PHYSICAL REVIEW B VOLUME 48, NUMBER 20 15 NOVEMBER 1993-II

Radiative lifetimes, quasi-Fermi-levels, and carrier densities in GaAs-(Ga,Al)As quantum-well photoluminescence under steady-state excitation conditions

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(Received 25 March 1993; revised manuscript received 30 June 1993)

A quantum-mechanical calculation of the carrier densities, electron and hole quasi-Fermi-levels, and various radiative decay times in GaAs-(Ga,Al)As quantum wells is performed, under steady-state excitation conditions, as functions of the cw laser intensity, temperature, well widths, and acceptor distribution in the well. We consider the radiative recombination of electrons with free holes and with holes bound at neutral acceptors. Our calculations—which have no free parameters—are in quantitative agreement in the intermediate laser-intensity regime at $T=300$ K with the results by Ding *et al.* [Appl. Phys. Lett. **60**, 2051 (1992)], who obtained the carrier density for multiple asymmetric coupled quantum wells through a fitting procedure that reproduced the total experimental photoluminescence intensity. Results for the carrier-dependent *e-h* recombination decay time are in good agreement with experimental data by Bongiovanni and Staehli [Phys. Rev. B **46**, 9861 (1992)].

I. INTRODUCTION

The area of semiconducting heterostructures has been intensively studied both theoretically and experimentally in the last few years. Modern growth techniques such as molecular-beam epitaxy (MBE) and metal-organic chemical-vapor deposition (MOCVD) have made possible the realization of high-quality samples with sharp interfaces, and controlled layer thicknesses. Carrier-confinement effects in these systems give rise to a variety of interesting phenomena in a much different range as compared to bulk materials. Potential device applications in optoelectronics, for instance, make understanding of the properties of semiconducting heterostructures a field of considerable technological and scientific importance. Superlattices, multiple quantum wells, isolated quantum wells (QW's), and heterostructures of GaAs and $\mathrm{Ga}_{1-x}\mathrm{Al}_x\mathrm{As}$ constitute, for a number of reasons, the most widely studied of these systems.

Photoluminescence (PL) is a powerful research technique in the understanding of the basic electronic properties of semiconductor systems.[1,2] Recombination processes associated to PL experiments carried out by using continuous-wave (cw) lasers or pulsed lasers in quasistationary or steady-state conditions have received some attention in recent theoretical and experimental work[3–8] on semiconducting heterostructures. Haug and Koch[3] theoretically investigated Coulomb interactions in the electron-hole (*e-h*) plasma of a semiconductor laser, and studied the reduction of the band gap with increasing carrier density, the *e-h* plasma screening of the Coulomb potential, and the enhancement of the optical interband transitions due to the attractive *e-h* interaction, and derived a diffusion equation for the carrier density. No comparison with experimental results is presented in their work. An experimental work on quasi-two-dimensional carrier systems of GaAs-(Ga,Al)As multiple-quantum-well structures was carried out by Schlaad *et al.*[4] under quasistationary excitation conditions using the pump and probe beam and luminescence spectroscopy, with the density and the reduced band gap determined via systematic evaluations of both gain and luminescence spectra. Their study of the higher subbands revealed that subband renormalization is due mainly to a direct occupation of the specific subband, and that the intersubband effects via Coulomb screening are negligible. The temperature dependence of the radiative and nonradiative recombination time constants in GaAs-(Ga,Al)As quantum-well structures were examined by Gurioli *et al.*[5] in PL experiments after both picosecond and continuous-wave (cw) excitation. They claimed that nonradiative processes play an important role and become dominant for $T \geq 100$ K, and that the radiative time constant increases by several orders of magnitude as the temperature is raised from $T=4$ K to room temperature. Bongiovanni and Staehli[6] investigated the density dependence of the *e-h* plasma decay time versus pair density in semiconductor quantum wells via a steady-state PL experiment. They showed that only radiative *e-h* recombination is important, and that nonradiative processes and plasma expansion have negligible effects on the total plasma lifetime. They also observed, at large plasma densities, a strong nonlinear reduction of the *e-h* capture rate. Ding *et al.*[7] have studied multiple narrow asymmetric coupled quantum wells (ACQW's) at room temperature, and obtained the intensity-dependent carrier density by a fitting procedure in order to reproduce the experimental cw-laser PL total intensity. They found that the dependence of the carrier density on laser intensity undergoes a clear transition from linear to square root for increasing laser intensities. In the case of acceptor-doped semiconducting heterostructures, several authors have treated the radiative recombination of quasi-two-dimensional electrons with bound holes both theoretically[8,9] and experimentally.[10]

The purpose of this work is to study the laser-intensity dependence of the radiative lifetimes, quasi-Fermi-levels, and carrier densities in GaAs-(Ga,Al)As QW's under steady-state cw-laser PL conditions via a quantum-mechanical calculation of the above quantities for a GaAs-(Ga,Al)As QW in a cw-laser PL experiment. Section II presents our theoretical model for treating quantum-well PL under steady-state excitation conditions. Results and a discussion are presented in Sec. III, and conclusions in Sec. IV

II. QUANTUM-WELL PHOTOLUMINESCENCE: THEORY UNDER STEADY-STATE CONDITIONS

In general, when a physical system is irradiated with light, the emitted optical radiation (photoluminescence) results from the excitation of the system to a nonequilibrium state.[1] For a semiconducting system, one may essentially distinguish three processes, namely (i) absorption of the exciting light with creation of *e-h* pairs, (ii) radiative recombination, and (iii) nonradiative recombination.

The final steady-state situation for the conduction-band electron density will occur when the rate of creation of photoexcited electron-hole pairs equates the rate of radiative and nonradiative *e-h* recombinations.

In this work, we are concerned with a single isolated GaAs-(Ga,Al)As QW of thickness L excited by a cw laser in a PL experiment. We consider acceptors as channels for radiative recombination, as it appears that carbon acceptors are common in several nominally undoped QW samples[10] (high-quality samples would correspond to a concentration of acceptors of about $10^{14}-10^{15}/\mathrm{cm}^3$). We follow Bongiovanni and Staehli[6] and Ding *et al.*,[7] and neglect Auger processes and excitonic recombination, although in the low-temperature regime excitonic recombination processes may be important, as we comment in the discussion in Sec. III. Other mechanisms, such as nonradiative recombination with interface traps (which may be relevant in the high-temperature regime), spatial diffusion, and drift,[7] will not be considered here. In what follows, we will work within the effective-mass approximation and use the parabolic-band model for describing both electrons and holes [for simplicity, we neglect the effect of coupling of the top four valence bands[11] of both GaAs and (Ga,Al)As and consider one heavy-hole band with a spherical carrier effective mass $m_v \simeq 0.3m_0$ which gives a bulk value[12] of 26 meV for the acceptor binding energy[13]]. We consider recombination of electrons in the $n=1$ conduction subband with free holes in the $n=1$ valence subband, and with holes bound to neutral acceptors distributed homogeneously inside the QW. The absorption of the exciting light creates *e-h* pairs. The electrons can emit photons by recombining with holes which are either free or bound to neutral acceptors. The rate equation for the change in the density of electrons per unit of area is given by

$$\frac{dn_e}{dt} = \omega_A - \omega_{\mathrm{cv}} - \omega_{cA} , \tag{2.1}$$

where ω_A, ω_{cv}, and ω_{cA} are the rate per unit area of interband absorption, electron radiative recombination with free holes, and bound holes, respectively. In the steady state, the carrier density n_e is determined by

$$\omega_A = \omega_{\mathrm{cv}} + \omega_{cA} , \tag{2.2}$$

which gives a transcendental equation relating n_e to the laser intensity, and which should be solved numerically, as we shall demonstrate below. The number of free holes at the valence subband may be readily obtained via charge conservation, if one takes into account the number of ionized acceptors, and therefore one may calculate the chemical potentials (or quasi-Fermi-levels) for electrons and holes and the various radiative lifetimes as functions of the laser intensity, temperature, well widths, and acceptor concentration. We now calculate the interband absorption (ω_A) and recombination (ω_{cv} and ω_{cA}) rates which appear in the above equations.

