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Calculations of the time taken for excitons to form in GaAs quantum wells

M H Zhang, Q Huang and J M Zhou

Institute of Physics, Chinese Academy of Sciences, Beijing 100080, People's Republic of China

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Abstract. The time taken for excitons to form has been calculated by solving the Boltzmann equation for an 80 Å GaAs quantum well. Our results show that longitudinal acoustic phonon emission is the most important relaxation mechanism for exciton formation from free-electron–hole pairs at low lattice temperatures. The time taken for excitons to form is found to depend on the excitation conditions: the excitation density, the electron–hole plasma temperature and the lattice temperature, and is approximately proportional to $1+k_BT_{eh}/E_b$ (where T_{eh} is the electron–hole plasma temperature and E_b is the exciton binding energy) at a fixed lattice temperature. For the excitation conditions used by Kumar *et al* (Kumar R, Vengurlekar A S, Prabhu S S, Shah J and Pfeiffer L N 1996 *Phys. Rev.* B **54** 4891), the calculated time is 108 ps, which is of the same order of magnitude as the experimentally deduced one of 50 ps. We find that the approximation in which the wave-vector component q_z of the LA phonon is taken as zero can lead to underestimation of the rate of exciton formation. A longer time of 150 ps for the exciton formation can be obtained using this approximation.

1. Introduction

Excitons in quantum wells (QWs) play an important role in optical spectra due to their high binding energies and enhanced oscillator strengths. Their formation dynamics has become a subject of considerable interest in recent years [1–4]. Under non-resonant excitation, free electrons and holes are generated and then relax into the lowest subbands, where they combine together to form excitons with various wave vectors ($\boldsymbol{k}_x$). Only when the excitons with large $\boldsymbol{k}_x$-vectors relax into a small region around $\boldsymbol{k}_x = 0$ can they undergo radiative decay. Thus the rise time of the exciton luminescence mainly reflects the relaxation and cooling processes of the excitons themselves [5], not the time taken for excitons to form from free-electron–hole pairs. Experimentally, there are several indirect ways to deduce the time taken for excitons to form [1–4, 6, 7]. Because the decay of the free-carrier density can be related to the exciton formation, one way to obtain this time is to measure the dependence of the intensity of the luminescence from free-electron–hole recombination on time. Recently, Kumar *et al* have carried out such a measurement for an 80 Å GaAs quantum well [6]. Their analysis gives a time constant of 50 ps for an excitation density of 4×10^{10} cm^{-2}. Yoon *et al* measured the change of the electron–hole (e–h) plasma temperature with time for a 210 Å GaAs quantum well [7]. Their results show that both LO and LA phonons might take part in the cooling of the electron–hole plasma. On the theoretical front, Selbmann *et al* have obtained a time of 50 ps for exciton formation in bulk GaAs using an ensemble Monte Carlo approach taking into account various scattering mechanisms [8]. By calculating the rate of transition of a free-e–h pair to an exciton state

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through longitudinal acoustic (LA) phonon emission, Thilagam *et al* obtained a time constant of a few 100 ps for QWs with widths of 2.5–20 nm [9]. So we might ask what the most important mechanism for exciton formation in QWs is. In this study, we have calculated the time taken for excitons to form in an 80 Å GaAs quantum well using a two-band model. For the wide quantum wells used by Yoon *et al*, both heavy and light holes should be considered. In this case, a two-band model is not sufficient; we defer their investigation to a future date. Our results show that the time taken for excitons to form should depend on the excitation conditions: the excitation density, the e–h plasma temperature and the lattice temperature. For the excitation conditions used in Kumar's experiment, LA phonon emission is the most important mechanism for exciton formation. Comparing with our results, we find that the approximations adopted in Thilagam's calculations can lead to a longer exciton formation time. Our calculated time is 108 ps, which is of the same order of magnitude as the one deduced from experiment.

The paper is organized as follows. The theoretical framework is given in section 2. In section 3, computational results and a discussion are presented. Finally in section 4, conclusions are given.

