Photocurrent studies of the carrier escape process from InAs self-assembled quantum dots

W.-H. Chang, T. M. Hsu, C. C. Huang, S. L. Hsu, and C. Y. Lai
Department of Physics, National Central University, Chung-Li, Taiwan 32054, Republic of China

N. T. Yeh, T. E. Nee, and J.-I. Chyi
Department of Electrical Engineering, National Central University, Chung-Li, Taiwan 32054, Republic of China

(Received 27 March 2000; revised manuscript received 31 May 2000)

We present a temperature- and bias-dependent photocurrent study of the excitonic interband transitions of InAs self-assembled quantum dots (QD's). It was found that the carrier escape process from QD's is dominated by hole escape processes. The main path for this hole escape process was found to be thermal-assisted hole tunneling, from the dot level to the GaAs barrier via the wetting layer as an intermediate state. Energy-dependent carrier tunneling from the QD's to the barrier was observed at low temperatures. Energy shifts due to the size-selective tunneling effect and the quantum-confined Stark effect are discussed and compared with the carrier redistribution effect in photoluminescence measurements.

Self-assembled quantum dots (QD's) have recently been of considerable interest due to their importance in low-dimensional physics and their applications[1] in optoelectronic devices such as QD lasers[2] and charge storage devices.[3–5] In most device applications, the carrier escape process from the QD's is very important. Electrical characterizations, such as capacitance-voltage (C-V) characteristics,[6] admittance spectroscopy,[7] and deep-level transient spectroscopy[8] have been employed to study this process. Optical characterizations, such as photoluminescence[9] (PL) and modulation reflectance spectroscopy,[10] have also been employed to investigate the thermal detrapping of carriers in QD's. In this paper, we present a carrier escape study from InAs self-assembled QD's by the use of photocurrent (PC) measurements. Complementary to the PL measurement, the collected QD PC signals provide an absorptionlike spectrum, which directly reflects information about carrier escapes from the QD's. The temperature- and bias-dependent PC measurements reveal the behavior of the carrier thermally assisted and electric-field-induced tunneling from the QD's to the GaAs barrier. From these results, the main carrier escape path is clarified and the effects of direct tunneling from the QD's at low temperatures are discussed.

The QD investigated sample was grown on (100)-oriented GaAs by solid-source molecular-beam epitaxy using the Stranski-Krastanow growth mode. A layer of self-assembled InAs QD's (~2.7 monolayers) capped with a 6 nm $In_{0.16}Ga_{0.84}As$ strain-reduced layer (SRL) were sandwiched between two 150-nm-thick intrinsic GaAs layers of a *p-i-n* structure. The growth temperature was 520 °C during the QD and SRL deposition processes, and 580 °C for the rest of the GaAs layers under As-stabilized conditions. The sample structure is schematically shown in the inset of Fig. 1. From transmission electron microscopy images, the QD's were found to be lens shaped, 3.5 nm high, with a 21 nm average diameter, and a dot density of $5\times10^{10}\,cm^{-2}$. 1 mm^2 mesas were deposited from Au-Be film with 500 μm^2 apertures as metallic top contacts. The PC measurements were excited using a 1 kW tungsten-halogen lamp combined with a 0.25 m monochromator, and detected using the standard lock-in techniques.

Figure 1 shows the close-circuit PC and a PL spectrum for the QD sample measured at room temperature (RT, $T=300\,K$). The PL spectrum shows clear luminescence peaks at 0.92 and 0.99 eV, which indicate the ground-state and first-excited-state transitions of the InAs QD's. In the PC spectrum, more spectral features are observed. A series of well-defined peaks arising from the QD interband absorption (QD0, QD1, QD2, etc.) are observed from 0.9 to 1.2 eV. The features that appear near 1.25 eV arise from interband absorption in the wetting layer (WL), while the broad peak at 1.33 eV is the quantum-well signal of the $In_{0.16}Ga_{0.84}As$ SRL. The increasing PC signal beyond 1.35 eV indicates the

