# Theory of well-width-dependent periodic variation in photoluminescence from $Al_xGa_{1-x}As/GaAs$ quantum wells

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The recently discovered periodic variation in photoluminescence (PL) efficiency is theoretically analyzed. The PL intensity $I_{PL}$ depends on the excitation intensity $I_{ex}$, in that it varies as a function of the width of the $Al_{0.3}Ga_{0.7}As/GaAs$ quantum well (QW), following a power law, i.e., $I_{PL} \propto I_{ex}^m$. The value $m$ was observed to vary periodically between 1 and 2 depending on width. Analysis makes it possible to correlate the $m=1$ and 2 cases to off-resonance and resonance, respectively, which occur between the highest QW level and the bottom of the conduction band of the barrier. In an off-resonance electrons are tightly trapped in the QW bottom after rapid relaxation; resonating electrons are free to escape from the well, if they are not recombined. Microscopic calculation of the relaxation of hot electrons reproduces the observed behavior of $m$ quite well.

## I. INTRODUCTION

Recent advances in MBE (molecular-beam epitaxy) technology have resulted in obtaining precisely controlled multilayered devices, working as a powerful means for investigating size-quantized energy levels, as well as energy transfer and relaxation schemes among them. Among the many investigations reported to date, it is quite important to view a periodic variation of the photoluminescence (PL) mode as a function of the relative position of the quantum level of concern to the quantum state into which carriers are first photoexcited. This new phenomenon was first reported by Mishima *et al.*[1] This phenomenon is called the QWIDDLE [quantum-well-width-dependent (photo) luminescence efficiency] effect.

It is well known that in tunneling spectroscopic study[2–4] an increase in tunneling probability is always observed when an applied voltage causes electrons to resonate between energy levels on both sides of the barrier. However, observed physical quantities should be the combined outcome of electron transfer and relaxation processes down to a stable state; in other words, resonantly transferred electrons are equally easy to back transfer, when they are unrelaxed.

From this viewpoint, electrons transferred from the photoexcited barrier states into a quantum well (QW) are one of the best tools for investigating the relation between resonance transfer and relaxation. Thus, the above-mentioned phenomenon is a good example of the fourth scheme for understanding relaxation processes precisely, in addition to tunneling spectroscopy and the hot carrier relaxation experiment within a single band, whether they are due to phonon emission[5] or interband photoemission.[6–10]

## II. SUMMARY OF EXPERIMENTAL RESULTS

The details of the experiment to be analyzed are well described in Ref. 1. Only its outline is described here. Various samples were prepared by MBE with dispersed well widths from 1.5 to 16 nm. The PL was measured at 77 K using an $Ar^+$ laser, whose wavelength is short enough to produce carriers within the $Al_xGa_{1-x}As$ barrier. The experiment performed at 4.2 K also showed similar behavior. The data for $x=0.3$ and at $T=77$ K are mainly discussed here.

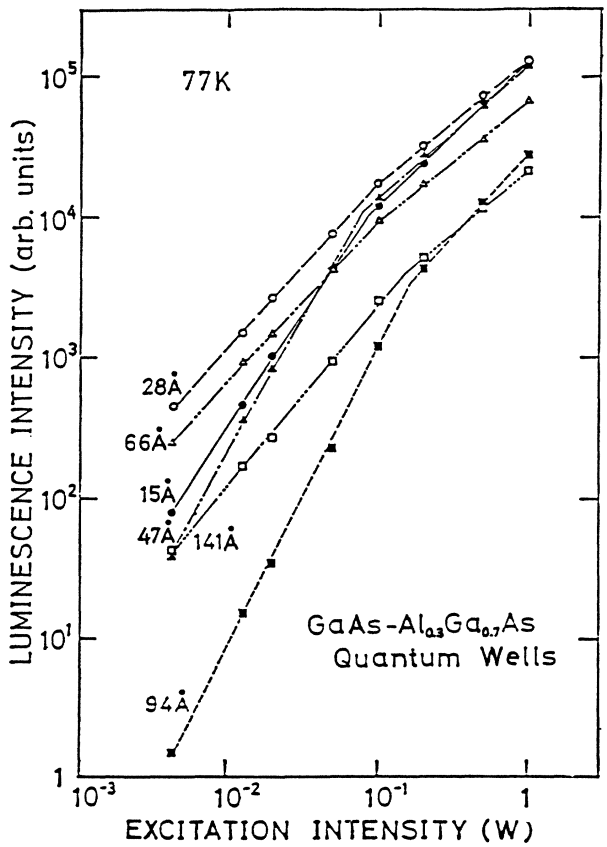


FIG. 1. PL intensity $I_{PL}$ versus excitation intensity $I_{ex}$ at 77 K. The parameters are well widths. From Ref. 1.

 

The observed PL intensity is highly dependent on well width. When the logarithm of $I_{PL}$ is plotted against the logarithm of excitation energy $I_{ex}$, as shown in Fig. 1, it is easy to derive a specific power index $m$ for each sample. The intensity region of interest is at rather low excitation levels. This is because no typical QWIDDLE effect is seen for much higher levels. The variation in $m$ is depicted in Fig. 2(a) by various symbols; it is seen that $m$ varies between 1 and 2 as a function of well width.

It should be stressed that the wavelength of PL peaks corresponded to the separation between the lowest electronic quantum level and the hole level, and the energy has been assigned to free exciton recombination, since GaAs QW's are prepared without intentional doping. As is seen in Fig. 2(b), no drastic change in PL energy occurred at the critical well widths ($L_z$'s), where $m$ jumped from 1 to 2. The first jump occurred at a width around 4.7 nm. Based on lack of data points around the width and ambiguity in determining widths, it is safe to assume the critical value to exist within $(17.5\pm0.5)a_0=4.76$ to 5.04 nm. $a_0=0.28$ nm is the monolayer thickness.

