Microwave modulation of exciton luminescence in $GaAs/Al_xGa_{1-x}As$ quantum wells

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We present a study of the photoluminescence (PL) modulation by microwave irradiation in $GaAs/Al_xGa_{1-x}As$ multiple quantum wells. At low temperatures, we observe that the inhomogeneously broadened PL band of (e1:hh1) excitons is split into low-energy negative-modulation and high-energy positive-modulation components. We interpret this spectral shape and its dependence on photoexcitation (laser) energy and on microwave power in terms of the following model: Photoexcited free electrons are heated by the microwave radiation and they impact activate the excitons into states with higher energies (without dissociating the excitons). This model is based on two types of exciton states: localized in the low-energy tail of the PL band and delocalized in its high-energy part. A well-defined exciton temperature is deduced from the anti-Stokes part of the microwave-modulated PL spectrum. It depends on the absorbed microwave power but is independent of the photoexcitation energy.

I. INTRODUCTION

The radiative recombination spectrum of photoexcited e-h pairs in semiconductors can be modified by applying microwave radiation. In undoped semiconductors and at low temperatures, the photoluminescence (PL) is due to bound and free excitons. Since the excitons are neutral, they do not interact directly with the microwave field. Instead, this field heats up the free carriers in the crystal,[1,2] which then affect the excitons in various ways. Several mechanisms have been proposed in order to explain the observed spectra. Impact ionization of free excitons by the heated electrons is proposed in Ge,[2] and of bound excitons in Si.[3] In very pure GaAs, impact ionization of both bound excitons and donor-acceptor pairs,[4] as well as a decrease in the cross section for exciton formation,[5] are observed. In bulk III-V semiconductor alloys, a different mechanism is invoked:[6] the heated electrons increase the lattice temperature, and this affects the PL spectrum. Microwave radiation effects on the PL spectrum of doped $GaAs/Al_xGa_{1-x}As$ (Ref. 7) and $In_xGa_{1-x}As/InP$ (Ref. 8) multiple quantum wells (MQW) were also observed. They were attributed to impact ionization of bound excitons, which is enhanced under cyclotron-resonance conditions.

We have studied the microwave-modulated PL spectra (hereafter denoted MPL) of undoped $GaAs/Al_xGa_{1-x}As$ MQW's, and compared them with those obtained under temperature modulation by directly heating the crystal (TMPL). The main purpose of this paper is to show that MPL spectroscopy is particularly useful in analyzing the degree of in-plane exciton localization in MQW's. As is well known, the (e1:hh1) 1S exciton band in MQW's is inhomogeneously broadened mainly by random interface fluctuations.[9,10] The spectral shape of the PL band reflects the exciton distribution among the lowest-energy states of the spatially fluctuating in-plane potential. The exciting (laser) radiation generates excitons and free carriers. Then, by applying a microwave field, those electrons (or holes) which are free to move in the QW plane are heated and impart their energy to the excitons. In this "impact activation" process, excitons are promoted into states with a higher kinetic energy without dissociation. This results in a redistribution of the excitons among the (lower-energy) localized states and the (higher-energy) delocalized ones. From the observed MPL spectra, and using a simple model for the exciton population rate equations, we estimate the rate coefficients of the hot electron-exciton impact activation and the exciton localization (trapping into localized states) as well as the exciton temperature.

The paper is laid out as follows. Section II describes the experimental procedure and results. Section III gives the model of exciton impact activation by heated electrons, and the analysis of the MPL spectra. Section IV is a summary.

II. EXPERIMENTAL PROCEDURE AND RESULTS

We studied several undoped $GaAs/Al_{0.33}Ga_{0.67}As$ MQW's. They were grown by molecular-beam epitaxy either without interruptions or with 75-sec interruption at each interface. (They will be denoted "unint" and "int," respectively). The $Al_xGa_{1-x}As$ barrier width is 200 Å and the GaAs well width is 50 Å. The samples were mounted in a microwave cavity which was placed in an immersion-type dewar. The ambient temperature was varied in the range of $T=2-30$ K. The samples were irradiated (through a small aperture in the cavity) by light from either a cw dye laser pumped by an Ar^+ laser or from a He-Ne laser. The laser beam was chopped at 3 kHz and the range of average intensity was $1-10^3$ mW/cm^2. The PL spectra were monitored by a spectrometer equipped with a cooled photomultiplier, and the signal was processed by a lock-in amplifier.