Let us consider the transition probability per unit time for valence to conduction subband transitions which is proportional to the square of the matrix element of the electron-photon interaction H_{int} between the wave functions of the initial (valence) and final (conduction) states, i.e.,

$$W = \frac{2\pi}{\hbar} \sum_i |\langle f|H_{\mathrm{int}}|i\rangle|^2 \delta(E_f - E_i - \hbar\omega) , \tag{2.3}$$

with $H_{\mathrm{int}} = (eA_0/m_0c)\hat{\varepsilon}\cdot\mathbf{p}$, where $\hat{\varepsilon}$ is the polarization vector in the direction of the electric field of the radiation, $\mathbf{p}$ is the momentum operator, $-e$ and m_0 are the free-electron charge and mass, respectively, A_0 is the amplitude of the photon vector potential,[14] and $\hbar\omega$ is the photon energy.

By considering transitions only between the $n=1$ valence and conduction subbands, summing over spins, and taking the electric field of the incident light polarized parallel to the interface of the QW, the rate per unit area of creating electron-hole pairs by interband absorption of photons from a cw laser is proportional to the intensity I of the laser and given by

$$\omega_A = 4\pi\alpha_f \frac{\mu_{\mathrm{cv}}(p_{\mathrm{cv}}^2/2)}{\eta(\omega) m_0^2 (\hbar\omega)^2} |\langle f_c(z)|f_v(z)\rangle|^2 I\theta(\hbar\omega - \varepsilon_g) , \tag{2.4}$$

where α_f is the fine-structure constant, μ_{cv} is the conduction-valence-band reduced mass, $\eta(\omega)$ is the refraction index, $p_{\mathrm{cv}} = |\langle s|\hat{p}_x|x\rangle|$ is the interband[15] matrix element, $\langle f_c(z)|f_v(z)\rangle$ is the overlap integral between the electron and hole envelope wave functions in the QW, $\theta(x)$ is the Heaviside unit-step function, and ε_g is the effective-band gap of the QW.

The radiative recombination of electrons in the $n=1$ conduction subband with holes in the $n=1$ valence subband involves photons with wave vectors $\mathbf{q}$ in any direction, and one must sum in three dimensions over directions and polarizations. Therefore, the coefficient ω_{cv} for spontaneous emission, in units of photons per area per second, is

$$\omega_{\text{cv}}=(2e^2/m_0^2)\sum_{\lambda}\int\frac{d^3\mathbf{q}}{(2\pi)^3}\frac{|\langle f_v(z)|f_c(z)\rangle|^2}{\eta^2(\omega')\omega'}(p_{\text{cv}}^2/2)[\varepsilon_{\lambda x}^2+\varepsilon_{\lambda y}^2]\int d^2\mathbf{k}_\perp n_c(k_\perp)n_v(k_\perp)\delta[\varepsilon_g-\hbar\omega'+\hbar^2k_\perp^2/2\mu_{\text{cv}}] \tag{2.5}$$

where λ is associated to the two photon-polarization directions, $\varepsilon_{\lambda x}$ and $\varepsilon_{\lambda y}$ are the x and y components of the polarization unit vector, $\omega'=cq/\eta$ and $\mathbf{q}$ are the frequency and the wave vector of the emitted photon, and

$$n_c(k_\perp)=1/(\exp\{[\varepsilon_c(k_\perp)-\mu_e]/k_BT\}+1) \tag{2.6}$$

is the occupation number of electrons in the conduction subband, μ_e is the conduction-subband chemical potential, and $\varepsilon_c(k_\perp)$ is the electron kinetic energy in the xy plane, with a corresponding expression for $n_v(k_\perp)$ (the occupation number of holes in the valence subband). By considering the densities per unit of area of electrons and holes, respectively,

$$n_e=2/(2\pi)^2\int d^2\mathbf{k}_\perp n_c(k_\perp)\ , \tag{2.7}$$

$$n_h=2/(2\pi)^2\int d^2\mathbf{k}_\perp n_v(k_\perp)\ , \tag{2.8}$$

one arrives at

$$\omega_{\text{cv}}=\tfrac{8}{3}\alpha_f(\varepsilon_g/\hbar)\frac{\eta(\varepsilon_g)(p_{\text{cv}}^2/2)}{m_0^2c^2}\times|\langle f_c(z)|f_v(z)\rangle|^2 n_0 Q(n_e,n_h)\ , \tag{2.9}$$

where

$$Q(n_e,n_h)=\int_0^\infty dx\frac{[1+(1+\gamma)(k_BT/\varepsilon_g)x]}{[\delta e^x+1][\sigma e^{\gamma x}+1]}\ , \tag{2.10a}$$

with

$$n_0=m_ck_BT/\pi\hbar^2\simeq 2.4\times 10^9(T/K)\ \text{cm}^{-2}\ , \tag{2.10b}$$

$$\delta=1/[\exp(n_e/n_0)-1]=\exp(-\mu_e/k_BT)\ , \tag{2.10c}$$

$$\sigma=1/[\exp(\gamma n_h/n_0)-1]=\exp(-\mu_h/k_BT)\ , \tag{2.10d}$$

$\gamma=m_c/m_v$ and m_c (m_v) being the conduction- (valence-) band effective mass. The density n_h of free holes at the valence subband—which of course depends on temperature—is determined by conservation of charge in such a way that it is equal to n_e plus the density of ionized acceptors. Notice that the conduction- (valence-) subband quasi-Fermi-level μ_e (μ_h) is measured from the bottom of the $n=1$ subband. One should notice that the quantities n_0 and n_0/γ essentially define the boundary between the nondegenerate ($n_e \ll n_0$ or $n_h \ll n_0/\gamma$) and degenerate ($n_e \gg n_0$ or $n_h \gg n_0/\gamma$) electron- and hole-gas regimes, respectively. Also, note that parameter δ (σ) gives a measure of the electron- (hole-) chemical potential (in units of k_BT) with respect to the conduction- (valence-) band edge. Therefore, one may define a low-intensity regime (for which $n_{e,h} \ll n_0$) as the nondegenerate regime for which Maxwell-Boltzmann statistics is suitable, i.e., when the laser intensity $I \ll I_0$, with

$$I_0\approx 2\eta^2(\varepsilon_g)\frac{\omega^2(\varepsilon_g/\hbar)k_BT}{3\pi^2c^2}\gamma=1.5\times 10^2(T/K)\ \text{W/cm}^2 \tag{2.10e}$$

obtained by setting $\omega_A=\omega_{\text{cv}}$, with $n_e=n_0$. Alternatively, the high-intensity regime $I \gg I_0$ would correspond to the degenerate regime for which one should use Fermi-Dirac statistics. Also, we follow Ding *et al.*[7] and ignore the difference between internal and external laser intensities when comparing theoretical and experimental results. Our results, therefore, are only determined within the order of magnitude that the internal laser intensity I in our equations should be adjusted in order to take into account reflection losses of the external pump laser radiation.[6,7]

