Temperature dependence of the optical properties of InAs/$Al_yGa_{1-y}As$ self-organized quantum dots

A. Polimeni, A. Patanè, M. Henini, L. Eaves, and P. C. Main
School of Physics and Astronomy, University of Nottingham, NG7 2RD Nottingham, United Kingdom
(Received 5 October 1998)

The photoluminescence properties of InAs/$Al_yGa_{1-y}As$ self-assembled quantum dots are studied as a function of temperature from 10 to 290 K. By varying the Al content the dot luminescence can be tuned over a wavelength range from 0.8 to 1.1 μm, and can be made thermally stable up to room temperature. The temperature dependence of carrier hopping between dots is discussed in terms of the depth of the dot confinement potential and the dispersion in dot size and composition. [S0163-1829(99)01207-2]

I. INTRODUCTION

Self-assembled quantum dots (QD's) in semiconductor heterostructures are of great interest because of their discrete atomlike energy levels, good optical properties, and promising device applications such as QD lasers.[1–3] Great attention has focused on the epitaxial growth of QD's based on (InGa)As/(AlGa)As heterostructures.[2,4,5] In particular, several authors have shown that an appropriate choice of growth conditions provides a means of tuning the optical properties of the dots in a controlled way.[5,6,7]

The effect of temperature T on the optical properties of self-assembled quantum dots is of great importance for their use in devices working at room temperature. Various papers have discussed the temperature dependence of the photoluminescence (PL) properties of QD's in InAs/GaAs.[8–14] $In_xGa_{1-x}As/GaAs$,[15–17] $Al_xIn_{1-x}As/Al_yGa_{1-y}As$ (Ref. 16), and ZnSe/ZnS heterostructures.[18] Similar studies were also performed on laser structures based on $In_{0.5}Ga_{0.5}As$ QD's embedded in (AlGa)As barriers.[19]

It is commonly stated that the dot-luminescence intensity quenches with temperature due to carrier thermal escape from the dot ground states to the wetting layer (WL) and to the states of the matrix material,[9,14,16] and/or nonradiative recombination centers.[12,17,18] An unusual temperature dependence of the linewidth and peak energy ($h\nu$) of the dot ensemble photoluminescence has been also reported:[8–14,16,18] with increasing T the PL linewidth goes through a minimum and the energy of the PL spectrum redshifts faster than the band gap of the dot material. These thermal effects are not expected in an ideal QD structure in which all the dots are decoupled from each other. They have been attributed to a thermally enhanced carrier relaxation between dots due to carrier thermionic emission[12,13,16,18] and/or carrier transport through the WL (Refs. 10, 13, and 14) and tunneling mechanisms.[8,9,13] Alternatively, they have been explained in terms of competition between ground- and excited-state transitions.[11]

This paper reports a systematic study of carrier-transfer phenomena in InAs/$Al_yGa_{1-y}As$ ($y=0-0.8$) self-assembled QD's. We investigate the emission energy and intensity of the PL as a function of temperature and find that an intense PL persists up to room temperature. This high thermal stability of the dot luminescence has been found in all samples, especially for high values of y. Also, changing the Al content allows us to study the role of the QD confining potential V_0 on the carrier hopping. At high T a carrier thermal redistribution between dots is observed and related to both V_0 and the intrinsic disorder of the dot ensemble by means of an analogy with carrier relaxation in disordered quantum wells (QW's).[20,21] In QW's the fluctuations of the well width and/or composition generate an in-plane inhomogeneous potential with local energy minima, which localize excitons analogous to carrier confinement in QD's. This results in optical properties common to both disordered two-dimensional systems and QD structures.