The samples were placed in the cavity in the region of maximal electric field, polarized in the QW plane. The 36-GHz microwave source was a Gunn diode, and its radiation was modulated by a *p-i-n* diode at 3 kHz. The microwave power reaching the sample was varied in the range of 10–100 mW. By measuring the power reflected from the sample we estimated that the actual absorption under laser illumination (by the whole sample, namely, both substrate and MQW) did not exceed 5% of the incident microwave power.

A. Excitation above the (*e*1:hh1) exciton band

Figure 1 shows the exciton PL [1(a)] and MPL [1(b)] spectra of a MQW with a well width of 50 Å (interrupted growth) observed at $T=2$ K. These spectra were obtained under excitation with a laser energy of $E_l=1.96$ eV. The MPL spectrum was obtained for a microwave power of about 5 mW absorbed by the whole sample. The maximal modulation under these conditions is $\Delta I/I \sim 15\%$ [Fig. 1(c)]. We observe that the MPL spectrum consists of a positive ΔI^+ in the high-energy part of the exciton band and a negative ΔI^- in the low-energy tail. The absolute value of both parts of ΔI increases linearly with increasing microwave power. The ratio between the integrated intensities is defined by

$$R=\frac{\int_{E_0}^{\infty}\Delta I^+(E)dE}{\int_{-\infty}^{E_0}\Delta I^-(E)dE}\ , \tag{1}$$

with E_0 being the energy at which $\Delta I(E_0)=0$. We find that $R\sim 3$ for the MQW's under study and for the whole range of microwave power used here. Figures 2(a) and 2(b) show, respectively, the PL and MPL spectra of a 50 Å/200 Å MQW (uninterrupted growth), observed at $T=5$ K, and obtained under the same excitation conditions as those of Fig. 1. We note that the MPL spectra obtained for the uninterruptedly grown MQW's are similar to those obtained for the interrupted growth samples.

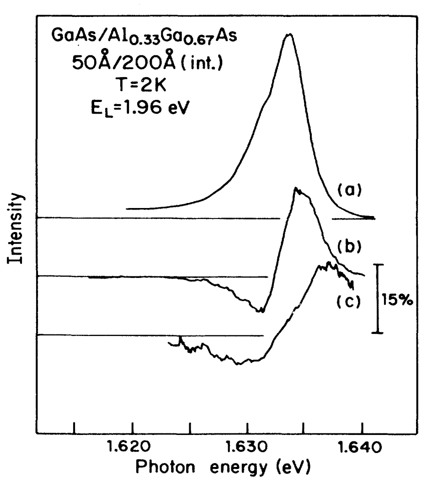

FIG. 1. (a) The photoluminescence spectrum [$I(E)$]. (b) The microwave-modulated photoluminescence spectrum [$\Delta I(E)$]. The microwave power absorbed by the whole sample is 5 mW. (c) $\Delta I(E)/I(E)$.

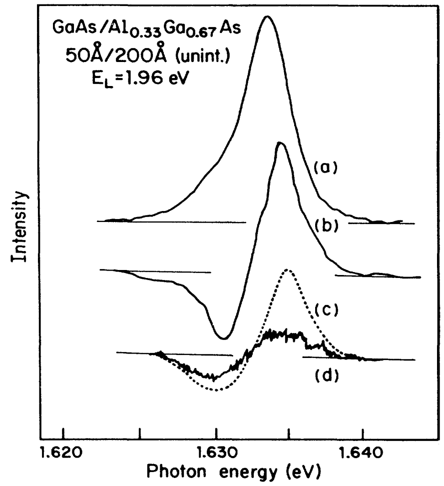

FIG. 2. The (a) photoluminescence and (b) microwave-modulated photoluminescence spectra observed at $T=5$ K. The microwave power absorbed by the whole sample is 5 mW. The two lowest curves are the temperature-modulated photoluminescence spectra obtained by subtraction: (c) $\Delta I(12,20,E)=I(T=20\text{ K},E)-I(T=12\text{ K},E)$ and (d) $\Delta I(5,8,E)$.