For very low laser intensities, an important recombination process is the emission of photons due to electron recombination with bound holes at acceptors. Assuming a distribution of acceptors inside the GaAs QW, and following a similar procedure as in the recombination with free holes, the rate per unit of area of radiative recombination of electrons in the $n=1$ conduction subband with holes bound at neutral acceptors is given by

$$\omega_{cA}=\frac{16}{3\hbar}N_A\alpha_f\frac{\eta(\varepsilon_g)(p_{cv}^2/2)}{m_0^2c^2}\times\int_{-L/2}^{L/2}dz_iJ_{cA}(\lambda,z_i)P(z_i)n_A[E_A(z_i)]\ , \tag{2.11}$$

with

$$J_{cA}(\lambda,z_i)=\frac{N^2(\lambda,z_i)}{N_v^2}\int\frac{d^2\mathbf{k}_\perp}{(2\pi)^2}n_c(k_\perp)K^2(z_i,\lambda,k_\perp)\times[\varepsilon_g-E_A(z_i)+\varepsilon_c(k_\perp)] \tag{2.12}$$

and

$$K(z_i,\lambda,k_\perp)=\int d^3r\, e^{i\mathbf{k}_\perp\cdot\rho}f_v(z)f_c(z)e^{-|\mathbf{r}-z_i\hat{z}|/\lambda}\ , \tag{2.13}$$

where N_A is the density of acceptors (number/cm^3), $P(z_i)$ is the probability distribution of acceptors, which we assume constant within the QW, i.e., $P(z_i)=1$,

$$n_A[E_A(z_i)]=2/\{2+\sigma\exp[-E_A(z_i)/k_BT]\} \tag{2.14}$$

is the probability distribution of having a hole bound to an acceptor at z_i, $E_A(z_i)$ is the acceptor binding energy, $N(\lambda,z_i)$ and λ are the normalization and variational parameters of the acceptor envelope wave function, and N_v is the normalization factor for the envelope wave function associated with the first valence subband.[13]

Finally, Eq. (2.2) should be supplemented by an equation determining the conservation of the charge, and relating the density of electrons at the conduction subband with the densities of holes at the valence subband and ionized acceptors, i.e.,

$$n_h = n_e + N_A \int_{-L/2}^{L/2} \{1 - n_A[E_A(z_i)]\} P(z_i) dz_i \ . \quad (2.15)$$

One should note that the above equation must be solved simultaneously with Eq. (2.2) in order to obtain the dependence on the laser intensity of the carrier densities, quasi-Fermi-levels for electrons and holes, and the various radiative decay times. In general, the decay lifetime corresponding to each recombination process is dependent on the laser intensity, or on the conduction-subband carrier density, and provides quantitative information on the relative importance of each recombination mechanism. In Sec. III, we denote by $T_{cv} = n_e/\omega_{cv}$ the lifetime of the electron radiative recombination with free holes, by $T_{cA} = n_e/\omega_{cA}$ the decay time related to recombination with bound holes at the acceptors, and by T_{tot} the total decay time

$$1/T_{tot} = 1/T_{cv} + 1/T_{cA} \ . \quad (2.16)$$

III. RESULTS AND DISCUSSION

In what follows, calculations were performed for a GaAs-$Ga_{1-x}Al_xAs$ single QW, and we assumed that the band-gap discontinuity in the heterostructure is distributed[16,17] about 60% (40%) on the conduction (valence) band with the total band-gap difference ΔE_g, between GaAs and $Ga_{1-x}Al_xAs$, given[18] as a function of the Al concentration x as $\Delta E_g(\mathrm{eV}) = 1.247x$. We have used $\eta(\omega) \simeq \eta(\varepsilon_g) \simeq 3.5$ for the index of refraction, and

$$E_g \ (\mathrm{eV}) = 1.519 - 5.405 \times 10^{-4} T^2/(T + 204) \quad (3.1)$$

for the temperature-dependent[19] bulk GaAs gap. Also, most of our results given for two typical QW widths, namely $L = 5$ and 10 nm, for low ($T = 2$ K) and high ($T = 300$ K) temperatures, for an Al concentration $x = 0.3$, and laser energy[6,7] $\hbar\omega = 1.75$ eV. Acceptor concentrations per unit of volume were considered to be $10^{16}-10^{17}/\mathrm{cm}^3$, or $10^{14}/\mathrm{cm}^3$ (for a high-quality sample). The corresponding acceptor concentration per unit of area would be given by $N_A^S = LN_A$.

The electron (n_e) and hole (n_h) densities at the conduction and valence subbands, respectively, are shown in Fig. 1 as functions of the laser intensity, and for different choices of the temperature, well widths, and acceptor concentrations. At low T [see Figs. 1(a) and 1(c)], $n_h = n_e$, since essentially no acceptors are ionized [cf. Eqs. (2.14) and (2.15)], and for $N_A \simeq 10^{16}-10^{17}/\mathrm{cm}^3$, one clearly sees that the dependence of the carrier density on the laser intensity undergoes a transition from linear (in the low-intensity regime) to square root (as the laser intensity increases), and to linear again (in the high-intensity regime). This low-temperature behavior was already pointed out by Mahan and Oliveira:[8] at very low intensities, i.e., $n_e/n_0 \ll 1$, the densities of electrons and free holes are very small, and most of the holes are bound to acceptors, and therefore electrons essentially recombine with bound holes, the carrier density being given by equating $\omega_A = \omega_{cA} \approx n_e(N_A^s/n_0)$, i.e., the density of free electrons (and holes) is proportional to the laser intensity. In this regime, any acceptors that are ionized by recombining with conduction electrons are quickly neutralized by the capture of free holes; also, it is apparent that increasing the concentration N_A of acceptors leads to a reduction in the carrier density. At intermediate laser intensities, when $n_e > N_A^S$, recombination with free holes according to Maxwell-Boltzmann statistics becomes dominant, and the carrier density is given by $\omega_A = \omega_{cv} \approx n_e(n_h/n_0)$, with $n_e \approx n_h$, and the carrier density exhibits a square-root behavior with the laser intensity. At high laser intensities, i.e., $n_e/n_0 \gg 1$, the electrons recombine with free holes, but one uses Fermi-Dirac statistics for the transition rate, $\omega_{cv} \approx n_e$, and the density of free electrons (and holes) is again proportional to the laser intensity.

For $T = 300$ K [see Figs. 1(b) and 1(d)] and low laser intensities, the electron density is very small, there are practically no bound holes at acceptors [note that in this regime and for the acceptor concentrations used, $n_h/n_0 \ll 1$; cf. Eq. (2.14)], and therefore the hole density n_h essentially equals the number N_A^S of acceptors per unit of area (i.e., n_h does not vary approximately with the laser intensity). In this regime, electrons essentially recombine with free holes according to Maxwell-Boltzmann statistics, n_e is given by $\omega_A = \omega_{cv} \approx n_e(N_A^S/n_0)$, and the density of free electrons presents a linear dependence on the laser intensity. Notice that, in this regime, although the linear dependence with the laser intensity is the same as in the low-T case, the recombination mechanism is different. For $T = 300$ K, taking into account the fact that $n_0 \approx 10^{12}/\mathrm{cm}^2$ (and $I_0 \approx 10^4$ W/cm^2) is much larger than for $T = 2$ K [cf. Eqs. (2.10b) and (2.10e)], the behavior at intermediate laser intensities is essentially the same as in the low-temperature case, and therefore one also observes a transition from linear to square-root dependence with intensity. For high laser intensities, there are essentially no ionized acceptors [notice that in this regime $n_e \approx n_h$, $n_h/n_0 \gg 1$, and holes bound to acceptors equilibrate with free holes despite the large temperature; cf. Eq. (2.14)], and electrons recombine with both bound and free holes, although again any acceptors which are ionized by recombining with conduction electrons are quickly neutralized by the capture of free holes; in this case, the carrier density is much larger than the density of acceptors, the main channel of recombination is with free holes, and follows Fermi-Dirac statistics. For $T = 300$ K, this last regime can be reached for very high laser intensities (such that $n_e/n_0 \gg 1$) and is not shown in Figs. 1(b) and 1(d); we shall discuss this regime again below. Also, it is worth noting that our results depends only weakly on the QW width through the matrix elements.