II. EXPERIMENT

Samples were grown by molecular-beam epitaxy on (100)-oriented GaAs substrates. First, a 0.7-μm-thick GaAs buffer layer was grown at 600 °C. Then the growth temperature T_G was reduced. Two different designs were considered. In the first one, three InAs layers were embedded in an $Al_yGa_{1-y}As$ matrix grown at $T_G=520$ °C. The average thickness L of each InAs layer is 1.8 ML. The three InAs layers are separated from each other by 20-nm-thick $Al_yGa_{1-y}As$ barriers. The Al content of the barrier is $y=0.0$, 0.15, 0.3, 0.4, 0.6, and 0.8. In the second design, three layers of dots ($L=1.8$ ML) are embedded each in a $Al_{0.3}Ga_{0.7}As/GaAs$ QW. The GaAs QW and the $Al_{0.3}Ga_{0.7}As$ barriers have a width equal to 10 nm. This structure is grown at $T_G=500$ °C. For both designs, a capping layer of 25 nm of GaAs completed the growth. The dot formation was controlled *in situ* by monitoring the reflection high-energy electron-diffraction pattern. The PL measurements were performed from $T=10$ K to room temperature (RT) ($T=290$ K). The optical excitation was provided by the 514.5-nm-line of an Ar^+ laser. The luminescence was dispersed by a $\frac{3}{4}$ m monochromator and detected by a cooled Ge diode.

III. RESULTS AND DISCUSSION

Figure 1 shows the PL spectra at room temperature for all samples investigated, recorded at the same excitation level of 80 W/cm^2. The luminescence spectra show an intense band due to electron-hole pair recombination from the dots. With

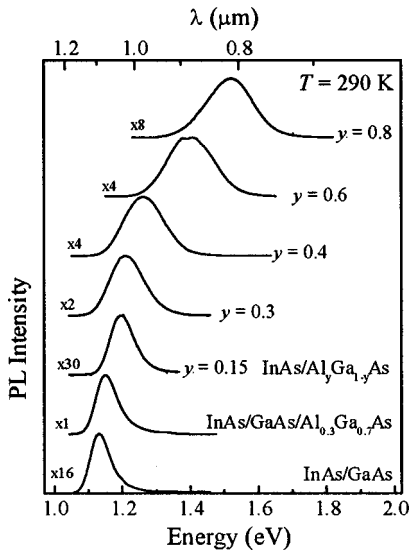

FIG. 1. Room-temperature ($T=290$ K) PL spectra of InAs/Al$_y$Ga$_{1-y}$As self-assembled quantum dots for different y.

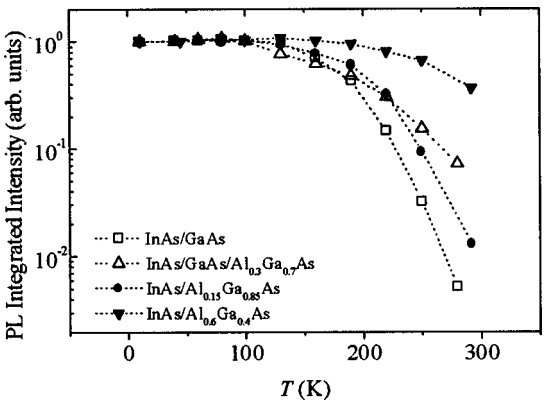

FIG. 2. Temperature dependence of the PL integrated intensity for InAs/GaAs (open squares), InAs/GaAs/Al$_{0.3}$Ga$_{0.7}$As (open triangles), InAs/Al$_{0.15}$Ga$_{0.85}$As (closed circles), and InAs/Al$_{0.6}$Ga$_{0.4}$As (closed triangles) quantum dots. The lines are guides for the eye.

increasing Al content the PL band blueshifts from 1.1 to 0.8 μm. The blueshift is attributed to three effects: (i) a deeper carrier-confining potential at higher values of y;[22] (ii) the formation of smaller dots: transmission electron microscopy studies performed on InAs/(AlGa)As QD's have shown that the dot diameter is smaller with respect to InAs/GaAs QD's;[5] and (iii) Al incorporation in InAs dots during the (AlGa)As overgrowth resulting in the formation of In$_z$Al$_{1-z}$As dots.[5,23] From the stronger blueshift of the luminescence peak energy for $y>0.3$ (see Fig. 1) we infer that the last two mechanisms are dominant for the higher Al concentrations.

