of an exciton on the excitation density $n_{e\text{-}h}$, which agrees very well with the observed square-law dependence of photoluminescence on the excitation density. This work is expected to be very useful in understanding the exciton formation processes, and in analyzing the experimental data of exciton luminescence in quantum wells.

This paper is organized as follows. In Sec. II, we have developed a theory to calculate the rate of formation of an exciton as a function of carrier densities, temperature, and wave vector $\mathbf{K}_\parallel$ in quantum wells. In Sec. III, the numerical results of the formation rate and time in GaAs quantum wells are presented and discussed.

II. THEORY OF FORMATION OF EXCITONS

In this section, we develop a theory of the exciton–LO-phonon interaction in quantum-well structures, suitable for studying the process of exciton formation. We assume that charge carriers are confined in quantum wells. The process of formation of excitons is considered as a quantum transition from an initial state of a photoexcited free electron-hole pair to a final state of an exciton formed due to electron-hole and charge-carrier–LO-phonon interactions.

The Hamiltonian of a pair of electrons and holes or/and an exciton interacting with LO phonons can be written by adding electron–LO-phonon and hole–LO-phonon interaction Hamiltonians as[21]

$$H_I(\mathbf{r}_e,\mathbf{r}_h)=\sum_{\mathbf{q}} C[(e^{i\mathbf{q}\cdot\mathbf{r}_e}-e^{i\mathbf{q}\cdot\mathbf{r}_h})\hat{b}_{\mathbf{q}}+\text{c.c.}], \tag{1}$$

where $\hat{b}_{\mathbf{q}}$ is the annihilation operator of a phonon with wave vector $\mathbf{q}$, $\mathbf{r}_e$ [$\mathbf{r}_h$] is the position coordinate of the electron [hole], and

$$C=ie\sqrt{\frac{\hbar\omega_{\mathrm{LO}}}{2\epsilon_0 V}\left(\frac{1}{\kappa_\infty}-\frac{1}{\kappa_0}\right)}\frac{1}{q}. \tag{2}$$

Here, ω_{LO} is the frequency of the LO phonon, V is the volume of the crystal, ϵ_0 is the permittivity of free space, and κ_0 is the low-frequency and κ_∞ the high-frequency relative dielectric constants. The motion of phonons is considered to be in 3D space, whereas that of excitons is in quasi-2D space.

Assuming the plane of the quantum well as the xy plane and introducing the center of mass and relative coordinates of an exciton in the plane of quantum wells as

$$\mathbf{R}_\parallel=\alpha_e\mathbf{r}_{e\parallel}+\alpha_h\mathbf{r}_{h\parallel}, \quad \mathbf{r}_\parallel=\mathbf{r}_{e\parallel}-\mathbf{r}_{h\parallel}, \tag{3}$$

with

$$\alpha_e=\frac{m^*_{e\parallel}}{M^*_\parallel}, \quad \alpha_h=\frac{m^*_{h\parallel}}{M^*_\parallel}, \quad M^*_\parallel=m^*_{e\parallel}+m^*_{h\parallel}, \tag{4}$$

the interaction Hamiltonian in Eq. (1) can be expressed as

$$H_I(\mathbf{r}_\parallel,\mathbf{R}_\parallel,z_e,z_h)=\sum_{\mathbf{q}_\parallel q_z} C[e^{i\mathbf{q}_\parallel\cdot\mathbf{R}_\parallel}(e^{i\alpha_h\mathbf{q}_\parallel\cdot\mathbf{r}_\parallel}e^{iq_z z_e} - e^{-i\alpha_e\mathbf{q}_\parallel\cdot\mathbf{r}_\parallel}e^{iq_z z_h})\hat{b}_{\mathbf{q}}+\text{c.c.}], \tag{5}$$

where z_e and z_h are the coordinates of the electron and hole, respectively, in the z direction perpendicular to the plane of the quantum well.