2. The theoretical framework

2.1. Equations for the distributions of free carriers

In this study, we do not consider hot-phonon effects; phonons are thought of as in an equilibrium state, with the lattice temperature T_L. Because laser-excited electrons and holes can quickly relax to the lowest subbands and into a quasi-thermal equilibrium state described with an e–h plasma temperature T_{eh} due to very fast carrier–carrier collisions, it is not necessary to include the excitation process; we can just study the decay of high-temperature electron–hole plasma in a two-band model due to exciton formation. Adopting the experimentally obtained value $T_L = 8.5$ K allows us to neglect exciton dissociation. In an 80 Å GaAs/$Al_{0.3}Ga_{0.7}$As QW, the lowest electron subband edge is at 44.3 meV and the lowest heavy-hole subband edge is at 10.67 meV relative to the fundamental gap of bulk GaAs of 1.519 eV. We have the following effective masses: $m_e = 0.0665$ and $m_h = 0.6165$. Here the heavy-hole effective mass is obtained from an energy band calculation using a Luttinger Hamiltonian, which then gives an exciton binding energy of $E_b = 10$ meV for the 1s state. We assume that free electrons and holes can be described by two-dimensional subbands with the following wave functions:

$$\varphi_{s,\boldsymbol{k}_s} = \frac{1}{\sqrt{A}} \exp(\mathrm{i}\boldsymbol{k}_s \cdot \boldsymbol{\rho}_s)\phi_s(z_s). \tag{1}$$

Here $s = \mathrm{e}, \mathrm{h}$, ϕ_s is the subband wave function of an electron with $s = \mathrm{e}$ or a hole with $s = \mathrm{h}$, and A is the normalization area. For the exciton, we only consider the 1s state, which is given by

$$\varphi_{ex,\boldsymbol{k}_x} = \frac{1}{\sqrt{A}} \exp(\mathrm{i}\boldsymbol{k}_x \cdot \boldsymbol{R})\sqrt{\frac{2}{\pi a^2}} \exp\left(-\frac{\rho}{a}\right)\phi_e(z_e)\phi_h(z_h) \tag{2}$$

with the exciton radius $a = 100$ Å. $\boldsymbol{R}$ and $\boldsymbol{\rho}$ are the centre-of-mass coordinate and the relative one of the electron and hole in the plane of the quantum well, respectively. We use the Boltzmann-equation approach to study the time evolution of the free-electron and free-hole distributions. For simplicity, we do not consider the band-to-band recombination

in the following. We have the following equation for the free-electron distribution:

$$\frac{\mathrm{d}f_e(\boldsymbol{k}_e)}{\mathrm{d}t} = \sum_{\boldsymbol{k}'_e} \Gamma^e_{in}(\boldsymbol{k}_e, \boldsymbol{k}'_e) f_e(\boldsymbol{k}'_e)(1 - f_e(\boldsymbol{k}_e)) - \sum_{\boldsymbol{k}'_e} \Gamma^e_{out}(\boldsymbol{k}_e, \boldsymbol{k}'_e) f_e(\boldsymbol{k}_e) \times (1 - f_e(\boldsymbol{k}'_e)) - \sum_{\boldsymbol{k}_x, \boldsymbol{k}_h} 2\Gamma(\boldsymbol{k}_x, \boldsymbol{k}_e, \boldsymbol{k}_h) f_e(\boldsymbol{k}_e) f_h(\boldsymbol{k}_h) + \left.\frac{\mathrm{d}f_e(\boldsymbol{k}_e)}{\mathrm{d}t}\right|_{cc}. \tag{3}$$

Here both the first and second term describe intrasubband scattering by phonons. For LA phonon scattering via deformation potential coupling, we consider both emission or absorption processes; for LO phonon scattering via polar interaction, we only consider emission processes because of the low lattice temperature. The third term in the above equation gives the rate of the formation of excitons; we calculate it by considering just the phonon emission processes. The factor 2 in this term is from the spin degeneracy. All of these three transition rates concerned with scattering of phonons can be calculated using the Fermi golden rule. The last term describes the change of the distribution due to carrier–carrier scattering; we will deal with it in the relaxation time approximation. Similarly, we have an equation for the distribution for free holes:

$$\frac{\mathrm{d}f_h(\boldsymbol{k}_h)}{\mathrm{d}t} = \sum_{\boldsymbol{k}'_h} \Gamma^h_{in}(\boldsymbol{k}_h, \boldsymbol{k}'_h) f_h(\boldsymbol{k}'_h)(1 - f_h(\boldsymbol{k}_h)) - \sum_{\boldsymbol{k}'_h} \Gamma^h_{out}(\boldsymbol{k}_h, \boldsymbol{k}'_h) f_h(\boldsymbol{k}_h) \times (1 - f_h(\boldsymbol{k}'_h)) - \sum_{\boldsymbol{k}_x, \boldsymbol{k}_e} 2\Gamma(\boldsymbol{k}_x, \boldsymbol{k}_e, \boldsymbol{k}_h) f_e(\boldsymbol{k}_e) f_h(\boldsymbol{k}_h) + \left.\frac{\mathrm{d}f_h(\boldsymbol{k}_h)}{\mathrm{d}t}\right|_{cc}. \tag{4}$$

