# High-temperature light emission from InAs quantum dots

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We study the photoluminescence (PL) properties of InAs/GaAs self-assembled quantum dots in a temperature range ($T=300-500$ K) above that reported to date. Various power excitation densities were used, allowing us to identify the important contribution of nonradiative channels in quenching the dot PL as the temperature is increased. The role played by the wetting layer on the dot PL intensity has been investigated in samples in which the separation of the dot and wetting layer levels is tuned by post-growth annealing. This experiment reveals that, at a high temperature (>300 K), the relative population of the dot and wetting layer levels is given by a Boltzmann distribution. © *1999 American Institute of Physics.* [S0003-6951(99)00432-5]

The study of the effect of temperature, $T$, on the optical properties of self-assembled quantum dots (QDs),[1,2] is of great importance for the exploitation of the zero-dimensional properties of the dots in optoelectronic devices working at room temperature.[3–5] Several mechanisms have been considered to explain the thermal behavior of the dot optical properties (i.e., the dot luminescence lineshape and intensity) and their effect on device performance. In particular, the deterioration of the QD optical efficiency with increasing $T$ has been found to be highly sensitive to the quality of the dot growth[6–9] and to the electronic structure of the QD.[10–13]

In this letter, we describe the high-temperature ($T=300-500$ K) photoluminescence (PL) properties of InAs self-assembled QDs embedded in a GaAs/(AlGa)As quantum well. This temperature regime, higher than those reported to date, allows us to identify clearly the main channels responsible for carrier depopulation of dot levels at typical device operation temperatures ($\geqslant$300 K). A study of the dot PL under various power excitation densities indicates the dominant role of nonradiative channels in the thermal quenching of the dot luminescence. We also investigate the role of the wetting layer (WL) formed beneath the dots in the redistribution of carriers with changing $T$. The analysis involves samples in which we vary the separation between the energy levels in the dots and in the WL. We find that the relative population of the dot and WL levels at high temperature (>300 K) is well described by the Boltzmann distribution and that the relative activation energy corresponds to the energy difference of the corresponding lines in the PL spectrum.

The structures investigated were grown by molecular beam epitaxy on (100)-oriented GaAs substrates. First, a 0.7-$\mu$m-thick GaAs buffer layer was grown at 600 °C. Then the growth temperature, $T_G$, was reduced to 500 °C and three layers of dots (InAs thickness, $L=1.8$ ML) were grown, each layer embedded in a $Al_{0.3}Ga_{0.7}As$/GaAs quantum well (QW) (see inset Fig. 1). The GaAs QW and the $Al_{0.3}Ga_{0.7}As$ barriers each have a width of 10 nm. A 25 nm GaAs capping layer completed the growth. The dot formation was controlled *in situ* by monitoring the reflection high-energy electron-diffraction pattern. Atomic force microscopy characterization of uncapped samples grown under the same conditions gave a dot density $\rho=2\times10^{11}\ \mathrm{cm}^{-2}$, average diameter $d=15$ nm and average height $h=1.5$ nm. The capped sample grown in this way is called S1. For samples S2 and S3, we applied a 30 min post-growth annealing under As overpressure at $T_A=630$ and 680 °C, respectively. PL measurements were performed from room temperature (RT) ($T=300$ K) up to $T=500$ K. The optical excitation was provided by the 514.5 nm line of an $Ar^+$ laser. The luminescence was dispersed by a 3/4 m monochromator and detected by a cooled Ge diode.

Figure 1 shows the normalized PL spectra of the unannealed sample S1 at various temperatures. The low- and high-energy bands are due to carrier recombination in the dot (QD) and WL levels, respectively. The temperature range (300–500 K) shown in Fig. 1 allows us to resolve clearly the emission from the WL and to establish quantitatively its relative weight with respect to that of the dots at different $T$. Previously this has not been possible because of the limited range of temperature investigated (<300 K).[2,6–13] Also note that a clear luminescence signal persists up to 500 K. This high thermal stability can be attributed to the low level of thermal escape of carriers from the dots and the WL towards the two-dimensional levels of the GaAs/(AlGa)As QW.[2,3] The effectiveness of this structure also has been exploited in a QD laser device in which we observe lasing action from the dots up to 400 K.[14]