We have also studied the thermally modulated PL spectra (TMPL), in order to compare them with the MPL spectra. The cw PL spectra were measured for a given excitation (laser) energy and power, and at various ambient temperatures. Then, the TMPL spectra were obtained by numerically subtracting the PL spectra observed at temperatures T_1 and T_2 ($T_2>T_1$):

$$\Delta I(T_1,T_2,E)=I(T_2,E)-I(T_1,E)\ . \tag{2}$$

Two TMPL spectra, observed under the same excitation conditions, are shown: $\Delta I(5,8,E)$ in Fig. 2(d) and $\Delta I(12,20,E)$ in Fig. 2(c). We note that while the TMPL spectra have a similar shape to that of the MPL spectra, the modulation depth of the former is smaller (for $T<20$ K) and so is the ratio of the positive to negative parts: $R(5,8)\sim 1$ and $R(12,20)\sim 2$. The MPL intensity is independent of temperature up to $T\sim 10$ K, then it decreases with increasing T and vanishes for $T\geq 30$ K.

B. Resonant excitation within the (*e*1:hh1) exciton band

Figure 3 shows the MPL spectra observed for three different laser energies (E_l) within the exciton band. (The laser intensity and microwave power are the same for all spectra.) We note that in all three cases there is a strong anti-Stokes intensity (MPL signal for $E>E_l$). Also, for $E_l<E_0$ (i.e., in the low-energy part of the PL band) and for absorbed microwave power greater than ~ 1 mW, ΔI changes its sign and becomes positive throughout the MPL spectrum. (There is a strong $\Delta I<0$ at E_l that is due to modulated Rayleigh scattering and it will be reported elsewhere.) Figure 4 compares the photoluminescence excitation (PLE) spectra, monitored in the low [4(b)] and high [4(c)] parts of the PL spectrum [4(a)]. The corresponding microwave-modulated PLE spectra

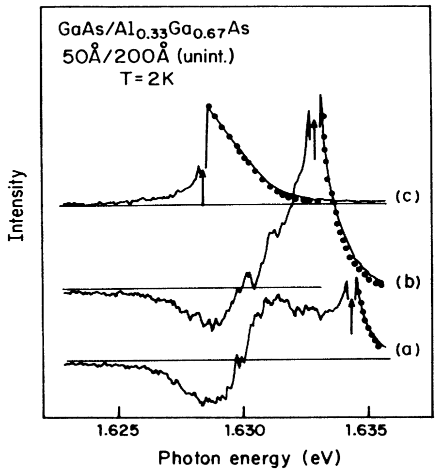

FIG. 3. The microwave-modulated photoluminescence spectra excited at the following laser energies (indicated by arrows): (a) $E_l = 1.6345$ eV, (b) $E_l = 1.6331$ eV and (c) $E_l = 1.6286$ eV. In all the spectra, the microwave power absorbed by the sample is 5 mW. The full lines are the experimental spectra and the dots are the calculated anti-Stokes spectra [Eq. (12)] corresponding to an exciton temperature of $T_{ex} = 11$ K.

are shown in Figs. 4(e) and 4(d), respectively. The main points to notice are (a) the sign of the modulated PLE spectrum is the same as that of the corresponding MPL signal: negative in the low-energy part of the PL band (for absorbed power < 1 mW) and positive in the high-energy part; and (b) the modulated PLE intensity in the energy range of $E_l > 1.64$ eV is stronger than that for

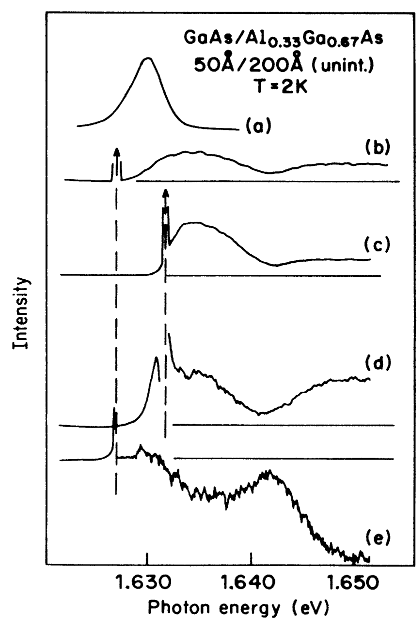

FIG. 4. The photoluminescence spectrum (a) and two excitation spectra monitored at (b) 1.627 eV and (c) 1.6315 eV. The microwave-modulated excitation spectra monitored at (d) 1.6315 eV and (e) 1.627 eV.

$E_l < 1.64$ eV, when compared with the corresponding PLE spectra.