Seeing the experimental results, the periodic variation in $m$ seems to stem from the periodic appearance of the highest quantum level coincident with the bottom of the barrier conduction band, i.e., with the top of the potential wall. Based on the effective-mass equation the critical thicknesses of the well occur at $L_z$'s defined by[11,12]

$$V_0/(2\hbar^2/m_w L_z^2)=[(n-1)\pi/2]^2, \quad n=1,2,3,\ldots . \tag{1}$$

$n$ is the quantum number for specifying each subband. It is worth noting that for $L_z=17a_0$ and $18a_0$ the barrier height $V_0$ is estimated to be 250 and 223 meV, respectively, if the effective mass in the GaAs well, $m_w$, is assumed to be $0.0665m_0$. Against the overall band-gap offset equal to 374 meV for $x=0.3$, the relative conduction-band offset becomes 66.8% and 59.6%, respectively. Thus estimated values agree well with those reported to date.[13–15]

The variation means that $m=2$ corresponds to a resonant condition, where PL efficiency is low. It is on this very point that the experiment has a unique significance. The relation of resonance to off-resonance looks different from that in tunneling spectroscopy. It is due to the interrelation between relaxation and transfer.

A fraction of this phenomenon had been observed by Shichijo *et al.*[16] They noted that under a critical thickness no PL from the well was observed except from the barrier. In order to explain the existence of such critical width, a couple of theoretical studies appeared,[17,18] the latter of which is scrutinized in the next section.

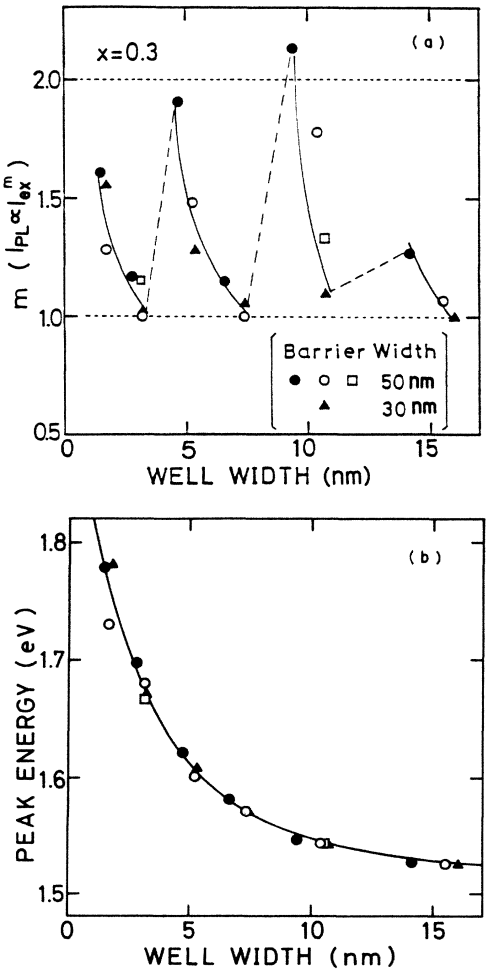


FIG. 2. (a) Observed $m$ values versus well width. $m$'s are obtained in plots of $I_{PL} \propto I_{ex}^m$. The curve is only a guide for the eye. (b) Observed photon energy versus well width. Both (a) and (b) are from Ref. 1.

## III. MODEL

We start from our model shown in Fig. 3, illustrating a variety of processes to consider. It describes relaxing schemes of electrons in the barrier (their density is denoted as $n_b$), in the highest subband ($n_w$), and in the lowest

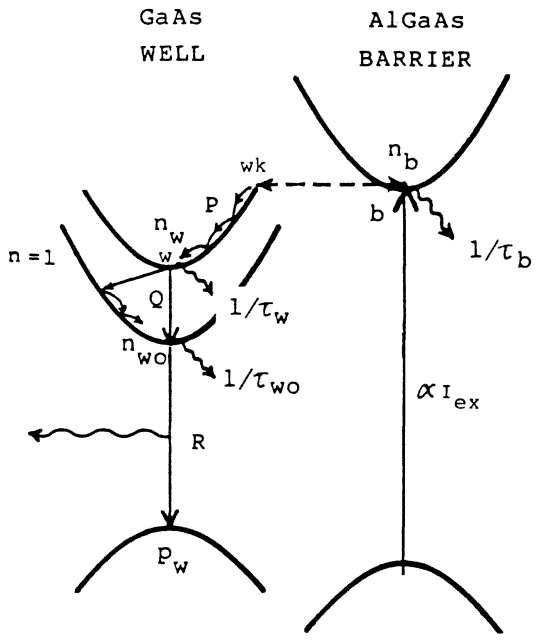


FIG. 3. Figure explaining the present model. The relation of QW levels to barrier band and relaxation schemes is denoted.

subband ($n_{w0}$), along with the recombination of $n_{w0}$ with holes ($p_w$). Electrons are generated by photoexcitation at a rate of $\alpha I_{ex}$, only within the barrier. Electronic states in the QW are quantized as shown schematically in Fig. 3. If holes are similarly quantized, the same processes should be considered for them as well. The horizontal axis denotes the wave vector *parallel* to the well, $k_{||}$.

Bastard[18] discussed the transmission probability of electrons in the barrier against the QW, taking the system simply as one dimensional and considering only the momentum $k_{b\perp}$ in the direction perpendicular to the QW.