We assume that in the initial state there is an electron and a heavy hole occupying their lowest subband in the quantum well, and then in the final state after the transition they form a heavy-hole exciton in the lowest exciton band. The field operator $\hat{\psi}^\dagger_{e\text{-}h}(\mathbf{R}_\parallel,\mathbf{r}_\parallel,z_e,z_h)$ describing the simultaneous creation of a pair of electrons and holes at $\mathbf{r}_e$ and $\mathbf{r}_h$, respectively, in the quantum well can be written as

$$\hat{\psi}^\dagger_{e\text{-}h}(\mathbf{R}_\parallel,\mathbf{r}_\parallel,z_e,z_h)=\frac{1}{A_0}\sum_{\mathbf{K}_\parallel\mathbf{k}_\parallel} e^{-i\mathbf{K}_\parallel\cdot\mathbf{R}_\parallel}e^{-i\mathbf{k}_\parallel\cdot\mathbf{r}_\parallel}\phi^*_e(z_e) \times\phi^*_h(z_h)\hat{d}^\dagger_{h,\alpha_h\mathbf{K}_\parallel-\mathbf{k}_\parallel}\hat{a}^\dagger_{c,\alpha_e\mathbf{K}_\parallel+\mathbf{k}_\parallel}, \tag{6}$$

where A_0 is the 2D area of quantum wells and $\hat{d}^\dagger_{h,\alpha_h\mathbf{K}_\parallel-\mathbf{k}_\parallel}$ and $\hat{a}^\dagger_{c,\alpha_e\mathbf{K}_\parallel+\mathbf{k}_\parallel}$ are hole and electron creation operators in the valence and conduction bands, respectively. The wave vector of the center-of-mass motion $\mathbf{K}_\parallel$ and the wave vector of the relative motion $\mathbf{k}_\parallel$ are defined in terms of electron and hole wave vectors $\mathbf{k}_{e\parallel}$ and $\mathbf{k}_{h\parallel}$ in the plane of the quantum well as

$$\mathbf{K}_\parallel=\mathbf{k}_{e\parallel}+\mathbf{k}_{h\parallel} \quad \text{and} \quad \mathbf{k}_\parallel=\alpha_h\mathbf{k}_{e\parallel}-\alpha_e\mathbf{k}_{h\parallel}.$$

$\phi_e(z_e)$ and $\phi_h(z_h)$ in Eq. (6) are, respectively, the electron and hole wave functions of their lowest-energy states of their motions along the z axis perpendicular to the wall of quantum wells, given by[23]

$$\phi_j(z_j)=\begin{cases} A_j\cos k_j z_j & \text{for} \quad |z_j|<\dfrac{L_z}{2} \\ B_j e^{-\beta_j(|z_j|-L_z/2)} & \text{for} \quad |z_j|>\dfrac{L_z}{2}, \end{cases} \quad j=e,h, \tag{7}$$

where A_j and B_j are constants to be determined from the boundary and normalization conditions and L_z is the quantum-well width. In the initial state, the excited electron and hole are regarded to be a noninteracting pair (free electron-hole pair).

By introducing the creation operator $\hat{B}^{\dagger}_{\mathbf{K}_\parallel}$ of an exciton with center-of-mass momentum wave vector $\mathbf{K}_\parallel$ in the plane of the quantum well and with its wave function $\psi_{\rm ex}=(1/\sqrt{A_0})e^{i\mathbf{K}_\parallel\cdot\mathbf{R}_\parallel}\phi_x(\mathbf{r}_\parallel)\phi_e(z_e)\phi_h(z_h)$, the field operator $\hat{\psi}^{\dagger}_{\rm ex}$ describing the creation of an exciton at $\mathbf{R}_\parallel,\mathbf{r}_\parallel,z_e,z_h$ can be written as

$$\hat{\psi}^{\dagger}_{\rm ex}(\mathbf{R}_\parallel,\mathbf{r}_\parallel,z_e,z_h)=\frac{1}{\sqrt{A_0}}\sum_{\mathbf{K}_\parallel} e^{-i\mathbf{K}_\parallel\cdot\mathbf{R}_\parallel}\phi^{*}_x(\mathbf{r}_\parallel) \times\phi^{*}_e(z_e)\phi^{*}_h(z_h)\hat{B}^{\dagger}_{\mathbf{K}_\parallel}, \tag{8}$$

where $\phi_x(\mathbf{r}_\parallel)$, the $1s$ state exciton wave function in the quantum-well plane, is chosen to be a trial wave function given by[23]

$$\phi_x(\mathbf{r}_\parallel)=\sqrt{\frac{2\beta^2}{\pi}}e^{-\beta r_\parallel}, \tag{9}$$

with β being the variational parameter.