The PL linewidth becomes broader with values of the linewidth W (measured as full width at half maximum) larger than 100 meV for $y>0.3$, consistent with the alloying effects and the consequent increase of dot compositional disorder. Finally, the dots embedded in the GaAs/Al$_{0.3}$Ga$_{0.7}$As QW have a PL linewidth and peak position similar to that of dots embedded in GaAs. This indicates that the outer QW with (AlGa)As barriers does not have a large effect on the energy position of the QD levels, as would be expected from the relatively weak additional confinement which it provides.

Figure 2 shows the temperature dependence of the integrated intensity of the PL for four representative samples. In the temperature range from 10 to 290 K the PL signals of dots embedded in GaAs and Al$_{0.15}$Ga$_{0.85}$As matrices decrease by factors of 200 and 80, respectively; the dots grown in the QW structure show a much smaller PL quenching of about an order of magnitude; for $y\geqslant 0.3$ the PL integrated intensity falls by less than a factor of 3 (for clarity we show the data only for $y=0.6$).

Because of the heterogeneous nature of the InAs/(AlGa)As heterostructure, which includes zero- (QD), two- (WL) and three-dimensional (barrier) structures, and the complex mechanisms that regulate the carrier transfer between them, a quantitative analysis of the thermal quenching of the PL from the QD is not straightforward. In particular, our data cannot be modeled simply in terms of an Arrhenius plot with one or more activation energies as is commonly the case for QW systems.[24,25] Also, the small decrease of the luminescence signal (within two orders of magnitude for all samples) does not allow us to extract *univocally* an activation energy.

We suggest that, for dots buried in (AlGa)As barriers with $y\geqslant 0.3$, the luminescence quenches with T mainly due to the presence of low-density nonradiative channels in the (AlGa)As barriers. In fact, the behavior of different samples ($y\geqslant 0.3$) is similar despite them having different QD energy levels. Also, the WL and the (AlGa)As energy levels are too distant from the dot ground state to have an appreciable thermal population (by assuming a wetting layer of about 1.3 ML, the corresponding excitonic energy position has been calculated to be more than 400 meV away from the dot emission energy). In contrast, for shallower confining potentials ($y=0$ and 0.15) the WL and the barrier may play an important role in the thermal decrease of the QD PL. In fact, in the PL spectra of samples having $y=0$ an increasing contribution of the carrier recombination from the WL is observed with increasing T.

The high quality of our samples allows us to investigate the optical properties of the dots in an extended range of temperatures and for different degrees of carrier localization in the dots (i.e., different Al contents of the matrix surrounding the dots). A decrease of the PL intensity by nearly an order of magnitude from $T=5$ to 300 K has been observed by Mukai, Ohtsuka, and Sugawara[17] for (InGa)As/GaAs QD's grown by atomic-layer epitaxy. Previous data on InAs dots buried in (AlGa)As matrices showed a PL signal only at temperatures below 200 K.[16] Also, it has generally reported that the luminescence signal of InAs/GaAs dots is extremely weak or absent for temperatures greater than 200 K.[8–14]

Figure 3(a) shows the temperature dependence of the linewidth W of the dot PL. The variation of W with T depends on the composition of the matrix surrounding the dots. For dots embedded in a GaAs matrix, W goes to a minimum value at a temperature $T\sim 150$ K in agreement with earlier work on similar samples.[8,9,11,13] For InAs/Al$_{0.15}$Ga$_{0.85}$As dots, the minimum shifts to $T\sim 250$ K. Finally, for $y\geqslant 0.3$, W smoothly decreases up to the highest T (in Fig. 3 only $y=0.3$ is shown for clarity). A similar effect has been predicted theoretically in disordered QW in which the inhomogeneous potential due to monolayer fluctuations and alloy disorder localizes carriers:[21] with increasing T excitons can