Our viewpoint differs. Even if there exists a well, the $k$ vector parallel to the well still remains a good quantum number, except for a constant-energy offset between the in and out sides of the QW. Accordingly, it cannot be discerned if a wave vector, $k_{||}$, in this direction is in the well or in the barrier. Thus, the points $b$ and $wk$ in Fig. 3 should be correlated in the sense that they are two crystal states, having the same energy and subject to the same quantization scheme.

Moreover, the number of $k_{||}$ states has a larger measure due to its two dimensionality than that of one-dimensional $k_{w(b)\perp}$ states. A couple of nearly vanishing $k_{b||}$ states are considered to be equivocally transferable into two $k_{w||}$ states: $+k_{||}$ and $-k_{||}$ of the same energy. This is due to electron-electron correlation. Thus, the transfer mechanism seems to be rather insensitive to the magnitude of energy offset, or accordingly, of the $k_{||}$ vector. The time constant of concern must be sufficiently small. Thus overall probability is given by the product of transfer probability from $b$ to $wk$ and the relaxation rate from $wk$ to the bottom of the QW. That is, $P_{bw}=P_{bw_k}P_{w_kw}$. The partial probability of the transfer process from point $b$ to $wk$, $P_{bw_k}$, will not be explicitly taken into account hereafter; $P_{bw}$ is exclusively taken to be the probability of an electron relaxing from the $wk$ point to the highest subband bottom in the well $b$, due to *intra*subband transition processes. We assume, thus, that $P_{bw_k}=P_0$ is roughly constant, independent of the width of the QW. $P_{wb}$ is that for the reverse process.

This is contrasted to the processes considered by Bastard.[18] In his case, electrons in virtual states within the QW region cause an *inter*band transition down to real subbands. The transition should be *real* phonon emitting processes.

Göbel *et al.*[19] made time-resolved PL spectroscopy measurements at 10 K in a similar device. They concluded that the time delay seen on PL from the QW of the order of 50 to 100 ps is due to that needed for photoexcited electrons to drift from the barrier to the QW, by analyzing simple rate equations. However, in their analysis two probabilities are mixed. The time delay is not specific to the transfer process. The delay seems to rather correspond to our $P_{bw}N_w$ (as will be discussed soon), though ambient temperatures are different.

The rate of PL is $R$. $Q$ is the relaxation rate of an electron from the highest subband to the lowest; it has two channels, as shown in Fig. 3. One is spontaneous infrared (IR) photon emission, and the other an *inter*subband transition followed by multiple phonon emission. All $\tau$'s are nonradiative relaxation times, regardless of the microscopic mechanism causing them. One of the most plausible of such mechanisms will be carrier relaxation, through deep traps within the energy gap. Since the PL intensity is plotted in Fig. 1 with its wavelength fixed, any luminescence whose wavelength is outside of the main spectrum is counted as a *non*radiative relaxation process.

Based on the present model the following rate equations are derived for electrons:

$$\dot{n}_b=-n_b/\tau_b-P_{bw}n_b(N_w-n_w)+P_{wb}n_w(N_b-n_b)+\alpha I_{ex}\ , \tag{2}$$

$$\dot{n}_w=-n_w/\tau_w+P_{bw}n_b(N_w-n_w)-P_{wb}n_w(N_b-n_b)-Qn_w(N_{w0}-n_{w0})\ , \tag{3}$$

$$\dot{n}_{w0}=-n_{w0}/\tau_{w0}+Qn_w(N_{w0}-n_{w0})-Rn_{w0}p_w\ . \tag{4}$$

Exactly speaking, similar equations must be considered for holes. However, if the band offset between GaAs and $Al_xGa_{1-x}As$ valence bands is small enough, especially for $x=0.3$, quantized levels are close to each other, and hence a single three-dimensional (3D) bandlike approximation seems to be valid. This holds true more plausibly, since holes have a heavier effective mass. In case $x=0.5$, the situation is a little different, as will be discussed later.

So long as only a single subband is concerned, or when the well width is less than the first critical thickness around 4.7 nm, $-Rn_wp_w$ replaces $-Qn_w(N_{w0}-n_{w0})$ in Eq. (3), and Eq. (4) is no longer necessary.

All $N$'s in the equations are effective densities of states. $N_b$ is given by integrating 3D $N_b^{(3)}$ up to the thickness of the barrier. When the excitation intensity is not extraordinarily strong and electrons are not degenerate, Boltzmann distribution occurs. However, it seems necessary to take a kind of exclusion principle into account, which is expressed by the factor $(N_w-n_w)$, and so on, when the excitation level is sufficiently high.[20] This formulation is approximately, not rigorously, justified in the Appendix. By using Fermi-Dirac statistics it is hard to write down simple rate equations as listed above; we are satisfied with a semiclassical statistics. The criterion for this approach is that the solutions, $n_{w(b)}$, eventually obtained should be such that $n_{w(b)}<N_{w(b)}$.

A stationary solution of the rate equations is of concern throughout this work. For simplicity, the solution of the case with only a single subband $n=1$ is explained. Two extreme cases called resonant and off-resonant conditions are discussed. The former case meets the vanishing energy separation $\varepsilon_k$ between the $wk$ point and the bottom $b$ (see Fig. 3). Off-resonance means that there is considerable $\varepsilon_k$, which is to be relaxed by emitting phonons.

### A. Resonant condition

In this case it is permitted to assume that $P_{bw}\cong P_{wb}=P$, then

$$n_w\cong\alpha I_{ex}PN_w/[1/\tau_b\tau_w+P(N_b/\tau_b+N_w/\tau_w)] \tag{5}$$

is obtained, provided the PL term $Rn_wp_w$ is sufficiently smaller than the other terms.