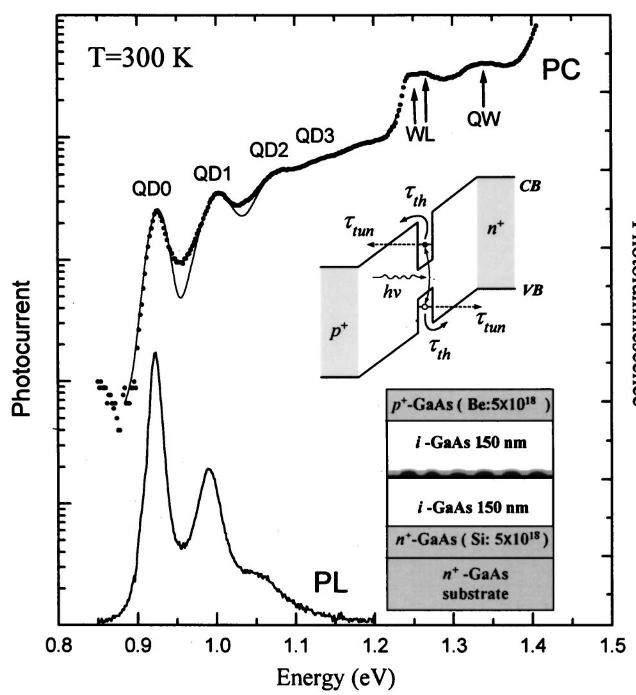

FIG. 1. The PL spectrum and the closed-circuit PC spectrum measured at RT. The labeled index indicates the ground and excited QD states. The inset shows the sample structure and the band diagram for the *p-i-n* structure investigated.

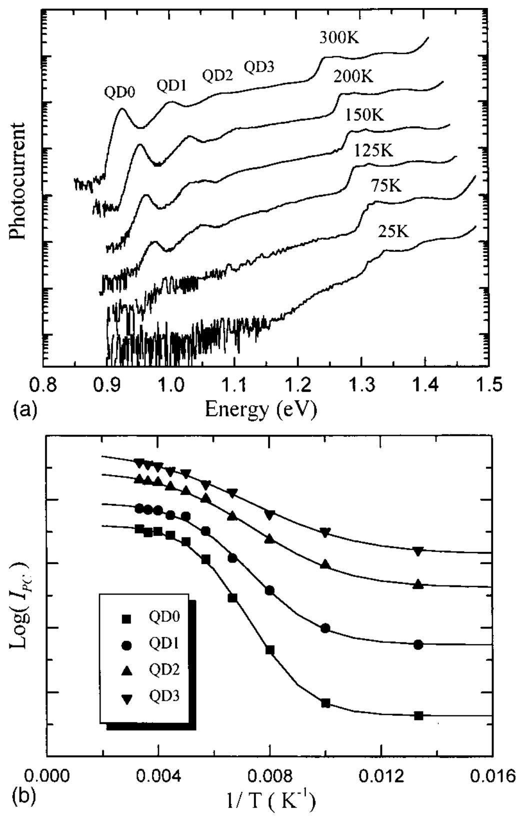

FIG. 2. The temperature-dependent PC spectra (a) and an Arrhenius plot of the QD PC intensity (b). The solid lines indicate the least-squares fits of Eqs. (1) and (2).

absorption tail of the GaAs bulk material due to the electric-field-induced Franz-Keldysh effect.

The temperature-dependent PC spectra measured at zero bias are shown in Fig. 2(a), and the QD PC intensities ($I_{\rm PC}$) vs $1/T$ are plotted in Fig. 2(b). In this figure, the temperature dependence of the QD PC intensity can be divided into three regimes. At low temperatures ($T<100\,{\rm K}$), the QD PC signals are very weak; only PC signals arising from the WL and the SRL are observed. With increasing temperature, the QD PC signals are gradually increased. At $T>230\,{\rm K}$, the increasing QD PC intensities become saturated. The QD PC strength is determined by the escape of photogenerated carriers from the dots to the barrier. When an electron-hole pair is created in a QD level, it can either recombine in the QD level or escape out of the QD. Once the carriers escape to the barrier, they will be accelerated by the built-in electric field and are collected by the top and rear electrodes as PC signals. Therefore, the competition between the recombination rate ($1/\tau_{\rm rec}$) and the escape rate ($1/\tau_{\rm esc}$) determines the collected QD PC strength. As shown in the inset of Fig. 1, carrier escapes from the QD state to the barrier consist of two processes: (i) direct tunneling through the triangular barrier induced by the built-in electric field and (ii) thermionic emissions from the dot level to the barrier. Therefore, the total escape rate may be given by $1/\tau_{\rm esc}=1/\tau_{\rm tun}+1/\tau_{\rm th}$, where $\tau_{\rm tun}$ and $\tau_{\rm th}$ are the tunneling and thermionic emission lifetimes, respectively. At low temperatures, $1/\tau_{\rm th}$ is very slow. The collected PC is determined by carrier tunneling through the triangular barrier, which is a temperature-independent process. If the tunneling rate $1/\tau_{\rm tun}$, of the photogenerated carrier is also slow compared to the recombination rate $1/\tau_{\rm rec}$, the low-temperature QD PC, signal is very weak. With increasing temperature, the carriers start to be activated, and the escape and recombination rates compete. As a result, the collected QD PC signal becomes more intense. At high temperatures, the increasing QD PC signals become saturated. This means that $1/\tau_{\rm esc}\gg 1/\tau_{\rm rec}$ at high temperatures and most of the photogenerated carriers have escaped from the QD's to be collected as PC signals before they recombine.