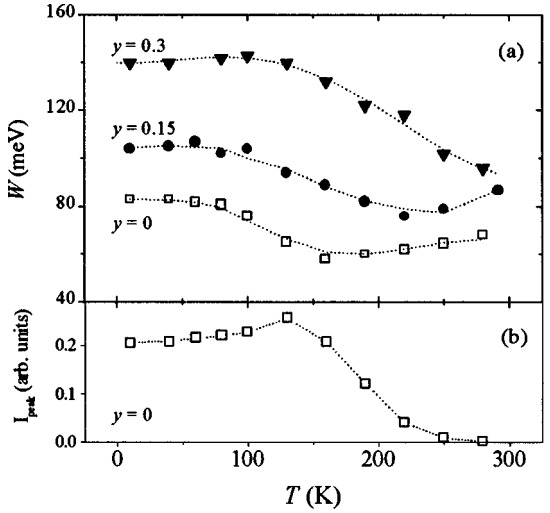

FIG. 3. (a) Temperature dependence of the PL full width at half maximum W of InAs dots embedded in matrices having different Al content; (b) PL peak intensity I_{peak} vs T for InAs/GaAs QD's. The lines are guides for the eye.

overcome shallow energy minima by thermal activation and fall down into deeper states.

It has been proposed that at low temperature, carriers are frozen randomly into the dot states and the PL spectrum reflects the absorption of the QD ensemble.[26] With increasing temperature, carriers may be thermally activated outside the dot into the wetting layer and/or the GaAs barrier and then relax into a different dot. Carrier hopping between dots favors a drift of carriers toward the dots having a higher binding energy, and hence a lower energy emission, resulting in a narrowing of the PL spectrum followed by a luminescence peak redshift larger than that expected for the thermal shrinkage of the InAs band gap with increasing T. Consistent with this explanation, the shift of the W minimum moves toward higher temperature for increasing y. In this case a larger thermal energy is necessary for the carriers to overcome the higher (AlGa)As barriers.

The carrier redistribution between dots is also evident from the temperature dependence of the PL peak intensity I_{peak}, as shown in Fig. 3(b) for the InAs/GaAs QD's. As the PL linewidth shrinks I_{peak} reaches a maximum due to carriers feeding into the lower energy dots from the higher energy ones.

Complementary information regarding the transfer mechanisms between dots can be inferred from the variation of the energy of the QD PL peak $h\upsilon$ with T. Figure 4(a) shows the dependence on T of the shift of $h\upsilon$ $\Delta(h\upsilon_{\mathrm{QD}})$, measured with respect to its value at $T=10\,\mathrm{K}$, for dots embedded in GaAs and $Al_{0.6}Ga_{0.4}As$. For comparison we also show the variation of the InAs band gap $\Delta(h\upsilon_{\mathrm{bulk}})$, calculated using Varshni's law with the InAs parameters.[27]

If carrier hopping between dots were absent, the QD PL peak position would follow the band-gap thermal shrinkage for all T, as has been seen in the temperature dependence of absorptionlike spectra.[28,29] However, the deviation of $\Delta(h\upsilon_{\mathrm{QD}})$ from $\Delta(h\upsilon_{\mathrm{bulk}})$ with increasing T is consistent with carrier redistribution into lower-energy dot states.

The difference between $\Delta(h\upsilon_{\mathrm{bulk}})$ and $\Delta(h\upsilon_{\mathrm{QD}})$ is a measure of the luminescence peak energy offset from the absorption energy maximum, which is usually referred to as the Stokes shift (SS) between luminescence and absorption lines. SS increases with T, and since the deeper $Al_yGa_{1-y}As$ barriers make the carrier hopping between dots less effective, the increase starts at high temperatures with increasing y [see Fig. 4(b)].