Referring to the fact that Eq. (5) is proportional to $I_{ex}$, $p_w$ is hereafter assumed to be similarly proportional to $I_{ex}$ like $p_w = \beta I_{ex}$. This is justified because all hole levels are close to the valence-band top for $x = 0.3$; hence, resonance always occurs of holes to the barrier.

Thus $I_{PL} \propto n_w p_w$ is proportional to $I_{ex}^2$ and, hence, $m = 2$. In general, this dependence on $I_{ex}$ is rather easy to understand. However, the dependence holds true when and only when electrons and holes resonate to their corresponding states in the barrier.

### B. Off-resonant condition

In this case $P = P_{bw} \gg P_{wb}$ is assumed. Then it is easy to obtain

$$n_b \cong 2\alpha I_{ex}\beta R / | P\alpha - R\beta(1/\tau_b + PN_w) | \quad (6)$$

and

$$n_w \cong PN_w n_b / (Pn_b + R\beta I_{ex}) \propto I_{ex}^0 . \quad (7)$$

It is concluded that in this case $I_{PL}$ is proportional to $I_{ex}^1$, since $n_w p_w \propto I_{ex}^1$. The power index is 1. Under an off-resonant condition the density of electrons within the well is insensitive to the change in excitation level. The increase in hot carriers in the barrier, $n_b$, certainly induces the increase in $I_{PL}$, although electrons within the QW, $n_w$, remain almost constant, limited by the maximum effective density of states. Any increase in $I_{ex}$ does cause an increase in $n_b$, but not in $n_w$, because the increase is just compensated for by that in $I_{PL}$.

The efficiency of PL defined by $R\beta I_{ex} n_w / \alpha I_{ex}$ is given by an intrinsic efficiency, $\eta = R\beta N_w / \alpha$, times the ratio $n_w / N_w$. Therefore, PL is more efficient under an off-resonant condition. Efficiency and value $m$ vary in an inverted manner.

For much higher excitation the QW subband seems unable to accommodate electrons beyond saturation. However, this becomes permissible by boosting the electron temperature within the QW. This is plausible when so many electrons are poured into the well and, therefore, electron temperature rises,[20] since $N_w$ is proportional to the temperature.

As for a general case, neither resonant nor off-resonant, we microscopically analyze the energy relaxation processes from the transferred state into the subband bottom.

The power index $m$ is defined by $m = d\ln(I_{PL}) / d\ln I_{ex}$. After separating the contribution from holes,

$$m = 1 + (I_{ex}/n_w) dn_w / dI_{ex} .$$

Except for the single subband case, this $n_w$ should be replaced by $n_{w0}$. That is,

$$(I_{ex}/n_{w0}) dn_{w0}/dI_{ex} = [(I_{ex}/n_w) dn_w/dI_{ex} - R\beta I_{ex}\tau_w/(1 + R\beta I_{ex}\tau_w)]/(1 + Qn_w\tau_R) . \quad (8)$$

Here, $1/\tau_R = 1/\tau_w + R\beta I_{ex}$.

## IV. MICROSCOPIC PROCESSES

Let us describe the model of microscopic processes determining $P_{bw}$ and $P_{wb}$ in the rate equations. When a transferred electron has a kinetic energy $\varepsilon_k$ large enough to emit at least one optic phonon of the energy of $\hbar\omega_{op}$, the first process is optic phonon emission, because of its quite short relaxation time of the order of 0.1 ps.[21] Otherwise, only multiple acoustic phonons can be emitted. The inelastic feature of the processes is essential in order to let the hot electrons relax into the subband bottom. This is the case, especially when the system consists of a single subband.

Even when multiple subbands must be considered, an intersubband transition due to optic-phonon emission is only a small fraction of the whole probability,[22,23] so long as $\varepsilon_k \gtrsim \hbar\omega_{op}$. In the opposite case with $\varepsilon_k < \hbar\omega_{op}$, an intersubband transition emitting an optic phonon will occur more easily than an intrasubband acoustic phonon process. The present study is concerned with the intrasubband transition exclusively

Processes are described by a group of Dyson equations, where several optic phonons are first emitted followed by multiple acoustic phonons, using the Born approximation. The relaxation time is given by $(2\pi/\hbar)\mathrm{Im}\Sigma(k,\varepsilon_k)$, where $\Sigma$ is the self-energy of the corresponding diagram shown in Fig. 4; thus, $P_{bw}N_w = (2\pi/\hbar)\mathrm{Im}\Sigma$. $P_{wb}/P_{bw} = \exp(-\varepsilon_k/k_B T_e)$ was assumed based on the detailed balance requirement.

The Dyson equation of a typical diagram is $\Sigma_*(\mathbf{k},\varepsilon) = \Sigma_{q\omega} g_q^2 \mathscr{G}_{**}(\mathbf{k}-\mathbf{q},\varepsilon-\omega)\mathscr{D}_{***}(\mathbf{q},\omega)$, where $\mathscr{G}$ and $\mathscr{D}$ are the temperature Green's functions for electrons and phonons, respectively. $g_q^2$ is the electron-phonon coupling. For $\varepsilon_k$ less than the dispersionless optic-phonon energy, $\hbar\omega_{op}$, only acoustic-phonon emission

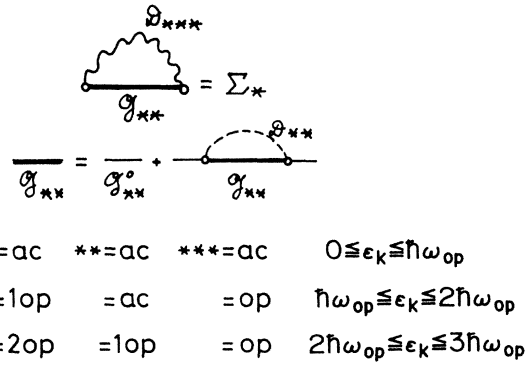


FIG. 4. Dyson equations to show microscopic relaxation processes. *, **, and *** stand for either the acoustic-phonon, one-optic-phonon, or two-optic-phonon nature of the propagator, according to the relative magnitude of $\varepsilon_k$ to $\hbar\omega_{op}$.

is possible within the same subband. Therefore, $\mathscr{G}_{**}$ should be the electronic Green's function with its self-energy determined by interaction with acoustic phonons. $\mathscr{D}_{***}$ is a Green's function for acoustic phonons in this case.