TABLE I. The fitting results shown in Fig. 2(b) to Eqs. (1) and (2) obtained by setting $\tau_{\rm rec}=1$ ns. The transition energies $E_{\rm WL}$ and $E_{{\rm QD}i}$ were obtained by a multiple Gaussian fit to the RT PC spectrum.

QD state	$E_{{\rm QD}i}$ (eV)	$E_{\rm GaAs}-E_{{\rm QD}i}$ (meV)	$E_{\rm WL}-E_{{\rm QD}i}$ (meV)	E_a (meV)	$1/\tau_{\rm tun}$ (s^{-1})
QD0	0.926	498	324	98.7	6.00×10^{7}
QD1	1.002	422	248	75.2	1.21×10^{8}
QD2	1.073	351	177	55.3	1.91×10^{8}
QD3	1.130	294	120	38.5	2.50×10^{8}

The temperature-dependent QD PC intensity can be modeled by a simple rate-equation approach. For a carrier generation rate G, the rate equation for the carrier population (n) in a given QD level is given by

$$\frac{dn}{dt}=G-\frac{n}{\tau_{\rm esc}}-\frac{n}{\tau_{\rm rec}}.$$

Under steady-state conditions, i.e., $dn/dt=0$, one can obtain the QD PC intensity as

$$I_{\rm PC}=\frac{n}{\tau_{\rm esc}}=\frac{G}{1+\tau_{\rm esc}/\tau_{\rm rec}}. \quad (1)$$

The major temperature dependence in $1/\tau_{\rm esc}$ is governed by $1/\tau_{\rm th}=1/\tau_0\exp(-E_a/kT)$, where τ_0 is the effective time constant for carriers scattered from the dot to the barrier, and E_a is the activation energy. Therefore, $1/\tau_{\rm esc}$ is given by

$$\frac{1}{\tau_{\rm esc}}=\frac{1}{\tau_{\rm tun}}+\frac{1}{\tau_0}\exp(-E_a/kT). \quad (2)$$

In Fig. 2(b), the solid lines indicate the least-squares fits of Eqs. (1) and (2) to the experimental QD PC intensities. It is worth mentioning that the rate constants $1/\tau_{\rm rec}$, $1/\tau_{\rm tun}$, and $1/\tau_0$ determined by Eq. (1) are not unique; only the ratios $\tau_{\rm rec}/\tau_{\rm tun}$ and $\tau_{\rm rec}/\tau_0$ can be uniquely determined. For InAs QD systems, the radiative recombination time constants are typically $\sim$1 ns,[9] as measured by time-resolved PL experiments. Therefore, we set the recombination lifetime to be $\tau_{\rm rec}=1$ ns in the fitting procedure, although this value may not be applicable in present case, since the built-in electric field in our sample might reduce the recombination rate significantly. However, this value is still a good approximation to estimate the relative lifetime of $\tau_{\rm tun}$ and τ_0 with respect to the referent $\tau_{\rm rec}$. In Table I, we list the fitting obtained parameters for different QD states. In this table, the deduced activation energies for the QD, states are smaller than the confinement energy $\Delta E=E_{\rm GaAs}-E_{{\rm QD}i}$, where $E_{\rm GaAs}$ and