Using Eqs. (5), (6), and (8), the interaction Hamiltonian for the formation of an exciton from a free electron-hole pair due to emission or absorption of a single phonon can be written in the second quantized form as

$$\begin{aligned}\hat{H}^{eh\rightarrow\rm ex}_I&=\int d\mathbf{R}_\parallel\int d\mathbf{r}_\parallel\int dz_e\int dz_h\hat{\psi}^{\dagger}_{\rm ex}H_I\hat{\psi}_{e\text{-}h}\\&=\frac{1}{\sqrt{A_0}}\sum_{\mathbf{K}_\parallel\mathbf{k}_\parallel\mathbf{q}_\parallel q_z}C[F^{0}_{-}(\mathbf{q}_\parallel,q_z,\mathbf{k}_\parallel)\hat{B}^{\dagger}_{\mathbf{K}_\parallel}\hat{b}_{\mathbf{q}}\\&\quad\times\hat{d}_{h,\alpha_h(\mathbf{K}_\parallel-\mathbf{q}_\parallel)-\mathbf{k}_\parallel}\hat{a}_{c,\alpha_e(\mathbf{K}_\parallel-\mathbf{q}_\parallel)+\mathbf{k}_\parallel}\\&\quad+F^{0}_{+}(\mathbf{q}_\parallel,q_z,\mathbf{k}_\parallel)\hat{B}^{\dagger}_{\mathbf{K}_\parallel}\hat{b}^{\dagger}_{\mathbf{q}}\hat{d}_{h,\alpha_h(\mathbf{K}_\parallel+\mathbf{q}_\parallel)-\mathbf{k}_\parallel}\\&\quad\times\hat{a}_{c,\alpha_e(\mathbf{K}_\parallel+\mathbf{q}_\parallel)+\mathbf{k}_\parallel}],\end{aligned} \tag{10}$$

where $\hat{\psi}_{e\text{-}h}$ is the conjugate operator of Eq. (6), and

$$F^{0}_{\mp}(\mathbf{q}_\parallel,q_z,\mathbf{k}_\parallel)=[F_e(\pm q_z)G_1(\mathbf{k}_\parallel\pm\alpha_h\mathbf{q}_\parallel) -F_h(\pm q_z)G_1(\mathbf{k}_\parallel\mp\alpha_e\mathbf{q}_\parallel)]. \tag{11}$$

Here $F_j(q_z)$ $(j=e,h)$ is a form factor obtained as

$$F_j(q_z)=\int dz_j|\phi_j(z_j)|^2e^{iq_zz_j},\quad j=e,h, \tag{12}$$

and G_1 is another form factor, obtained using the variational wave function in Eq. (9), as

$$G_1(\mathbf{k}_\parallel+\alpha\mathbf{q}_\parallel)=\int d\mathbf{r}_\parallel\phi^{*}_{\rm ex}(\mathbf{r}_\parallel)e^{i(\mathbf{k}_\parallel+\alpha\mathbf{q}_\parallel)\cdot\mathbf{r}_\parallel} =\sqrt{\frac{8\pi}{\beta^2}}\left[\left(\frac{|\mathbf{k}_\parallel+\alpha\mathbf{q}_\parallel|}{\beta}\right)^2+1\right]^{-3/2}. \tag{13}$$

The first term of Eq. (10) corresponds to the formation of an exciton due to an LO-phonon absorption, and the second term corresponds to that due to an LO phonon emission.

Here we consider a case where free electrons and holes, excitons, and phonons are initially assumed to occupy their states with occupation numbers $f^{e}_{\mathbf{k}_e}$, $f^{h}_{\mathbf{k}_h}$, $f^{\rm ex}_{\mathbf{k}_{\rm ex}}$, and $n_{\mathbf{q}}$ with their wave vectors $\mathbf{k}_e$, $\mathbf{k}_h$, $\mathbf{k}_{\rm ex}$, and $\mathbf{q}$, respectively. A transition is considered to take place from the initial state to the final state in which one free electron-hole pair changes into an exciton due to Coulomb and phonon interactions. That means in the final state the occupation numbers of states of all four, free electrons and holes and excitons and phonons, change by 1.