In this circumstance hot electrons with a finite $\mathbf{k}$ vector can also recombine with holes in band-to-band indirect transition. This will broaden the apparent linewidth. The transition is neglected here; the electrons are assumed to make only a direct transition from the bottom of the subband.

On the other hand, when $\varepsilon_k$ is larger than $\hbar\omega_{\rm op}$, an optic phonon is emitted. Hence, $\mathscr{D}_{***}$ is for an optic phonon, while $\mathscr{G}_{**}$ has its self-energy part, including all orders of acoustic-phonon interaction. For higher energies, the possible processes are similarly described; they are symbolically shown in Fig. 4.

The equations to be solved read[24]

$$\Sigma(\mathbf{k},\varepsilon)=-(2\pi)^{-4}\int d\mathbf{q}\int d\omega\{[\mathrm{Im}G_R(\mathbf{k}-\mathbf{q},\varepsilon-\hbar\omega)]D_R(\mathbf{q},\omega)\tanh[(\varepsilon-\hbar\omega)/2k_BT_{\rm ph}] + G_R(\mathbf{k}-\mathbf{q},\varepsilon-\hbar\omega)[\mathrm{Im}D_R(\mathbf{q},\omega)]\coth(\hbar\omega/2k_BT_{\rm ph})\}\ . \tag{9}$$

$G_R$ and $D_R$ are the retarded Green's functions for electron and phonon, respectively, corresponding to $\mathscr{G}$ and $\mathscr{D}$. The hot phonon effect is considered only through the probability of the phonons' occupation.

### A. In case $\varepsilon_k$ is such that $0\leqq\varepsilon_k\leqq\hbar\omega_{\rm op}$

After a tedious integration, $\Gamma_{ac}=\mathrm{Im}\Sigma_{ac}$ is given by

$$\Gamma_{ac}=(4\pi)^{-1}(m_w\Xi^2/\rho b_n)(m_w/\hbar^2\sqrt{\varepsilon_k})\pi/b\ , \tag{10}$$

where $b=(2m_ws^2)^{1/2}/k_BT_{\rm ph}$ and $\Xi=7$ eV is the deformation potential, $\rho=5.7$ g/cm$^3$ the 3D mass density, and $s=5.22\times10^5$ cm/s the velocity of sound in GaAs.[25] The value $b_n$ is the span of the localized electron probability in a direction perpendicular to the well, introduced by Price.[26] The definition is $\pi/b_n=\int|I_n(q)|^2dq$, in terms of the Fourier transform $I_n(q)$ of the probability density of the wave function.

### B. In case $\varepsilon_k$ is such that $\hbar\omega_{\rm op}\leqq\varepsilon_k\leqq2\hbar\omega_{\rm op}$

The transferred electron first emits an optic phonon and subsequently multiple acoustic phonons. In such a case it is difficult to perform an integration analytically. It suffices to numerically integrate

$$\Gamma_{1\rm op}(k,\varepsilon_k)=(\gamma/4\pi)\hbar\omega_{\rm op}(\hbar\omega_{\rm op}/\varepsilon_k)^{1/2}[N(\hbar\omega_{\rm op})+1]\times\int dq\,|I_n(q)|^2\int dQ/(q^2+Q^2) \times\mathrm{Im}\ln|[\hbar\omega_{\rm op}+\varepsilon_Q+2(\varepsilon_k\varepsilon_Q)^{1/2}-i\Gamma_{ac}]/[\hbar\omega_{\rm op}+\varepsilon_Q-2(\varepsilon_k\varepsilon_Q)^{1/2}-i\Gamma_{ac}]|\ . \tag{11}$$

This expression is for unscreened electron–optic-phonon interaction, which will give the shortest relaxation time. In ordinary systems containing considerable electrons, screening works and relaxation time becomes longer. $N(\hbar\omega_{\rm op})$ is the probability of occupation of optic phonons at $T_{\rm ph}$. $\Gamma_{ac}$ appears in the logarithm because of the Dyson equation described in Fig. 4. The $\Gamma_{ac}$ is the same that is calculated in Sec. IV A. In this equation $\varepsilon_Q=\hbar^2Q^2/2m_w$ and $\gamma$ is the Fröhlich's parameter for longitudinal polar optic-phonon–electron interaction: $\gamma=0.071$ for GaAs.[25]

When the width of the QW is sufficiently thin, the characteristic phonon energy is possibly enhanced through interaction with the cladding $Al_{0.3}Ga_{0.7}As$ layers where $\hbar\omega_{\rm op}=47$ meV. However, according to Sawaki,[27] phonons within the QW are well localized, if the width exceeds several monolayers. Accordingly, $\hbar\omega_{\rm op}$ is considered equal to bulk value,[28] 36.7 meV, here.