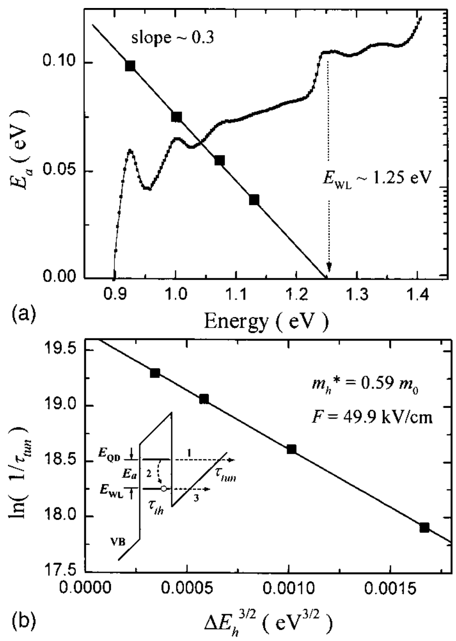

FIG. 3. (a) A plot of the activation energy vs the transition energy and the RT PC spectrum. (b) A plot of the fitted $\ln(1/\tau_{tun})$ vs $(\Delta E_h)^{3/2}$. The inset shows a schematic diagram of direct hole tunneling (1) and thermally assisted hole tunneling (2 and 3).

E_{QDi} are the band-gap energy and the transition energy for the GaAs barrier and the bound QD state, respectively. If the main carrier thermal escape process is exciton dissociation from the bound QD state to the GaAs barrier, the fitted E_a should be quite close to ΔE. However, the smaller E_a values suggest that the escape of photogenerated carriers may not be direct exciton dissociation processes. Furthermore, the E_a values obtained are three times smaller than $E_{WL}-E_{QDi}$. Therefore, we suggest that these smaller activation energies correspond to the escape of the less confined carriers. For lens-shaped InAs QD's, several authors have pointed out that the hole confinement ΔE_h is weaker than the electron confinement ΔE_e.[11] To further clarify the dominant hole escape processes, we plot the fitted E_a vs E_{QDi} in Fig. 3(a). In this figure, we also include the measured PC spectrum for comparison. It is interesting that the relationship between the E_a values obtained and the measured E_{QDi} is found to be linear with a slope of $\sim$0.3. By extrapolating this linear relation to $E_a=0$ eV, the corresponding transition energy is found to be 1.25 eV. When compared with the PC spectrum, this energy just coincides with the WL transition energy. This observation indicates that the path for the dominant hole escape processes is thermally assisted tunneling from the QD hole state to the GaAs valence band via the WL as an intermediate state [inset in Fig. 3(b)]. Moreover, the slope of $\sim$0.3 suggests that the valence-band offset ratio may be roughly $\sim$30%, i.e., $\Delta E_e:\Delta E_h \approx 70:30$, which is consistent with the ratio of 2:1 obtained from capacitance spectroscopy,[12] and also in agreement with a recent report of 70:30 for single InAs monolayers in a GaAs system.[13]

In Table I, the obtained direct tunneling rates $1/\tau_{tun}$ are in the range of $10^7-10^8\ \mathrm{s}^{-1}$; this is significantly slower than the recombination rate ($10^9\ \mathrm{s}^{-1}$). This reveals that the low-temperature QD PC should be weak, since both direct and thermally assisted tunneling rates are minimized. In fact, the

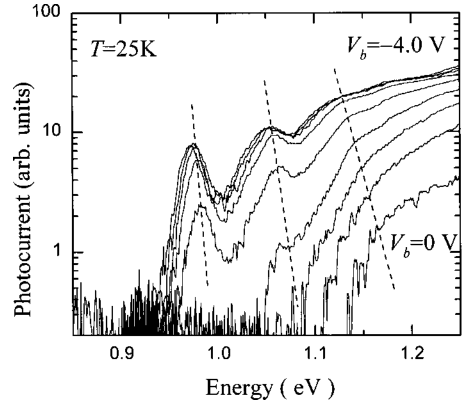

FIG. 4. The bias-dependent PC spectra measured at $T=25$ K. The bias voltages (V_b) for these spectra from the bottom to the top traces are 0, −0.5, −1, −1.5, −2, −2.5, −3, −3.5, and −4 V, respectively.