III. ANALYSIS AND MODEL

In order to analyze the observed MPL and its distinct features when compared with the TMPL spectra, we begin by summarizing the main properties of $(e1{:}hh1)1S$ excitons in $GaAs/Al_xGa_{1-x}As$ MQW's. These excitons are subjected to spatial potential fluctuations which arise mainly from interface roughness.[9,10] Consequently, the exciton band is inhomogeneously broadened with the low-energy tail of the PL band corresponding to localized excitons. The higher-energy part of the PL band appears in the low-energy tail of the absorption spectrum [or the PLE spectra, cf. Figs. 4(a)–4(c)] and is due to the radiative recombination of delocalized excitons. Several recent studies[11,12] show that these delocalized excitons move freely over regions of the QW in which the well width is approximately uniform. These regions ("interface islands") have a larger area than πa_{1S}^2, where a_{1S} is the two-dimensional exciton Bohr radius in the $1S$ state. There is still microroughness on the scale of a few lattice constants, but, to a first approximation, the delocalized excitons are considered to have a well-defined in-plane wave vector ($\mathbf{k}_\parallel$). For still higher kinetic energies, the exciton wave function extends over the whole QW plane. Figure 5(a) presents a schematic description of the types of $(e1{:}hh1)1S$ excitons. The localized and delocalized excitons are represented by two sets of levels, while the extended excitons are described by a parabolic dispersion curve. The total lifetimes of these two types of excitons are denoted τ_L and τ_F, respectively. (The total lifetime is given by $\tau^{-1} = \tau_{rad}^{-1} + \tau_{nr}^{-1}$ where τ_{rad} is the radiative lifetime and τ_{nr} is the nonradiative lifetime.) The exciton density of states [$\rho(E)$ in Fig. 5(b)] has an exponentially decreasing low-energy tail and a constant value for high energies (two-dimensional dispersion).

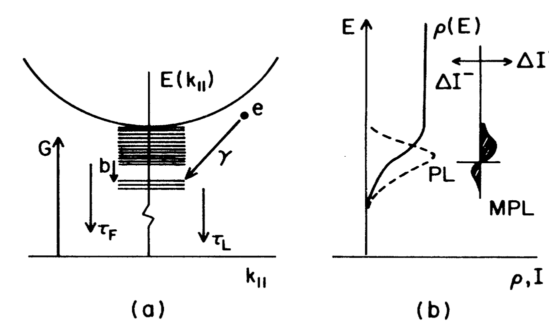

FIG. 5. A schematic description of the model: (a) Two types of exciton states are described by horizontal bars: localized, having a lifetime of τ_L, and delocalized, with τ_F. The extended exciton states are described by a parabolic dispersion with respect to the in-plane $k_\parallel$. γ is the rate coefficient of impact activation of localized excitons by the hot electrons. The rate coefficient of exciton trapping from delocalized into localized states is denoted by b. (b) The density of states, $\rho(E)$, photoluminescence (PL), and its microwave-modulated [ΔI^+ and ΔI^-)] spectra.

Only the localized and delocalized excitons recombine radiatively at low temperatures. Under cw excitation the PL band appears in the low-energy part of $\rho(E)$, and its shape is determined by two main factors. The first is the density of states of those excitons that can interact with photons [having energies in the (e1:hh1) spectral range]. While all the localized excitons can interact with these photons, only those delocalized excitons which have $\mathbf{k}_{\parallel}\sim 0$ can do so. The second factor includes all the dynamic processes that the excitons undergo during their lifetime. These include exciton scattering between interface islands, acoustic phonon-assisted relaxation within the translational states of a given island, and exciton trapping into localized states.

All these dynamic processes are faster than the exciton radiative lifetimes.[13,14] Microwave and thermal modulations activate dynamic processes which modify the exciton distribution among the localized and delocalized states. The observed MPL and TMPL spectra indicate that in the MQW's under study, localized excitons acquire a higher energy and are promoted into the delocalized exciton part of the PL band [Fig. 5(b)]. Since the excitons are neutral particles, they do not interact directly with the microwave field. Rather, this field heats up free carriers,[1,15] mainly electrons, which then impart some of their excess energy to the excitons. [We schematically describe this process by the arrow marked γ in Fig. 5(a)]. Since the MPL spectral range coincides with that of the PL spectrum, and it spans about one-half the exciton binding energy, the hot electrons merely increase the exciton energy without ionizing it. It is thus an "impact activation" process.