### C. In case $\varepsilon_k$ is such that $2\hbar\omega_{\rm op}\leqq\varepsilon_k\leqq3\hbar\omega_{\rm op}$

The calculation proceeds similarly. For this time $\Gamma_{2\rm op}$ is calculated in terms of $(G_{1\rm op})^{-1}=(G^0_{1\rm op})^{-1}-\Sigma_{1\rm op}$. $\Gamma_{1\rm op}=\mathrm{Im}\Sigma_{1\rm op}$ appears in the logarithm instead of $\Gamma_{ac}$ in Eq. (11). $\Sigma_{1\rm op}$ has been calculated in Sec. IV B.

## V. RESULTS AND DISCUSSION

The calculated $m$ is plotted in Fig. 5 as a function of well width by a solid line. Also plotted is $n_w/N_w$, in a similar way, by a dashed line, which is proportional to PL efficiency, since $R\beta I_{\rm ex}n_w/\alpha I_{\rm ex}\propto n_w/N_w$. The $m$'s should be read on the left scale, while $n_w/N_w$ are on the right scale. In all calculations it is taken that $\tau_b=\tau_w=\tau_{w0}=\tau$, as the simplest assumption. Since it is not known how long $\tau$ is experimentally, the value is chosen so as to adjust calculations to observations. In this plot $\tau$ is assumed to be 10 ps.

$\tau_{is}$ is the intersubband relaxation time, which consists of a spontaneous IR photon emission and an intersubband transition with emitting optic and/or acoustic phonons, from the highest to the lowest subband. $\tau_{is}$ is equated to $1/QN_w$, which is tentatively assumed to be 50 ps. All calculations are not highly sensitive to this value, provided it is taken to be longer than 3 times $\tau$. According to West and Eglash,[29] the matrix element of the dipole moment $\langle ez\rangle$ is calculated to be around 20 $e$ Å for the intersubband transition. This value gives $\tau_{is}$ a spontaneous photoemission time of several $\mu$s.[30] On the other hand, an intersubband optic-phonon emission time seems to give around several tens ps,[22,23] which predominates over the

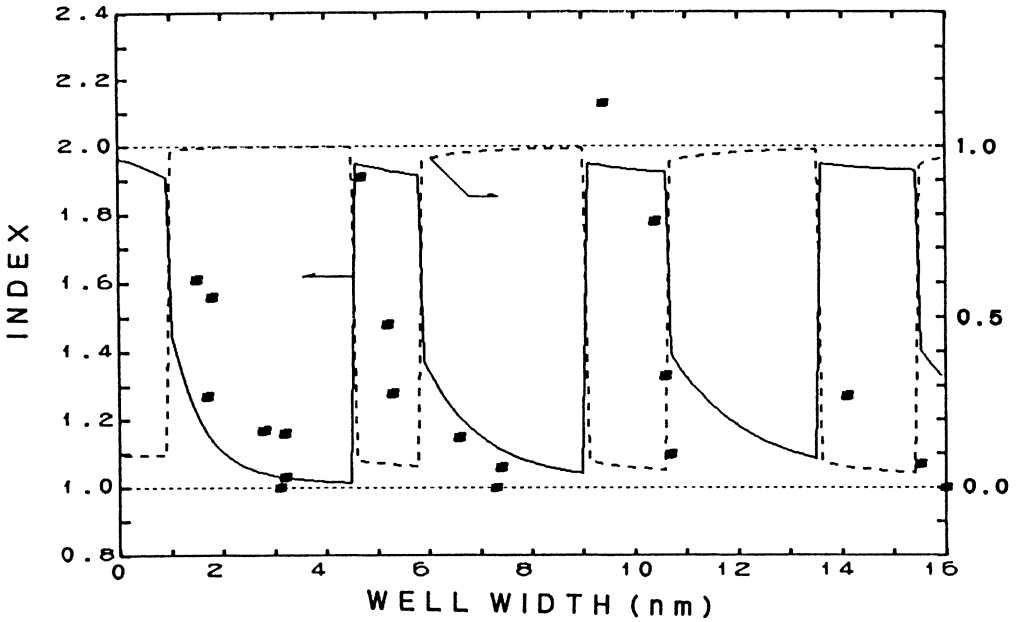


FIG. 5. Power index $m$ versus well width. The solid curve is calculated for $\tau=10$ ps, $\tau_{is}=50$ ps, $i=\tau\alpha I_{ex}/N_w=4.5$, $T_e=T_b=450$ K, and $\hbar\omega_{op}=36.7$ meV. The scale for the solid curve is on the left. The dashed curve stands for calculated $n_w/N_w$, which is proportional to PL efficiency, whose scale is shown on the right. $T_{ph}$ is assumed to be 100 K here and in Fig. 6.

former process. A further investigation on this point will be reported elsewhere.[31]

The calculations illustrated in Fig. 5 are for an excitation level given by a parameter $i=\tau\alpha I_{ex}/N_w=4.5$. The excitation-dependent behavior of $m$ is explained in the following.

In Fig. 1 it is seen that the power index is insensitive to $I_{ex}$ from 50 mW down to a value lower than $\sim 3$ mW. This region spreads over a span of a factor of more than 20. It is seen in Fig. 1 that the data beyond $I_{ex}=50$ mW do not show the QWIDDLE effect and experimenters believe that, there, the lattice temperature rises because of poured-in radiation energy. Just below the region it is natural to assume that temperature, especially that for electrons, is considerably high; thus taken, $T_e$ is 450 K. As was previously mentioned, it is expected that there exists an almost saturation (degeneracy) in the lowest subband under a high excitation level. However, electrons are relieved of saturation by elevating their temperature.