Figure 2 displays the dependence on the laser intensity of the electron and hole quasi-Fermi-levels (or chemical potentials) for different choices of well widths, and acceptor concentrations at low ($T = 2$ K) and high ($T = 300$ K) temperatures. The electron (μ_e) and hole (μ_h) quasi-Fermi-levels are calculated via Eqs. (2.10c) and (2.10d), i.e.,

$$\mu_e = k_B T \ln[\exp(n_e/n_0) - 1] \ , \quad (3.2a)$$

$$\mu_h = k_B T \ln[\exp(\gamma n_h/n_0)-1]\,, \tag{3.2b}$$

and their variation with the laser-intensity may readily be obtained through the dependence (on I) of the carrier densities previously discussed. For $T=2$ K, and in the low-laser-intensity regime ($n_e/n_0 \ll 1$), one therefore has $\mu_e = k_B T \ln(I/I_a)$, where I_a is a constant (for $N_A \neq 0$; n_e varies linearly with I), and $\mu_e = (k_B T/2)\ln(I/I_b)$, where I_b is another constant (for $N_A = 0$; n_e has a square-root dependence on I), with similar expressions for μ_h. This behavior is clearly seen in the curves of Figs. 2(a) and 2(c), where the change of slope by a factor of 2 is apparent when one goes from $N_A = 0$ to $10^{17}/\text{cm}^3$, or when one goes from low to intermediate laser intensities for doped QW's. For high laser intensities ($n_e/n_0 \gg 1$), $\mu_e = k_B T(n_e/n_0) \approx \exp[\ln(I/I_c)]$, where I_c is a constant and the quasi-Fermi-level increases exponentially (similar results are valid for μ_h). Of course, the electron chemical potential μ_e is always larger than the hole chemical potential μ_h due to the differences between the electron and hole effective masses.

In the high-temperature regime [$T=300$ K; see Figs. 2(b) and 2(d)], and for undoped QW's and low (and intermediate) laser intensities, $n_e = n_h$, $n_e/n_0 \ll 1$, and $\mu_e = (k_B T/2)\ln(I/I_b)$, with a similar result for μ_h, whereas for doped QW's, as n_e is proportional to I, $\mu_e = k_B T \ln(I/I_a)$, and n_h is practically constant with the hole quasi-Fermi-level essentially unchanged as the laser intensity increases. In the intermediate-laser-intensity regime, results for doped QW's are essentially the same for $N_A = 0$ as for $n_e \approx n_h$, with a square-root dependence on the laser intensity, and therefore $\mu_e \approx (k_B T/2)\ln(I/I_b)$,

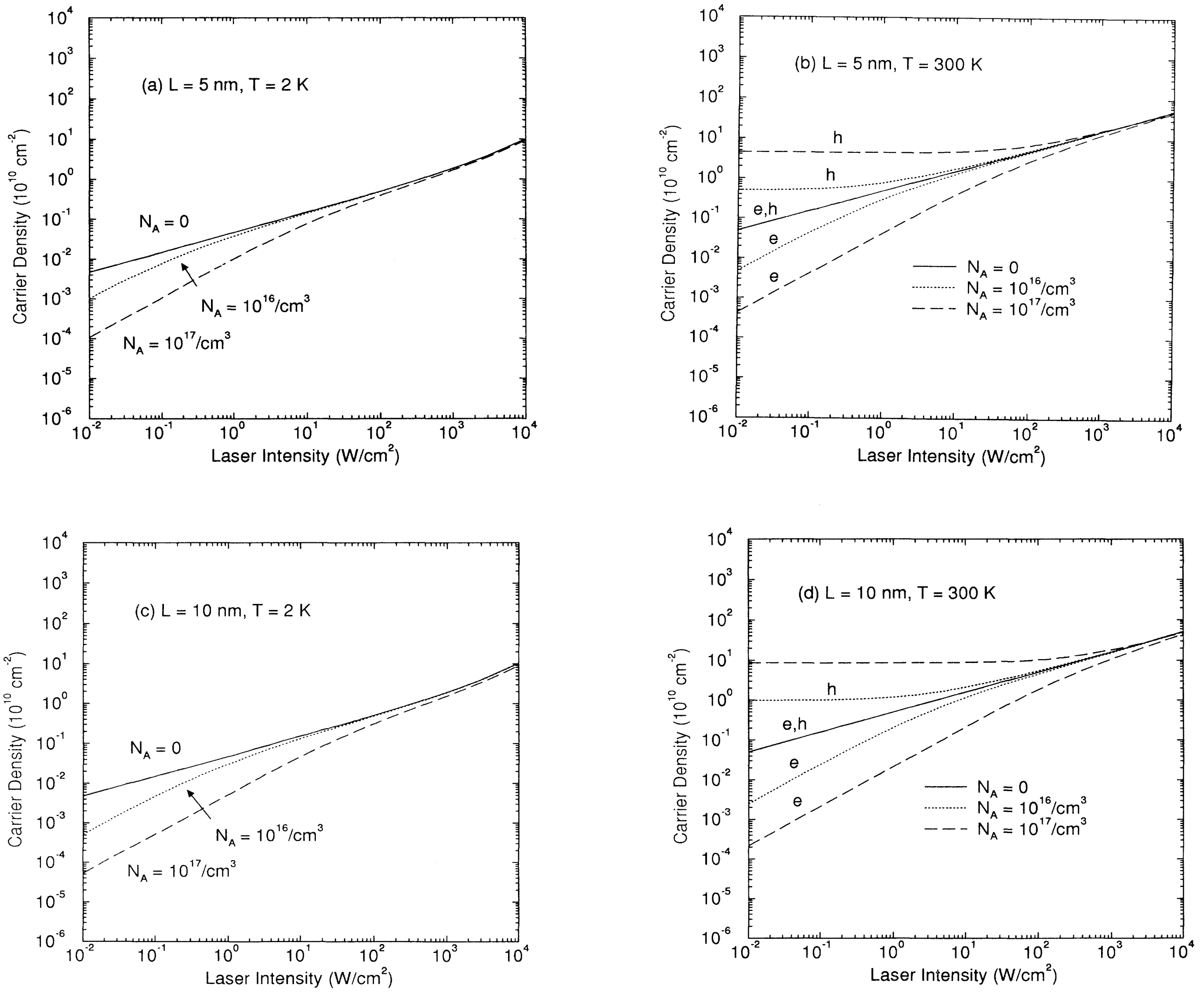

FIG. 1. Electron (hole) density for the $n=1$ conduction (valence) subband as a function of laser ($\hbar\omega = 1.75$ eV) intensity for a GaAs-Ga$_{0.7}$Al$_{0.3}$As QW with various homogeneous distributions N_A of acceptor impurities inside the QW. Results are presented for two well sizes $L=5$ [(a) and (b)] and 10 nm [(c) and (d)] and in the low- ($T=2$ K) and high- ($T=300$ K) temperature regimes. Curves for the electron and hole densities at $T=2$ K are essentially the same.

with a corresponding result for μ_h.