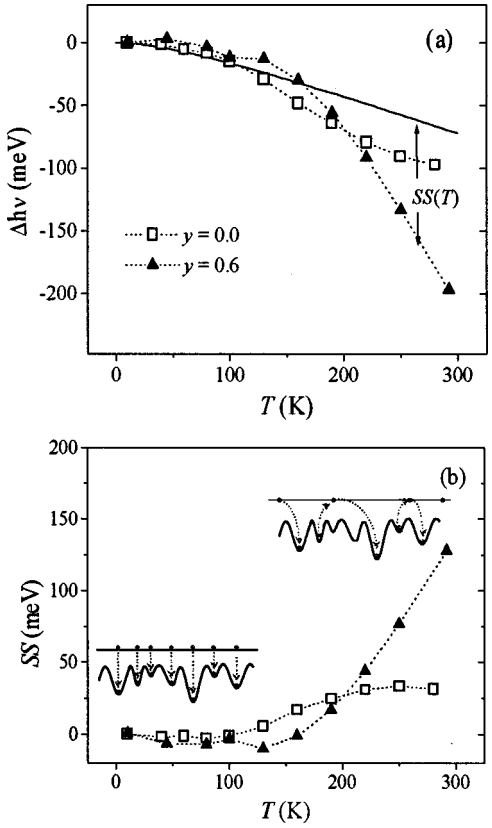

FIG. 4. (a) Dependence on T of the QD PL peak energy $\Delta(h\upsilon_{\mathrm{QD}})$ measured with respect to its value at $T=10\,\mathrm{K}$ for dots embedded in GaAs (open squares) and $Al_{0.6}Ga_{0.4}As$ (closed triangles). For comparison the temperature dependence of the InAs band gap is also shown (continuous line). The difference between $\Delta(h\upsilon_{\mathrm{QD}})$ and $\Delta(h\upsilon_{\mathrm{bulk}})$, SS($T$), is indicated by the double-arrowed line. (b) SS dependence on T for the same samples shown in the upper panel. The insets sketch the carrier relaxation in the dots for low (left) and high (right) temperature regimes.

While at low temperatures carriers recombine from the full range of the dot states, at room temperature they can diffuse more easily in the growth plane and occupy the lowest dot-energy states. Therefore, at RT, SS becomes a good estimate of the QD size dispersion and can be related to the QD absorption linewidth. Figure 5 shows the value of SS at RT as a function of the dot luminescence linewidth measured at 10 K, $W(10\,\mathrm{K})$. As discussed above, the latter quantity can be considered a good estimate of the width of the energy distribution of the dots.

SS shows a linear dependence on $W(10\,\mathrm{K})$ (see diagram). As shown in Fig. 5, the same linear relation between SS and $W(10\,\mathrm{K})$ holds also for InAs/GaAs self-assembled QD's grown under different conditions and with a different degree of disorder. The smallest values of SS are found in systems with the narrowest QD PL linewidth. Note that SS(T) would be zero in the limit of an ensemble of identical dots having the same size and composition. A linear fit to the data gives

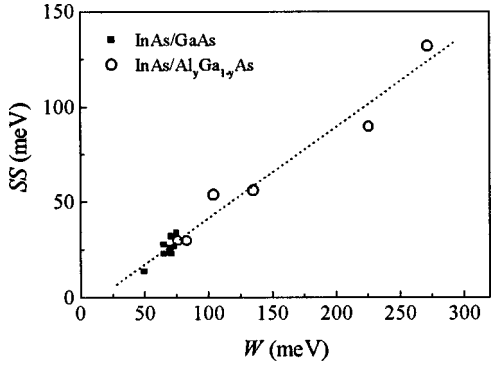

FIG. 5. RT Stokes shift, SS (290 K), of the QD luminescence vs dot PL linewidth measured at 10 K, *W*. Different symbols refer to different series of samples. The InAs/GaAs self-assembled quantum dots considered (full squares) have been grown by using different growth temperatures, substrate orientations, and/or a growth interruption after the InAs deposition. The dashed line is a linear fit to the data.

a slope of about 0.5 and an intercept of 15 meV (see Fig. 5). The same kind of proportionality between the Stokes shift and the absorption linewidth has been found in disordered quantum wells and described by a topological model.[20]

Our optical data show several similarities with those obtained for disordered QW's.

In quantum wells interface and compositional disorder generates a fluctuating potential that may localize excitons. In particular, in (AlGa)Ga/GaAs QW's local-probe techniques have shown δ-like luminescence spectra originating from excitons localized at the QW interface fluctuations.[30–32]

The carrier redistribution between the localized states of the QW depends on temperature. At $T=0$ K carriers populate randomly these states. On the contrary, an initial increase of *T* may favor the carrier relaxation on lower-energy states.[21] As a consequence, the SS may increase with *T* just as we find for QD's. However, because the energy scale of the potential fluctuations in two-dimensional systems is much less than for self-assembled dots, the temperature scale where SS increases is much lower for quantum wells. Also, a narrowing of the exciton luminescence linewidth has been theoretically predicted[21] and experimentally observed[33] in (InGa)As/GaAs quantum wells.