For lower excitation, $T_e$ should be sufficiently low near the lattice temperature. Since the experiment is done at 77 K, a slightly higher temperature equal to 100 K is as-

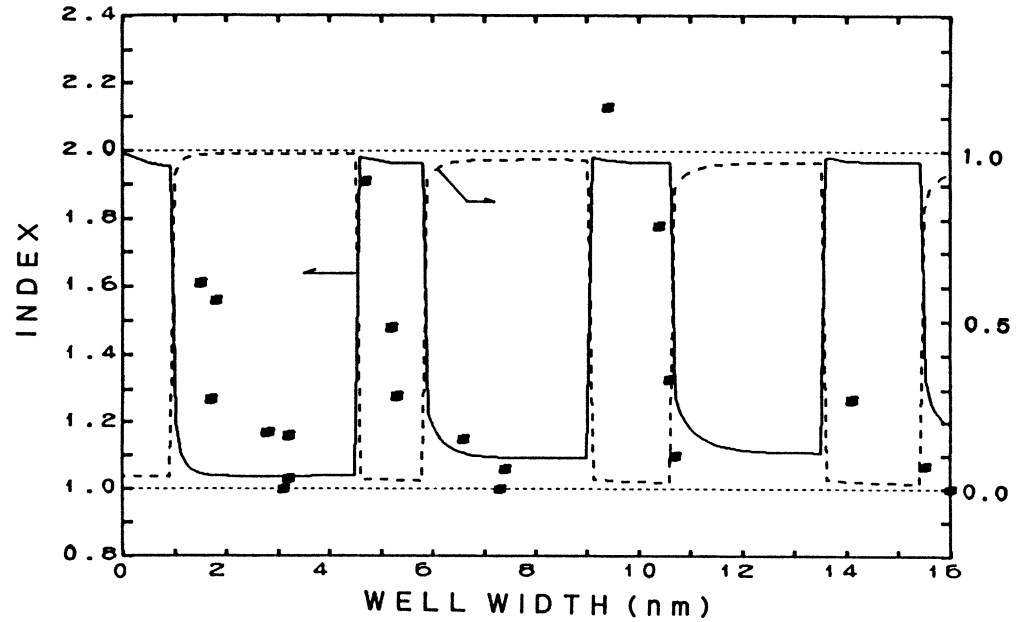


FIG. 6. Same as Fig. 5. $i=\tau\alpha I_{ex}/N_w=1.5$ and $T_e=T_b=100$ K are assumed in this case.

sumed for the calculation shown in Fig. 6. There, $i=\tau\alpha I_{ex}/N_w=1.5$. Therefore, both cases have a difference in $I_{ex}$ of a factor of 20, since $N_w \propto T_e$. From the similarity between Figs. 5 and 6 the constancy of $m$ for $I_{ex}$ up to 50 mW, as shown in Fig. 1, is explainable.

The results obtained so far indicate that the electron system is easily heated within QW's.[20] Otherwise, the behavior of $m$ versus well width would be quite different and the region where $m$'s have constant values would be much smaller than it really is.

As is seen in Figs. 5 and 6, bents occur from $m \cong 2$ to lower values. They are caused by the fact that below the widths corresponding to the bents, $\varepsilon_k$ is too small to emit an optic phonon. Beyond the widths the most dominant relaxation is optic-phonon emission, and relaxation time becomes rapidly shortened. The contrast is clearer at lower electron temperature $T_e$ (Fig. 6).

The present problem seems to be an interesting example of the hot phonon effect. It is seen in Eq. (10) that the transition rate for acoustic phonons is proportional to the phonon temperature, $T_{ph}$. Hence, in the QW width region where $\varepsilon_k$ is less than $\hbar\omega_{op}$ and acoustic-phonon emission is dominant, the temperature effect should appear on the curves in Figs. 5 and 6. Really, the gradients in the region near $m=2$ vary depending on $T_{ph}$, though they are not contrasted in the figures. On the other hand, for optic phonons, the transition rate is given in terms of $(1+N_{ph})$; since $1 \gg N_{ph}$, it becomes insensitive to $T_{ph}$, unless $k_B T_{ph}$ is comparable to $\hbar\omega_{op}$. Thus it is difficult to conclude definitely that the existence of a hot phonon effect is indispensable in order to explain the present effect.

In both cases calculated $\tau$ was assumed to be between the typical relaxation time for acoustic-phonon emission ($\sim$500 ps) and that for the optical-phonon process ($\sim$0.5 ps). For shorter $\tau$'s than 0.5 ps, $m$ values always fall near 2, because then no microscopic process is reflected on $m$'s. As a matter of course for longer $\tau$'s, nonradiative processes no longer work. Thus, it is concluded that $\tau$ is at least longer than the time corresponding to the optic-phonon process to explain the observation. For a much stronger excitation, $m=2$ peaks drop; the power index is almost equal to 1 over all widths, as is really seen in the experiment (see Fig. 1).

The present theoretical analysis interprets observations fairly well, in spite of its quite simple modeling. For much closer agreement, refinements in the model would be necessary. For nearly degenerate electrons, a Fermi-Dirac statistical treatment should replace the present semiclassical Boltzmann statistics in terms of effective density of states. Energy levels, if solved using a self-consistent potential method, would give a better agreement.

Recently, Brum and Bastard[32] studied the trapping rate of electrons into the QW, following Bastard's idea,[18] taking into account newly phonon emitting processes from virtual levels, which are first occupied by transferred electrons from the barriers. However, due to their calculation the trapping efficiency as a function of well width is in contradiction to the observation. (It can be said that it gives a rather completely inverted tendency.) This discrepancy seems to be caused by their model of transfer mechanism, that plays only a minor role in the present phenomenon. Zohta[33] is the other to consider a similar process (i.e., electron transfer in the direction perpendicular to the QW). Though his result seem to be noncontradicting, his mathematics is not convincing.