The total *e-h* radiative recombination time ($T_{\rm tot}$), and the *e-h* radiative recombination times with free holes ($T_{\rm cv}$) and bound holes (T_{cA}) are given as functions of the laser intensity (Fig. 3) and electron density (Fig. 4). The total *e-h* recombination time was calculated via Eq. (2.16), i.e., $T_{\rm tot}=T_{\rm cv}T_{cA}/(T_{\rm cv}+T_{cA})$. As $T_{\rm cv}=n_e/\omega_{\rm cv}$ and $T_{cA}=n_e/\omega_{cA}$, for $T=2$ K and low laser intensities ($n_e/n_0 \ll 1$), $T_{\rm cv}\approx 1/n_e$ [cf. Figs. 4(a) and 4(c)], and therefore $T_{\rm cv}\approx 1/I$ for doped QW's and $\approx 1/I^{1/2}$ for $N_A=0$ [cf. Figs. 3(a) and 3(c)], whereas T_{cA} (for $N_A\neq 0$) is essentially constant (there are no ionized acceptors and $\omega_{cA}\approx n_e$) and inversely proportional to the number of acceptors per unit of area N_A^S for all laser intensities. For high laser intensities ($I \gg 300$ W/cm^2; $n_e/n_0 \gg 1$), the conduction-electron gas is degenerate, $\omega_{\rm cv}\approx n_e$, and $T_{\rm cv}$ tends to a limiting value. For $T=300$ K, and low laser intensities (cf. Figs. 3 and 4), the behavior of the electron–free-hole recombination time is $T_{\rm cv}\approx 1/n_e$ for undoped QW's, the same as in the low-temperature regime, whereas $T_{\rm cv}$ is essentially constant for acceptor-doped samples, due to the fact that there are practically no bound holes at acceptors, and the number of holes at the valence subband is independent of the laser intensity. In this regime, as $\omega_{cA}\approx n_e$, then T_{cA} is essentially constant. In the high-temperature case, as the laser intensity increases, one reaches the regime (for intermediate laser intensities, i.e., $I\approx 10^3$–10^4 W/cm^2) for which $n_e\approx n_h$ (see Fig. 1), and therefore the result for electron–free-hole recombination time $T_{\rm cv}$ for doped samples tends to be the same as values obtained for undoped QW's (cf. Figs. 3 and 4). For intermediate laser intensities and $T=300$ K, the electron–bound-hole recombination time T_{cA} decreases with laser intensities as $\omega_{cA}\approx N_A^S n_e n_h$, and T_{cA} is inversely proportional to the product $N_A^S n_h$. Notice that this intermediate-laser-intensity regime occurs roughly for laser intensities such that $N_A^S < n_e < n_0$, and therefore it depends on the temperature and concentra-

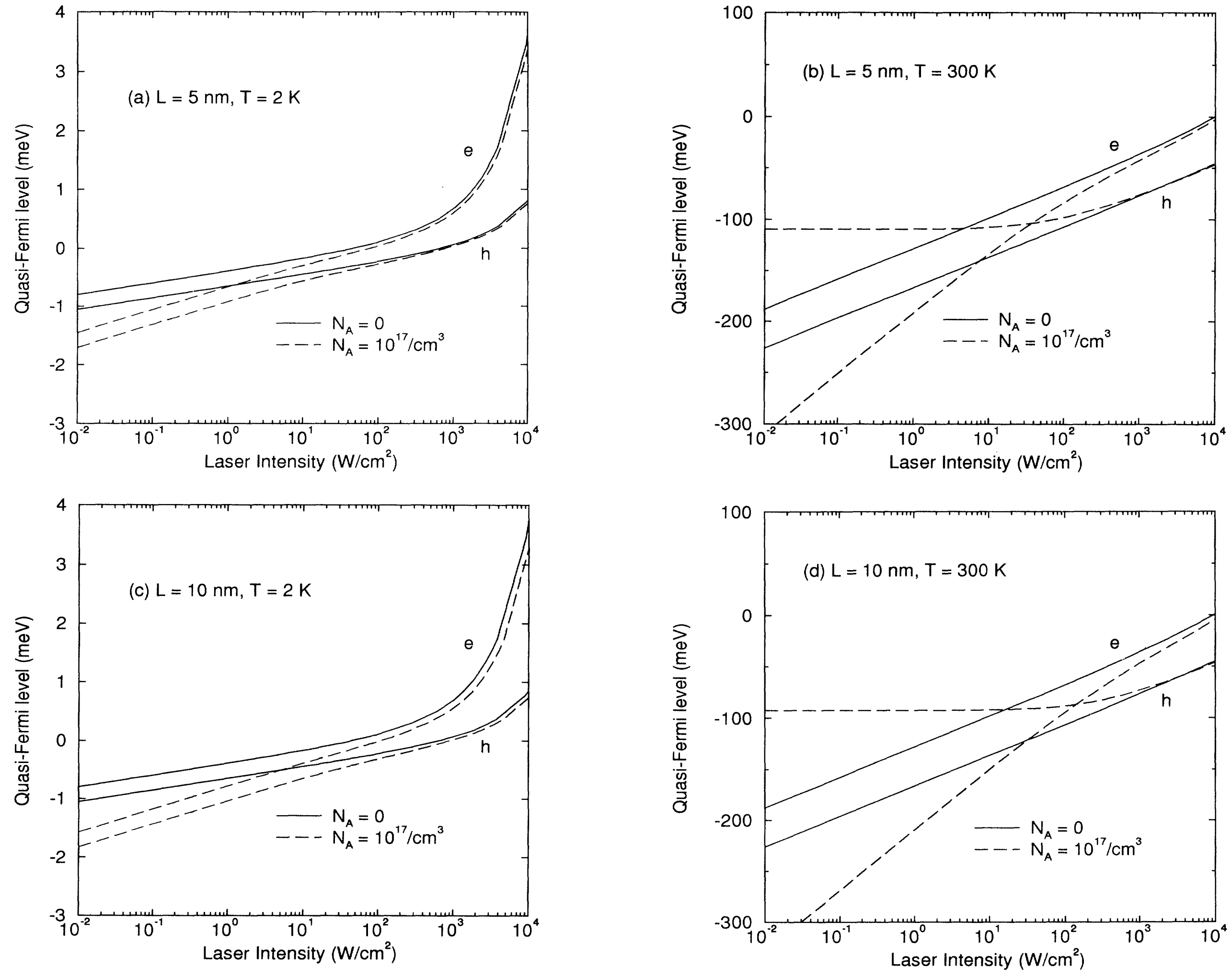

FIG. 2. Electron (hole) quasi-Fermi-levels for the $n=1$ conduction (valence) subband as a function of laser ($\hbar\omega=1.75$ eV) intensity for a GaAs-Ga$_{0.7}$Al$_{0.3}$As QW in the undoped case (full curves) and for a homogeneous distribution $N_A=10^{17}/{\rm cm}^3$ of acceptor impurities inside the QW (dashed lines). Results are presented for two well sizes $L=5$ [(a) and (b)] and 10 nm [(c) and (d)] and in the low- ($T=2$ K) and high- ($T=300$ K) temperature regimes.