Using the Fermi golden rule and the interaction operator in Eq. (10), the rate of exciton formation from a free electron-hole pair involving a single phonon with wave vector $\mathbf{q}$ is obtained as

$$\begin{aligned}&W(\mathbf{K}_\parallel\pm\mathbf{q}_\parallel,q_z,\mathbf{k}_\parallel\rightarrow\mathbf{K}_\parallel)\\&\quad=\frac{1}{A_0}\frac{2\pi}{\hbar}|CF^{0}_{\pm}(\mathbf{q}_\parallel,q_z,\mathbf{k}_\parallel)|^2f^{h}_{\alpha_h(\mathbf{K}_\parallel\pm\mathbf{q}_\parallel)-\mathbf{k}_\parallel}\\&\qquad\times f^{e}_{\alpha_e(\mathbf{K}_\parallel\pm\mathbf{q}_\parallel)+\mathbf{k}_\parallel}(f^{\rm ex}_{\mathbf{K}_\parallel}+1)\\&\qquad\times\left(n_{\mathbf{q}}+\frac{1}{2}\pm\frac{1}{2}\right)\delta(E_x-E_{e\text{-}h}\pm\hbar\omega_{\rm LO}),\end{aligned} \tag{14}$$

where the $+$ sign represents emission of phonon and the $-$ represents absorption, and

$$E_x=\frac{\hbar^2|\mathbf{K}_\parallel|^2}{2M^{*}_\parallel}-E_b \tag{15}$$

and

$$E_{e\text{-}h}=\frac{\hbar^2|\mathbf{K}_\parallel\pm\mathbf{q}_\parallel|^2}{2M^{*}_\parallel}+\frac{\hbar^2|\mathbf{k}_\parallel|^2}{2\mu^{*}}, \tag{16}$$

where E_b is the exciton binding energy and $\mu^{*}_\parallel$ $(1/\mu^{*}_\parallel=1/m^{*}_{e\parallel}+1/m^{*}_{h\parallel})$ the reduced mass of the electron-hole pair.

As the time of formation of an exciton is relatively longer than that of the scattering of excitons, electrons, and holes,[24] it can be assumed that electrons and holes are in the quasithermal equilibrium before and after the transition. With this assumption, we can replace the occupation numbers $f^{h}_{\alpha_h(\mathbf{K}_\parallel\pm\mathbf{q}_\parallel)-\mathbf{k}_\parallel}$ and $f^{e}_{\alpha_e(\mathbf{K}_\parallel\pm\mathbf{q}_\parallel)+\mathbf{k}_\parallel}$ of hole and electron states, respectively, by their thermal average values obtained from the Fermi-Dirac distribution as[25]

$$f^{j}_{\mathbf{k}_j}=[e^{[E_j(\mathbf{k}_j)-\mu_j]/k_BT_j}+1]^{-1},$$

$$\mu_j=k_BT_j\ln(e^{\hbar^2\pi n_j/m^{*}_{j\parallel}k_BT_j}-1),\quad j=e,h, \tag{17}$$

where μ_j, T_j, and n_j denote the chemical potential, temperature, and 2D density of electrons and holes, respectively. Also assuming that excitons and phonons are in thermal equilibrium as bosons before and after the transition, the occupation number, $n_{\mathbf{q}}$, of phonon and $f^{\rm ex}_{\mathbf{K}_\parallel}$ of exciton states can be replaced by their average values obtained from the Bose-Einstein distribution as

$$n_{\mathbf{q}}=[e^{\hbar\omega_{\rm LO}/k_BT}-1]^{-1},\quad f^{\rm ex}_{\mathbf{K}_\parallel}=[e^{[E(\mathbf{K}_\parallel)-\mu_{\rm ex}]/k_BT_{\rm ex}}-1]^{-1}, \tag{18}$$

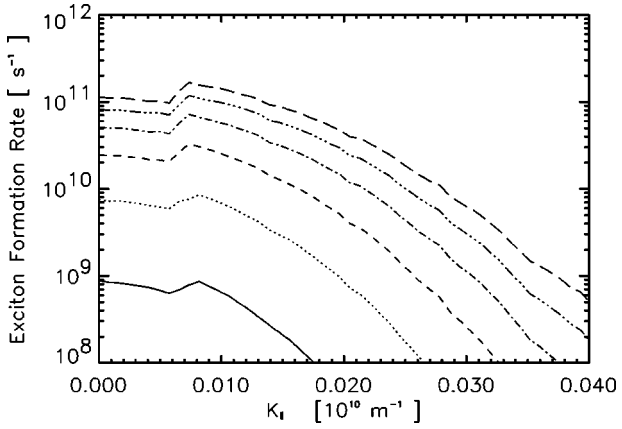

FIG. 1. The formation rate of an exciton in GaAs quantum wells as a function of the center-of-mass wave vector $\mathbf{K}_\parallel$ for $L_z = 80$ Å, $n_{e\text{-}h} = 1\times10^{10}$ cm^{-2}, and different temperatures, i.e., $T_{e\text{-}h} = 30$ K (solid curve), $T_{e\text{-}h} = 40$ K (dotted curve), $T_{e\text{-}h} = 50$ K (dashed curve), $T_{e\text{-}h} = 60$ K (dash-dot curve), $T_{e\text{-}h} = 70$ K (dash-dot-dot-dot curve), and $T_{e\text{-}h} = 80$ K (long dashed curve).