Two-dimensional structures and self-assembled QD's can be described using similar models. Both systems can be modeled as the two limiting situations of a disordered heterostructure, where a weak (QW) or a strong (QD) in-plane inhomogeneous potential localizes carriers. Unusual thermal effects are observed due to carrier localization and thermally enhanced carrier hopping. However, due to the different degree of carrier localization in QW and QD systems these effects manifest themselves in markedly different temperature regimes in the two cases: 10–30 K for QW and 100–200 K for QD.

IV. CONCLUSIONS

We have carried out a systematic study of the PL properties of InAs/Al$_y$Ga$_{1-y}$As self-assembled QD's from 10 to 290 K and over the compositional range $y=0-0.8$. All structures show a strong luminescence intensity up to room temperature with a wavelength emission at 290 K tunable between 1.1 to 0.8 μm. In particular, for an Al content greater than 15% the QD PL integrated intensity quenches very slowly with increasing *T* (by less than an order of magnitude). The high quality of the samples allowed us to study carrier-relaxation phenomena up to room temperature and in QD structures having different carrier-confining potential barriers. We observe evidence for carrier hopping between dots similar to the behavior seen and predicted in highly disordered quantum wells.

ACKNOWLEDGMENTS

This paper and one of us (L.E.) were supported by the Engineering and Physical Sciences Research Council (United Kingdom).

[1] Y. Arakawa and H. Sakaki, Appl. Phys. Lett. **40**, 939 (1982).
[2] N. N. Ledentsov, V. A. Shchukin, M. Grundmann, N. Kirstaedter, J. Böhrer, O. Schmidt, D. Bimberg, V. M. Ustinov, A. Yu. Egorov, A. E. Zhukov, P. S. Kop'ev, S. V. Zaitsev, N. Yu. Gordeev, Zh. I. Alferov, A. I. Borovkov, A. Kosogov, S. S. Ruminov, P. Werner, U. Gösele, and J. Heydenreich, Phys. Rev. B **54**, 8743 (1996).
[3] A. Polimeni, M. Henini, A. Patanè, L. Eaves, P. C. Main, and G. Hill, Appl. Phys. Lett. **73**, 1415 (1998).
[4] J. Y. Marzin, J. M. Gérard, A. Izraël, D. Barrier, and G. Bastard, Phys. Rev. Lett. **73**, 716 (1994).
[5] R. Leon, S. Fafard, D. Leonard, G. L. Merz, and P. M. Petroff, Appl. Phys. Lett. **67**, 521 (1995).
[6] J. M. García, T. Mankad, P. O. Holtz, P. J. Wellman, and P. M. Petroff, Appl. Phys. Lett. **72**, 3172 (1998).
[7] S. Malik, C. Roberts, R. Murray, and M. Pate, Appl. Phys. Lett. **71**, 1987 (1997).
[8] D. I. Lubyshev, P. P. González-Borrero, E. Marega, Jr., E. Petiprez, N. La Scala, Jr., and P. Basmaji, Appl. Phys. Lett. **68**, 205 (1996).
[9] Z. Y. Xu, Z. D. Lu, X. P. Yang, Z. L. Yuan, B. Z. Zheng, J. Z. Xu, W. K. Ge, Y. Wang, and L. L. Chang, Phys. Rev. B **54**, 11 528 (1996).
[10] L. Brusaferri, S. Sanguinetti, E. Grilli, M. Guzzi, A. Bignazzi, F. Bogani, L. Carraresi, M. Colocci, A. Bosacchi, P. Frigeri, and S. Franchi, Appl. Phys. Lett. **69**, 3354 (1996).
[11] H. Lee, W. Yang, and P. C. Sercel, Phys. Rev. B **55**, 9757 (1996).
[12] Y. T. Dai, J. C. Fan, Y. F. Chen, R. M. Lin, S. C. Lee, and H. H. Lin, J. Appl. Phys. **82**, 4489 (1997).
[13] A. Patanè, M. G. Alessi, F. Intonti, A. Polimeni, M. Capizzi, F. Martelli, M. Geddo, A. Bosacchi, and S. Franchi, Phys. Status Solidi A **164**, 493 (1997).
[14] Y. Tang, D. H. Rich, I. Mukhametzhanov, P. Chen, and A. Madhukar, J. Appl. Phys. **84**, 3342 (1998).
[15] R. P. Mirin, J. P. Ibbetson, K. Nishi, A. C. Gossard, and J. E. Bowers, Appl. Phys. Lett. **67**, 3795 (1995).