In order to facilitate the numerical calculation, we introduce a discretization in wave-vector space to transform the above equations into a set of differential equations. Due to isotropy, the distributions of electrons and holes only depend on the absolute value of the wave vector; we find that an equispaced discretization (an energy interval of $\Delta = 0.5$ meV is used) in the energy space is more convenient. Taking the average of the scattering rate in the wave-vector space and then summing over the final exciton states [11], we can change the above equations into the following forms:

$$\frac{\mathrm{d}f_e(E_{ei})}{\mathrm{d}t} = \sum_{ej} \Gamma^e_{in}(E_{ei}, E_{ej}) f_e(E_{ej})(1 - f_e(E_{ei})) - \sum_{ej} \Gamma^e_{out}(E_{ei}, E_{ej}) \times f_e(E_{ei})(1 - f_e(E_{ej})) - \sum_{hj} 2m_h \Gamma(E_{ei}, E_{hj}) f_e(E_{ei}) f_h(E_{hj}) + \frac{f_e(E_{ei}) - F_e(\mu_e, T_{eh})}{\tau_{cc}} \tag{5}$$

$$\frac{\mathrm{d}f_h(E_{hi})}{\mathrm{d}t} = \sum_{hj} \Gamma^h_{in}(E_{hi}, E_{hj}) f_h(E_{hj})(1 - f_h(E_{hi})) - \sum_{hj} \Gamma^h_{out}(E_{hi}, E_{hj}) \times f_h(E_{hi})(1 - f_h(E_{hj})) - \sum_{ej} 2m_e \Gamma(E_{ej}, E_{hi}) f_e(E_{ej}) f_h(E_{hi}) + \frac{f_h(E_{hi}) - F_h(\mu_h, T_{eh})}{\tau_{cc}}. \tag{6}$$

In the above two equations, the indices ei, ej, hi and hj are used to label different energy intervals. In addition, we have made the relaxation time approximation for the carrier–carrier scattering. $F_s(\mu_s, T_{eh})$ $(s = \mathrm{e}, \mathrm{h})$ is a Fermi distribution with chemical potential μ_s and temperature T_{eh}. Because τ_{cc} is very short (50 fs for bulk GaAs) [12], we choose $F_s(\mu_s, T_{eh})$ $(s = \mathrm{e}, \mathrm{h})$ to give the same energy and the same number of particles of the

system at the current time. The scattering rate $\Gamma(\boldsymbol{k}_x, \boldsymbol{k}_e, \boldsymbol{k}_h)$ in equation (3) and equation (4) is transformed as follows. First we define the scattering rate $\Gamma_k(k_e, k_h)$:

$$\Gamma_k(k_e, k_h) \equiv \sum_{\boldsymbol{k}_x} \frac{1}{(2\pi)^2} \int_0^{2\pi} \mathrm{d}\theta_e \int_0^{2\pi} \mathrm{d}\theta_h \ \Gamma(\boldsymbol{k}_x, \boldsymbol{k}_e, \boldsymbol{k}_h)$$
$$= \frac{A}{(2\pi)^4} \int_0^{\infty} k_x \ \mathrm{d}k_x \int_0^{2\pi} \mathrm{d}\theta_x \int_0^{2\pi} \mathrm{d}\theta_e \int_0^{2\pi} \mathrm{d}\theta_h \ \Gamma(\boldsymbol{k}_x, \boldsymbol{k}_e, \boldsymbol{k}_h). \tag{7}$$