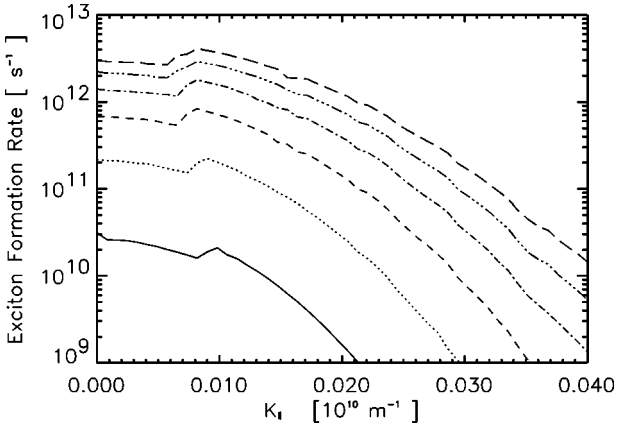

FIG. 2. The formation rate of an exciton in GaAs quantum wells as a function of the center-of-mass wave vector $\mathbf{K}_\parallel$ for $L_z = 80$ Å, $n_{e\text{-}h} = 5\times10^{10}$ cm^{-2}, and different temperatures, i.e., $T_{e\text{-}h} = 30$ K (solid curve), $T_{e\text{-}h} = 40$ K (dotted curve), $T_{e\text{-}h} = 50$ K (dashed curve), $T_{e\text{-}h} = 60$ K (dash-dot curve), $T_{e\text{-}h} = 70$ K (dash-dot-dot-dot curve), and $T_{e\text{-}h} = 80$ K (long dashed curve).

where $\mu_{\rm ex}$ is the chemical potential of 2D excitons, and T and $T_{\rm ex}$ correspond to lattice and exciton temperatures, respectively. The chemical potential $\mu_{\rm ex}$ can be expressed in terms of exciton temperature $T_{\rm ex}$ and 2D exciton density $n_{\rm ex}$ as[26]

$$\mu_{\rm ex} = k_B T_{\rm ex} \ln[1 - e^{-2\pi n_{\rm ex}\hbar^2/(gM_\parallel^* k_B T_{\rm ex})}], \tag{19}$$

where g is the spin degeneracy factor of 2D excitons.

Finally, the total rate of formation of an exciton with wave vector $\mathbf{K}_\parallel$ is obtained by summing the rate in Eq. (14) over all $\mathbf{k}_\parallel$, q_z, and $\mathbf{q}_\parallel$. The total rate $W_+(\mathbf{K}_\parallel)$ thus obtained gives the inverse of the time of formation, $\tau_f(\mathbf{K}_\parallel)$, of an exciton with wave vector $\mathbf{K}_\parallel$ as

$$\frac{1}{\tau_f(\mathbf{K}_\parallel)} = W_+(\mathbf{K}_\parallel) = \sum_{\mathbf{q}_\parallel, q_z, \mathbf{k}_\parallel} W(\mathbf{K}_\parallel + \mathbf{q}_\parallel, q_z, \mathbf{k}_\parallel \rightarrow \mathbf{K}_\parallel). \tag{20}$$

III. RESULTS AND DISCUSSION

We have calculated the formation rate of excitons in the [001] GaAs/$Al_{0.3}Ga_{0.7}As$ quantum wells by using the material parameter as $m_{e\parallel}^* = m_{e\perp}^* = 0.0665 m_e$ ($m_e = 9.109\,53 \times 10^{-31}$ kg), $m_{h\parallel}^* = 0.11 m_e$, $m_{h\perp}^* = 0.33 m_e$, $\kappa_0 = 12.9$, $\kappa_\infty = 10.9$, $\hbar\omega_{\rm LO} = 36.2$ meV,[27] and $T = 4.2$ K. For simplicity, we set the exciton wave vector in the direction of the x axis, i.e., $\mathbf{K}_\parallel = K_\parallel \hat{\mathbf{x}}$, which is not expected to cause any loss of generality, because the exciton dispersion relation is considered to be parabolic and isotropic in the plane (xy plane) of quantum wells. We assume that there are no excitons in the system in the initial state, i.e., $f_{\mathbf{K}_\parallel}^{\rm ex} = 0$. The densities of the photogenerated electron and hole can be considered to be the same ($n_e = n_h = n_{e\text{-}h}$) in an intrinsic quantum well. We also assume that electrons and holes are at the same temperature, i.e., $T_e = T_h = T_{e\text{-}h}$.