[16] S. Fafard, S. Raymond, G. Wang, R. Leon, D. Leonard, S. Charbonneau, J. L. Merz, P. M. Petroff, and J. E. Bowers, Surf. Sci. **361**, 778 (1996).
[17] K. Mukai, N. Ohtsuka, and M. Sugawara, Appl. Phys. Lett. **70**, 2416 (1997).
[18] Y. Wu, K. Arai, and T. Yao, Phys. Rev. B **53**, R10 485 (1996).
[19] A. E. Zhukov, V. M. Ustinov, A. Yu. Egorov, A. R. Kovsh, A. F. Tsatsulnikov, N. N. Ledentsov, S. V. Zaitsev, N. Yu. Gordeev, P. S. Kop'ev, and Zh. I. Alferov, Jpn. J. Appl. Phys., Part 1 **36**, 4216 (1997).
[20] F. Yang, M. Wilkinson, E. J. Austin, and K. P. O'Donnell, Phys. Rev. Lett. **70**, 323 (1993).
[21] R. Zimmermann and E. Runge, Phys. Status Solidi A **164**, 511 (1997).
[22] M. Grundmann, O. Stier, and D. Bimberg, Phys. Rev. B **52**, 11 969 (1995).
[23] O. Baklenov, D. L. Huffaker, A. Anselm, D. G. Deppe, and B. G. Streetman, J. Appl. Phys. **82**, 6362 (1997).
[24] J. D. Lambkin, D. J. Dunstan, K. P. Homewood, L. K. Howard, and M. T. Emeny, Appl. Phys. Lett. **57**, 1986 (1990).
[25] S. Weber, W. Limmer, K. Thonke, R. Sauer, K. Panzlaff, and G. Bacher, Phys. Rev. B **52**, 14 739 (1995).
[26] R. Heitz, M. Grundmann, N. N. Ledentsov, L. Eckey, M. Veit, D. Bimberg, V. M. Ustinov, A. Yu. Egorov, A. E. Zhukov, P. S. Kop'ev, and Zh. I. Alferov, Appl. Phys. Lett. **68**, 361 (1996).
[27] Y. P. Varshni, Physica (Amsterdam) **34**, 149 (1967).
[28] L. Aigouy, T. Holden, F. R. Pollak, N. N. Ledentsov, V. M. Ustinov, P. S. Kop'ev, and D. Bimberg, Appl. Phys. Lett. **70**, 3329 (1997).
[29] Photocurrent measurements performed on InAs/GaAs self-assembled quantum dots show that the photocurrent signal peak of the dots follows the thermal shift of the InAs band gap [I. E. Itskevich (unpublished)].
[30] H. F. Hess, E. Betzig, T. D. Harris, L. N. Pfeiffer, and K. W. West, Science **264**, 1740 (1994).
[31] K. Brunner, G. Abstreiter, G. Böhm, G. Tränkle, and G. Weimann, Appl. Phys. Lett. **64**, 3320 (1994).
[32] D. Gammon, E. S. Snow, B. V. Shanabrook, D. S. Katzer, and D. Park, Phys. Rev. Lett. **76**, 3005 (1996).
[33] Photoluminescence and photoluminescence excitation measurements performed on (InGa)As/GaAs QW's [A. Patanè, M. Grassi Alessi, M. Capizzi, and F. Martelli (unpublished)].