The electrons, which are available to interact with the microwave field, must be free to move over a large enough distance in the QW plane in order to acquire kinetic energy. The modulated PLE spectra [Figs. 4(e) and 4(d)] indicate that such electrons are present even for excitation (laser) energies deep in the tail of $\rho(E)$, namely, for $E_l < E_{\rm gap}$. We thus conclude that electrons are excited directly from (below gap) trap states, and that they are free to move over large distances—presumably over the same interface islands where delocalized excitons are excited. This is consistent with a recent observation of separately localized electron-hole pairs in MQW's.[16] We also note that the modulated PLE spectrum for E_l high in the conduction band is more intense than for E_l in the exciton band, when compared with the PLE spectrum [cf. Figs. 4(b) and 4(e), and similarly, Figs. 4(c) and 4(d)]. This can be interpreted in terms of a higher free carrier generation rate, which results in a more efficient microwave modulation.

According to this analysis, it is now clear why the microwave modulation is more efficient than that of the crystal temperature (for $T\leq 20$ K, Fig. 2). The free electrons, which are heated by the microwave radiation, are efficient in activating the localized excitons into the delocalized states. Since the delocalized states correspond to large interface regions in which the exciton translational states are the same (only the relative "localization" energy is different), an effective exciton temperature is established throughout the MQW. As we shall see below, this temperature is higher than that of the lattice. On the other hand, increasing the crystal temperature results in a thermal activation of the excitons with only a slight redistribution due to tunneling between the localized states.

We now turn to putting these ideas into a quantitative model. Consider first the case of excitation energy well above the band gap (as in Figs. 1 and 2). Excitons are generated at a rate of G cm^{-2} sec^{-1}. As the density of delocalized exciton states is much larger than that of the localized states, we assume that only the former are directly generated by the exciting laser beam. If n_F denotes the sheet density of all delocalized excitons, then the trapping rate into all the localized states is $(-bn_F)$. The main effect of microwave irradiation is impact activation of the localized excitons (sheet density of n_L) at a rate of $(-\gamma n_L)$ into the band of delocalized excitons. Then, under the nonsaturation conditions of our experiments, namely, R of Eq. (1) is independent of microwave power, the rate equations are

$$\frac{dn_F}{dt}=G-\frac{n_F}{\tau_F}-bn_F+\gamma n_L \ , \tag{3}$$

$$\frac{dn_L}{dt}=-\frac{n_L}{\tau_L}+bn_F-\gamma n_L \ . \tag{4}$$

In these equations we neglect any dependence of τ_F, τ_L, b, and γ on exciton energy. This simplified model will thus yield only average values of these parameters for the two types of excitons. In the absence of microwave irradiation, the rate equations are

$$\frac{dn_F^0}{dt}=G-\frac{n_F^0}{\tau_F}-bn_F^0 \ , \tag{3$'$}$$

$$\frac{dn_L^0}{dt}=-\frac{n_L^0}{\tau_L}+bn_F^0 \ . \tag{4$'$}$$

Under steady-state conditions, these four equations are solved for the following physical quantities:

$$\frac{\Delta n_F}{|\Delta n_L|}\equiv\frac{n_F-n_F^0}{|n_L-n_L^0|}=\frac{\tau_F}{\tau_L} \ , \tag{5}$$

$$\delta_F\equiv\frac{\Delta n_F}{n_F^0}=\frac{b\gamma\tau_F\tau_L}{1+b\tau_F+\gamma\tau_L} \ , \tag{6}$$

$$\delta_L\equiv\frac{\Delta n_L}{n_L^0}=-\frac{\gamma\tau_L}{1+b\tau_F+\gamma\tau_L} \ , \tag{7}$$

$$\frac{\delta_F}{|\delta_L|}=b\tau_F \ . \tag{8}$$

These quantities are related to the following measured ones:

$$R=\frac{\Delta n_F}{|\Delta n_L|} \ , \tag{9}$$

$$\frac{\int_{E_0}^{\infty}\Delta I^{+}(E)dE}{\int_{E_0}^{\infty}I(E)dE}=\delta_F \ , \tag{10}$$

and similar relations for δ_L and $\delta_F/|\delta_L|$. According to this model, R is independent of microwave power (represented by the parameter γ), as observed experimentally.