direct hole tunneling rate is exponentially proportional to the confined barrier height (ΔE_h) and the electric field (F), and is given by[14]

$$\frac{1}{\tau_{tun}} \sim \exp\left(-\frac{4}{3}\frac{\sqrt{2m^*}(\Delta E_h)^{3/2}}{e\hbar F}\right), \tag{3}$$

where m^* is the effective mass and e is the electron charge. As a first approximation, the hole confinement energy can be roughly obtained by using the valence-band offset ratio $Q_V=30\%$, i.e., $\Delta E_h \approx Q_V(E_{GaAs}-E_{QDi})$. Therefore, a plot of $\ln(1/\tau_{tun})$ vs $(\Delta E_h)^{3/2}$ can determine the built-in F. This is shown in Fig. 3(b). By using Eq. (3), if the m^* is chosen to be the heavy-hole effective mass $m_{hh}=0.59m_0$,[15] we obtain $F=49.9$ kV/cm. This value agrees fairly well with the built-in F of 49 kV/cm, calculated by assuming a built-in voltage of 1.5 V in the *p-i-n* structure investigated. It also confirms the proposed hole escape processes, since if we use the electron confinement and electron effective-mass parameters in Eq. (3), the deduced F disagrees completely with the built-in F shown by this structure.

At low temperatures, the suppressed PC signals can be recovered by applying a reverse bias to increase the F exhibited in the intrinsic GaAs layer. In Fig. 4, we display the bias-dependent PC spectra measured at $T=25$ K. When the bias voltage $V_b=0$ V, the QD PC is very weak. With an increasing reverse bias, the suppressed QD PC recovers gradually from the higher-energy QD excited states to the QD ground state. These results clearly demonstrate the behavior of energy-dependent tunneling from the QD's. Note that the energy for the QD PC signals is redshifted with increasing reverse bias. The reasons for this energy redshift are twofold:[16] (i) the size-selective tunneling effect and (ii) the quantum confined Stark effect (QCSE). For a given QD level, the carrier tunneling rates from smaller dots (with higher energies) are faster than those from larger dots. Therefore, carrier tunneling from the high-energy side of the inhomogeneous distribution occurs at a lower applied field. When the applied electric field is high enough, the carrier tunneling rates from all the QD's are fast enough, so this selective tunneling effect disappears and only the QCSE contributes to the energy redshift at the higher electric fields.[16]

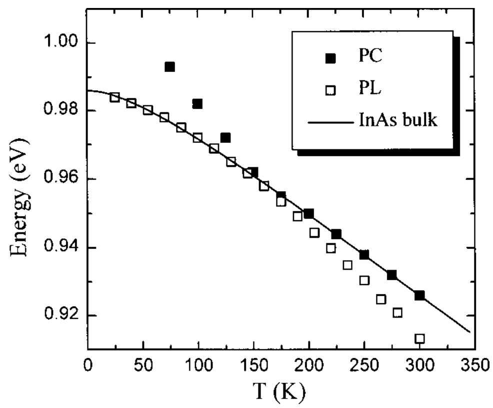

FIG. 5. The temperature-dependent QD peak energies of the PC (E_{PC}), PL (E_{PL}), and the InAs bulk calculated by the Varshini empirical equation.

Before concluding, it is worth remarking that the QD transition energies measured by PC and PL are slightly different. As shown in Fig. 1, the QD peak energies for the RT PC spectrum were slightly higher (~5–15 meV) than for the PL spectrum. Chu *et al.*[17] have ascribed this energy blueshift in the RT PC spectrum to energy-dependent tunneling of carriers through the triangular barrier. However, carrier redistribution among the QD's would also lead to a redshift in the RT PL spectrum.[18] To clarify these two mechanisms, in Fig. 5 we compare the temperature-dependent QD peak energies of the PC (E_{PC}) and the PL (E_{PL}). In this figure, we also include temperature-dependent band-gap shifts in the InAs bulk (E_{InAs}) calculated by the Varshini empirical equation. In this figure, except at temperatures ~150 K, the observed E_{PC} is higher than the E_{PL} in both the low- and high-temperature regions. However, compared with E_{InAs}, it is clear that the energy blueshift in E_{PC} due to energy-dependent carrier tunneling occurs only at $T<150$ K. At higher temperatures ($T>150$ K), E_{PC} will gradually follow E_{InAs}, while E_{PL} becomes redshifted to lower energies due to the carrier redistribution among the QD's. These results clearly demonstrate that the energy difference between RT PC and PL arises from the carrier redistribution effect in PL experiments. We emphasize that the carrier escape process from the QD's is dominated by thermally activated processes at RT. The direct tunneling processes are negligible so the RT PC spectrum directly reflects the absorption spectrum.