With increasing temperature, the overall PL intensity decreases and the relative weight of the WL emission increases with respect to that of the dots. Figure 2 shows the temperature dependence of the total integrated PL intensity for a variation of power densities ($P$) over more than two orders of magnitude. The stronger decay rate of the total luminescence with increasing $T$ (dot and WL bands) for lower $P$ indicates a carrier emission out of the InAs confined states toward nonradiative centers, which tend to saturate with increasing $P$. The nonradiative centers are probably located in the (AlGa)As barrier layers which are grown at 500 °C in order to avoid In segregation and desorption from the QDs. This growth temperature is well below the optimum value of ~600 °C generally used for optoelectronic-grade

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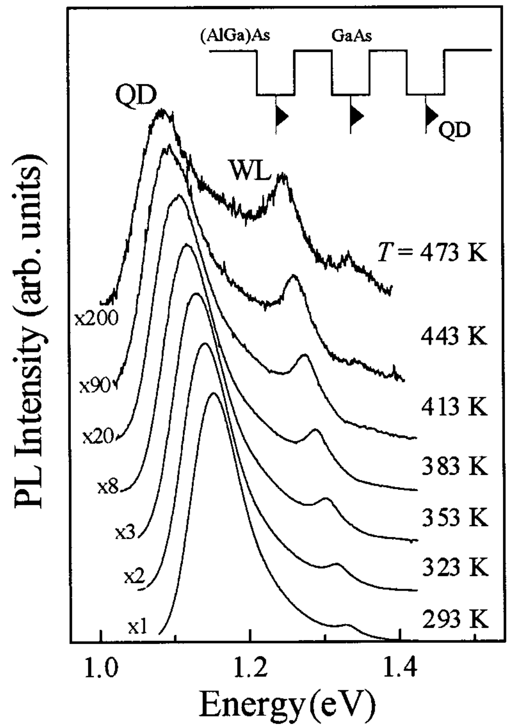


FIG. 1. Normalized PL spectra of sample S1 at different temperatures. The excitation power is $P = 15\ \mathrm{W/cm^2}$. The low- (QD) and high-energy (WL) bands are due to carrier recombination in the dot and WL levels, respectively. The inset sketches the conduction band and profile of the sample.

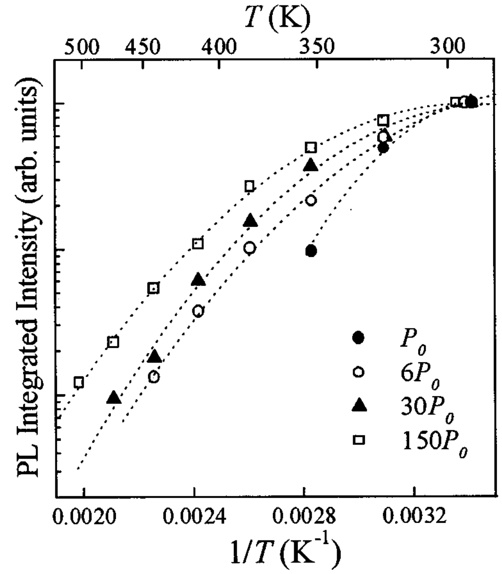


FIG. 2. PL integrated intensity for sample S1 as a function of temperature for different laser power densities. $P_0$ is equal to $0.5\ \mathrm{W/cm^2}$. Data are normalized to their value at RT ($T = 293$ K) for comparison purposes. Dotted lines are guides for the eye.

(AlGa)As.[15] The presence of nonradiative recombination centers makes a quantitative analysis of the thermal quenching of the dot PL rather complicated. In particular, the integrated intensity of the QD line alone cannot be modeled simply in terms of an Arrhenius plot with a well-defined activation energy. These results highlight the importance of nonradiative channels in determining the luminescence intensity and indicate the relevance of the power excitation in any analysis of the dependence of the dot luminescence on temperature.[13]

The increasing intensity of the WL emission with respect to that of the dot with increasing temperature indicates a redistribution of carriers from the dots towards the two-dimensional WL. The WL acts as a drain of carriers and possibly as an intermediate channel for carrier capture into nonradiative recombination centers. In order to investigate carrier recycling between the WL and the QDs, we studied the ratio, $R$, between the WL and the QD PL peak intensities for a set of samples, in which the energy separation between the QD and WL bound states is different for the different samples.