From Eqs. (5) and (9), and the measured value of $R \sim 3$ we obtain $\tau_F \sim 3\tau_L$. We note that the parameters τ_F and τ_L represent average values of the delocalized and localized exciton lifetimes, respectively. Reported[17,18] values range between 150 and 400 psec, for QW's with similar parameters to those studied here. Our main interest is in obtaining values for the parameters b and γ, again averaged over the respective type of exciton states. Therefore we take the full range of reported lifetimes as representative values for both τ_F and τ_L. Now, from Figs. 1(b) and 1(c) we obtain $\delta_F/|\delta_L| \sim 1$ and $|\delta_L| \sim 0.2$. Using Eqs. (7) and (8) yields $b^{-1} \sim \tau_F$ and $\gamma^{-1} \sim 2\tau_L$.

The average trapping time range, $b^{-1} \sim (1.5-4) \times 10^{-10}$ sec, can be compared with the time required for excitons to reach the maximum PL intensity. This was studied by Damen *et al.*,[14] using time-resolved spectroscopy and was found to be $\tau_f \sim 4 \times 10^{-10}$ sec for a MQW with 80-Å wells at $T = 5$ K. This agreement between the two different types of measurements is further supported by the observation[14] that τ_f decreases with increasing temperatures. We observed that the MPL signal vanishes for $T \geq 30$ K, and therefore thermalization overcomes the microwave modulation.

Finally, we consider the MPL spectra which are observed under resonant excitation within the exciton band (Fig. 3). In all cases there is a strong anti-Stokes MPL which is due to excitons that are impact activated into higher-energy states. For the case of delocalized states, only excitons with $\mathbf{k}_\parallel \sim 0$ will radiate. Therefore the anti-Stokes spectrum, in the delocalized exciton range, reflects the redistribution of $\mathbf{k}_\parallel \sim 0$ excitons. We note that also for $E_l < E_0$ and for the entire range of absorbed microwave power, the anti-Stokes MPL is a smooth spectrum with $\Delta I > 0$. We then propose that the subset of delocalized excitons with $\mathbf{k}_\parallel \sim 0$ (namely, those which can recombine radiatively) and the localized ones have a common density-of-states function which we approximate by a Gaussian:

$$\rho'(E) = \frac{1}{(\pi \Delta E^2)^{1/2}} \exp\left\{ -\left[\frac{E-E'}{\Delta E} \right]^2 \right\} . \quad (11)$$

Then, assuming a Boltzman distribution of the impact-activated excitons among the $\mathbf{k}_\parallel = 0$ states, their population is

$$n(E, T_{ex}) = \rho'(E) \exp\left\{ -\frac{E-E_l}{kT_{ex}} \right\} . \quad (12)$$

T_{ex} is the exciton temperature, expected to be higher than that of the lattice. Since the anti-Stokes spectrum $\Delta I(E > E_l) \propto n(E)$, we fit all the spectra shown in Fig. 3 with the same set of parameters. The dots in the figures are the calculated MPL intensities, using the following parameter values: $E' = 1.6315$ eV, $\Delta E = 2.9$ meV, and $T_{ex} = 11$ K. This temperature is obtained for the maximal microwave power absorbed by the whole sample (5 mW). For lower absorbed power, lower T_{ex} are obtained with the same values of E' and ΔE. From this we conclude that the model summarized by Eqs. (11) and (12) presents an adequate description of the exciton states and their population under microwave modulation. We also note that the change of sign observed in the MPL spectrum for $E_l < E_0$ and absorbed power higher than 1 mW is also explained by this model. For these laser energies, excitons are created in localized states, with limited transfer into lower-energy states (at $T = 2$ K). Under hot electron impact activation, the excitons are promoted into delocalized states from where they can reach all the localized states and thus increase their luminescence.

IV. SUMMARY

In this work we showed that the main mechanism of microwave-modulation photoluminescence in undoped $GaAs/Al_xGa_{1-x}As$ MQW's is impact activation of excitons by hot electrons. Thermal modulation is a much weaker process in the ambient-temperature range of $2 < T \leq 20$ K. We interpreted the observed MPL in terms of a redistribution of excitons in localized and delocalized states within the inhomogeneously broadened $(e1\text{:hh}1)1S$ band. The model is consistent with a description of the delocalized states as those of a free translational motion of the exciton in large interfacial islands. The localized states arise from short-range fluctuations in these islands. The analysis of the spectra yielded average values of the hot electron-exciton impact activation rate coefficient and the trapping rate coefficient of delocalized excitons into localized states. Both are of the order of the inverse exciton lifetime.

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