In Fig. 1 are plotted the calculated values for the formation rate of an exciton as a function of exciton wave vector $\mathbf{K}_\parallel$ for different values of charge-carrier temperatures in quantum wells with a well width of $L_z = 80$ Å and a charge-carrier density of $n_{e\text{-}h} = 1\times10^{10}$ cm^{-2}. For charge-carrier temperature $T_{e\text{-}h} \lesssim 30$ K, the formation of an exciton occurs dominantly at $\mathbf{K}_\parallel = 0$, but for $T_{e\text{-}h} \gtrsim 30$ K it occurs at nonzero $\mathbf{K}_\parallel \simeq 0.0082\times10^{10}$ m^{-1}. Our results indicate that first the formation rate of an exciton decreases with increasing $\mathbf{K}_\parallel$, and then it increases to a peak value at about $\mathbf{K}_\parallel \simeq 0.0082\times10^{10}$ m^{-1}, after which it decreases continuously as the exciton wave vector $\mathbf{K}_\parallel$ increases (see Fig. 1). The results of Fig. 1 are in agreement with those of Damen *et al.*[7] that excitons are formed dominantly at $\mathbf{K}_\parallel \neq 0$, but our results also suggest that the formation process depends on $T_{e\text{-}h}$ and $n_{e\text{-}h}$. For instance, according to Fig. 1, an exciton can be dominantly formed at $\mathbf{K}_\parallel = 0$ at low carrier temperatures $T_{e\text{-}h} \lesssim 30$ K for a carrier density of $n_{e\text{-}h} = 1\times10^{10}$ cm^{-2}. However, at higher carrier temperatures, the situation can be different. For example, for a fixed carrier density of $n_{e\text{-}h} = 1\times10^{10}$ cm^{-2}, by raising the temperature to $T_{e\text{-}h} = 40$ K, we find that the formation time $\tau_f = 133$ ps at $\mathbf{K}_\parallel = 0$, which is slightly slower than $\tau_f = 118$ ps found at $\mathbf{K}_\parallel = 0.0082\times10^{10}$ m^{-1}. Likewise, if the temperature is raised further to $T_{e\text{-}h} = 50$ K, it is found again that $\tau_f = 40$ ps at $\mathbf{K}_\parallel = 0$ higher than $\tau_f = 30$ ps at $\mathbf{K}_\parallel = 0.0074\times10^{10}$ m^{-1}. This trend seems to continue even at higher temperatures as well (see Fig. 1).

We have plotted in Fig. 2 the calculated formation rate of an exciton as a function of exciton wave vector $\mathbf{K}_\parallel$ at six different carrier temperatures, from 30 K to 80 K, at a charge-carrier density of $n_{e\text{-}h} = 5\times10^{10}$ cm^{-2} in a GaAs quantum-well width $L_z = 80$ Å. Although the dependence of the formation rate of an exciton on the center-of-mass wave vector for $n_{e\text{-}h} = 5\times10^{10}$ cm^{-2} is similar to that for $n_{e\text{-}h} = 1\times10^{10}$ cm^{-2}, as it is clear from Figs. 1 and 2, the formation rates for $n_{e\text{-}h} = 5\times10^{10}$ cm^{-2} are relatively larger than those for $n_{e\text{-}h} = 1\times10^{10}$ cm^{-2}.

Figure 3 shows the formation rate of an exciton as a function of exciton wave vector $\mathbf{K}_\parallel$ at $T_{e\text{-}h} = 60$ K for three different well widths, 80, 150, and 250 Å, and two different charge-carrier densities. For all three quantum wells, the maximum formation rate occurs at nonzero $\mathbf{K}_\parallel$. This is ob-