The inset of Fig. 3 (right side) shows the PL spectra recorded at $T = 110\,°\mathrm{C}$ and $P = 15\ \mathrm{W/cm^2}$ of sample S1 and of the two annealed samples S2 and S3. The dot luminescence undergoes a blueshift and a linewidth narrowing moving from sample S1 to S3. In contrast, the WL peak position does not change. Similar observations have been already reported by other groups[16,17] and we will not discuss them further here. In our investigation, the annealing is simply a means of tuning the energy separation, $\Delta E_{\mathrm{PL}}$, between the dot and WL energy position as measured from the PL spectra. The effect of different values of $\Delta E_{\mathrm{PL}}$ is readily observed on the temperature dependence of $R$ shown on the left side of Fig. 3. Values of $R$ may be fitted by $A\exp(-\Delta E/k_BT)$, where $A$ is a constant. The best fit to the data gives $\Delta E = 160$, 110, and 40 meV for samples S1, S2, and S3, respectively. These values are in remarkable agreement with the experimental values $\Delta E_{\mathrm{PL}}$ (see the insert of Fig. 3). Similar results were obtained for different power densities.

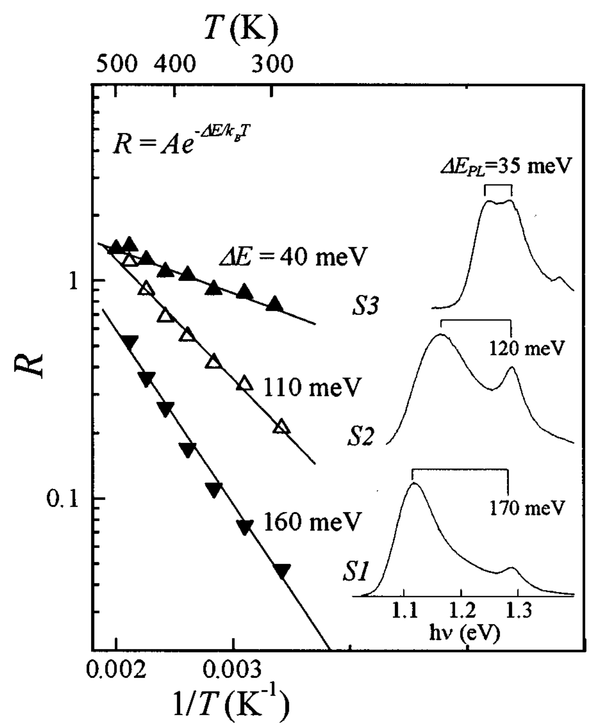


FIG. 3. Ratio, $R$, between the WL and the QD PL intensity as a function of temperature for samples S1, S2, and S3. The straight lines are fit to the data by the formula $A\exp(-\Delta E/k_BT)$. The fit value of $\Delta E$ is reported in the figure for each sample. The insert shows the PL spectra at $T = 383$ K and $P = 15\ \mathrm{W/cm^2}$ for the samples studied. Also the energy distance between the WL and the QD PL band is shown.

Our data can be explained in terms of a quasi-equilibrium distribution of carriers between the dots and the WL levels at high $T$. In fact, the QD (WL) PL intensity can be written as[18]

$$I_{\mathrm{QD(WL)}} = B_{\mathrm{QD(WL)}} n_{\mathrm{QD(WL)}} p_{\mathrm{QD(WL)}}, \tag{1}$$

where $n$ and $p$ are the electron and hole populations of the QD (WL) levels and $B$ is a term given by the probability for radiative recombination. If both types of carrier obey Boltzmann statistics at the lattice temperature $T$, the ratio $R = I_{\mathrm{WL}}/I_{\mathrm{QD}}$ can be rewritten as

$$R = (B_{\mathrm{WL}}/B_{\mathrm{QD}}) C_e C_h e^{-(\Delta E_e + \Delta E_h)/k_B T} \sim A e^{-\Delta E/k_B T}, \tag{2}$$

where $C_e$ ($C_h$) is the ratio of the electron (hole) effective density of states of the wetting layer to that of the dots, $\Delta E_e$ ($\Delta E_h$) is the energy spacing between the WL and QD electron (hole) levels, and $\Delta E$ ($= \Delta E_e + \Delta E_h$) corresponds to the energy difference between the electron-hole recombination in the WL and the dots. Since the $T$-dependence of $B$ and $C$ is much weaker than exponential, $R$ can be approximated by $\exp(-\Delta E/k_B T)$, consistent with the results shown in Fig. 3. This analysis remains valid when an excitonic recombination from the dots and the WL levels is considered. In this case, $R$ can be described in terms of the relative population of two (excitonic) levels separated by $\Delta E$.