Here θ_x, θ_e and θ_h are the polar angles of $\boldsymbol{k}_x$, $\boldsymbol{k}_e$ and $\boldsymbol{k}_h$ in the plane of the quantum well. Then we transform $\Gamma_k(k_e, k_h)$ into the energy space; it takes the form

$$\Gamma(E_{ei}, E_{hj}) = \frac{A(\Delta E)}{2\pi\hbar^2} \Gamma_k\left(\sqrt{\frac{2m_e E_{ei}}{\hbar^2}}, \sqrt{\frac{2m_h E_{hj}}{\hbar^2}}\right). \tag{8}$$

$\Gamma(E_{ei}, E_{hj})$ is just the scattering rate in equations (5) and (6) describing the exciton formation. The scattering rates in equations (5) and (6) are computed once at the fixed lattice temperature and tabulated. The angular integrals in these scattering rates are evaluated by means of the Gauss–Legendre integrating formula and other integrals by the simple trapezoidal rule. Then, given initial distributions for electrons and holes, we can solve the above equations by the fourth-order Runge–Kutta method. In the following calculation, the commonly used parameters of GaAs are adopted [8], and the lattice temperature $T_L = 8.5$ K is adopted.

2.2. Scattering rates

It is very easy to find intraband phonon scattering rates in the literature [8, 11], so we only give the rate of exciton formation in the following.

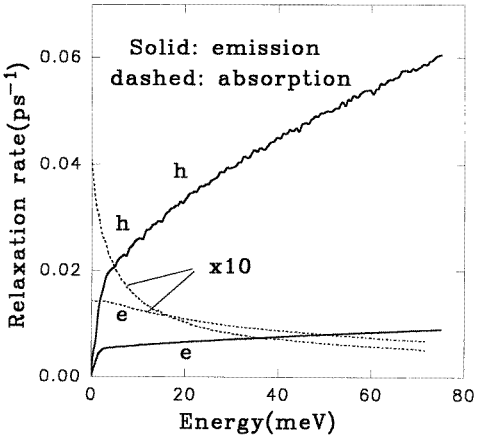

Figure 1. Rates of intraband relaxation due to LA phonon emission or absorption for electrons and holes.

The short time (100–200 fs) taken for intraband relaxation due to LO phonon emission to occur makes this relaxation mechanism mainly act only before the formation of a quasi-thermal electron–hole plasma. We do not study this initial stage here. Although we include this relaxation mechanism in our calculation, it is not important in the present case. In figure 1, we present the intraband relaxation rates due to LA phonon emission or absorption

for free electrons and holes. We can see that the electron scattering rate is about 0.005 ps^{-1}, while the hole scattering rate is much larger, about 0.03 ps^{-1} in the physically important energy range.

The picture of exciton formation is as follows: an electron ($\boldsymbol{k}_e$, E_e) and a hole ($\boldsymbol{k}_h$, E_h) in the continuum as a whole emit or absorb a phonon $\boldsymbol{Q} = (\boldsymbol{q}_\parallel, q_z)$ and are transformed into an exciton state ($\boldsymbol{k}_x$, E_x). Thus, the exciton formation rate can be calculated by means of the Fermi golden rule [8]:

$$\Gamma_\pm(\boldsymbol{k}_x, \boldsymbol{k}_e, \boldsymbol{k}_h) = \frac{2\pi}{\hbar} \sum_{\boldsymbol{Q}} |D^i|^2 \left(n_{\boldsymbol{Q}} + \frac{1}{2} \pm \frac{1}{2} \right) \delta_{\boldsymbol{k}_x \pm \boldsymbol{q}_\parallel, \boldsymbol{k}_e + \boldsymbol{k}_h} \delta(E_x - E_e - E_h \pm \hbar\omega_{\boldsymbol{Q}}). \tag{9}$$

Here the upper and lower indices denote phonon emission and absorption, respectively. $E_x = \hbar^2 k_x^2/2M - E_b$ is the energy of the exciton, with $M = m_e + m_h$; $E_e = \hbar^2 k_e^2/2m_e$ and $E_h = \hbar^2 k_h^2/2m_h$ are the energy of the electron and hole, respectively. $\hbar\omega_{\boldsymbol{Q}}$ is the energy of the phonon. The coupling constant D^i is given by

$$D^i = C^i_{e\boldsymbol{Q}} S_x[\beta(\boldsymbol{k}_e \mp \boldsymbol{q}_\parallel) - \alpha \boldsymbol{k}_h] F_e(q_z) - C^i_{h\boldsymbol{Q}} S_x[\beta \boldsymbol{k}_e - \alpha(\boldsymbol{k}_h \mp \boldsymbol{q}_\parallel)] F_h(q_z). \tag{10}$$