At last, another experiment is referred to where samples have a different composition $x=0.5$ for the barrier. Most observed $m$'s were less than 1 to around 0.5 in that case. Based on the present formulation and the fact that a potential wall due to the barrier valence band is sufficiently high for holes, they should have a definite quantized subband structure as well; they are sometimes off-resonant to the valence band. Consequently, the present assumption, $p_w=\beta I_{ex}$, no longer holds. The hole contribution to $I_{PL}$ can also be between 0 and 1 like the electron contribution, and the variation in $m$ should be doubly periodic. Because of a large effective mass $m^*$, the critical thicknesses for holes must be small [see Eq. (1)]. Further experimental study is needed to substantiate this point.

In summary, a couple of rate equations along with microscopic transition probabilities between the bottom of the QW subbands and that of the barrier conduction band explained the QWIDDLE effect, i.e., the anomalously oscillating behavior of the power index $m$ defined by $I_{PL} \propto I_{ex}^m$ between 1 and 2, when the QW's are confined by $Al_{0.3}Ga_{0.7}As$. Throughout calculations reproducing observations, only two unknown quantities, nonradiative and intersubband transition relaxation times, are assumed, which are of quite reasonable orders of magnitude. At higher excitation levels the density of carriers within QW states increases enough to make $n_w$ close to $N_w$ in such a manner that a higher temperature $T_e$ is necessarily attributed to the electron system. This temperature boost possibly indicates that the power index $m$ is kept almost constant over a wide range of excitation levels.

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## APPENDIX

Since the Fermi-Dirac (FD) statistics is not easy to handle analytically unless the system is almost completely degenerate, it is advantageous to establish a semiclassical statistics, effectively taking into account a kind of FD nature. It is so, especially in such equations as the rate equations. In order to justify Eqs. (2)–(4), the ordinary golden rule formula is considered for the optic-phonon emission rate, as an example. The process is sketched in Fig. 7.

In the equation,

$$\Sigma_{\mathbf{k},\mathbf{q}}[1/\tau(\mathbf{k},\mathbf{q})]f_{\mathbf{k}+\mathbf{q}}(1-f_{\mathbf{k}})=(2\pi/\hbar)\Sigma\,|M_{\mathbf{q}}|^2 f_{\mathbf{k}+\mathbf{q}}(1-f_{\mathbf{k}})(1+N_{\mathbf{q}})(1/\pi)\mathrm{Im}[1/(\varepsilon_{\mathbf{k}+\mathbf{q}}-\varepsilon_{\mathbf{k}}-\omega_{op}+i\Gamma)]\,, \tag{A1}$$

$f_{w+q}=\Sigma_j(-1)^j\exp[-j\beta_b(\varepsilon_k+\omega_{op}-\zeta_b)]$ and $f_w=\Sigma_j(-1)^j\exp[-j\beta_w(\varepsilon_k-\zeta_w)]$ are the FD functions for electrons just after having been transferred from the barrier and for those within the QW, respectively. $\beta_b=1/k_BT_b$ and $\beta_w=1/k_BT_w$ specify temperatures similarly in the barrier and the QW. $\zeta_b$ and $\zeta_w$ stand for the chemical potentials. For both energies such as $\beta_b(\varepsilon_k+\omega_{op}-\zeta_b)>1$ and $\beta_w(\varepsilon_k-\zeta_w)>1$, the following final expression for

$$\Sigma_{\mathbf{k}}[1/\tau(\mathbf{k},\mathbf{q})]f_{\mathbf{k}+\mathbf{q}}(1-f_{\mathbf{k}})$$

reads

$$[1/\tau(0,q)](D_w/D_b)n_b\exp[-\beta_b(\omega_{op}-\Delta)]$$
$$\times\{1-[\beta_b/(\beta_b+\beta_w)]\exp\beta_w\zeta_w\}\ , \quad \text{(A2)}$$

when $1/\tau(k,q)$ is slowly varying with respect to $\mathbf{k}$ in comparison to exponential function, and $N_w=D_w/\beta_w$, $n_w=N_w\exp(\beta_w\zeta_w)$, and $N_b=D_b/\beta_b$, as well as $n_b=N_b\exp[-\beta_b(\Delta-\zeta_b)]$ are utilized. The values within the curly brackets in the above equation (A2) are obviously proportional to $1-n_w/N_w$, as long as $T_w\gg T_b$. Otherwise, $N_w$ must be the renormalized effective density of states, including the temperature effect.

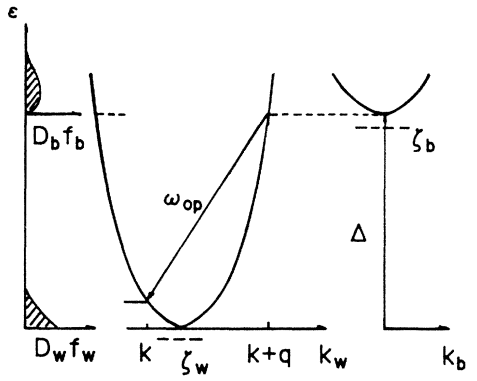


FIG. 7. Schematic diagram of the optic-phonon emission process.

For $\beta_w\zeta_w\gtrsim 0$, all calculations should be performed based on the FD statistics itself. For the present problem, effective temperatures higher than the ambient one are assumed, and the rate equations are considered to be justified so long as the solved $n_w$ does not surpass $N_w$.


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