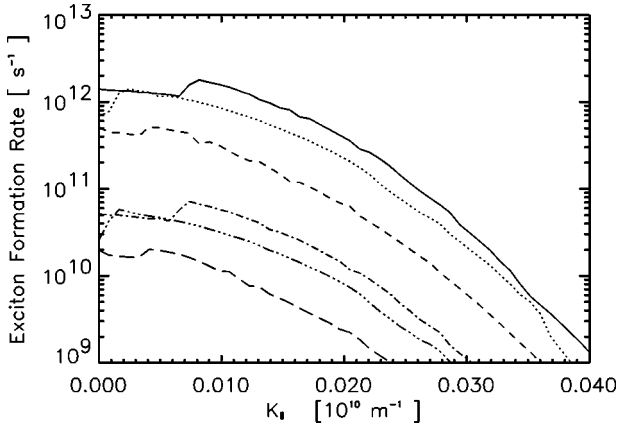

FIG. 3. The formation rate of an exciton in GaAs quantum wells as a function of the center-of-mass wave vector $\mathbf{K}_\parallel$ at $T_{e\text{-}h}=60$ K for different well widths and charge-carrier densities, $L_z=80$ Å, $n_{e\text{-}h}=5\times10^{10}$ cm^{-2} (solid curve), $L_z=150$ Å, $n_{e\text{-}h}=5\times10^{10}$ cm^{-2} (dotted curve), $L_z=250$ Å, $n_{e\text{-}h}=5\times10^{10}$ cm^{-2} (dashed curve), $L_z=80$ Å, $n_{e\text{-}h}=1\times10^{10}$ cm^{-2} (dash-dot curve), $L_z=150$ Å, $n_{e\text{-}h}=1\times10^{10}$ cm^{-2} (dash-dot-dot-dot curve), and $L_z=250$ Å, $n_{e\text{-}h}=1\times10^{10}$ cm^{-2} (long dashed curve).

vious from Fig. 3, which shows that for quantum-well widths 80 and 150 Å the maximum formation rate occurs at $\mathbf{K}_\parallel=0.0074\times10^{10}$ m^{-1} and 0.0016×10^{10} m^{-1}, at the carrier density of $n_{e\text{-}h}=1\times10^{10}$ cm^{-2}, which change to 0.0081×10^{10} m^{-1} and 0.0025×10^{10} m^{-1}, respectively, at the carrier density of 5×10^{10} cm^{-2}. However, for the well of $L_z=250$ Å, the maximum rate occurs at $\mathbf{K}_\parallel=0.0041\times10^{10}$ m^{-1}, for both the carrier densities. For quantum wells of widths $L_z=80$ Å and $L_z=250$ Å, the formation rates first decrease slightly, then increase to a maximum, and then decrease again continuously as $\mathbf{K}_\parallel$ increases. However, the formation rate for $L_z=150$ Å quantum wells first increases up to its maximum and then decreases continuously with increasing $\mathbf{K}_\parallel$. The formation time corresponding to the maximum formation rate in the $L_z=150$ Å quantum well is obtained as $\tau_f=17$ ps and 0.7 ps at the carrier density of $n_{e\text{-}h}=1\times10^{10}$ cm^{-2} and 5×10^{10} cm^{-2}, respectively. Such a trend that the formation time corresponding to the maximum rate becomes shorter with the increase in the carrier density is obtained in all three well widths.

In Fig. 4, we have plotted the rate of formation of an exciton as a function of charge-carrier density $n_{e\text{-}h}$, by varying it from 1×10^8 to 5×10^{10} cm^{-2} at $T_{e\text{-}h}=60$ K for the three well widths. From the calculated results of the formation rate, we have carried out curve fittings using the relation $W(n_{eh})=bn_{e\text{-}h}^x$, where b and x are fitting parameters. Our results give $x\approx2$ as shown in Fig. 4. In other words, our theory shows a square-law dependence of the formation rate of an exciton on $n_{e\text{-}h}$. This provides a theoretical confirmation of the observed experimental square-law dependence of the photoluminescence on excitation density obtained by Strobel *et al.*[9]