tion of acceptors. For high temperatures, as the laser intensity further increases, the conduction-subband dispersion $\varepsilon_c(k_\perp)$ and $K(z_i,\lambda,k_\perp)$ [see Eqs. (2.11)–(2.13)] dependences on $k_\perp$ become important in the evaluation of ω_{cA} due to the recombination of large wave-vector electrons such as $n_e/n_0 \gg 1$. In this case, one may show that the rate per unit of area of radiative recombination of electrons with bound holes at acceptors, ω_{cA}, is essentially independent of the laser intensity (and of n_e), and therefore $T_{cA} \approx n_e$, as clearly seen in Fig. 5(b) for $N_A = 10^{14}$ and $10^{17}/\mathrm{cm}^3$. The importance of a proper description of the conduction-subband dispersion $\varepsilon_c(k_\perp)$ in the calculation of the radiative recombination of electrons with free holes ω_{cv} is also clearly seen in Fig. 5(c) by comparing the full curve for the total decay time T_{cv} with the dotted-dashed curve which was calculated by ignoring the dispersion with $k_\perp$ in the linear term of Eq. (2.10a). The results in Fig. 5 are for $T = 155$ K, and an $L = 122$ Å GaAs-Ga$_{0.77}$Al$_{0.23}$As QW, appropriate for calculating the *e-h* total recombination lifetime, and comparing with experimental results by Bongiovanni and Staehli[6] for a high-quality quantum structure consisting of six 122-Å-wide wells of GaAs separated by 180-Å-wide barriers of a Ga$_{0.77}$Al$_{0.23}$As alloy. Notice that our theoretical results for the total radiative recombination lifetime are in good quantitative agreement with the experimental data,[6] whereas the calculated results ignoring the linear term of Eq. (2.10a) lead to a saturated value of the recombination lifetime, a behavior also obtained in the calculation by Bongiovanni and Staehli.[6] In this regime (high temperature and laser intensities, and $n_e/n_0 \gg 1$), the main *e-h* recombination channel is with free holes, and one must use Fermi-Dirac statistics, and $\omega_{\mathrm{cv}} \approx (n_e/n_0) + \alpha(n_e/n_0)^2$, where $\alpha \approx k_B T/2\varepsilon_g$, and $T_{\mathrm{cv}} \approx 1/[1 + \alpha(n_e/n_0)]$.

In Fig. 6, the density of electrons (per unit of volume) in the conduction subband is shown as a function of the cw laser intensity for an $L = 6.5$ nm GaAs-Ga$_{0.6}$Al$_{0.4}$As

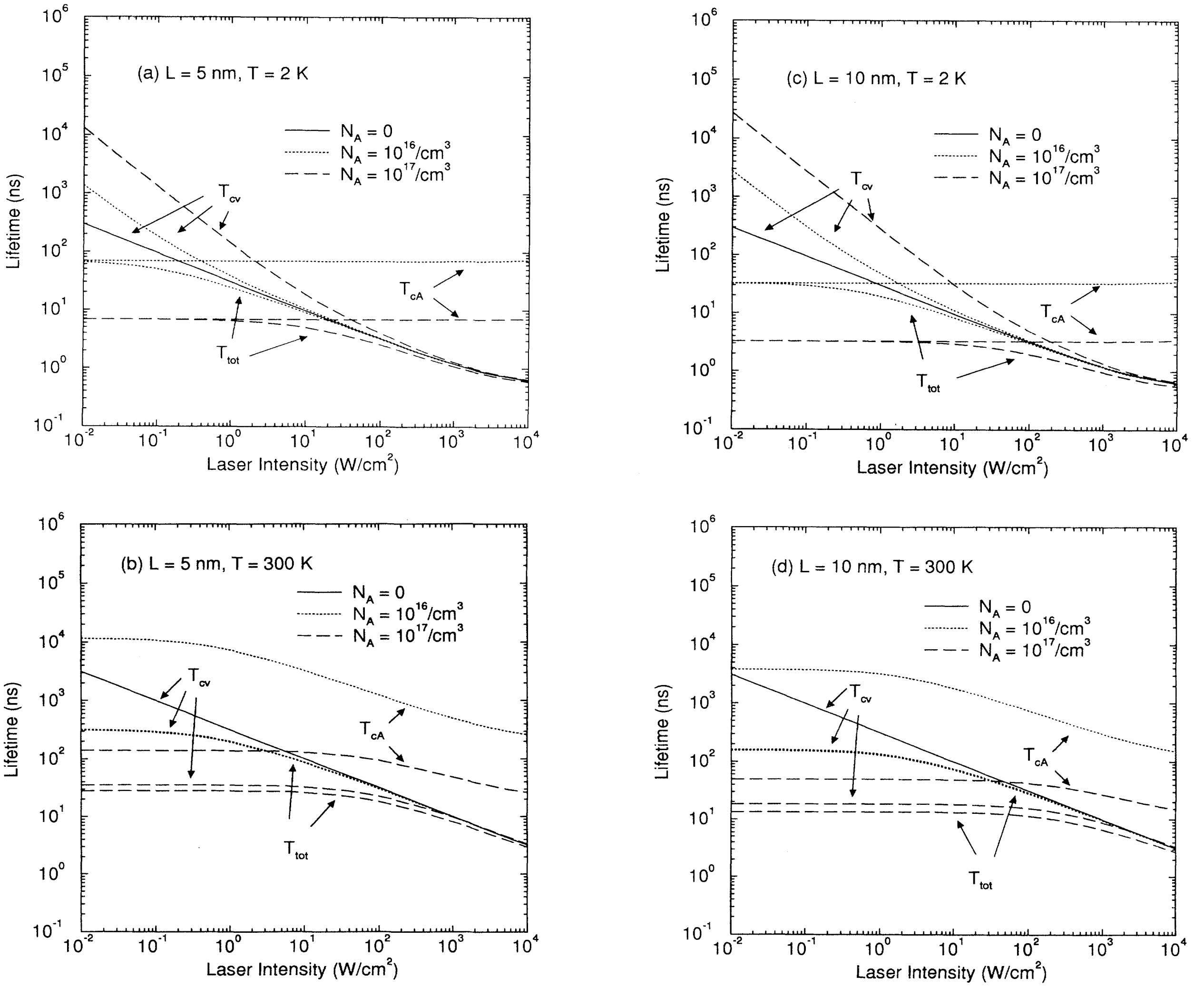

FIG. 3. Laser-intensity dependence of radiative lifetimes for *e-h* pair recombination for a GaAs-Ga$_{0.7}$Al$_{0.3}$As QW with various homogeneous distributions N_A of acceptor impurities inside the QW. The lifetime of the electron radiative recombination with free holes is denoted by T_{cv}, that with holes bound at neutral acceptors by T_{cA}, and the total radiative lifetime by T_{tot}. Results are presented for two well sizes $L = 5$ and 10 nm and in the low- ($T = 2$ K) and high- ($T = 300$ K) temperature regimes.