Our analysis shows the important role played by the WL levels in thermal escape of carriers from the dots. This mechanism has to be taken into account, for example, in the realization of QD laser devices working at RT, where it is responsible for the increase of the threshold current density with temperature[5] and for the blueshift of the maximum laser gain from the dot toward the WL emission energy.[19]

In conclusion, we have performed a study of the PL properties of self-assembled QDs at temperatures above 300 K. Our investigation reveals the importance of nonradiative channels in the decay of the total luminescence intensity and the importance of the power density employed in the analysis of the dot PL thermal quenching. A study performed on structures having different electronic properties reveals a carrier redistribution with temperature between the dots and the WL levels where the relative populations follow the Boltzmann distribution.

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[1] D. Bimberg, M. Grundmann, and N. N. Ledentsov, *Quantum Dot Heterostructures* (Wiley, New York, 1999).

[2] A. Polimeni, A. Patanè, M. Henini, L. Eaves, and P. C. Main, Phys. Rev. B **59**, 5064 (1999), and references therein.

[3] M. V. Maximov, I. V. Kochnev, Yu. M. Shernyakov, S. V. Zaitsev, N. Yu. Gordeev, A. F. Tsatsul'nikov, A. V. Sakharov, I. L. Krestnikov, P. S. Kop'ev, Zh. I. Alferov, N. N. Ledentsov, D. Bimberg, A. O. Kosogov, P. Werner, and U. Gösele, Jpn. J. Appl. Phys., Part 1 **36**, 4221 (1997).

[4] A. Polimeni, M. Henini, A. Patanè, L. Eaves, P. C. Main, and G. Hill, Appl. Phys. Lett. **73**, 1415 (1998).

[5] A. Patanè, A. Polimeni, M. Henini, L. Eaves, P. C. Main, and G. Hill, J. Appl. Phys. **85**, 625 (1999).

[6] 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).

[7] K. Mukai, N. Ohtsuka, and M. Sugawara, Appl. Phys. Lett. **70**, 2416 (1997).

[8] Y. Wu, K. Arai, and T. Yao, Phys. Rev. B **53**, R10485 (1996).

[9] 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).

[10] 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**, 11528 (1996).

[11] Y. Tang, D. H. Rich, I. Mukhametzhanov, P. Chen, and A. Madhukar, J. Appl. Phys. **84**, 3342 (1998).

[12] 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).

[13] H. Lee, W. Yang, and P. C. Sercel, Phys. Rev. B **55**, 9757 (1996).

[14] A. Patanè, A. Polimeni, M. Henini, L. Eaves, P. C. Main, P. Smowton, P. Blood, and G. Hill (unpublished).

[15] M. Ilegems, in *The Technology and Physics of Molecular Beam Epitaxy*, edited by E. H. C. Parker (Plenum, New York, 1985), p. 83.

[16] R. Leon, Y. Kim, C. Jagadish, M. Gal, J. Zou, and J. H. Cockyne, Appl. Phys. Lett. **69**, 1888 (1996).

[17] A. O. Gosogov, P. Werner, U. Goesele, N. N. Ledentsov, D. Bimberg, V. M. Ustinov, A. Yu. Egorov, A. E. Zhukov, P. S. Kop'ev, N. A. Bert, and Zh. I. Alferov, Appl. Phys. Lett. **69**, 3072 (1996).

[18] J. I. Pankove, *Optical Processes in Semiconductors* (Dover, New York, 1975).

[19] F. Heinrichsdorff, M. H. Mao, N. Kirstaedter, A. Krost, D. Bimberg, A. O. Kosogov, and P. Werner, Appl. Phys. Lett. **71**, 22 (1997).