We have also calculated the formation rate of light-hole excitons as a function of $\mathbf{K}_\parallel$ using the effective masses as $m_{\mathrm{lh}\parallel}^*=0.20m_e$ and $m_{\mathrm{lh}\perp}^*=0.09m_e$ (Ref. 27) for quantum wells of width $L_z=80$ Å. The formation rate thus obtained for a light-hole exciton is comparable with that of a heavy-hole exciton near the zero center-of-mass wave vector. For instance, $n_{e\text{-}h}=1\times10^{10}$ cm^{-2} and $T_{e\text{-}h}=50$ K, the formation rate of a light-hole exciton is slightly larger than that of a heavy-hole exciton for $\mathbf{K}_\parallel$ in the range $0\leqslant\mathbf{K}_\parallel\lesssim0.0063\times10^{10}$ m^{-1} and $\mathbf{K}_\parallel\gtrsim0.0142\times10^{10}$ m^{-1}. However, the reverse of this apparently occurs in the range 0.0063×10^{10} m$^{-1}\lesssim\mathbf{K}_\parallel\lesssim0.0142\times10^{10}$ m^{-1}, and the latter becomes larger than the former. The maximum rate for a light-hole exciton is 2.7×10^{10} s^{-1} at $\mathbf{K}_\parallel=0.0010\times10^{10}$ m^{-1} and that for a heavy-hole exciton 3.3×10^{10} s^{-1} at $\mathbf{K}_\parallel=0.0074\times10^{10}$ m^{-1}. The corresponding formation times are 37 ps for the light-hole exciton and 30 ps for the heavy-hole exciton. On the other hand, at $\mathbf{K}_\parallel=0$ the formation rate and time of the light-hole exciton are 2.6×10^{10} s^{-1} and 38 ps, and those for the heavy-hole exciton are 2.5×10^{10} s^{-1} and 40 ps, respectively. Therefore, in this case, we conclude that the formation of a light-hole exciton is dominant near zero values of $\mathbf{K}_\parallel$ whereas that of a heavy-hole exciton is dominant at larger values of $\mathbf{K}_\parallel$.

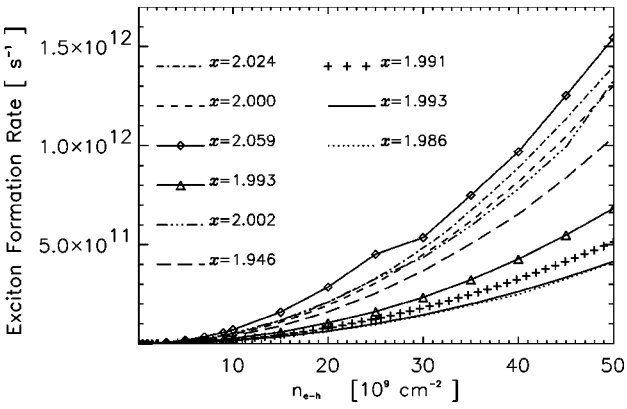

FIG. 4. The formation rate of an exciton in GaAs quantum wells as a function of the carrier density $n_{e\text{-}h}$ at $T_{e\text{-}h}=60$ K for different well widths and center-of-mass wave vectors, $L_z=80$ Å, $\mathbf{K}_\parallel=0$ (dash-dot curve), $L_z=150$ Å, $\mathbf{K}_\parallel=0$ (solid curve with triangles), $L_z=250$ Å, $\mathbf{K}_\parallel=0$ (++++ curve), $L_z=80$ Å, $\mathbf{K}_\parallel=0.003\times10^{10}$ m^{-1} (dashed curve), $L_z=150$ Å, $\mathbf{K}_\parallel=0.003\times10^{10}$ m^{-1} (dash-dot-dot-dot curve), $L_z=250$ Å, $\mathbf{K}_\parallel=0.003\times10^{10}$ m^{-1} (solid curve), $L_z=80$ Å, $\mathbf{K}_\parallel=0.0075\times10^{10}$ m^{-1} (solid curve with diamonds), $L_z=150$ Å, $\mathbf{K}_\parallel=0.0075\times10^{10}$ m^{-1} (long dashed curve), and $L_z=250$ Å, $\mathbf{K}_\parallel=0.0075\times10^{10}$ m^{-1} (dotted curve). The numbers x in the figure are obtained from the curve fitting $W(n_{e\text{-}n})=bn_{e\text{-}h}^x$.

In summary, we have presented the theory of formation processes of an exciton due to the LO-phonon interaction as a function of exciton wave vector $\mathbf{K}_\parallel$, charge-carrier temperature $T_{e\text{-}h}$, and charge-carrier density $n_{e\text{-}h}$ in quantum wells. The theory developed is applied to GaAs quantum wells. We have found that the formation of an exciton occurs dominantly at a nonzero $\mathbf{K}_\parallel$, and it depends on the charge-carrier density and temperature. It is also found that the behavior of the formation of an exciton depends significantly on the width of the quantum wells. Our results also illustrate very clearly the square-law dependence of the formation rate of an exciton on $n_{e\text{-}h}$ in agreement with the square-law dependence of photoluminescence on the excitation density.

ACKNOWLEDGMENTS

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