The superscript i denotes the mechanism of scattering. The parameters α and β are defined as $\alpha = m_e/M$, $\beta = m_h/M$. S_x is the overlap of the exciton 1s wave function with the plane-wave states in the exciton continuum, and is given by

$$S_x(\boldsymbol{q}) = 2\sqrt{\frac{2\pi a^2}{A}} \frac{1}{(1 + (qa)^2)^{3/2}}. \tag{11}$$

$F_e(q_z)$ and $F_h(q_z)$ are defined as

$$\begin{aligned} F_e(q_z) &= \int \mathrm{d}z_e \ |\phi_e(z_e)|^2 \exp(\mathrm{i} q_z z_e) \\ F_h(q_z) &= \int \mathrm{d}z_h \ |\phi_h(z_h)|^2 \exp(\mathrm{i} q_z z_h). \end{aligned} \tag{12}$$

$C^i_{s\boldsymbol{Q}}$ specifies the interaction of particles with lattice vibrations. We consider the Fröhlich interaction with LO phonons of frequency ω_{LO}; we have

$$|C_{e,\boldsymbol{Q}}|^2 = |C_{h,\boldsymbol{Q}}|^2 = \frac{2\pi e^2}{V} \hbar\omega_{LO} \left(\frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_0} \right) \frac{1}{(q_\parallel^2 + q_z^2)}. \tag{13}$$

The deformation potential coupling to LA phonons is given by

$$|C_{s,\boldsymbol{Q}}|^2 = \frac{\hbar}{2\rho u_l V} D_s^2 \sqrt{(q_\parallel^2 + q_z^2)} \qquad (s = \mathrm{e}, \mathrm{h}). \tag{14}$$

In the above equations, ϵ_∞ and ϵ_0 are the high- and low-frequency dielectric constants, D_s the deformation potential, ρ the crystal density, u_l the velocity of sound and V a normalization volume.

The scattering rate $\Gamma(\boldsymbol{k}_x, \boldsymbol{k}_e, \boldsymbol{k}_h)$ describing the rate of exciton formation in equations (3) and (4) is

$$\Gamma(\boldsymbol{k}_x, \boldsymbol{k}_e, \boldsymbol{k}_h) = \Gamma_+(\boldsymbol{k}_x, \boldsymbol{k}_e, \boldsymbol{k}_h) + \Gamma_-(\boldsymbol{k}_x, \boldsymbol{k}_e, \boldsymbol{k}_h). \tag{15}$$

At the low lattice temperature, the process of phonon absorption is very slow, so in the following we just retain the first term in the right-hand side of the above equation. The rate of the exciton formation due to LO phonon emission can be calculated easily. However, computing the rate due to LA phonon emission is very time consuming. The usual approximation is to set the wave-vector component of the phonon, q_z, equal to zero [9, 10]. This is equivalent to setting $F_e(q_z) = 1$, $F_h(q_z) = 1$ in equation (10) and $q_z = 0$, $V = AL_w$ in equation (14). Here L_w is the width of the quantum well. Our calculation shows that this approximation can cause large errors.

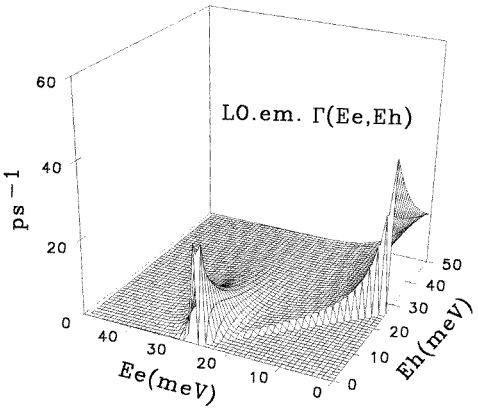

Figure 2. The rate of exciton formation $\Gamma(E_e, E_h)$ due to LO phonon emission.