QW at $T=300$ K. We performed the calculation for both undoped and doped ($N_A=10^{16}/\text{cm}^3$) QW's. The widths of the GaAs QW and the concentration of acceptor impurities were chosen in order to model the QW structure studied by Ding *et al.*;[7] of course, as they have studied undoped *multiple narrow* ACQW's, and we are considering a *single isolated* QW, our results should be compared to theirs in a qualitative way. One clearly sees from Fig. 6 that by considering a concentration of acceptor impurities one has more radiative decay channels, which leads to a decrease in the steady-state electron density in the conduction subband. Also, it is apparent that, for $N_A=10^{16}/\text{cm}^3$, the dependence on laser intensity undergoes a clear transition from linear to square root. For very low laser intensities, where the density of electrons is very small, most of the acceptors are ionized, and electrons in the conduction subband emit photons by recombining with free holes: the electron density at the conduction subband is essentially given by equating $\omega_A=\omega_{\text{cv}}$ with ω_{cv} proportional to n_e (and to $n_h \approx N_A$), and therefore the carrier density is proportional to the laser intensity. When the laser intensity increases (with a corresponding increase in the free-electron and hole densities), the steady-state carrier density in the conduction subband becomes proportional to the square root of the laser intensity, as $n_h \approx n_e$. One should notice that our calculation for $N_A=10^{16}/\text{cm}^3$ compares well with the experimental results of Ding *et al.*[7] in the intermediate-laser-intensity regime ($I > 10^2$ W/cm^2). Although at very low laser intensities, we found the same carrier-density linear

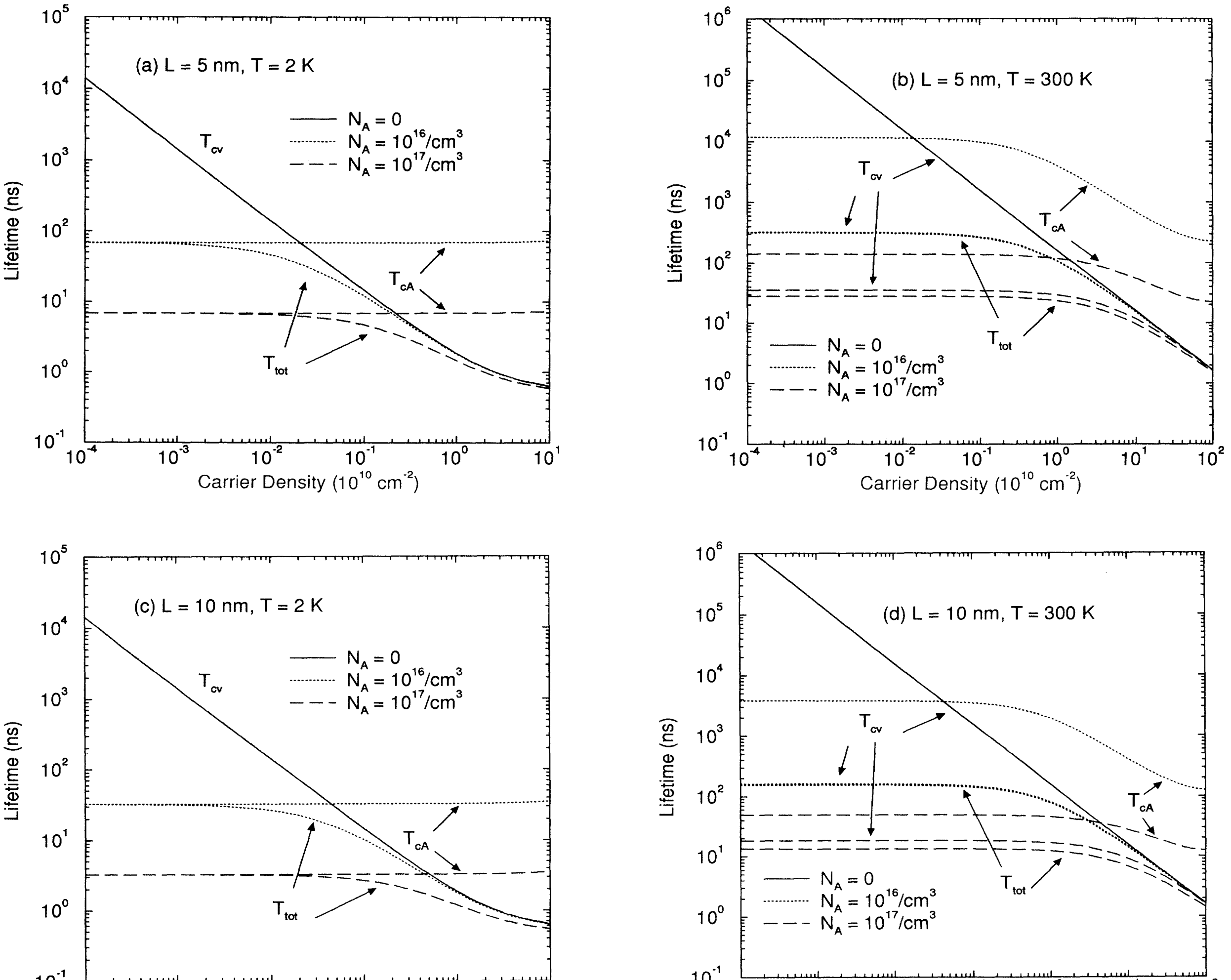

FIG. 4. Electron-density dependence of radiative lifetimes for *e-h* pair recombination for a GaAs-Ga$_{0.7}$Al$_{0.3}$As QW with various homogeneous distributions N_A of acceptor impurities inside the QW. The lifetime of the electron radiative recombination with free holes is denoted by T_{cv}, that with holes bound at neutral acceptors by T_{cA}, and the total radiative lifetime by T_{tot}. Results are presented for two well sizes $L=5$ and 10 nm, and in the low- ($T=2$ K) and high- ($T=300$ K) temperature regimes.

dependence on intensity due to radiative recombination of electrons with free holes influenced by the presence of ionized acceptors (notice that several nominally undoped GaAs-(Ga,Al)As QW samples[10] have presented extrinsic photoluminescence due to carbon acceptors), in the work of Ding *et al.*[7] recombination mechanism is dominated by nonradiative recombination of free carriers at nearly saturated interface traps. As argued by Ding *et al.*,[7] this mechanism is dominant in the low-laser-intensity regime and is responsible for the dependence of the *photoluminescence intensity* on the laser intensity undergoing a transition from square-law to linear dependence as the laser intensity increases. One should point out that in the case of Ding *et al.*,[7] one would not observe the acceptor extrinsic photoluminescence feature at $T=300$ K, as most of the acceptors are ionized at the laser intensities considered ($I<135$ W/cm^2).