In summary, we present the temperature- and bias-dependent PC measurements for an InAs self-assembled QD system. It is found that the carrier escape process is dominated by thermal-assisted hole tunneling, from the dot levels to the GaAs barrier, via the WL as an intermediate state. We also demonstrate the energy-dependent tunneling of carriers from QD's at low temperatures. Both the size-selective tunneling effect and the quantum-confined Stark effect are found to be important for the transition energy shift in PC measurements.

This work was supported in part by the National Science Council of the Republic of China under Grant No. NSC 88-2112-M-008-003.

[1] Y. Arakawa and H. Sakaki, Appl. Phys. Lett. **40**, 939 (1982).
[2] N. Kirstaedter, O. G. Schmidt, N. N. Ledentsov, D. Bimberg, V. M. Ustinov, A. Yu. Egorov, A. E. Zhukov, M. V. Maximov, P. S. Kop'ev, and Zh. I. Alferov, Appl. Phys. Lett. **69**, 1226 (1996).
[3] J. J. Finley, M. Skalitz, M. Arzberger, A. Zrenner, G. Böhm, and G. Abstreiter, Appl. Phys. Lett. **73**, 2618 (1998).
[4] M. C. Bödefeld, R. J. Warburton, K. Karrai, J. P. Kotthaus, G. Medeiros-Ribeiro, and P. M. Petroff, Appl. Phys. Lett. **74**, 1839 (1999).
[5] T. Lundstrom, W. Schoenfeld, H. Lee, and P. M. Petroff, Science **286**, 2312 (1999); W. V. Schoenfeld, T. Lundstrom, P. M. Petroff, and D. Gershoni, Appl. Phys. Lett. **74**, 2194 (1999).
[6] P. N. Brounkov, A. Polimeni, S. T. Stoddart, M. Henini, L. Eaves, P. C. Main, A. R. Kovsh, Yu. G. Musikhin, and S. G. Konnikov, Appl. Phys. Lett. **73**, 1092 (1998).
[7] R. J. Luyken, A. Lorke, A. O. Govorov, J. P. Kotthaus, G. Medeiros-Ribeiro, and P. M. Petroff, Appl. Phys. Lett. **74**, 2486 (1999).
[8] C. M. A. Kapteyn, F. Heinrichsdorff, O. Stier, R. Heitz, M. Grundmann, N. D. Zakharov, D. Bimberg, and P. Werner, Phys. Rev. B **60**, 14 265 (1999).
[9] Weidong Yang, Roger R. Lowe-Webb, Hao Lee, and P. C. Sercel, Phys. Rev. B **56**, 13 314 (1997).
[10] T. M. Hsu, W.-H. Chang, K. F. Tsai, J.-I. Chyi, N. T. Yeh, and T. E. Nee, Phys. Rev. B **60**, R2189 (1999).
[11] R. J. Warburton, C. S. Dürr, K. Karrai, J. P. Kotthaus, G. Medeiros-Ribeiro, and P. M. Petroff, Phys. Rev. Lett. **79**, 5282 (1997).
[12] K. H. Schmidt, G. Medeiros-Ribeiro, M. Oestreich, P. M. Petroff, and G. H. Döhler, Phys. Rev. B **54**, 11 346 (1996).
[13] Raffaele Colombelli, Vincenzo Piazza, Antonio Badolato, Marco Lazzarino, Fabio Beltram Winston Schoenfeld, and Pierre Petroff, Appl. Phys. Lett. **76**, 1146 (2000).
[14] G. Vincent, A. Chantre, and D. Bois, J. Appl. Phys. **50**, 5484 (1979).
[15] M. A. Cusack, P. R. Briddon, and M. Jaros, Phys. Rev. B **56**, 4047 (1997).
[16] P. W. Fry, I. E. Itskevich, D. J. Mowbray, M. S. Skolnick, J. J. Finley, J. A. Barker, E. P. O'Reilly, L. R. Wilson, I. A. Larkin, P. A. Maksym, M. Hopkinson, M. Al-Khafaji, J. P. R. David, A. G. Cullis, G. Hill, and J. C. Clark, Phys. Rev. Lett. **84**, 733 (2000).
[17] L. Chu, M. Arzberger, A. Zrenner, G. Böhm, and G. Abstreiter, Appl. Phys. Lett. **75**, 2247 (1999).
[18] A. Polimeni, A. Patanè, M. Henini, L. Eaves, and P. C. Main, Phys. Rev. B **59**, 5064 (1999).