3. Results and discussion

The map of $\Gamma(E_e, E_h)$ due to LO phonon emission is presented in figure 2. There are two regions in the E_e–E_h plane with large transition rates. The first one is of low electron energy and high hole energy. Due to the fast relaxation of holes, this region is in fact not important. The second region is around the point with $E_e + E_b = \hbar\omega_{LO}$ and low hole energy. This region is found to be important only at high electron–hole plasma temperature. The maps of $\Gamma(E_e, E_h)$ due to LA phonon emission without and with the approximation $q_z = 0$ made are presented in figures 3(a) and 3(b). It can be seen that $\Gamma(E_e, E_h)$ depends mainly on E_e, in contrast with the intraband LA phonon scattering. In the latter case, the electron has a smaller scattering rate than the hole, as shown in figure 1. The electrons with low energy are easier to combine with holes to form excitons. Making the approximation $q_z = 0$ leads to underestimation of the transition rate, and thus a longer exciton formation time, which will be discussed in the following. We point out that the value of $\Gamma(E_e, E_h)$ depends on the specific discretization in the energy space, as, according to our definition, $\Gamma(E_e, E_h)$ is proportional to ΔE. It is $m_e \ll m_h$ that causes the part of $\Gamma(E_e, E_h)$ due to LA phonon emission to depend weakly on E_h. To understand this point, let $\alpha = 0$, $\beta = 1$, and define

$$\begin{aligned}\Gamma_{\pm}(\boldsymbol{k}_e, \boldsymbol{k}_h) \equiv \sum_{k_x} \Gamma_{\pm}(\boldsymbol{k}_x, \boldsymbol{k}_e, \boldsymbol{k}_h) &= \frac{2\pi}{\hbar} \sum_{\boldsymbol{Q}} |D^i|^2 \left(n_{\boldsymbol{Q}} + \frac{1}{2} \pm \frac{1}{2} \right) \\ &\times \delta\left(\frac{\hbar^2 \boldsymbol{q}_{\parallel}{}^2}{2M} \mp \frac{\hbar^2 \boldsymbol{q}_{\parallel} \cdot (\boldsymbol{k}_e + \boldsymbol{k}_h)}{M} - E_b - \frac{\hbar^2 (\beta \boldsymbol{k}_e - \alpha \boldsymbol{k}_h)^2}{2\mu} \pm \hbar\omega_{\boldsymbol{Q}} \right) \\ &\approx \frac{2\pi}{\hbar} \sum_{\boldsymbol{Q}} |D^i|^2 \left(n_{\boldsymbol{Q}} + \frac{1}{2} \pm \frac{1}{2} \right) \delta\left(\frac{\hbar^2 \boldsymbol{q}_{\parallel}{}^2}{2M} - E_b - \frac{\hbar^2 \boldsymbol{k}_e{}^2}{2\mu} \pm \hbar\omega_{\boldsymbol{Q}} \right). \end{aligned} \tag{16}$$

Here $M = m_e + m_h$, and $\mu = m_e m_h/(m_e + m_h)$. In the last line of the above equation, we have let $\alpha = 0$, $\beta = 1$ in the equation for the coupling constant D^i, and we have taken the term $\mp\hbar^2 \boldsymbol{q}_{\parallel} \cdot (\boldsymbol{k}_e + \boldsymbol{k}_h)/M$ as zero in the delta function, to ensure energy conservation, because the average of this term is zero. The final result clearly shows the part of $\Gamma(E_e, E_h)$ due to LA phonon emission to depend mainly on E_e. In figure 4 we present $\Gamma(E_e, E_h = 0)$ at different lattice temperatures $T_L = 8.5$ K, 77 K and 300 K. The value of $\Gamma(E_e, E_h = 0)$