In the discussion of our results for carrier densities, radiative lifetimes, and quasi-Fermi-levels, excitonic processes were not considered, although excitonic recombination should be important at low temperatures. According to Gurioli *et al.*,[5] at low temperatures it is important to consider the recombination process involving excitons localized at crystal defects. Pickin and David,[21] Ridley,[22] and Ping,[23] among others, have modeled the exciton-formation process via a recombination term of electrons and holes proportional to their joint density, or $\omega_x \approx n_e n_h$. Bastard[15] has discussed the relative importance, at low temperatures and with increasing laser intensity, of the electron recombination with bound holes as compared with excitonic recombination. Accordingly,[5,21–23] when excitonic processes are dominant (with

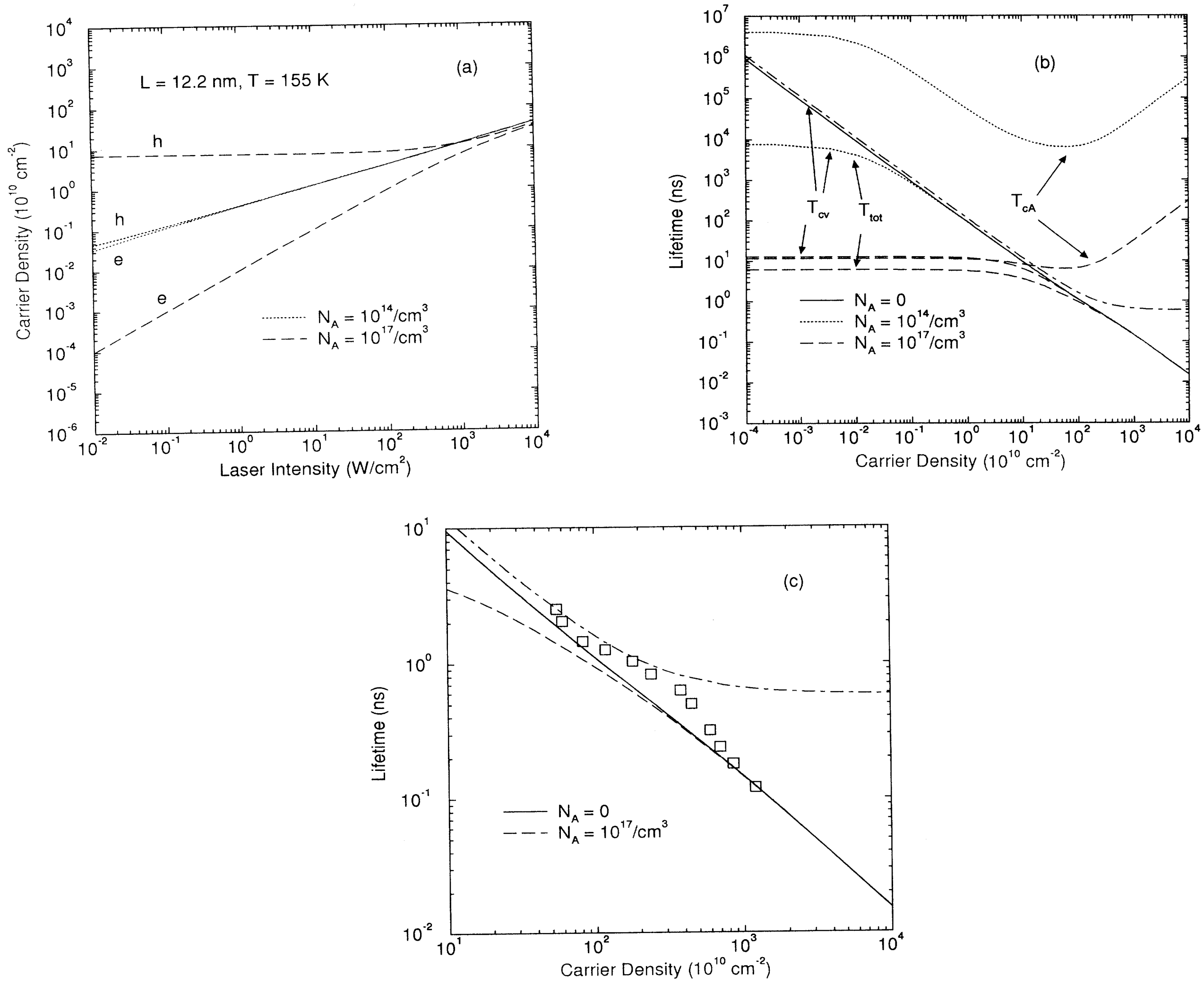

FIG. 5. Results for an $L=12.2$ nm GaAs-Ga$_{0.77}$Al$_{0.23}$As QW with various homogeneous distributions N_A of acceptor impurities inside the QW: (a) electron (hole) density vs laser intensity; (b) electron-density dependence of radiative lifetimes for *e-h* pair recombination. The lifetime of the electron radiative recombination with free (bound) holes is denoted by T_{cv} (T_{cA}), and the total radiative lifetime by T_{tot}; and (c) total radiative lifetimes vs electron density, with experimental results by Bongiovani and Staehli (Ref. 6) represented by squares, and dotted-dashed curve corresponding to theoretical lifetime results calculated for an undoped QW by setting to zero the linear term in Eq. (2.10a). All results are shown at $T=155$ K.

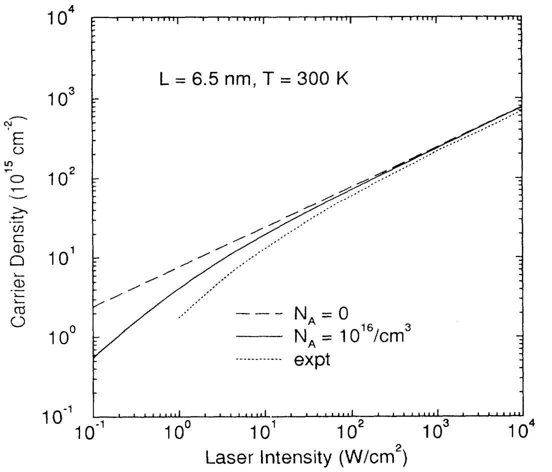

FIG. 6. Electron density at $T=300$ K for the $n=1$ conduction subband as a function of laser intensity for an $L=6.5$ nm GaAs-$Ga_{0.6}Al_{0.4}As$ QW in the undoped case (dashed curve) and for an homogeneous distribution $N_A=10^{16}/cm^3$ of acceptor impurities inside the QW (full line). The dotted curve corresponds to the experimental results by Ding *et al.* (Ref. 7). Note that the electron density is now given in cm^{-3}.

low concentration of impurities, low temperatures, and not too high optical excitation conditions), the behavior of laser intensity with carrier densities, radiative lifetimes, and quasi-Fermi-levels calculated in this work may be substantially affected (in this regime, for instance, the carrier density should behave as the square root of the laser intensity). Therefore, we would like to emphasize that as we have not taken into account excitonic processes, the results of this work in the low-temperature range and for low laser intensities ($I \ll I_0$) should be viewed with caution, since in this regime excitonic recombination may be important.[5,15,20–23] In the high-laser-intensity regime, the screening of the Coulomb interaction leads to exciton ionization, and excitonic processes may be neglected.[2]

IV. CONCLUSIONS

Summing up, quantum-mechanical calculations of the carrier densities, electron and hole quasi-Fermi-levels, and various radiative decay times in GaAs-(Ga,Al)As quantum wells is performed, under steady-state excitation conditions, as functions of the cw-laser intensity, temperature, well widths, and acceptor distribution in the well. We consider radiative recombination of electrons with free holes and with holes bound at neutral acceptors. To our knowledge, this is the first calculation which considers the effects of temperature and of the acceptor distribution in the well. Results for the carrier-dependent *e-h* recombination decay time are in good agreement with experimental data from Bongiovanni and Staehli.[6] Our calculations—which have no free parameters—in the intermediate-laser-intensity regime are in quantitative agreement at $T=300$ K with the results by Ding *et al.*,[7] who obtained the carrier density for multiple ACQW's through a fitting procedure which reproduced the total experimental PL intensity. For low temperatures, there are no experimental results to compare with our calculations, which show three different behaviors of the intensity dependence of the carrier density for $N_A \approx 10^{16}/cm^3$ (although these results should be viewed with caution, as no excitonic recombination processes are included in our calculation).

ACKNOWLEDGMENT

M.D.L. and L.E.O. acknowledge support by Brazilian Agencies CNPq, FAPESP, and FAEP-UNICAMP.

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