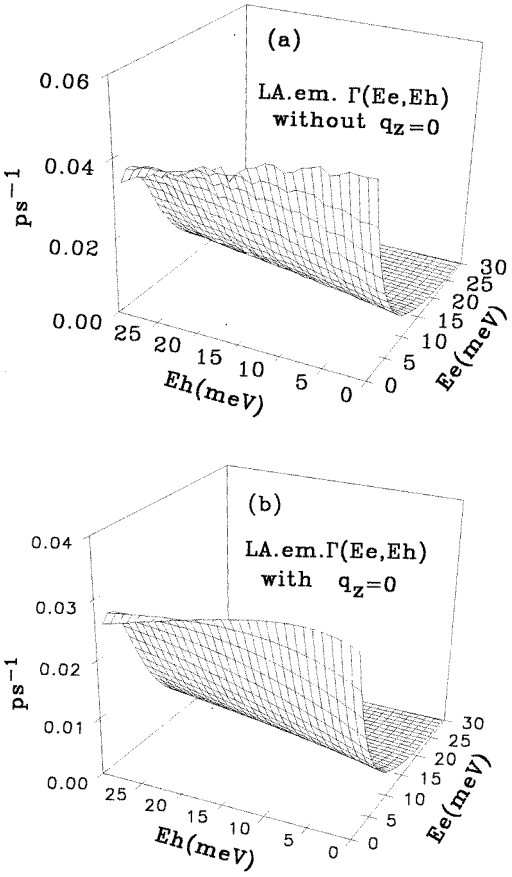

Figure 3. The rate of exciton formation $\Gamma(E_e, E_h)$ due to LA phonon emission. (a) Without making the approximation $q_z = 0$; (b) with the approximation $q_z = 0$ made.

increases very quickly with the increase of T_L. On the other hand, the time taken for excitons to dissociate will become short at high T_L. To study these processes, one must also include the time evolution of excitons; we defer this problem to study at a future date.

Next we study the decay of the density of free-electron–hole pairs. We consider three cases: (a) corresponding to the experimental conditions used by Kumar *et al*: excitation density $n = 4 \times 10^{10}$ cm^{-2} and initial plasma temperature $T_{eh} = 50$ K; (b) $n = 4 \times 10^{10}$ cm^{-2}, $T_{eh} = 80$ K; and (c) $n = 2 \times 10^{10}$ cm^{-2}, $T_{eh} = 50$ K. For comparison, for each case we compute the decay of the density of free-electron–hole pairs without making the approximation $q_z = 0$ and with this approximation made. We adopt $\tau_{cc} = 50$ fs as the carrier–carrier scattering time. As this time is far shorter than other characteristic times concerned with emission and absorption of LA phonons, the specific choice made for its value does not change the final results very much. In figure 5 we present the decay of the free-electron–hole pair density for the above three cases. The characteristic time τ_f of the decay is obtained by fitting the decay of the pair density to

$$n = n_0 \frac{\tau_f}{t + \tau_f} \tag{17}$$

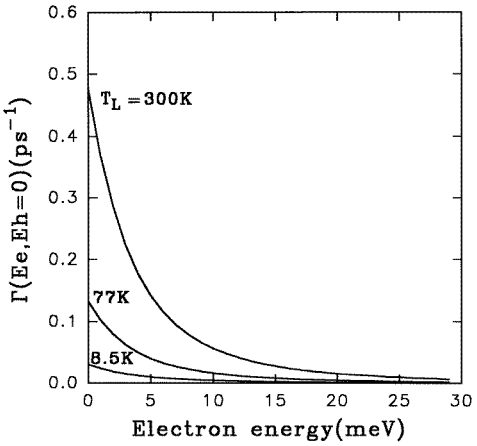

Figure 4. The rate of exciton formation $\Gamma(E_e, E_h = 0)$ due to LA phonon emission at $T_L = 8.5$ K, 77 K and 300 K. The approximation $q_z = 0$ is adopted.

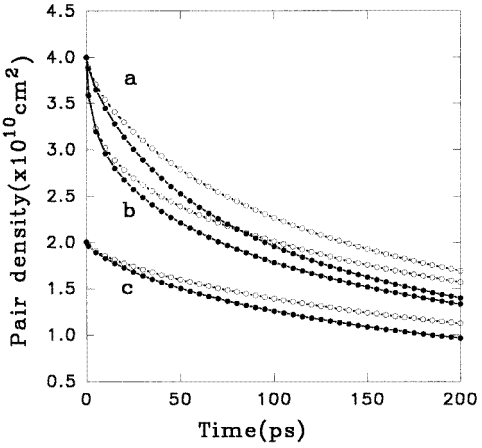

Figure 5. The time dependence of the free-electron–hole pair density for three sets of initial parameters: (a) $n = 4 \times 10^{10}$ cm^{-2}, $T_{eh} = 50$ K; (b) $n = 4 \times 10^{10}$ cm^{-2}, $T_{eh} = 80$ K; and (c) $n = 2 \times 10^{10}$ cm^{-2}, $T_{eh} = 50$ K. For each case: filled circles: without making the approximation $q_z = 0$; open circles: with the approximation $q_z = 0$ made, for the component q_z of the wave vector of the LA phonon.

which is the solution of the following equation describing the bimolecular exciton formation:

$$\frac{\mathrm{d}n}{\mathrm{d}t} = -\frac{n^2}{\tau_f n_0}. \tag{18}$$

The results are 108 ps, 113 ps and 192 ps for cases (a), (b) and (c) without making the approximation $q_z = 0$, respectively. In contrast, they are 150 ps, 155 ps and 267 ps when the approximation $q_z = 0$ is used. For case (b), the fast decay during the initial time of 10 ps is caused by LO phonon emission; the slow decay at later times is completely due to LA phonon emission. For cases (a) and (c), only slow decay due to the process of LA phonon emission is present. From this result, we have following conclusions.

(1) The time taken for excitons to form due to LO phonon emission is very short, i.e. less than 10 ps.

(2) The time taken for excitons to form is mainly determined by LA phonon emission—108 ps or so according to the experimental conditions.

(3) Making the approximation $q_z = 0$ leads to a longer exciton formation time, i.e. 150 ps or so from our numerical calculation.

In order to understand the above results qualitatively, according to figure 3 and figure 4, we approximate the rate of exciton formation due to LA phonon emission by

$$\Gamma(E_e, E_h) = C(T_L) \exp\left(-\frac{E_e}{E_0}\right). \tag{19}$$

Here $C(T_L)$ is a coefficient depending on T_L, and E_0 is an energy parameter and is approximately equal to the exciton binding energy E_b from physical considerations. We assume that the pair density can be described by a Boltzmann distribution:

$$F_s(E_s) = \frac{2\pi n\beta}{m_s} \exp(-\beta E_s) \qquad (s = \text{e, h}). \tag{20}$$

Here $\beta = 1/k_B T_{eh}$, m_s ($s = $ e, h) is the effective mass and n is the two-dimensional density. Considering the slow change of T_{eh}, and ignoring the intraband scattering terms, we obtain the following relation for time constant τ_f in equation (18) in a short time:

$$\tau_f \propto 1 + \frac{k_B T_{eh}}{E_b} \tag{21}$$

at a fixed lattice temperature. τ_f describes the time taken for excitons to form. Over long times, we must consider the time dependence of T_{eh}. This equation tells us how the exciton formation time depends on T_{eh} and E_b.

Now we compare our result with those derived by others. The calculated exciton formation time is 108 ps, which is of the same order of magnitude as, but longer than, the time of 50 ps deduced experimentally by Kumar *et al*. The underlying reason for this is still not clear. It might be because of non-radiative combination [13], which is not included in our calculation. On the other hand, our calculated time is shorter than the one calculated by Thilagam *et al* [9], whose theory does not show how T_{eh} affects the time taken for excitons to form. The calculation of the time taken for excitons to form in bulk GaAs by Selbmann *et al* shows that relaxation with LO phonons involved is more efficient than that involving LA phonons, contrary to the results of the present work. This may be due to the different ranges of the lattice temperature T_L considered in the two calculations. Selbmann *et al* used a high value of T_L, for which the relaxation processes of LO phonon emission and absorption are more efficient. We consider a low value of T_L; in this case relaxation involving LA phonons becomes more important.

Our above-described approach for calculating the rate of exciton formation in quantum wells is in essence an extension of the work of Selbmann *et al* and is phenomenological. It is very desirable to establish it using the theory of the non-equilibrium Green's function. Due to the phenomenology and the approximations made, such as neglecting the light emission recombination and the dissociation of excitons, the present approach may not be quantitatively reliable; however, it does give some degree of qualitative understanding of the rate of exciton formation in quantum wells. It can qualitatively describe the early stage of the exciton formation, and is only valid in the case of low T_L and high T_{eh}.

4. Conclusions

In conclusion, we have calculated the time taken for excitons to form in an 80 Å GaAs quantum well. Our results show that this time is determined mainly by the process of LA phonon emission at low lattice temperatures. The calculated time is 108 ps or so, which is of the same order of magnitude as, but longer than, the experimentally deduced one. We show that the formation time is approximately proportional to $1 + k_B T_{eh}/E_b$ at a fixed lattice temperature. It would be desirable to include the dynamics of excitons explicitly and to describe carrier–carrier collisions taking into consideration realistic screening effects. Such work is under way.

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